# 13th JEMS Joint European Magnetic Symposia

**27th August to 1st September 2023**

Faculty of Medicine, UCM

**M A D R I D**

# BOOK OF ABSTRACTS

www.jems2023.es

13th JEMS Joint European Magnetic Symposia

## ORGANIZERS

instituto iMdea nanociencia — EXCELENCIA SEVERO OCHOA

EMa The EUROPEAN MAGNETISM ASSOCIATION

## IN COLLABORATION WITH

MADRID CONVENTION BUREAU

GRUPOPACIFICO The power of meeting

## SUPPORTED BY

UNIVERSIDAD COMPLUTENSE MADRID

UAM Universidad Autónoma de Madrid

## SILVER SPONSOR

Quantum Design EUROPE

## SPONSORS & EXHIBITORS

CIQTEK

PASSENGER FOR SUSTAINABLE AND EFFICIENT RE-FREE MAGNETS EU H2020 Project (Ref. 101003914)

Qnami

evicomagnetics

IMA Precision in Magnetics

METALPINE THE ART OF SPHERICAL POWDER

MONOLITHOS CATALYSTS - RECYCLING - INNOVATION

nanotech SOLUTIONS

UK Magnetic Society

Lake Shore CRYOTRONICS

MBN nanomaterialia

## OUTLINE

# PLENARY SESSIONS

# Shaping the future: energy-efficient magnetic memory and neuromorphic technologies

*Jordi Sort[1,2], Zhengwei Tan[1], Nicolau López[1], Zheng Ma[1], Monalisha Peda[1], Julius de Rojas[1], José L. Costa-Krämer[3], Alberto Quintana[4], Eva Pellicer[1], Josep Nogués[2,5], Enric Menéndez[1]*

[1]*Departament de Física, Universitat Autònoma de Barcelona, 08193 Bellaterra, Spain*

[2]*ICREA, Pg. Lluís Companys 23, 08010 Barcelona, Spain*

[3]*IMN-Instituto de Micro y Nanotecnología (CNM-CSIC), 28760 Tres Cantos, Madrid, Spain*

[4]*Institut de Ciència de Materials de Barcelona (ICMAB-CSIC), Bellaterra 08193, Spain*

[5]*Catalan Institute of Nanoscience and Nanotechnology (ICN2), CSIC and BIST, Campus UAB, Bellaterra, 08193 Barcelona, Spain*

Controlling magnetism with voltage has an enormous potential to boost energy efficiency in nanoscale magnetoelectric devices since the use of electric fields (instead of magnetic fields or electric currents) minimizes Joule heating effects and reduces the overall device power consumption. In recent years, we have demonstrated the possibility to induce reversible, non-volatile changes in the magnetic properties (coercivity, remanent magnetization and saturation magnetization) of nanoporous films consisting of metal alloys (e.g., CuNi, FeCu) or oxides (e.g., $FeO_x$, $CoFe_2O_4$), by applying an electric field through a liquid electrolyte gate at room temperature [1,2]. In addition, we have made significant progress in the field of magneto-ionics (i.e., voltage-driven ion transport in magnetic materials), which has traditionally relied on controlled migration of oxygen or lithium ions. Here, I will show that voltage-driven transport of nitrogen ions can be also triggered at room temperature in transition metal nitride (CoN, FeN, CoMnN and CoFeN) films via liquid electrolyte gating [3,4]. Nitrogen magneto-ionics can induce reversible ON-OFF transitions of ferromagnetic states at faster rates and lower threshold voltages than oxygen magneto-ionics. This is due to the lower activation energy needed for ion diffusion and the lower electronegativity of nitrogen with cobalt, compared with oxygen. Remarkably, and in contrast to oxygen magneto-ionics, nitrogen transport occurs uniformly through a plane-wave-like migration front, without the assistance of diffusion channels, which is particularly interesting for the implementation of multi-stack memory devices. Furthermore, we will show that both oxygen and nitrogen magneto-ionics can be used to emulate some important neuromorphic/synaptic functionalities (spike amplitude-dependent plasticity, spike duration-dependent plasticity, long term potentiation/depression). By tuning ion cumulative effects of DC and pulsed voltage actuation (at frequencies in the range 1 – 100 Hz), learning, memory retention, forgetting and self-learning by maturity (post-stimulated learning) can be mimicked. The latter can serve as a logical function for the device to decide between self-learning or forgetting emulation, at will, post-voltage input. This constitutes a novel approach to emulate some specific neural functionalities (*e.g.*, learning under deep sleep), that are challenging to achieve using other classes of materials currently employed for neuromorphic computing applications.

## Acknowledgements

Financial support by the European Research Council (REMINDS 2021-Advanced Grant, Agreement No. 101054687), the European Union's Horizon 2020 research and innovation programme (European Training Network, BeMAGIC ETN/ITN Marie Skłodowska-Curie grant No. 861145, the Spanish Research Agency (PID2020-116844RB-C21 and PDC2021-121276-C31) and the Catalan Government though the AGAUR (2021-SGR-00651) is acknowledged.

## References

[1] A. Quintana et al., *Adv. Funct. Mater*. **27** (2017) 1701904.
[2] C. Navarro-Senent et al., *APL Mater.* **7** (2019) 030701.
[3] J. de Rojas et al., *Nat. Commun.* **11** (2020) 5871.
[4] J. de Rojas et al, *ACS Appl. Mater. Interfaces* **13** (2021) 30826–30834.
[5] Z. Tan et al., *Mater. Horiz.* **10** (2023) 88–96.

# Orbitronics, from present to future with orbit currents induced by charge/spin currents, FMR or light for torques or TeraHz emission.

Albert Fert [1], Henri-Yves Jaffrès [1*]
[1] Unité Mixte de Physique, CNRS, Thales, Université Paris-Saclay, 91767 Palaiseau, France

To start in the usual situation of orbit currents generated by charge currents, I will present experiments [1] showing the strong enhancement of the current-induced torques on the Co layer of a Pt/Co bilayer by the addition of an Al layer on Co (Al protected from oxidation). The enhancement is predominant for the FL torque, up to factor of 9. The interpretation [2] comes from ab-initio calculations showing large Co orbital moments in the interfacial Co layer with a helical texture similar to the spin texture on the surface of topological insulators (see Fig.1). The calculation of the resulting torques leads to a good agreement between the calculated and experimental torques.

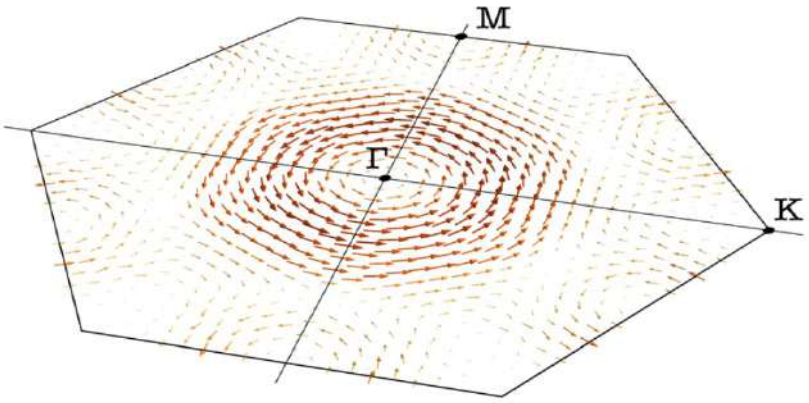



Fig.1. *Helical locking of orbit moment **L** (arrows) with momentum **k** in Brillouin zone in the plane of the Co layer at the interface with Al and in a small energy range close to Fermi energy. A similar texture between spin **S** and **k** in the plane is of much smaller amplitude.*

In another set of experiments [3] on light-induced teraHz emission by NM/F bilayers (NM = nonmagnetic metal, deposited on glass substrate, F = ferromagnetic metal or alloy), we show that, in contrast with results for other ferromagnets, Ni leads to large teraHz signals with the same polarity for NM = Pt, Ta, W and also a large teraHz signal for Cu. After measurements of the Anomalous Hall Effect (AHE) contributions on Ni single layers, we can rule out an explanation of our results by AHE and ascribe the teraHz signals of the NM/Ni bilayers to a significant light-induced production of orbit current by Ni followed by the predominance of Inverse Orbit Hall Effect (OOHE) on Inverse Spin hall Effect (ISHE). This property of Ni is consistent with ab-initio calculations and FMR-induced orbit currents with Ni.

**References:**

[1] S. Khrishnia, A. Fert, H. Jaffrès et al., https://doi.org/10.1021/acs.nanolett.2c05091
[2] S. Nikolaev et al (In preparation). S.Nikolaev (Osaka Univ.) and collaborations with M. Blanco, A. Arnau (UPV and DIPC at San Sebastian), M. Chshiev (Spintec, Grenoble), H. Jaffrès and A. Fert.
[3] arxiv:2208.01866 and submitted, Y. Xu et al., (collaboration between MIIT Key Lab. of Spintronics and Hefei IRI, Beihang University at Hefei with UMR CNRS-Thales at University Paris-Saclay).

# Permanent Magnet Alternatives: An Effective Strategy for Mitigating Supply Chain Risks?

*Thomas A. Lograsso* [1,*]

[1] *Critical Materials Institute, Ames National Laboratory, 134 Wilhelm Hall, Ames IA 50014*

*lograsso@ameslab.gov

Substitutions are at the heart of the clean energy transition. While most people think substitution is about a finding a replacement material as good as the one that is critical, in fact, this is just one aspect of the substitution strategy. Substitution, the replacement of one item for another, can be applied at four levels. First is element-for-element substitution. This can be total replace a critical element with one that is readily accessible in the quantities needed. Second is a material-by-material substitution. This is what is commonly thought of when substitution is thought of i.e., an "drop in" alternate that is better, cheaper, and less reliant on critical materials. History tells us this is a rare case indeed. Third is a process-by-process substitution and is often associated with manufacturing innovations that are driven by lowering costs through energy reduction, reduced material usage or environmental improvements. The Critical Materials Institute has led the United States efforts to develop substitute permanent magnets with reduced or no rare earth content. Our approach is three-fold. We look to substitute for the rare earth elements with select elemental substitutions that reduce the critical rare earth content while retaining magnetic anisotropies. We look for other alternative formulations that make use of "abundant rare earths" and iron. This strategy targets magnet performance between the rare earth magnets and non-rare earth magnets, i.e., the so-called gap magnet. Our third approach is to utilize advanced manufacturing for process improvements that can maximize magnetic performance while minimize wastage.

**Acknowledgements**

Critical Materials Institute is an Energy Innovation Hub funded by the U.S. Department of Energy, Office of Energy Efficiency and Renewable Energy, Advanced Manufacturing Office, Ames National Laboratory is operated for the U.S. Department of Energy by Iowa State University of Science and Technology under Contract No. DE-AC02-07CH11358.

# A new spin on neural networks

J. Grollier, CNRS,Thales, Université Paris-Saclay, Palaiseau, France

Neuromorphic computing aims to accelerate artificial intelligence and reduce its power consumption by a factor of 100 by drawing inspiration from the architecture of the brain. This ambitious undertaking requires inventing new materials for synaptic and neural functions, and assembling these nanodevices into systems capable of running state-of-the-art machine learning algorithms. The challenges are to densely integrate and connect hundreds of millions of neural and synaptic components, and have them compute and learn with high accuracy and low energy despite device imperfections. An emerging branch of research studies how such physical neural networks can be assembled by leveraging physical principles, and how they could learn to perform cognitive tasks by directly using physical principles, such as energy minimization.

In this talk, I will provide an overview of the challenges of neuromorphic computing, explain why spin-based systems could help solve these challenges, and detail our recent progress towards this goal. I will focus on two examples that illustrate how we can exploit the multifunctionality of spintronic devices to form hardware neural networks that can be efficiently scaled up. In the first one, the vectors of information are nanoscale moving skyrmions that mimic neuro-transmitters. In the second one, we exploit radio frequency signals and frequency multiplexing to enhance the network's connectivity.

# Current-induced magnetization switching of ferromagnets, ferrimagnets and noncollinear antiferromagnets

Pietro Gambardella

Department of Materials, ETH Zurich, Hönggerbergring 64, 8093 Zurich, Switzerland

Spin-orbit coupling allows for efficient transfer of angular momentum between lattice, orbital, and spin degrees of freedom in magnetic heterostructures. This in turn enables unprecedented control over magnetization reversal and domain wall motion in a variety of materials and devices [1]. Illustrative examples include 3-terminal magnetic tunnel junctions, in which the combination of spin-orbit torques, spin transfer torque, and voltage control of magnetic anisotropy leads to sub-ns magnetization reversal with very narrow switching distributions [2,3], current-induced domain wall and skyrmion motion in insulating magnetic layers [4,5], and the possibility to perform logic operations using current-driven domain walls [6,7]. Further, spin-orbit torques provide a practical means to investigate fast spin dynamics in materials with antiferromagnetic interactions. Time-resolved measurements of current-induced magnetization switching of rare-earth transition-metal ferrimagnetic alloys performed by Hall effect [8] and x-ray imaging [9] show that an electric current excites the magnetization of the different sublattices asynchronously in time and inhomogeneously in space depending on the strength of the antiferromagnetic coupling. This behavior originates from the unequal transfer of angular momentum from the current to the rare-earth and transition-metal sublattices, which leads to a master-agent type of dynamics akin (but different) to that observed for ultrafast optical excitations. The electrical manipulation of chiral antiferromagnetic order in a topological antiferromagnet such as $Mn_3Sn$, instead, proceeds via a two-step demagnetization-remagnetization process that lasts several tens of ns and is governed by the temporal overlap of current-induced heating and spin-orbit torques generated by a Pt or W layer adjacent to it [10]. These results illustrate advantages and limitations of electric currents for the manipulation of spin textures in different magnetic materials.

## References

[1] A. Manchon et al., *Current-induced spin-orbit torques in ferromagnetic and antiferromagnetic systems,* Rev. Mod. Phys. **91**, 035004 (2019).
[2] E. Grimaldi et al., *Single-shot dynamics of spin-orbit torque and spin transfer torque switching in 3-terminal magnetic tunnel junctions*, Nat. Nanotech. **15**, 111 (2020).
[3] V. Krizakova et al., *Spin-orbit torque switching of magnetic tunnel junctions for memory applications,* J. Mag. Magn. Mater. **562**, 169692 (2022).
[4] S. Vélez et al., *High-speed domain wall racetracks in a magnetic insulator,* Nat. Comm. **10**, 4750 (2019).
[5] S. Vélez et al., *Current-driven dynamics and ratchet effect of skyrmion bubbles in a ferrimagnetic insulator*, Nat. Nanotech. **17**, 834 (2022).
[6] Z. Luo et al., *Current-driven magnetic domain-wall logic*, Nature **579**, 214 (2020).
[7] Z. Luo et al., *Field- and current-driven magnetic domain-wall inverter and diode*, Phys. Rev. Applied **15**, 034077 (2021).
[8] G. Sala et al., *Real-time Hall-effect detection of current-induced magnetization dynamics in ferrimagnets*, Nat. Comm. **12**, 656 (2021).
[9] G. Sala et al., *Asynchronous current-induced switching of rare-earth and transition-metal sublattices in ferrimagnetic alloys*, Nat. Mater. **21**, 640 (2022).
[10] G. K. Krishnaswamy et al., *Time-dependent multistate switching of topological antiferromagnetic order in $Mn_3Sn$,* Phys. Rev. Appl. **18**, 024064 (2022).

## Magnetic hyperthermia in combination with other anti-tumoral therapeutic approaches: from the preparation of the magnetic nanomaterials to their preclinical studies

Teresa Pellegrino
Italian Institute of Technology, via Morego 30, 16163, Genoa, Italy

In Magnetic hyperthermia (MHT) magnetic nanoparticles act as heat transducers to convert magneto-energy into heat when exposed to alternating magnetic fields (AMF). At AMF of clinical use (100 kHz and up to 24kA/m) and the magnetic actuation of the nanoparticles has no tissue-depth attenuation enabling to activate magnetic nanoparticle located at deep tumor sites. This provides more selective heat treatment with less side effects which, furthermore, can be combined with other therapeutic approaches to couple toxic effects towards cancer cells.

This talk aims to provide an overview of our last five years research efforts to combine MHT with chemotherapy, intrinsic nanoparticle cytotoxicity, internal radiotherapy and more recently immunotherapy. I will first focus on the latest progress on non-hydrolytic methods for the preparation of magnetic nanoparticles and our strategies to assemble them in well-defined nanostructures with the aim of optimizing their heat performance in MHT. Then, I will introduce thermo-responsive polymer coated magnetic nanoplatforms as drug carriers platform for doxorubicin with MHT heat-mediated drug release mechanism. Finally, I will present our magnetic nano systems for combining MHT with internal radiotherapy or for dual therapy based on magneto-thermia and photo-thermia. Throughout the presentation, I will discuss our preclinical results to evaluate the magnetic hyperthermia efficacy and bio-distribution for some of the best performing materials we are developing.

# SEMY-PLENARY SESSIONS

# Development of high performance permanent magnets; elements criticality, new demands, and extrinsic magnetic properties

*H. Sepehri-Amin*, Xin Tang, J. S. Zhang, A. Bolyachkin, T. Ohkubo, K. Hono*
*National Institute for Materials Science, 1-2-1 Sengen, Tsukuba, 305-0047, Japna*
*h.sepehriamin@nims.go.jp

Permanent magnets are widely used in green energy conversion applications. They therefore play an important role in achieving net-zero $CO_2$ emissions in our society. In order to maintain sustainable production of permanent magnets in the long term, it is necessary to eliminate the dependence of permanent magnets on critical elements such as Dy and to diversify the use of rare earths while maintaining sufficiently high coercivity and energy product in the magnets.

We will first present our fundamental research on the coercivity mechanism of Nd-Fe-B based permanent magnets which has provided us guideline to develop high coercivity Dy-free Nd-Fe-B magnets for applications in hybrid/electric vehicle traction motors and wind turbine generators. We will show how grain boundary/interface engineering in the hot formed Nd-Fe-B magnets has resuled in a high coercivity of 2.5 T, a remanent magnetisation of 1.32 T and excellent thermal stability of the coercivity [1]. Furthermore, it is discussed how the emergence of new applications for the permanent magnets has opened up new requirements, *i.e.* moderate room temperature coercivity and flat first order reversal curves for their applications in variable-magnetic-force motors, which can be achieved by reducing the grain size and using light rare earths in the hot-deformed (Nd,Ce,La)-Fe-B permanent magnets [2].

In the second part of the talk, we will discuss the potential of Fe-rich $SmFe_{12}$ based magnets [3], and the current challenges to realise these materials as new permanent magnets [4-5].We will show our recent success in realising a sufficiently large coercivity of 1.0 T in rare earth lean $SmFe_{12}$ based anisotropic sintered magnets (Fig. 1) assisted by machine learning [6]. Based on detailed microstructural characterisations, modelled thin films and micromagnetic simulations, the optimal microstructure that can lead to higher coercivity and remanent magnetisation in the $SmFe_{12}$-based magnets will be discussed.

## References

[1] M. Korent *et al.* Scripta Mater. **205** (2021) 114206.
[2] X. Tang, H. Sepehri-Amin, *et al.* Acta Mater. **228** (2022) 117747.
[3] K. Ohashi *et al.* J. Appl. Phys. **64** (1988) 5714-5716.
[4] P. Tozman, H. Sepehri-Amin, *et al.* Acta Mater. **153** (2018) 354.
[5] H. Sepehri-Amin *et al.* Acta Mater. **194** (2020) 337.
[6] J.S. Zhang *et al.* Acta Mater. **238** (2022) 118228.

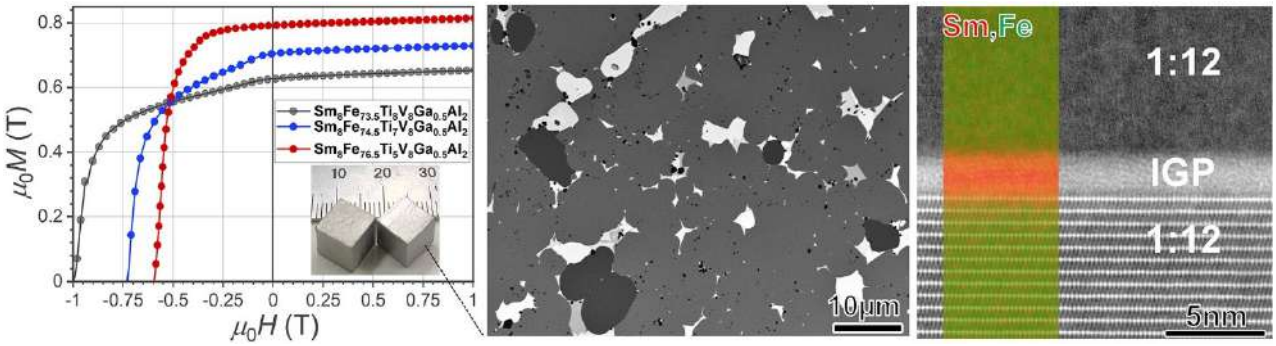



Figure 1: Realizing coercvity in anisotropic $Sm(Fe,Ti,V)_{12}$-based sintered magnets; demagnetization curves of $Sm_8Fe_{73.5+x}Ti_{8-x}V_8Ga_{0.5}Al_2$ at. % (x=0-3) sintered magnets, and backscattered electron (BSE) SEM image, high-angle annular dark-field (HAADF) scanning transmission electron microscopy (STEM) image, and superimposed STEM-EDS maps of Sm and Fe showing the overal microstructure of the magnet with coercivity of 1.0 T.

# Spin Manipulation based on Orbital Angular Momentum

*YoshiChika Otani[1]*, Junyeon Kim[2]*

[1] *Center for Emergent Matter Science, RIKEN, Wako, Saitama, Japan*

[2] *Institute for Solid State Physics, The University of Tokyo, Kashiwa, Chiba, Tokyo, Japan*

*yotani@riken.jp; yotani@issp.u-tokyo.ac.jp

A novel platform for spin manipulation based on orbital angular momentum (OAM) is preparing the next spintronics innovation. We could create a nonzero OAM at surfaces and interfaces with structural inversion asymmetry or in centrosymmetric bulks, similar to how spins are generated by the inverse spin Galvanic effect. These generation mechanisms, however, are unrelated to the spin-orbit coupling (SOC) strength, highlighting the distinction from spin generation mechanisms. Several recent experimental reports suggest that the nonzero OAM could be used for highly efficient spin manipulation. These reports show light metal elements can also be used for OAM-induced spin manipulation.

Furthermore, the collaborative spin manipulation by the OAM and the spin angular momentum (SAM) reveals a new mechanism. Here we review so far reported experimental and theoretical new findings, including our works on OAM-induced or OAM–assisted spin manipulation. All these new findings indicate that the OAM-induced spin manipulation opens the door to novel spintronics in terms of mechanism and materials.

**Acknowledgments**

JK and YO appreciate the financial support from JSPS KAKENHI (Grant No. 19K05258 and 19H05629).

## Single nuclear spin, a fascinating platform for quantum experiments

Franck Balestro. Néel Institut - CNRS, France

The realization of a universal quantum computer is one of the major scientific objectives of the 21st century, as its promises are revolutionary: inviolable cryptography, higher computing power, simulation processes inaccessible to conventional technologies ... Its principle is based on qubit, a two-level quantum system, a quantum analogue of the classical bit. Today, the challenge is to increase the number of qubits in interaction to achieve more complex and more efficient quantum information protocols. In this context, molecular magnets of nanometric size are of major interest. The information is carried by the direction of magnetization which is multiple, unlike conventional magnets which have only two. They thus make it possible to have d-states quantum devices or qudits. These multi-level devices could the processes involved in the manipulation of quantum spin more efficient. The use of these would also simplify some computational tasks, and thus the circuits required to realize a quantum computer

In this context, it is possible to fully control a multilevel system based on a single molecular magnet. Proving that it is possible to read-out and manipulate the four-level spin of the terbium nucleus of a molecule was a first step to show the long decoherence time of a single nuclear spin which is by nature strongly isolated from its environment.

Among those properties, superposition of states and phase interference are two fundamental mechanisms from which quantum computing can benefit. We explored these properties by applying interference protocols involving the phases of the four nuclear spin states. A first measurement makes it possible to know the coherence time of the coherent superposition of a quantum system. Applied to a coherent superposition of three nuclear spin states, this protocol can be generalized to any given qudit. Using another protocole in a physical system that evolves adiabatically and cyclically, one can highlight, a phase that depends on the entire evolution during a cycle.

# New Frontiers in Nanoscale Magnetism: Towards Three Dimensional Materials and Devices

*Amalio Fernández-Pacheco*[1,2]*

[1] *Instituto de Nanociencia y Materiales de Aragón, CSIC-University of Zaragoza, C/Pedro Cerbuna 12, 50009, Zaragoza, Spain*

[2] *SUPA, School of Physics & Astronomy, University of Glasgow, G12 8QQ, Glasgow, UK*

*amaliofp@unizar.es

The expansion of nanomagnetism to three dimensions provides exciting opportunities to explore new physical phenomena and opens great prospects to create 3D magnetic devices for green computing technologies [1-3].

In this talk, I will present some of our recent works dedicated to the investigation of three dimensional artificial magnetic materials, including multilayered and complex-shaped geometries. The talk will give an overview of the new methods we have developed to fabricate [4,5] and characterize [6,7] these nanomaterials, and some of the new functionalities obtained. This includes the creation of localized spin textures, topological defects and stray fields exploiting a combination of geometrical effects and inter-element interactions [8-10], the automotive 3D motion of domain walls [11], an unconventional angular dependence of magnetotransport effects in 3D circuits [12], and the generation of chiral spin interactions across interlayers via interfacial effects in synthetic antiferromagnetic multilayers [13].

**Acknowledgements**

I would like to thank all my group members and collaborators contributing to the work I will be presenting. This work is supported by the European Community under the Horizon 2020 Program, Contract No. 101001290 (3DNANOMAG).

**References**

[1] R. Streubel et al, J. Phys. D: Appl. Phys. 49 363001 (2016).
[2] A. Fernández-Pacheco et al, Nature Comm. 8, 1 (2017).
[3] D. Makarov et al, Adv. Mater. 34, 2101758 (2021).
[4] L. Skoric et al, Nano Letters 20, 184 (2020).
[5] D. Sanz-Hernández et al, Nanomaterials 8, 483 (2018).
[6] D. Sanz-Hernández et al, ACS Nano 11, 11066 (2017).
[7] M.A. Cascales-Sandoval et al, arXiv:2212.02975
[8] D. Sanz-Hernández et al, ACS Nano 14, 8084 (2020).
[9] C. Donnelly et al, Nature Nanotechnol. 17, 136 (2022).
[10] J. Fullerton et al, Nanotechnology 34, 125301 (2023).
[11] L. Skoric et al, ACS Nano 16, 8860 (2022).
[12] F. Meng et al, ACS Nano 15, 6765 (2021).
[13] A. Fernández-Pacheco et al, Nature Mater. 18, 679 (2019).

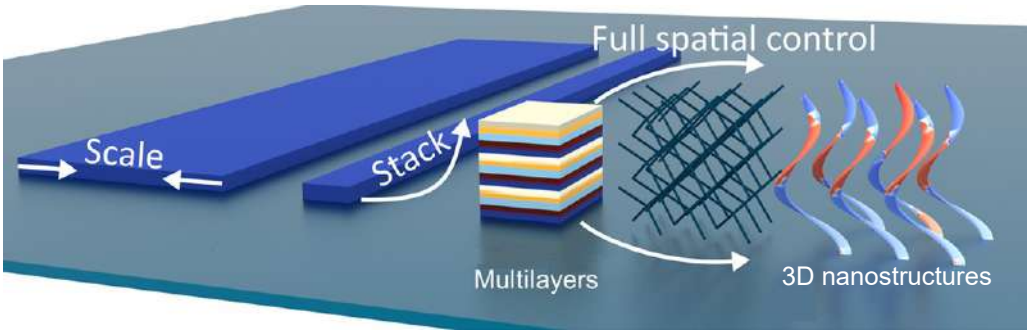



Figure: Towards 3D nanomagnetic materials and devices.

# High performance spintronic devices from microwave technology to computing

*Giovanni Finocchio*[1]

[1] *University of Messina, Viale F. Stagno D'Alcontres 31, Messina, Italy*

*gfinocchio@unime.it

In this talk, I will present recent advances achieved in the development of spintronic microwave detectors, oscillators and amplifiers based on magnetic tunnel junctions (MTJs). I will review the main applications of those devices for computing including the realization of Ising machines based on probabilistic computing with p-bits. The spintronic technology takes advantage of the manipulation of the electron spin together with its charge. This technology potentially combines important characteristics such as ultralow power needs, compactness (nanoscale size) and it is CMOS-compatible. Spintronics has different success stories such as the head read for magnetic hard drive and the recent spin-transfer-torque magnetic random access memories. The latter are realized with MTJs which are devices composed by two ferromagnets separated by a ultrathin isolating material. The resistance of this device depends on the relative orientation of the magnetization of the two ferromagnets and in particular the configuration where the magnetization are parallel or antiparallel can code the binary information. Together with memory developments, which are already in the market and integrated within the CMOS processes by main foundries (INTEL, SAMSUNG, GlobalFoundries), MTJs can be used for the development of auto-oscillators and very high efficient detectors. In detail, I will show the applications of spintronic diodes based on MTJs for energy harvesting, sensors and RF detectors and what it is expected to achieve in the next three years for integration with CMOS-technology. I will also present, theoretical predictions on how voltage controlled magnetocrystalline anisotropy (VCMA) can be used to excite linear and parametric resonant modes in easy-axis antiferromagnetic materials AFMs with perpendicular anisotropy, thus opening the way for an efficient electrical control of the Néel vector, and for electrical detection of THz dynamics. [1] In particular, I will focus on two key results: (i) VCMA parametric pumping experiences the so-called "exchange enhancement" of the coupling efficiency and, thus, is 1-2 orders of magnitude more efficient than microwave magnetic fields or spin-orbit-torques, and (ii) zero-field parametric resonance, which cannot be achieved by other parametric pumping mechanisms in AFMs with out-of-plane easy axis.

The latter part of the talk will focus on probabilistic computing which is one direction to implement Ising Machines. Probabilistic computing is a computational paradigm using probabilistic bits (p-bits), unit in the middle between standard bit and q-bits. I will show how to map hard combinatorial optimization problems (Max-Sat, Max-Cut, etc) into Ising machine and how to implement those in spintronic technology. [2] [3]

**Acknowledgements**

This work was supported under the project number 101070287 — SWAN-on-chip — HORIZON-CL4-2021-DIGITAL-EMERGING-01, the project PRIN 2020LWPKH7 funded by the Italian Ministry of University and Research., and by the PETASPIN association (www.petaspin.com).

**References**

[1] R. Tomasello, R. Verba, V. Lopez-Dominguez, F. Garesci, M. Carpentieri, M. Di Ventra, P. Khalili Amiri, and G. Finocchio, *Antiferromagnetic Parametric Resonance Driven by Voltage-Controlled Magnetic Anisotropy*, Phys. Rev. Appl. **17**, 034004 (2022).

[2] N. A. Aadit, A. Grimaldi, M. Carpentieri, L. Theogarajan, J. M. Martinis, G. Finocchio, and K. Y. Camsari, *Massively Parallel Probabilistic Computing with Sparse Ising Machines*, Nat. Electron. **5**, 460 (2022).

[3] A. Grimaldi, L. Sánchez-Tejerina, N. Anjum Aadit, S. Chiappini, M. Carpentieri, K. Camsari, and G. Finocchio, *Spintronics-Compatible Approach to Solving Maximum-Satisfiability Problems with Probabilistic Computing, Invertible Logic, and Parallel Tempering*, Phys. Rev. Appl. **17**, 024052 (2022).

# Magnetism at the Limit: from Skyrmions to Antiferromagnets in Model-type Systems, Studied with STM

*Kirsten von Bergmann*

*Department of Physics, University of Hamburg*

*kirsten.von.bergmann@physik.uni-hamburg.de

Complex magnetic order arises due to the competition of different magnetic interactions. Often the dominant interaction is the isotropic pair-wise exchange between neighboring atomic magnetic moments. With additional sizable contributions from the Dzyaloshinskii-Moriya-interaction or from the exchange between more distant neighbors any length-scale spin texture ranging from ferromagnetic to antiferromagnetic order can form. Spin-polarized scanning tunneling microscopy (SP-STM) is well suited to investigate complex magnetic order down to the atomic scale [1].

Recently we have investigated prototype antiferromagnetic states in hexagonal magnetic layers, ranging from collinear via coplanar to non-coplanar [2]. In such systems higher-order exchange interactions can play a role for the formation of the magnetic ground state as they can favor either multi-q states or uniaxial states depending on the sign. In such systems we have experimentally observed a new type of antiferromagnetic domain wall with perpendicularly oriented nearest neighbor magnetic moments in it's center [3]. Also nano-scale two-dimensionally modulated magnetic states can arise due to higher-order magnetic interactions at zero magnetic field [4,5]. In such systems these higher-order interactions also can also stabilize collinear counterparts of nanoskyrmion lattices, reminescent of nano-scale ferrimagnets [5]. The balance between these different magnetic interactions creates endless possibilites for complex magnetic order, either as magnetic ground state or at magnetic defects such as domain walls, possibly having an impact not only on static magnetic order but also on dynamical properties and response to currents.

**Acknowledgements**

I gratefully acknowledge financial support from the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) via projects no. 402843438 and no. 418425860. In addition this project has received funding from the European Union's Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement No 955671.

**References**

[1] K. von Bergmann et al., *J. Phys.: Cond. Mat.* **26**, 394002 (2014).
[2] J. Spethmann et al., *Phys. Rev. Lett.* **124**, 227203 (2020).
[3] J. Spethmann et al., *Nature Commun.* **12**, 3488 (2021).
[4] S. Heinze et al., *Nature Phys.* **7**, 713 (2011).
[5] M. Gutzeit et al., *Nature Commun.* **13**, 5764 (2022).

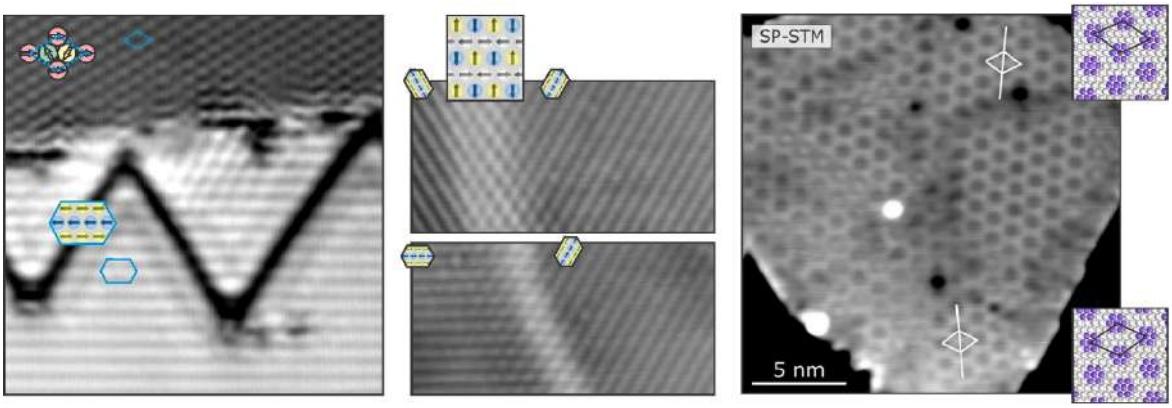



Figure: SP-STM measurements of different atomic- or nano-scale magnetic states; arrows and colors indicate the direction of atomic magnetic moments.

# A Simple Model of Anisotropy in Compositionally-Disordered Rare Earth Magnets

*Christopher E. Patrick[1], L. H. Lewis[2,3] and Julie B. Staunton[4]**

[1] *Department of Materials, University of Oxford, Oxford OX1 3PH, UK*

[2] *Department of Chemical Engineering, Northeastern University, Boston, MA 02115, USA*

[3] *Department of Mechanical and Industrial Engineering, Northeastern University, Boston, MA 02115, USA*

[4] *Department of Physics, University of Warwick, Coventry CV4 7AL, UK*

*J.B.Staunton@warwick.ac.uk

"One-twelve" magnets with compositions RFe12-xTx (R = Rare earth, T = transition metal) are under intense investigation as potential rare earth-lean permanent magnets [1]. The transition metal T is added in order to stabilize the magnet in bulk, and randomly substitutes Fe atoms. These substitutions break the point symmetry at the rare earth site, such that different rare earth atoms are subject to different crystal field potentials, depending on the number and proximity of T dopants. Since the crystal field determines the magnetocrystalline anisotropy, this phenomenon means a nominally crystalline material may nonetheless contain a distribution of rare earth atoms with inhomogeneous single-ion anisotropies.

In order to understand and quantify this effect, we have carried out first-principles calculations of crystal-field coefficients in compositionally-disordered systems, using a numerically stable method developed previously [2]. We have found that these high-level calculations can be represented by a model which is sufficiently simple that it can be incorporated into large-length scale simulations and statistical analyses. As such, our work provides a route to achieving more realistic simulations of magnetization reversal and domain wall propagation in magnets where the crystalline symmetry is disrupted by substitutional defects.

**Acknowledgements**

We gratefully acknowledge the support of the UK EPSRC, Grants No. EP/M028941/1 and EP/W021331/1. This work is also supported in part by the U.S. Department of Energy, Office of Basic Energy Sciences under Award Number DE SC0022168 and by the U.S. National Science Foundation under Award ID 2118164.

**References**

[1] P. Tozman, H. Sepehri-Amin, K. Hono, Scripta Mat. **194** (2021) 113686.

[2] C. E. Patrick, J. B. Staunton, J. Phys.: Condens. Matter **31** (2019) 305901.

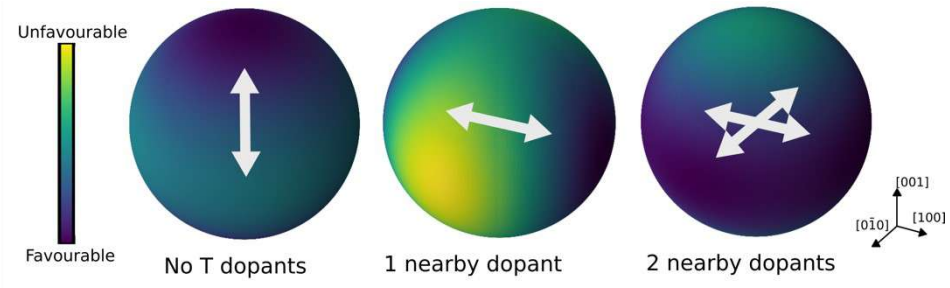



Figure 1: Effect of T-doping on the single ion anisotropy of the rare earth. Arrows represent the easy directions of magnetization, which coincide with the (energetically-favourable) dark blue regions.

# Synthetic-Ferrimagnetic Co/Gd Systems for Spintronics

*R. Lavrijsen*[*1], *T.J. Kools*[1], *P. Li*[1], *Y.L.W. van Hees*[1], *H. Pezeski*[1], *M.C.H. de Jong*[1], *J. Hintermayer*[1], *B. Koopmans*[1]

[1]*Department of Applied Physics, Eindhoven University of Technology, Eindhoven, Netherlands*

*r.lavrijsen@tue.nl

Ferrimagnets [1] and then in particular for this contribution; layered synthetic ferrimagnets based on heterostructures of Co and Gd bear promise for bridging the gap between volatile information in the photonic domain and non-volatile information in the magnetic domain [2,3]. The layered nature of the synthetic approach allows for strong spin-orbitronic effects and single pulse all-optical switching (AOS), the engineering of these, and hence new ways to address the underlying physical mechanisms at play.

In this talk I will introduce the material system, discuss growth considerations and then expand on a few detailed studies. Specifically, I will (i) discuss the relevant effects that should be taken into account when designing synthetic ferrimagnets based on Co and Gd for spintronic applications [4,5]. (ii) Introduce our results on fast current-induced domain wall motion up to 2 km/s in quad-layers [6]. This will then be combined with work on (iii) single shot AOS focusing on the relatively low threshold fluence and ion irradiation studies [7,8]. (iv) Finally, I will give an outlook of where we think the synthetic ferri-magnetism field is heading and elaborate on ongoing studies to characterise the interfacial Dzyaloshinskii-Moriya interaction of these systems for chiral magnetism. All these components will be discussed from a materials engineering viewpoint.

Our studies establish Co/Gd-based synthetic ferrimagnets to be a unique material platform which opens up many routes of inquiry into the underlying competing mechanisms and its applications [1].

**References**

[1] Y. Zhang, X. Feng, Z. Zheng, Z. Zhang, K. Lin, X. Sun, G. Wang, J. Wang, J. Wei, P. Vallobra, Y.Hu, Z Wang, L. Chen, K. Zhang, Y. Xu, W. Zhao, Appl. Phys. Rev. 10, 011301 (2023)
[2] H. Pezeshki, P. Li, R. Lavrijsen, M. Heck, E. Bente, J. van der Tol, B. Koopmans, Accepted at Phys. Rev. Appl. (2023)
[3] F.E. Demirer, Y. Baron, S. Reniers, D. Pustakhod, R. Lavrijsen, J. van der Tol, B. Koopmans, Nanophotonics 0165 (2022)
[4] T.J. Kools, M.C. van Gurp, B. Koopmans, R. Lavrijsen, Appl. Phys. Lett. 121, 202405 (2022)
[5] T.J. Kools, Y.L.W. van Hees, K. Poissonnier, P. Li, B. Barcones Campo, M.A. Verheijen, B. Koopmans, R. Lavrijsen, In-Preparation (2023)
[6] P. Li, T.J. Kools, B. Koopmans, R. Lavrijsen, Adv. Elec. Mater. 9(1), 2199-160X (2022)
[7] P. Li, J.W. van der Jagt, M. Beens, J. Hintermayr, M.A. Verheijen, R. Bruikman, B. Barcones, R. Juge, R. Lavrijsen, D. Ravelosona, B. Koopmans, Appl. Phys. Lett. 121, 172404 (2022)
[8] Y.L.W. van Hees, B. Koopmans, R. Lavrijsen, Appl. Phys. Lett. 120, 252401 (2022)
[9] L. Wang, H. Cheng, P. Li, Y. Liu, Y.L.W. van Hees, R. Lavrijsen, X. Lin, K. Cao, B. Koopmans, W. Zhao, PNAS 119(24) (2022)

# SYMPOSIUM 01.
## MAGNETIC MATERIALS FOR ENERGY APPLICATIONS.
## S1. INVITED ORAL PRESENTATIONS

# Potential of Magnetic Shape Memory Ribbons in Magnetic Refrigeration

*P. Álvarez-Alonso[1]*, R. López-Antón[2], J.P. Camarillo[3], D. Salazar[4], V.A. Chernenko[4]*

[1] *Departamento de Física, Universidad de Oviedo, C/Calvo Sotelo, 18, Oviedo, Spain.*
[2] *Departamento de Física Aplicada, Universidad de Castilla-La Mancha, Av. Camilo José Cela, 10, Ciudad Real, Spain.*
[3] *Universidad Autónoma de Zacatecas, Ramón López Velarde 801, Col. Centro, Zacatecas, Mexico.*
[4] *BCMaterials, Barrio Sarriena s/n, Lejona, Spain.*

*alvarezapablo@uniovi.es

Magnetic refrigeration is an ecoalternative to gas compression/expansion refrigerant technology that is on the final stage to be economically competitive for end-user applications and already available for industrial processes. It is based on the magnetocaloric effect (MC), that is, changes in isothermal entropy/adiabatic temperature in MC materials associated with the application of a magnetic field [1]. Although other caloric effects, such as the elastocaloric effect, are interesting for solid-state refrigeration purposes, the MC effect has concentrated the main interest of the scientific community until now [2].

Among the candidates as MC refrigerants, Magnetic Shape Memory Alloys (MSMAs) are outstanding functional materials that can experience large MC effect associated with the thermo-magnetically driven martensitic transformation that they undergo [3]. In particular, giant MC effects have been achieved in Heusler-type Ni-Mn-X (X=Ga, In, Sn) alloys, leading them to the podium of the MC materials [4]. Although in recent decades MC materials were thoroughly investigated in bulk form to discover their fascinating properties and develop applications for energy harvesting, actuators, or sensors, scientific and technological interest is currently focused on materials with unconventional geometries such as thin films [5]. In this sense, engineering MSMA in ribbon shape is a successful strategy to manufacture low-cost specimens showing both good mechanical properties and fast heat exchange as a result of the increase in surface-to-volume ratio.

In this work, we will uncover the advantages and remarkable potential of Ni-Mn-based Heusler-type ribbons for magnetic refrigeration. We will summarize our latest results comparing their properties with those of the bulk counterparts, paying special attention to the influence of the specific microstructure of the ribbons on their magnetic properties.

**Acknowledgements**

The support received from the following organizations is gratefully acknowledged: Asturian government, and European FEDER (project number: AYUD/2021/51822) and the Basque Government Department of Education (project: IT1479-22).

**References**

[1] J. Romero Gómez, R. Ferreiro Garcia, A. De Miguel Catoira, M. Romero Gómez, *Renew. Sustain. Energy Rev.* **17** (2013) 74–82.
[2] H. Johra, C. Bahl in: L. Itard, L. Hensen-Centnerová, A. Boerstra, P. Bluyssen (Eds.), Proc. CLIMA2022 14th REHVA HVAC World Congr., TU Delft Open, Delft (2022) 1–8.
[3] T. Bachaga, J. Zhang, M. Khitouni, J.J. Sunol, *Int. J. Adv. Manuf. Technol.* **103** (2019) 2761–2772.
[4] V. Franco, J.S.S. Blázquez, J.J.J. Ipus, J.Y.Y. Law, L.M.M. Moreno-Ramírez, A. Conde, *Prog. Mater. Sci.* **93** (2018) 112–232.
[5] A.A. El-Gendy, J.M. Barandiarán, R.L. Hadimani, Magnetic nanostructured materials: From lab to fab, 1st edition, Elsevier, Cambridge, MA, 2018.

# Cross-coupling contribution to the multicaloric effect in magnetic materials

*Lluís Mañosa*[1]*, *Enric-Stern Taulats*[1], *Adrià Gràcia-Condal*[1,2], *Antoni Planes*[1]

[1] *Departament de Física de la Matèria Condensada, Universitat de Barcelona, 08028 Barcelona, Catalonia. Spain*

[2] *CELLS-ALBA Synchrotron, 08290 Cerdanyola del Vallès. Catalonia. Spain.*

*lluis.manosa@fmc.ub.edu

Multicaloric effects result from the application or removal of diverse external fields and they are enhanced in materials with strong coupling between different degrees of freedom. Such a response is synergic when the monocaloric effects are both conventional (or both inverse) while it is non-synergic when one of the monocaloric effects is conventional and the other is inverse. Importantly, in all cases the multicaloric properties do not result from the simple addition of the monocaloric data because there is a contribution from the interplay between degrees of freedom (cross-coupling term). We will analyse the contribution of such a cross-coupling term to the multicaloric isothermal entropy change in both synergic and non-synergic materials. We will first discuss several model examples and then we will illustrate the realistic situation for prototype magnetostructural materials with synergic and non-synergic magnetocaloric and mechanocaloric effects [1-4].

**Acknowledgements**

Financial support from MCIN/AEI/10.13039/50110001103 (Spain) under Grants PID2020-113549RB-I00/AEI and IJC2020-043957-I, and from AGAUR (Catalonia) under Project SGR00328 is acknowledged. A.G acknowledges financial support from Universitat de Barcelona under a Margarita Salas fellowship, supported by the Spanish Ministry of Universitites with funding from the European Union under the NextGeneration EU program.

**References**

[1] E. Stern-Taulats et al. Phys. Rev. B **89** (2014) 214105.

[2] A. Gràcia-Condal et al. Appl. Phys. Rev. **7** (2020) 041406.

[3] A. Gràcia-Condal et al. Phys. Rev. Mater. 6 (2022) 084403.

[4] L. Mañosa et al. J. Phys. Energy (2023). To be published,

# Impact of Chemical Disorder on Magnetic and Vibrational Properties of Magnetocaloric Heusler alloys: First-Principles Insight

*Olga N. Miroshkina[1]*, Benedikt Eggert[1], Johanna Lill[1], Benedikt Beckmann[2], David Koch[2], Francesco Cugini[3,4], Simone Chicco[3], Fabio Orlandi[5], Giuseppe Allodi[3], Pietro Bonfà[3], Roberto De Renzi[3], Franca Albertini[4], Massimo Solzi[3,4], Franziska Scheibel[2], Andreas Taubel[2], Katharina Ollefs[1], Mojmir Šob[6,7], Martin Friák[7], Oliver Gutfleisch[2], Heiko Wende[1], Markus E. Gruner[1]*

[1] *Faculty of Physics and CENIDE, University of Duisburg-Essen, Lotharstr. 1, 47057 Duisburg, Germany*
[2] *Institute of Materials Science, Technical University of Darmstadt, Alarich-Weiss-Straße 16, 64287 Darmstadt, Germany*
[3] *SMFI Department, University of Parma, Parco Area delle Scienze 7/A I-43124 Parma, Italy*
[4] *IMEM-CNR Institute, Parco Area delle Scienze 37/A, I-43124 Parma, Italy*
[5] *ISIS Pulsed Neutron Facility, STFC, Chilton, Didcot, Oxfordshire OX11-0QX, UK*
[6] *Faculty of Science, Masaryk University, Kotlářská 2, CZ-611 37 Brno, Czech Republic*
[7] *Institute of Physics of Materials, Czech Academy of Sciences, Žižkova 22, CZ-616 62 Brno, Czech Republic*
* olga.miroshkina@uni-due.de

The development of high performance magnetocaloric materials is one of the major challenges for magnetic refrigeration technology, which is considered as an efficient and ecologically friendly alternative to gas refrigeration. The promising class of materials for magnetic cooling devices is multifunctional Heusler alloys. Consisting mostly of abundant elements, they possess many multifunctional properties, which depend strongly on chemical and anti-site disorder.

We investigate the vibrational properties and complex magnetic ordering mechanisms in magnetocaloric Ni-Mn-(In,Sn) Heusler alloys with different variants of transition-metal atoms distribution by means of density functional theory (DFT), which provides an accurate and efficient characterization of the relation between structure, magnetism, and electronic properties in these materials. Through the large-scale DFT calculations combined with NRIXS and Mössbauer spectroscopy, we were able to disentangle the vibrational contributions of the main group element in a near-stoichiometric $Ni_2MnSn$ [1]. We showed the evidence that the inversion of optical modes at Γ involving the displacement of Ni and the heavier Sn atoms, which was predicted previously for other Ni-Mn-based Heusler compounds, is also a characteristic property of $Ni_2MnSn$. Sn partial vibrational density of states (VDOS) are found to be sensitive to magnetic and chemical disorder on the transition metal site, which results in a distinctive redistribution and broadening of the Sn-VDOS.

Another example of the impact of the partial disorder can be found in Ni-Mn-In system, where Mn-excess atoms are introduced on Ni and main-group-element sublattices. Together with Sn-doping, this positional disorder leads to increasing magnetic inhomogeneity, which results in an effective magnetic decoupling of 4*a* and 4*b* sublattices and different magnetic ordering temperatures for them [2]. Increasing Sn content in Ni-Mn-(In,Sn) weakens ferromagnetic (FM) Ni-Mn exchange and strengthens antiferromagnetic (AFM) $Mn_{4a}$-$Mn_{4b}$ coupling due to the increasing valence electron concentration. Thus, we found that the revealed composition-dependent competition of the effective FM and AFM coupling between the sublattices can be tuned by the variation of the main-group element in combination with the modified 3*d* metal atoms distribution and be employed to control the magnetization of the transition metal sublattice. These results show the possibility of fine-tuning of Heusler materials properties via redistribution of the transition metal atoms caused by deviation from the stoichiometry and anti-site disorder, increasing the range of their potential applications.

## Acknowledgements

This work was funded by the DFG within TRR 270, Project-ID 405553726. Calculations were performed using the MagnitUDE HPC of the University of Duisburg-Essen and Lichtenberg II HPC of TU Darmstadt.

## References

[1] O. N. Miroshkina, B. Eggert, J. Lill, B. Beckmann, D. Koch, M. Y. Hu, T. Lojewski, S. Rauls, F. Scheibel, A. Taubel, M. Šob, K. Ollefs, O. Gutfleisch, H. Wende, M.E. Gruner, M. Friak, *Phys. Rev. B* **106** (2022) 214302.
[2] F. Cugini, S. Chicco, F. Orlandi, G. Allodi, P. Bonfá, V. Vezzoni, O. N. Miroshkina, M. E. Gruner, L. Righi, S. Fabbrici, F. Albertini, R. De Renzi, M. Solzi, *Phys. Rev. B* **105** (2022) 174434.

# Experiments and simulations for physics-informed machine learning to design neodymium-iron-boron permanent magnets

*Alexander Kovacs[1]*,Johann Fischbacher[1], Harald Oezelt[1],Qais Ali[1],Markus Gusenbauer[1], Masao Yano[2], Noritsugu Sakuma[2], Akihito Kinoshita[2], Tetsuya Shoji[2], Akira Kato[2], Yuan Hong[3], Stephane Grenier[3], Thibaut Devillers[3], Nora M. Dempsey[3], Thomas Schrefl[1]*

[1] *Christian Doppler Laboratory for magnet design through physics informed machine learning, Department of Integrated Sensor Systems Wr. Neustadt, University for Continuing Education, Krems, Austria*
[2] *Advanced Materials Engineering Division, Toyota Motor Corporation, Susono, Japan*
[3] *Université Grenoble Alpes, CNRS, Grenoble INP, Institut Néel, Grenoble, France*
*alexander.kovacs@donau-uni.ac.at

Rare-earth elements such as neodymium, terbium, and dysprosium are critical to the performance of permanent magnets which are embedded into hybrid and electric traction motors or wind turbines. To address the supply risk of those elements, we use machine-learning techniques to search for magnetic materials with less neodymium content and no terbium or dysprosium content [1]. During this search it is intended that the performance of the magnet will be preserved.
We developed machine-learning methods to aid materials design by integrating physical models to bridge the length scale gap from atomistic to micrometer-sized granular microstructures in 2-14-1 permanent magnets. We use data assimilation to combine data from experiments and simulations to create machine-learning models capable of optimizing the chemical composition and microstructure of the magnet. We demonstrate (i) a model predicting temperature dependent intrinsic material properties based on their chemical composition and (ii) another model predicting the coercive field dependence based on measured x-ray diffraction patterns.
We apply partial-least squares regression, a well understood method similar to principal components analysis, that aid in understanding and interpreting the outcomes of machine learning predictions [2,3]. A variable importance factor [4] is computed to demonstrate how specific design variables influence the magnetic properties.
High-throughput x-ray diffraction measurements on compositionally graded sputtered films were analyzed with partial least squares regression and the most important peaks for coercivity highlighted. We demonstrate how high-performance Nd-lean magnets can be realized and characterized using machine learning models.

## Acknowledgements

The financial support by the Austrian Federal Ministry of Labour and Economy, the National Foundation for Research, Technology and Development and the Christian Doppler Research Association is gratefully acknowledged. Ab initio data are provided by the National Institute of Advanced Industrial Science and Technology and the Institute for Solid State Physics, the University of Tokyo under “Program for Promoting Researches on the Supercomputer Fugaku”.

## References

[1] Kovacs A, Fischbacher J, Oezelt H, Kornell A, Ali Q, Gusenbauer M, Yano M, Sakuma N, Kinoshita A, Shoji T, Kato A, Hong Y, Grenier S, Devillers T, Dempsey NM, Fukushima T, Akai H, Kawashima N, Miyake T and Schrefl T (2023) Physics-informed machine learning combining experiment and simulation for the design of neodymium-iron-boron permanent magnets with reduced critical-elements content. Front. Mater. 9:1094055. doi: 10.3389/fmats.2022.1094055 (Times New Roman 10 points, left-justified)
[2] Geladi, P., and Kowalski, B. R. (1986). Partial least-squares regression: A tutorial. Anal. Chim. Acta 185, 1–17. doi:10.1016/0003-2670(86)80028-9
[3] Eriksson, L., Trygg, J., and Wold, S. (2014). A chemometrics toolbox based on projections and latent variables. J. Chemom. 28, 332–346. doi:10.1002/cem.2581
[4] Tobias, R. D., et al. (1995). “An introduction to partial least squares regression,” in Proceedings of the twentieth annual SAS users group international conference (Cary, NC, USA): SAS Institute Inc.), 1250–1257.

# SYMPOSIUM 01.
## MAGNETIC MATERIALS FOR ENERGY APPLICATIONS.
## S1. ORAL PRESENTATIONS

# Processing Conditions for High Coercivity in (Sm,Zr)(Fe,Co,Ti)$_{12}$ Particles Prepared via Calcium Reduction

*Chaoya Han[1], A.M. Gabay[2]*, Chaoying Ni[1], G.C. Hadjipanayis[2,3]*

[1] *University of Delaware, 201 DuPont Hall, Newark, Delaware, USA*
[2] *University of Delaware, 217 Sharp Lab, Newark, Delaware, USA*
[3] *Northeastern University, 360 Huntington Avenue, Boston, Massachusetts, USA*
*gabay@udel.edu

Monocrystalline $Sm_{1-x}Zr_x(Fe,Co)_{12-y}Ti_y$ particles of the $ThMn_{12}$ crystal structure prepared via calcium reduction of mechanically activated metal oxides at 1200 °C [1] had been recently shown to develop a record-high room-temperature coercivity of 16.3 kOe [2]. Curiously, these high coercivity values are only attainable if the $ThMn_{12}$-type compound (1:12) forms in a Sm-depleted environment following a primary ($\alpha$-Fe) solid solution. If, on the other hand, the same 1:12 compound is preceded by a Sm-enriched phase of the $TbCu_7$ type (or a high-temperature phase of the $Th_2Ni_{17}$ type [3]), the coercivity in the particles of a similar size reaches only half of the above value. In this work, products of a partial reduction synthesis, collected after 15–50% of the time required for the reduction to complete, were subjected to TEM characterization to better understand the difference in the 1:12 phase obtained via slightly different processing routes that ultimately led to the same particle morphology but large differences in coercivity. The 1:12 phase formation in the Sm-depleted environment has been confirmed to occur at the expense of the primary ($\alpha$-Fe) phase [see Figure (a)]. In contrast, the 1:12 phase forming alongside the Sm-enriched 1:7/2:17 hexagonal phase initially emerges as very Ti-rich. Its evolution in this case involves major changes in the chemical composition that may result in an imperfect crystal structure. A TEM image of particles annealed for 8 seconds indicates that the formation of 1:12 phases is preceded by the 1:7/2:17 phase [see Figure (b)].

## Acknowledgement

This work was supported by the U.S. Department of Energy Office of Science, Award No. DE SC0022168.

## References

[1] A.M. Gabay, G.C. Hadjipanayis, *Scr. Mater.* 196 (2021) 113760.
[2] A.M. Gabay, C. Han, B.T. Lejeune, K.W.S. Jinn, C.Y. Ni, L.H. Lewis, G.C. Hadjipanayis, "KOA-01. Formation of $ThMn_{12}$-type phase and conditions for high coercivity in (Sm,Zr)(Fe,Co,Ti)$_{12}$ alloys," *MMM 2022 Conference, Book of Abstracts*, p. 341.
[3] G.V. Ivanova, G.M. Makarova, Ye.V. Shcherbakova, Ye.V. Belozerov, *J. Alloys Compd.* 224 (1995) 29.

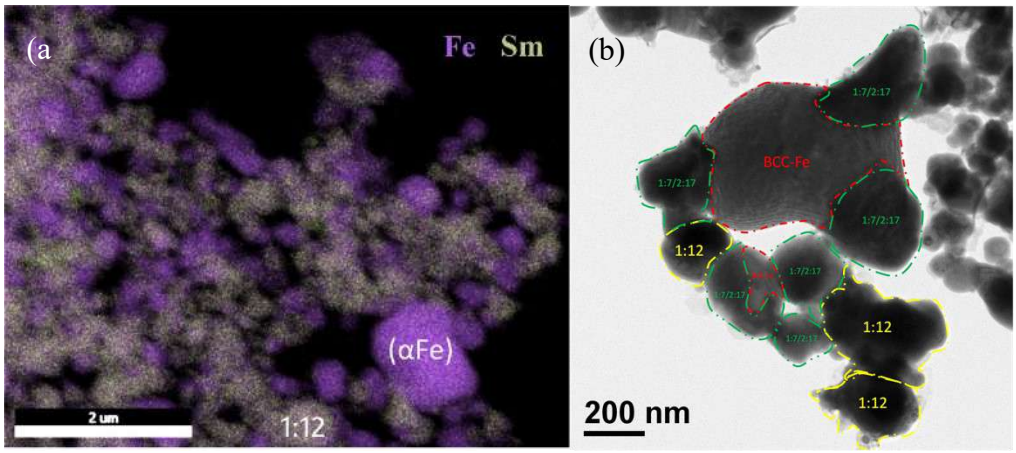



Figure: TEM characterization of Sm-Zr-Fe-Co-Ti nanoparticles produced via incomplete reduction diffusion: (a) superimposed Fe and Sm element maps of particles reduced for 30 s in Sm-depleted environment – showing 1:12 particles growing at the expense of (α-Fe) particles; (b) TEM image of particles reduced for 8 s in Sm-rich environment – showing coexisting particles of $\alpha$-Fe, 1:12 and 1:7/2:17 phases.

# Nanocrystalline $SmFe_{12}$-based magnets prepared by high pressure hydrogen milling

*I. Dirba[1]*, A. Aubert[1], H. Sepehri-Amin[2], K. Hono[2] and O. Gutfleisch[1]*

[1] *Functional Materials, Institute of Materials Science, Technical University of Darmstadt, 64287 Darmstadt, Germany*

[2] *Elements Strategy Initiative Center for Magnetic Materials, National Institute for Materials Science, Tsukuba 305-0047, Japan*

*imants.dirba@tu-darmstadt.de

$SmFe_{12}$-based compounds possess promising intrinsic magnetic properties, even superior to those of $Nd_2Fe_{14}B$ phase at elevated temperatures. However, the challenge is to demonstrate that those intrinsic properties can be further developed into practically relevant extrinsic ones by proper processing. Our previous work has shown that hydrogenation disproportionation desorption recombination process can be employed for preparation of ultrafine-grained $SmFe_{12}$-based alloys [1]. Here we demonstrate for the first time that milling under high pressure hydrogen (HPRM in evico magnetics high pressure vial) can be used to obtain even further grain size reduction via reversible hydrogen absorption-desorption process.

The starting alloys were $SmFe_{11}Ti$ doped with Co, Ga, V and Cu aimed to enhance both the intrinsic and extrinsic magnet properties. Milling was carried out at 450 rpm under a hydrogen pressure of 100 bar for 5 h with 10 mm balls and ball-to-powder ratio of 10:1. This results in disproportionation of the $SmFe_{12}$-based phase into $SmH_2$, $Fe_2Ti$ and α-Fe (Fig. 1a 'as HPRM'). Hydrogen desorption-recombination (DR) process was carried out at 700 °C - 1000 °C temperatures for 30 min under continuous vacuum pumping. Depending on the temperature, recombination leads to formation of a phase with $ThMn_{12}$-type or $TbCu_7$-type crystal structure. Using subsequent nitriding treatment, it was attempted to obtain strong uniaxial anisotropy. However, Fig. 1b shows that in the case of $Sm(Fe_{0.8}Co_{0.2})_{10.5}Ga_{0.2}Cu_{0.3}Ti$ composition, nitriding results in formation of α-Fe as well as $Sm_2O_3$ and thus low coercivity. Further nitriding process optimization is necessary to avoid soft magnetic phases as well as oxidation of the fine powders and thus obtain higher coercivities.

## Acknowledgements

We acknowledge funding by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation), Project ID No. 405553726-TRR 270.

## References

[1] I. Dirba, H. Sepehri-Amin, T. Ohkubo, K. Hono, Acta Mater. **165**, 373–380 (2019).

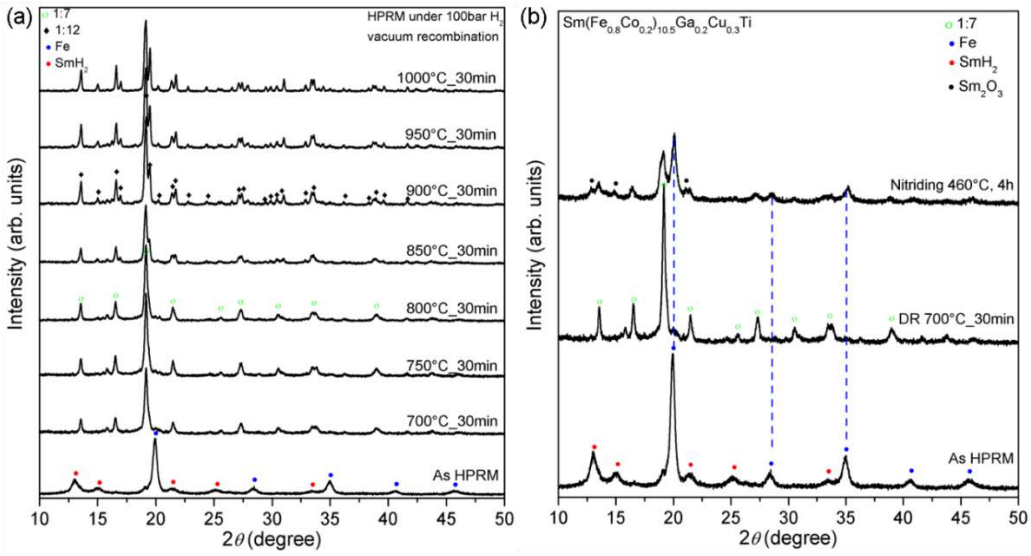



Figure 1: (a) XRD results for $Sm(Fe_{0.8}Co_{0.2})_{10.5}Ga_{0.2}Cu_{0.3}Ti$ composition after HPRM and hydrogen desorption treatments (a) and subsequent nitriding (b).

# Magnetic Properties of the Ge, Re and Cr Doped $Fe_5SiB_2$ Compound

*M. Casadei[1,2*], M. Alsayed[1], L. Tincani[1], G. Trevisi[2], S. Fabbrici[2], R. Cabassi[2], G. Allodi[3], C.D. Fernández[2], S. Sanna[1], F. Albertini[2]*

[1]*Department of Physics, University of Bologna, viale Berti Pichat 6/2, Bologna, Italy*
[2] *IMEM-CNR, Via delle Scienze 37, Parma, Italy*
[3] *Department of Mathematical, Physical and Computer Science, University of Parma, Parco Area delle Scienze 7/A, Parma, Italy*
*matteo.casadei34@unibo.it

The permanent magnet market, at the current state, is essentially split into two types of materials. On one end we have cheap but low performing ferrite magnets, on the other end we have the more expensive and better performing rare earth (RE) magnets. RE magnets play a key role in the transition from fossil fuels towards renewable energy alternatives, since they are largely used in electric motors, generators and energy converters. However, more concerns have been risen regarding their supply risks, volatile prices and environmental impact. One way to reduce the demand for critical materials is to develop a new class of cheap magnets, called "gap magnets" [1], possessing intermediate properties, that could replace RE magnets in those application that do not require extreme performances.

Since iron is one of the most abundant elements on Earth, iron-based magnets are desirable for this new class of materials. $Fe_5SiB_2$ is a noticeable candidate due to its high saturation magnetization and Curie temperature. However, its magnetic anisotropy energy is too low for practical applications. It has been reported, both experimentally and theoretically, that the anisotropy energy can be tuned by chemical substitution of Si or Fe with other elements [2,3]. We synthesized and characterized polycrystalline samples of $Fe_5SiB_2$ with different levels of Ge, Re and Cr substitutions. Here we report on the main magnetic properties of these compounds, such as Curie temperature, saturation magnetization and anisotropy field as measured by magnetometry, Singular Point Detection (SPD) and Nuclear Magnetic resonance experiments. The effect of chemical substitutions on the magnetic properties and their dependence on doping levels will be discussed. In particular, we show that the temperature dependence of the anisotropy field measured by SPD from RT down to 77 K indicates that this compound does not have an easy axis of magnetization at high temperature, as commonly reported in literature [4], but rather an easy plane or easy cone configuration.

## References

[1] Coey, J. M. D. "Permanent magnets: Plugging the gap." *Scripta Materialia* 67.6 (2012): 524-529.A. Gapito, I. Saias, *Phys. Rev. X* **7** (2017) 031037.
[2] Clulow, Rebecca, et al. "Magnetic and structural properties of the Fe5Si1-xGexB2 system." *Journal of Solid State Chemistry* 316 (2022): 123576.
[3] McGuire, Michael A., and David S. Parker. "Magnetic and structural properties of ferromagnetic Fe5PB2 and Fe5SiB2 and effects of Co and Mn substitutions." *Journal of Applied Physics* 118.16 (2015): 163903.
[4] Wäppling, R., et al. "Magnetic properties of Fe5SiB2 and related compounds." *Le Journal de Physique Colloques* 37.C6 (1976): C6-591.

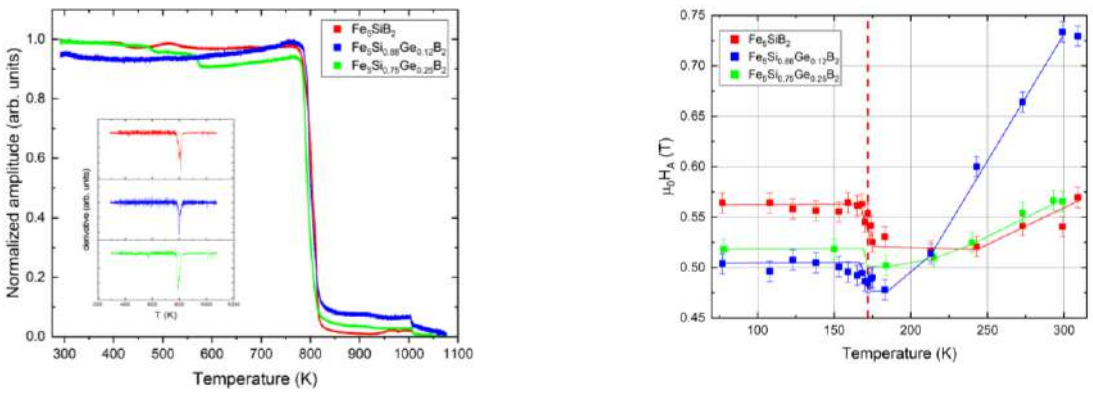



Figure: Thermomagnetic (left) and SPD (right) measurements of the $Fe_5Si_{1-x}Ge_xB_2$ compound.

# Kinetics of the Magnetoelastic Phase Transition in Fe49Rh51

*A. Beleza[1]*, M. L. Arreguín-Hernández[2], J. L. Sánchez Llamazares[2,3], P. Álvarez Alonso[2], J. P. Araújo[1], J. H. Belo[1]*

[1] *IFIMUP, Departamento de Física e Astronomia, Faculdade de Ciências, Universidade do Porto, Rua do Campo Alegre s/n, 4169-007 Porto, Portugal*

[2] *Instituto Potosino de Investigación Científica y Tecnológica A.C., Camino a la Presa San José 2055, Col. Lomas 4a, San Luis Potosí, SLP 78216, México*

[3] *Departamento de Física, Universidad de Oviedo, C/Federico García Lorca 18, 33007 Oviedo, Spain*

*up201804896@up.pt

Material optimisation for environmentally friendly caloric applications, such as magnetic refrigeration and thermomagnetic energy harvesting, may be brought about by the use of magnetocaloric materials presenting magnetostructural first-order phase transitions, FOPTs. While these materials exhibit large entropy and adiabatic temperature changes linked to their magnetic-field driven FOPT, in some cases, such as in La(Fe,Si)13 alloys, magnetization may present a significant relaxation with time at temperatures close to the phase transition after an applied field is paused [1]. We have conducted a thorough characterization of this phenomenon in a bulk Fe49Rh51 alloy by designing an experiment that carefully isolates the relevant parameters involved, such as temperature, magnetic field intensity and magnetic field sweep rate. To this end, for a set of temperatures below the AFM to FM transition, a magnetic field was applied at intensities close to the critical field (onset of the AFM-FM transition), and the field sweep rate to reach a given value varied. Then, after pausing the field at a certain value, the magnetization's relaxation was measured as a function of time.

As shown in the Figure below, the relaxation time of Fe49Rh51's magnetization in the AFM state was observed to last over 1000 seconds when the field is driven at 70 mT/s. In contrast with the behaviour previously observed in La(FeSi)13, Fe49Rh51 exhibits an increase in the magnetization's relaxation time as the magnetic field sweep rate increases. Lastly, we also report on the magnetization's time derivative immediately after the magnetic field is halted. It is shown that it exhibits a linear relation with the applied field for a range of values past but near the critical field, as well as a sweep rate dependent saturation speed for intensities surpassing this range.

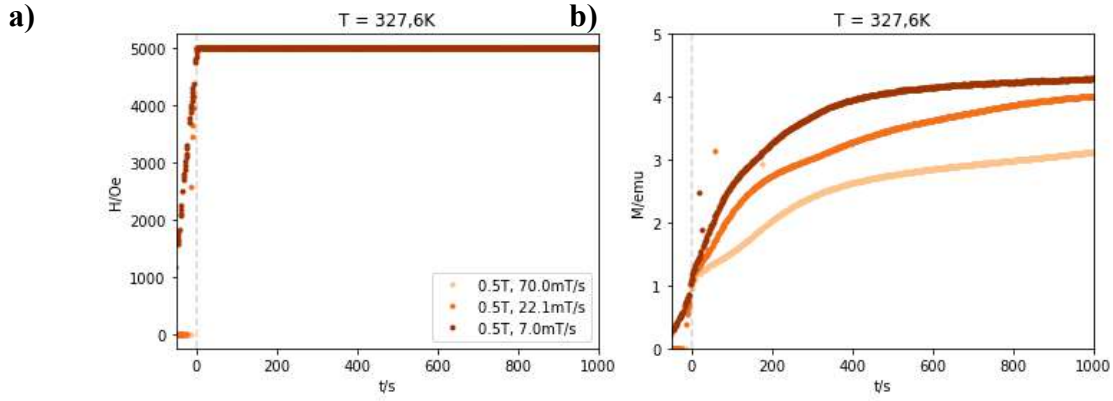



Figure: **a)** Profile of the magnetic field applied isothermally to an Fe49Rh51 sample in the AFM state, driven at different magnetic field sweep rates. **b)** The resulting relaxation of magnetization over time. As the magnetic field sweep rate is increased, an increase in relaxation time is observed.

**Acknowledgements:** We would like to acknowledge FCT, for the funded projects PTDC/EME-TED/3099/2020 (IFIMUP), UIDP/04968/2020 - Programático, UIDB/04968/2020, NECL - NORTE-010145-FEDER- 022096 and CERN/FISTEC/0003/2019.

## References

[1] Lovell, E., Pereira, A. M., Caplin, A. D., Lyubina, J. & Cohen, L. F. (2015). Dynamics of the First-Order Metamagnetic Transition in Magnetocaloric La(Fe,Si)13: Reducing Hysteresis. Advanced Energy Materials, 5(6), 1401639. doi:10.1002/aenm.201401639

# Fast and giant modulation of magnetism in 3d-4f magnets

*X.L.Ye[1,2*], Zhibo Zhao[2], H. Singh[1],,Fengkai Yan[3], H.B Zhang[1], H. Geßwein[1], M.R. Chellali[2], R. Witte[2], A. Molinari[2], K. Skokov[1], D. Raabe[3], B. Gault[3] , R. Kruk[2], O. Gutfeisch[1], H. Hahn 2*

[1] *Department of Material Science, Technical University Darmstadt, 64287 Darmstadt, Germany*

[2]*Institute of Nanotechnology, Karlsruhe Institute of Technology, 76344 Eggenstein-Leopoldshafen, Germany*

[3] *Max-Planck-Institut für Eisenforschung GmbH (MPIE), 40237 Düsseldorf, Germany*

xinglong.ye@tu-darmstadt.de

Controlling magnetism and magnetic properties by applying small voltages have been vigorously pursued in magneto-electric actuation, spintronics, information processing and data storage due to its ultralow power consumption. However, in ferromagnetic metals and alloys, which are core materials in important technologies, the control of magnetic properties has been limited to the scale of atomic layers in surface/interface due to strong electric field screening and the change of magnetism are usually small [[i]]. Recently, we proposed the concept of controlling magnetism through electrochemically-driven insertion/extraction of hydrogen atoms into/from interstitial sites of metal structure. The uniqueness of this concept lies in controlling the absorption and desorption of electrically neutral hydrogen atoms by means of electrochemical potentials. We showed that by applying voltages of 1 V, the magnetocrystalline anisotropy and coercivity of Sm-Co permanent magnets in micrometer-sized powders can be reversibly altered by more than 1 T [[ii],[iii]]. For the first time, voltage-assisted and -controlled magnetization reversal has been achieved at room temperature in permanent magnets. However, there are still challenges for this approach. Firstly, the aqueous electrolyte that provides hydrogen atoms will be difficult to be integrated into devices. Secondly, the dynamics of the tuning process and the mechanism behind the huge change of magnetic properties remain unclear. Targeting these challenges, in this talk we have dispensed with the use of aqueous electrolyte and developed the solid-state electrochemical system to realize the voltage-control of hydrogen insertion/extraction in a completely solid-state system. This enabled us to reversibly manipulate the coercivity by a magnitude of more than 2 T and the switching time can be reduced to milliseconds. Our work may take the voltage-control of magnetism truly beyond the field-effect devices, which can enable the development of novel magnetoelectric devices

**References:**

[1] F. Matsukura, Y. Tokura, H. Ohno, Nature Nanotech.10, 209-220 (2015).

[2] X.L.Ye, H. Singh, H.B Zhang, H. Geßwein, M.R. Chellali, R. Witte, A. Molinari, K. Skokov, O. Gutfeisch, H. Hahn, R. Kruk. Nature Comm. 11, 4849 (2020).

[3] X.L.Ye, F.K. Yan, L. Schaefer, D. Wang, H. Geßwein, W. Wang, M.R.Chellali, L. Stephenson, K. Skokov, O. Gutfleisch, D. Raabe, H. Hahn, B. Gault, R. Kruk. Adv. Mater. 33, 2006853 (2021).

.

# Cu-doped Sm-Co Alloy Nanopowder Prepared by Induction Thermal Plasma Process

*Kwangjae Park[1], Yusuke Hirayama[1*], and Jian Wang[1]*

[1] *National Institute of Advanced Industrial Science and Technology (AIST), Nagoya, Japan*

*hirayama.yusuke@aist.go.jp

For developing high-performance permanent magnets, particle size refinement is the one way to exploit the potential of the compounds. The mechanical milling process and chemical synthesis method are widely used to prepare fine rare-earth (RE) alloy particles [1, 2]. Among the various methods, the induction thermal plasma (ITP) process [3], which is a gas evaporation process, is a unique method to prepare the RE alloy nanopowder with sizes smaller than 300 nm.

In the previous study, the Sm-Co alloy nanopowder was successfully synthesized by the ITP process [4]. The anisotropic magnetic behavior and the particle formation mechanism of the Sm-Co nanopowder were demonstrated by the experimental results and numerical analysis. Using this Sm-Co nanopowder as a starting material, the isotropic nanocrystalline Sm-Co bulk magnet with a giant coercivity of 5.2 T and remarkable thermal stability was successfully consolidated [5]. The outstanding magnetic properties resulted from the well-organized microstructure with better crystallinity obtained by using the Sm-Co nanopowder. However, the prepared Sm-Co "nanopowder" by the ITP process consists of only 36.7 wt% of the $SmCo_5$ phase. In order to increase the content of the $SmCo_5$ phase and obtain the magnetically harder nanopowder, Cu doping into $SmCo_5$ is effective way since the solidus line of the $SmCo_5$ phase is shifted to a lower temperature region by doping Cu into the Sm-Co alloy [6]. Therefore, in this work, the third element Cu was doped to prepare the Sm-Co nanopowder as close as single-phase by decreasing a composition distribution.

The Sm-Co-Cu nanopowder was prepared by the ITP process using the mixed raw powders in the ratio of Sm:Co:Cu=1:4.5:0.5 at%. The X-ray diffraction (XRD) profile of the Sm-Co-Cu nanopowder indicates that almost only the $SmCo_5$ phase was detected as an alloy phase. From Rietveld refinement, the Sm-Co-Cu nanopowders yielded a $SmCo_5$ phase content of ~60 wt%, about 1.6 times that of the Sm-Co nanopowder synthesized without Cu doping. Scanning transmission electron microscopic (STEM) observation clearly showed the formation of the Sm-Co-Cu alloy particles. The coercivity of the Sm-Co-Cu nanopowder was measured as 1.9 T at 300 K. The Sm-Co-Cu nanopowder magnetic aligned by an external magnetic field of 2 T displayed sufficient crystallographic orientation. In addition, the Sm-Co-Cu nanopowder has better anisotropic magnetic behavior compared to that of the Sm-Co nanopowder without Cu doping. Based on the results, we suggested that the ITP process has a high potential to prepare not only binary but also ternary RE alloy nanopowder for developing high-performance permanent magnets.

## Acknowledgements

This work is supported by JSPS International Research Fellow. The authors would like to thank Kazumi Hayakawa for TEM sample preparation.

## References

[1] B. Cui, A. Gabay, W. Li, M. Marinescu, J. Liu and G. Hadjipanayis, *J. Appl. Phys.* **107** (2010) 09A721.
[2] Y. Hirayama, A. Panda, T. Ohkubo and K. Hono, *Scr. Mater.* **120** (2016) 27-30.
[3] M. Shigeta and A. B. Murphy, *J. Phys. D: Appl. Phys.* **44** (2011) 174025.
[4] K. Park, Y. Hirayama, M. Shigeta, Z. Liu, M. Kobashi and K. Takagi, *J. Alloys Compd.* **882** (2021) 160633.
[5] K. Park, Y. Hirayama, J. Wang and M. Kobashi, *Scr. Mater.* **218** (2022) 114847.
[6] A. Perry, *J. Less-Common Met.* **51** (1977) 153-162,

# Improved saturation magnetization in Ce-doped $Sm_2Fe_{17}N_3$ powder

*Akihide Hosokawa[1]*, Wataru Yamaguchi[1], Yusuke Hirayama[1], Kimihiro Ozaki[1]*

[1] *National Institute of Advanced Industrial Science and Technology, 4-205 Sakurazaka Moriyama-ku, Nagoya, Japan*

*aki.hosokawa@aist.go.jp

$Sm_2Fe_{17}N_3$ compound is a promising candidate material that possesses high saturation magnetization, high Curie temperature, and high anisotoropy field being a potential competitor for Nd-Fe-B magnets at elevated temperatures. Although it has been said that this compound is extremely difficult to consolidate into bulk magnets, we recently reported that high-coercivity $Sm_2Fe_{17}N_3$ sintered magnets can be produced by development of what is called low-oxygen powder metallurgical processes [1]. Quite recently, Toyota motor corporation reported a possibility that saturation magnetization and anisotropy field can be improved by doping of Ce, La and Co to $Sm_2Fe_{17}N_3$ compound [2]. However, the saturation magnetization for Ce-free $Sm_2Fe_{17}N_3$ is 1.51 T for $Sm_2Fe_{17}N_3$ in the patent embodiment which is not as high as the already reported value (1.57 T) in the liretrature. This is probably because the nitridation in the patent embodiments was carried out by nitrogen gas. It is quite possible that nitridation is imperfect in that work since the nitriding ability of nitrogen gas is inferior to that of ammonia. In this work, Ce-doped $Sm_2Fe_{17}N_3$ powder was nitrided by ammonia gas so that the nitrogen concentration is precicsely controlled to exploit the full potential of the intrinsic properties of the magnetic powder truly has.

Raw coarse Ce-doped $Sm_2Fe_{17}$ powder was supplied from Toyota motor corporation. A part of the powder was nitrided by mixture gas of ammonia and hydrogen to produce $(Sm_{1-x}Ce_x)_2Fe_{17}N_3$ powder. The other part of the powder was pulverized by jet-milling or ball-milling to reduce the particle size down to 1.3 ~ 3.0 μm. A part of the pulverized powders were subjected to magnetic alignment and direct current sintering. The obtained pulverized fine powders and sintered magnets were subjected to the characterizations of laser-diffraction particle size analyzer, oxygen-nitrogen analysis, scanning electron microscope (SEM), X-ray diffractometry (XRD), vibrating sample magnetometer (VSM) equipped with superconductivity magnets with maximum field of 90 kOe.

In both the properly nitrided coarse powder and fine powders, it was confirmed that $Sm_2Fe_{17}N_3$ structure was obtained by X-ray diffractometry. VSM measurements also revealed that the $(Sm_{0.8}Ce_{0.2})_2Fe_{17}N_3$ powder exhibits the saturation magnetizatoin at room temperature is 169.9 emu/g, being superior to that of the $Sm_2Fe_{17}N_3$ compound (163 emu/g). Among the several pulverized powders, one of the powders exhibited $H_c$ = 8.4 kOe and $(BH)_{max}$ = 34 MGOe. However, it was found that the improved saturation magnetization by Ce doping cannot be kept high after pulverization processes. In fact, the magnetization degradatiaon became more serious when the pulverized powder was sintered. Consequently, the $(BH)_{max}$ of the sintered magnet exhibited 18 MGOe, which is not as high as the ones for dopant-free $Sm_2Fe_{17}N_3$ magnets previously produced by our low oxygen powder metallurgical processes [3]. Although the sequences of nitridation and pulverization was changed to see if possible improvement of performance, it was found that the process route wherein pre-nitrided coarse powder was pulverized by jet-milling was able to produce the powder having the highest $(BH)_{max}$ and coercivity.

## Acknowledgements

The authors appreciate Toyota Motor Corporation for supplying raw powder. This presentation is based on results obtained from the Future Pioneering Program "Development of magnetic material technology for high-efficiency motors" commissioned by the New Energy and Industrial Technology Development Organization (NEDO).

## References

[1] K. Takagi, Y. Hirayama, S. Okada, W. Yamaguchi, K. Ozaki, *Science and Technology of Advanced Materials* **1** (2021) 150
[2] Japanese patent, JP 2022053187
[3] K. Takagi, R. Soda, M. Jinno, W. Yamaguchi, *Journal of Magnetism and Magnetic Materials* **506** (2020) 166811

# Extending the Application of Exponent $n$ Criteria to Evaluate Ambiguities in Complicated Magnetocaloric Materials

*Jia Yan Law[1]*, Luis M. Moreno-Ramírez[1], Victorino Franco[1]*

[1] *Department of Condensed Matter Physics, ICMS-CSIC, University of Seville, P.O. Box 1065, 41080 Seville, Spain*

*jylaw@us.es

Since the discovery of the fingerprint [1] of first-order thermomagnetic phase transitions (FOPT) in the magnetocaloric response of materials, the so-called exponent $n$ quantitative criterion, the community has been applying it to evaluate their newly developed materials and performance [2]. This criterion is not limited to any theoretical model, does not require a fitting procedure, enhancing its versatility and, at the same time, eliminated the ambiguities encountered by the conventional methods when evaluating the order of phase transition (such as the presence of thermal hysteresis, universal curve scaling, and Banerjee's criterion). Aside from addressing these limitations with the FOPT fingerprint, i.e. an overshoot of $n$>2 near the transition, exponent $n$ also achieves another breakthrough: identifying the critical point where the first-order crossovers to second-order phase transition (SOPT). This is a crucial regime for many high-performance magnetocaloric materials as it combines the advantages of both FOPT and SOPT, i.e., large MCE without hysteresis. A different viewpoint on this advantage might contend that identifying the location of the borderline can be difficult.

In this talk, we present the exponent $n$ criteria to address the ambiguities faced in studying high-performance La(Fe,Mn,Si)$_{13}$H and Mn-Fe-P-Si magnetocaloric materials undergoing magnetoelastic transitions: (i) exhibiting very low hysteresis [3] and (ii) with the presence of impurity magnetic phases near the FOPT [4], respectively. For case (i), the least hysteretic sample has a strong resemblance to SOPT characteristics but demonstrates a FOPT character according to the $n$ criteria [1,5]: FOPT fingerprint of $n$>2 overshoot and $n$ at the transition temperature, $n_{\mathrm{transition}}$, below 0.4 (equals to 0.37). This slight difference in the $n_{\mathrm{transition}}$ values denote close proximity to the critical point, which also explains the least hysteresis observed in the series. These claims will be supported by complementary data. In case (ii), as $n = 1$ at ferromagnetic state, the presence of magnetic impurities near the temperature range where FOPT occurs affects the overshoot associated to the FOPT: the existence of an overshoot remains, but it reaches a different maximum value, as demonstrated with numerical simulations.

**Acknowledgements**

Work supported by Grant funded by PID2019-105720RBI00/AEI/10.13039/501100011033, Consejería de Economía, Conocimiento, Empresas y Universidad de la Junta de Andalucía (grant P18-RT-746), and Air Force Office of Scientific Research (FA8655-21-1-7044). JYL acknowledges EMERGIA fellowship from Junta de Andalucía (ref. EMC21_00418).

**References**

[1] J.Y. Law et al., *Nat. Commun.* **9** (2018) 2680.

[2] J.Y. Law et al., *J. Appl. Phys.* **133** (2023) 040903.

[3] L.M. Moreno-Ramírez et al., *Under review* (2023).

[4] V. Franco et al., *In preparation* (2023).

[5] V. Franco et al., J. Phys. D: Appl. Phys. **50** (2017) 414004.

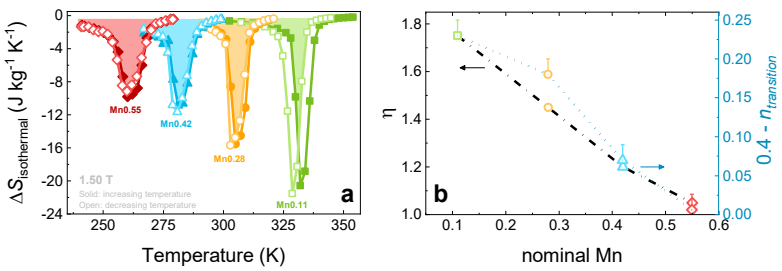



Figure: (a) The least hysteretic sample among the La(Fe,Mn,Si)$_{13}$H series show strong resemblance to SOPT characteristics. (b) The $n$ criteria reveals its FOPT character andproximity to the critical point.

# Enhanced Spin Seebeck Effect via Voltage-Controlled Oxygen Manipulation

Jeong-Mok Kim[1†], Seok-Jong Kim[1,2†], **Min-Gu Kang[1,3†]**, Jong-Guk Choi[1], Soogil Lee[1], Jaehyeon Park[2], Cao Van Phuoc[4], Kyoung-Whan Kim[5], Kab-Jin Kim[2], Jong-Ryul Jeong[4], Kyung-Jin Lee[2★] and Byong-Guk Park[1★]

[1] *Department of Materials Science and Engineering, KAIST, Daejeon 34141, Korea,*[2] *Department of Physics, KAIST, Daejeon 34141, Korea,*[3] *Department of Materials, ETH Zurich, Zurich 8093, Switzerland,* [4] *Department of Materials Science and Engineering, Chungnam National University, Daejeon, 34134, Korea,*[5] *Center for Spintronics, Korea Institute of Science and Technology, Seoul 02792, Korea*

*† These authors equally contributed to this work*

★ **Correspondence to:** bgpark@kaist.ac.kr (B.-G.P.) & kjlee@kaist.ac.kr (K.-J.L.)

Spin Seebeck effect (SSE) [1] is referring the thermoelectric conversion via a magnon current in magnetic systems, where a thermoelectric voltage is generated in a direction perpendicular to the temperature gradient. SSE has potential to offer efficient thermoelectric devices because the transverse geometry of SSE enables to utilize waste heat from a large-area source by greatly simplifying the device structure. However, for widespread application, the thermoelectric conversion efficiency of SSE needs to be further improved. In this work, we show that the SSE substantially enhances by oxidizing a ferromagnet in normal metal/ferromagnet/oxide structures. In W/CoFeB/$AlO_x$ structures, voltage-controlled interfacial oxidation of CoFeB modifies the SSE, resulting in the enhancement of thermoelectric signal by an order of magnitude. We describe a mechanism for the enhancement that results from a reduced exchange interaction of the oxidized region of ferromagnet, which in turn increases a temperature difference between magnons in the ferromagnet and electrons in the normal metal and/or a gradient of magnon chemical potential in the ferromagnet. Our results will invigorate research for thermoelectric conversion by suggesting a promising way of improving the SSE efficiency.

## Acknowledgements

This work was supported by support from Samsung Research Funding Center of Samsung Electronics under Project Number SRFC-MA1802-01 and the National Research Foundation of Korea (NRF) (NRF-2022R1A4A1031349). K.-J.L. was supported by the NRF (NRF-2020R1A2C3013302). K.-W.K. was supported by the KIST institutional program.

## References

[1] Uchida, K. et al. Observation of the spin Seebeck effect. *Nature* **455**, 778–781 (2008).

# Strong converse magnetoelectric coupling in FeGa/PMN-PT epitaxial thin films

*Adrián Begué[1,2], Miguel Ciria[1]**

[1] *Instituto de Nanociencia y Materiales de Aragón (INMA), CSIC-Universidad de Zaragoza, Zaragoza 50009, España.*

[2] *Universidad Complutense de Madrid (UCM), dpto. de Física de Materiales, Madrid 28071, España*

*miguel.ciria@csic.es

Magnetoelectric (ME) heterostructured materials with strain-induced coupling between ferroelectric and magnetostrictive components provides the larger converse ME coupling for low energy consumption applications [1]. In this contribution we present the ME properties of crystalline FeGa films grown on PMN-PT crystals with (110) and (001) cuts. FeGa alloys, around the galfenol composition, are interesting because of their large magnetoelastic coupling, with low magnetic anisotropy values which tend to conceal strain-induced effects. Crystalline layers are obtained at the growing temperature of 150 ºC using a MgO buffer layer. FeGa films on the (001) crystal surface grown on the (001) plane and display a large inverse ME coefficient above $10^{-5}$ s/m [2], with the easy direction switching between in-plane [100] and [010] directions by the application of an external electric field, which activates a uniaxial anisotropy (see Figure 1). The films deposited on (011)PMN-PT display the (112) surface and an intrinsic uniaxial anisotropy where the sign depends on the composition of the layer through the sign of the magnetoelastic stress $B_2$, and the residual strain. The effect of the induced strain is weak and appears as minor modifications of the coercive field or the remanent magnetization.

## Acknowledgements

This work was supported by Spanish MICINN (Grants No. TED2021-131064B-I00 and PID2021-124734OB-C21). A.B. would like to acknowledge the funding received from the Ministry of Science and Innovation for the Ph.D. contract BES-2016-076482, as well as the funding received from the Ministry of Universities and the European Union-Next Generation for the Margarita Salas fellowship.

## References

[1] Jia-Mian Hu and Ce-Wen Nan, *APL Mater.* **7** (2019) 080905.

[2] A. Begué and M. Ciria, *ACS Applied Materials & Interfaces* **13** (2021) 6778-6784.

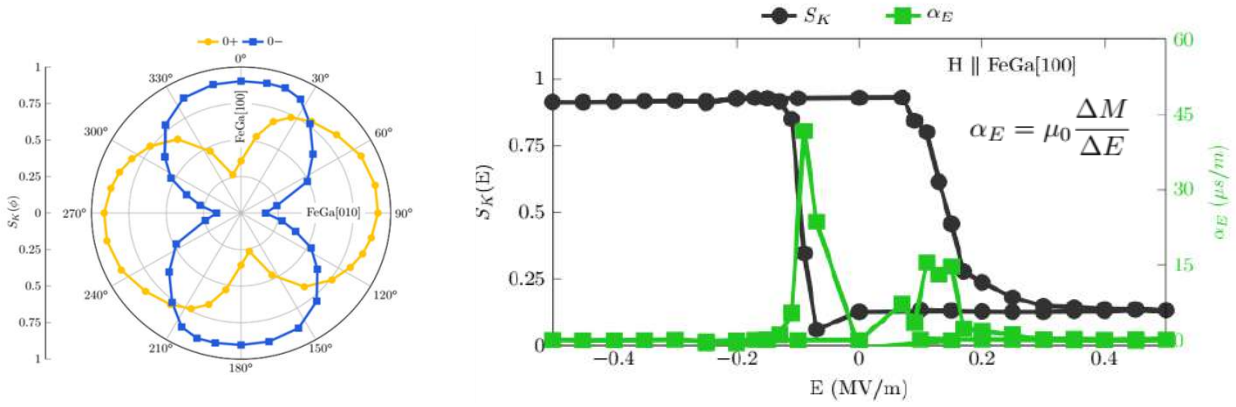



Figure 1. Left: Squareness, $S_K$, as a function of the angle after applying a positive (0+) and negative (0-) pulse of electric field. Right: $S_K$ and the inverse ME coefficient as a function of the applied electric field.

# Enhancing Magneto-Ionics in Electrolyte-Gated Co oxide Thin Films by Fine-Tuning Electrolyte Composition

*Eva Pellicer*[1]*, *Sofia Martins*[1], *Zheng Ma*[1], *Nicolau López-Pintó*[1], *Aitor Arredondo-López*[1], *Xavier Solans-Monfort*[2], *Luis Rodriguez-Santiago*[2], *Mariona Sodupe*[2], *Enric Menéndez*[1], *and Jordi Sort* [1,3]

[1] *Departament de Física, Universitat Autònoma de Barcelona, E-08193 Cerdanyola del Vallès, Spain*

[2] *Departament de Química, Universitat Autònoma de Barcelona, E-08193 Cerdanyola del Vallès, Spain*

[3] *Institució Catalana de Recerca i Estudis Avançats (ICREA), Pg. Lluís Companys 23, E-08010 Barcelona, Spain*

*eva.pellicer@uab.cat

Significant progress has been made in recent years to improve voltage-driven ion motion rates and cyclability, which are the two main bottlenecks to exploit the full potential of magneto-ionics. This has been achieved by proper selection of moving ion species ($O^{2-}$, $N^{3-}$, $H^{+}$, $F^{-}$, etc.), target materials, heterostructure designs, and voltage actuation protocols. However, room-temperature oxygen magneto-ionics is still too slow for relevant applications. Improvements are still required to further increase oxygen motion speeds and to reduce the required threshold voltages. In particular, for electrolyte-gated magneto-ionic films of tens of nanometers in thickness, voltages of several tens of volts are often needed [1].

In this study, we demonstrate that oxygen motion in electrolyte-gated 15 nm-thick Co oxide film is significantly increased via 'electrolyte engineering'. In other words, we tune the composition of the electrolyte instead of manipulating the Co oxide thin film or the working conditions, which is the most referenced pathway to improved magneto-ionics in literature. In particular, potassium iodide (KI), potassium chloride (KCl) and calcium tetrafluoroborate ($Ca(BF_4)_2$), are added to anhydrous propylene carbonate (PC) to modulate the ionic strength and, in turn, the electric field at the Co oxide/electrolyte interface by means of the built-in electric double layer. Specially for KI-containing PC, a 35-fold increase of the magneto-ionic rate is observed as compared to plain PC, and importantly, under a relatively low bias voltage of –1.5 V [2]. The time evolution of magnetization has been experimentally followed and quantified for each case. The formation of metallic Co clusters upon negative biasing of the Co oxide film is probed by X-ray photoelectron spectroscopy. *Ab initio* molecular dynamics simulations support the observed findings, namely that the effects observed for KI are greater than for KCl, which is a result one would not expect a priori.

## Acknowledgements

The authors acknowledge financial support from the European Union's Horizon 2020 research and innovation programme BeMAGIC under the Marie Sklodowska-Curie grant agreement No 861145, the Spanish Government (PID2020-116844RB-C21, PDC2021-121276-C31 and PID2020-112715GB-I00), and the Generalitat de Catalunya (2021-SGR-292 00651 and 2017-SGR-1323). The Red Española de Supercomputación (QHS-2022-2-0029) is also acknowledged.

## References

[1] S. Martins, J. de Rojas, Z. Tan, M. Cialone, A. Lopeandía, J. Herrero-Martín, J. L. Costa-Krämer, E. Menéndez, J. Sort, *Nanoscale* **14** (2022) 842.

[2] S. Martins, Z. Ma, X. Solans-Monfort, M. Sodupe, L. Rodriguez-Santiago, E. Menéndez, E. Pellicer, J. Sort, *Nanoscale Horiz.* **8** (2023) 118.

# Hysteresis Losses Limiting First-Order Phase Transition Materials in Cryogenic Caloric Cooling

*B. Beckmann[1]*, D. Koch[1], L. Pfeuffer[1], T. Gottschall[2], A. Taubel[1], E. Adabifiroozjaei[1], O. N. Miroshkina[3], S. Riegg[1], T. Niehoff[2], N. A. Kani[1], M. E. Gruner[3], L. Molina-Luna[1], K. P. Skokov[1], O. Gutfleisch[1]*

[1] *Institute of Materials Science, Technical University of Darmstadt, 64287 Darmstadt, Germany*

[2] *Dresden High Magnetic Field Laboratory, Helmholtz-Zentrum Dresden-Rossendorf, 01328 Dresden, Germany*

[3] *Faculty of Physics, University of Duisburg-Essen, 47057 Duisburg, Germany*

*benedikt.beckmann@tu-darmstadt.de

First-order phase transition materials, such as all-d-metal Ni(-Co)-Mn-Ti Heusler alloys [1], are highly promising candidates for energy-efficient solid-state refrigeration as large multicaloric effects can be achieved upon driving the respective phase transition with an external stimulus. However, first-order materials suffer from reduced cyclic caloric effects, mechanical failure due to large volume changes, and hysteresis losses.

In this work [2], we investigate the hysteresis losses at cryogenic temperatures using Ni(-Co)-Mn-Ti as a model material system. We analyze the responses of the magnetic, structural and electronic subsystems to the temperature- and magnetic field-induced martensitic transformation, showing an abnormally increased magnetic hysteresis width at cryogenic temperatures (Fig. 1 (a)). Based on this, we reveal the detrimental effect of hysteresis losses on the adiabatic temperature change, leading to substantial irreversible heating of the caloric material (Fig. 1 (b)). Most importantly, this phenomenon is universal, it applies to any first-order material with hysteresis, effectively limiting their utilization for gas liquefaction at cryogenic temperatures.

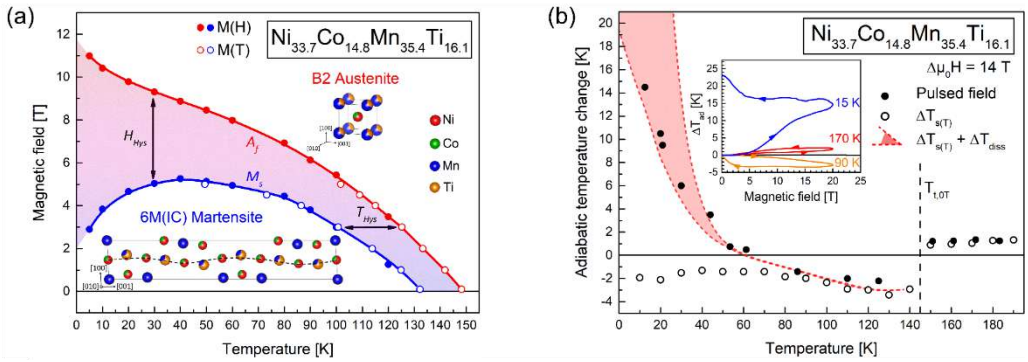



Figure 1: (a) Temperature- and magnetic field-dependent phase diagram of $Ni_{33.7}Co_{14.8}Mn_{35.4}Ti_{16.1}$ with austenite finish $A_f$, martensite start $M_s$, thermal hysteresis width $T_{Hys}$ and magnetic hysteresis width $H_{Hys}$. (b) Temperature dependence of the adiabatic temperature change $\Delta T_{ad}$ in $Ni_{33.7}Co_{14.8}Mn_{35.4}Ti_{16.1}$ measured with pulsed magnetic fields and determined with $s(T)$ based on $c_p$ measurements ($\Delta T_{s(T)}$) in a magnetic field change of 14 T. The estimated effect of dissipation losses is given by $\Delta T_{diss}$. The upper limit is neglecting and the lower limit is considering adiabatic boundary conditions. The zero-field martensite to austenite transformation temperature is given by $T_{t,0T}$. The inset shows the magnetic field dependence of the measured adiabatic temperature change for a pulsed magnetic field of 20 T at 15, 90, and 170 K.

## Acknowledgements

We acknowledge financial support by the Deutsche Forschungsgemeinschaft (DFG) within the CRC/TRR 270 (Project-ID 405553726) and BEsT (Project-ID 456263705) as well as by the European Research Council (ERC) under the European Union's Horizon 2020 research and innovation programme (Grant No. 743116). We acknowledge the support of the HLD at HZDR, member of the European Magnetic Field Laboratory (EMFL).

## References

[1] A. Taubel, B. Beckmann, L. Pfeuffer, N. Fortunato, F. Scheibel, S. Ener, T. Gottschall, K. P. Skokov, H. Zhang, O. Gutfleisch, *Acta Mater.* **201** (2021) 425-434

[2] B. Beckmann, D. Koch, L. Pfeuffer, T. Gottschall, A. Taubel, E. Adabifiroozjaei, O. N. Miroshkina, S. Riegg, T. Niehoff, N. A. Kani, M. E. Gruner, L. Molina-Luna, K. P. Skokov, O. Gutfleisch, *Acta Mater.* **246** (2023) 118695

# Concomitant Thermochromic and Phase-Change Effect in a Switchable Spin Crossover Material for Efficient Passive Control of Day and Night Temperature Fluctuations

[1]Jose Sánchez Costa*, [1]Esther Resines-Urien, [2]Miguel Ángel García García-Tuñón, [3]Mar García-Hernández, [4]Jose Alberto Rodríguez-Velamazán, [1]Ana Espinosa,*

[1]IMDEA Nanociencia, C/ Faraday 9, Madrid 28049, Spain, Tel: 912998888.
[2]Instituto de Cerámica y Vidrio, CSIC, C/Kelsen s/n, Madrid 28240, Spain.
[3]Instituto de Ciencia de Materiales de Madrid, CSIC, C/Sor Juana Inés de la Cruz, 3, 28049, Madrid, Spain.
[4]Institut Laue-Langevin, 71 avenue des Martyrs, BP 156, 38042, Grenoble Cedex 9, France.
*jose.sanchezcosta@imdea.org

Nowadays, building thermalization is a widespread human necessity, accounting for 28% of energy-related carbon dioxide ($CO_2$) worldwide emissions.[1] These $CO_2$ emissions are a major contributor to climate change, which has become one of the biggest concerns of humankind.[2] Thus, the growing energy-saving and environmental protection demand have prompted the development and implementation of more energy efficient and environmentally friendly thermalization technology. In this regard, remarkable efforts have been focused on the implementation of passive thermal regulation systems, that can be incorporated directly into windows,[3] roofs, or walls of buildings and operate without the need for electricity.[4] Here, it is demonstrated that the heat generated by the sun is sufficient to produce a partial spin transition in an spin crossover (SCO) material. SCO materials exhibit a reversible transition, between the high spin and low spin electronic states through the application of external stimuli such as temperature.[5] This SCO leads to a cooling effect with respect to other materials, due to an increase in light reflection resulting from the color change (from pink to white) and the energy absorption associated with the spin transition. In addition, when the material is cooled, a dampening of the temperature decrease is produced due to the energy release associated with the spin transition. Therefore, these materials can be used to reduce temperature fluctuations, and could potentially be implemented for passive temperature control in buildings. Interestingly, SCO materials are remarkably stable upon cycling and highly versatile, allowing for the design of compounds adapting the intended properties (transition temperature and hysteresis) for the desired climatic conditions and comfort temperature.

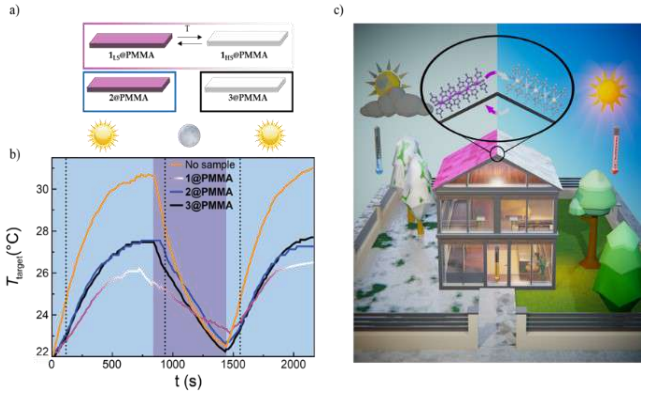



Figure 1. a) Compounds employed in the experiment: 1 (that displays a SCO with a change of color between white at high temperatures and pink at low temperatures), 2 (pink) and 3 (white). b) Temperature in the sensor vs. exposure time with on-off solar simulator cycles c) Illustration of the working scheme

## References

[1] International Energy Agency "The Critical Role of Buildings, Perspectives for the Clean Energy Transition – Analysis", https://www.iea.org/reports/the-critical-role of-buildings (accessed: November 2020).
[2] Crowley, T. J.; Berner, R. A.; Science 2001, 292, 870-872.
[3] Wang, S.; Zhou, Y.; Jiang, T.; Yang, R.; Tan, G.; Long, Y.; Nano Energy 2021, 89, 106440.
[4] Granqvist, C. G.; Adv. Mater. 2003, 15, 1789-1803.
[5] Resines-Urien, E.; García García-Tuñón, M. A.; García-Hernández, M.; Rodríguez-Velamazán, J. A.; Espinosa, A.; Sánchez Costa, J. Advanced Science, 2022, 24, 2202253

# Magnetic Shape Memory Heuslers for Low-grade Heat Harvesting

*L.Gallo[1,2]*, F.Cugini[1,2], G. Garulli[2], S. Fabbrici[1], G. Trevisi[1], D. Olivieri[2], M. Solzi[1,2], F. Albertini[1]*

[1] *IMEM-CNR, Via delle Scienze 37, Parma, Italy*

[2] *Department of Mathematical, Physical and Computer Science, University of Parma, Parco Area delle Scienze 7/A, Parma, Italy*

*lorenzo.gallo@unipr.it

Manufacturing and industrial processes lead to considerable waste heat and most of it is emitted at less than one hundred Celsius degrees. The conversion of this large amount of untapped heat into useful energy might represent an important contribution to the energy transition of our society [1]. To date, the harvesting of low-grade heat is only based on thermoelectric materials. However, such materials are too expensive for industry use and they are characterized by a low thermodynamical efficiency for thermal sinks near room temperature. The identification of new magnetic materials, that exploit the thermomagnetic cycle for the conversion of low-grade heat, might introduce a promising technology for fully sustainable energy production. Such materials must have as higher as possible magnetization change and thermal diffusivity in the working temperature range of their applications. In addition, they should be possibly free of critical elements and cost-effective [2].

We synthesized, characterized, and tested a set of Ni-Mn-Z Heusler alloys (with Z = Sn, In, Cu-Ga) for low-grade heat recovery. The selected compounds are characterized by a second-order Curie magnetic transition between 40–80 °C to maximize the change of magnetization in the typical working temperature range of a thermomagnetic generator for low-grade heat harvesting. We chose to work around the second order transition because, even if a first-order magnetostructural martensitic transition leads to a sharper variation of the magnetization, the thermal hysteresis of such transition can lower the efficiency of the thermomagnetic cycle. Finally, we reduced the compounds to powders and we realized some rotor-shape functionalized graphene-based composites [3] to test them directly on a thermomagnetic generator developed in situ.

The wide tunability of the magnetic interactions and critical temperatures of the synthesized Heuslers have been investigated, revealing the potential of this class of materials for thermomagnetic energy harvesting. The obtained tunability of the Curie temperature of the compounds opens the possibility to find the most appropriate material for every temperature working range of the heat conversion device. The strict correlation between the magnetization change across the transition and the efficiency of the thermomagnetic generator will be also discussed.

## References

[1] C. Formann et al., *Renew. Sustain. Energy Rev.* **57** (2016) 1568–1579.

[2] D. Dzekan, A. Waske, K. Nielsch, and S. Fähler, *APL Materials* **9** (2021) 011105.

[3] C. Coppi et al., *Adv. Eng. Mater.*, **25** (2023) 2200811

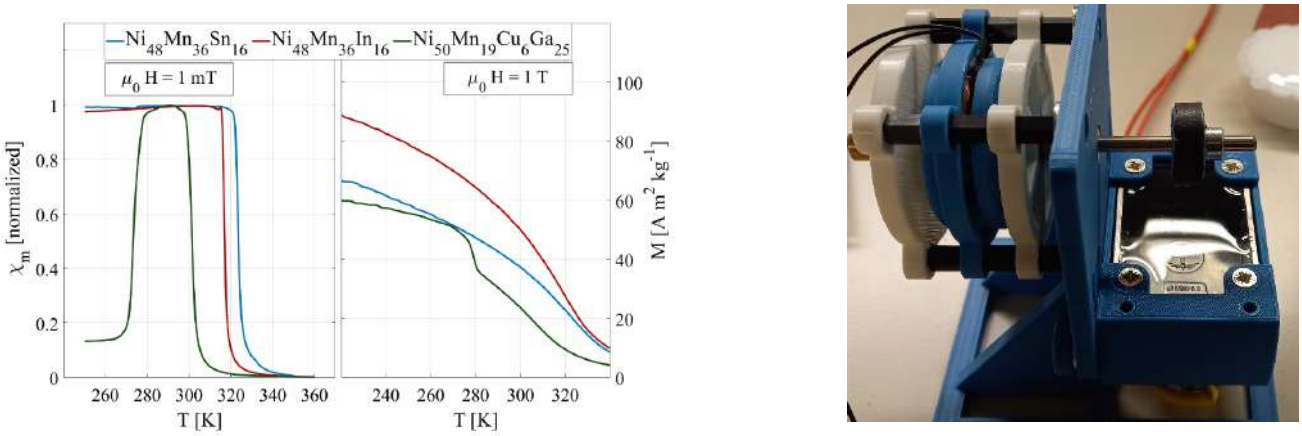



Figure: (left) magnetic measurements of the three Heusler compounds NiMnSn (blue), NiMnIn (red), NiMnCuGa(green). In the left graph are reported the magnetic susceptibilities over temperature with an applied field of $10^{-3}$ T, and in the right graph are reported the isolfied curves with an applied field of 1 T. (right) The thermomagnetic generator developed in situ as thermomagnetic materials tester.

# Probing the kinetics of $LaFe_{11.6}Si_{1.4}$ magnetic field-induced (iso)structural transitions with X-ray Diffraction

*A. Beleza [1], R. Almeida [1], B. Beckmann[2], A. M. dos Santos[3], J. E. Araújo[1], M. D. Frontzek[3], E. Lovell[4], J. Amaral[5], O. Gutfleisch[2], K. Skokov[2], AM. M. Abeykoon[6],* ***J. H. Belo[1]****  *(jbelo@fc.up.pt)*

[1] *IFIMUP, Departamento de Física e Astronomia da Faculdade de Ciências da Universidade do Porto, Rua do Campo Alegre, 687, 4169-007 Porto, Portugal*

[2] *Institute of Material Science, Technical University of Darmstadt, 64287 Darmstadt, Germany*

[3] *Neutron Scattering Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831-6475, USA*

[4] *Camfridge Ltd., Cambridge CB22 3GN, UK*

[5] *Department of Physics and CICECO, University of Aveiro, Campus of Santiago, 3810-193 Aveiro, Portugal*

[6] *National Synchrotron Light Source II, Brookhaven National Laboratory Upton, NY 11973, USA*

Magnetic refrigeration, one of the most promising alternatives to conventional cooling/heating technology, makes use of magnetocaloric effect (MCE) which is maximized in materials that exhibit a strong magnetovolume coupling associated with first-order phase transitions (FOPTs), such as $Gd_5(Si,Ge)_4$ and $La(Fe,Si)_{13}$ [1]. In order to optimize a magnetic heat pump efficiency, one approach is to enhance its frequency – and for that the fundamental knowledge about the time required for the materials to undergo these FOPTs is crucial. From recent litereature, it has been demonstrated that magnetic transitions can range up to 100s and strongly depends on the sample shape, temperature and magnetic field [2]. Since FOPT transitions occur in strong spin-lattice coupled materials, it is expected that, in parallel with the magnetization time-dependent evolution, there must be a crystal structure time-dependent evolution.

In this work, we present for the first time (to the best of our knowledge) time-dependent evolution of the cubic structure of a $LaFe_{11.6}Si_{1.4}$ sample across its magnetovolume transition probed by Synchrotron XRD as a function of temperature, magnetic field, time, and direction of the transition. The time-scales and the evolution profiles of the lattice parameter as a function of time (Figure 1 b)) are shown to strongly depend on the field and temperature. Remarkably a strong asymmetry was also observed between the timescales of the transition triggered by increasing field (typically few hundreds of seconds) versus decreasing field (below 1 second). Through free energy estimates of a compressible Ising model system [3], it is shown that this assymetry correlates with the free energy barrier between stable and metastable states. In the field decreasing process, this barrier is small, or even non-existent, in contrast to the field increasing process.

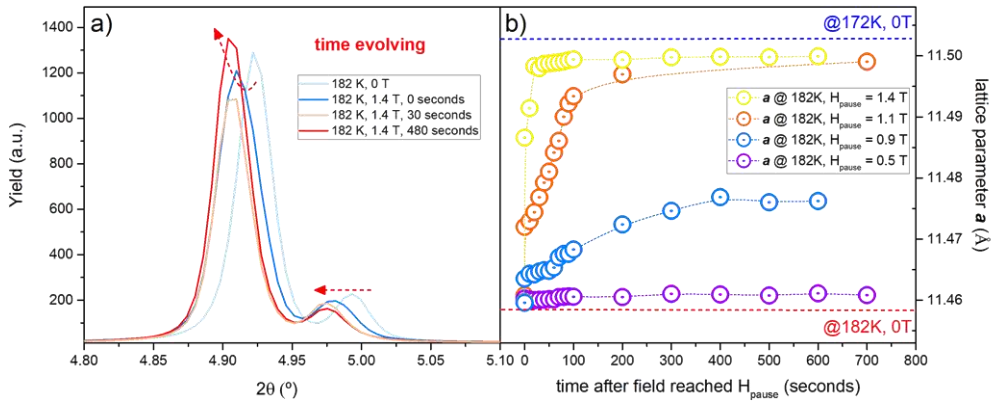



Figure 1: a) Time and field evolution of $LaFe_{11.6}Si_{1.4}$ XRD most intense peak. b) Time evolution of $LaFe_{11.6}Si_{1.4}$ lattice parameter at $T = T_C + 4\ K = 182\ K$ for a set of four different magnetic field values $H_{pause}$.

## Acknowledgements:

We would like to acknowledge FCT, for the funded projects PTDC/EME-TED/3099/2020 (IFIMUP), project CICECO-Aveiro Institute of Materials, UIDB/50011/2020, UIDP/50011/2020 & LA/P/0006/2020 and the the Deutsche Forschungsgemeinschaft (DFG) within the CRC/TRR 270 (Project-ID 405553726). A portion of this research used resources at the Spallation Neutron Source, a DOE Office of Science User Facility operated by the Oak Ridge National Laboratory.

## References

[1] T. Gottschall, K. Skokov, et al. *Adv. Ener. Mater., 9, 1901322 (2019);* [2] E. Lovell, A. M. Pereira, D. Caplin, J. Lyubina, L.F. Cohen, *Adv. Ener. Mater. 14016394* (2014); [3] J. S. Amaral and V. S. Amaral, Front. Mater 10 1037396 (2023).

# Dissecting complexity of phase transitions in first-order multi-caloric materials

*K.P. Skokov*[1], A.Yu. Karpenkov[1], D.Yu. Karpenkov[1], A. Aubert[1], B. Beckmann[1], H. Wende[2], K Ollefs[2], A. Rogalev[3], F. Wilhelm[3], and . O. Gutfleisch[1]*

[1]*Functional Materials, Technical University of Darmstadt, 64287 Darmstadt, Germany*
[2]*Faculty of Physics, University of Duisburg-Essen, 47057 Duisburg, Germany*
[3]*European Synchrotron Radiation Facility, 38043 Grenoble, France*
*Konstantin.Skokov@tu-darmstadt.de

Being driven by rapidly increasing demand for new high-tech and energy-efficient cooling devices, the multi-stimuli responsive functional materials with a strong interplay between their structural, magnetic, and electronic degrees of freedom have been recognized as a new family of promising materials for applications in emerging alternative solid-state refrigeration technologies [1]. In this context, materials with a first-order phase transition, where an application of the different generalized thermodynamic forces/fields is accompanied by large discontinuities and abrupt changes in their conjugate variables are of particular interest. For rational design of these materials, it is vitally important to know in detail, how different subsystems of the solid interplay during the transition, which system triggers the phase transition and how this mutual entanglement interaction can be responsible for the resulting magnetocaloric effect.

Our work shows a new pathway to disentangle the interplay between the structural, magnetic and electronic degrees of freedom, and is the next step towards a complete understanding of the driving forces of the transition, together with comprehension of the origin of thermal hysteresis in magnetic phase-change materials. We have built several original experimental setups for simultaneous measurement of macroscopic physical properties (magnetization, magnetostriction, resistivity, temperature change) in isothermal or adiabatic conditions [2], [3]. These devices were used for study materials with first-order magneto-structural phase transitions, such as $La(Fe,Si)_{13}$ (Fig. 1), Heusler alloy, FeRh and $RCo_2$, where the quantitative determination of elastic and magnetoelastic coupling constants is indispensable to understand the nature of field- and stress-driven phase transformation.

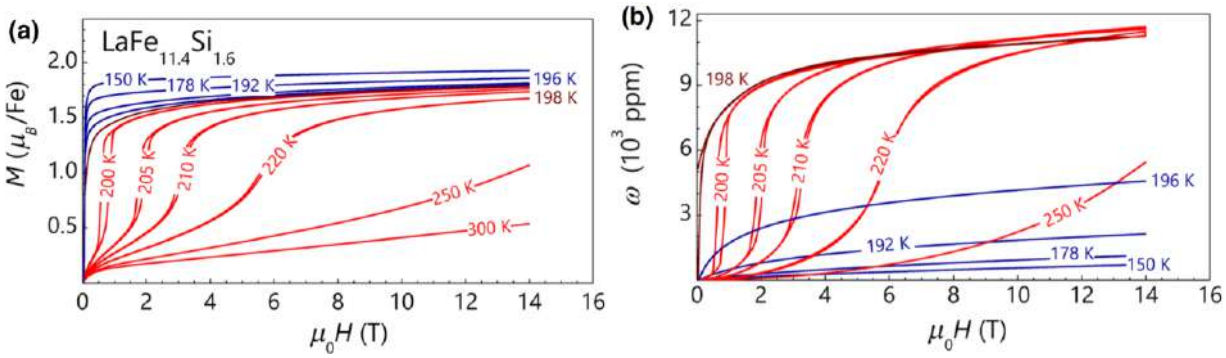



Figure 1: (a) field dependences of magnetization and (b) field dependences of magnetovolume effect, both measured simultaneously on polycrystalline $LaFe_{11.4}Si_{1.6}$

## Acknowledgements

We acknowledge financial support by the Deutsche Forschungsgemeinschaft (DFG) within the CRC/TRR 270 (Project-ID405553726).

## References

[1] O. Gutfleisch, T. Gottschall, M. Fries, D. Benke, I. Radulov, K. P. Skokov, H. Wende, M. Gruner, M. Acet, P. Entel and M. Farle, *Phil. Trans. R. Soc.* A 374 20150308 (2016)

[2] D. Y. Karpenkov, A. Y. Karpenkov, K. P. Skokov, I. A. Radulov, M. Zheleznyi, T. Faske and O. Gutfleisch. *Phys. Rev. Appl.* **13**, 034014 (2020).

[3] A. Aubert, K. Skokov, G. Gomez, A. Chirkova, I. Radulov, F. Wilhelm, A. Rogalev, H. Wende, O. Gutfleisch and K. Ollefs, *IEEE Trans. Instrum. Meas.* **71**, 1–9 (2022).

# Strategies to decrease the thermal hystéresis in NiMnInCo(Cu) metamagnetic shape memory alloys

*P.La Roca* [*1,2,3] , *J. López-García* [1,4] , *V.Sánchez-Alarcos* [1,3] , *V. Recarte* [1,3] , *J. I. Pérez- Landazábal*[1,3]
[1] *Institute for Advanced Materials and Mathematics (INAMAT2), Universidad Pública de Navarra, Pamplona 31006, Spain*
[2] *Centro Atómico Bariloche (CNEA), CONICET, Bariloche 8400, Argentina*
[3] *Science Department, Universidad Pública de Navarra, Pamplona 31006, Spain*
[4] *Department of Physics, Faculty of Science, University of Oviedo, Oviedo 33007, Spain*
* paulo.laroca@cab.cnea.gov.ar

The current refrigeration technology, based on gas vapour-compression cycles, damages the ozone layer and is reaching its technical limits of efficiency. On the other side, solid state refrigeration technology based on magnetocaloric (MC) materials is one of the most promising green technologies to replace conventional processes. In this way, Ni–Mn-based Heusler alloys exhibiting both long-range magnetic ordering and thermoelastic martensitic transformation (MT) have been intensively investigated over recent years due to their magnetocaloric properties [1,2]. However, there are important challenges to solve such as the reduction of the energy losses linked to both thermal and magnetic hysteresis and the achievement of reversible transformations with even higher MCE. In this work, different strategies have been used to decrease the thermal hystéresis (~15-20K) in NiMnInCo metamagnetic shape memory alloys. In particular, the composition,the long-range ordering and size and microstructure of the particles will be analyzed.
The change of the chemical composition (adding Cu) combined with an adecuate thermal treatment to promote long-range atomic ordering, decreases the thermal histérisis of bulk alloys from 20-15K to 6-7K [3]. This chemical and structural modifications enhance the crystallographic compatibility between austenite and martensite, bringing about a remarkable reduction of thermal hysteresis. Also, the ordering increases the magnetization of the austenite without changing the transformation temperatures, doubles the sensitivity of the transformation to magnetic fields (the Claussius-Clapeyron slope increases from 2.1 to 3.9 K/T), and improves the magnetocaloric effect, the reversibility and finally, enhances the refrigeration capacity [3]. Subsequently, microparticles have obtainded by hand crash. The high density of microstructural defects induced by grinding have been recovered and quasi-monocrystalline particlles were obtained. This last step allow to reduce the thermal hysteresis from 6K (Bulk) to 3K.

**Acknowledgements**

"Agencia Estatal de Investigación (AEI), Ministerio de Ciencia, Innovación y Universidades" (Projects number RTI2018-094683-B-C54 (MCIU/AEI/FEDER, UE)). P. La Roca has received funding from "la Caixa" and "Caja Navarra" Foundations, under agreement LCF/PR/PR13/51080004.

**References**

[1] X. Moya, S. Kar-Narayan, N.D. Mathur, Nat. Mater. 13 (2014) 439-450
[2] J. Liu, T. Gottschall, K.P. Skokov, J.D. Moore, O. Gutfleisch, Nat. Mater. 11 (2012) 620-626.
[3] P. La Roca, J. López-García, V. Sánchez-Alarcos, V. Recarte, J.A. Rodríguez-Velamazán, J.I. Pérez-Landazábal, Journal of Alloys and Compounds 922, 166143 (2022)

# Effects of Nanoindents on the Martensitic Transformation of Heusler Films

*M. Takhsha[1]*, F. Casoli[1], C. Minnert[2], S. Bruns[2], E. Bruder[2], R. Cabassi[1], K. Durst[2], O. Gutfleisch[3], and F. Albertini[1]*

[1] *IMEM-CNR, Parco Area delle Scienze 37/A, 43124 Parma, Italy*
[2] *Physical Metallurgy, TU-Darmstadt, Alarich-Weiss-Str. 16, 64287 Darmstadt, Germany*
[3] *Functional Materials, TU-Darmstadt, Alarich-Weiss-Str. 16, 64287 Darmstadt, Germany*
*milad.takhsha@imem.cnr.it

Magnetic-shape-memory (MSM) Heusler compounds are promising for a variety of applications in actuating, sensing, energy harvesting, spintronics and multicaloric cooling technologies. Multifunctionality of these materials originates from a reversible martensitic phase transformation, which is typicaly accomponied by some undesirable characteristics (e.g. thermal hysteresis and broad transition). Those prevent full exploitation of MSM materials towards applications [1 and references therein].

Over the last decade, we have investigated martensitic phase trasformation of Ni-Mn-Ga MSM Heusler films for gaining a comprehensive insight into the effects of different factors, processes and stimuli such as the lateral size, film thickness, martensitic interfaces, twins and solid-state dewetting process on the phase transformation of the material [1-5]. These can eventually help us designing the material with an optimal phase trasformation characteristics.

In this study, we apply indentation forces by a Berkovich tip covering two orders of magnitude (0.1-10.0 mN) as the sources of explicit deformations in epitaxial Ni-Mn-Ga films on MgO(001) substrate. We evaluate the nucleation, growth and annihilation of the martensitic phase in the nanometric and micrometric scales by atomic/magnetic force microscopy imaging in temperature as a function of a series of indentation forces and different distances from the indents. The focus of the work is to study the links between the indentation force and the local impacts of the indents on the martensitic transformation route of the material.

We observe a local increase of the martensitic transformation temperature (up to around 5 K) as a function of the applied forces that follows a non-linear regime, reaching a plateau by increasing the applied force. The observed effect is local and almost disappears for distances longer than 500 nm from the deformed regions around the indents called pile-ups beyond which the material transforms similarly to the pristine sample. The local increase of transformation temperatures as a function of nanoindentation forces occurs in both the cooling and the heating curves. Therefore, no considerable thermal hysteresis variation is observed in the transition of the material. The major portion of the temperature increase is obtained at the martensite-fraction range between zero to fifty percent, resulting in “broadening” of the transition close to the indents. The results are discussed by highlighting the shear mechanism of the martensitic transformation and the pinning effect of the substrate [1].

## References

[1] M. Takhsha Ghahfarokhi, F. Casoli, C. Minnert, S. Bruns, E. Bruder, R. Cabassi, K. Durst, O. Gutfleisch, F. Albertini, *Acta Mater.* **245** (2023) 118603.
[2] M. Takhsha Ghahfarokhi, F. Celegato, G. Barrera, F. Casoli, P. Tiberto, F. Albertini, *Crystals* **12** (2022) 1826.
[3] M. Takhsha Ghahfarokhi, A. Chirkova, F. Maccari, F. Casoli, S. Ener, K. P. Skokov, R. Cabassi, O. Gutfleisch, F. Albertini, *Acta Mater.* **221** (2021) 117356.
[4] M. Takhsha Ghahfarokhi, J. A. Arregi, F. Casoli, M. Horký, R. Cabassi, V. Uhlíř, and F. Albertini, *Appl. Mater. Today* **23** (2021) 101058.
[5] Takhsha Ghahfarokhi, F. Casoli, S. Fabbrici, L. Nasi, F. Celegato, R. Cabassi, G. Trevisi, G. Bertoni, D. Calestani, P. Tiberto, F. Albertini, Acta Mater. 187 (2020) 135

.

# A Gd based MEMS-scale thermomagnetic generator

*Morgan Almanza[1], Nesrine Belkadi[2], Jean-Eudes Duvauchelle[3], Erika Fontana[4], Doan Nguyen Ba[1,3], Loïc Becerra[3], Thibaut Devillers[4], Nora M. Dempsey[4], Massimiliano Marangolo[3], Fabien Parrain[2], Martino LoBue[1*]*

[1] *Université Paris-Saclay, ENS Paris-Saclay, CNRS, SATIE, F-91190 Gif-sur-Yvette, France*
[2] *Université Paris-Saclay, CNRS, Centre de Nanosciences et de Nanotechnologies, 91120, Palaiseau, France*
[3] *Sorbonne Université, CNRS, Institut des NanoSciences de Paris, UMR7588, 75252 Paris, France*
[4] *Université Grenoble Alpes, CNRS, Grenoble INP, Institut Néel, 38000 Grenoble, France*

*martino.lo-bue@ens-paris-saclay.fr

The demand for small, lightweight, and autonomous power supply generators to sustain low-power wireless sensor nodes, wearable devices, and implants has been continuously rising for more than one decade. At the same time, as the cheapest, and most ubiquitous energy source available in anthropized environments is ultralow waste heat (i.e. in a temperature range from 25°C to 80°C) [1], its efficient scavenging represents a key, and urgent technological goal. Recently, a self-actuating thermomagnetic generator (TMG), using as an active substance a 5 µm Heusler film, and working over a 138°C temperature difference, has been reported [2]. Its power-output of 118 $mWcm^{-3}$, surpasses the state-of-the-art of small scale powering solutions, mostly relying on thermoelectric generators [2].

Here we present a TMG prototype somewhat inspired by the one reported in [2] but scaled down in size, and working over a smaller temperature gradient. Reducing the temperature difference while keeping a high power output has been obtained through improved thermal management. Working over 20°C, at 100 Hz, we estimate an available power of the order of 10 $mWcm^{-2}$. From this standpoint the device is an excellent candidate for ultralow heat harvesting. The size has been scaled down through optimization of the field source making the device the smallest and lightest TMG prototype reported so far. To do this we used a thick, free-standing, flexible, Gd film [3], jointly with a patterned NdFeB hard-magnet film [4] to get highly confined fields, with strong gradients, over displacements of tens of micrometers. Combining the properties of the Gd film and NdFeB micro-magnets, we identified the optimal size and elastic force to get the micro-device working through a self-oscillating (i.e. fully autonomous) cycle driven by a suitable balance between elastic and magnetic forces. Mechanical to electric energy conversion is obtained through a laser-cut piezoelectric spring. Experiments show that a frequency up to 100 Hz is achievable working over a small temperature difference, with the hot side at 30°C and heat sink at 10°C. This result is a key step towards higher output power. We shall present the main characteristic of the device with a particular focus on its self-actuating dynamics, and on the prospects to achieve further improvements of its throughput.

**Acknowledgements**

This research has received funding from the French National Research Agency under the project HiPerTher-Mag (ANR-18-CE05-0019), and from the financial support of the LabEx LaSIPS (ANR-10-LABX-0032-LaSIPS) under the "Investissements d'avenir" program (ANR-11-IDEX-0003).

**References**

[1] M. Luberti, R. Gowans, P. Finn, and G. Santori, Energy **238** (2022) 121967.
[2] Gueltig et al., Adv. Energy Mater. **7** (2017) 1601879.
[3] D. Nguyen Ba, Y. Zheng, L. Becerra, M. Marangolo, M. Almanza, and M. LoBue, Phys. Rev. Appl. **15** (2021) 064045.
[4] F. O. Keller, R. Haettel, T. Devillers, and N. M. Dempsey, IEEE Trans Mag 58 (2022) 2101005

# Anomalous Nernst effect in FeGa film and bulk samples

*Alessandro Sola[1]*, Elena Olivetti[1], Vittorio Basso[1], Federica Celegato[1], Marco Coisson[1], Gabriele Barrera[1], Gajanan Pradhan[1], Abdeslem Fnidiki[2], Jean Juraszek[2] and Paola Tiberto[1]*

[1] *Istituto Nazionale di Ricerca Metrologica, Strada delle Cacce 91, 10135 Torino, Italy*
[2] *UNIROUEN, INSA Rouen, CNRS, GPM, Normandie Université, 76000 Rouen, France*
*a.sola@inrim.it

The development of heat flux sensors based on planar architectures as well as thermoelectric modules for energy harvesting and efficient cooling can benefit from the research on transverse thermoeletric effects [1]. In this framework, Fe-based compounds and alloys play a fundamental role because of their relatively large anomalous Nernst effect (ANE) thermopower, their availability and ease of being produced in thin films [2]. Recently, the ANE of $Fe_{100-x}Ga_x$ has been investigated as a function of the Ga concentration [3], with the thermal gradient along the in-plane direction and the magnetic field along the out-of-plane direction of of the film under test.

In our work, we investigate the ANE thermopower of $Fe_{100-x}Ga_x$ films and we compare their values with those of polycrystalline bulk samples, with Ga concentrations in the range $x$ = 18–30. We adopt a measurement configuration whose geometry is compatible with the design of heat flux sensor devices. This includes the measurement of the ANE voltage dependence on the heat currents through the surface of the sample, in the out-of-plane direction, and the temperature dependence of the ANE thermopower for typical working temperature values. By assuming the value of the thermal conductivity of the active material as equal to 15 $WK^{-1}m^{-1}$, we find a value of the ANE thermopower equal to -2.9 μV/K in a 200 nm thick $Fe_{70}Ga_{30}$ film, compatible with the highest values reported so far in the literature [2,3,4].

## Acknowledgements

The authors gratefully acknowledge the financial support of the Italian Ministry of University and Research (MUR) under the Next-Generation Metrology project.

## References

[1] Boona, Stephen R., Hyungyu Jin, and Sarah Watzman. "Transverse thermal energy conversion using spin and topological structures." *Journal of Applied Physics* 130.17 (2021): 171101.
[2] Sakai, Akito, et al. "Iron-based binary ferromagnets for transverse thermoelectric conversion." *Nature* 581.7806 (2020): 53-57.
[3] Nakayama, Hiroyasu, et al. "Mechanism of strong enhancement of anomalous Nernst effect in Fe by Ga substitution." *Physical Review Materials* 3.11 (2019): 114412.
[4] Feng, Zili, et al. "Giant and Robust Anomalous Nernst Effect in a Polycrystalline Topological Ferromagnet at Room Temperature." Advanced Functional Materials (2022): 2206519.

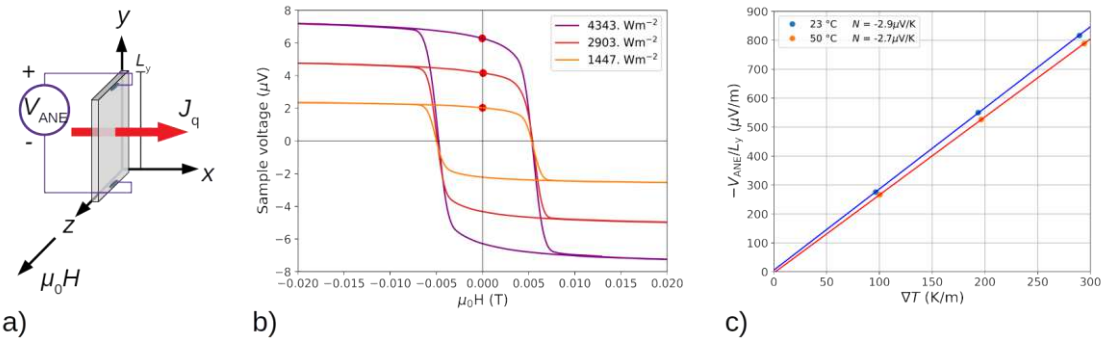



Figure: a) Scheme of the measurement system. b) Example of the ANE voltage as a function of the applied magnetic field at different values of heat current for a $Fe_{70}Ga_{30}$ film (thickness = 200 nm): the values of ANE voltage for the evaluation of the ANE thermopower have been measured at magnetic remanence (red dots on the loops). c) ANE thermopower from the same sample at room temperature and at 50°C.

# Microstructure Design to Tailor Multicaloric Materials for a Novel Multi-Stimuli Cooling Cycle

*A. Taubel[1]*, L. Pfeuffer[1], F. Scheibel[1], N. Shayanfar[1], B. Beckmann[1], T. Gottschall[2], K. P. Skokov[1] and O. Gutfleisch[1]*

[1] *TU Darmstadt, Institute of Material Science, Alarich-Weiss-Str. 16, 64287 Darmstadt, Germany*

[2] *Dresden High Magnetic Field Laboratory (HLD-EMFL), Helmholtz-Zentrum, Germany*

*andreas.taubel@tu-darmstadt.de

Magnetocaloric refrigeration attracts a lot of attention since it can be more energy efficient and environmentally friendly than current vapor compression technology. Many efforts are guided towards a reduction of the thermal hysteresis to enhance the cyclic magnetocaloric effect in first-order materials. An alternative solution has been proposed, which benefits from the thermal hysteresis by using two stimuli (magnetic field and uniaxial stress) to trigger the phase transition in a multicaloric material [1].

In this work, we present the material development for tailored chemistry and microstructure for the multi-stimuli cooling cycle. We follow different approaches for an increased mechanical stability for reliable cyclic performance combined with optimum thermal hysteresis width and large caloric effects:

(1) Tailoring microstructure by processing, where we use Ni-Mn-In Heusler alloys to produce samples with different grain sizes and degree of texture to study the influence on mechanical stability and stress-sensitivity of the phase transition. We can achieve for Ni-Mn-In a cyclic multicaloric effect of 4.1 K in 1.9 T and 55 MPa, which exceeds the corresponding magnetocaloric effect by more than 200 % [2]

(2) Tailoring microstructure by doping, where we introduce secondary phases in Ni-Mn-In to study mechanical stability and caloric response reaching a stable elastocaloric effect over 16.000 cycles [3,4]

(3) Using mechanically stable Ni-Co-Mn-Ti with intrinsically strong chemical bondings and developing thermal hysteresis and caloric effect by chemistry and processing [5].

In order to test the developed materials under application near conditions, we developed a novel multicaloric testbed which allows the direct measurement of the multicaloric material performance in an “exploiting-hysteresis cycle”. Using this purpose-built device we study the multicaloric performance of FeRh under different pulsed magnetic fields and uniaxial loads. We demonstrate that in FeRh a cyclic multicaloric effect of 2.5 K can be achieved whereas its magnetocaloric counterpart in similar fields is negligibly small.

**Acknowledgements**

This work was supported by the ERC Advanced Grant "CoolInnov" (No 743116), and the CRC/TRR 270 “HoMMage” (DFG).

**References**

[1] T. Gottschall et al., *Nature Mat.* **17**, 929–934 (2018)
[2] L. Pfeuffer et al., *Acta Materialia* **217** 1175157 (2021)
[3] L. Pfeuffer et al., *Acta Materialia* **221**, 117390 (2021)
[4] F. Scheibel et al., *Journal of Applied Physics* **133**, 075104 (2023)
[5] A. Taubel et al., *Acta Materialia* **201**, 425-434 (2020)

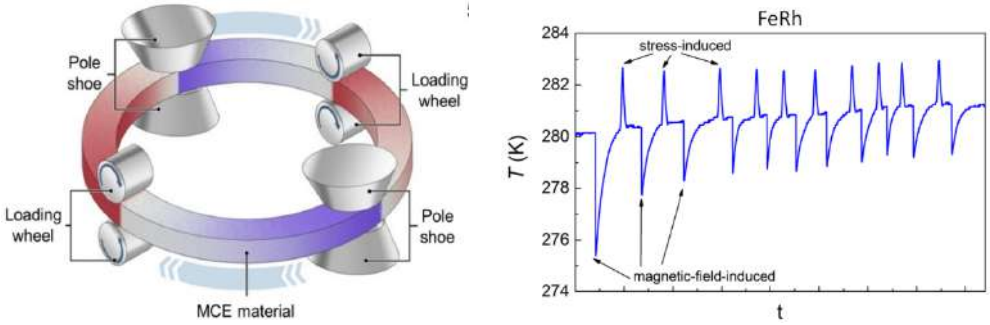



Figure 1: Schematic of the multi-stimuli cooling cycle (left) and cyclic temperature change of FeRh in a testbed for subsequent application of magnetic field and uniaxial stress (right)

# Attempting to Circumvent Inert Atmospheres in Recycling End-of-Life NdFeB Magnets

*C. Granados-Miralles*[1]*, *A. Berja*[1], *L. Grau*[2], *A. Serrano*[1], *J. López-Sánchez*[1], *M.A. García*[1], *C. Burkhardt*[2], *A. Quesada*[1]

[1] *Instituto de Cerámica y Vidrio, CSIC, 28049 Madrid, Spain*

[2] *Institute for Precious and Technology Metals, Pforzheim University, 75175 Pforzheim, Germany*

*c.granados.miralles@icv.csic.es

Permanent magnets are behind the operation of countless devices, including many of the modern technologies enabling the transition towards a climate-resilient sustainable green future, such as wave and wind power generators or electric vehicle motors. When seeking for maximizing the energy efficiency while minimizing the size (weight) of these devices, magnets based on rare-earth elements (REEs) are preferred. However, the REEs used in permanent magnets (e.g., Nd, Dy, Pr, …) have long held the top positions on the list of the most critical materials with the highest supply risk, and the situation is only expected to worsen, with the EU forecasting a tenfold increased demand by 2050.[1] In this framework, booting the recycling rates of REE magnets and optimizing the current recycling procedures is crucial to ensure the sustainability of clean energy production and use.

Recycling NdFeB magnets from end-of-life (EOL) products is a promising solution to meet the high demand expected for the coming years while minimizing the harmful environmental impact of REEs extraction. The present study employs NdFeB powders recovered from household appliances through HDDR (Hydrogenation-Decrepitation-Desorption-Recombination). A common procedure after the HDDR treatment is jet-milling the obtained NdFeB down to grain sizes of 3–5 µm, which may be then aligned, compressed and sintered to manufacture fully-dense recycled magnets.[2]

In the present work, we have explored an alternative method to the costly jet-milling procedure. The two-step milling protocole developed here has allowed reducing the NdFeB average grain size down to 4.83 µm, which is competitive compared with the values attained by jet-milling, while in an attempt to simplify the process, the powders have been manipulated in air. The NdFeB powders have been extensively studied before and after the various steps. Rietveld analysis of powder X-ray diffraction (PXRD) data and X-ray absorption spectroscopy (XAS) have allowed following the changes in composition, microstructure, local structure and crystallite size, unravelling the formation of Nd-rich phases (oxides and hydroxides) and other secondary phases, while field-emission scanning electron microscopy (FE-SEM) and confocal Raman microscopy (CRM) measurements have unveiled the sample morphology and the distribution of secondary phases.Air exposure yielded a partial oxidation of the material (estimated below 10%) but we have been able to confirm that most NdFeB remains in a metallic state. Ongoing work includes the reduction and re-structuring of the oxidized recycled powders back to pure metallic NdFeB with the correct microstructure.

**Acknowledgements**

This research was funded by the european project INSPIRES (INtelligent and Sustainable Processing of Innovative Rare-Earth MagnetS) supported by the EIT Raw Materials and co-funded by the European Union.

**References**

[1] *Critical Raw Materials Resilience: Charting a Path towards greater Security and Sustainability*, 2020.

[2] A. Walton, H. Yi, N. A. Rowson, J. D. Speight, V. S. J. Mann, R. S. Sheridan, A. Bradshaw, I. R. Harris, A. J. Williams, *J. Clean. Prod.* 104 (2015) 236–241.

# A Comprehensive TEM Investigation of FeRh Alloy in Antiferromagnetic State

*E. Adabifiroozjaei[1], F. Maccari[1], L. Schäfer[1], N. Shayanfar[1], I. Dirba[1], R. Winkler[1], T. Jiang[1], O. Recalde[1], A. Zintler, A. Chirkova[2], L. Pfeuffer[2], A. Kovacs[3], M. Gruner[4], K. Skokov[2], O. Gutfleisch[2], and L. Molina Luna[1]*

[1]*Advanced Electron Microscopy, Department of Materials- and Earth Sciences, Technical University of Darmstadt, Germany*

[2]*Functional Materials, Department of Materials- and Earth Sciences, Technical University of Darmstadt, Darmstadt, Germany*

[3]*Ernst Ruska-Centre for Microscopy and Spectroscopy with Electrons and Peter Grünberg Institute, Forschungszentrum Jülich, 52425 Jülich, Germany*

[4]*Faculty of Physics and Center for Nanointegration Duisburg–Essen (CENIDE), University of Duisburg–Essen, Duisburg 47057, Germany*

*e.adabifiroozjaei@aem.tu-darmstadt.de

Fe50Rh50 alloys are known to have a B2 ordered structure with an antiferromagnetic (AFM) to ferromagnetic (FM) transition at near room temperature. The transition is isostructural with about 1% change in volume and is accompanied by giant magnetoresistive and magnetocaloric effects [1]. The majority of the previously reported works do not provide extensive local atomic structure investigations. Thus, the structural characteristics of the alloy at the AFM or FM states remains controversial [2]. Since FeRh alloys can be considered as amember of betta alloy series (CdAu, TiNi, Fe-C, etc…), it is expected that they present a premartensite structure followed by a martensite structure upon cooling at cryogenic temperature[3]. The martensite was also predicted by extensive first principal calculations[2,4]. However, so far, no evidence has been given regarding the formation of either pre-martensite or martensite structures in the Fe50Rh50 alloy. Therefore, in the present work, we use various TEM techniques (CTEM, HRTEM, STEM (HAADF), and EDS) to investigate the FeRh 50/50 alloy nanostructure locally. Our HRTEM results clearly show systematic presence of modulations along three principal directions ([001], [-110], and [111]), while the overall structure of the alloy still match perfectly with the B2 structure. The modulations are present along certain reflexes (100 and 110), as is confirmed by further HAADF-STEM imaging. High resultion HAADF data also showed that the modulations are caused by displacement of Fe and Rh atoms. Such displacement is belived to be caused by phonon softening along [110]. We confirmed this using dnesity functional theory (DFT) calculation. From our results, we conclude that the B2 alloy of FeRh 50/50 at the AFM state possess a pre-martensite structure.

**Acknowledgements**

We acknowledge financial support by the Deutsche Forschungsgemeinschaft (DFG) within the CRC/TRR 270 (Project-ID 405553726).

**References**

[1] A. Chirkova, F. Bittner, K. Nenkov, N. V. Baranov, L. Schultz, K. Nielsch, T. G. Woodcock, *Acta Mater.* **2017**, *131*, 31.

[2] M. Wolloch, M. E. Gruner, W. Keune, P. Mohn, J. Redinger, F. Hofer, D. Suess, R. Podloucky, J. Landers, S. Salamon, F. Scheibel, D. Spoddig, R. Witte, B. Roldan Cuenya, O. Gutfleisch, M. Y. Hu, J. Zhao, T. Toellner, E. E. Alp, M. Siewert, P. Entel, R. Pentcheva, H. Wende, *Phys. Rev. B* **2016**, *94*, 174435.

[3] K. Otsuka, X. Ren, *Prog. Mater. Sci.* **2005**, *50*, 511.

[4] J. Kim, R. Ramesh, N. Kioussis, *Phys. Rev. B* **2016**, *94*, 180407.

# Data-Mining Approach to the Search for Novel Rare-Earth-Free High-Performance Permanent Magnets

*Alena Vishina[1]*, Olle Eriksson[1,2], Heike C. Herper[1]*

[1] *Department of Physics and Astronomy, Uppsala University, Box 516, SE-75120 Uppsala, Sweden*
[2] *School of Science and Technology, Örebro University, SE-701 82 Örebro, Sweden*
*alena.vishina@physics.uu.se

With an increasing concern for climate change, many countries have transition to green energy sources and electric transportation as one of their priorities. It causes a spike in demand for high-performance permanent magnets (PM), which are used in large amounts in wind turbines and electric motors. At the same time, all the high-performance PM currently in use contain rare-earth (RE) elements, which brings another set of problems. REs are subject to high supply risks, with their prices being high and unpredictable, are not recycled efficiently, and are often mined with a high risk for the environment. Hence, there is a growing interest in finding new RE-free or RE-lean magnetic materials, which should still demonstrate similar high performance.

We have introduced high-throughput and data mining density functional theory (DFT) approach to the search for RE-free PMs [1]. Filtering through the Inorganic Crystal Structure Database (ICSD) of materials preciously synthesized experimentally, we are looking for the materials with ferromagnetic (FM) ground state, high saturation magnetization ($M_S$ > 0.5 T), high uniaxial magnetocrystalline anisotropy (> 1 MJ/m$^3$), and high Curie temperature ($T_C$ > 400 K). When a promising material is found, an attempt is often made to improve its characteristic by various element substitution.

A combination of a *3d*- and a *p*-element of the periodic table was considered in our recent work [2]. Here, we looked specifically into three of the materials found as a result of the high-throughput search in an attempt to improve some of their properties. Those are $Fe_2C$, $Mn_2MoB_4$, and $Mn_2WB_4$ (black squares in Fig. 1). For example, $Fe_2C$, with its good magnetic characteristics of $M_S$ = 1.31 T, MAE = 0.8 MJ/m$^3$, and $T_C$ = 900 K, is reported to be unstable. We have attempted to improve its stability by alloying it with the other magnetic elements. The optimal additional element was found to be Mn, as it improved the stability, along with all the magnetic characteristics (see the purple pentahedron in Fig. 1) [2].

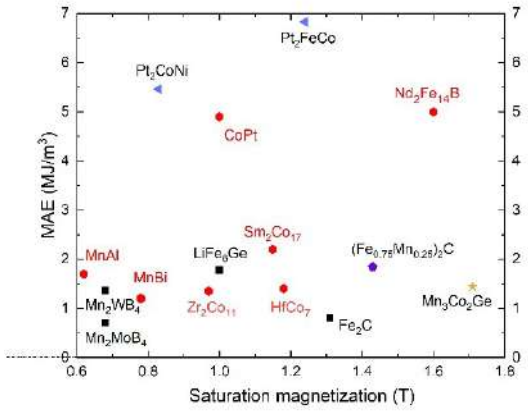



Figure 1: MAE and saturation magnetization of the materials discovered as a result of our data-mining searches. For comparison, red hexagons show the data for some of the currently used RE permanent magnets and several RE-free PM

## References

[1] A. Vishina, O. Yu. Vekilova, T. Björkman, A. Bergman, H. C. Herper and O. Eriksson Phys. Rev. B 101, 094407 (2020)
[2] A. Vishina, O. Eriksson, H.C. Herper, Materials Research Letters, 11, 76 (2022)

# Direct synthesis of highly ordered $L1_0$-FeNi nanoparticles from crystalline Ni-Nitroprusside complexes

*G. Varvaro[1]*, P. Imperatori[1], S. Laureti[1], D. Peddis[2,1], F. Locardi[2], M. Ferretti[2], C. Cannas[3], M. Sanna Angotzic[3], N. Yaacoub[4], and A. Capobianchi[1]**

[1] *CNR-ISM, nM²-Lab, Monterotondo Scalo (Roma), 00015, Italy*
[2] *Un- Genova, DCCI, nM²-Lab, Via Dodecaneso 31, 1-16146 Genova, Italy*
[3] *Un. Cagliari and INSTM, DSCG, S.S. 554, bivio per Sestu, 09042, Monserrato, Italy*
[4] *Le Mans Université, CNRS UMR-6283, Avenue O. Messiaen, Le Mans, 72085, France Le Mans*
*gaspare.varvaro@ism.cnr.it; aldo.capobianhi@ism.cnr.it

The $L1_0$-FeNi binary alloy is a promising candidate for next generation rare earth-free permanent magnets (PMs) [1], which can revolutionize the high-performance PM market currently dominated by the Nd-Fe-B [2]. However, the fabrication of the $L1_0$ phase is extremely challenging owing to the low atomic mobility below the chemical order/disorder transition temperature that kinetically limits the formation of the $L1_0$ phase [3]. Despite many efforts, the experimental results are still far from the theoretical predictions and the proposed approaches mainly involve complex and expensive protocols, which cannot be easily scaled-up for bulk production and/or result in a low proportion of the $L1_0$ phase [3].

To overcome current limitations, we exploited an effective and easily scaled-up chemical synthesis method, already successfully applied for other $L1_0$ alloys [4], which is based on the use of crystalline precursor complexes consisting of an ordered arrangement of the elements on alternating atomic planes that resembles the atomic arrangement of the $L1_0$ structure. The perfect atomic order of the precursors allows reducing the energy required to order the atoms thus driving the formation of the $L1_0$ phase that can be obtained by low- temperature reduction in $H_2$ atmosphere. To apply this concept to the $L1_0$ FeNi alloy, crystalline Ni-Nitroprusside complexes with a 1:1 ratio of Fe and Ni were used as precursors [5]. Carbon coated FeNi alloy nanoparticles (20 – 120 nm) with a >55 % of $L1_0$ phase (as determined by Mossbauer analysis), quite high coercivity (up to 65 mT) and large saturation magnetization (~ 135 $Am^2/kg$, close to the bulk value) were obtained in the best experimental conditions. Despite the coercivity is still far from optimal for a high-performance permanent magnet, the results clearly prove the effectiveness and high potential of the developed strategy, which can be exploited, after further optimization, for mass production of highly ordered $L1_0$-FeNi nanoparticles for next generation critical-element-free permanent magnets.

## References

[1] J. Cui et al., Acta Mater. **158** (2018), 11.
[2] J. M. D. Coey, Engineering **6** (2020), 119.
[3] S. Mandal et al., Crit. Rev. Solid State Mater. Sci. (2022).
[4] G. Varvaro et al., J. Alloys Compd. **846** (2020), 846.
[5] A. Capobianchi, P. Imperatori and G. Varvaro, Patent Application Filed #102022000024852 (2022).

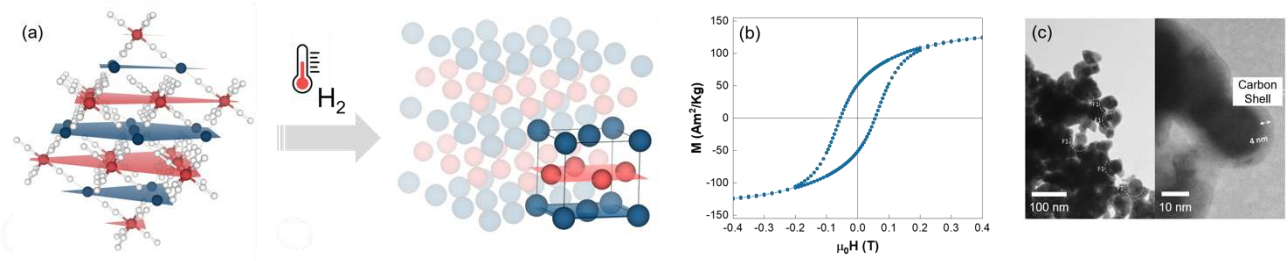



Figure: (a) Schematic representation of the synthesis process: from crystalline Ni-Nitroprusside complexes (left) to $L1_0$-FeNi alloy (right). (b) Representative M(H) loop of FeNi powders and (c) corresponding TEM image.

# Effect of aging on the magnetic and physical properties of consolidated Mn-Al-C bulk magnets

*Semih Ener[1]*, Ulysse Rocabert[1], Fernando Maccari[1], Alex Aubert[1], Guixiang Qin[2], Andres Martin-Cid[3], Martin Krengel[4], Alberto Bollero[3] and Oliver Gutfleisch[1]*

[1] *Functional Materials, Material and Geoscience Department, Technical University of Darmstadt, Alarich-Weiss-Str. 16, Darmstadt, 64287, Germany,*

[2] *Less Common Metal Ltd. Unit 2 Hooton Park, North Road, Ellesmere Port, Cheshire CH65 1BL, United Kingdom*

[3] *Group of Permanent Magnets and Applications, IMDEA-Nanociencia, Madrid, 28049, Spain*

[4] *WILO SE, Wilopark 1, 44267 Dortmund, Germany*

*semih.ener@tu-darmstadt.de

Rare earth (RE)-free magnetic materials may play a vey important role towards the reduction of Europe's dependency to the critical elements like light- and heavy-rare-earths. For tackling this problem a European project consortium with 21 partners including leading industries [1]. In this project we carry out studies on RE-free hard magnetic materials such as Sr-hexaferrites and Mn-Al-C systems. Mn-Al-C is one of the promising material systems which fits to the concept of the gap magnets. The tetragonal τ-MnAl phase has the potential of theoretical energy products approximately of 100 kJ/m$^3$ [2].

In this work, we investigated the effect of different aging conditions on the structural, microstructural, chemical and magnetic properties of the hot compacted $Mn_{56.2}Al_{42.3}C_{1.5}$ samples. High phase purity Mn-Al-C powders were prepared by the the company Less Common Metal (LCM) using conventional casting following a KEK milling and classification process. Powders of < 300 µm (particle size) were used for the hot compaction experiments. A series of consolidation trials were carried out at different pressure and temperature conditions to end with bulk Mn-Al-C magnets. The details towards an optimization of the process will be discussed in this presentation. Structural (including phase evolution), microstructural and magnetic studies were carried out on the consolidated samples.

Together with the magnetic and structural characterization, investigations on the corrosion behavior have been assessed. Figure shows voltammetry measurement results under two different chemical environments with different pH levels. These results indicate that the τ-MnAl phase oxidizes relatively easier in alcaline conditions. Considering the pH level of tab water (between 6.5 and 8.6) evaluating the corrosion sensibility of this material seems relevant prior to utilization in aqueous environment. Similar to the consolidated samples, structural, microstructural and magnetic characterizations of the aged samples were carried out. The results will be discussed in detail in view of future applications of this RE-free permanent magnet alternative.

## Acknowledgements

Authors acknowledge support from EU Horizon 2020 research and innovation programme through the project PASSENGER (grant agreement Nº 101003914).

## References

[1] EU H2020 *PASSENGER* Project: https://passenger-project.eu/

[2] J.M.D. Coey "New permanent magnets; manganese compounds", J. Phys.: Condens. Matter 26 (2014) 064211.

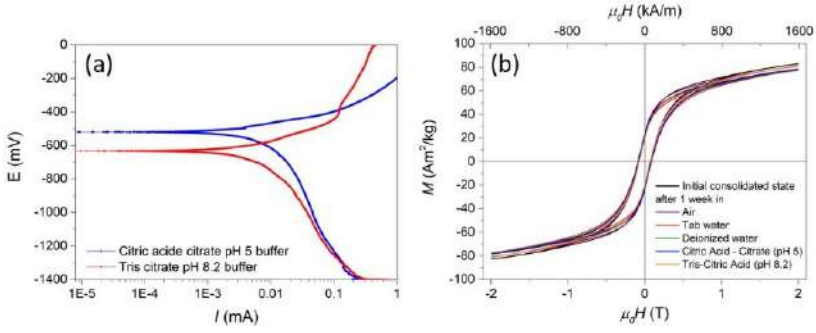



Figure: (a) Linear sweep voltammetry and (b) room temperature hysteresis measurements of $Mn_{56.2}Al_{42.3}C_{1.5}$ samples.

# Ferrite-Based Magnets by High Pressure Consolidation

*C. de Julián Fernández[1]*, B. Belec[2,1], D. Delmonte[1], R. Cabassi[1], E. Giglioli[1], B. Muzzi[3,4], M. Albino[3,4], A. Učakar[5], C. Sangregorio[3], F. Albertini[1], P. Jenuš[5]*

[1] *IMEM- CNR, Parco Area delle Scienze 37/A, 43124 Parma, Italy*
[2] *University of Nova Gorica, Vipavska 13, Vipavska, Slovenia*
[3] *ICCOM – CNR, Via Madonna del Piano 10, Sesto Fiorentino, Italy*
[4] *Dept. of Chemistry "U. Schiff", University of Florence and INSTM, Via della Lastruccia 3, Sesto Fiorentino, Italy*
[5] *Dept. Nanostructured Materials, Jožef Stefan Institute, Jamova 39, Ljubljana, Slovenia*
*cesar.dejulian@imem.cnr.it

M-type hexaferrite magnets constitute together the rare-earth magnets the most employed magnets in the world. Even if ferrite magnets have smaller energy product and lower magnetization saturation than rare-earth permanent magnets (PMs), currently ferrite magnets represent the most widely used PMs, covering 80% of the PM market production [1,2]. Recently, a strong effort is being performed to improve the magnetic properties of ferrites with the scope of substituting partially rare-earth magnets. Most of the strategies involve the nanostructuration of the ferrites and/or the development of hybrid compounds [3]. A bottleneck for the production of these magnets is that standard sintering process requires high temperatures and oxidizing atmosphere that produces the destruction of the nanostructure and several chemical changes. Different novel strategies as out-of-equilibrium or cold sintering processes are mainly considered[2,4].

In our presentation we will show the production of dense ferrite-based magnets by high pressure multi-Anvil press at low temperatures. This press applies quasi-isotropic pressures in the range up to 20 GPa and temperatures up to 1200°C. We demonstrate that the consolidation of micrometric hexaferrite ($SrFe_{12}O_{19}$) powders is possible at temperatures below 1000°C, below the standard sintering temperatures. In addition, dense hybrid magnets were obtained composed of micrometric hexaferrites and soft high magnetization metal Fe or FeCo NPs applying pressures up to 6 GPa and low temperatures (250°C). A deep study, including structural, morphological and magnetic characterizations, has been performed to determine the influence of the temperature and pressure consolidation conditions in the properties of these novel magnets. Magnetic characterizations show that hybrid magnets exhibit larger magnetization than ferrites and single step hysteresis loops. Single Point Detection characterizations indicate that the anisotropy field of the hybrid magnets is similar to that of the ferrite magnets. These results suggest that the two moieties componing the high pressure consolidated magnets have similar properties than the original micro and nano powders, but they are magnetic coupled during the reversal process. High pressure consolidation appears as a promising technique to obtain nano-based metal-oxide hybrid magnets with promising hard properties.

**Acknowledgements**

This research was supported by EU- H2020 AMPHIBIAN Project (H2020-NMBP-2016-720853) and by the European Union - NextGenerationEU (National Sustainable Mobility Center CN00000023, Italian Ministry of University and Research Decree n. 1033 - 17/06/2022, Spoke 11 - Innovative Materials & Lightweighting).

**References**

[1] R. C. Pullar, *Progr. Mater. Sci.* **57** (2012) 1191
[2] C. Granados-Miralles, P. Jenuš, *J. Phys. D Appl. Phys.* **54** (2021) 303001.
[3] C. de Julián Fernández *et al. J. Phys. D Appl. Phys*. **54** (2021), 15300
[4] A. Serrano *et al., J. European Cer. Soc*. **42** (2022) 1014.

# EFFICIENT RECYCLING OF STRONTIUM FERRITE MAGNETS FROM END-OF-LIFE APPLIANCES

*A. Berja[1*], C. Granados-Miralles[1], J.F. Fernández[1] and A. Quesada[1]*

[1] *Instituto de Cerámica y Vidrio (CSIC), 28049, Madrid, Spain*

*alba.berja@csic.es

Strontium ferrite ($SrFe_2O_{19}$) permanent magnets account today for 80% in volume of the market due to their excellent properties and important advantages over other families of magnets [1,2]. These permanent magnets are a great alternative to rare-earth (REE) permanent magnets, well-known for being critical raw elements, whose extraction produce an enormous impact on the environment. Therefore, the development of these REE-free magnets is of great significance nowadays, as they are the most natural candidate for substitution of REE in a variety of applications such as electric motors, sensors, and home appliances [3]. Its demand is expected to increase in the next 5-10 years, so it is crucial to ensure the sustainability of the ferrite magnet value chain. This work is based on the recycling of strontium ferrite magnets obtained from end-of-life home appliances in Europe, as a first step to implementing a circular economy loop in the ferrite magnet value chain in Europe.

We have established a succesful process for recycling sintered Strontium Ferrite magnets back into powder based on milling and annealing cycles that achieve the reduction of the particle size and competitive magnetic properties after a final thermal annealing. In addition, we conditioned the recycled powder for fabricating the first batch of recycled ferrite magnets by injection moulding, obtaining properties similar to commercial injection moulded ferrite magnets. The laboratory-scale method used to obtain the recycled powder is easily implementable and up-scalable at industrial level.

## Acknowledgements

This research was funded by the european project INSPIRES (INtelligent and Sustainable Processing of Innovative Rare-Earth MagnetS) supported by the EIT Raw Materials and co-funded by the European Union.

## References

[1] Pullar, R. C. (2012). Hexagonal ferrites: A review of the synthesis, properties and applications of hexaferrite ceramics. *Progress in Materials Science*, *57*(7), 1191–1334.

[2] de Julian Fernandez, C., Sangregorio, C., de la Figuera, J., Belec, B., Makovec, D., & Quesada, A. (2020). Topical Review: Progress and Prospects of Hard Hexaferrites for Permanent Magnet Applications. *Journal of Physics D: Applied Physics*.

[3] de Campos, M. F., & Rodrigues, D. (2016). High Technology Applications of Barium and Strontium Ferrite Magnets. *Materials Science Forum*, *881*, 134–139.

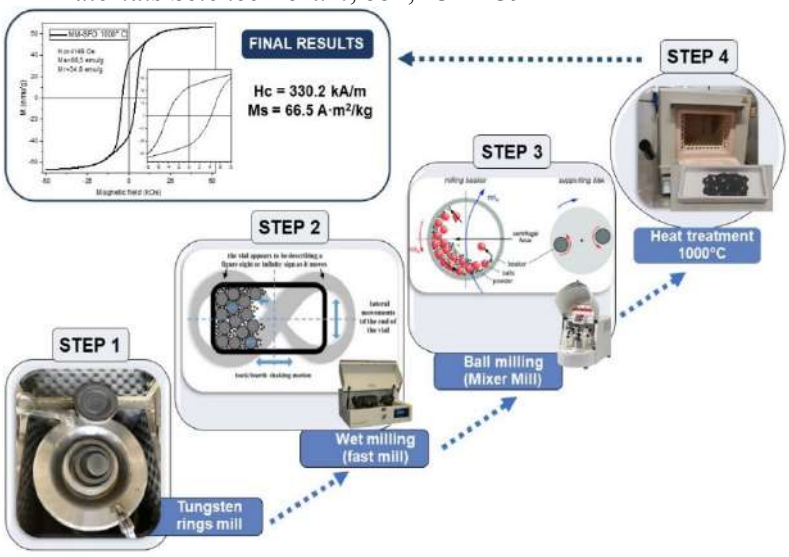



Figure 1: Image showing the steps of the recycling proccess of the strontium ferrite magnets

.

# SYMPOSIUM 01.
## MAGNETIC MATERIALS FOR ENERGY APPLICATIONS.
## S1. ORAL POSTERS

# Entropy Change Reversibility Near The Triple Point

*T. Samanta[1]*, C. Taake[1], and L. Caron[1, 2]*

[1] *Faculty of Physics, Bielefeld University, PO Box 100131, D-33501 Bielefeld, Germany*

[2] *Helmholtz-Zentrum Berlin für Materialien und Energie, Berlin 12489, Germany*

* tsamanta@physik.uni-bielefeld.de

Energy-efficient greener magnetic refrigeration technology based on magnetocaloric effect is sought for its applicability for over two decades. However, the unavoidable thermal hysteresis loss often reduces the cyclic magnetic cooling performance significantly for magnetocaloric materials exhibiting a first-order phase transition. Therefore, a better reversible magnetocaloric effect is desirable for practical applications (particularly in the 2 T magnetic field range). In this context, the nature of the phase transition responsible for the magnetocaloric effect in $MnNi_{1-x}Co_xGe_{0.97}Al_{0.03}$ ($x$=0.20-0.50) compounds has been studied.

Isostructural substitution of MnCoGe (Collinear ferromagnet) with MnNiGe (Spiral antiferromagnet) can create a fascinating phase transition scenario with a magnetic instability near 50:50 substitution [1]. The magnetic phase transition remains second-order and the structural transition occurs in the paramagnetic state. Small amounts of Al substituting Ge can lower the structural phase transition temperature, resulting in a coupled first-order magnetostructural transition in $MnNi_{1-x}Co_xGe_{0.97}Al_{0.03}$ ($x$=0.20-0.50) near room temperature. In this system a magnetic instability combined with a magnetostructural transition results in a triple point, where three different types of first-order magnetic phase transition boundaries coincide [(i) magnetostructural transition between ferromagnetic (orthorhombic)-paramagnetic (hexagonal), (ii) magnetostructural transition between antiferromagnetic (orthorhombic)-paramagnetic (hexagonal) and (iii) magnetoelastic transition between antiferromagnetic (orthorhombic)-ferromagnetic (orthorhombic)]. Most interestingly, compositions in the vicinity of the triple point are found to have an enhanced reversible low-field magnetocaloric effect ($|\Delta S_{rev}|$=6.9 J/kg K for $\Delta\mu_0 H$=2 T). Moreover, $|\Delta S_{rev}|$ reaches values of 17.2 and 24.5 J/kg K for $\Delta\mu_0 H$=5 and 7 T, respectively, which is rather rare in the studied class of materials and also comparable or even larger than observed in other well-known giant magnetocaloric materials.

It has been found that magnetic frustration plays a most crucial role in the reversibility of the $|\Delta S_{rev}|$. A sudden release of magnetic frustration due to composition-dependent transformation from a frustrated antiferromagnetic to an ordered ferromagnetic state is responsible for a better $|\Delta S_{rev}|$ in $MnNi_{1-x}Co_xGe_{0.97}Al_{0.03}$ near the triple point. It can be inferred from the fundamental insights behind the observed reversibility in $|\Delta S_{rev}|$ that a similar reversibility is also expected for other functional properties near the triple point and it would be likely invariant irrespective of material classes.

**Acknowledgements**

This work was carried out in the framework of the Joint Lab BiBer and was supported by the BMBF joint project DiProMag FKZ 13XP5 120B.

**References**

[1] S. Nizioł, A. Bombik, W. Bażela, A. Szytuła, D. Fruchart, *Solid State Commun*. **42** (1982) 79.

# Up-Scaled Coercivity Development Of Cast And Milled MnAlC Alloy

*Jorge Vergara-Ortega[1]*, Andres Martin-Cid[1], Carla Muñoz-Rodríguez[1], Ester M. Palmero[1], Guixiang Qin[2], Ian Higgins[2], Chris Hall[2], Enrico Forlin[3], Lorenzo Bianchin[3], Semih Ener[4], Oliver Gutfleisch[4], Alberto Bollero[1].*

[1] *Group of Permanent Magnets and Applications, IMDEA Nanociencia, C/ Faraday 9, Madrid 28049, Spain*
[2] *Less Common Metals Ltd. Unit 2 Hooton Park, North Road, Ellesmere Port, Cheshire CH65 1BL, U. K.*
[3] *MBN Nanomaterialia S.p.A., Via G. Bortolan 42, 31050 Vascon di Carbonera, TV, Italy*
[4] *Technical University of Darmstadt, Alarich-Weiss-Str. 16, Darmstadt, 64287, Germany*
*jorge.vergara@imdea.org

τ-phase MnAl is an excellent rare earth-free PM candidate [1] due to its properties such as an estimated maximum energy product of $(BH)_{max}$ = 12 MGOe [2] at room temperature and its density of 5.2 g/cm$^3$ (by comparison with 7.6 g/cm$^3$ for $Nd_2Fe_{14}B$).

Using casting technology and then pulverized by KEK milling, Less Common Metals (LCM) was able to produce powder with particle size under 90 μm and 300 μm, respectively. This powder produced in an industrial environment was confirmed to show a nearly 100% content of τ-MnAlC by VSM and XRD measurements. IMDEA's "flash-milling" method [3] was used in a first set of experiments, considering milling times between 30 to 840 seconds. This processing allowed a two-fold increase in coercivity respect to the precursor, maintaining a remanence of 34 Am$^2$kg$^{-1}$. High-energy ball milling (HEBM) tests were performed by a proprietary process of MBN at a pilot scale (limited at 0.5 kg per test), demonstrating the viability of up-scaling the process and increasing the precursor particle size, achieving a maximum coercivity of 0.35 T (i.e. 4.4-fold increase respect to the starting material) in times under 1 hour. Considering previous results [4,5], powders were subsequently annealed at a moderate temperature (550ºC for 10 min) under $N_2$ atmosphere to recrystallize τ-phase and develop β-phase to optimize the magnetic properties.

Corrosion assessment was done in distilled water at room temperature and heating in air at 550ºC, showing promising results after 84 days in water and after 10 min of heat treatment in air.

## Acknowledgements

Authors acknowledge support from EU Horizon 2020 research and innovation programme through the project *PASSENGER* (grant agreement Nº 101003914). IMDEA thanks support from MICINN through *NEXUS* project (PID2020-11521RB-C21). E.M.P. acknowledges support from AEI - Juan de la Cierva - Incorporación program (IJC2020-043011-I/MCIN/AEI/10.13039/501100011033) and NextGeneration EU/PRTR.

## References

[1] Y. Jia *et al.*, *Acta Mater.* **245** (2023) 118654.
[2] J.H. Park *et al.*, *J. Appl. Phys.* **107** (2010) 09A731.
[3] J. Rial *et al., J. Phys. D. Appl. Phys.* **50** (2017) 105004.
[4] J. Rial, E.M. Palmero, A. Bollero, *Engineering* **6** (2020) 173-177.
[5] J. Rial *et al., Acta. Mater.* **157** (2018) 42-45.

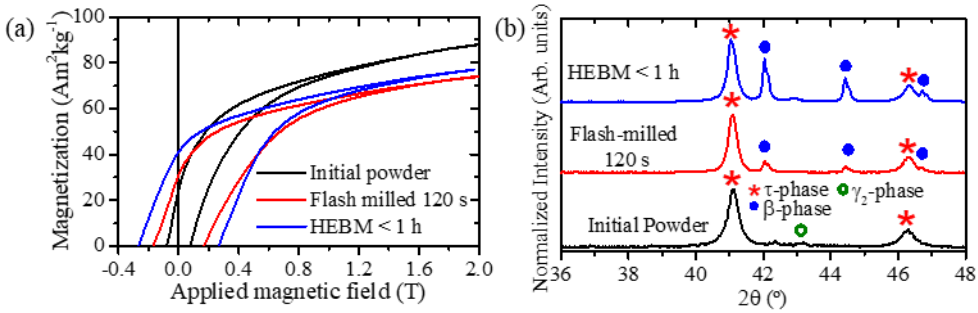



Figure 1: (a) First and second quadrants of the VSM hysteresis loop; (b) XRD patterns for the initial powder, powder flash-milled for 120 seconds, and powder obtained via up-scaled HEBM for a milling time below 1h. Milled samples after annealing at 550ºC.

# Induced Magnetism in Hybrid Improper Ferroelectric $Sr_3Sn_2O_7$ Replacing Sn by Magnetic Atoms

*J. Blasco[1]*, V. Cuartero[1,2], D. Gracia[1], J. A. Rodriguez-Velamazán[3], I. Puente-Orench[3], J. Herrero-Martín[4], S. Lafuerza[1], J. L. García-Muñoz[5], and G. Subías[1]*

[1] *Instituto de Nanociencia y Materiales de Aragón, Dpto. de Física de la Materia Condensada, CSIC-Universidad de Zaragoza, C/ Pedro Cerbuna 12, Zaragoza, Spain*

[2] *Dpto. de Ciencia y Tecnología de Materiales y Fluidos, Universidad de Zaragoza, C/ María de Luna 3, Zaragoza, Spain*

[3] *Institut Laue-Langevin, Grenoble Cedex 38042, France*

[4] *ALBA Synchrotron Light Source, Carrer de la Llum 2-26, Cerdanyola del Vallès, Barcelona, Spain*

[5] *Institut de Ciència de Materials de Barcelona, ICMAB-CSIC, Campus UAB, Bellaterra, Spain*

*jbc@unizar.es

Nowadays, the study of hybrid improper ferroelectric (HIF) materials is increasing considerably as a way to obtain new multiferroic compounds [1], which are promising materials to obtain ultra-low-power electronic devices. One of the latest HIF materials discovered is $Sr_3Sn_2O_7$ [2]. This compound belongs to the family of $(AO)(ABO3)_2$ Ruddlesden-Popper (R-P) perovskite oxides and it is very interesting because it exhibits both ferroelasticity and ferroelectricity with a relatively low coercive field in single-crystals [2]. In order to extend the functionality of this material, we have explored the possibility of inducing magnetism by replacing Sn with magnetic atoms. We have prepared $Sr_{3-x}La_xSn_{2-x}M_xO_7$ (M = Cr, Fe, Mn) compounds up to x = 1, where the simultaneous replacement of Sr by La allows preserving the electrical neutrality in the unit cell.

The new compounds are single phase showing solid solutions and the patterns can be refined using the $A2_1am$ space group of the parent compound [2], that is, the crystallographic structure remains non-centrosymmetric. These samples show a paramagnetic behavior at high temperature obeying the Curie-Weiss law. Negative values of the Weiss constant for M = Fe or Cr in the paramagnetic region suggest antiferromagnetic (AFM) correlations that increase with doping. However, M = Mn samples present positive values of this constant that reveal the occurrence of ferromagnetic correlations.

At low temperatures, a large deviation from the Curie-Weiss law is observed for all samples. Below 6 K, magnetic irreversibility between zero-field-cooled and field-cooled conditions are found for $Sr_{3-x}La_xSn_{2-x}Mn_xO_7$ samples with $x \geq 0.75$, *i.e.* above the percolation limit ($x \approx 0.66$). Isothermal measurements, M(H), reveal a deviation from linear behavior in samples doped with Fe or Cr at 5 K. This result could be associated with an easy polarization of the magnetic moments by an external magnetic field. However, a clear spontaneous magnetization is observed in the M(H) loops for Mn-based samples. Although magnetic saturation is not achieved at 5 T, the magnetic moment values at these fields are close to 3 $\mu_B$/*at. Mn*, suggesting the occurrence of a long range ferromagnetic ordering. The hysteresis loops are supported by X-ray circular magnetic dichroism measurements and the X-ray absorption spectra at the Mn $L_{2,3}$ edges confirm the presence of mostly $Mn^{3+}$ cations in these samples. This result seems to contradict the Goodenough-Kanamori rules that predict AFM interactions between $Mn^{3+}$ ions but it is strongly reminiscent of the ferromagnetism described in simple perovskites when $Mn^{3+}$ is diluted with nonmagnetic ions [3,4]. The present work suggests a similar ferromagnetic vibronic-superexchange mechanism for these R-P phases.

## Acknowledgements

We acknowledge the Spanish Ministerio de Ciencia e Innovación for financial support (Projects PID2021-124734OB-C21 and PID2021-124734OB-C22) co-financed by ERDF from EU. We thank the ILL and SpINS (experiment CRG-2990), and ALBA synchrotron (experiment 2023027292-IHR) for granting beam time.

## References

[1] N. A. Benedek and C. J. Fennie, *Phys. Rev. Lett.* **106** (2011) 107204.
[2] X. Xu *et al.*, *Adv. Funct. Mater.* **30** (2020) 2003623.
[3] J. Blasco *et al.*, *Phys. Rev B* **66** (2002) 174431.
[4] J. B. Goodenough *et al.*, *Sol. State Sci.* **4** (2002) 297.

# Nanocrystalline strontium ferrite permanent magnet material with high coercivity and enhanced thermal stability

Adrián Fernández-Calzado[1], Ester M. Palmero[1], Cristina M. Montero[1], Semih Ener[2], Oliver Gutfleisch[2], Darko Makovec[3], Jean-Marc Grenèche[4], Paolo Bernardi[5], Alberto Bollero[1]

[1] Group of Permanent Magnets and Applications, IMDEA Nanoscience, 28049 Madrid, Spain
[2] Functional Materials, Material Science, TU Darmstadt, 64287 Darmstadt, Germany
[3] Department for Materials Synthesis, Jožef Stefan Institute, Jamova 39, SI-1000 Ljubljana, Slovenia
[4] Institut des Molécules et Matériaux du Mans (IMMM UMR CNRS 6283), Le Mans Université, Avenue Olivier Messiaen, Le Mans F-72085, Cedex 9, France
[5] Industrie ILPEA Spa, Viale Industria 887, 21020 Malgesso-Varese, Italy
*adrian.fernandez@imdea.org

Hard ferrite magnets might find very soon an increased number of market applications in the electromobility sector based on improved magnetic properties combined with the possibility of making a redesign to take full advantage of the new achievements [1]. This possibility will contribute to aliviate the volatile situation of Europe's dependence on critical raw materials in the permanent magnet sector.

This study shows the possibility of developing high-coercivity strontium ferrite powder with an excellent performance at low temperature and with no need to use critical raw elements (e.g. La and Co). This has been possible starting from a commercial ferrite and nanostructuring it (Figs. 1a,b) by the self-developed "flash milling" method [2,3]. The creation of a Sr-ferrite/hematite nanocomposite has led to a room temperature coercivity above 475 kA/m with increasing the milling time (up to 30 min) (Fig. 1c). Addition of $Fe_2O_3$ powder prior to milling has allowed to reduce the required processing time and to end with a low-temperature high-coercivity nanocomposite: 430 kA/m measured at -100ºC (Fig. 1d), thus opening the path for future applications.

**Acknowledgements**

Authors acknowledge support from EU's Horizon 2020 research and innovation program through the project PASSENGER (GA Nº101003914). IMDEA thanks support from MICINN through the NEXUS project (PID2020-11521RB-C21). E.M.P. acknowledges support from AEI through Juan de la Cierva Incorporación program (IJC2020-043011-I/MCIN/AEI/10.13039/501100011033) and EU by NextGeneration EU/PRTR.

**References**

[1] A. Bollero and E.M. Palmero, "Recent advances in hard - ferrite magnets" in *Modern Permanent Magnets*, J.J. Croat and J. Ormerod, Eds. Elsevier (2022) 65-112.
[2] F.J. Pedrosa *et al.*, RSC Adv. **6** (2016) 87282-87287.
[3] A. Bollero et al., "Ferrite type materials and process for the production thereof". Publication number: WO/2018/211121. Int. Application nº: PCT/EP2018/063222, 2018.

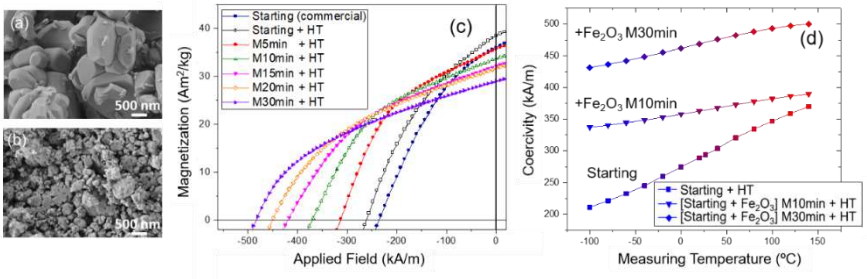



Figure 1: (a) and (b) SEM images of starting (commercial) powder and this same powder after milling for 40 min. (c) Room temperature VSM hysteresis loops measured for the precursor powder and that after milling (different durations) and heat treatment (1000ºC). Evolution of coercivity between -100ºC and 140ºC for the starting (commercial) precursor and the synthesized Sr-ferrite/hematite nanocomposites.

# High-entropy alloys as the thermoelectric materials

*A. Dzubinska[1]*, M. Varga[1], M. Reiffers[2] and R. Varga[1]*

*[1] CPM-TIP, University of Pavol Jozef Safarik, Tr. SNP 1, 040 11 Kosice, Slovakia*

*[2] Fac. of Hum. and Nat. Sciences, University of Presov, Ul. 17. novembra 1, 080 01 Presov, Slovakia*

*andrea.dzubinska@upjs.sk

Preparing new progressive materials is one of the first steps towards developing materials research at the edge of our knowledge, using the latest knowledge of modern physics and chemistry. Knowing the behaviour of materials under certain physical conditions enables the modification of their properties in order to search for application possibilities gradually. Our work focuses primarily on high-entropic alloys that will fulfil the function of thermoelectric materials. The demand for TE has been increasing over the past two decades. Developing industrially viable materials for global use is a challenge for research workers. Thermoelectric materials must meet several criteria to be suitable for this type of application [1].

Entropy is a physical quantity that measures the degree of chaos. High-entropic alloying is a new and effective strategy for reducing the lattice thermal conductivity of thermoelectric materials. Generally, high-entropy alloys contain five or more elements in a relatively high concentration (5-35 at. %). In this research, we need to examine the suitability of our chosen compositions – TiNiSb and HfNiSb for individual materials or the use of experimental methodologies to which we have access. Current knowledge shows [e.g. 1-3] that this class of materials is suitable for practice for their, e.g. mechanical properties.

High-entropic alloys offer great complexity due to severe lattice distortions, point defects, or precipitation of secondary phases due to efficient phonon scattering while maintaining the high mobility of the numerous conduction electrons. In addition to efficient means for phonon scattering, high-entropy alloys usually have high symmetry crystal structures such as face-centred cubic (*fcc*), centre-centred cubic (*bcc*), or in some cases, hexagonal closest-packed (*hcp*) structures.

In this work, we have studied two compositions of Heusler alloy TiNiSb and HfNiSb. The scanning electron microscopy with EDX function confirms stochiometric composition. From the XRD data point of view, HfNiSb alloy crystallizes in an orthorhombic *Pnma* space group that has been published previously [4], and TiNiSb has a cubic *F-43m* space group corresponding to previously published Heusler compounds [5]. The magnetic measurements, and thermoelectric data will be shown and discussed.

The authors expects that the interdisciplinarity in this work will lead to developing and improving technologies, and their implementation in practice will be easier and possible in earlier times.

## Acknowledgements

This research work was supported partially by VEGA 1/0404/21; VEGA 1/0705/20; APVV-16-0079 and VVGS-2022-2117.

## References

[1] A. Karati, M. Nagini, S. Ghosh, R. Shabadi, K. G. Pradeep, R. Ch. Malik, B. S. Murty, U. V. Varadaraju, *Nature Scientific Reports* **9** (2019), 5331.

[2] E. P. George, D. Raabe, R. O. Ritchie, *Nature Review Materials* **4** (2019), 515-534.

[3] Y. Pei, X. Shi, A. La Londe, H. Wang, L. Chen, G. J. Snyder, *Nature* **473** (2011), 66.

[4] G. Melnyk, E. Bauer, P. Rogl, R. Skolozdra, E. Seidl, *Journal of Alloys and Compounds* **296** (2000), 1-2.

[5] A. Szytula, Z. Tomkowicz, M. Turowski, *Acta Physica Polonica A* **44** (1973), 147-149.

# Control of Co Occupied Sites in La–Co Substituted M-type Ferrites

*H. Nakamura[1]*, H. Ohta[2], R. Kobayashi[1], T. Waki[1], Y. Tabata[1], H. Ikeno[3], C. Mény[4]*

[1] *Department of Materials Science and Engineering, Kyoto University, Kyoto 606-8501, Japan*

[2] *Department of Molecular Chemistry and Biochemistry, Faculty of Science and Engineering, Doshisha University, Kyotanabe 610-0321, Japan*

[3] *Department of Materials Science, Graduate School of Engineering, Osaka Metropolitan University, Sakai, Osaka, 599-8570, Japan*

[4] *Institut de Physique et Chimie des Matériaux de Strasbourg (IPCMS), UMR7504, CNRS-Université de Strasbourg, 23 rue du Loess, F-67034 Strasbourg Cedex 02, France*

*nakamura.hiroyuki.2w@kyoto-u.ac.jp

Base materials of hard ferrite magnets are ferrimagnetic M-type ferrites $A$Fe$_{12}$O$_{19}$ ($A$ = Sr, Ba, Ca, …) with five crystallographycally different Fe sites; majority-spin 12k, 2a and 2b sites and minority-spin 4f$_1$ and 4f$_2$ sites. When the Fe sites are replaced with a small amount of Co and the $A$ site with La for charge compensation, the coercivity and magnetization increase together, and the magnets have been commercialized as high-performance ferrite magnets [1]. The performance is improved because Co has an orbital component in the magnetic moment and mainly occupies minority-spin sites. However, it is known that the degree of performance improvement associated with Co substitution depends on the $A$ ion. This is reminiscent of the fact that Co occupied sites or the Co site distribution differs depending on the $A$ ion. Previous studies have shown that Co mainly occupies the 4f$_1$ minority-spin site and the 12k and 2a majority-spin sites [2], but we found that only Co occupying the 4f$_1$ site contributes to the improvement of uniaxial magnetic anisotropy [3]. Therefore, it is expected that the difference in performance for the same amount of Co depending on the $A$ ion is due to the difference in the 4f$_1$ site occupancy of Co. In this study, Co distribution was experimentally evaluated by $^{59}$Co-NMR and Co preferentially occupied sites were evaluated by DFT calculations for systems with different $A$ ions.

La–Co co-substituted M-type ferrites $A$Fe$_{12}$O$_{19}$ ($A$ = Ca, Sr, and Ba, ion size is $Ca^{2+} < Sr^{2+} < Ba^{2+}$) with Co composition around 0.2 were subjected to $^{59}$Co-NMR. The results show that Co occupies the 4f$_1$, 2a, and 12k sites, and that the smaller $A$, the more Co tends to occupy the 4f$_1$ minority-spin site, which is effective in enhancing uniaxial anisotropy. Furthermore, DFT calculations of non-doped $A$Fe$_{12}$O$_{19}$ and the supercells (2 × 2 × 1 of the unit cell) in which 1/96 of $Fe^{3+}$ is replaced by $Co^{2+}$ were performed to predict the stable structure and the Co occupation sites. The results show that regardless of $A$, Co is most stable when it occupies the 4f$_1$ site, followed by the 2a and 12k sites with energy differences on the order of 100 meV, and that Co practically does not occupy the 2b and 4f$_2$ sites. As the $A$ ion becomes smaller, the distribution of energy when Co occupies each Fe site tends to broaden, and the Co occupancy of the 4f$_1$ site also increases. The site selectivity of Co can be roughly explained as a result of the difference in uniaxial strain along the $c$ axis associated with the difference in $A$, but the influence of $A$ ions differs between the R and S blocks in the unit cell, and local strain also has a secondary effect on the Co distribution.

Based on the above results, to improve the performance (anisotropy and magnetization) of La-Co co-substituted M-type ferrite magnets with limited Co content, it is effective to select as small $A$ ions as possible to concentrate Co in the tetrahedral coordination of the 4f$_1$ site.

## Acknowledgements

This work was supported by the Elements Strategy Initiative Center for Magnetic Materials (ESICMM), Grant Number JPMXP0112101004, through the Ministry of Education, Culture, Sports, Science and Technology (MEXT), Japan, and JSPS KAKENHI 17K06793 and 22H00263.

## References

[1] K. Iida, Y. Minachi, K. Masuzawa, M. Kawakami, H. Nishio, H. Taguchi, *J. Magn. Soc. Jpn.* **23** (1999) 1093, Y. Kobayashi, S. Hosokawa, E. Oda, S. Toyota, *J. Jpn. Soc. Powder Powder Metall.* **55** (2008) 541.

[2] Y. Kobayashi, E. Oda, T. Nishiuchi, T. Nakagawa, *J. Ceram. Soc. Jpn.* **119** (2011) 285.

[3] H. Nakamura, T. Waki, Y. Tabata, C. Mény, *J. Phys. Mater.* **2** (2019) 015007.

# Tuning Size of W-type Hexaferrite with Novel Salt Matrix Synthesis

*Jens Plum Frandsen[1,2]*, Mogens Christensen[1,2]*

[1] *Interdisciplinary Nanoscience Center Aarhus University, Gustav Wieds Vej 14, Aarhus, Denmark*
[2] *Department of Chemistry Aarhus University, Langelandsgade 140, Aarhus, Denmark*
*jensplum@inano.au.dk

The hexaferrites constitute a large part of the permanent magnet market[1] with the bulk part constituted of the M-type hexaferrite (MHF). The W-type hexaferrite (WHF) is a candidate phase for improving the current ferrite magnets owing to a theoretical 38% higher saturation magnetisation ($M_s$). The current challenge is obtaining sufficient coercivity ($H_c$). Elevated calcination temperatures (≥ 1200 °C) are needed for phase formation,[2] causing excessive grain growth which reduces $H_c$.

A novel salt matrix (SM) synthesis strategy was recently developed for tailoring the crystallite size of MHF [3, 4]. The difference between the conventional synthesis route and the SM strategy is illustrated in the figure below. The SM strategy was adapted for WHF to produce nanocrystalline powder with the aim of increasing $H_c$. $SrZn_2Fe_{16}O_{27}$ and $SrNiZnFe_{16}O_{27}$ were synthesized utilizing both $K_2SO_4$ and $Na_2SO_4$ as matrix at calcination temperatures between 1150 °C and 1300 °C. Characterization with powder X-ray diffraction showed that the SM strategy yielded nanocrystalline WHF, while scanning electron microscopy revealed the presence of large particles resulting in a sample with a bimodal size distribution.

The magnetic properties of the powder samples were investigated by measuring the magnetic hysteresis with vibrating sample magnetometry. The WHF from the $K_2SO_4$ matrix had a high saturation magnetization up to 88 $Am^2kg^{-1}$ for $SrZn_2Fe_{16}O_{27}$ approaching other reported $M_s$ of 91 $Am^2kg^{-1}$ for WHF[5]. The coercivity, however, was low between 25-40 $kAm^{-1}$. The samples synthesized from $Na_2SO_4$ matrices generally had lower $M_s$ and higher $H_c$ compared to the samples from the $K_2SO_4$ matrices. The coercivity was not improved by the tuning of the crystallite sizes and the low value might be a consequence of a low magnetocrystalline anisotropy. The non-magnetic $Zn^{2+}$-ions in the structure might disrupt the superexchange interaction and lower the magnetocrystalline anisotropy.

## References

[1] J. Ormerod, Chapter 12 in Modern Permanent Magnets, J. Croat and J. Ormerod Eds.: *Woodhead Publishing*, 2022, pp. 403-434.
[2] M. I. Morch, J. V. Ahlburg, M. Saura-Muzquiz, A. Z. Eikeland, and M. Christensen, *IUCrJ*, vol. 6, no. Pt 3, pp. 492-499, 2019
[3] A. Z. Eikeland, M. Stingaciu, A. H. Mamakhel, M. Saura-Muzquiz, and M. Christensen, Sci Rep, vol. 8, no. 1, p. 7325, 2018
[4] J. Svane, "Development and Investigation of Novel Solid Salt Matrix Sythesis of SrFe12O19," master's degree, Department of Chemistry, Aarhus University, Aarhus, 2022.
[5] S. Ram and J. C. Joubert, Journal of Magnetism and Magnetic Materials, vol. 99, no. 1-3, pp. 133-144, Sep 1991

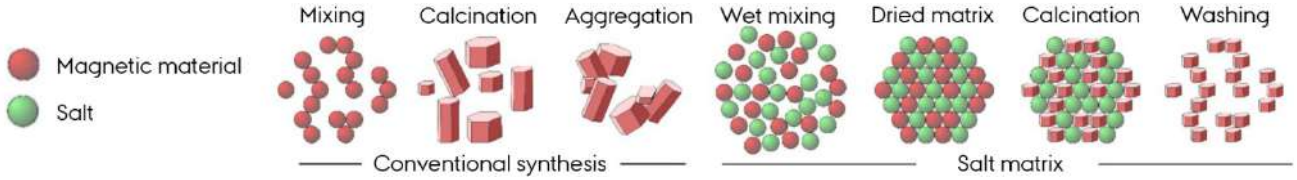



Figure: Sketch of conventional and salt matrix synthesis route. Red bodies indicate magnetic particles or precursor, and green bodies indicate salt particles.

# Magnetic properties of amorphous Fe-Si-Cr-B-C alloy by melt-spinning and selective laser melting for electrical machines

*G. Barrera[1], E. Ferrara[1], P. Tiberto[1*], I. Gallino[2], A. H. Ghavimi[2], R. Busch[2], M. Rodriguez[3], M. T. Pérez-Prado[3]*

[1] *INRiM, Strada delle Cacce, 91, Torino, Italy*
[2] *Saarland University, Campus C6.3, Saarbrücken, Germany*
[3] *IMDEA, Calle Eric Kandel, 2, Getafe, Madrid, Spain*
*p.tiberto@inrim.it

Amorphous soft-magnetic materials play an important role as core constituents in improving the energy transformation efficiency of electrical machines and passive electrical components [1].

Although the melt-spinning process remains one of the main techniques for obtaining amorphous soft-magnetic ribbons, new and efficient production methods based on additive manufacturing have been developed in recent years. These techniques allows to overcome technical limitations characteristic of casting processes and also to print complex 3D geometries.

In this work, room temperature hysteresis properties have been studied in Fe-Si-Cr-B-C alloy obtained in ribbon shape and in 3D printed cubic-shape by different casting techniques.

Ribbons were obtained by a conventional melt-spinning process, in which the pre-alloy was first inductively melted in a quartz tube equipped with a nozzle under vacuum and then injected onto a rotating copper wheel by insufflating high-purity Ar. The produced ribbons have thickness around 40 μm.

On the other hand, the 3D printed cubic samples were obtained by additive manufacturing via Selective Laser Melting using powder of the same alloy as precursor. The effect of changing printing parameters, such as laser power (20-60 W) and scan speed (350-650 mm/s), has been investigated. The processing conditions in the Selective Laser Melting have in fact a crucial role on the microstructure of the printed parts and therefore on their magnetic properties.

This can be observed in the optical images reported in Figure 1 a) where the dependence of morphology (surface roughness, porosity, density, ...) on the processing parameters is evident. The sample having the higher homogeneity is KS12 obtained with a scan speed of 350 mm/s and a power of 50 W.

Room temperature quasi-static hysteresis loops of all printed samples and as-cast ribbon were measured by VSM magnetometry. The corresponding magnetic polarization $J$ (T) values are reported in Figure 1 a). The highest value of $J$, around 1.34 T, has been measured in the ribbon (dotted line), while in printed samples it is seen to increase with increasing sample homogeneity. Hysteresis curves of $Fe_{72.5}Si_{11.05}Cr_{2.23}B_{11.14}C_{3.09}$ ribbon and KS12 printed sample are compared in Figure 1 b). The ribbon displays a magnetically softer behavior with respect to the 3D KS12 printed sample: in the inset, the coercivity appears to be around one order of magnitude larger, and initial susceptibility lower in the 3D printed KS12 sample with respect to the amorphous ribbons.

A digital wattmeter has also been exploited to measure hysteresis losses behavior of ribbons as a function of frequency in the range 1Hz-1kHz at peak induction $J = 0.5$ T.

## References

J. M. Silveyra, E. Ferrara, D. L. Huber, T. C. Monson, *Science*, **362** (2018), 6413.

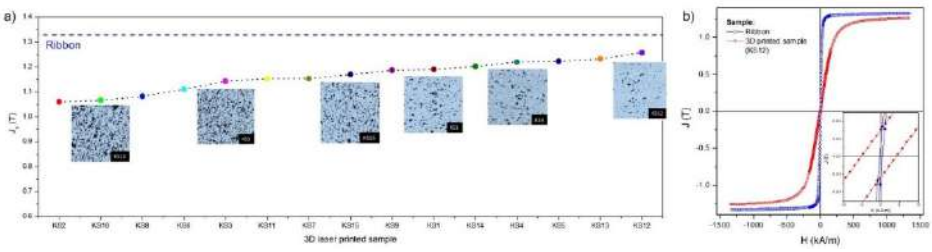



Figure: a) Saturation magnetization behavior of 3D printed samples (circles) and ribbon (dotted line). Optical images of the printed sample surface are shown. b) Quasi-static hysteresis curves of KS12 printed sample and ribbon. In the inset, an enlargement of the low field region.

# Sustainability Assessment of the Manufacture of NdFe-based Materials for Permanent Magnet Applications

*Andrés García-Franco [1,2*], James Rosero-Romo [1,2], Bosco Rodríguez-Crespo[1,2], Paula G. Saiz[1], Jorge Lago[2], Daniel Salazar [1]*

[1] *BCMaterials, Basque Center for Materials, Applications and Nanostructures, UPV/EHU Science Park, Barrio Sarriena s/n, 48940 Leioa, Spain.*
[2]*University of the Basque Country, UPV/EHU Science Park, Barrio Sarriena s/n, 48940 Leioa, Spain.*
**andres.garcia@bcmaterials.net*

The growing interest in new materials for permanent magnet applications stems from the key role they play in the energy efficiency of technological devices ranging from electric motors and wind turbines to hard drives.

Alloys such as NdFeB and $SmCo_5$ are among the materials currently used in these applications. However, its high content of critical raw materials (CRMs), such as rare earth elements or Co, has led in recent years to a renewed scientific effort to find alternative, eco-friendly materials or processes that match their performance. For this purpose, we study two different approaches on NdFe-based materials: i) Magnets with a ThMn12-type structure and rare-earth content and other CRMs are considered prime targets [1] and ii) new recycling methods are making it possible to reuse permanent magnets with properties similar to the originals.

From a sustainability point of view, we have compared magnetic performance, power consumption and required materials of both NdFe-based magnets. For the first compound, we synthesized $NdFe_{11}(Mo)$ samples by arc-melting and melt-spinning, subsequently a nitrogenation process of the powdered ribbons was done to develop coercivity in these samples. For the second, we prepare melt-spun ribbons from scrap NdFeB magnets. Both materials were embedded and oriented in a resin to obtain bonded magnets to compare their magnetic properties.

Crystalline structure, microstructure and elemental composition of bonded magnets were studied by X-ray diffraction and Scanning electron microscopy with energy-dispersive X-ray detector, respectively. Magnetic properties were determined by dc magnetometry using a vibrating sample magnetometer. Our study confirms that both alternatives are efficient ways of making permanent magnets in a sustainable alternative way.

## References

[1] A. Aubert, R. Madugundo, A.M. Schönhöbel, D. Salazar, J.S. Garitaonandia, J.M. Barandiaran, G. Hadjipanayis, Structural and magnetic properties of Nd-Fe-Mo-(N) melt-spun ribbons with ThMn12 structure, Acta Materialia, **195** (2020) 519-526.

# Investigating the Effect of Magnetic Short Circuiting on Alignment of Platy Hexaferrite

*Jacob Løwe Valentin*[1]*, *Mogens Christensen*[1]

[1] *Aarhus University Institute of Chemistry & iNANO, Langelandsgade 140, 8000 Aarhus C, Denmark*

*jval@inano.au.dk

Permanent magnets are important for the green transition, as they are key ingredients in power generation and electrical motors [1,2]. One of the measures of magnetic strength for permanent magnets is the remanent magnetisation ($M_r$), which is greatly affected by the crystallographic orientation of the grains within the sapmle, known as the texture. It has been shown previously that anisotropically shaped crystallites can be oriented without the need for external magnetic fields [1,3].

To map the effect of different compaction methods on the texture of strontium hexaferrite ($SrFe_{12}O_{19}$), four different uniaxial pressing- and sintering methods were compared. Firstly, the compaction and sintering step was separated into a cold compaction (CP) and a cold compaction followed by sintering (CPS). The cold pressed samples showed weak texture and low remanence magnetisation.

The hot compaction methods employed were spark plasma sintering (SPS) and induction pressing (IP). IP and SPS both showed remarkably sharper texture than the cold pressed pellets, with corresponding improvements in remanence magnetisation. It is hypothesised that the alignment is due to compaction above the Curie temperature of the material. Above the Curie temperature the samples are paramagnetic and magnetic short circuiting can be avoided..

## Acknowledgements

Parts of this research were carried out at Petra III @ DESY Hamburg, and Alexander Schökel is thanked for assistance during beamtime at P02.1. Frederik Holm Gjørup and Cecilie Grønvaldt are thanked for fruitful discussions and guidance with data treatment.

## References

[1] C. Granados-Miralles and P. Jenuš, J. Phys. D: Appl. Phys. **54**, 303001 (2021).

[2] O. Gutfleisch, M. A. Willard, E. Brück, C. H. Chen, S. G. Sankar, and J. P. Liu, Adv. Mater. **23**, 821 (2011).

[3] J. Thomas-Hunt, A. Povlsen, H. Vijayan, C. G. Knudsen, F. H. Gjørup, and M. Christensen, Dalton Trans. **51**, 3884 (2022).

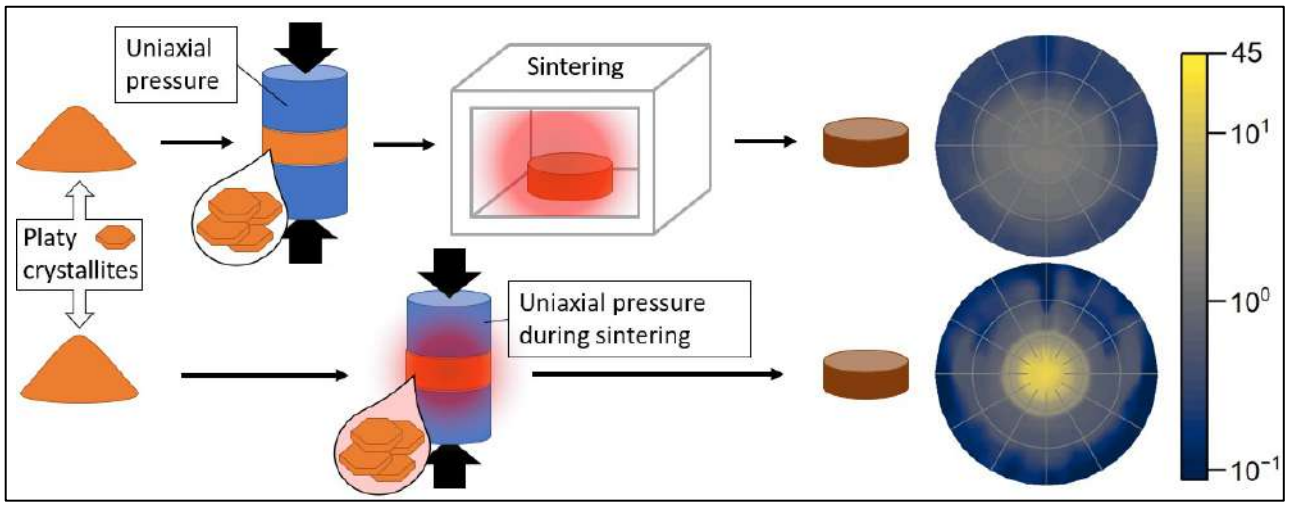



Figure: shows a cold sintering procedure (CPS, top row) compared with a hot compaction procedure (bottom row). Hot compaction produces much sharper texture compared with the cold compaction, as can be seen in the pole figures (perspective along pressing direction). Scale on the right shows texture strength in m.r.d. on a logarithmic scale.

# Synthesis of Magnetite from Natural Pyrite for Energy Conversion Devices

*Vanina G. Franco,[1,2,3,4] Adriana E. Candia,[1,4] Jorge M. Nuñez[1,5], Miguel Rengifo[1,2], Jorge Lobo Checa,[1,2] Myriam H. Aguirre[1,2,6*]*

*[1] Laboratorio de Microscopias Avanzadas, - Universidad de Zaragoza, Campus Río Ebro, Edificio I+D+i, C/ Mariano Esquillor, s/n, 50018, Zaragoza, España*

*[2]Instituto de Nanociencia y Materiales de Aragón, CSIC, Campus Río Ebro, Edificio I+D+i, C/ Mariano Esquillor, s/n, 50018, Zaragoza, España*

*[3] Departamento de Física, Facultad de Ingeniería Química, Universidad Nacional del Litoral, Santiago del Estero 2829, 3000, Santa Fe, Argentina*

*[4]Instituto de Física del Litoral, CONICET-UNL, Güemes 3450, 3000, Santa Fe, Argentina*

*[5]Instituto de Nanociencia y Nanotecnología, CNEA-CONICET, S. C. Bariloche 8400, Río Negro, Argentina*

*[6]Departamento de Física de la Materia Condensada, Universidad de Zaragoza, C/ Pedro Cerbuna 12, 50009, Zaragoza, España*

**maguirre@unizar.es*

Starting from magnetic materials, and using the properties associated with the presence of spins, materials with thermomagnetic conversion can be designed [1]. Direct thermoelectric conversion (DTC) has a secondary role as an energy source, but its development is very important as it offers a carbon-free alternative, which is crucial to contribute to mitigate energy consumption and environmental impact [2]. Pyrite ($FeS_2$) is one of the most abundant minerals in the earth's crust, as well as being easy to extract and inexpensive, which makes it a material of choice for technological applications. By heat treatment, it is possible to generate a thin film of magnetite ($Fe_3O_4$) that confers magnetic properties of the layer material. However, for these materials to be considered for potential applications, it is necessary to clearly understand the interrelationships between their structure, magnetic behaviour and Seebeck, spin Seebeck and Anomalous Nernst effect. In this work, we prepare and characterise a layered material from a natural mineral. Pyrite single-crystals were sectioned and chemically treated for cleaning, annealed at different temperatures and exposure times, under normal conditions of pressure and aerobic atmosphere. The chemical species of each surface film were determined by XPS; the structure and composition of the different materials generated using different techniques of transmission electron microscopy and EELS spectroscopy. The magnetic properties were characterised by MFM and magnetometry measures, and the charge carriers through Seebeck effect, thermal spin pumpimg by Spin Seebeck Effect. XPS and EELS spectra, revealed the presence of oxidised Fe and Oxygen species. TEM and STEM images revealed different structural arrangements for the magnetite which correlates with the magnetisation measurements. The topography and phase MFM images showed significant differences in the configuration of the magnetic domains of each material. The behaviour in relation to the Seebeck effect correlates with the structural data. From the above, we can infer that the use of pyrite as a raw material for the development of thermoelectric materials is possible given the advantages and results obtained in the present study.

## Acknowledgements

Author acknowled the financial support of MSCA- RISE ULTIMATE-I project Nº 101007825 and MELON project Nº 872631 from European Commission.

## References

[1] A. Portavoce *et al., Scientific Reports Open Access,* 13 (2023) 172.
[2] M. Culebras *et al*., *Applied Surface Science,* 615 (2023) 156432.

# AlCoFeNiCu$_x$ ($x$ = 0.6–3.0) High-Entropy Alloys: Combining Soft Magnetism with Zero-Magnetostriction

*Primož Koželj[1,2]*, Jože Luzar[1], Stanislav Vrtnik[1], Pavol Priputen[3], Marián Drienovský[3], Andreja Jelen[1], Magdalena Wencka[4], Darja Gačnik[1], Peter Mihor[1], Bojan Ambrožič[1], Goran Dražić[5], Anton Meden[6], Janez Dolinšek[1,2]*

[1] *Jožef Stefan Institute, Jamova cesta 39, Ljubljana SI-1000, Slovenia*
[2] *Faculty of Mathematics and Physics,University of Ljubljana, Jadranska cesta 19, Ljubljana SI-1000, Slovenia*
[3] *Faculty of Materials Science and Technology in Trnava, Slovak University of Technology in Bratislava, ul. Jána Bottu 25, Trnava SK-917 24, Slovak Republic*
[4] *Institute of Molecular Physics, Polish Academy of Sciences, Mariana Smoluchowskiego 17, Poznań PL-60-179, Poland*
[5] *National Institute of Chemistry, Hajdrihova 19, SI-1000 Ljubljana, Slovenia*
[6] *Faculty of Chemistry and Chemical Technology, University of Ljubljana, Večna pot 113, Ljubljana SI-1000, Slovenia*
* primoz.kozelj@ijs.si or primoz.kozelj@fmf.uni-lj.si

High-entropy alloys [1] are metallic systems composed of multiple principal elements in equimolar or near-equimolar concentrations, where the high entropy of mixing stabilizes a solid solution on a simple lattice (bcc, fcc, hcp) as the main phase in the material. A potential future application of these alloys in the context of their physical properties are as soft magnetic materials [2,3,4] in low-frequency AC magnetic devices, e.g. transformers. While some high-entropy alloys such as FeCoNiPdCu (presented also at JEMS 2022) exhibit properties which are roughly comparable with commercial magnetically soft materials, the properties would still need to be improved to a large extent to make them attractive for real applications.

Motivated by the above we combined our investigation of the magnetic properties of the high-entropy-alloy series of materials AlCoFeNiCu$_x$ ($x$ = 0.6–3.0) with measurements of their magnetostriction at room temperature [5]. All of the materials have coercivities lower than 1000 A m$^{-1}$, so that they truly are magnetically soft materials. The saturation magnetic polarization is largest ($J_s \approx 0.9$ T) at the smallest copper content $x$ = 0.6 and decreases with increasing copper content $x$. The magnetostriction goes from relatively large positive values at small copper content to small negative values at high copper content. The best combination of properties is obtained in the AlCoFeNiCu$_{2.0}$ alloy, which has a relatively low coercivity of $H_c \approx 650$ A m$^{-1}$, a decent saturation polarization $J_s \approx 0.55$ T, while also exhibiting zero magnetostriction $\lambda_s = 0$. The alloys for $x$ = 2.5 and 3.0 have similar properties, so we may also take the whole range $x$ = 2.0–3.0 as magnetically soft materials with negligible magnetostriction. The good magnetostrictive properties obtained in the AlCoFeNiCu$_x$ high-entropy alloys can be intrepreted as a consequence of the changing microstructure with copper content $x$. Proceeding from mainly bcc phase at low $x$, we obtain increasing amounts of two fcc phases at higher copper content $x$, which at a value of $x$ = 2.0 seems to cause the compensation of the negative and positive magnetostrictions of the constituent phases and gives zero magnetostriction.

All in all, this contribution will present AlCoFeNiCu$_{2.0}$, which is one of the first high-entropy alloys that combines soft magnetic properties with zero magnetostriction. Perhaps this is an indication that the large space of possible high-entropy alloys might harbour an exceptional magnetically soft material which would not expand and contract in applied DC fields at 50 or 60 Hz and thus minimize sound pollution at audio frequencies. We will conclude by briefly comparing our results to the few others available in literature [6,7,8].

## References

[1] J.W. Yeh, et al., *Adv. Eng. Mater.* **6** (2004) 299 (2004).
[2] Y. Zhang, T.T. Zuo, Y.Q. Cheng, P.K. Liaw, *Sci. Rep.* **3** (2013) 1455.
[3] T. Kang, S. Wu, M. Wang, J. Wang, X. Fan, Y. Lu, *Appl. Phys. A* **127** (2021) 829.
[4] P. Koželj, S. Vrtnik, A. Jelen, M. Krnel, D. Gačnik, G. Dražić, A. Meden, M. Wencka, D. Jezeršek, J. Leskovec, S. Maiti, W. Steurer, J. Dolinšek, *Adv. Eng. Mater.* (2019) 1801055.

[5] J. Luzar, P. Priputen, M. Drienovský, S. Vrtnik, P. Koželj, A. Jelen, M. Wencka, D. Gačnik, P. Mihor, B. Ambrožič, G. Dražić, A. Meden, J. Dolinšek, *Adv. Mater. Interfaces* **9** (2022) 2201535.
[6] C. Chen, et al., *J. Magn. Magn. Mater.* **502** (2020) 166513.
[7] Y. Zhang, et al., *Metals* **9** (2019) 382.
[8] T.T. Zuo, et al., *Intermetallics* **100** (2018), 1.

# Temperature First Order Reverse Curve (T-FORC) Analysis Applied to Fe-Rh Alloys

*K. Padron-Aleman[1]*, M. Rivas[1,2], J.C. Martínez-García[1,2], P. Alvarez Alonso[1], J.H. Belo[3], J.L. Sánchez Llamazares[1,4],*

[1]*Departamento de Física, Universidad de Oviedo, Calvo Sotelo 18, 33007 Oviedo, Spain*

[2]*IUTA, Universidad de Oviedo, 33203 Gijón, Spain*

[3] *IFIMUP, Departamento de Física e Astronomia, Faculdade de Ciências, Universidade do Porto, Rua do Campo Alegre s/n, 4169-007 Porto, Portugal*

[4] *Instituto Potosino de Investigación Científica y Tecnológica A.C., Camino a la Presa San José 2055, Col. Lomas 4a, San Luis Potosí, SLP 78216, México*

*uo297061@uniovi.es

Near equiatomic Fe-Rh alloys present a giant magnetocaloric effect (GMCE) near room temperature with the greatest adiabatic temperature changes ever reported [1]. These alloys undergo a magnetic structure change from antiferromagnetic (AFM) to ferromagnetic (FM) (and vice versa) when the cell volume changes by around 1 % upon heating/cooling [2]. One of their distinctive features is the strong dependence of the magnetoelastic transition characteristics on preparation conditions [3]. Recently, Franco *et al.* have proposed the determination of temperature first-order reversal curves (T-FORC) distributions as a new method to get meaningful information about several characteristics of first-order phase transitions (FOPT) materials such as identification of inhomogeneities and asymmetries of the heating and cooling transformation branches, among others [3, 4]. In this work, we have obtained the T-FORC distributions for the FM-AFM and AFM-FM branches of a $Fe_{50}Rh_{50}$ alloy exhibiting a broad transition. As a matter of example, figures below show the recoil $M(T)$ curves measured from different return temperatures inside the thermal hysteresis loop at 2 T for the FM-AFM transition and the resulting T-FORC distribution.

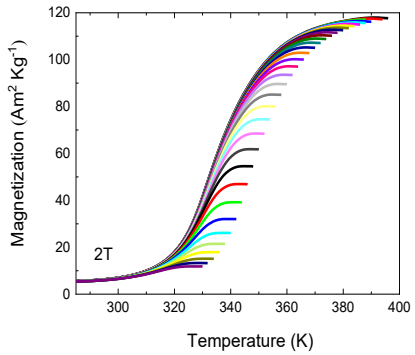



Fig. 1: M(T) curves for each return temperature at 2T.

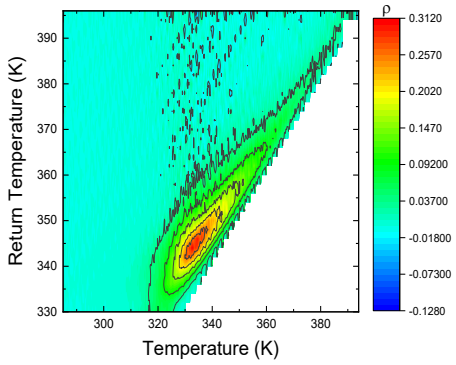



Fig. 2: T-FORC distribution for the FM-AFM transition.

## Acknowledgements

Work supported by MCIU, AEI, European FEDER (MCIU-19-RTI2018- 094683-B-C52), Principado de Asturias (SV-PA-21-AYUD/2021/51822), SEP-CONACyT, Mexico (project A1-S-37066), and LINAN (IPICyT). J.L.S.LL. acknowledges for the support received from the EU-NGEU, MIU, and Plan de Recuperación, Transformación y Resiliencia (Maria Zambrano program; Ref: MU-21-UP2021-030 71741542).

## References

[1] M.P. Annaorazov, K.A. Asatryan, S.A. Nikitin, A.M. Tishin, A.L. Tyurin, *Cryogenics.* **32** (1992) 867.
[2] F. de Bergevin, L. Muldawer, *Compt. Rend.* **252** (1961) 1347.
[3] M.L. Arreguín-Hernández, J.L. Sánchez Llamazares, *J. Alloys. Compd.* **871** (2021) 159586.
[4] V. Franco, T. Gottschall, K. P. Skokov, O. Gutfleisch, *IEEE Magn. Lett.* **7** (2016) 6602904.
[5] Á. Díaz-García, J.Y. Law, F. Albertini, S. Fabbrici, V. Franco, *J. Alloys. Compd.* **867** (2021) 159184.

# Magnetic coupling between a Co film and a barium hexaferrite film

*G.D. Soria[1], E. García-Martín[2], S. Ruiz-Gómez[3], C. Gutiérrez-Cuesta[2*], J.F. Marco[2], J.E. Prieto[2], C. Granados-Miralles[4], E.M. Trapero[2], S. Sánchez[5], M. Foerster[6], L. Aballe[6], J. de la Figuera[2] and A. Quesada[4]*

[1] *CIEMAT , Madrid E-28040, Spain*

[2] *Instituto de Química Física "Rocasolano", CSIC, Madrid E-28006, Spain*

[3] *Max-Planck-Institut für Chemische Physik fester Stoffe, 01187 Dresden, Germany*

[4] *Instituto de Cerámica y Vidrio, CSIC, Madrid E-28049, Spain*

[5] *Instituto de Estructura de la Materia, Madrid E-28006, Spain*

[6] *Alba Synchrotron Light Facility, CELLS, 08290 Barcelona, Spain*

*cgutierrez@iqfr.csic.es

Barium hexaferrite ($BaFe_{12}O_{19}$, BFO) is used as a permanent magnet in many different devices due to its low cost and its high coercivity. However, the moderate saturation magnetization of BFO means that the energy product is orders of magnitude smaller than the one that rare-earth-based magnetic materials offer. To improve its properties, a commonly proposed strategy to enhance the energy product is to combine BFO as a magnetically hard component (BFO) together with a soft phase in order to improve the remanent magnetization without a high loss in coercivity. Nonetheless, the results obtained in other hard/soft systems (SFO/Co bilayers) have pointed out the difficulty of taking advantage of this coupling magnetic regime [1]. In this research, we focus on two steps to investigate the Co/BFO coupling in a bilayer system: first, we sought to obtain BFO films with an in-plane magnetic easy axis to avoid shape anisotropy competition, and second, we deposit Co on top of such a BFO film while monitoring both the BFO and Co magnetic domains.

Following a similar recipe used for strontium hexaferrite [2], thin films with in-plane easy axes have been obtained by RF magnetron sputtering followed by subsequent annealing in air. Their structure and composition were characterized by Mössbauer, Raman spectroscopy, X-ray diffraction and vibrating sample magnetometry (VSM), confirming their in-plane magnetization. On top of it we have grown a 5 nm thick Co magnetic soft layer by molecular beam epitaxy. We have then analysed the resulting bilayer system by X-ray absorption spectroscopy (XAS), and X-ray magnetic circular dichroism (XMCD) coupled to photoemission electron microscopy (PEEM), as well as by Mössbauer spectrocopy and VSM.

## Acknowledgements

This work is supported by the Grants PID2021-124585NB-C31 and TED2021-130957B-C54 funded by MCIN/AEI/10.13039/501100011033 and by "ERDF A way of making Europe" and by the "European Union NextGenerationEU/PRTR"

## References

[1] G.D. Soria, J.F. Marco, A. Mandziak, S. Sánchez-Cortés, M. Sánchez-Arenillas, J.E. Prieto, J. Dávalos, M. Foerster, L. Aballe, J. López-Sánchez, J. C. Guzmán-Mínguez, C. Granados-Miralles, J. de la Figuera, A. Quesada, *J. Phys. D Appl. Phys.* **53** (2020) 344002

[2] G.D. Soria, C. Granados-Miralles, A. Mandziak, P. Janus, M. Saura-Muzquiz, M. Christensen, M. Foerster, L. Aballe, J.F. Fernandez, J. de la Figuera, A. Quesada, *J. Phys. D: Appl. Phys.* **54** (2021) 054003

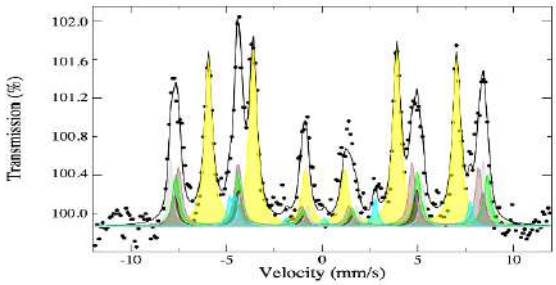



Figure: Mössbauer spectrum of the Co/BFO film.

# Effect of Various Cutting Techniques on Magnetic Properties of Fully Processed NO Electrical Steels

*I. Petryshynets[1]*, F. Kováč[1], J. Füzer[2], P. Molidrán[3], L. Falat[1]*

[1] *Institute of Materials Research, Division of Metallic Systems, Slovak Academy of Sciences, Watsonova 47, 04001 Košice, Slovakia*

[2] *Institute of Physics, Faculty of Science, Pavol Jozef Safarik University, Park Angelinum 9, 041 54 Košice, Slovakia*

[3] *Department of Computer Support of Technology, Faculty of Mechanical Engineering, Technical University of Košice, Mäsiarska 74, 040 01 Košice, Slovakia*

*ipetryshynets@saske.sk

Non-oriented (NO) electrical steels belong to the group of soft magnetic materials and their excellent magnetic properties are similar when magnetized along any direction in the plane of a sheet. The main application is for magnetic cores of rotating electrical machines which are characterized by high efficiency [1]. The magnetic characteristics of magnetic core segments prepared from NO electrical steel strongly rely on grain size, texture, chemical composition steel, mechanical defects, and the quality of the cutting edge. Manufacturing the magnetic core of electrical machines affects the magnetic properties of the electrical steels in terms of inducing stresses near the cutting edge. Since mechanical cutting is the most efficient manufacturing technology, the possible improvement of the manufacturing process by reducing deterioration can enable the full application potential of the magnetic materials. The knowledge of the degree of the deterioration of magnetic property is important for designing electrical machines in terms of magnetic field and energy loss calculations [2].

In this paper, magnetic loss behavior in fully processed NO electrical steels prepared by mechanical shearing, laser cutting, and wire electric discharge machining before and after heat treatment is studied. As experimental material, three vacuum-degassed NO steels were used with the following content of silicon Si = 1.2 wt.%, Si = 2.4 wt.%, and Si = 3.6 wt.%, respectively. The solely designed shear-cutting tool was used for punching ring-shaped samples with outer and inner radii of 25mm and 15mm, respectively. Also, the size of the same experimental samples in form of toroids was cut by laser beam and wire electric discharge machine. The evolution of microstructure and texture near investigated cut edges which were obtained after carrying out different cutting techniques and then heat treatment according to the long-term annealing process was carried out by SEM and EBSD analysis. The analysis of mechanical and thermal stresses around the cutting edges obtained after punching and laser cutting was released by nanoindentation and local misorientation maps (EBSD).

The magnetic measurements of investigated samples in DC and AC magnetic field conditions have clearly indicated that the deterioration of the magnetic properties depends on the geometry of the parts at cutting. The power losses data have shown that the nature of the resulting magnetic behavior of prepared samples is different for mechanical cutting and cutting by laser. It was also demonstrated that the process of deterioration at cutting by laser of small parts affects the cutting edges, which are opposite to each other, in a different way. This is due to the different characters of the process of mechanical cutting and cutting by laser. Finally, the observed changes in the magnetization behavior are correlated with the microstructure and intrinsic parameters of the materials. The different character of the induced residual stresses at mechanical cutting and cutting by laser is briefly discussed.

**Acknowledgements**

This work was carried out within the research projects under contract No. APVV-18-0207, APVV-21-0418, and VEGA 2/0106/21.

**References**

[1] A. J. Moses, *Scripta Materialia.* **67** (2012) 560-565.

[2] J. Füzer, S. Dobák, I. Petryshynets, P. Kollár, J. Slota, F. Kovač. *Materials*., **14**, (2021), 6893.

# Structural And Magnetic Properties of Hexagnoal $Mn_{3-x}Fe_xSn$ Prepared By Melt-Spun Techniques

*1st_Wuliji. Hanggai[1]*, 2nd_N. H. van Dij[1], 3rd_E. Brück[1]*

*[1]Fundamental Aspects of Materials and Energy Group, Department of Radiation Science and Technology, Faculty of Applied Sciences, Delft University of Technology, Mekelweg 15, 2629JB, Delft, The Netherlands*

*H.Gai@tudelft.nl

Heusler compounds have always been the focus of attention due to their fascinating physical properties and potential applications in spintronics,[1] half-metals,[2] ferromagnetic shape memory alloys,[3] and magnetocaloric effects.[4]A classic composition is defined as the $X_2YZ$ structure, where X and Y are *3d*-transition or rare-earth metals and Z is a non-metals element. The important one among them is $Mn_2$-based Heusler alloys, which have received widespread attention recently. Gasi et al. reported the exchange-spring magnetic behavior in $Mn_2FeGa$ alloy.[5] Liu et al. also found the giant exchange bias up to 0.132 T at 5 K in this alloy.[6] Luo et al. predicted theoretically that $Mn_2FeZ$ Heusler compounds would present the half-metallic feature for Z=Al and Sb.[7]

In this work, change the Mn-Fe ratio on structure, magnetic property and magnetocaloric effects of Mn-rich $Mn_{3-x}Fe_xSn$ (x = 0.8-1.4) compound are studied. According to the XRD results, for the all samples crystallize in the $Mn_3Sn$ type hexagonal structure, space group is $P6_{3/mmc}$, the impurity phase was $Mn_2Sn$ hexagonal phase, and according to the XRD pattern refinement results, the impurity phase content of Fe=0.8-1.4 is 1-2 *wt*%. The transition temperature of the compounds significantly increase with Fe content, from 250 K at $x = 0.8$ to 396 K at $x = 1.4$. For an increasing Fe content the maximum isothermal magnetic entropy change ($-\Delta Sm$) increased from of 1.0 $Jkg^{-1}K^{-1}$ to 1.4 $Jkg^{-1}K^{-1}$ for a field change 2 T. The *n*-value is used to define the type of magnetic phase transition. The magnetic entropy change scales with the magnetic field as $\Delta S_M \propto Hn$ in the vicinity of the phase transition. The index of the magnetic field can be expressed as $n = \frac{\mathrm{d}\ln \Delta S_M}{\mathrm{d}\ln H}$. If the maximum *n*-value is greater than 2, the material behavior corresponds to a first-order phase transition(FOMT), and if it is less than 2, it corresponds to a second-order phase transition(SOMT). With the Fe content increase from 0.8 to 1.4, the maximum of *n*-value near the phase transition is found to be less than 2 for all samples, this means during Fe content increase from 0.8 to 1.4 phase transition is the second-order phase transition(SOMT).

## Acknowledgements

Thanks for all of the FAME group members.

## References

[1] T. Jungwirth, X. Marti, P. Wadley, and J. Wunderlich, *Nat. Nanotechnol.* **11** (2016) 231.
[2] R. Weht and W. E. Pickett, *Phys. Rev. B* **60** (1999) 13006.
[3] I. Takeuchi, O. O. Famodu, J. C. Read, M. A. Aronova, K.-S. Chang, C. Craciunescu, S. E. Lofland, M. Wuttig, F. C. Wellstood, L. Knauss, and A. Orozco, *Nat. Mater.* **2** (2003) 180.
[4] P. J. Shamberger and F. S. Ohuchi, *Phys. Rev. B* **79** (2009) 144407.
[5] T. Gasi, A. K. Nayak, J. Winterlik, V. Ksenofontov, P. Adler, M. Nicklas, and C. Felser, *Appl. Phys. Lett.* **102** (2013) 202402.
[6] Z. H. Liu, Y. J. Zhang, H. G. Zhang, X. J. Zhang, and X. Q. Ma, *Appl. Phys. Lett.* **109** (2016) 032408.
[7] H. Z. Luo, H. W. Zhang, Z. Y. Zhu, L. Ma, S. F. Xu, G. H. Wu, X. X. Zhu, C. B. Jiang, and H. B. Xu, J. *Appl. Phys.* **103** (2008) 083908.

# Magnetocaloric Effect at the Metamagnetic Spin-Spiral - Ferromagnetic Transition in $Au_2Mn$

*Elvina_Dilmieva[1]*, Andrei_Rogalev[1], Fabrice_Wilhelm[1], Jean-Paul_Kappler[2], Alex_Aubert[3], Konstantin_Skokov[3]*

[1] *European Synchrotron Radiation Facility, 71 Avenue des Martyrs, 38043 Grenoble, France*

[2] *Synchrotron Soleil, L'Orme des Merisiers, 91190 Saint-Aubin, France*

[3] *Technical University of Darmstadt, 16 Alarich-Weiss-Str. 64287 Darmstadt, Germany*

*elvina.dilmieva@esrf.fr

The research activity in the field of magnetocaloric materials is fueled by the expectation that new advanced refrigerants may be found whose magnetocaloric effect (MCE) will significantly surpass that of Gadolinium metal [1]. Recently this research is boosted by a possibility of using MCE in eco-friendly gas liquefaction at cryogenic temperatures [2]. Structural, magnetic and electronic subsystems of a material contribute differently to theMCE and definite answer to the question of the driving force is still to be found [3]. Therefore, a possibility to disentangle the role of each of these degrees of freedom is essential from both fundamental and technological points of view..

$Au_2Mn$ compound is remarkable system that could help to elucidate the roles of different subsystems to the MCE. It exhibits the 1st order metamagnetic (field induced) transition from a spin-spiral state [4,5] to a ferromagnetic one, which is not accompanied by the structural changes [6] (Fig.1). In this work we present the results of thorough study of magnetic properties of $Au_2Mn$ across the metamagnetic transition. Using low temperature and high field magnetometry, it is shown that at temperatures below 100 K, there a hidden transition to a magnetic state with high magnetic anisotropy. Direct and indirect metods were exploited to study the inverse MCE under fields up to 14 T. The extremely low value ($\Delta S \sim 0.04$ J/(kgK) at $\mu_0 H = 2$ T) of the MCE is unambiguously measured. Since there are no structural changes at the metamagnetic transition, we can assume that this subsystem does not contribute to MCE. To clarify the role of electronic subsystem we have performed X-ray magnetic circular dichroism (XMCD) measurements at the $L_{2,3}$-edges of *Au* and at the *K*-edge of *Mn* across the metamagnetic transitions. The XMCD results showed the presence of a finite magnetic moment on Au atoms, induced via hybridization of the 5*d* states with 3*d* orbitals of Mn atoms. It should be noted that the spin-to-orbital moment ratio of Au is the same for the spin-spiral and the ferromagnetic states. This, in turn, allows us to conclude that there is no significantchanges in the electron sublattice during the metamagnetic transition. Therefore, the electronic subsystem does not contribute either to the MCE. Finally, we can conclude that small value of the MCE in $Au_2Mn$ is due to the magnetic subsystem only. The obtained results are discussed from a more general point of view of influence of metamagnetic transitions on magnetocaloric properties.

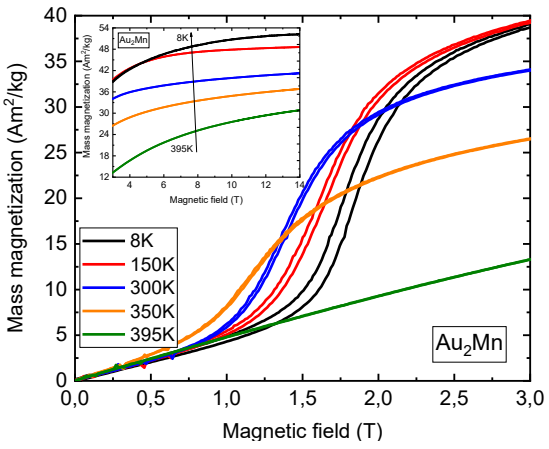



Figure: Magnetization curves at applied field up to 3 T and 3-14 T (inset) in the temperature range from 8 K to 395 K for a $Au_2Mn$ polycrystalline.

## References

[1] V.I. Zverev, A.M. Tishin, M.D. Kuz'min, *J. Appl. Phys.***107** (2010) 043907.

[2] P.B. Castro, K. Terashima, T.D. Yamamoto, et.al., *NPG Asia Materials*,**12** (2020) 35.

[3] A.S. Komlev, D.Y. Karpenkov, R.R. Gimaev, A. Chirkova et al., *J. Alloy Compd.* **898**, (2022) 163092.

[4] L. Liu, M.J. Pascale, J.B. Leao, C.M. Brown, *Phys. Rev. B* **96** (18), (2017) 184429.

[5] J. Glasbrenner, K.M. Bussmann, I.I. Mazin, *Phys. Rev. B* **90** (14) (2014) 14442.

[6] D.P. Morris, R.P. Preston, *Proc. Phys. Soc. B* **69**, (1956) 849.

# Impact of different numerical approaches on the magnetocaloric effect simulation

*C. R. Fernandes [1]*, J. Ventura [1] and D. J. Silva [1]*

[1] *IFIMUP - Institute of Physics for Advanced Materials, Nanotechnology and Photonics of University of Porto, Porto, Portugal*

*up201404999@fc.up.pt

The implementation of the magnetocaloric effect in new models of magnetic refrigeration is part of the demand for sustainable alternatives to the conventional refrigerators. Modeling, simulation, and optimization are crucial processes prior to the construction of a new prototype [1, 2]. Here, we present a comparison between the three most used numerical methods to simulate the magnetocaloric effect: continuous, discrete temperature change step and heat source obtained from adiabatic temperature.

We considered a simple system that consists of a gadolinium (Gd) sample submerged by water, as shown in Fig. 1. Gadolinium and water are rectangles of sizes 0.2x0.6 cm and 0.4x3 cm. To compare the three numerical methods, we performed the 2D simulations with the software COMSOL Multiphysics.

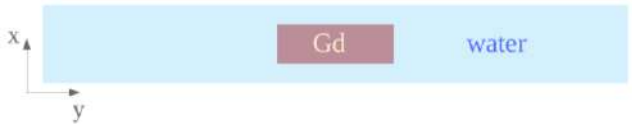



Fig 1 Simulated system.

Table 1 presents an overview and the main features of each method. A t-dependent smooth square wave was used for the continuous method, and a square wave was used for the discrete and $\Delta T_{ad}$ power source methods. The continuous method has the most expensive computational cost, due to the large number of operations, while the discrete is the faster. The impact of $\Delta t$ in the results is larger in the continuous method and is lower in the other two methods. The results show a lower variation with the number of elements in the continuous and $\Delta T_{ad}$ power source methods. The discrete method can only be applied to small time steps, but is the fastest method and the $\Delta T_{ad}$ power source method can be applied in the entire range and is the one that presents the best results for larger time steps.

| | Continuous T change step | Discrete T change step | ΔT ad power source |
|---|---|---|---|
| **Computational cost** | +++ | + | ++ |
| **Impact Δt** | +++ | ++ | + |
| **Impact of number of grid elements** | + | +++ | + |

Tab 1 Overview of the three numerical methods used to simulate the magnetocaloric effect (+++ represents large and + low).

## Acknowledgements

This work was funded by FEDER funds through the COMPETE 2020 Programme and National Funds through FCT – Portuguese Foundation for Science and Technology under projects UID/NAN/50024/2019, PTDC/EME-SIS/31575/2017-POCI-01-0145-FEDER-031575 and PTDC/EME-TED/3099/2020.

## References

[1] 1. K. Nielsen et. al. Int J of Ref, 34, 603–616 (2011)
[2] 2. D. J. Silva et. al. Int J Energy Res, 45, p.18498- 18539 (2021)

# Phase Transitions and the Magnetocaloric Effect

*Julieth C. Patiño[1], Nilson A. de Oliveira[1*]*

[1] *Rio de Janeiro State University, Physics Institute, Rua São Francisco Xavier 524, Rio de Janeiro, Brazil*

*nilson@uerj.br

The magnetocaloric effect is characterized by the entropy change in an isothermal process ($\Delta S_{iso}$) and the temperature change in an adiabatic process ($\Delta T_{ad}$) upon variation of the applied magnetic field. Usually, the curves of these quantities exhibit a sharp peak around the magnetic phase transition and fade away outside this region. However, unusual behavior such as the table-like effect with an almost constant value of the entropy change in a wide range of temperatures, the anomalous effect where the entropy variation exhibits a change of sign along the temperature axis etc, have been observed in materials with more than one kind of phase transition.

Based on the above, in this work, we theoretically discuss the behavior of the entropy change and the magnetocaloric effect in magnetic compounds with more than one magnetic phase transition. For this purpose, we use a model Hamiltonian with more than one magnetic sublattice. In this multiple magnetic sublattices model are included terms to account for the inter and intra-sublattice interactions between the local moments and the coupling with the applied magnetic field. Firstly, for the sake of simplicity, we consider a model with only two magnetic sublattices and perform systematic calculations to theoretically discuss the role of the magnetic interactions on the behavior of the magnetocaloric quantities $\Delta S_{iso}$ and $\Delta T_{ad}$. The results of this analysis show that the behavior of these quantities depends on the relation between the interacting model parameters. For instance, the table-like behavior takes place when the system undergoes two magnetic phase transitions in a short range of temperatures. On the other hand, the anomalous effect as well as the structure with two peaks occur due to the competition between the magnetizations of the sublattices. Afterward, we apply the model to discuss the magnetocaloric effect in the real compounds $R_2Cu_2Cd$ and $RFe_2$ where R stands for rare earth ions. Our calculations show that the entropy change in the compound $Dy_2Cu_2Cd$ exhibits two peaks, with the first one at low temperature associated with a kind of spin reorientation and the other one at higher temperature is associated with ferromagnetic-paramagnetic phase transition. On the other hand, in the compound $TmFe_2$, our calculations show anomalies in the entropy change curve which is associated with the ferrimagnetism of the material and the existence of a compensation temperature. All of these theoretical results are in reasonable agreement with the available experimental data.

**Acknowledgments**

We acknowledge the Brazilian agencies CNPq and Faperj for financial support.

# Structural and Magnetic Properties of Potential Multiferroic Fe/Ti-based Ruddlesden-Popper Phases

*V. Cuartero[1,2*], G. Subías[1], G. Gorni[3], E. Bazarkina[4], L. Amidani[4], and J. Blasco[1]*

[1] *Instituto de Nanociencia y Materiales de Aragón, Dpto. de Física de la Materia Condensada, CSIC-Universidad de Zaragoza, C/ Pedro Cerbuna 12, Zaragoza, Spain*

[2] *Dpto. de Ciencia y Tecnología de Materiales y Fluidos, Universidad de Zaragoza, C/ María de Luna 3, Zaragoza, Spain*

[3] *ALBA Synchrotron Light Source, Carrer de la Llum 2-26, Cerdanyola del Vallès, Barcelona, Spain*

[4] *ESRF-The European Synchrotron. Rue des Martyrs. Grenoble (France)*

*vcuartero@unizar.es

There is a current need on the search for new electronic devices with lower energy consumption. In this context, multiferroic materials offer great opportunities when they show magnetoelectric coupling at ambient conditions. Unfortunately, it is hard to find ferromagnetic ground states in ferroelectrics at room temperature and therefore many efforts are focused on the search for alternative ways of promoting ferroelectricity. This includes novel mechanisms such as hybrid improper ferroelectricity (HIF) [1].

In particular, $ALn_2Fe_2O_7$ (A=Ca, Sr and Ln=Tb, Dy) series of compounds are antiferromagnetic Ruddlesden-Popper phases with a perovskite bilayer adopting a centrosymmetric structure (space group *Amam*). Interestingly, when doping the antiferromagnetic $[1-x](Ca_{0.6}Sr_{0.4})_{1.15}Tb_{1.85}Fe_2O_7$ compound with non-magnetic $[x]CaTiO_3$, the polar $A2_1am$ structure becomes stable at and below room temperature for $0 \leq x \leq 0.3$ [2]. Furthermore, a weak ferromagnetic canting of the $Fe^{3+}$ moments gives rise to a net magnetization coupled to the appearance of the polar structure.

In order to investigate the role of the rare earth on this phenomena, we have substituted Tb with Dy, and we studied $[1-x](Ca_{0.6}Sr_{0.4})_{1.15}Dy_{1.85}Fe_2O_7 + [x]CaTiO_3$ series of compounds (x=0.1, 0.15, 0.17, 0.25 and 0.3). In this case, the polar structure $A2_1am$ is found at room temperature, but the evolution of the lattice parameters slightly differs from Tb series. The substitution of $Tb^{3+}$ with $Dy^{3+}$ decreases the unit cell volume and the tolerance factor accordingly. This factor reduction favors the rotation of $BO_6$ (B=Fe, Ti) octahedra and consequently, the HIF. Furthermore, the slope change in the c/b ratio as a function of Ti content (x), marking the occurrence of the ferroelectric phase [2], is observed at lower x-values in the Dy-based series. In contrast to Tb series, an anomaly in magnetic susceptibility is observed at 129 K only for x=0.3 and magnetic measurements on x=0.1, 0.17 and 0.2 samples agree with a paramagnetic ground state following Curie-Weiss law in a wide temperature range.

The local structure of Fe and Ti ions in both Tb and Dy series has been probed by X-ray absorption spectroscopy (XAS) at Fe and Ti K edges, respectively. For the Dy series, we observe that the Debye-Waller (DW) factor of Fe-O first shell increases as temperature does in all cases. The temperature evolution of the DW factors agree with the Einstein model along the full temperature range meaning there is no sign of any static distortion in the $FeO_6$ octahedra in all the compositions. High energy resolution fluorescence detection-XAS (HERFD-XAS) at the Fe K-edge in the Tb series do not reveil any displacive transition either, when crossing the non-polar/polar structural phase transition observed by neutron diffraction [2]. Furthermore, HERFD-XANES measurements were undertaken in all samples (Tb and Dy) at the Ti K edge, and also agree with the absence of any further local distortion on $TiO_6$ on the polar phase supporting an order-disorder mechanism for the ferroelectric transition.

**Acknowledgements**

We acknowledge the Spanish Ministerio de Ciencia e Innovación for financial support (Projects PID2021-124734OB-C21 and PID2021-124734OB-C22) co-financed by ERDF from EU. We thank the ESRF and BM20 (exp. HC-4927), and ALBA synchrotron (exp. 2021095417) for granting beam time.

**References**

[1] N. A. Benedek et al., *Phys. Rev. Lett* **106** (2011) and *Dalton Trans*. **44**, 10543 (2015)

[2] M.J. Pitcher et al., *Science* **347**, 420 (2015).

# Optimisation of RF rectification effect in magnetic tunnel junction for energy harvesting applications

*Luana C. Benetti*[1]*, *Alex S. Jenkins*[1], *Ricardo Ferreira*[1]

[1]*International Iberian Nanotechnology Laboratory, Av. Mestre José Veiga 4715-330, Braga, Portugal*

*luana.benetti@inl.int

Wireless sensors are becoming an increasingly ubiquitous aspect of modern society as the Internet of Things paradigm becomes a standard part of many emerging technologies, however recent focus has turned to the energy consumption of the billions of IoT sensor nodes. In order for the realisation of a 'Green IoT', low-power sensor nodes are essential, to extend node lifetime and reduce carbon footprint, and one possible solution is wireless radio-frequency energy harvesting. Magnetic tunnel junctions (MTJ) have been previously proposed as nanoscale radio-frequency rectifiers for energy harvesting applications [1], specifically in the sub-µW power regime.

In this presentation, we will present various aspects for optimising the rectification effect in magnetic tunnel junctions. The linear resistance dependence with magnetic field of MTJ devices has been demonstrated to act as RF rectifying signal [2]. A linear response is obtained when the free layer is orthogonal to the pinned layer. In order to achieve the desired linearity, one of the strategies is to optimize the MTJ stacks shown in Fig. a), by tuning the thickness of the CoFeB free layer, where it is possible to change its magnetic orientation from in-plane to out-of-plane. Electrical measurements performed in sub-µm patterned MTJ devices have shown broadband rectified voltage, Fig. b), which covers several frequency bands whose background signals can be utilised for energy harvesting, i.e. mobile phones, Bluetooth and WiFi.

## References

[1] S. Hemour, Y. Zhao, S. Member, C. Henrique, P. Lorenz, D. Houssameddine, Y. Gui, C. Hu, and K. Wu, Towards Low-Power High-Efficiency RF and Microwave Energy Harvesting, *IEEE Transactions on Microwave Theory and Techniques* **62**, 965 (2014).

[2] M. Tarequzzaman, A. S. Jenkins, T. Böhnert, J. Borme, L. Martins, E. Paz, R. Ferreira, and P. P. Freitas, Broadband voltage rectifier induced by linear bias dependence in CoFeB/MgO magnetic tunnel junctions, *Appl. Phys. Lett.* **112**, 252401 (2018).

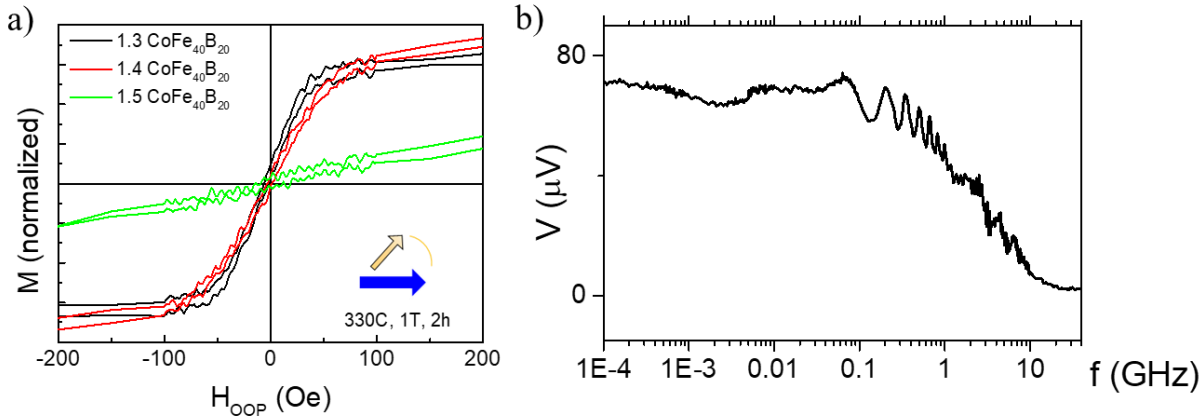



Figure: a) out-of-plane magnetization curves for samples with different free layer thicknesses after annealing b) broadband rectified voltage of a 1 µW excitation signal.

# *Ab Initio* Quantification of Electronic and Magnetoelastic Mechanisms of First-order Transitions: Application to $La(Fe_xSi_{1-x})_{13}$

*Eduardo Mendive Tapia[1]*, Christopher Patrick[2], Tilmann Hickel[3,4], Jörg Neugebauer[3], and Julie Staunton[5]*

[1] *Departament de Física de la Matèria Condensada, Facultat de Física, Universitat de Barcelona, Martí i Franquès 1, E-08028 Barcelona, Catalonia*

[2] *Department of Materials, University of Oxford, Parks Road, Oxford OX1 3PH, United Kingdom*

[3] *Department of Computational Materials Design, Max-Planck-Institut für Eisenforschung, 40237 Düsseldorf, Germany*

[4] *BAM Federal Institute for Materials Research and Testing, 12489 Berlin, Germany*

[5] *Department of Physics, University of Warwick, Coventry CV4 7AL, United Kingdom*

*corresponding author: e.mendive.tapia@ub.edu

$La(Fe_xSi_{1-x})_{13}$ and derived quaternary compounds are well-known for their giant, tunable, magneto- and barocaloric responses [1,2], used in the development of more efficient and environmentally friendly cooling technologies [3]. Caloric effects in $La(Fe_xSi_{1-x})_{13}$ are associated with a first-order paramagnetic-ferromagnetic phase transition near room temperature with low hysteresis, which remarkably shows a large spontaneous volume change together with itinerant electron metamagnetic features. While magnetovolume effects are well-established mechanisms driving first-order transitions [4], purely electronic sources have a long, subtle history and remain poorly understood. Here we apply a disordered local moment picture [5,6] to quantify electronic and magnetoelastic effects at finite temperature [7,8] in $La(Fe_xSi_{1-x})_{13}$ from first-principles. Here we obtain results in very good agreement with experiment and demonstrate that the magnetoelastic coupling drives the first-order character and causes at the same time a huge electronic entropy contribution to the caloric response in this important materials class.

## Acknowledgements

E. M. T. acknowledges funding received from the European Union's Horizon 2020 research and innovation programme under the Marie Sklodowska-Curie grant agreement No. 101025767, and computing time provided at the NHR Center NHR4CES at RWTH Aachen University (project number p0020248). This work is supported by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) under Projektnummers 405621160 and 405621217. J. B. S. acknowledges support from EPSRC (UK) Grant No. EP/M028941/1 (PRETAMAG project).

## References

[1] A. Fujita *et al.*, Phys. Rev. B **65**, 014410 (2001).
[2] Ll. Mañosa *et al.*, Nature Communications **2**, 595 (2011).
[3] X. Moya *et al.*, Nature Materials **13**, 439 (2014).
[4] C. P. Bean and D. S. Rodbell, Phys. Rev. **126**, 104 (1962).
[5] B. L. Gyorffy *et al.*, J. Phys. F: **15**, 1337 (1985).
[6] C. E. Patrick and J. B. Staunton, Electronic Structure **4**, 017001 (2022).
[7] E. Mendive-Tapia and J. B. Staunton, Phys. Rev. B **99**, 144424 (2019).
[8] E. Mendive-Tapia, J. Neugebauer, and T. Hickel, Phys. Rev. B **105**, 064425 (2022).

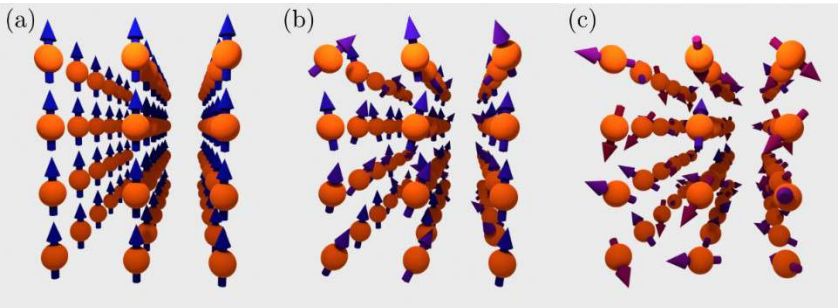



Figure: Disordered local moment picture for a ferromagnetic state, implemented within Density functional theory.

# RE-TM alloy fine powder preparation through induction thermal plasma process for higher performance permanent magnet

*Y. Hirayama[1*], K. Park[1], J. Wang[1], S. Tsurumi[2], M. Shigeta[2], Z. Liu[1], K. Takagi[1] and K. Ozaki[1]*
[1] *National Institute of Advanced Industrial Science and Technology, Nagoya, Japan*
[2] *Tohoku University, Sendai, Japan*
*hirayama.yusuke@aist.go.jp

The average particle size of nanoparticles produced by induction thermal plasma (ITP) is distributed in the range of 10-300 nm. This size can greatly draw out the potential of the ferromagnetic compound as a permanent magnet. However, it is difficult to obtain under micronsized- powder by the conventional break-down process, such as the jet-milling method [1, 2]. Therefore, we focus on the ITP process as a new process to prepare the nanopowders of ferromagnetic compounds.

For the ITP process, the starting materials should be single micron-sized powders. Since rare earth metal powder for use as a starting material is not commercially available, micron-sized powders of rare earth metals such as Y, Ce, Sm, Nd, etc and their alloys were prepared by using a skull gas atomizing system with a water-cooled copper crucible (CCGA-0.8, SINFONIA TECHNOLOGY Co., Ltd). Note that this facility can dissolve metal with a high melting point and a high reactivity without reacting to the crucible and prepare a metal fine powder.

Ferromagnetic nanoparticles synthesized here by the ITP are often single crystals with an average particle size of 100 nm or less. When trying to prepare alloy nanoparticles of metal pairs with large difference of vapor pressures and surface tensions, for example, Sm-Co powder prepared by ITP has a core-shell structure of Co-core/Sm-shell or Co-core/SmCo-shell. According to the numerical analysis based on a binary aerosol formation–growth model [3, 4], this is understood by the coagulation process of Sm which differs greatly from that of Fe or Co, and the coagulation process of Sm continues even after the core becomes a solid phase[5]. In addition, we succeeded in fabricating nanoparticles with a metastable structure due to the rapid cooling effect [6], which is a characteristic of the ITP process. It was also found that all these obtained nanoparticles can be sufficiently oriented by an external magnetic field of 9 T. For $TbCu_7$-type Sm-Fe-N nanoparticles, it was found that there is a degree of crystal orientation of 90.7% along *c*-axis. This was the first step toward obtaining an anisotropic magnet having a metastable structure, which had been difficult to anisotropically aligned. A bulk magnet was obtained by sintering the synthesized Sm-Co nanoparticles, and its huge coercivity of 5.2 T was achieved[7].

## Acknowledgements

The authors wish to thank Kazumi Hayakawa for technical assistance with the TEM microsample preparation using the focused ion beam.

## References

[1] K. Park, Y. Hirayama, W. Yamaguchi, M. Kobashi, K. Takagi, IEEE Transactions on Magnetics 58(2) (2022) 1-5.
[2] K. Takagi, Y. Hirayama, S. Okada, A. Hosokawa, W. Yamaguchi, KONA Powder and Particle Journal 40 (2023) 74-93.
[3] M. Shigeta, T. Watanabe, Powder Technology 288 (2016) 191-201.
[4] M. Shigeta, T. Watanabe, Nanomaterials 6(3) (2016) 43.
[5] K. Park, Y. Hirayama, M. Shigeta, Z. Liu, M. Kobashi, K. Takagi, Journal of Alloys and Compounds 882 (2021) 160633.
[6] Y. Hirayama, M. Shigeta, Z. Liu, N. Yodoshi, A. Hosokawa, K. Takagi, Journal of Alloys and Compounds 873 (2021) 159724.
[7] K. Park, Y. Hirayama, J. Wang, M. Kobashi, Scripta Materialia 218 (2022) 114847.

# Cold Compated $Fe_{16}N_2$ Permanent Magnets

*N. G. Akdogan[2,3], Sylvie Le Floch[4], D. Le Roy[4], and O. Akdogan[1,3,*]*

*[1]Faculty of Engineering and Natural Sciences, Bahcesehir University, 34353 Istanbul, Turkey*

*[2]Faculty of Engineering, Piri Reis University, 34940 Istanbul, Turkey*

*[3]NANOTerial Technology Corporation, Istanbul, Turkey*

*[4]Univ Lyon, Université Claude Bernard Lyon 1, CNRS, Institut Lumière Matière, Campus LyonTech - La Doua, F-69622, Lyon, France*

*ozan.akdogan@eng.bau.edu.tr

Problems with the difficulty in accessing strategically important rare earth metals raised the awareness of the permanent magnet industry to shift its focus from Nd-Fe-B-based alloys to rare earth-free alternatives. Among many prominent rare earth-free hard ferromagnetic alloys, $Fe_{16}N_2$ is promising due to its giant magnetic saturation and relatively high magnetocrystalline anisotropy thus the expected high magnetic energy and abundant constituent elements. In this work, powders with 97% $Fe_{16}N_2$ phase and room temperature coercivity of up to 1050 Oe has been successfully synthesized. [1, 2] The synthesis method is as follows: Fe flake powder was produced by surfactant-assisted ball milling of irregularly shaped Fe powder. Followed by heat treatments for oxidation and subsequent reduction back to iron was performed. The resultant porous Fe flake with a high surface area was nitrogenated under $NH_3$ gas at 150-180 C up to 12 h. The synthesized powder was cold compacted under 2 GPa to a 4 mm cylinder with 80% density while preserving the $Fe_{16}N_2$ phase (Figure 1). The developed rare earth-free permanent magnet has wide applicability in many industries.

**Acknowledgements**

The work was supported by BAP.2022.01.35 (Bahcesehir University).

**References**

[1] Zirhli, O., Gunduz Akdogan, N., Odeh, Y.N., Misirlioglu, I.B., Devlin, E. and Akdogan, O. (2020), Fabrication and Characterization of $Fe_{16}N_2$ Micro-Flake Powders and Their Extrusion-Based 3D Printing into Permanent Magnet Form. Adv. Eng. Mater., 22: 2000311.

[2] N.G. Akdogan, O. Zirhli, M. Gerin, Sylvie Le Floch, D. Le Roy, O. Akdogan, Production and characterization of compacted Fe16N2 permanent magnets, Acta Materialia, Volume 235, 2022, 118064

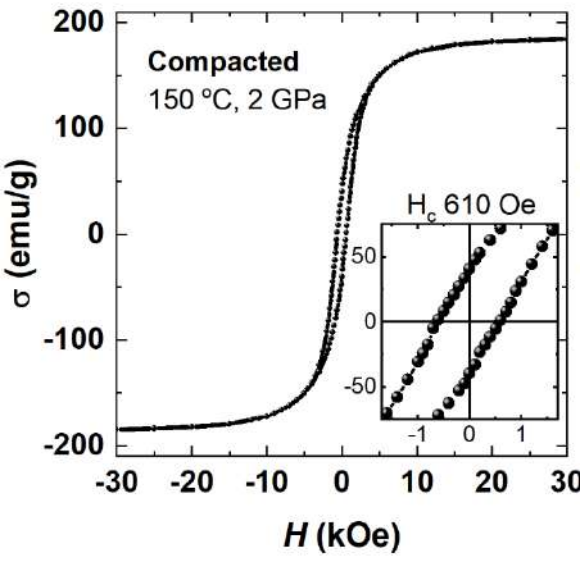



**Figure 1:** Room Temperature Hysteresis loop of $Fe_{16}N_2$ permanent magnet compacted at 150 °C and 2 GPa.

# Two-step sintering of $SrFe_{12}O_{19}$ ceramics: a more sustainable production of ferrite permanent magnets

*A. Quesada[1]*, J.C. Guzmán Minguez[2], C. Granados-Miralles[1], V. Fuertes, J.F. Fernández*

[1] *Instituto de Cerámica y Vidrio (CSIC), Kelsen 5, 28049, Madrid, Spain*

*a.quesada@icv.csic.es

With an annual production amounting to 800 kilotons, ferrite magnets constitute the largest family of permanent magnets in volume, a demand that will only increase as a consequence of the rare-earth crisis. With the global goal of building a climate-resilient future, strategies towards a greener manufacturing of ferrite magnets are of great interest. A new ceramic processing route for obtaining dense Sr-ferrite sintered magnets is presented here. Instead of the usual sintering process employed nowadays in ferrite magnet manufacturing that demands long dwell times, a shorter two-step sintering is designed to densify the ferrite ceramics. As a result of these processes, dense $SrFe_{12}O_{19}$ ceramic magnets with properties comparable to state-of-the-art ferrite magnets are obtained. In particular, the $SrFe_{12}O_{19}$ magnet containing 0.2% PVA and 0.6% wt $SiO_2$ reaches a coercivity of 164 kA/m along with a 93% relative density. A reduction of 31% in energy consumption is achieved in the thermal treatment with respect to conventional sintering, which could lead to energy savings for the industry of the order of $7.10^9$ kWh per year [1]. Ongoing work is being devoted to further lower sintering temperatures and times.

## Acknowledgements

This work is supported by the Spanish Ministerio de Economía y Competitividad through Projects no. RTI2018-095303-A-C52 MAT2017-86450-C4-1-R, MAT2015-64110-C2-1-P, MAT2015-64110- C2-2-P, FIS2017-82415-R and through the Ram´on y Cajal Contract RYC- 2017-23320; and by the European Comission through Project H2020 no. 720853 (AMPHIBIAN).

## References

[1] J.C. Guzmán Mínguez *et al.*, *Ceram. Inter.* **47** (2021) 31765.

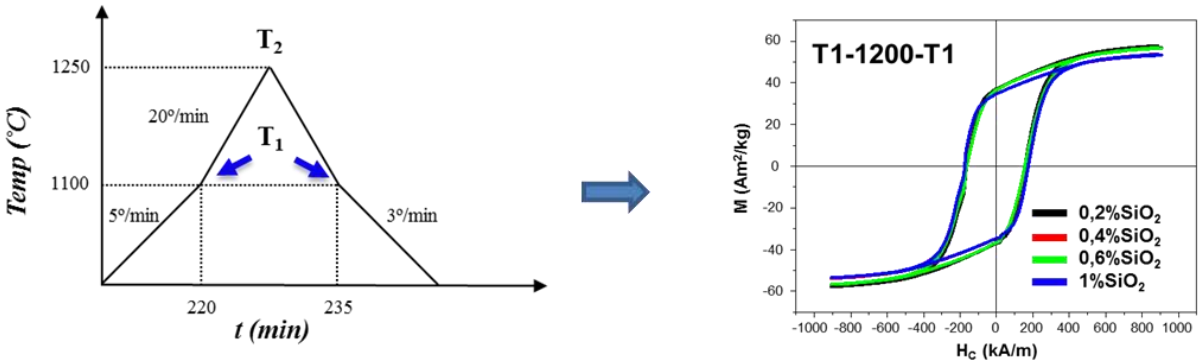



Figure: (Left) Sintering cycle showing temperature as a function of time. (Right) Magnetization curves of the sintered magnets for different additive contents using T1 = 1100 ºC and T2 = 1200 ºC.

# Bulk Rare-Earth Free MnBi-based Permanent Magnets by Severe Plastic Deformation

*L. Weissitsch[1], S. Wurster[1], H. Krenn[2], R. Pippan[1], A. Bachmaier[1,*]*

[1]*Erich Schmid Institute of Materials Science, Austrian Academy of Sciences, Leoben, Austria*
[2]*Institute of Physics, University of Graz, Graz, Austria*
* andrea.bachmaier@oeaw.ac.at

The public awareness in green technologies, thus an increasing demand for wind power plants and electric mobility fosters the relevance of high-performance permanent magnets (PMs) on an economical level. Furthermore, scientific achievements within this field are of paramount importance. This includes the development of novel material systems but also the reduction and substitution of elements necessary for PM production, such as the significant amount of cobalt and rare-earth elements (REE), already recognized as critical materials by international institutes [1].

In former works, we already demonstrated the successful generation of the REE-free intermetallic α-MnBi phase, which exhibits exceptional hard magnetic properties but its formation is a challenging task [2]. Based on high-pressure torsion (HPT), a method of severe plastic deformation, this technique allows to overcome current processing limitations known from metallurgical manufacturing routes. Moreover, the production of large volume-scale samples is possible, exhibiting outer dimensions of several mm to cm, whereas the microstructure simultaneously features particle and grain sizes in the range between several micrometres to tens of nanometres, respectively.

In the present study, we focus on the α-MnBi phase formation which is found to be enhanced after HPT-deformation and magnetic field assisted thermal treatments. The annealing procedure, including the applied external magnetic field, is monitored by in-situ synchrotron X-ray diffraction and the α-MnBi phase formation is correlated with different HPT-deformation grades. The α-MnBi phase formation is associated to the defect density and enhanced diffusion processes. Results obtained by means of electron microscopy and SQUID magnetometry emphasize a positive influence of HPT-deformation prior annealing. Finally, we discuss the influence of adding Sb prior HPT, which stabilizes and supports the α-MnBi phase formation.

## Acknowledgements

This project has received funding from the European Research Council (ERC) under the European Union's Horizon 2020 research and innovation programme (Grant agreement No. 101069203 and grant agreement No 757333).

## References

[1] Cui, J., Kramer, M., Zhou, L., Liu, F., ... & Sellmyer, D. *Acta Materialia* **158** (2018) 118-137.

[2] Gabay, A. M., Hadjipanayis, G. C., & Cui, J. *Journal of Magnetism and Magnetic Materials*, ***495*** (2020) 165860.

# Designing magnetocaloric materials for hydrogen liquefaction with light rare-earth Laves phases

*Wei Liu[1]*, Konstantin Skokov[1], Tino Gottschall[2], Eduard Bykov[2], Oliver Gutfleisch[1]*

[1]*Institute of Materials Science, TU Darmstadt, Darmstadt, 64287, Germany, wei.liu@tu-darmstadt.de*

[2]*Helmholtz-Zentrum Dresden-Rossendorf (HZDR), Dresden, 01328, Germany*

*wei.liu@tu-darmstadt.de

Global climate change caused by greenhouse gas emissions threatens human society. The urgent need for actions to stop global climate change requires changing our way of using energy, including using hydrogen to replace fossil fuels. Recently, we have seen a growing interest in magnetocaloric hydrogen liquefaction. As an emerging technology with high efficiency, magnetocaloric hydrogen liquefaction has the potential to serve as an important building block in the infrastructure for hydrogen energy as liquid hydrogen is important for efficient hydrogen transportation and storage.

Our work focuses on rare-earth magnetocaloric material for liquefying hydrogen, as they are one of the most potential candidates.[1] Via a comprehensive review of the maximum magnetocaloric effect (MCE) with respect to $T_C$ for heavy rare-earth intermetallic compounds, the feature that giant MCE can be achieved in the vicinity of the condensation point of hydrogen is discovered. Two trends, namely increasing maximum $\Delta S_T$ and $\Delta T_{ad}$ with respect to decreasing $T_C$, are summarized. These findings are rationalized by a mean-field approach demonstrating the correlations between maximum $\Delta S_T$ and $\Delta T_{ad}$ and $T_C$. [2]

Inspired by the observation that giant MCE can be achieved near 20 K, a light rare-earth intermetallic series for magnetocaloric hydrogen liquefaction with large MCEs covering the temperature range of 77 ~ 20 K is developed, providing cheaper choices for magnetocaloric hydrogen liquefaction. The findings and methods in this work could help with designing affordable magnetocaloric materials for large-scale applications of magnetocaloric hydrogen liquefaction. [3]

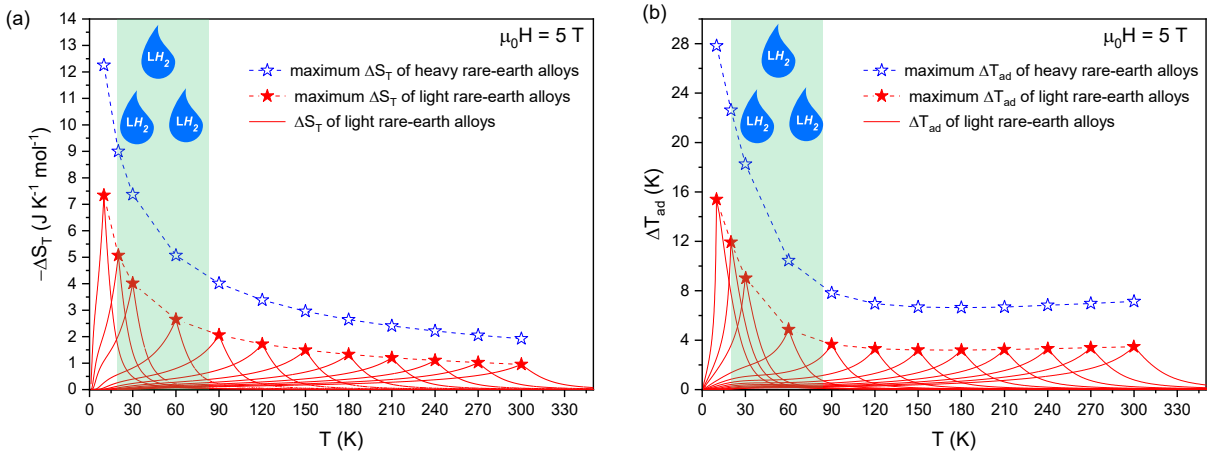



Figure 1 Isothermal magnetic entropy changes for the idealized Nd- and Dy- alloy systems in magnetic fields of 5 T

## Acknowledgements

We gratefully acknowledge supports from the DFG through the Würzburg-Dresden Cluster of Excellence on Complexity and Topology in Quantum Matter-*ct.qmat* (EXC 2147, Project ID 39085490), the CRC/TRR 270 (Project-ID 405553726) and the Project-ID 456263705, from the ERC under the European Union's Horizon 2020 research and innovation program (Grant No. 743116, Cool Innov), and the Clean Hydrogen Partnership and its members within the framework of the project HyLICAL (Grant No. 101101461).

## References

[1] W. Liu, F. Scheibel, K. Skokov, O. Gutfleisch et al., Appl. Phys. Lett. **119** (2021) 022408

[2] W. Liu, T. Gottschall, K. Skokov, O. Gutfleisch et al., Appl. Mater. Today **29** (2022) 101624.

[3] W. Liu, T. Gottschall, K. Skokov, O. Gutfleisch et al., J. Phys.: Energy (under review)

# Magnetocaloric properties of $Y_{1-x}R_xFe_2H(D)_y$ compounds ($R$= Rare Earth)

*V. Paul-Boncour[1*], T. Mazet[2], A. N'Diaye[1], E. Alleno[1], K. Provost[1], F. Couturas[1], V. Shtender[1]*

[1] *Université Paris Est Créteil, CNRS, ICMPE, 2 Rue Henri Dunant F-94320 Thiais, France*

[2] *Université de Lorraine, CNRS, IJL, F-54000 Nancy, France*

*valerie.paul-boncour@cnrs.fr

Hydrogen insertion allows to tune the magnetic properties of rare earth ($R$) and transition metal ($M$) intermetallic compounds, not only by increasing the cell volume but also by modifying the electronic properties and changing the density of state at the Fermi level [1]. For example it allows to tune the Curie temperature $T_C$ of $La(Fe,Si)_{13}$ giant magnetocaloric materials from 200 K to room temperature for magnetic refrigeration application in ambient conditions [2]. On the otherhand hydrogen insertion in $RFe_2$ and $RMn_2$ compounds induces complex structural and magnetic phase diagrams with several phases of different structures and a strong evolution of the magnetic properties versus H content [1].

$YFe_2$ compounds can absorb up to 5 H/f.u. with a decrease of $T_C$ and an augmentation of the Fe moment at 4.2 K up to $x$ =3.5 and a decrease for larger H content [3]. Unusual giant isotope effect has been observed on the magnetic properties of $YFe_2(H,D)_{4.2}$ compounds which show a ferromagnetic-antiferromagnetic (FM-AFM) transition temperature at $T_{M0}$ of 84 K for the deuteride and 131 K for the hydride [4]. They have the same H(D) content and cristallize in the same monoclinic structure but the hydride has a 0.8 % larger cell volume than the deuteride which can explain the shift of $T_{M0}$ [4]. This first order FM-AFM transition is related to a itinerant electron metamagnetic behaviour and displays a large magnetocaloric effect at $T_{M0}$ [5].The substitution of Y by another larger $R$ element ($R$ =Nd, Pr, Gd) allow to tune $T_{M0}$ to larger values but, whatever the rate of $R$ element, $T_{M0}$ remains below 200 K, which is still too low for practical application such as room temperature refrigeration. Several $Y_{1-x}R_xFe_2(H,D)_y$ compounds have therefore been synthetized to search for compounds with magnetic transitions near room temperature and having a large magnetocaloric effect. For this purpose a structural and magnetic phase diagram of $Y_{0.9}Gd_{0.1}Fe_2H_y$ compounds has been determined by X-ray diffraction using synchrotron radiation (XRD-SR), differential scanning calorimetry (DSC) and magnetic measurements for H content between 3 and 5 H/f.u.. Five different hydrides with different crystal structures related to H ordering have been identified, separated by two phase ranges [6]. Hydrides with $3 < y < 3.9$ crystallize in monoclinic and cubic structures, with $T_C$ near room temperature accompanied by a small variation of their magnetic entropy variation [7]. Interestingly, the hydrides with monoclinic structure display also an inverse inverse magnetocaloric effects near room temperature due to a first order transition towards a cubic structure. The correlations between structural and magnetic transitions and the magnetocaloric properties will be detailed for $Y_{1-x}R_xFe_2(H,D)_y$ compounds ($R$ = Nd, Pr and Gd) and H content between 3 and 5 H/f.u..

## Acknowledgements

We are thankful to the synchrotron SOLEIL for the allocated beam time on the CRISTAL beam line (proposal 20160868), and particularly to Erik Elkaim as local contact.

## References

[1] G. Wiesinger and G. Hilscher, Handbook of Magnetic Materials; **17**, edited by K. H. J. Buschow, Elsevier North-Holland, Amsterdam, (2008), p. 293-456.
[2] S. Fujieda, A. Fujita, K. Fukamichi, Y. Yamazaki, and Y. Iijima, Appl. Phys. Lett. **79,** (2001) 653
[3] V. Paul-Boncour and S. Matar, Phys. Rev. B **70,** 184435 (2004).
[4] V. Paul-Boncour, M. Guillot, G. Wiesinger, and G. André, Phys. Rev. B **72,** (2005) 174430.
[5] V. Paul-Boncour and T. Mazet, J. Appl. Phys. **105** (2009) 013914.
[6] V. Paul-Boncour, K. Provost, E. Alleno, A. N'Diaye, F. Couturas, et al., J. Alloy. Compd.**, 896** (2021) 163016.
[7] V. Paul-Boncour, K. Provost, T. Mazet, A. N'Diaye, E. Alleno, et al., J. Alloy. Compd., 907 (2022) 164390.

# SYMPOSIUM 02.
## MAGNETIC MATERIALS FOR INFORMATION STORAGE AND SENSING TECHNOLOGIES.
## S2. INVITED ORAL PRESENTATIONS

# Optimizing Magnetic Sensors with Smart Shapes and High Gain Flux Concentrator

*C. Baraduc[1]*, S. Manceau[1,2], S. Dounia[3], T. Brun[1,2], C. Ducruet[3], P. Sabon[1], C. Cavoit[2], G. Jannet[2], J.R. Childress[3], L. Prejbeanu[1], M. Kretzschmar[2]*

[1] *SPINTEC, UGA-CEA-CNRS, 17 rue des martyrs, Grenoble, France*
[2] *LPC2E, CNRS-Université d'Orléans, 3A av. de la recherche scientifique, Orléans, France*
[3] *Crocus Technology, 3 av. Doyen Louis Weil, Grenoble, France*
*claire.baraduc@cea.fr

Spintronic devices allow the realization of integrated magnetic field sensors with much better performances than Hall effect semiconductor devices. The optimization of a sensor depends on the desired performances in terms of field range, linearity and detectivity. Even if the macrospin sensor remains the reference device, other approaches have also been proposed, in particular the use of spin structures such as domain-walls or vortices. In all cases, hysteresis, sensitivity and noise level are essential issues [1].

We illustrate the richness and diversity of the approaches on two very different examples of sensors based on magnetic tunnel junctions: a sensor with a wide field range (typically 10 mT) and a sensor to detect sub-nanoTesla fields. For the first one, we take advantage of the naturally linear response of the vortex sensor [2]: under planar field, the vortex core moves perpendicularly to the field, which induces a linear response of the magnetoresistive sensor in a field range smaller than the annihilation field. Nevertheless, we may observe some irreproducibility of the signal, which is associated with the hysteresis of the magnetic cycle. We show that we can suppress the hysteresis if the chirality of the vortex is controlled by a judicious choice of the junction shape.

The second example corresponds to the development of sensors for ultra-small field detection. In this case, we use micron-sized junctions, in which the magnetization of the free layer is maintained in the uniform state by soft pinning. This weak exchange coupling is achieved by indirect contact with an antiferromagnetic layer via a spacer with controlled thickness. By carefully tuning the exchange field value, we show that we can improve the sensitivity of symmetric response junctions. Then the use of a flux concentrator can significantly enhance the sensor sensitivity at low field. The flux concentrator design has been optimized to reach high gain values thanks to a particularly narrow air-gap of 10 µm. We demonstrate an amplification gain of a factor 350 with a 6 µm thick flux concentrator realized by electro-chemical deposition. This result opens the way to the development of integrated magnetic field sensors for the detection of ultra-low fields. Such sensors would advantageously compete with current flux sensors, which are much larger and more expensive.

## Acknowledgements

This work has been supported by CNES R&T and ANR-22-CE42-0020 project MAROT

## References

[1] P. Freitas, R. Ferreira, S. Cardoso, *Proc. IEEE* **104** (2016) 1894.
[2] D. Suess et al., *Nat. Elec.* **1** (2018) 362

Figure: Magnetic field sensor for sub-nT field detection

# Emerging Magnetic Sensors based on amorphous magnetic microwires

*P. Marin*

*Instituto de Magnetismo Aplicado, Departamento de Física de Materiales. Universidad Complutense de Madri, A VI km 22,500 28230 Las Rozas (Madrid) SPain*

*mpmarin@fis.ucm.es

Already today, but in the very near future our life will depend enormously on sensors. Robotic technologies, home automation, telemedicine, the autonomous car, the internet of things are developed around sensors. All these applications include gas sensors, biosensors and sensors for medical applications. In this type of devices the transducer plays a fundamental role but the most developed technologies are based on resistive materials and acoustic waves. However, the technologies based on magnetic materials, although being very promising for the development of contactless sensors, are still in is infancy.

In recent years, much interest and effort have been devoted to develop soft magnetic materials due to their technological potential [1]. Amorphous microwires are one of them most widely studied soft materials due to their outstanding properties as giant magnetoimpedance effect [2], bistability, ferromagnetic resonance, and magnetoelastic resonance [3]. It is easy, also, to find much literature regarding microwave-related applications of microwires [4]. This kind of work gives experimental evidence showing that the microwave scattering by a single microwire depends on the magnetic permeability with sufficient strength to be experimentally detected as an effect of the giant magnetoimpedance. This dependence is used to show the potential of such microwire as a wireless field and/or stress sensor. Experimental results are followed by a theoretical approach where the influence of the microwire magnetic state in its microwave reflection features is taken into account. The aim of the present work is to show the physical fundamentals and the possibilities offered by magnetoelastic materials as sensor transducers. In particular, biosensors based on magnetoelastic resonance are shown as well as the importance of the giant magnetoimpedance effect in microwaves domain for the development of remotely detectable safety labels, sensors with biomedical applications as for the detection of blood pressure or for the wireless detection of collagen concentration or even structural health monitoring of structures.

## Acknowledgements

The present work has been supported by the Ministerio Espanol de Ciencia e Innovacion (MICINN) through the projects:, RTI2018-095856-B-C21, and RTI2018-095303-A-C52; and Comunidad de Madrid, Spain, by S2018/NMT-4321 NANOMAGCOST.

## References

[1] P. Marín, M. López, P. Agudo, M. Vázquez, and A. Hernando, *Sensors Actuators, A Phys.* **91** (2001) 1.

[2] A. Gonzalez, V. Zhukova, P. Corte-Leon, A. Chizhik, M. Ipatov, J. M. Blanco, A. Zhukov, *Sensors* ***22*****(3)** (2022) 1053.

[3] C. Herrero-Gómez, P. Marín, and A. Hernando, *Appl. Phys. Lett.* **103** (2013) 14.

[4] C. Herrero-Gómez, A. M. Aragón, M. Hernando-Rydings, P. Marín, and A. Hernando, *Appl. Phys. Lett.***105** (2014) 9.

# SYMPOSIUM 02.
## MAGNETIC MATERIALS FOR INFORMATION STORAGE AND SENSING TECHNOLOGIES.
## S2. ORAL PRESENTATIONS

# Citrate-Coated Manganese Ferrite Nanoparticles and Nanoclusters for Magnetic Biosensing

*V. Pilati[1,2]*, M. Salvador[1,3], J. L. Marqués-Fernández[1], G. Castellanos1[1], L. B. Fraile[1], B. X. Díaz[1], F. G. da Silva[2], C. Kern[2], J. Depeyrot[2], J.C. Martínez-García[1], M. Rivas[1]*

[1] *Departamento de Física & IUTA - Universidad de Oviedo, Lab. Invest. 2, Edificio Departamental Este, Campus de Viesques, 33203 Gijón, Spain*

[2] *Laboratório de Nanosciência Ambiental e Aplicada & Instituto de Física - Universidade de Brasília, Brasília, Brazil*

[3] *Departamento de Energía, Medioambiente y Salud - Instituto de Ciencia de Materiales de Madrid, Madrid, Spain*

*pilativanessa@uniovi.es

Manganese ferrite nanoparticles (MFNPs) are currently envisioned as theranostic nanomaterials [1]. Recent research has indicated that superparamagnetic MFNPs induce a high $T_2$ contrast ability in MRI [2]. In addition, it has been suggested that these materials would increase the success of conventional oncological treatment through magnetic hyperthermia and the generation of reactive oxygen species [1]. Both applications rely on MFNPs' intrinsic magnetic and structural properties.

In the present work, we propose using superparamagnetic MFNPs for magnetic biosensing. Said nanomaterials present high saturation magnetization and low magnetocrystalline anisotropy. Depending on their dimensions, they are superparamagnetic at room temperature and exhibit high initial magnetic susceptibility. Both aspects play a key role in increasing the signal in radiofrequency (RF) inductive sensors [3,4]. Furthermore, since the dimensions of these nanomaterials are compatible with most biological entities such as proteins and viruses, they can tag molecules of interest, providing their isolation from the biological matrix, concentration in a small volume by a field gradient, magnetic detection and quantification.

We synthesized $MnFe_2O_4$ nanoparticles and nanoclusters by hydrothermal coprecipitation. Synthesis parameters were varied to obtain different mean sizes. Nanoparticles and clusters were functionalized with citric acid to achieve colloidal stability and provide -COOH groups on the particle surface for binding to biorecognition molecules (as antibodies). Samples were characterized by XRD, TEM, DLS, ICP, and SQUID measurements. The detection performance of such nanomaterials was measured by using RF capacitive-inductive sensors. We correlated the signal to the MFNP and cluster sizes and magnetic properties.

Our results point out that Mn ferrite nanoparticles and nanoclusters are promising tags to increase the sensitivity of biomolecule detection and quantification. This would be useful for point-of-care testing devices such as lateral flow assays (like the COVID-19 rapid diagnostic tests), which provide the early diagnosis of several diseases, and detection of allergens and toxins in food and environmental pollutants.

## Acknowledgements

This work was partially founded by the Ministry of Science and Innovation of the Spanish Government through grant PLEC2022-009490, the University Technological Institute of Asturias (IUTA) under grant SV-22-GIJON-18, and the Government of the Principality of Asturias under projects FICYT/IDI/2021/000100 and FICYT/IDI/2021/00027 3. M.S. was supported by a "Severo Ochoa" fellowship (Consejería de Educación y Cultura del Gobierno del Principado de Asturias, grant BP19-141) and by the Margarita Salas fellowship financed by the European Union-NextGenerationEU and the Plan for Recovery, Transformation and Resilience.

## References

[1] C.R. Kalaiselvan et al. *Coord. Chem. Rev.* **473** 214809 (2022).
[2] S. Díez-Villares et al. *Adv. Healthc. Mater.* **10** 1–12 (2021).
[3] M. Salvador et al. *Analyst* **145** 5905–5914 (2020).
[4] J.L. Marqués-Fernández et al. *Sensors* **23** (2023)

# Magnetic Field Sensor with SOT Based Offset Compensation and High Sensitivity TMR Readout

*Sebastian Zeilinger[1,2]*, Johannes Guettinger[1], Armin Satz[1], Dieter Suess[2]*

[1] *Infineon Technologies Austria, Siemensstraße 2, Villach, Austria*

[2] *University of Vienna, Kolingasse 14-16, Vienna, Austria*

*sebastian.zeilinger@univie.ac.at

For highly accurate magnetic field sensors, a high signal to noise ratio and a low offset are crucial. In this work we present a sensor concept which combines the high accuracy of TMR readout with the offset cancellation enabled via spin orbit torque (SOT). This allows the design of a magnetic field sensor which is on the one hand capable of offset compensation and on the other hand highly sensitive. Our sensor consists of a ferromagnetic (FM) layer with an in-plane magnetic anisotropy on top of a heavy metal (HM) layer. The TMR stack consists of an oxide layer and a reference layer which are placed on top of the FM layer (see Fig. 1). The reference layer is magnetized in the x-direction and therefore enables the measurement of the x-magnetization in the FM layer via TMR effect. The device utilizes a spin current, generated via an electrical current flowing in the HM layer. This spin current acts on the magnetization of the ferromagnetic layer via the SOT effect. By flipping the SOT current direction the measured effect on the magnetization also switches the sign. The subtraction of these two signals lead to an active offset compensation and therefore to an offset reduced signal. In addition we characterize the sensor response as a function of external bias fields $B_{bias}$ to illustrate the tunability of the sensitivity and linear range by shape anisotropy. We can show with increasing bias field the linear range will increase and as a result the sensitivity decreases.

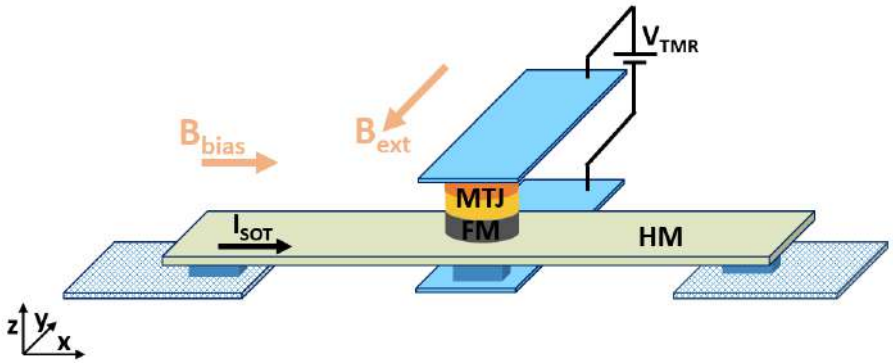



Figure 1: Structure of the sensitive element (SOT line including the TMR junction)

# Modulation of perpendicular magnetic anisotropy in CoFeB/MgO structure by applying tensile strain

*Takeaki GOKITA[1]*, Ryota NAGIRA[1], Taiga TORII[1], Tomohiro KOYAMA[1,2,3] Daichi CHIBA[1,2,3,4]*

[1] *SANKEN, Osaka Univ., 8-1, Mihogaoka, Ibaraki, Osaka, Japan*
[2] *CSRN, Osaka Univ., 1-3, Machikaneyamacho, Toyonaka, Osaka, Japan*
[3] *OTRI, Osaka Univ., 1, Machinaneyamacho, Toyonaka, Osaka, Japan*
[4] *SRIS, Tohoku Univ., 2-1-1, Katahira, Aoba-ku, Sendai, Japan*
*t.gokita11@sanken.osaka-u.ac.jp

Spintronics devices formed on flexible substrates allow us to acquire mechanical information via the magnetoelastic effect and magnetoresistance effect [1]. Recently, we have demonstrated the spin-reorientatuion transition (SRT) between perpendicular and in-plane magnetic anisotropy by applying uniaxial/biaxial tensile strain to Co/Pd giant magnetoresistive devices [2]. The SRT caused by the strain can serve as new types of strain sensors. The sensitivity of them is expected to be improved by realizing this function in a CoFeB/MgO-based magnetic tunnel junctions with higher magnetoresistance ratio [3]. In this study, we investigated the strain control of perpendicular magnetic anisotropy (PMA) in a CoFeB/MgO structure.

CoFeB(1.2 nm)/MgO(2.0 nm) structure with Ta buffer and cap layers was direcrly formed on a flexible substrate by rf-sputtering at room temperature. The film was patterned into a Hall bar structure using maskless exposure apparatus and Ar-ion milling. The sample was annealed at 300°C for 1 hour to introced the PMA. The biaxial tensile strain $\varepsilon_b$ was applied to the sample with an auto tensile machine. We measured anomalous Hall resistance $R_{AHE}$ under several $\varepsilon_b$ values. Figure 1 shows the normalized $R_{AHE}$ ($R_{AHE}^n$) as a function of perpendicular external magnetic field $H_z$. Clear increase in the saturation field is observed when $\varepsilon_b$ = 2.0% is applied, indicating that easy axis swiching from perpendicular to in-plane by the strain occurs in the CoFeB/MgO system. Moreover, the $R_{AHE}^n$ - $H_z$ curve obtained after returning $\varepsilon_b$ to 0% is overlapped with the initial curve. Thus, reversible control of the strain-induced SRT is possible in this system.

**Acknowledgements**

This work was supported by JST CREST (Grant No. JPMJCR20C6), JST A-step (Grant No. JPMJTR20T7), and Spintronics Research Network of Japan.

**References**

[1] S. Ota, A. Ando, D. Chiba, *Nat. Electron.* **1**, 124-129 (2018).
[2] H. Matsumoto, S. Ota, A. Ando, D. Chiba, *Appl. Phys. Lett.* **114**, 132401 (2019)..
[3] S. Ikeda, K. Miura, H. Yamamoto, K. Mizunuma, H. D. Gan, M. Endo, S. Kanai, J. Hayakawa, F. Matsukura, H. Ohno, *Nat. Mater.* **9**, 721-724 (2010).

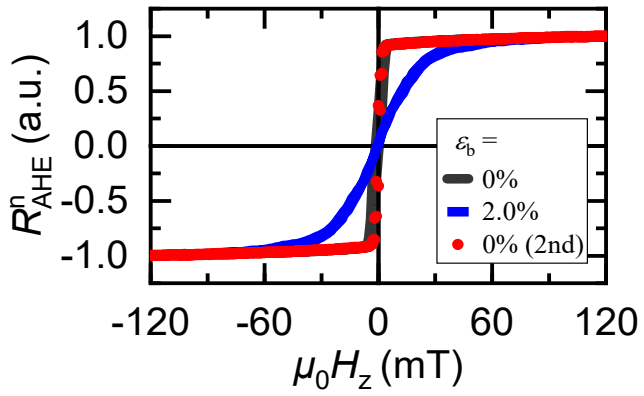



Figure 1 Normalized anomalous Hall resistance $R_{AHE}^n$ as a function of perpendicular external field $H_z$ under applying biaxial tensile strain $\varepsilon_b$.

# Mixed sensors for biomedical imaging

*Claude Fermon*[1], *Aurélie Solignac*[1] *and Myriam Pannetier Lecoeur*[1]
[1] *SPEC, CEA, CNRS, Université Paris-Saclay, CEA Saclay 91191 Gif-sur-Yvette Cedex, France*
*Claude.fermon@cea.fr

The association of a well designed X-Magneto-Resistance (X=Giant or Tunnel; GMR or TMR) and superconducting flux to field transformer allows achieving femtotesla detectivity [1]. These sensors, called mixed sensors, are good candidates for magnetoencephalography, which requires fT- sensitivity for low frequency (<10kHz) signals or very low field Magnetic Resonance Imaging (MRI), where the signal to detect is at higher frequencies (typically few hundred of kHz) but requires even better detectivities down to the subfT, ie. from 0.1 to 0.01fT/√Hz. To target the applications requirements, improvements have to be performed on the sensitivity and on the design of mixed sensors.
We present three advances to tackle these requirements (higher sensitivity, better detection at low frequencies). The first is to use flux-flux untuned or tuned transformers which add an extra gain of sensitivity. This approach is successfully used for performing MRI at very low fields with present detectivities in the range of 0.3-0.5fT/sqrt(Hz).
The second is to use modulation of supercurrents with Joule effect heaters. This technique improves performances of mixed sensors at low frequencies by pushing the detected field by the magnetoresistive sensor at high frequencies.
The third is to lower the working temperature which becomes now rather easy with cryogenic free dilution systems developed for quantum computing. GMR sensors have been successfully used at temperature down to 50mK and present an excellent detectivity [2].

**References**

[1] M. Pannetier, C. Fermon, G. Le Goff, J. Simola, and E. Kerr. *Femtotesla magnetic field measurement with magnetoresistive* sensors. Science, **304**(5677) :1648-1650 (2004).
[2] J. Vallejo Bustamante , N.J. Wu, C. Fermon, M. Pannetier-Lecoeur, T. Wakamura, K. Watanabe and H. Bouchiat. Science, **374**(6573), 1399-1402 (2021).

# Single domain FeCoSiB magnetoelectric composites for low magnetic field sensing

*D. Seidler[1*], P. Hayes[2], L. Thormählen[2], E. Quandt[2], D. Meyners[2], J. McCord[1]*

[1]*Nanoscale Magnetic Materials, Institute for Materials Science, Kiel University, Kaiserstraße 2, 24143 Kiel*
[2]*Inorganic Functional Materials, Institute for Materials Science, Kiel University, Kaiserstraße 2, 24143 Kiel*

*dese@tf.uni-kiel.de

Magnetoelectric (ME) cantilever-based sensors [1-4] have the potential to measure small magnetic fields, such as biomagnetic signals in medical applications. To achieve a low limit of detection (LOD), it is crucial to reduce all noise sources, including dominant magnetic noise caused by the activity of magnetic domain walls during sensor operation. This is especially of importance as ME sensors rely on magnetic modulation schemes.

In this work, we present various schemes of single-domain magnetic multilayer stacks designed to eliminate domain-induced magnetic noise in ME sensors. We employ a scalable approach using magnetostatically coupled $(Fe_{90}Co_{10})_{78}Si_{12}B_{10}$ "free" layers (FL) in combination with exchange-biased antiferromagnetic-ferromagnetic $(MnIr/FeCoSiB)_n$ multilayers (EB-ML) for layerwise magnetization alignment in the millimeter-sized cantilevers. The approach enables nearly unbiased magnetically sensitive layers to exhibit single-domain behavior while maintaining high magnetic field sensitivity. The inverse ME composite resonator sensor concept used is described in detail in [3]. It provides sensitivity at zero magnetic bias fields. The ME sensors are characterized in terms of their magnetic properties in relation to their sensor performance. For this purpose, magnetic bias curves are measured from which the sensors are initialized for maximum sensitivity. In addition, the sensors are characterized in terms of total noise, sensor signal, and LOD.

An example stack with the corresponding bias curve is shown in Figure 1. With the achieved eliminaton of magnetic domain walls we obtain a LOD of 20 $pT \cdot Hz^{-0.5}$ for low frequency magnetic signals at 10 Hz for the sensor shown in Figure 1. Strategies to improve the sensor performance based on different FL and EB-ML arrangements are discussed in detail.

## Acknowledgements

This work was funded by the German Research Foundation (DFG) through the CRC 1261.

## References

[1] S. Salzer, V. Röbisch, M. Jovičević Klug, P. Durdaut, J. McCord, D. Meyners, J. Reermann, M. Höft, R. Knöchel, *IEEE Sensors Journal* **18**, 596 (2018)

[2] M. Jovičević Klug, L. Thormählen, V. Röbisch, S. D. Toxværd, M. Höft, R. Knöchel, E. Quandt, D. Meyners, and J. McCord, *Appl. Phys. Lett.* **114**, 192410, (2019)

[3] P. Hayes, M. Jovičević Klug, S. Toxværd, P. Durdaut, V. Schell, A. Teplyuk, D. Burdin, A. Winkler, R. Weser, Y. Fetisov, M. Höft, R. Knöchel, J. McCord, and E.Quandt, *Sci Rep.* **9**, 16355 (2019)

[4] E. Elzenheimer, P. Hayes, L. Thormählen, E. Engelhardt, A. Zaman, E. Quandt, N. Frey, M. Hoft, G. Schmidt, *IEEE Sensors Journa*l (2023) doi:10.1109/JSEN.2023.3237910

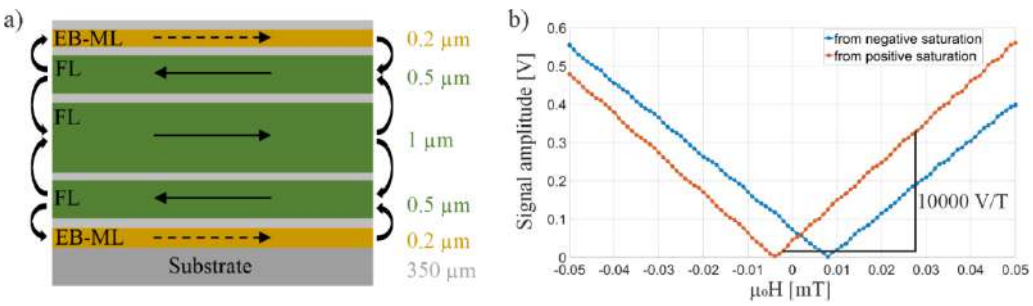



Figure 1: (a) Example of a magnetic multilayer stack and corresponding sensor signal as a function of magnetic bias fields around zero field, showing high sensitivity.

# Novel Linearization Strategy for Magnetoresistve Sensors Based on Artificial Nano-void Arrays

*Pedro D. R. Araujo[1,2]*, R. Macedo[1], S. Cardoso[1,2], D. C. Leitao[3]*

[1] *Instituto de Engenharia de Sistemas E Computadores – Microsistemas e Nanotecnologias (INESC MN) Lisbon, Portugal*

[2] *Instituto Superior Téncico, Universidade de Lisboa, Av. Rovisco Pais, 1000-029 Lisboa, Portugal*

[3] *Department of Applied Physics, Eindhoven Universitiy of Technology, Eindhoven, The Netherlands*

*paraujo@inesc-mn.pt

Tunning of the intrinsic thin film magnetic anisotropy can offer an extra degree of freedom when fabricating magnetic sensors. The ability to locally modify the magnetic anisotropy, either its strength or the easy axis direction, will directly impact key operation parameters in tunneling magnetoresistive devices (TMR) such as linearity, coercivity or the sensing direction [1]. In this work, we will use artificial defects such as voids in the magnetic thin films to locally pin magnetic domains walls due to the presence of local stray fields. Depending on the shape and the lattice arrangement of these non-magnetic voids, we are able to manipulate the magnetic anisotropy [2], and ultimately tailor the electrical output response R(H). For that, a fabrication process is presented to allow the inclusion of micro or nanometric holes precisely at the sensing layer of TMR devices. Electron beam lithography and ion beam milling were used to define the non-magnetic holes with dimensions ranging from 100 nm to 500 nm – figure 1 a. Their depth and profile were evaluated by atomic force microscopy. The impact of the artificial voids was accessed via the transfer curve R(H) at room temperature. Clear changes in the linearity and sensing range were observed (figure 1 b) within a 14 mT range, compared to a device without any nanovoid. In addition, higher field R(H) loops up to 200 mT (not shown) present significant differences in tunneling magnetoresistance ratio. For exemple, an increase from a magnetoresistance ratio of 4.5 % with no nanovoids to 8% was obtained when a quadrangular lattice of 200 nm diameter voids distanced of 400 nm was patterned. The origin of such increase will be discussed in the framework of current perpendicular to plane devices. This work explores a new path to tune the linear range of TMR sensors without compromising the sensor electrical output [3] and which can be done selectively at wafer level to extend the window of magnetic field senstivity.

## Acknowledgements

INESC MN acknowledges FCT for UID/05367/2020. P. D. R Araujo acknowledges FCT grant PD/BD/150391/2019 within the doctoral program AIM.

## References

[1] Salaheldeen, M., et al., *Nanotechnology* 31.48 (2020): 485708.
[2] Krupinski, M., et al., *J. Mag. Magn. Mat.* 518 (2021): 167448.
[3] Silva, A. V., et al., *Eur. Phycs. J. App. Phys.*, 72(1), 10601. (2015).

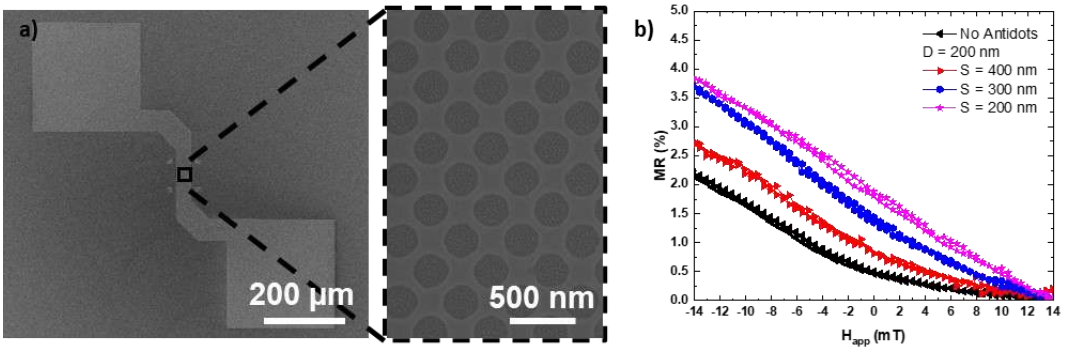



Figure 1: a) Example of a fabricated TMR sensor at the nanovoid fabrication level and b) corresponding R(H) transfer curves for a device with circular nanovoids with 200 nm diameter (D) and distances (S) from 200 nm to 400 nm.

# shifting-discrete magnetic racetrack memory device based on chirality switching

*Shen Li*[1]*, *Pingzhi Li*[1], *Reinoud Lavrijsen*[1],

[1] *Department of Applied Physics, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, The Netherlands*

*s.li2@tue.nl

Magnetic racetrack memory is the archetype research focus of domain wall memory since its first experimental demonstration. Due to its non-volatility, high storage capacity and flexible design, one potential application of racetrack is to replace hard disk drives whose capacity growth is slowing down. However, there are still many challenges before domain wall (DW) devices become commercially available [1-2]. First, the DW movement needs to be highly controllable. The distance the DW moves is proportional to the duration of DW driving stimulus (e.g. magnetic field or spin torque). Precise control of pulse timing usually requires more complex circuits and higher costs. Second, the operating power consumption of DW devices needs to be reduced. The current spin-transfer torque or spin-orbit torque current used to drive DWs is still too large to make DW devices commercially viable.

Hence, different methods are explored to reform the DW motion strategy to address reproducible DW motion. The Dzyaloshinskii–Moriya interaction (DMI), as one of the origins for chiral magnetism, could be game-changer in this respect. It however requires modulating the sign and size of the DMI. In this work, we propose an ultralow power and controllable magnetic racetrack memory device, which is driven by domain-wall chirality-switching and spin current. Until now, several feasible methods of DMI sign control, which gives rise to the aforementioned chirality switching, have been reported, such as ferroelectric proximity effect [3], mechanical strain [4], etc. Interesting simulation results based on chirality switching such as driving domain walls have been proposed. However, this does not allow for a unidirectional motion of all ‘bits’ in the racetrack [5]. We propose to generate chirality switching by ferroelectric polarization switching, while a small symmetry breaking spin current (introduced via a spin-orbit-torque) is used to induce the unidirectional motion during the DW switching process. First, we establish an all-oxide trilayer model based on existing experimental results and demonstrate the feasibility of using it as a racetrack device. We also simulate the DW displacement under different initial and final DMI values, perpendicular magnetic anisotropy, saturation magnetization and damping constant to fit the actual experimental situation. Furthermore, a comparison of various DW motion methods using the same model parameters is made to verify the performance superiority of chirality switching mechanism. These results qualify that our device has the potential to become an ultralow power prototype device based on DW chirality control for racetrack memory [6].

**References**

[1] R. Blasing, A.A. Khan, P.Ch. Filippou, C. Garg, F. Hameed, J. Castrillon, and S.S.P. Parkin, Proc. IEEE 108, 1303 (2020).
[2] P. Li, T.J. Kools, B. Koopmans, and R. Lavrijsen, Adv Elect Materials 2200613 (2022).
[3] Wang, L.; Feng, Q.; Kim, Y.; Kim, R.; Lee, K. H.; Pollard, S. D.; Shin, Y. J.; Zhou, H.; Peng, W.; Lee, D.; Meng, W.; Yang, H.; Han, J. H.; Kim, M.; Lu, Q.; Noh, T. W. Nature Mater 2018, 17 (12), 1087–1094.
[4] N.S. Gusev, A.V. Sadovnikov, S.A. Nikitov, M.V. Sapozhnikov, and O.G. Udalov, Phys. Rev. Lett. 124, 157202 (2020).
[5] Chen, J.; Dong, S. Phys. Rev. Lett. 2021, 126 (11), 117603.
[6] Shen Li et al [in-preparation, 2023].

# Role of interface phenomena in the Integration of Ferroelectric thin films on graphene/Co/HM stacks

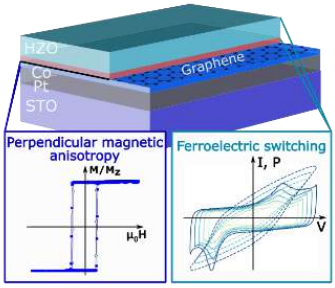
*Iciar Arnay,*‡ Suzanne Lancaster,† Ruben Guerrero,‡,⊥ Adrian Gudín,‡ Alejandra Guedeja-Marrón,¶ Jose Manuel Diez Toledano,§ Jan Gärtner,† Alberto Anadón,‡ Maria Varela,¶ Julio Camarero,§ Thomas Mikolajick,†,‖ Paolo Perna,‡ and Stefan Slesazeck†*

*‡IMDEA Nanociencia, c/ Faraday 9, 28049 Madrid, Spain*
*†NaMLab gGmbH, Nöthnitzer Str. 64a, 01187 Dresden, Germany*
*¶Departamento de Física de Materiales and Instituto Pluridisciplinar, Universidad Complutense de Madrid, Ciudad Universitaria, 28040 Madrid, Spain*
*§Departamento de Física de la Materia Condensada & Departamento de Física Aplicada & Instituto Nicolás Cabrera, Universidad Autónoma de Madrid, 28049 Madrid, Spain*
*‖Institute of Semiconductors and Microsystems, Technische Universität Dresden, Nöthnitzer Str 64., 01187 Dresden, Germany*
*⊥Current address: University of Castilla-La Mancha, 13001 Ciudad Real, Spain*
*iciar.arnay@imdea.org

The integration of ferroelectric thin films with graphene (gr) spintronics potentially allows the realization of highly efficient, electrically tuneable, non-volatile memories through control of the interfacial spin-orbit driven interaction occuring at gr/Co interfaces deposited on heavy metal (HM) supports. Here, the integration of ferroelectric $Hf_{0.5}Zr_{0.5}O_2$ (HZO) on gr/Co/HM epitaxial stacks is investigated via the implementation of several nucleation methods in atomic layer deposition. It has been shown that the gr-ferromagnet interface enhances interfacial perpendicular magnetic anisotropy (PMA) in Co/HM stacks[1]. Moreover, such Gr/Co interfaces present sizeable Dzyaloshinskii-Moriya interaction (DMI)[2] which is critical for stabilizing skyrmionic spin textures[3] and is particularly interesting in the case of gr, as it may be efficiently electrically tuned. The integration of ferroelectrics on these stacks allows the potential for new device geometries exploiting the interfacial Rashba SOC [4]. Nevertheless, the direct deposition of HZO directly on gr shows poor nucleation so no conformal film is formed. This is solved by adding an interlayer between gr and HZO [5]. By employing in-situ $Al_2O_3$ as a nucleation layer sandwiched between HZO and gr, the HZO demonstrates a remanent polarization (2Pr ) of 19.2 µC/cm2. Using an ex-situ, naturally oxidized sputtered Ta layer for nucleation, 2Pr could be controlled via the interlayer thickness, reaching maximum values of 28 µC/cm2 with low coercive fields[6].These results open up pathways for the integration of HZO and other $HfO_2$-based ferroelectrics directly on transfer-free gr achieving high remanent polarization at room temperature.

## Acknowledgements

This project has received funding from the FLAG-ERA JTC 2019 grant SOgraphMEM through AEI/MICINN (Spain, PCI2019-111867-2) and DFG (Germany, MI 1247/18-1; Community of Madrid (CM) through Project P2018/NMT-4321, by MICINN through Projects RTI2018-097895-B-C42, 43 and PID2021-122980OB-C51,52, and by the 'Severo Ochoa' Programme CEX2020-001039-S. AG and IA ackowledge CM (PEJD-2017-PREIND-469020 and PEJD-2019-POST/IND-15343) and JMD,MICINN (BES 2017-080617).




## References

[1] M. Blanco-Rey, et al., ACS Appl. Nano Mater. **4** (2021) 5, 4398–4408
[2] F. Ajejas, et al., Nano letters **18** (2018) 5364–5372; H. Yang, et al., Nat. mater. **17** (2018) 605–609.
[3] P. Olleros-Rodríguez, et al., ACS Appl. Mater. Interfaces **12** (2020) 25419–25427.
[4] B. Muniz, et al. Submitted (2023) preprint at https://arxiv.org/abs/2206.04351; A. Anadón, et al., APL Mater. **9** (2021) 061113; D. Marchenko, et al., Nat. Comm. **3** (2012) 1–6;
[5] S. Lancaster at al. 2022 IEEE Intern. Symp. on Applications of Ferroelectrics (ISAF)
[6] S. Lancaster, et al. ACS Appl. Mater. Interfaces (2023) (Accepted) preprint at https://arxiv.org/abs/2208.14061.

Figure: Schematic view of the hybrid FE/Gr/FM/HM stacks showing ferroelectric switching and PMA

# Nanostructured Lanthanum Manganite Films for Fast Strong Magnetic Field Sensors

*Nerija Zurauskiene[1,2]*, Vakaris Rudokas[1], Voitech Stankevic[1,2], Skirmantas Kersulis[1], Justas Dilys[1], Vilius Vertelis[1,2], Mindaugas Viliunas[3], Valentina Plausinaitiene[1,4]*

[1] *Department of Functional Materials and Electronics, Center for Physical Sciences and Technology, Sauletekio ave. 3, 10257 Vilnius, Lithuania*

[2] *Faculty of Electronics, Vilnius Gediminas Technical University, Sauletekio ave. 11, 10223 Vilnius, Lithuania*

[3] *Faculty of Physics, Vilnius University, Sauletekio ave. 9, 10222 Vilnius, Lithuania*

[4] *Faculty of Chemistry and Geosciences, Vilnius University, Naugarduko Str. 24, 03225 Vilnius, Lithuania,*

*nerija.zurauskiene@ftmc.lt

Magnetoresistive (MR) sensors are widely used in consumer, automotive, medical, industrial and other applications [1]. *Yole group* report on magnetic sensor market and technology [2] predicted an increase of MR sensors market at the expense of widely used Hall sensors in different application areas, especially when enhanced sensitivity, detectivity and miniaturization is required. The magnetoresistive sensors, as they are broadly understood, usually include xMR (AMR, GMR, TMR) devices, and their operation range of magnetic fields is less than tens of millitesla. However, advanced scientific, medical and industrial equipment requires magnetic field sensors capable of measuring stronger magnetic fields (1–10 Tesla) in a wide range of temperatures. Furthermore, application of high pulsed magnetic fields for scientific or industrial applications requires fast, highly sensitive magnetic sensors with nano-microscale dimensions. It was demonstrated, that magnetic sensors based on colossal magnetoresistance effect in polycrystalline manganite films are able to measure pulsed magnetic field magnitude independently on magnetic field orientation in respect to the sensor's plane [3]. Such sensors, called CMR-B-scalar sensors [4], were used for measurement of magnetic field distribution in electromagnetic launchers and nondestructive pulsed magnets, when duration of magnetic field pulses was more that millisecond [3]. However, for shorter pulses, for example, in magnetic forming and welding systems or plasma science applications, the sensors measuring microsecond duration magnetic fields with high accuracy and temporal resolution are required. Therefore, to use manganite films for the development of fast magnetic sensors, the magnetic memory effects have to be minimized. Moreover, to avoid parasitic signal due to electromotive forces appearing in the transmission line of the sensor during measurement of short-pulsed magnetic field, a special recording system has to be developed.

In this study, the results of magnetoresistance and memory effects of thin nanostructured lanthanum manganite films are presented and discussed. La-Sr-Mn-O films were grown by Pulsed-injection metal organic chemical vapor deposition technique onto polycrystalline $Al_2O_3$ and $SiO_2$ substrates with different thicknesses in the range of 60–360 nm. The magnetoresistance of these films was measured in pulsed magnetic fields up to 20 T, while the resistance relaxation after the switch-off of the magnetic field was recorded up to 10 T. It will be demonstrated how magnetoresistance values (sensor's sensitivity) and memory effects of lanthanum manganites could be tuned in a wide range of magnetic fields and temperatures by changing the substrate and thickness of the films. Moreover, a magnetic field recording system using a bipolar-pulsed voltage supply with a frequency up to 12.5 MHz and a 16-bit ADC with a sampling rate of 25 MHz will be presented and discussed. The measurements of several microsecond duration magnetic fields by using a sensor based on the investigated manganite films will be presented.

## References


[1] C. Zheng, K. Zhu, S. Cardoso de Freitas, J.-Y. Chang, J. E. Davies, P. Eames, P. P. Freitas, O. Kazakova, C. G. Kim, C.-W. Leung, S.-H. Liou, A. Ognev, S. N. Piramanayagam, P. Ripka, A. Samardak, K.-H. Shin, S.-Y. Tong, M.-J. Tung, S. X. Wang, S. Xue, X. Yin, P. W. T. Pong, *IEEE Trans. Magn.*, **55** (2019) 0800130.

[2] Yole Intelligence. Magnetic Sensor 2022 Report. 2022. Available online: https://www.yolegroup.com/product/report/magnetic-sensor-2022/# (accessed on 16 March 2023).

[3] T. Stankevič, L. Medišauskas, V. Stankevič, S. Balevičius, N. Žurauskienė, O. Liebfried, M. Schneider, *Review of Scientific Instruments*, **85** (2014) 044704.

[4] N. Žurauskienė, *Sensors* **23** (2023) 2939.

# Effect of size and Sr content on the detectivity of anisotropic magnetoresistive sensors patterned in $La_{1-x}Sr_xMnO_3$ thin films

*R. Solís [1,2]*, A. Vera[2], L. Enger[1], V. Pierron[1], B. Guillet[1], J.M. Díez[2], L. Pérez[2], M.T. Gonzalez[2], R. Miranda[2], J. Camarero[2], S.K. Chaluvadi[3], V. Polewczyk[3], A.Y. Petrov[3], G. Vinai[3], L. Braglia[3], P. Torelli[3], P. Orgiani[3], P. Perna[2], S. Flament[1], L. Méchin[1]*

[1] *Normandie Univ, UNICAEN, ENSICAEN, CNRS, GREYC, 14000 Caen, France*
[2] *IMDEA Nanociencia, Campus de Cantoblanco, 28049 Madrid, Spain*
*3 Istituto Officina dei Materiali (IOM)−CNR, Laboratorio TASC, I-34149 Trieste, Italy*
*raul.solis@imdea.org

Manganite perovskites have attracted the attention of the scientific community due to their interesting electronic and magnetic properties. The concentration of a dopant cation will affect heavily the physical and structural properties of the material, changing, among other things, its Curie temperature. $La_{1-x}Sr_xMnO_3$ (LSMO) deposited on vicinal $SrTiO_3$ (STO) substrates, was chosen for fabricating Anisotropic Magnetoresistance (AMR) sensors due to its very low intrinsic noise in the low-frequency region. As the final goal of these devices is to be able to use them for biomedical applications, it is essential for them to operate at body temperature (~310 K) and in the low frequency region. The LSMO thin films were deposited using Pulsed Laser Deposition (PLD) and Molecular Beam Epitaxy (MBE) and were patterned in Wheatstone bridge geometry using laser lithography and Ion Beam Etching (IBE).

Following the promising results previously achieved with detectivity as low as 200 pT·Hz$^{-1/2}$ [1, 2], new pathways for optimization have been implemented in order to simplify the required setup and get lower detectivity values while keeping the core configuration, ideas, and simplicity of fabrication intact. The use of vicinal substrates induces uniaxial magnetic anisotropy with the easy axis parallel to the the step edges. First we will present results obtained with AMR sensors made of 45 nm thick epitaxial $La_{2/3}Sr_{1/3}MnO_3$ deposited on 4º vicinal STO(001) with different sizes for the Wheatstone bridges. The usual bridge size which we refer to as 100% has 300 µm length and 100 µm width and is scaled down to 50% and 75% and scaled up to 200%. It is confirmed that the detectivity varies inversely with the square root of the Wheatstone bridge volume (figure 1). Second, we will show the effect of different dopant concentrations ($0.15 \leq x \leq 0.45$) of $La_{1-x}Sr_xMnO_3$ deposited on 4º vicinal STO(001) on AMR and noise [3].

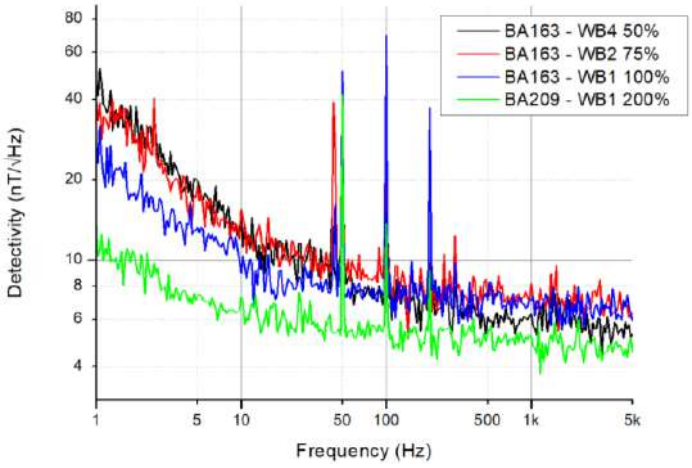



Figure 1: Effect of the size of the Wheatstone bridges on the detectivity for 10V bias.

## References

[1] L. G. Enger *et al.*, *IEEE Trans. Magn.*, 58, 2, 1-4 (2022) 4001204
[2] L.G. Enger *et al.*, *ACS Appl. Mater. Interfaces*, 5, 2, (2023) 729–739.
[3] S. K. Chaluvadi *et al.*, *ACS Omega, 7,* 17 (2022) 14571-14578

# SYMPOSIUM 02.
## MAGNETIC MATERIALS FOR INFORMATION STORAGE AND SENSING TECHNOLOGIES.
## S2. POSTERS

# Extraordinary magnetoresistance in a 2-terminal structure

*Jérémy Létang[1]*, Stefano Lumetti[1], Perla Malagò[1],Stefan Dan Costea[2], Wolfgang Hauer[3], Jürgen Kosel[1], Michael Ortner[1]*

[1] *Silicon Austria Labs GmbH, Europastraße 12, 9524 Villach, Austria*
[2]*Eaton European Innovation Center, Bořivojova 2380, 252 63, Roztoky u Prahy, Czech Republic*
[3] *Eaton Industries GmbH, Scheydgasse 42, 1215 Vienna, Austria*
*jeremy.letang@silicon-austria.com

State-of-the-art fault current limiters are based on active components or inductors, and are large and expensive devices. Here, we propose a new passive and miniaturized magnetoresistance-based system [1] strongly reducing device size and cost. The extraordinary magnetoresistance (EMR) effect was chosen as it exhibits the largest resistance ratio among the known magnetoresistive (MR) phenomena, reaching values up to $10^7$ % [2].

Many works have studied EMR in high-mobility materials (like semiconductors [3], graphene [4] and heterostructures [5]) mainly for magnetic field sensing [2, 5]. For current limiter applications, only 2-terminal geometries are applicable. Through 2D finite element simulations (COMSOL), we have studied several geometry and material parameter combinations to understand if and how EMR-based current limiters can be realized. For instance, we have designed different planar shapes and sizes for the electrodes (such as van der Pauw disk [3], multibranch geometry [2], bar type geometry [5] or stripes). For 2-terminal systems, our simulations show that the EMR ratio reaches its largest value in simple sandwich designs, that maximize the resistance variation between current paths where the electrons cross the metal-semiconductor interface tangentially and normally, respectively. Comparing the EMR ratio values reported in the literature [2, 3, 5] with those resulting from our simulations, we suppose that the higher EMR values displayed by 4-terminal systems arise from a combination of Hall and EMR effects. 2-terminal systems, where the Hall effect is absent, exhibit instead lower EMR ratios.

Simulations show that EMR saturates with lateral extension of such 2-terminal systems. This EMR saturation varies as a function of electron mobility and out-of-plane magnetic field according to the following empirical relation: $EMR = 19.1(\mu B)^2$. This is in agreement with previous works [3]. It shows that it is possible to reach high EMR values of about 2000 % at a magnetic field of 1 T when the semiconductor materials mobility is around $10^5$ $cm^2/(Vs)$. Such values are very promising for current limiter components. This behavior can also be found with a simple 2-channel model which describes 2-terminal systems, where electrons follow the least resistive path. It fits to the simulation data and gives $EMR = 8480l/(0.29+l)$, with $l$ being the lateral extension of our system.

In summary, this work investigates a new passive and compact curent limiter device concept based on the EMR effect. With proper mobility and good interfaces, an EMR ratio as high as 2000% can be achieved in a 2-terminal system, which could make this technology suitable to replace state-of-the-art fault current limiter components.

## References

[1] P. Kopejtko, EP Patent EP3 961 925A1 (2022)
[2] T. H. Hewett and F. V. Kusmartsev, *Physical Review B*, **82** (2010) 21, 212404
[3] S. A. Solin et al., *Science*, **289** (2000) 5484, 1530-1532
[4] S. Pisana et al., *Nano Letters*, **10** (2010) 1, 341–346
[5] J. Sun and J. Kosel, *Materials*, **6** (2013) 2, 500–516

# Growth of Atomically Flat Insulating MgO Films on Fe(001) using Interface Oxygen Pre-Coating

*Toyo Kazu Yamada[1,2]*, Nana Nazriq[1], Kyosei Ishii[1], Peter Krüger[1]*

[1] *Department of Materials Science, Chiba Univ., 1-33, Yayoi, Inage, Chiba, Japan*

[2] *Molecular Chirality Research Centre, Chiba Univ., 1-33, Yayoi, Inage, Chiba, Japan*

*toyoyamada@faculty.chiba-u.jp

The Fe/MgO interface is one of the most promising junctions for spin filtering, which has brought about tunneling magnetoresistance (TMR) devices and provided the MgO-based magnetoresistive random access memory (MRAM) application [1]. One crucial problem from the application point of view is that on Fe(001), MgO grows in the three-dimensional Volmer-Weber mode rather than in the layer-by-layer (Frank-van der Merwe) mode. As a consequence, nanometer-sized MgO clusters are formed, resulting in large interface roughness, which is detrimental to device performance.

This is caused by the strong interation between Fe(001) 3*d* surface states and adsorbed molecular states. For example, the oxidization process of the Fe(001) in vacuum ($10^{-6}$ Pa) produced $Fe_2O_3$, $Fe_3O_4$, and FeO phases at different temperatures [2]. Also, this strong bonding could stop the thermal diffusion of adsorbed organic molecules [3,4].

Here, we succeeded in introducing a 1×1 oxygen monolayer film at the interface between the MgO films and the Fe(001) substrate [5]. This interface engineering drastically modifies the MgO growth and increases the MgO island size by a factor of 10 as compared to MgO grown directly on Fe(001) (see Figure). As a result, regularly shaped and atomically flat MgO islands are obtained. By using our novel method, the MgO/Fe interface roughness is dramatically reduced, which should lead to improved device performance. All experiments were performed using ultra-high vacuum (UHV) low temperature (4.6 K) scanning tunnelling microscopy and spectroscopy (STM/STS) setup.

We also show, by a combination of tunnelling spectroscopy and first-principles calculations, that the MgO islands provide an efficient way to electronically decouple organic molecular films from the metal substrate with interesting perspectives for organic semi-conductor junctions.

## References

[1] S. Yuasa, T. Nagahama, A. Fukushima, Y. Suzuki, K. Ando, *Nat. Mater.* **3** (2004) 868.
[2] T. K. Yamada, Y. Sakaguchi, L. Gerhard, W. Wulfhekel, *Jap. J. Appl. Phys.* **55** (2016) 08NB14.
[3] T. K. Yamada, Y. Yamagishi, S. Nakashima, Y. Kitaoka, K. Nakamura, *Phys. Rev. B* **94** (2016) 195437.
[4] E. Inami, M. Shimasaki, H. Yorimitsu, and T. K. Yamada, *Scieitific Reports* **8** (2018) 353.
[5] N. K. M. Nazriq, P. Krueger, T. K. Yamada, *Appl. Surf. Sci.* **618** (2023) 156628.

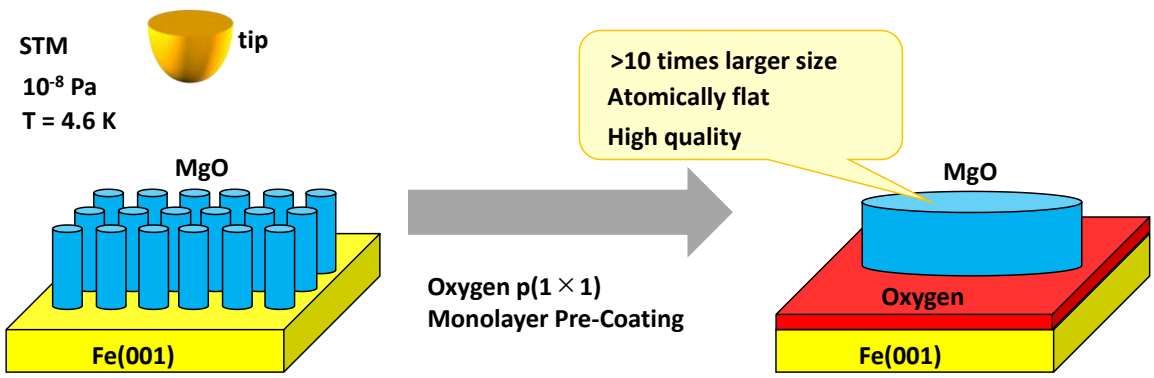



Figure: STM study of the growth of atomically-flat MgO islands on Fe(001)-p(1×1)O substrate.

# Study of the magnetoresistive behaviour of LSMO thin films towards optimise AMR biosensors

*I.García-Manuz *[2], R. Solís [1,3], J.M. Diez [2], G. De Arana [1], J. L. F. Cuñado[1], A. Gudín[1], L.G. Enger [3], B. Guillet [3], L. Méchin [3], S. Flament [3], P. Perna [1] and J.Camarero[1,2]*

[1]*IMDEA-Nanoscience, Campus de Cantoblanco, 28049 Madrid, Spain*

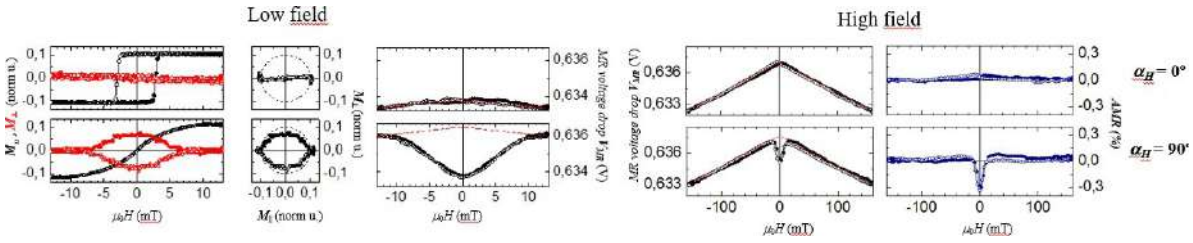
[2]*Dpto. Física de la Materia Condensada, IFIMAC, and Instituto "Nicolás Cabrera", Universidad Autónoma de Madrid, 28049Madrid, Spain*

[3]*CNRS GREYC (UMR 6072) ENSICAEN UNICAEN, 14050 Caen, France*

*E-mail: ines.garciam@uam.es*

The advance in nanotechnology promises to achieve the dream of being able to detect synaptic signals under physiological conditions in real time. Spintronic technologies are currently used in the fabrication of magnetic sensors because of their low fabrication cost, robustness, temperature stability and scalability. AnisotropicMagnetoresistive (AMR) sensors comprise of a single ferromagnetic material which are a much less complex fabrication than giant magnetoresistive (GMR) and tunnel magnetoresistive (TMR) technologies. In particular, engineered AMR sensors based on epitaxial $La_{1-x}$ $Sr_xMnO_3$ (LSMO) films grown on top of vicinal (001)-oriented SrTiO3 (STO) single crystal substrates [1] show both highly sensitivity performances and suitable for industrial implementation [2,3]. The understanding of both magnetic and magnetoresistance features and their temperature dependencies will enable the optimisation of future tailored devices for biosensor applications.

In this work, magnetic and transport features of epitaxial LSMO films grown on vicinal STO substrates with different vicinal angles are investigated. Simultaneuos vectorial-resolved magneto-optic Kerr effect (vMOKE) and magnetoresistance (MR) measurements have been acquired at different applied field angles ($\alpha_H$) and different temperatures ($T$) close to Curie temperature [4]. In brief, MR clearly shows two contributions: one isotropic (red dashed curve in central panels) and the other anisotropic (right panels). At a given $T$, the former displays the same linear drop of the resistivity as the magnetic field increases for all $\alpha_H$ conditions, and it is related with the collosal magnetoresistance (CMR). Remarkably, CMR is better identified at large magnetic fields while the anisotropic magnetoresistance (AMR) contribution is at low fields. AMR originates from competing magnetic anisotropy terms: uniaxial $K_U$ (growth-induced) and biaxial $K_2$ (magnetocrystalline). The study allows to figure out which is the optimal sample to be used as sensors.




**References:**

[1] P. Perna et al. Adv. Funct. Mater. 27, 1700664 (2017)

[2] L.G. Enger et al., IEEE Trans. Magn., **58**, no. 2, 1-4, (2021). **ByAxon**-FETOPEN

[3] Luiz G. Enger et al. ACS Appl. Electron. Mater. 2023, 10.1021/acsaelm.2c01096

[4] I.García-Manuz et al., to be published

# Tuning the Perpendicular Magnetic Anisotropy of CoFeB and $Co_2MnGa$ for Voltage Tuneable Magnetoresistive Sensors

*Atif Islam,[1,2*] Anmol Mahendra,[1,2,3] Susant Acharya,[1,2] Yao Zhang, [1,2] Simon Granville[1,2]*

[1]*Robinson Research Institute, Victoria University of Wellington, Wellington, 6140 New Zealand*
[2]*MacDiarmid Institute for Advanced Materials and Nanotechnology, Wellington, 6012 New Zealand*
[3]*National Isotope Centre, GNS Science, PO Box 30368, Lower Hutt 5010, New Zealand*

*atif.islam@vuw.ac.nz

Magnetic materials with switchable magnetic domains and anisotropy are ideal for applications in the area of spintronics[1]. However, for sensing applications, the existing magnetoresistive (MR) sensors are limited by their sensing axis, which is restricted to only one direction. To overcome this limitation, it is crucial to tune the magnetic anisotropy of the reference layer of the thin film stack used in the MR sensors. The most efficient and reliable is the electrochemical effect[2] which uses ionic liquid to apply a gate voltage to tune magnetic anisotropy in magnetic materials[3]. This method has the added advantage of low joule heating dissipation and low power consumption of magnetic devices[4]. Here, I will present the tuneability of perpendicular magnetic anisotropy (PMA) in the two stacks, 'MgO/$Co_2MnGa$/Pd' and 'MgO/CoFeB/W'. This tuneability achieved through ionic liquid gating (ILG), and the change was recorded using a magneto-optic Kerr system (MOKE). Both stacks reduced their PMA upon the application of gate voltage through the ionic liquid. The CoFeB-based stack can completely switch from PMA to in-plane magnetic anisotropy and this change was reversible. In contrast, $Co_2MnGa$ could only reduce its PMA through gate voltage and remained in that state without any reversibility. Our results demonstrate that the ILG of thin film stacks based on reference layers of CoFeB and $Co_2MnGa$ is very useful for making the sensing axis tuneable in MR sensors.

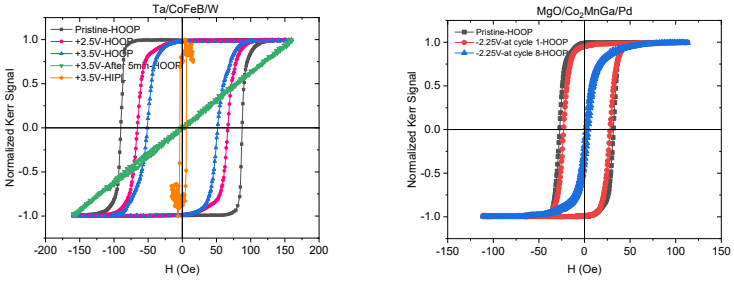



*Figure 1 Hysteresis curves of both stacks measured at various gate voltages using MOKE. The reduction of PMA signal is prominent for both stacks. HOOP is hysteresis with out-of-plane magnetic field and polar mode of MOKE. HIPL is hysteresis with in-plane magnetic field and longitudinal mode of MOKE.*

**Acknowledgements**
Ministry of Business, Innovation and Employment (MBIE) New Zealand endeavour fund research programme: "**Voltage tuneable magnetoresistive sensors**"

**References**

[1] A. Barrera *et al.*, *Adv. Sens. Res.*, vol. 2200042, p. 2200042, 2022.
[2] C. Song *et al.*, *Prog. Mater. Sci.*, vol. 87, pp. 33–82, 2017.
[3] A. J. Tan *et al.*, *Nat. Mater.*, vol. 18, no. 1, pp. 35–41, 2019.
[4] C. Navarro-Senent *et al.*, *APL Mater.*, vol. 7, no. 3, 2019.

# Magnetic sensors based on MTJs with thick amorphous free layer

*E. Paz[1]*, A. Talantsev[1], A. Jenkins[1], R. Ferreira[1]*

[1] *INL – International Iberian Nanotechnology Laboratory, Av. Mestre José Veiga s/n, 4715-330 Braga, Portugal*

*elvira.paz@inl.int

Magnetic sensors are widely used in our daily lives [1], resulting in a significant amount of research on improving their properties, not only to have a better sensitivity but also: to increase their thermal stability [2] to be used under harsh conditions; and to reduce their dimensions [3][4] to have better spatial resolution and reduced energy consumption. For magnetic sensors based on magnetic tunnel junctions (MTJ), the sensitivity depends on the volume of the sensing layer, in theory the bigger the area and the thicker the MTJ the lower the noise of the sensor, but in reality there is a trade-off [5]. Amorphous materials attracted [6] as the sensing layer of an MTJ as they have been shown to be a soft magnetic materials in layers with thicknesses of a few hundreds of nanometers [5]. In this work a linear response has been achieved in bulk with one magnetic annealing, where the field was applied perpendicular to the field direction during the deposition, allowing the possibility of making large sensors with a good linear response in order to decrease the noise of the sensor.

In this work we are going to present how we can control the linear range of an MTJ with the stack Buffer / 20 IrMn / 2.6 $CoFe_{30}$ / 0.825 Ru / 1.8 $CoFe_{40}B_{20}$ / MgO / 2.0 $CoFe_{40}B_{20}$ / 0.21 Ta / 100 CoFeSiB / Top lead, where all the thicknesses are in nm, $CoFe_{30}$ percentage is weight and $CoFe_{40}B_{20}$ is atomic percentage. We manage to control the linear range by modifying the MTJ shape, both with and without aspect ratio, with one single annealing process. As it can be seen in the figure b), the coercive field and the tunnel magnetoresistance are almost constant, independent of the shape or size of the MTJ sensor, whereas the linear range controlling the sensitivity can be tuned between 3 and 54 mT by adjusting these parameters.

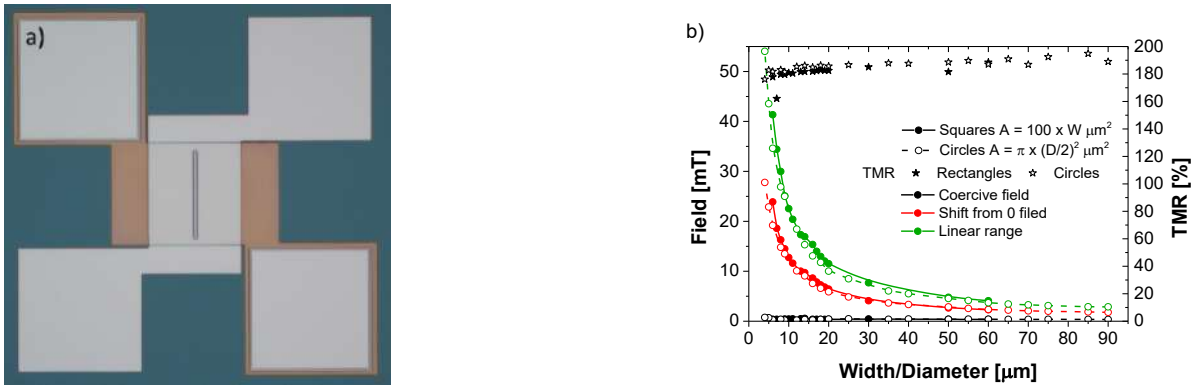



Figure: a) Optical microscope image of one device b) Characteristic fields and tunnel magnetoresistance of sensors depending on their shape.

## Acknowledgements

The work is supported by the project NeWeSt - New generation of cyberphysical Weighing Systems, with the reference nº POCI-01-0247-FEDER-069716, co-funded by COMPETE 2020 - Operational Programme for Competitiveness and Internationalisation under the PORTUGAL 2020 Partnership Agreement, through the European Regional Development Fund (ERDF).

## References

[1] X. Liu, K. H. Lam, K. Zhu, et *al.*, *IEEE Trans. Magn.* **55** (2019) 0800222
[2] B. Lim, M. Mahfoud, P. T. Das, et *al.*, *APL Mater.* **10** (2022) 051108
[3] E. Raymenants, O. Bultynck, D. Wan, et *al.*, *Nat. Electron.* **4** (2021) 392–398
[4] K. Wu, M. C. J. Cheeran, J. P. Wang, et *al.*, *ACS Appl. Nano Mater.* **3** (2020) 9560–9580
[5] M. Oogane, K. Fujiwara, A. Kanno, et *al.*, *Appl. Phys. Express* **14** (2021) 123002
[6] L. Huang, Z. H. Yuan, B. S. Tao, et *al.*, *J. Appl. Phys.* **122** (2017) 113903

# High Sensitivity Amplification in Symmetric Response Magnetic Tunnel Junction with Flux Concentrators

*T. Brun[1,2*], S. Manceau[1,2], C. Ducruet[3], P. Sabon[1], C. Cavoit[2], G. Jannet[2], L. Prejbeanu[1], M. Kretzschmar[2], C. Baraduc[1]*

*[1]SPINTEC, Univ. Grenoble Alpes, CNRS, CEA, 17 avenue des Martyrs, Grenoble, France*
*[2]LPC2E, CNRS & Université d'Orléans, 3a avenue de la recherche scientifique, Orléans, France*
*[3]Crocus Technology, 3 Av Doyen Louis Weill, Grenoble, France*
* thomas.brun@cea.fr

Magnetic Tunnel junctions (MTJs) are often used as magnetic sensors due to their high sensitivities but are limited by their 1/f noise. Detecting sub-nanoTesla magnetic fields is therefore challenging. A possible route to achieve this goal is to amplify the sensor sensitivity by using a high gain flux concentrator. Here we demonstrate a gain of 350 with the addition of Flux Concentrators (FCs) on Symmetric Response MTJs (SR-MTJs).

The challenge in increasing the sensitivity of the MTJ itself lies in the compromise between the ability of the free layer magnetization to rotate freely and the need to keep the magnetization uniform without domain formation. This is a key point as reducing the noise of the sensor requires increasing the magnetic volume and thus using micron-size junctions. The solution we developed for our symmetric response MTJ is to use a soft-pinned free layer [1]. We precisely control the exchange coupling strength between the free layer and an IrMn layer by inserting a thin Ru spacer layer. The corresponding exchange field value is measured with a vibrating sample magnetometer.

With a soft-pinned free layer, we obtained SR-MTJs with a sensitivity 3.6 %/mT. Our experimental results are well described by an analytical model based on Stoner-Wohlfarth model considering exchange, Zeeman and shape anisotropy energies. We also included in the model any possible misalignment due to the process (orientation of the junction, direction of the magnetic field during annealing) or to the measurement.

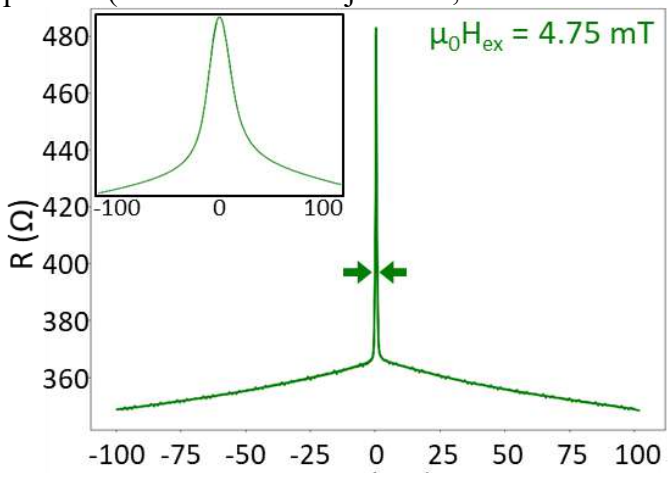



Fig. 1 Symmetric response of a soft-pinned junction with flux concentrators aligned with the hard axis. The high gain result in a strong increase of the sensitivity at low magnetic field. Inset: same junction without flux concentrators. The exchange soft-pinning field is 4.75 mT.

Finally, to strongly increase the sensitivity, 6µm-thick NiFe flux concentrators were electrodeposited around the MTJs. Their elongated shape and narrow air-gap of 10 µm are the key points to obtain the highest amplification. We have measured a gain of 350 for our most recent fabrication [2]; the effect of the flux concentrators is shown in Fig. 1 with the response of the SR-MTJ without FC in inset. The FC very high gain results in a drastic increase of sensitivity on small fields range.

To conclude, using the high gain flux concentrators we achieve a sensitivity of 1260 %/mT. This result paves the way towards the detection of very small fields with an integrated sensor. For space missions, such a miniature sensor could advantageously replace currently used magnetometers with a significant reduction of mass [3].

**Acknowledgements**

This work has been supported by CNES R&T and ANR-22-CE42-0020 project MAROT.

**References:**

[1] R. Ferreira, E. Paz, and P. P. Freitas, *IEEE Transaction on Magnetics* **48**, (2012) 3719.
[2] S. Cardoso et al., *Microsyst Technol* no. **20**, (2014) 793.
[3] A. Bocheux, C. Cavoit, and M. Mouchel, *IEEE (SAS 2016) Proceedings*, 149 (2016).

# Programmable multistate memory and spin logic cell

*Yufeng Tian*, Yanxue Chen, Lihui Bai, and Shishen Yan*

[1] *School of Physics, State Key Laboratory of Crystal Materials, Shandong University, Jinan 250100, P. R. China*

* yftian@sdu.edu.cn

Emerging data-intensive applications in fields including machine learning and neural networks require high-performance and energy-efficient computation for operations such as automatic driving, pattern recognition, and disease diagnosis. Controllable spin-orbit torque based nonvolatile memory is highly desired for constructing energy efficient reconfigurable logic-in-memory computing suitable for emerging data-intensive applications. Here we report our investigation of IrMn/Co/Ru/CoPt/CoO heterojunction as a promising candidate for both multistate memory and programmable spin logic applications. The studied heterojunction can be programmed into four different magnetic configurations at-will by controlling both the in-plane exchange bias between IrMn and Co and the out-of-plane exchange bias between CoPt and CoO. Moreover, based on the controllable exchange bias effect, 10 states nonvolatile memory and multiple logic-in-memory functions have been demonstrated as shown in Figure 1. These findings indicate that IrMn/Co/Ru/CoPt/CoO heterojunction can be used as a building block for next-generation logic-in-memory and multifunctional, multidimensional spintronic devices[1].

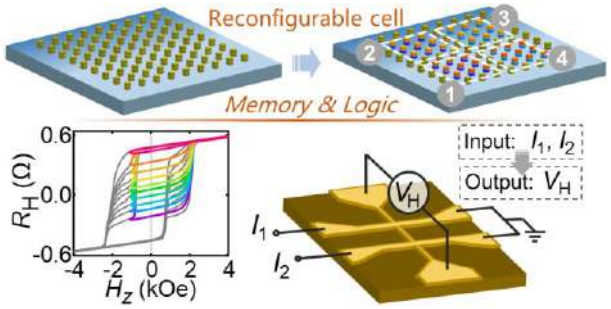



Figure 1. Top: schematic illustration of programmable nonvolatile memory for very large scale integrated processor applications, where every IrMn/Co/Ru/CoPt/CoO heterojunction can be programmed at-will into four different magnetic configurations. Bottom: schematic demonstration of 10 states memory and spin logic applications.

## Acknowledgements

This work was supported by the National Natural Science Foundation of China (Grant Nos. 12174219, 11774199, and 51871112), the 111 Project B13029 and the Major Basic Research Project of Shandong Province ZR2020ZD28.

## Reference

[1] Y. B. Fan, X. Han, X. N. Zhao, Y. N. Dong, Y. X. Chen, L. H. Bai, S. S. Yan, and Y. F. Tian, *ACS Nano* **16** (2022) 6878-6885.

# SYMPOSIUM 03.
## BIOMAGNETISM AND BIOMEDICAL APPLICATIONS.
## S3. INVITED ORAL PRESENTATIONS

# *In vivo* thin GMR probes for magnetic recording of single-neuron action potentials

Chloé Chopin[1], Frederike J. Klein[2,*], Patrick Jendritza[2,3,*], Mohsen Parto-Dezfouli[2], Aurélie Solignac[1], Claude Fermon[1], Pascal Fries[2,4], Myriam Pannetier-Lecoeur[1 *].

[3] SPEC, CEA, CNRS, Université Paris-Saclay, CEA Saclay 91191 Gif-sur-Yvette Cedex, France

[2] Ernst Strüngmann Institute (ESI) for Neuroscience in Cooperation with Max Planck Society, Deutschordenstraße 46. 60528 Frankfurt, Germany

[3] International Max Planck Research School for Neural Circuits, Frankfurt, Germany

[4] Donders Institute for Brain, Cognition and Behaviour, Kapittelweg 29, 6525 EN Nijmegen, the Netherlands

*myriam.pannetier-lecoeur@cea.fr

Neuronal currents reflect part of the information transmission in the brain and nervous system and are widely measured through electrophysiology tools at the surface or within the tissues. They have also a magnetic counterpart of very small amplitude, which can be detected at large scale for ensembles of neurons in magnetoencephalography, by means of femtotesla-sensors such as Superconducting Quantum Interference Devices.

In the quest for detecting at local scale, and furthermore *in vivo,* the magnetic signature of an action potential of a single neuron, spintronics sensors can be very good candidates since they bring sensitivity and miniaturization.

We fabricated very thin (25µm) probes, called magnetrodes, containing GMR elements [1], and combined them with standard tungsten electrodes to have a tool with the potential to detect both electric and magnetic signals *in vivo*.

Here we present the *in vivo* experiments performed in the rat hippocampus with these thin magnetrodes. We show how, with specific spike sorting procedures, we could access both electric and magnetic action potentials, and were able to detect single-cell magnetic recordings [2].

**Acknowledgements**

This work was supported by EU (FP7-600730-Magnetrodes) and ANR-DFG Project NeuroTMR (17-CE19-0021-01). This work was partly supported by the French RENATECH network.

**References**

[1] C. Chopin *el al.* https://dx.doi.org/10.1021/acssensors.0c01578

[2] F.J. Klein *et al. Under preparation.*

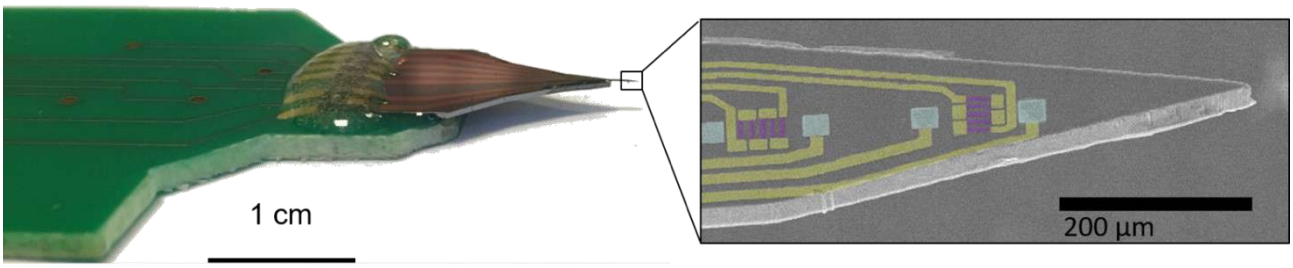



Figure: Magnetrode device, 25µm thick, containing two GMR sensing elements at the tip, and used to record locally *in vivo* magnetic signals associated with single-neuron action potentials.

# Tuning magnetic particle features and field conditions for biomedical treatments

*M. Angelakeris*[1]*

[1] *School of Physics, Aristotle University, 54124, Thessaloniki, Greece*

*agelaker@auth.gr

Magnetic fields and particles are nowadays an everyday practice in modern biomedical treatments. Biomedical treatments may be categorized in three major pillars: Handling, Diagnosis and Therapy. For many years magnetic field modes (Static or Dynamic) are used to handle (Biomagnetic Separation) or diagnose (MRI: Magnetic Resonance Imaging). More recently, magnetic nanoparticles came into play either as treatment enhancers or "cargo" carriers promoting variable therapeutic schemes (e.g., Drug Delivery, Magnetic Particle Hyperthermia). The diversity of magnetically driven applicability schemes urges not only for handling to diagnostic and therapeutic probes but for their combinatory exploitation as well. Moreover, effectiveness of a biomedical modality also puts additional constraints based on physiochemical interactions will living matter, typically hindering effective in-vivo performance. The impact of magnetic fields and particles is driven by their magnetic interaction and according to the treatment, certain prerequisites should be followed with respect to a). particle properties, b). field conditions and c). bio-performance. Generally, the stronger the field-particle interaction the more efficient the treatment.

To start with materials, particle properties will be discussed within the framework of their structure, stoichiometry and morphology focusing on their nanoscopic features. Namely, single, or two-phase magnetic nanoparticles of tuneable sizes, shapes, arrangements, will be evaluated in conjunction with prompt delivery of their adequate "cargo" at specific sites [1].

The second critical point has to do with the field conditions required to provide an effective scheme. Again, issues imposed by field amplitude, frequency, and modes (DC, AC, Pulsed) will be discussed with respect to Brezovich-Atkinson criterion and potential side-effects [2]. Variable field modes in Magnetic Particle Hyperthermia and Magneto Mechanical Actuation [3] will be proposed as alternatives to promote enhanced efficiency yet with mitigation of side-effects [4].

Finally, the beneficiary role of magnetic nanoparticles and fields will be evaluated beyond the in-silico case towards in-vitro, ex-vivo and in-vivo studies where aspects such as biocompatibility, and sustainability should be circumvented without sparing treatment efficiency [5, 6]. Certain applicable case studies of magnetic field-material combinations for handling, diagnosis, and therapy studies and theranostic schemes [7] will be proposed.

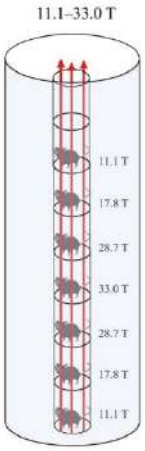
## References

[1] M. Angelakeris, *Biochimica et Biophysica Acta - General Subjects* **1861**(6) (2017) 1642–1651.
[2] A.R. Tsiapla et al., *Nanomedicine* **16**(11) (2021) 895-907.
[3] N. Maniotis et al. *Journal of Magnetism and Magnetic Materials* **470** (2019) 6–11.
[4] A. R. Tsiapla et al. *International Journal of Hyperthermia* **38**:1 (2021).
[5] K. Spyridopoulou et al. *Nanotechnology* **29**(17) (2018) 175101.
[6] A.R. Tsiapla et al., *Magnetochemistry* 8(2), (2022) 21.
[7] R. Tzoneva et al. *Magnetochemistry*, 8(10) (2022) 117.

# Magnetobiology and Magnetomedicine: Effects of Moderate and High Magnetic Fields on Living Organisms

*Vitalii Zablotskii[1,2]*, Tatyana Polyakova[1], Xin Zhang[2,3]*

[1] *Institute of Physics of the Czech Academy of Sciences, Na Slovance 2,18221, Prague, Czechia*
[2] *International Magnetobiology Frontier Research Center (iMFRC), Science Island, China*
[3] *High Magnetic Field Laboratory, CAS Key Laboratory of High Magnetic Field and Ion Beam Physical Biology, Hefei Institutes of Physical Science, Chinese Academy of Sciences, Hefei, China*
*zablot@fzu.cz




The biological and therapeutic effects of low magnetic fields have been the subject of numerous studies for over a hundred years. However, investigations of the biological effects of moderate and high magnetic fields are still at an early stage. Several new biological and therapeutic effects of moderate and high magnetic fields (MFs) will be discussed. Effects of a high static MF on lung cancer-bearing mice were examined in [1]. In mice treated 88 h with a 9.4 T static magnetic field, tumor growth and DNA synthesis were significantly inhibited, G2 cell cycle was arrested, while the ROS and P53 levels were increased. Surprisingly, the application of ultra-high static MF (up to 33 T) causes an anti-depressive effect on mice, enhancing the levels of oxytocin and c-Fos in the mice's brains [2]. This was the first attempt to apply ultrahigh static MFs to living organisms. It is important for clinical applications to treat a number of myopathies associated with the defective calcium regulation in muscle cells that exposure of skeletal muscle cells to a complex spatiotemporally modulated 70 mT magnetic field triggers a significant increase in cytosolic $Ca^{2+}$ levels leading to actin polymerization [3]. We found that low-frequency (4.2 Hz) rotating 0.4 T MF not only reduced the F-actin filament in cells, but also interfered with actin polymerization and depolymerization in vitro, which showed the direct action of the MF on actin dynamics [4]. This implies that low-frequency rotating MFs could inhibit breast cancer metastasis through F-actin modulation and its related pathways. A high static MF can control the diffusion of biologically active molecules including oxygen, hemoglobin, and drugs, thereby affecting many physiological processes in organisms, e.g., wound healing [5]. We suggest several new physical mechanisms of the MF impacts on endothelial and cancer cells by the MF interaction with chains of biogenic and non-biogenic magnetic nanoparticles on cell membranes [6]. The physiological sequences of the MF – cell interactions for organisms in health and disease are discussed. A prolonged exposure to a high static MF (1.0–8.6 T) with magnetic gradient 10 T/m can have negative effects on diabetic mice, especially mice with severe T1D, whereas quasi-uniform 9.4 T MFs did not produce the same effects, providing important information for the future development and clinical application of MFs, especially high-field MRI [7]. The analysis performed in [8] showed that specific ion channels in cells can be turned off and on by remotely applying a high gradient magnetic field, thus modulating the cell membrane potentials. The suggested model and mechanisms provide a general framework for identifying possible hidden mechanisms of biomagnetic effects associated with modulation of ion channel activity by high gradient static magnetic fields. The described effects and their underlying mechanisms are general and should take place in a large family of biological effects of MFs. The results are of great importance for further developing novel approaches in cell biology, cell therapy and medicine.

## Acknowledgements

Project is funded from the Mobility Program budget of the Czech Academy of Sciences and Chinese Academy of Sciences.

## References

[1] X. Yang et al., *Translational Oncology* **14** (2021) 101103.
[2] Y. Lv et al., *J Magn Reson Imaging* **56** (2022) 354.
[3] M. R. Ayala et al., *Biomaterials* **163** (2018) 174.
[4] X. Ji et al., *Research* (2023) (in press), DOI: 10.34133/research.0080.
[5] V. Zablotskii et al., *Cells* **11** (2022), 81.

[6] O. Gorobets et al., *Bioelectrochemistry* **153** (2023) 108390.
[7] B. Yu et al., *Zoological Research* **44**(2) (2023) 249.
[8] V. Zablotskii et al., In book: "Biological Effects of Static Magnetic Fields", Publisher: Springer Nature Singapore, 2023, Ed. Xin Zhang. 420p. ISBN: 9789811988684.

# SYMPOSIUM 03.
## BIOMAGNETISM AND BIOMEDICAL APPLICATIONS.
## S3. ORAL PRESENTATIONS

# Stress monitoring in intervertebral disc using bistable microwires.

*D. Kozejova[1], M. Kohan[2] , S. Lancos[2] , R. Hudak[2], R. Varga[1,3]**

[1] *Inst. Phys., Fac. Sci., UPJS, Park Angelinum 9, Kosice, Slovakia*

[2] *Dep. of Biomed. Eng. Meas., Fac. Mech. Eng., TUKE, Letna 1/9, 042 00, Kosice, Slovakia*

[3] *RVmagnetics a.s., Nemcovej 30, 040 01, Kosice, Slovakia*

*rastislav.varga@upjs.sk

For decades, implantable sensors have been used in research to provide comprehensive understanding of the biomechanics of the human musculoskeletal system [1]. An implantable, freestanding, minimally invasive, intervertebral disc pressure sensor would vastly improve the knowledge of spinal biomechanics, the understanding of spinal disease and clinical indications for surgical interventions in disc-related pathology [2].

According to our previous study [3] it is possible to use bistable magnetic microwire for contactless sensor of the pressure. Based on this possibility, the new concept for measuring the pressure in intervertebral discs implants using bistable magnetic microwire as minimally invasive implatable sensor has been created.

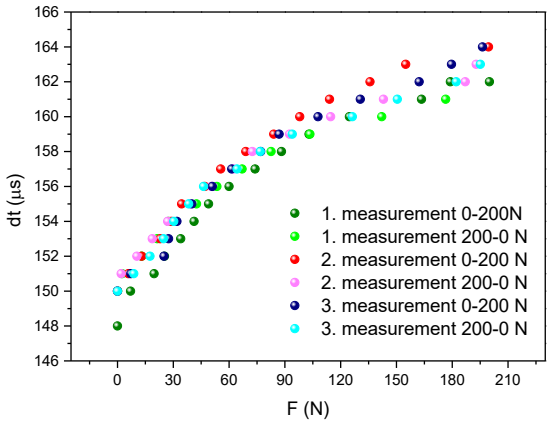



Figure: Magnetic response (switching time change) of bistable microwire under applied load.

Bistable microwires were glued on the surface of 3D printed intervertebral disc made of Polylactide (PLA). Measurements were made in series of 3 runs with a gradual increase in the load from 0 to 200 N and a gradual release of the load from the finished position 200N to 0 N. Repeated measurements of this dependence show that with an increase in the force, the magnetic response increases monotonously. The observation of the measurements shows flattening of the intervertebral disc (see figure). occurs and the horizontally located microwire is stretching. On the other hand, the measurement shows that torsion occurs when the load is applied.

## Acknowledgements

This publication is the result of the project implementation SMARTfix ITMS2014+: 313011BWQ1 and CEMBAM ITMS2014+: 313011V358 supported by the Operational Programme Integrated Infrastructure funded by the European Regional Development Fund. This work was partially supported by Slovak Grant Agencies APVV-16-0079 and VEGA 1/0180/23

## References

[1] Ledet EH, D'Lima D, Westerhoff P, et al. Implantable sensor technology: from research to clinical practice. The Journal of the American Academy of Orthopaedic Surgeons. 2012, 20(6):383-392.

[2] Moore MK, Fulop S, Tabib-Azar M, Hart DJ. Piezoresistive pressure sensors in the measurement of intervertebral disc hydrostatic pressure. Spine J. 2009, 9(12):1030-4.

[3] D. Kozejova, R. Varga, Bistable magnetic microwire for contactless sensor of intracranial pressure, JMMM, 569; 2023, 170473.

# Measuring the Heating Power of Magnetic Nanoparticles

*Lise Grüner Hanson[1], Bianca Laura Hansen[1], Thomas Veile[1], Mathias Zambach[1], Niels Bech Christensen[1] and Cathrine Frandsen[1]**

*[1]Department of Physics, Technical University of Denmark, Kongens Lyngby, Denmark*

**fraca@fysik.dtu.dk*

The hysteretic properties of magnetic nanoparticles allow for heating which can be utilised in a localized cancer treatment called magnetic hyperthermia. Clinical trials with magnetic hyperthermia have already been conducted [1]. For magnetic hyperthermia to be efficient it is of great importance to improve the heating power of the magnetic nanoparticles.

Within the field of magnetic hyperthermia, there is a need for an accurate measurement procedure for quantifying the heating power of magnetic nanoparticles in order to support comparison between different candidates of particles [2]. Usually, this quantity is measured through AC-calorimetry in which a magnetic nanoparticle solution is placed inside an induction coil, which provides an alternating magnetic field while a thermometer records the temperature inside the sample [3].

A recent inter-laboratory study found that this standard method for quantifying the heating power is subject to large systematic errors of unknown origin [2]. Across the 17 participating state-of-the-art laboratories the intrinsic loss power (heating power normalized by the frequency times the squared amplitude of the applied magnetic field) displays a standard deviation of the mean of 40 %. This situation is highly problematic for comparing and developing new efficient magnetic nanoparticles and highlights the need for an accurate standard measurement protocol for quantifying the heating power.

In this contribution, we present a newfound importance of sample environment (Fig. 1 left and centre). We demonstrate how heat transfer from the sample can be analysed (Fig. 1 right) and how local temperature increments in the sample insulation can influence the measured heating power. Based on our results we propose strategies for obtaining better estimates for the heating power of magnetic nanoparticles.

## References

[1] Chang D., et al. Frontiers in pharmacology **9** (2018) 831.
[2] Wells J., et al. International Journal of Hyperthermia **38** (2021) 447
[3] Wildeboer, R. R., et al. Journal of Physics D: Applied Physics **47** (2014) 495003
[4] Hanson L.G., et al. Submitted (2022)

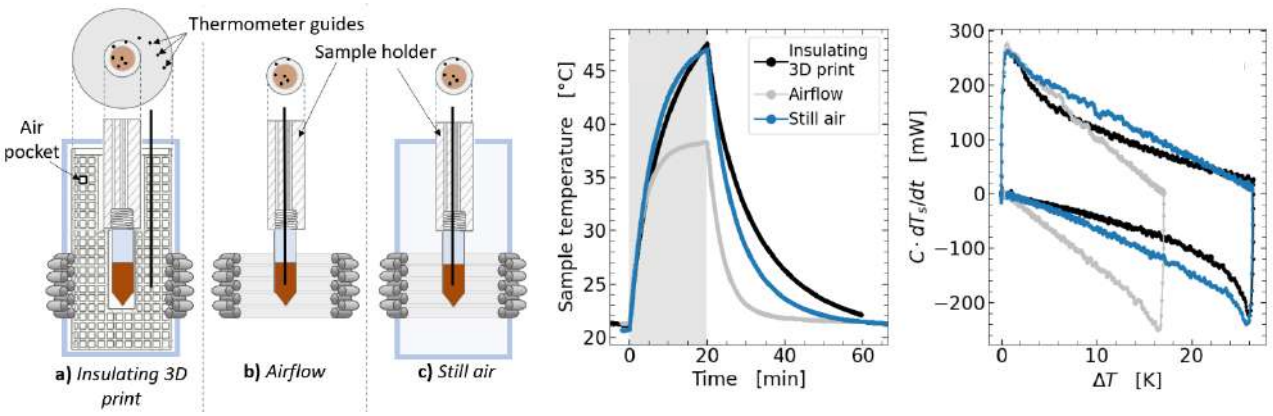



Fig 1: **Left:** AC calorimetric setups with three different sample environments. **Centre:** Sample temperatures measured in the three different sample environments seen in the left figure. **Right:** The data in the central plot converted into a plot of the time derivative of the temperature vs temperature change. From [4].

# Nanoparticle-mediated magnetic hyperthermia improves thrombolytic therapy in thrombosis

*David Cabrera[1]*, Maneea E Sharifabad[1], Jacob A Ranjbar[2], Neil D Telling[1] and Alan GS Harper[2]*

[1] *School of Pharmacy and Bioengineering, Guy Hilton Research Centre, Keele University. Thornburrow Drive, Harshill, Stoke-on-Trent ST4 7QB, United Kingdom*

[2] *School of Medicine, Keele University, David Weatherall Building, ST5 5BG, United Kingdom*

*d.c.cabrera@keele.ac.uk

Ischemic stroke is one the main causes of death and disability in developed countries. Strokes occur when the blood supply to a part of the brain is interrupted due to the blockage of blood vessels, leading to potential irreversible tissue damage. To re-establish blood and prevent further damage in the affected brain tissue, intravenous injection of the clot-busting agent tissue plasminogen activator (tPA) is provided to breakdown a blood clot. However this thrombolytic therapy is currently restricted by its short time window of efficacy and the risk of uncontrolled haemorrhage. Here, we investigated whether platelet-targeted magnetic hyperthermia (MH) induced by Iron Oxide Nanoparticles (IONPs) could be used to improve the efficacy of enzyme-mediated thrombolysis. PAC-1, an antibody that recognises activated platelets taking part in blood coagulation, was conjugated to IONP to produce platelet-targeted IONPs. *Ex vivo* generated human blood clots were exposed to tPa in the presence or absence of MH. MH was found to enhance the clot-lysis action of tPa by reducing the weight of tPa + MH treated clots to 75.6 ± 1.5 % of untreated control clots, compared to 81.3 ± 2.2 % clots treated with tPa alone ($n = 15$, $P < 0.05$)[1]. Through 3D reconstruction of confocal images, platelet-targeted MH was found to increase permeability of blood clots to 70 kDa fluorescent dextran, which has similar molecular weight to that of tPa (Figure 1). Electron microscopy images revealed that localised MH enhances tPa-mediated clot disruption. Remarkably, viability tests evidenced no increase in cell death of HUVEC 3D cell cultures, which are those located in the internal layer of blood vessels, when exposed to f-IONP-mediated MH. Platelet-targeted magnetic nanoparticles could act as an adjuvant to enhance the efficacy of current clot busting therapies through the induction of localised MH. Nanoparticles-mediated MH could be also explored for improving the treatment of other cardiovascular conditions such as Venous Thromboembolism.

## Acknowledgements

This work was supported by the Wellcome Trust [207617/Z/17/Z and 218671/Z/19/Z]

## References

[1] D. Cabrera, M. Eizadi Sharifabad, J. A. Ranjbar, N. D. Telling, and A. G. S. Harper. *Journal of Thrombosis and Haemostasis,* vol. 20, no. 11, pp. 2556-2570, 2022.

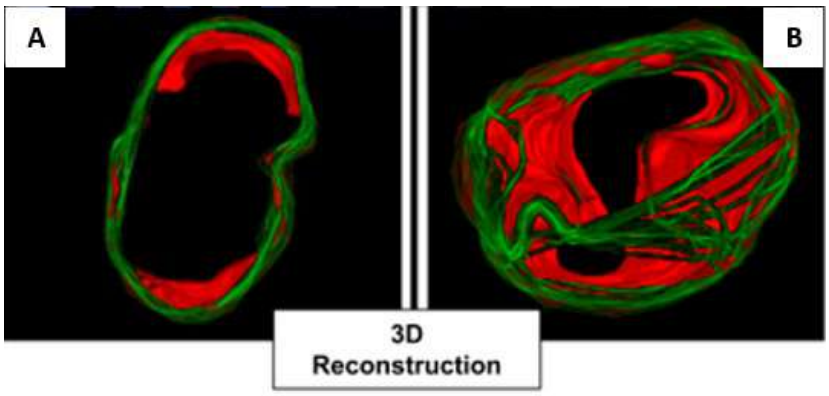



Figure 1: 3D reconstruction of a blood clot (A) untreated or (B) treated with nanoparticle-mediated magnetic hyperthermia. Green colour: Blood clot contour; Red colour: fluorescent dextran with similar molecular weight to that of clot-busting enzymes. *Extracted from ref. 1*

# Magnetic Microwire and Static Magnetic Field Modulates Neuron Outgrowth

*Katelyn Neuman[1]*, Xiaoyu Zhang[1], Brian T. Lejeune[1], Dominic Pizzarella[1], Manuel Vázquez[2], Abigail Koppes[1], Laura H. Lewis[1], Ryan Koppes[1]*

[1] *Northeastern University, Boston, USA*

[2]*Instituto de Ciencia de Materiales de Madrid, CSIC, Madrid, 28049 Spain*

*neuman.k@northeastern.edu

Injuries to the peripheral nervous system are estimated to occur in 2-5% of trauma hospital admissions and can severely impact patients' quality of life [1]. Current surgical interventions are limited and, in ~50% of cases, ineffective at restoring patients to their previous functional level [2]. Our current research aims to improve outcomes by understanding and harnessing *magnetic microwires and magnetic stimulation, which increased total neruron outgrowth while strongly biasing growth parallel to the wire.* These results build on our previous work highlighting the promise of static low magnitude (~15 mT) magnetic fields, increasing neurite outgrowth by 60% [3]. Additionally, linear topographical cues have been demonstrated throughout the literature to strongly influence neuron directional outgrowth, but never using highly responsive magnetic microwires [4, 5].

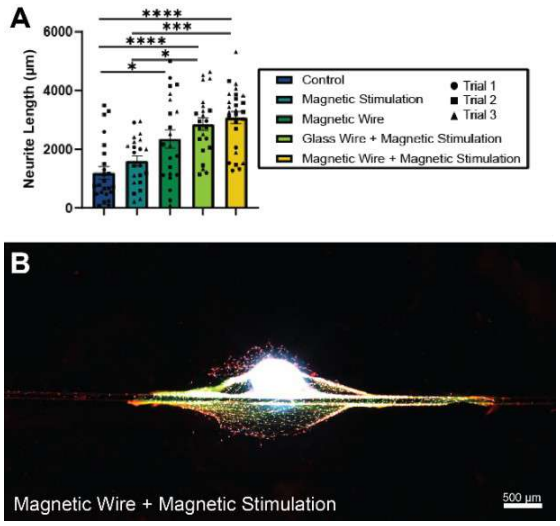



Figure 1: (A) Comparison of the longest neurite for all five experimental groups. Each point represents one DRG replicate. Significant differences are denoted with *, ***, and ****. Error bars indicate the standard error of mean (SEM). (B) DRG on magnetic wire + magnetic stimulation. Green fluorescence labels neurites (neurofilament-heavy), red fluorescence labels glia cells (S100β), and blue fluorescence labels cell nuclei (counterstained with DAPI).

These magnetic microwires (diameter ~80 microns), fabricated using the modified Taylor-Ulitovsky method [6], are composed of $Fe_{73}Si_{11}B_{13}Nb_3$, metallic glass core coated with inert pyrex glass. This class of material is extraordinarily responsive to small (< 100 A/m) applied external magnetic fields, with a saturation induction around 1.6 T of the metallic core, coercivity in the range of several A/m, and permeability up to around $10^5$. Magnetic field conditions with and without the magnetic wire were modeled using Ansys Simulation Software. Dorsal root ganglia (DRGs) were subjected to five different parameters: control (no magnetic stimulation, no wire), magnetic stimulation, magnetic wire, glass wire + magnetic stimulation, magnetic wire + magnetic stimulation. After five days, DRGs were fixed, immunocytochemistry was performed, and imaged. Images were analyzed for total neurite outgrowth, neurite density, longest outgrowth, and directional biases. Further, changes in gene expression between groups will be evaluated with RNA sequencing. Data was assessed with the Kruskal-Wallis Test with a post hoc Dunn's multiple comparison test.

The combination of the magnetic wire and magnetic field stimulation resulted in significant increase in total neurite outgrowth (by 120%), neurite density (by 500%), and the longest neurite extension (by 95%) compared to the unstimulated controls. Additionally, magnetic wire + magnetic stimulation drove a strong directional bias, with 50% more growth parallel to the wire. This study furthers our knowledge of the interactions between neurons and magnetic materials/fields, bringing us one step closer to improving patient outcomes after nerve injuries.

**Acknowledgement**

This research was conducted under the auspices of Northeastern University, the Fulbright Scholar Program and ICMM/CSIC, Madrid, Spain.

## References

[1] L. Robinson, *Muscle, and Nerve.* **23**(2000) 863-873
[2] J. Thomson, *Plast. Reconst. Surg.* **2**(2005) 116
[3] K. Neuman, X. Zhang, C. Mills, R. Koppes, L. Lewis, A. Koppes, *J. Neural Eng.* **20**(2023) 016020
[4] D. Hoffman-Kim, J. Mitchel, R. Bellamkonda, *Annu. Rev. Biomed. Eng.* **12**(2010) 203-231
[5] F. Liu, J. Wu, T. Zheng, Q. Han, Y. Liang, … Y. Yang, *Stem Cells Int.* (2021) 8124444
[6] M. Vázquez, in *Handbook Magn. Mater.(H.Kronmüller & S. Parkin eds.) Wiley Publ.* **4**(2007) 2193

# Synthetic Antiferromagnetic Nanoplatelets: Surface Chemistry and Physical Properties for Magnetomechanical Cell Manipulation

*Mariia Efremova[1]*, Jianing Li[1], Marc A. Verschuuren[2], Bert Koopmans[1], Reinoud Lavrijsen[1]*

[1] *Department of Applied Physics and Science Education, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, the Netherlands*

[2] *SCIL Nanoimprint Solutions, Philips Group Innovation, HTC 11, 5656 AE Eindhoven, the Netherlands*

*m.efremova@tue.nl

Spatiotemporal application and modulation of mechanical forces in biological systems are of great interest for studying a wide range of mechanotransduction processes involved in therapy, including cancer mechano-immunoengineering [1]. Remote cell manipulation can be implemented in an external magnetic field by using magnetic nanoparticles that serve as the "nanotransducers," converting the energy of a magnetic field into a mechanical force in a 1-100 pN range [2]. To maximize this force (or a torque), magnetically anisotropic structures (for example, platelet-shaped) are preferred [3].

In this work, we study synthetic antiferromagnetic (SAF) nanoplatelets (NPs) with perpendicular magnetic anisotropy for future magnetomechanical applications. Our group recently published the protocol for fabricating SAF NPs of 123.3 ± 3.3 nm diameter based on substrate conformal imprint lithography [4]. Yet, this fabrication route requires certain modifications and development of the functionalization steps to make SAF NPs stable and biocompatible while retaining their magnetic properties.

In particular, introducing gold layers at the top and bottom of the SAF stack (Figure 1a) does not affect its magnetic characteristics, namely the antiferromagnetic state with two clear on/off switches (Figure 1b). It also allows for the surface modification of NPs with thiol-containing ligands (Figure 1c). In my talk, I will discuss our current strategies to fabricate and functionalize the gold-coated SAF NPs circumventing the associated issues that include the adjustment of non-magnetic layers, hydrophobicity of the as-deposited Au surface, colloidal instability of SAF NPs, and others. Finally, I will give an overview of using (gold-coated) SAF NPs for magnetomechanical cell manipulation, including recent examples from the literature.

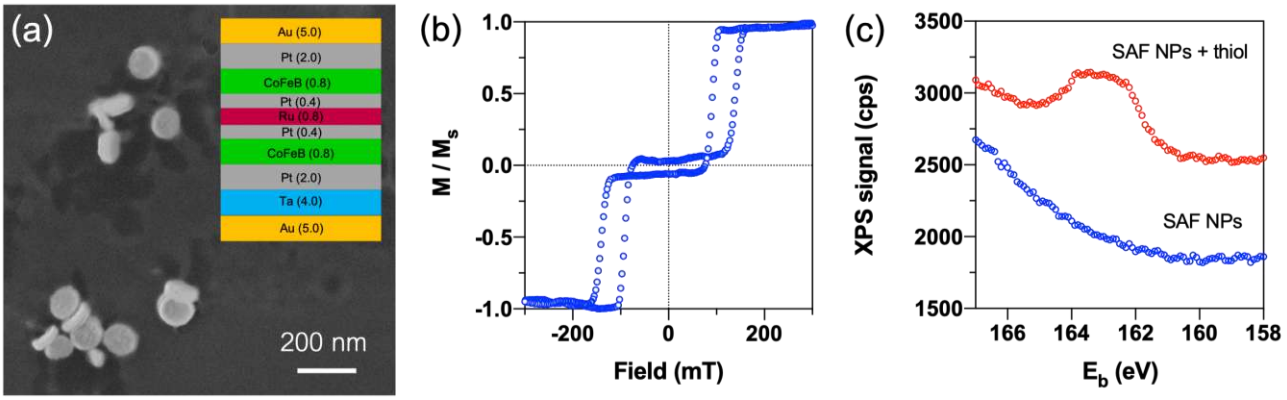



Figure 1. Gold-coated SAF NPs for magnetomechanical applications: a) SEM image of the NPs after the release, the inset shows the layered structure with thicknesses in nanometers; b) Hysteresis loop of a SAF thin film, MOKE measurement; c) XPS data on the patterned SAF NPs before and after the functionalization with a thiol (11-mercaptoundecanoic acid).

## Acknowledgements

This project has received funding from the European Union's Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement no. 899987.

## References

[1] M. Huse, *Nat. Rev. Immunol.* **17** (2017) 679–690.
[2] J.-U. Lee, W. Shin, Y. Lim, et al. *Nat. Mater.* **20** (2021) 1029–1036.
[3] M. Goiriena-Goikoetxea, D. Muñoz, I. Orue, et al. *Appl. Phys. Rev.* **7** (2020) 011306.
[4] J. Li, P. van Nieuwkerk, M.A. Verschuuren, et al. *Appl. Phys. Lett.* **121** (2022) 182407.

# $Fe_3O_4$-$SiO_2$ mesoporous core/shell nanoparticles for potential magnetic field-induced ibuprofen-controlled release

*A. López-Ortega*[1,2,*], *L. García*[1,2], *E. Garayo*[1,2], *I. Galarreta*[1], *G. Cruz-Quesada*[1,2], *J. J. Garrido*[1,2], *C. Gómez-Polo*[1,2], *J. I. Pérez-Landazábal*[1,2]

*1Dept. de Ciencias, Universidad Pública de Navarra, Campus de Arrosadia, 31006 Pamplona, Spain.*
*2Institute for Advanced Materials and Mathematics (INAMAT2), Universidad Pública de Navarra, Campus de Arrosadia, 31006, Pamplona, Spain.*
*lopezortega.alberto@gmail.com

Hybrid magnetic nanoparticles made up of iron oxide, $Fe_3O_4$, presenting high magnetization and large surface area hold great potential for multimodal therapiesas such as tragered drug delivery, magnetic resonance imaging, and detection, diagnosis, and magnetic field–assisted radiation treatment of cancerous cells. [1,2] In this aim, we have prepared a multimodal hybrid core/shell nanoparticles composed of $Fe_3O_4$ core and mesoporous $SiO_2$ shell through a two steps seeded-mediation growth synthesis, combining solvothermal and sol-gel approaches and using organic molecules as porous scaffold template. [3] As-synthesized nanoparticles have been characterized by means of transmission electron microscopy, x-ray powder diffraction, nitrogen adsorption-desorption isotherms, Fourier transform-infrared spectroscopy, AC and DC magnetometry, calorimetric hyperthermia measurements, thermogravimetric analysis, and ultraviolet-visible spectroscopy. The system presents a particle diameter of 30(5) nm (9 nm core and 10nm shell thickness) with superparamagnetic character, saturation magnetization of 32 emu/g, and a strong AC magnetic-field-induced heating response (SAR = 27 W/g, measured at 400 Oe and 307 kHz). Using ibuprofen as a model drug, the large surface area (231 $m^2/g$) of the porous structure presents high molecule loading capacity (10 % in weight) and controlled drug release efficiency (67 %) can be achieved using an external AC magnetic field in short time periods (5 min). In addition, it has been demonstrated that the magnetic field induced drug release shows an enhanced efficiency in comparison with sustained release at fixed temperatures (47 and 53 % for 37 and 42 ºC) even if the temperature reached at 5 minutes magnetic field exposition is well below (31 ºC). [3]

## References

[1] X. Lu, *et al.* RSC Advances 7, (2017) 32049
[2] N. Lee, *et al.* Chem. Rev. 19 (2015) 10637
[3] L. García, *et al.* Langmuir 39 (2023), 211

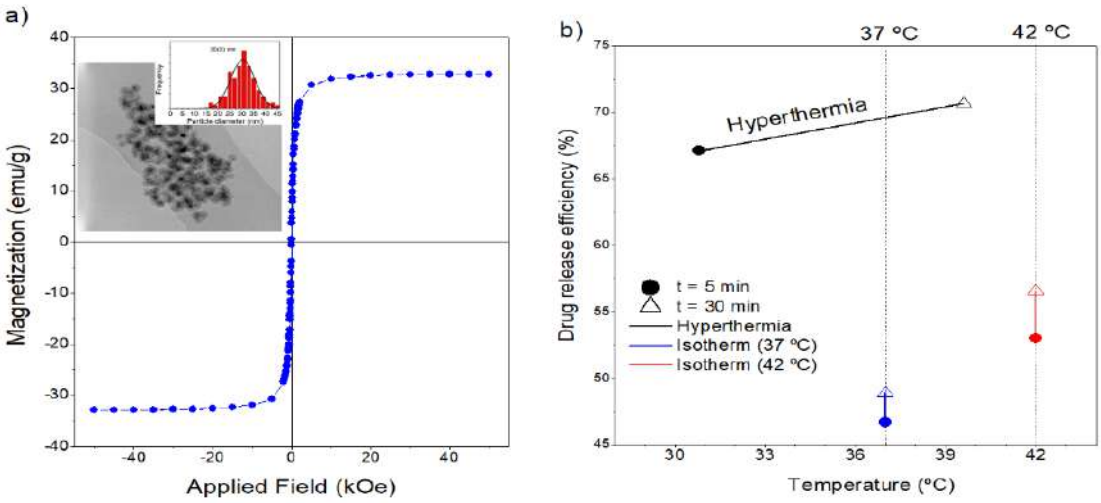



Figure 1. (a) Room temperature hysteresis loop, transmission electron image and particle size histogram for hybrid core/shell nanoparticles. (b) Comparative study of non-cumulative ibuprofen release experiments between magnetic field-induced and isothermal approaches.

# Studying the magnetic hyperthermia effect of magnetosomes in a three-dimensional tumour model

*Alicia Gascón Gubieda[1], Lucía Gandarias[1], Danny Villanueva-Álvaro[2], Alaine Urrutia[1], Alicia Muela[1], M. Luisa Fernández-Gubieda[2], Ana García-Prieto[3], Ana Abad Díaz de Cerio[1]*

[1]*Depto. Inmunología, Microbiología y Parasitología, Universidad del País Vasco (UPV/EHU), Leioa, Spain*

[2]*Depto. Electricidad y Electrónica, Universidad del País Vasco (UPV/EHU), Leioa, Spain*

[3]*Depto. Física Aplicada, Universidad del País Vasco (UPV/EHU), Bilbao, Spain*

*alicia.gascon@ehu.eus

Magnetotactic bacteria (MTB) are a group of bacteria that can align themselves along the earth's magnetic field thanks to organelles called magnetosomes. Magnetosomes are magnetic nanoparticles enveloped by a lipid bilayer membrane that can be made of magnetite ($Fe_3O_4$) or greigrite ($Fe_3S_4$) of high chemical purity, and have very uniform morphology and a narrow size distribution. These properties, together with their low toxicity and their bio-compatibility make them good candidates for many biomedical applications, such as magnetic hyperthermia [1,2,3].

However, most of the studies on the medical applications of magnetosomes so far have been performed on 2D cell cultures, which lack many of the properties of tumours and living organisms, and further development of magnetosomes as hyperthermia agents will require studies in more biomimetic systems. For this purpose we have employed spheroids of the human lung carcinoma cell line A549. These are 3D tumour models with properties, such as nutrient and oxygen gradients, that mimic those of human solid tumours. Our study had two aims: first to determine how magnetosomes are degraded in 3D tumour models to evaluate how long they can be used for treatment; and second, to determine the effectiveness of magnetosomes as hyperthermia agents in this system.

To study the degradation of magnetosomes from Magnetospirillum gryphiswaldense, A549 cells were incubated with them for 2 h at a concentration of 30 µg/ml and then placed on low-attachment plates for the formation of spheroids. The degradation was followed by means of magnetometric measurements and Fe K- edge X-ray Absorption Near Edge Structure spectroscopy (XANES) at the ESRF synchrotron (Grenoble, France). Both techniques revealed that the magnetite of magnetosomes is degraded very slowly within spheroids, with only 30% of the iron degraded from magnetite to maghemite, and up to 90% of the magnetic signal still present 30 days after internalisation.

To test the effectiveness of magnetic hyperthermia in 3D tumour models, we incubated the spheroids with magnetosomes and immediately exposed them to an alternating magnetic field of 157 Hz and 48 mT for 45 minutes. We found that the treatment resulted in lethality of up to 40% in the first 48 h after a single hyperthermia session.

The low degradation rate of magnetosomes in tumour-models, combined with the high effectivity of magnetosomes for magnetic hyperthermia, suggests that magnetosomes could be used for long-term hyperthermia treatments.

## References

[1] E. Alphandéry, *et al. Int J Hyperthermia*, 29 (8) 801-809 (2013)
[2] A. Muela, *et al. Journal of Physical Chemistry C* 120, 24437-24448 (2016) [3] D. Gandia *et al. Small 15*, 1902626 (2019)

# Mitigating SAR Dependence on Measurement Devices in Magnetic Nanoparticle Hyperthermia

*Sergiu Ruta[1], Yilian Fernández-Afonso[2], Samuel E. Rannala[3], M. Puerto Morales[4], Sabino Veintemillas-Verdaguer[4], Carlton Jones[5], Lucía Gutiérrez[2], David Serantes[6*] and Roy W. Chantrell[3]*

[1]*College of Business, Technology and Engineering, Sheffield Hallam University, United Kingdom*
[2]*Department of Analytical Chemistry, Universidad de Zaragoza, Spain*
[3]*Department of Physics, University of York, United Kingdom*
[4]*Materials Science Institute of Madrid (ICMM/CSIC), Spain*
[5]*nanoTherics Ltd Brookside Farm, Dig Lane, Warrington, WA2 0SH, United Kingdom*
[6]*Applied Physics Department, Universidade de Santiago de Compostela, Spain*
*david.serantes@usc.es

Accurately understanding the heating performance of magnetic nanoparticles (MNPs) under AC fields is crucial for the development of hyperthermia-based applications. Typically, the specific absorption rate (SAR) is used to describe this performance, which is obtained from the temperature change (ΔT) versus time (t) curve. However, such estimates can be highly uncertain, resulting in vastly different SAR values for the same particles measured on different equipment or in different laboratories [1]. This lack of control hinders the advancement of MNP-mediated heat-triggered technologies.

Here, we propose a new protocol to reduce the dependence of SAR estimates on specific measurement devices. First, we have identified the problems associated to the calorimetric methods, such as the inhomogenous temperature distribution along the sample or the different heat loss mechanisms. Then, we have developed an alternative way of analyzing the T(t) curves, changing from the initial slope, generally used in the most extended approaches, to the analysis of the peak generated once the AC field is switched off. This approach minimizes the influence of the apparatus' thermal properties on SAR determination. This "peak analysis" allows performing very fast measurements, so a further development of the methodology includes to perform very rapid field on/off switches resulting in a zigzag-shaped ΔT (t) curve (see Figure). This type of measurement allows quickly obtaining a large number of SAR values in a short period of time. Moreover, it can also allow tracking possible SAR variations over time. The proposed methodology has been experimentally tested using three different devices achieving better interlaboratory comparison then when the classical methods to calculate SAR are used.

Overall, we propose a change of paradigm in the way SAR is calculated, aiming to improve the reliability of magnetic nanoparticle heating performance measurements.

## References

[1] Wells, J., Ortega, D., Steinhoff, U., Dutz, S., Garaio, E., Sandre, O., ... & Radiomag Consortium. *Int. J. Hyperthermia*, **38** (2021), 447-460.

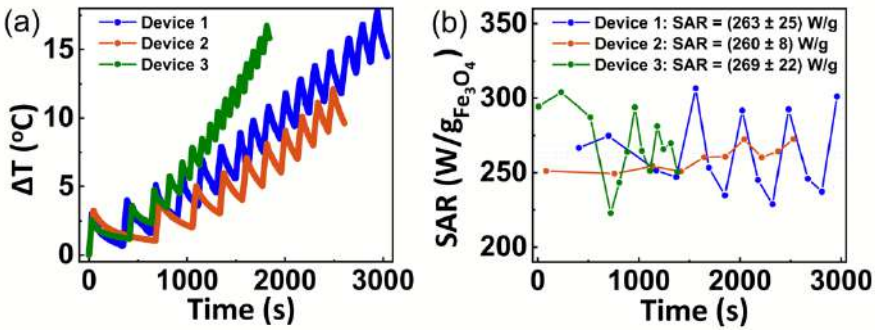



Figure: (a) Zig-zag measurement of the heating properties of magnetic nanoparticles performed with three different devices. (b) SAR values calculated using the "peak analysis" from the measurements in panel (a).

# Synthetic Antiferromagnet Disk Particles for Hyperthermia

*S. Scheibler[1], I. K. Herrmann[2], S. Helbig[3], D. Suess[3], M. Krupiński[4], M. Graeser[5], and H. J. Hug[1,6*]*

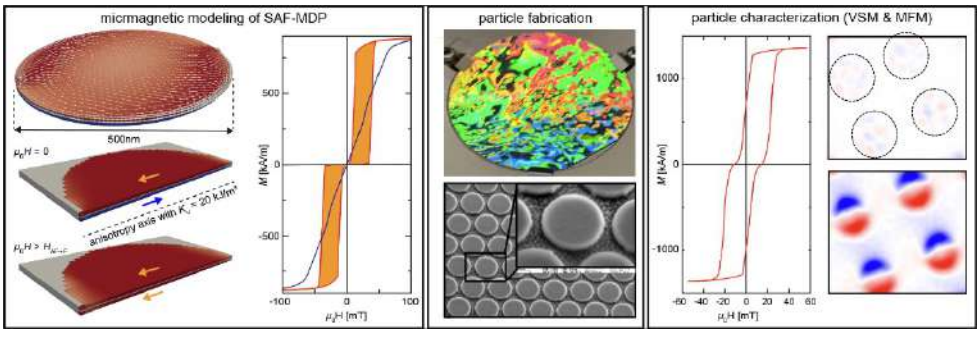
[1] *Empa, Überlandstrasse 129, 8600 Dübendorf, Switzerland*
[2] *ETH Zürich, Sonnegstrasse 3, 8092 Zürich, Switzerland*
[3] *Physics of Functional Materials, University of Vienna Kolingasse 14-16, 1090 Wien, Austria*
[4] *The Henryk Niewodniczanski Institute, Radzikowskiego 152, 31-342 Kraków, Poland*
[5] *Universität zu Lübeck, Ratzeburger Allee 160, and Fraunhofer Research Institution of Individualized and Cell-based Medical Engineering , IMTE, Mönkhofer Weg 239a, 23562 Lübeck*
[1] *Department of Physics, University of Basel, 4056 Basel, Switzerland*

hans-josef.hug@empa.ch

Chemically synthesized superparamagnetic nanoparticles (SP-MNP) have limited heat generation due to the low magnetization of the typically used oxidic materials, wide particle size distribution, and consequently narrow shape of their magnetic hysteresis loop. To overcome these limitations, top-down fabricated disk-shaped particles were designed that either contain a single magnetic layer forming a magnetic vortex (V-MDP) or synthetic antiferromagnet particles (SAF-MDP) consisting of at least one pair of antiferromagnetically-coupled ferromagnetic (F) layers [1].

Current approaches using SAF-MDP, which consist of ferromagnetic layers with in-plane magnetization, do not exhibit hysteretic losses. In contrast, a small hysteretic loss was demonstrated with V-MDP arising from the annihilation/nucleation of the vortex. SAF-MDP with perpendicular anisotropy include at least two stacks of Co/Pt or Co/Pd multilayers that are antiferromagnetically coupled through the RKKY interaction of a thin Ru interlayer. Li et al. [2] recently reported $M(H)$-loops with substantial hysteretic losses, but the field required to switch the particles from their antiferromagnetically aligned ground state to the ferromagnetic state ($H_{AF\rightarrow F}$) remained rather large for hyperthermia applications. Furthermore, the particles consist of expensive non-magnetic materials, which limits their potential for economically viable applications.

In our work [3], we utilize micromagnetic modelling to optimize the system the parameters of in-plane SAF-MDP to achieve an antiferromagnetically coupled (zero-moment) ground state and an abrupt switching into a ferromagnetically aligned state in an applied field $H_{AF\rightarrow F}$ to maximize the hysteretic loss. The primary difference from earlier approaches is that the ferromagnetic layers have an in-plane uniaxial anisotropy that is strong enough to align the magnetic moments, resulting in an antiferromagnetically coupled ground state via stray fields. This particle design also achieves an $H_{AF\rightarrow F}$ of less than 50 mT, which is compatible with the oscillatory fields used in hyperthermia. The magnetic multilayer was sputter-deposited onto a 50 nm-thick Ge sacrificial layer on a silicon wafer. Self-assembled polystyrene spheres were used as an etch mask for the successive nanopatterning of disk-shaped islands. Finally, these islands were detached from the supporting wafer by dissolving the Ge layer. Using vibrating sample and Kerr magnetometry, as well as high-resolution in-field magnetic force microscopy, we analyzed the magnetic properties of the SAF-MDP. The results closely matched the design goals and micromagnetic simulation results. We observed a turn-on/turn-off magnetism of the SAF-MDP and hysteretic losses that approached the theoretical limit given by the magnetic material with the highest saturation magnetization and a perfectly rectangular hysteresis loop. Moreover, experiments mapping the hysteretic loss of SAF-MDP suspensions for different operation conditions revealed a substantially higher heating efficiency than that obtained with SP-MNP.




## References:

[1] M. Goiriena-Goikoetxea, Appl. Phys. Rev. **7** (2020) 011306

[2] J. Li, P. van Nieuwkerk, M. A. Verschuuren, B. Koopmans, & R. Lavrijsen, Appl. Phys. Lett. 121 (2022) 182407

[3] H. J. Hug and D. Suess (2021) Patent Nr. PCT/EP2021/083572.

# Targetable thermomagnetophoretic nanopumps for theranostics

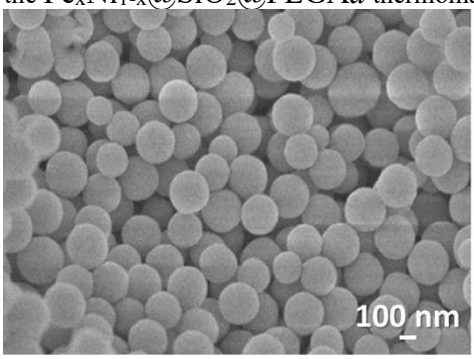
*Carlos Frade-González,[1,2] Ricardo Fernández Cisnal,[2] Francisco José García-Cozar,[2,3] Daniel Ortega[1,2,3]*

[1] *Dept. of Condensed Matter Physics, University of Cádiz, 11510 Puerto Real, Spain*
[2] *Institute of Research and Innovation in Biomedical Sciences of Cádiz (INiBICA), 11002 Cádiz, Spain*
[3] *Dept. of Biomedicine, Biotechnology and Public Health, University of Cádiz, 11003 Cádiz, Spain*
[4] *IMDEA Nanoscience, 28049 Madrid, Spain*

*daniel.ortega@uca.es

Despite their approval for human use, nanoparticles of ferrimagnetic iron oxides - namely maghemite ($\gamma$-$Fe_2O_3$) and magnetite ($Fe_3O_4$) - often underperform for drug release and hyperthermia, thus requiring higher doses. This is partly due to the magnitude of their magnetic moment as well as their magnetic anisotropy [1].
We have designed a thermomagnetophoretic nanopump for controlled heating and drug release. This nanopump consists of several layers, namely: (i) a core consisting of a controlled aggregate of $Fe_xNi_{1-x}A_y$ nanoparticles coated with a chelator that prevents Ni desorption; (ii) a layer of amorphous silica for the containment of the aggregates – either in liquid suspension or compacted, as needed -; (iii) a layer of poly(lactic-co-glycolic acid) derivative (PLGA*d*), a thermosensitive polymer with shape memory that retain and release the biomolecules of interest; (iv) optional surface linkers to facilitate the bonding of targeting biomolecules. By applying an external ac magnetic field, the core aggregates generate a thermophoretic effect that leads to an externally actuated and progressive PLGA destructuring, and hence to the controlled release of the inner biomolecules.

**Figure 1:** SEM image of the $Fe_xNi_{1-x}$@$SiO_2$@PLGA*d* thermomagnetophoretic nanopumps




**Acknowledgements**

We acknowledge support from the Ministry of Science through the grant PID2020-117544RB-I00 and the Ramón y Cajal grant RYC2018-025253-I.

**References**

[1] C. Blanco-Andújar, F. J. Teran and D. Ortega “Current outlook and perspectives on magnetic hyperthermia mediated by iron oxide nanoparticles” in Iron oxide nanoparticles for biomedical applications: synthesis, functionalization and application, Ed. Sophie Laurent and Morteza Mahmoudi (2017) Elsevier

# From the role of interactions in the specific absorption rate to the determination of the temporal temperature rise profile in magnetic hyperthermia

*François Vernay[1]*, Vincent Russier[2], Jean-Louis Déjardin[1] and Hamid Kachkachi[1]*

[1] *Laboratoire PROMES CNRS (UPR-8521) & Université de Perpignan Via Domitia, Rambla de la thermodynamique, Tecnosud, 66100 Perpignan, France*
[2] *ICMPE CNRS (UMR-7182) & Université Paris-Est Créteil, 2-8 rue Henri Dunant, 94320 Thiais, France*
*francois.vernay@unv-perp.fr

Magnetic hyperthermia consists in converting electromagnetic energy into heat by applying an external AC magnetic field to an assembly of magnetic nanoparticles. This results in a very localized temperature rise which can be useful in medicine and catalysis. However, several fundamental aspects must be fully analyzed in order to better understand how to control the temperature space-time profile within magnetic nanoparticle assemblies. In particular, in view of building multi-scale sensible models for future developments in magnetic hyperthermia, two important points need to be clarified: 1) the role of dipolar interactions (DI) in the optimization of the specific absorption rate (SAR), and 2) the heat diffusion within the assembly and through its interface with the surrounding environment.
Here, we address comprehensively these two issues. We first show how to determine the SAR of the assembly both semi-analytically (weak DI [1,2]) and numerically (higher concentration – strong DI [3]) to systematically investigate the effect of density and non-linear terms. The obtained SAR then serves as a source in the balance equation for heat diffusion, taking account of Newton's law of cooling at the interface between the sample and its immediate vicinity [4]. The temporal profile of the temperature elevation is confronted with experimental data for maghemite and magnetite ferrofluids, providing a basis for the rationalization of the dependence of the Newton coefficients on the relevant physical parameters.

**Acknowledgments**

The authors acknowledge the support of the French Agence Nationale de la Recherche (ANR), ANR "NanoHype", under grant ANR-21-CE09-0043-01

**References**

[1] J.-L. Dejardin, F. Vernay, M. Respaud, H. Kachkachi, J. Appl. Phys. **121**, 203903 (2017)
[2] J.-L. Déjardin, F. Vernay, H. Kachkachi, J. Appl. Phys. **128**, 143901 (2020)
[3] V. Russier, J. J. Alonso, I. Lisiecki, A.T. Ngo, C. Salzemann, S. Nakamae, and C. Raepsaet, Phys. Rev. B **102**, 174410 (2020)
[4] J.-L. Déjardin, and H. Kachkachi, Journal of Magnetism and Magnetic Materials, 556, 169354 (2022)

# Exploiting Nanoparticle Dynamic Magnetization for Biosensing in Liquids

*E. Sanz-de Diego,[1] P. Palacios-Alonso,[1] N. Silvestri,[2]. E.J. Artés-Ibáñez[1,3] R. Delgado-Buscalioni[4], T. Pellegrino,[2] A. L. Cortajarena[1,5,6], ,and F. J. Teran[1,7]*

[1] *iMdea Nanociencia. Calle Faraday, 9. Ciudad Universitaria de Cantoblanco, 28049 Madrid, Spain*

[2] *Istituto Italiano di Tecnologia, Via Morego 132, Genoa, Italy*

[3] *Nanotech Solutions, 40150 Villacastín, Spain*

[4] *Dpto Física Teórica Materia Condensada, Universidad Autónoma de Madrid, 28049 Madrid, Spain*

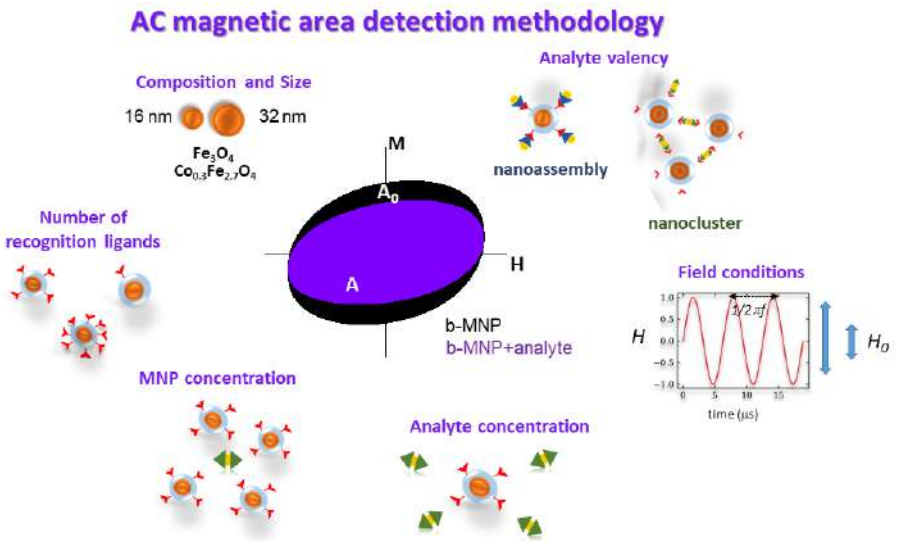
[5] CIC biomaGUNE, Paseo Miramón 182, 20014 Donostia-San Sebastian. Spain

[6] *Ikerbasque, Basque Foundation for Science, 48009 Bilbao, Spain*

[7] *Nanobiotecnología (iMdea-Nanociencia), Unidad Asociada al Centro Nacional de Biotecnología (CSIC), 28049 Madrid, Spain*

*francisco.teran@imdea.org

Biological markers are measurable indicators of biological or physiological states. Current detection technologies require exhausting sample preparation or complex detection methodologies. Such drawbacks are being overcome by recent progress in nanoscience and nanomaterials. Among the latest, nanoparticles have been widely employed in different diagnosis, therapeutic, and sensing applications. In particular, magnetic nanoparticles (MNPs) have been proven to transduce biomolecular recognition of target biomarkers in fluids. Here, we present a novel, quick, low cost and versatile methodology for magnetic detection of biomarkers in biological fluids (i.e. buffer saline, and blood plasma), based on the variation of the AC hysteresis loops of MNPs after interacting with an analyte. Such measurable variations are observed after short MNP-analyte incubation times, and are associated with the increase of hydrodynamic volume and magnetic dipolar interactions. The displayer is an AC magnetometer taking few seconds (< 5s) for measurement and requiring reduced magnetic suspension volumes. Among all these suitable features, the most important aspect of our novel method is that the sensitivity of biomarker detection is modulated down to 10 nM not only by the analyte-ligand interaction affinity, but by different parameters of employed magnetic display system (see Figure below). We have succeeded to perform the proof of concept of this novel and versatile magnetic detection methodology, whose potential relies on exploiting distinct strategies to probe changes in magnetic relaxation processes (i.e. Néel and Brownian) of MNPs dispersed in fluids. Indeed, AC magnetometry has a novel technological biosensing potential beyond its use as magnetic characterization technique.




**Acknowledgments**

Authors acknowledges financial support from PEJ-2020-AI/IND-19394, M-ERANET 2018 (PCI2019-103600), PID-2020-117080RB-C51, and PID- 2020-117080RB-C53

# Magnetosomes Detection In Liquid By Magnetic Force Microscopy

*Jorge Marqués-Marchán[1]*, Pablo Ares[2], Miriam Jaafar[2,3], Alicia Gascón Gubieda[4], Ana Abad[4] ,Eider Berganza[1], María Luisa Fdez-Gubieda[5,6], Agustina Asenjo[1]*

[1] *Instituto de Ciencia de Materiales de Madrid, CSIC, 28049 Madrid, Spain*
[2] *Dpto. de Física de la Materia Condensada and Condensed Matter Physics Center (IFIMAC). Universidad Autónoma de Madrid 28049, Madrid, Spain*
[3] *Instituto Nicolás Cabrera, INC Cantoblanco, Madrid, Spain*
[4] *Dpto. Inmunología, Microbiología y Parasitología, Universidad del País Vasco (UPV/EHU), Leioa, Spain*
[5] *Dpto. Electricidad y Electrónica, Universidad del País Vasco (UPV/EHU), Leioa, Spain*
[6] *BCMaterials, Basque Center for Materials, Applications and Nanostructures, UPV/EHU, Spain*
*jorge.marques@csic.es

Atomic force microscopy (AFM) is a powerful technique for the nanoscale characterization in a broad range of areas, such as nanomedicine, biophysics and nanobiotechnology. These subjects require the AFM measurements to be performed in physiological conditions, thus in a liquid environment. Magnetic force microscopy (MFM) is usually performed in vacuum or air conditions. Though the applicability of MFM in liquid has been demonstrated, the signal-to-noise ratio is decreased compared to vacuum and air [1].

Magnetotactic bacteria (MTB) are microorganisms that biomineralize magnetic nanoparticles surrounded by a lipid bilayer membrane (so-called magnetosomes) forming a chain along the axis of the cell, which allows bacteria alignment under a magnetic field. Due to their properties, MTB have potential applications in medicine, for example in cancer therapies [2]. Up to now, MFM characterization of MTB has been a challenging task due to the fact that magnetosomes are small compared to bacteria and they are localized inside their membrane, causing that their magnetic signal is hardly detected, even in air conditions. The customization of AFM probes with magnetic material has exhibited better results in air (Figure) [3], but the detection of the magnetic signal in liquid environments has not been yet observed for these MTB. More recently, magnetic nanorods grown by Focused Electron Beam Induced Deposition (FEBID) [4] on non-magnetic AFM tips have shown an improvement of the signal-to-noise ratio for the magnetic signal in liquids [5].

In this work, magnetic nanorods have been grown on non-magnetic AFM commercial tips. These probes are used to perform MFM measurements on MTB bacteria in a liquid environment. The detection of magnetosomes signal in these liquid conditions constitutes a significant advance in thedevelopment of MFM for biological applications.

## References

[1] P. Ares, et al., Small, 11(36), 4731-6 (2015)
[2] M. L. Fdez-Gubieda, et al., J. Appl. Phys., 128, 070902 (2020)
[3] E. Berganza Eguiarte, A study of complex magnetic configurations using Magnetic Force Microscopy. Universidad Autónoma de Madrid (2018)
[4] M. Jaafar, et al., patent PCT/ES2018/070709 and WO19086745
[5] M. Jaafar, et al., Nanoscale,12, 10090-10097 (2020)

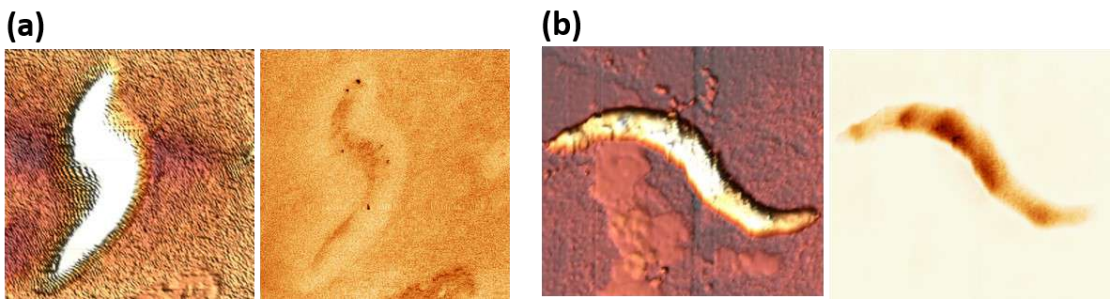



Figure: Topography (left) and MFM (right) images of MTB measured in air with (a) a commercial Nanosensors PPP-MFMR probe and (b) a customized commercial Olympus Biolever Mini AFM probe with magnetic coating.

# Magnetic Particles Imaging: Study of the Inverse Problem by Machine Learning

*Marco Coïsson[1]*, Gabriele Barrera[1], Federica Celegato[1], Paolo Allia[1], Paola Tiberto[1]*

[1] *INRIM, strada delle Cacce 91, 10135 Torino (TO), Italy*

*m.coisson@inrim.it

Magnetic Particles Imaging (MPI) is a promising imaging technique exploiting magnetic nanoparticles with typical diameters in the 10-20 nm range. Nanoparticles, driven in correspondence of tumor masses, are triggered with a static magnetic field gradient to form a sensitive region that is excited to produce a measurable signal by an external, low-intensity ac electromagnetic field. By scanning with a suitable device (an antenna) over the patient's body, maps of the magnetic response of the magnetic nanoparticles can be obtained, which can be used to identify the tumoral masses and characterise some of their properties. With respect to other imaging diagnostic techniques, MPI does not use ionising radiation or intense magnetic fields, and is therefore attracting much attention.

The maps acquired during scanning report the real and imaginary components of the third harmonic of the signal detected by an antenna that is capturing the magnetic response of the magnetic nanoparticles excited by the rf field. For each point of the map, the signal does not come only from the particles immediately underneath the antenna, but it is the integral of the contribution of all the particles, according to their distribution and their distance from the antenna. The solution of the direct problem, i.e. the calculation within the sensitive region of the real and imaginary parts maps given the initial nanoparticles distribution and their magnetisation vs. field curve, is straightforward (although time consuming), whereas the inverse problem, i.e. the calculation of the nanoparticles distribution from the real and imaginary parts maps, is not trivial and may be severely affected by incomplete or inaccurate knowledge of the physical properties of the nanoparticles [1]. Nonetheless, it is the inverse problem that is mostly relevant for diagnostic applications.

In this work we approach the solution of the inverse problem of MPI exploiting a machine learning model. First, nanoparticles with different size and magnetic properties (blocked or superparamagnetic), distributed in one or more clusters, with different shape and size, are randomly generated with a numerical approach (an example of nanoparticles distribution is given in Figure 1(a)). Then, by using the cyclic magnetisation associated to the considered nanoparticles, the complex third harmonic response (system function, Figure 1(b)) is calculated for each point of the map, and numerically integrated (Figure 1(c) and (d)). A large dataset is therefore compiled, whose entries are the real and imaginary parts of the signal, used as inputs, and the initial nanoparticles distribution, used as output, of a convolutional neural network. After training the machine learning model, it is possible to test its predictions by comparing the true (numerically generated) nanoparticles distribution, and the calculated one (Figure 1(e)). The model is shown to efficiently reconstruct isotropic and anisotropic nanoparticle distributions (including bimodal or multimodal classes) without the need of entering the details of the magnetic properties of the particles themselves (which are often difficult to check and control in a living environment).

## References

[1] G. Barrera, P. Allia, P. Tiberto, Phys. Rev. Appl. 19 (2022) 034029

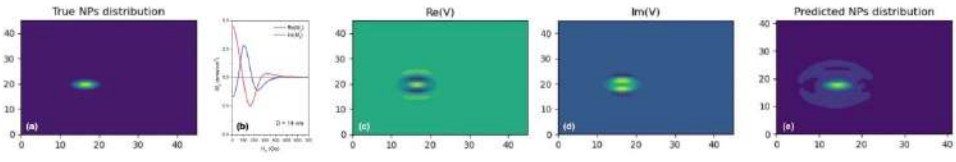



Figure 1: (a) Simulated disrtribution of magnetic nanoparticles. (b) Calculated third harmonic response at the peak of the distribution. (c) Real part and (d) imaginary part of the integrated third harmonic maps. (e) Recalculated magnetic nanoparticles distribution by a convolutional neural network.

# Probing AC Magnetic Properties of Magnetic Nanoparticles Inside Biological Cells Using Magneto-Optic Faraday Effect

*Xuyiling Wang[1]*, Ying Yang[1], Neil Telling[1]*

[1] *School of Pharmacy and Bioengineering, Keele University, Stoke-on-Trent, ST5 5BG, UK*

*x.wang3@keele.ac.uk

Magnetic nanoparticles (MNPs), exposed to high frequency alternating magnetic fields, can generate localized heat and induce death of cancer cells; an effect termed magnetic hyperthermia (MH). The magnetic-optic Faraday effect (MOFE) is the rotation of light polarisation through materials magnetised in a direction parallel to the light propagation. Here, a scanning laser microscope based on MOFE, developed in-house, was used to determine the AC magnetisation of nanoparticles in biological samples in situ. It could measure AC hysteresis loops under frequencies up to 500 kHz and field up to 50 mT with high spatial resolution (<1 μm). In addition, a fluorescence imaging module allows simultaneous biological structural and functional imaging.

By mapping the magneto-optical AC susceptibility across cell regions, magnetic images of MNPs within the cells in situ were obtained (Figure 1: Centre). AC hysteresis loops under different frequencies from aggregates of MNPs within cells were also investigated by this microscope. Analysis of AC hysteresis loops obtained from different nanoparticles in suspension form, as well as in different intra and extra-cellular locations in biological samples, revealed differences in their AC magnetic properties. Such effects have been suggested previously from bulk measurements of cellular suspensions [1], but have not been resolved at the cellular level until now. The ability to probe this cellular level magnetic information from the MNPs, enables the determination of their AC magnetic properties in situ. This is essential to promote magnetic hyperthermia by informing on how the heating effect can be optimised inside cancer cells.

## References

[1] Cabrera D, Coene A, Leliaert J, Artés-Ibáñez EJ, Dupré L, Telling ND, Teran FJ. Dynamical magnetic response of iron oxide nanoparticles inside live cells. ACS nano. 2018 Mar 6;12(3):2741-52.

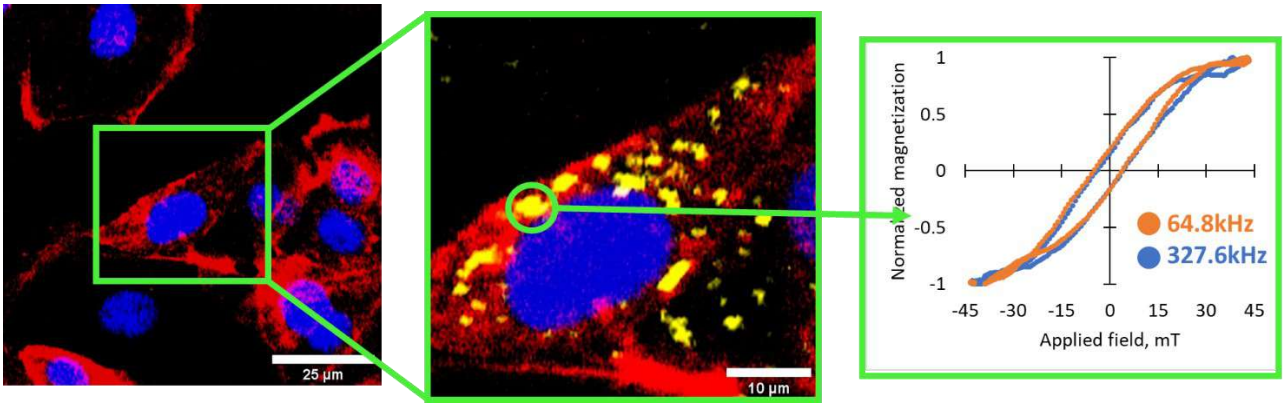



Figure 1: (Left) Fluorescence image of fixed human breast cancer cell line (MCF-7): labelling F-actin (red) and nucleus (blue). (Centre) Zoom-in fluorescence image superimposed with magnetic mapping (yellow) by measuring AC susceptibility under 64.8kHz using the in-house developed multifunctional microscope. (Right) Hysteresis loops under two frequencies (64.8kHz, 327.6kHz) measured from MNPs aggregated inside cells

# SYMPOSIUM 03.
## BIOMAGNETISM AND BIOMEDICAL APPLICATIONS.
## S3. ORAL PRESENTATIONS

# Open design very low field MRI for preterm babies

*Denitsa Nencheva*[1*], *Claude Fermon*[1]

[1] *CEA,CNRS, Université Paris-Saclay, 91191 Gif-sur-Yvette, France*

*denitsa.nencheva@cea.fr

One baby out of ten is born preterm and it may face many health complications. The ischemic stroke is one of them and is often diagnosed belatedly during a routine ultrasound exam, when irreversible neurological changes have already taken place. Our goal is to develop an alternative imaging tool, allowing for a safe and efficient scanning environment for neonatological applications. Our open very low field MRI (VLFMRI) is compatible with preterm baby's incubators, which will be inserted between the main field's coils, allowing for continuous image acquisition.

The set-up is low cost with minimal acoustic noise and low specific absorption rate. To design it, we have developed a software, based on Biot-Savart's law, calculating, and optimizing the $B_0$'s and gradients' coils. The |MRI is made of symmetrical big (2 m diameter) and small coils (64 cm diameter) generating a static $B_0$ field of 8 mT with 7ppm on 10x10x30cm$^3$. Our gradients Gx, Gy are rectangular coils placed symmetrically on both sides of the set-up, generating 290µT/m with 0.45% linearity error. Gz mimics the main field's geometry and creates 500µT/m with 0.11% linearity error.

The key challenge at very low magnetic fields is the low signal that requires extremely low noise sensors. We are developing two different approaches, one based on mixed superconducting-spintronic sensors [1] the other based on optimized tuned coils. The latter is presently better with achieved detectivities of $0.03\ ft/\sqrt{Hz}$. The current design is a rectangular double-layered surface tuned coil (23.5x17 cm$^2$) with 28 turns in Litz wire, spaced at 3 mm between each turn. Its intrinsic resistance ($R_{DC}$+$R_{AC}$) is 170 mΩ, with a quality factor Q=476.

The neonatology MRI system is already operational, and first images have been performed on phantoms. Our future goal is to implement fast acquisition sequences for producing 3D images in a reasonable scanning time.

View of the 8 mT open MRI during the installation process.

# Synthesis of protein microbubbles endowed with iron oxide nanocrystals for magnetic manipulation and heat delivery

*J. N. Majcherkiewicz[1,2], H. Zhu[3], F. Cavalieri[4], M. Ashokkumar[3], V. Salgueiriño[1,2]*

[1] *Departamento de Física Aplicada, Universidade de Vigo, 36310 Vigo, Spain*

[2] *CINBIO, Universidade de Vigo, 36310 Vigo, Spain*

[3] *School of Chemistry, University of Melbourne, Vic 3010, Australia*

[4] *Dipartimento di Scienze e Tecnologie Chimiche, Università degli Studi di Roma Tor Vergata, 00133, Rome, Italy*

*julia.natalia.majcherkiewicz@uvigo.es

Since magnetic nanoparticles when exposed to an alternating magnetic field can generate heat, they are great candidates for applications in which thermal energy is essential to accomplish the anticipated experimental purpose. In nanomedicine, this phenomenon is used for localized cancer therapy (magnetic hyperthermia), complementing or being a promising alternative to chemotherapy and radiotherapy. [1][2][3] For achieving better biocompatibility and gaining additional features, the magnetic nanoparticles must be functionalised by different coatings or immersed in hybrid systems. With this in mind, albumin protein microbubbles obtained by a sonochemical method were chosen to attain a hybrid system, which hosts iron oxide nanoparticles (two samples with different size and shape) in the protein shell, and become endowed with magnetic properties for manipulation and/or heat delivery applications. [4][5]

**Acknowledgements**

The authors acknowledge the financial support from, the Spanish Ministerio de Ciencia e Innovación under project PID2020-119242RB-I00 and the European Union H2020-MSCA-RISE-2019 PEPSA-MATE project.

**References**

[1] S.-H. Noh, S.H. Moon, T.-H. Shin , Y. Lim, J. Cheon, *Nano Today* **13** (2017) 61–76

[2] E. A. Périgo, G. Hemery, O. Sandre, D. Ortega, E. Garaio, F. Plazaola, F. J. Teran, *Applied Physics Reviews* **2** (2015) 041302

[3] S. Dutz, R. Hergt, *Nanotechnology* **25** (2014) 452001

[4] B. Bhargawa, V. Sharma, M.-R. Ganesh, F. Cavalieri, M. Ashokkumar, B. Neppolian, A. Sundaramurthy, *Ultrasonics Sonochemistry* **86** (2022) 106016

[5] J. N. Majcherkiewicz, H. Zhu, F. Cavalieri, M. Ashokkumar, V. Salgueiriño (in preparation, 2023)

# Influence of the magnetosome morphology in the magnetic hyperthermia response of different magnetotactic bacteria species

*Danny Villanueva-Alvaro[1]*, Lucía Gandarias[2], David Gandía[1], Lourdes Marcano[3], Alicia G. Gubieda[2], Ana Abad[2], Javier Alonso[4], Ana García Prieto[5], David de Cos[5], Ma Luisa Fdez-Gubieda[1,6].*

*[1]Dpto. Electricidad y Electrónica, Universidad del País Vasco, 48940 Leioa, Spain.*
*[2]Dpto. Inmunología, Microbiología y Parasitología, Universidad del País Vasco, 48940 Leioa, Spain.*
*[3]Departmento de Física, Facultad de Ciencias, Universidad de Oviedo, Oviedo 33007, Spain.*
*[4]Departamento CITIMAC, Universidad de Cantabria, Santander 39005, Spain.*
*[5]Dpto. de Física, Universidad del País Vasco, Spain.*
*[6]Basque Center for Materials Applications and Nanostructures (BCMaterials), 48940 Leioa, Spain.*

* dannyyosmar.villanueva@ehu.eus

Magnetotactic bacteria (MTB) are microorganisms able to align along the geomagnetic field lines due to their ability to biomineralize intracellular magnetic nanoparticles, called magnetosomes, that organize forming a chain. The morphology, composition and size of the magnetosomes are characteristic of each species and define the magnetic response of MTB and their potential use as nanorobots for cancer therapies such as magnetic hyperthermia.

In this work, we study the suitability of the *Magnetovibrio blakemorei* (MV-1) species as a magnetic hyperthermia agent. The MV-1 bacteria synthesize elongated magnetite magnetosomes with truncated hexa-octahedral morphology (~35x35x53 $nm^3$). In order to reveal any relationship between magnetosomes morphology and heating efficiency of MV-1 bacteria, we used different MTB species with other magnetosomes morphology: *Magnetospirillum gryphiswaldense* (MSR-1) [1] and *Magnetospirillum magneticum* (AMB-1) [2]. MSR-1 and AMB-1 bacteria synthesize magnetite magnetosomes with a truncated octahedral morphology (~40nm) with a slight distortion. The main difference between both species lies in the arrangement of the magnetosomes in the chain: while MSR-1 present a full chain, AMB-1 presents a fragmented one. We evaluated the heating capacity of MV-1 by measuring the specific absorption rate (SAR) using a homemade AC magnetometer [3]. The SAR values were obtained from the area of the AC hysteresis loops measured at a frequency of 132 kHz and with magnetic field amplitudes ranging up to 90mT.

The evolution of SAR/f as a function of the applied field amplitude is depicted in Fig.1 within the values of MSR-1 and AMB-1 for comparison. The same trend is observed for the three species: at low field the SAR/f values are negligible, whereas above a threshold field SAR/f exhibits a rapid increase reaching saturation at 40, 53 and 80 mT with saturation values of 8, 12 and 16.4 W/gkHz for MSR-1, AMB-1 and MV-1, respectively. The main difference observed should be related to the shape anisotropy of the different morphologies studies.

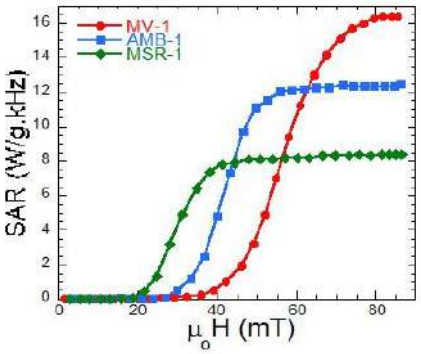



Fig.1: Specific absorption rate normalized by frequency as a function of the magnetic field amplitude for MV-1, AMB-1 and MSR-1 species.

## Acknowledgements

Grant PID2020-115704RB-C31 funded by MCIN/AEI/ 10.13039/501100011033 and, as appropriate, by ESF Investing in your future.

We thank J. A. Garcia, I. Rodrigo and I. Orue for the magnetometric SAR measurements.

## References

[1] D. Gandía, L. Gandarias, I. Rodrigo, et al. *Small* **15**, 1902626 (2019).
[2] D. Gandía, et al. *ACS Appl. Mater. Interfaces* **15**, 1, 566–577 (2023).
[3] I. Rodrigo, et al. *International Journal of Hyperthermia*, Vol. **37**, no 1, 976-991, (2020).

# Optimization of a microwave-assisted continuous-flow setup for rapid $Fe_3O_4$ nanoparticles synthesis

*K. Simeonidis[1]*, T. Asimakidou[1], C. Virgiliou[1], N. Maniotis[1], M.P. Morales[2], S. Veintemillas Verdaguer[2]*

*[1]Analytical Chemistry Laboratory, Department of Chemical Engineering, Aristotle; University of Thessaloniki, Thessaloniki, Greece*

*[2]Materials for Medicine and Biotechnology Group, Instituto de Ciencia de Materiales de Madrid, CSIC, Madrid, Spain*

*ksime@physics.auth.gr

Advances in nanotechnology brought significant progress in various technological fields including biomedical sciences. The use of engineered nanoparticles for drug delivery, diagnosis and treatment of deseases, imaging and biomarkers detection are the most pronounced cases of this research trend that frequently reaches clinical levels. However, avaibility of nanoparticles able to comply with the standards for handling in the health sector is not always an easy condition to fullfil mostly due to details of their synthesis procedure. Ideally, combining rapid, inexpensive, highly automated, green, aqueous processes, with minimum energy consumption and by-products release operable by non-specialized staff, is a prerequisite to qualify a synthesis method as a good manufacturing practice valid for medical facilities.

In this context, this work examines the translation of a well-known production method for magnetite nanoparticles, based on the oxidative precipitation of $FeSO_4$ in aqueous media into a continuous-flow process, which implements complete control of operating parameters and products' quality, managing to minimize the duration of the rate-controlling thermal ageing step. Particularly, transformation of the green rust intermediate form into the $Fe_3O_4$ nanocrystals was carried out by heating in a microwave system adapted to a continuous flow reactor. In a first approach, a tank reactor (CSTR) was placed into the microwave oven and continuously fed with the green rust precursor. Following this scheme, $Fe_3O_4$ nanoparticles with diameter around 30 nm were successfully produced by applying a residence period of less than 10 min. Surprisingly, by using a plug-flow tube reactor (PFR), an extremely high heating rate was succeeded and well-defined magnetic nanoparticles were produced with a residence period of less than 30 s. Monitoring and optimization of the procedure was assisted by computational fluid dynamics. The potential of produced nanoparticles was evaluated by studying their behavior for in vivo intravenous administration in mice and their efficiency as agents for detecting metabolic processes.

## Acknowledgements

Research is funded by the IMAGINE: Implementing MAgnetic targeting of nano-Guided ImmuNE cells project funded by European Science Foundation Fight Kids Cancer 2020 programme. The research project was supported by the Hellenic Foundation for Research and Innovation (H.F.R.I.) under the "2nd Call for H.F.R.I. Research Projects to support Post-Doctoral Researchers" (Project Number: 00046 MagnoSorb).

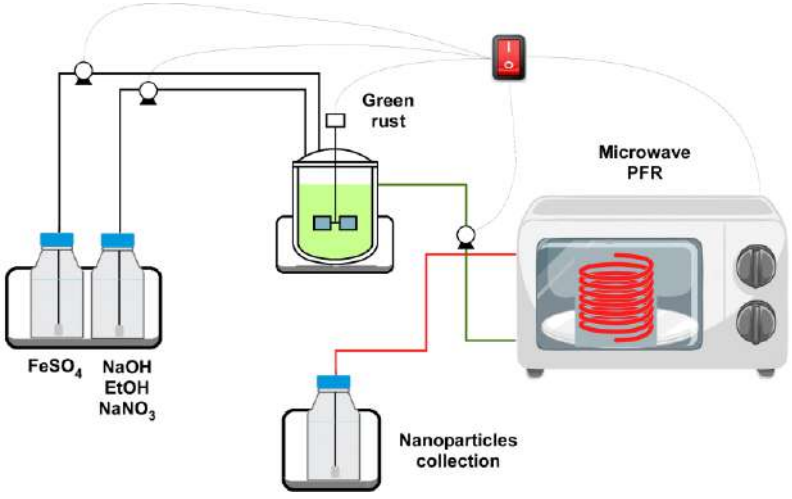



Figure: Microwave-assisted setup for continuous flow oxidative precipitation with a plug-flow reactor.

# A New Therapy for Adiposopathy Based on Low Frequency AC Field Applications on Magnetic Nanoparticles

*Elena Balica*[1,2*], *Martin Albino*[2,3], *Alessio Gabbani*[3,4], *Beatrice Muzzi*[2,3], *Claudia Innocenti*[2] *and Claudio Sangregorio*[2,3]

[1] *INSTM and Dept. of Industrial Engineering, University of Florence, via di S. Marta 3, 50019 Sesto Fiorentino, Italy;*

[2] *INSTM and Dept. of Chemistry U. Schiff, University of Florence, via della Lastruccia, 3, 50019 Sesto Fiorentino, Italy*

[3] *ICCOM-CNR, via Madonna del Piano 10, 50019 Sesto Fiorentino, Italy;*

[4] *University of Pisa, via Lungarno Antonio Pacinotti 43, Pisa, Italy*

e-mail: elena.balica@unifi.it

Magnetic nanoparticles (MNPs) have been the subject of intense research, not only for fundamental research but also for their potentiality in a wide number of practical applications such as energy storage, catalysis, photonics, electronics, or biomedicine. [1-3] In particular, MNPs are constantly showing their great potential to revolutionize classical medical treatments and therefore the number of new biomedical applications has been increasing exponentially over time.

Adiposopathy is one of the most widespread pathologies in EU member states. It is promoted by fat accumulation (adiposity) and a sedentary lifestyle in genetically susceptible patients. At the present, this pathology is treated by invasive interventions, such as bariatric surgery or liposuction procedures, or by pharmacological treatments aimed at addressing some effects of adiposopathy, such as hypertension or Type 2 diabetes, but not the disease itself. For this, we propose a radical new approach for the treatment of pathological adipose tissue, based on the application of MNPs and the induction of magneto-mechanical stress applying an external LF-AC magnetic field that activates cell apoptosis or lipolysis, avoiding side effects associated with standard routes. Iron oxide MNPs of different shapes are of particular interest in the precise determination of their shape-dependent properties and functionalities in biomedical applications. Compared with spherical and octahedral MNPs, iron oxide nano-octopods offer higher heating efficiency, enhanced anisotropy, and higher contrast in magnetic resonance imaging (MRI) while magnetite nanorods offer longer blood circulation times, stronger interaction with tumors, enhanced retention at tumor sites, improved targeting efficiency, making them excellent candidates to induce mechanical damage of cells. [4]

In this contribution we present the synthesis of iron oxide MNPs of different sizes and shapes (rods and octapods) by colloidal chemistry to be used as stress inductors in adipocytes. The MNPs were characterized by X-Ray Diffraction (XRD), Transmission Electron Microscopy (TEM), and superconducting quantum interference device (SQUID) magnetometry. Also, we present a prototype of an LF-AC generator for inducing magneto-mechanical damages. This device is easy to handle and transport and can be used for different cell cultures and biological targets. The $Fe_3O_4$ MNPs were synthesized to be used as stress inductors in adipocytes.

**References**

[1] A. K. Gupta, A.S. Curtis, *Biomaterials* **26(18)** (2005) 3995-4021.
[2] A. Jordan et al., *Journal of Magnetism and Magnetic Materials* **225** (2001) 118-126.
[3] T. Neuberger et al., *Journal of Magnetism and Magnetic Materials* **293** (2005) 483-496.
[4] Nemati, Z. et al., *J. Phys. Chem.* **120** (2016) 8370–8379.

# Multi-detector platform for analyzing the interaction of magnetic nanoparticles in biological environments

*Amani Remmo[1]*, Norbert Löwa[1], Lena Kampen[2], Antje Ludwig[2], Frank Wiekhorst[1]*

*[1] Physikalisch-Technische Bundesanstalt, Abbestrasse 2-12, 10587 Berlin, Germany*

*[2] Charité-Universitätsmedizin Berlin, Division of Cardiology and Angiology, Charitéplatz 1, 10117 Berlin, Germany*

*amani.remmo@ptb.de

Quantification of magnetic nanoparticles (MNP) in biological systems like cells is of importance for the development of novel biomedical applications [1]. Surface and charge as well as MNP size are considered to be key factors for the biomedical applicability of MNP. Therefore, characterization of MNP in environments that mimic physiological conditions is of great importance. For this reason, the development of new biomedical applications of MNP requires powerful techniques capable of measuring all relevant structural and magnetic properties of MNP in complex media. The coupling of chromatographic separation techniques with complementary detectors in a contained system allows real-time and detailed characterization of MNP, as shown in [2]. In this work, we used this multi-detector platform to characterize the interaction between MNP and biological environments. We applied centrifugal flow-field fractionation (CFFF, *Postnova Analytics GmbH*) to separate the MNP in different media according to their mass. The fractions were then magnetically and physically characterized by the detector array consisting of UV/Vis detection for concentration determination, dynamic light scattering (DLS) for hydrodynamic size determination, and magnetic particle spectroscopy (MPS) for dynamic magnetic characterization. Using this setup, MNP changes were studied in deionized water ($H_2O$), Fetal Calf Serum (FCS, *Biochrom*), Roswell Park Memorial Institute 1640 medium (RPMI 1640 Medium, *Gibco*), and Dulbecco's Modified Eagle Medium (DMEM high glucose, *Thermofisher*). All these media are widely used for experiments on human and animal cells. Investigations were carried out on various commercial MNP systems. After separation of the MNP sample with respect to particle mass and instantaneous determination of hydrodynamic size (DLS signal), concentration (UV/Vis signal) and magnetism (MPS signal), the measurements were evaluated. The results for the commercial MNP system Synomag-COOH (SynC, *Micromod Partikeltechnologie*) with a citrate surface and a hydrodynamic diameter of 30 nm, in $H_2O$, 10% FCS+RPMI and DMEM are shown in Fig. 1. In $H_2O$ (black curve), SynC exhibits a narrow size distribution (Fig. 1a), which changes for DMEM (green) and becomes much broader for 10% FCS in RPMI (purple). These changes also reduce the magnetic signal of the detected size classes (Fig. 1b).

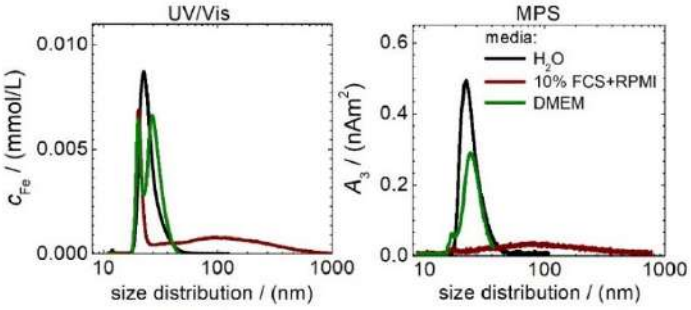



Figure 1: a) Size dependent UV/Vis signal ($c_{Fe}$) and b) MPS signal ($A_3$) of SynC30 in three biological media $H_2O$ (black), 10% FCS in RPMI (purple) and DMEM high glucose (green) as obtained by the multi-detector platform.

Our results show that the multi-detector platform is suitable for studying the interaction of MNP in different physiological media. Due to the separation of the MNP sample prior to multi-modal characterization, this method is significantly more powerful and reliable compared to commonly used integral measurement methods. The platform provides important insight into the physics of MNP in biological environments and can be used as a standard analytical method for the control of MNP in biomedical applications.

**Acknowledgements:** This project was partially funded by the German Research Foundation (DFG) within the research grant "CellMPI" (455706279) and the collaborative research centre "Matrix in Vision" (SFB 1340/2 2022, no 322486779, projects A02 and B02).

**References:**

[1] L. Trahms, 'Biomedical applications of magnetic nanoparticles', *Lect. Notes Phys.*, vol. 763, 2009.

[2] A. Remmo, N. Löwa, J. Peter, and F. Wiekhorst, 'Physical characterization of biomedical magnetic nanoparticles using multi-detector centrifugal field-flow fractionation', *Curr. Dir. Biomed. Eng.*, vol. 7, no. 2, 2021.

# Fabrication of 3D magnetoresponsive nano/microstructures

*Eider Berganza[1,2]*, George Mathew[2], Jorge Marqués-Marchán[1], Alejandro Gomez-Roca[3], Evgeniy Boltynjuk[2], Agustina Asenjo[1], Michael Hirtz[2]*

[1] *Instituto de Ciencia de Materiales de Madrid (ICMM), Consejo Superior de Investigaciones Científicas (CSIC), Sor Juana Ines de la Cruz 3, 29048 Madrid, Spain*

[2] *Institute of Nanotechnology (INT) & Karlsruhe Nano Micro Facility (KNMF), Karlsruhe Institute of Technology (KIT), Hermann-von-Helmholtz-Platz 1, 76344 Eggenstein-Leopoldshafen, Germany*

[3]*Instituto Catalan de Nanociencia y Nanotecnología, Campus de la, Universitat Autònoma de Barcelona, Edifici ICN2, Av. de Serragalliners, s/n, 08193 Bellaterra, Barcelona.*

*eider.berganza@csic.es

The emergence of new 3D printing technologies together with the development of miniaturization techniques holds promise to start a new industrial and technological era through the incorporation of complexed shaped multifunctional microstructures. From the Internet of Things (IoF) environments to integration of theranostic in-vivo devices, many technological fields will benefit from the development of stimuli-responsive microstructures. In particular, the capability to respond to external magnetic fields possess some inherent advantages in comparison to other stimuli (thermal, chemical, electrical, etc.), such as the possibility for remote control or high controllability.[1] Despite the rapid development of 3D printing of magnetoresponsive materials on the macroscale, fabrication of micron/nano sized structures is still an emerging field.

In view of this, we present two different approaches that we are developing towards the creation of 3D polymeric magnetoresponsive structures (Figure 1). (a) Direct printing of nanoparticles embedded in a polymeric matrix via FluidFM[2], a technology based on the combination of microfluidics with atomic force microscopy (AFM), in which a hollow cantilever, with an aperture at the tip apex, can be used for precisely localized liquid dispensing. (b) The combination of scaffolds fabricated via 2-photon lithography with physical vapour deposition of ferromagnetic alloys to render them responsive to magnetic field.[3]

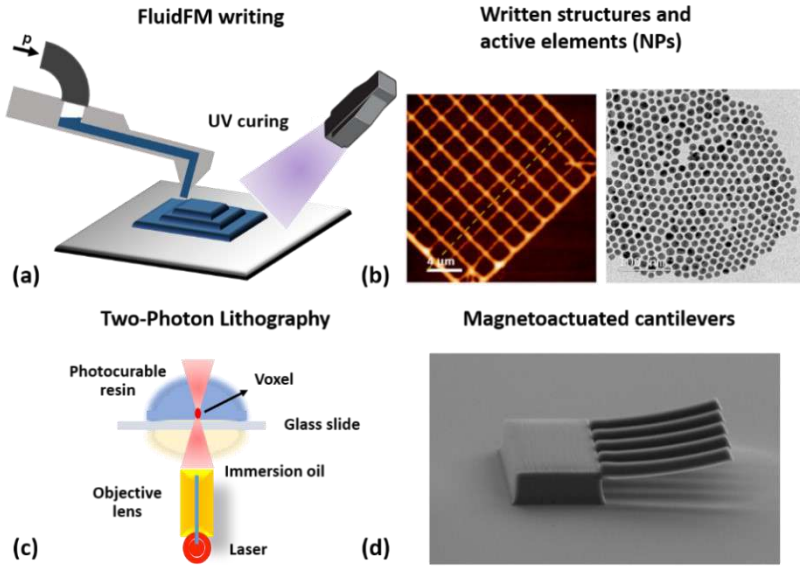



Figure 1. Sketches of (a) FluidFM for direct writing of (b) polymeric matrices with magnetic nanoparticles. (c) Two-photon lithography to fabricate (d) magnoactuated cantilevers.

## Acknowledgements

E.B. acknowledges the Karlsruhe Institute of Technology for a postdoctoral fellowship in the frame of the YIG Prep Pro program and the Spanish Ministry of Science for a Juan de la Cierva Incorporation fellowship.

## References

[1] K.J. Merazzo et al., *Mater. Horizons* **8** (2021) 2654–2684.
[2] T.G. Pattison, et al., *Nat. Commun.* **13** (2022) 1-7.
[3] F. Qiu et al. *Adv. Funct. Mater.* **25**, (2015) 1666–1671.

# Biosensor Based on Nanolayered Structures for SARS-COV2 Virus Detection

*Nerija Zurauskiene*, Vilius Vertelis, Arunas Stirke, Eivydas Andriukonis, Andrius Maneikis, Skirmantas Kersulis, Voitech Stankevic*

*Department of Functional Materials and Electronics, Center for Physical Sciences and Technology, Sauletekio ave. 3, 10257 Vilnius, Lithuania*

*nerija.zurauskiene@ftmc.lt

Currently, the main method for detecting SARS-CoV-2 virus is the molecular diagnostic test qRT-PCR, which is relatively slow, because it takes about 48 hours to perform and requires expensive and complex testing techniques. To ensure rapid community testing other ways to detect the virus are needed. Therefore, it is relevant and important to develop simple, non-invasive tools and methods that allow tests to be performed on anyone without special preparation [1]. To develop the biosensor prototype for direct virus detection from the patient's exhaled air we have chosen nanolayered structures based on spin valves [2]. These are giant magnetoresistive devices which operation is based on spin selective electron scattering between ferromagnetic layers with thicknesses in nanometer range. As their properties can be tuned by many factors, they can be designed to fit specific biosensing applications. In this contribution, we tested several spin valve structures deposited using ultra-high vacuum magnetron sputtering with varing layer thicknesses and different magnetic materials, namely CoFe and $Co_2MnSi$, in order to achieve high magnetic field sensitivity and large magnetoresistance. The spin valves were shaped as 6 mm × 2 µm meandering channels and incoroporated into a microfluidic system. The biosensing of the device is based on the detection of the shift in the magnetoresistance characteristics due to the presence of magnetic nanoparticles joined with the virus on the surface of the structure. Top layer of the structure was passivated with a polimer in order to anchor covalantenly binded polyclonal antibodies to the surface. Inactivated SARS-CoV-2 Whole Virus was used as a model. To detect and measure concetration of the attached virus, superparamagnetic nanoparticles funcionalized with monoclonal antibodies against virus were used. The prototype of final device for detection of viruses or other airborne patogens in exhaled air will be presented and discussed.

**Acknowledgements**

This work was partly supported by the European Regional Development Fund (project No 01.2.2-LMT-K-718-05-0015) under a grant agreement with the Research Council of Lithuania (LMTLT), funded as the European Union's measure in response to COVID-19 pandemic.

**References**

[1] D. Su, K. Wu, R. Saha, C. Peng, and J.-P. Wang, *Micromachines*, **11** (2019) 34.

[2] W. Qiu, L. Chang, Y.-C. Liang, J. Litvinov, J. Guo, Y.-T. Chen, B. Vu, K. Kourentzi, S. Xu, T.R. Lee, Y. Zu, R.C. Willson, and D. Litvinov, *Sensors and Actuators A: Physical*, **265** (2017) 174.

# Iron Oxide Nanoparticles as Chemo-thermal Agents for Effective Cancer Therapy

*Rosalía López-Méndez[1*], Nuria Lafuente-Gómez[1], Marina París[1,2], Yurena Luengo[1], Aida Serrano[2], Julio Camarero[1,3], Gorka Salas[1], Álvaro Somoza[1] and Ana Espinosa[1,4]*

[1] *IMDEA Nanociencia, C/ Faraday 9, 28049 Madrid, Spain*

[2]*Departamento de Electrocerámica, Instituto de Cerámica y Vidrio, ICV-CSIC, Kelsen 5, 28049 Madrid, Spain*

[3]*Departamento de Física de la Materia Condensada and Instituto 'Nicolás Cabrera', Universidad Autónoma de Madrid, 28049 Madrid, Spain*

[4]*Instituto de Ciencia de Materiales de Madrid, Consejo Superior de Investigaciones Científicas, calle Sor Juana Inés de la Cruz 3, 28049-Madrid, Spainoints)*

*rosalia.lopez@imdea.org

Nanomedicine seeks to develop new efficient therapeutic tools which can overcome the limitations of current treatments based on the use of nanomaterials. These nanoparticles can overcome cellular and physiological barriers and provide favorable biodistribution, bioavailability and improved pharmacokinetics of desired drugs [1]. They can also target entities in the tumor environment and induce physical therapeutic effects after being stimulated by a remote signal, allowing for site-specific treatment. These treatments can limit unwanted side effects by preventing damage to surrounding healthy cells. One of the most promising treatments is hyperthermia. Hyperthermia therapy is an anticancer clinical practice based on elevation of the tumor temperature, driving malignant cells and tissues up to the cytotoxic level, that is, 42-48 °C [2]. In addition, cell resistance against traditional treatments, such as chemotherapy or radiotherapy, can be reduced. In this work, we report the synthesis of a multimodal therapeutic system based on iron oxide nanoparticles (IONPs) loaded with a chemotherapeutic agent, doxorubicin (DOX). IONPs are widely used in biomedicine and have been approved as contrast agents for magnetic resonance imaging (MRI) [3]. Furthermore, IONPs are excellent candidates for magnetic hyperthermia (MHT). Therefore, we propose a multimodal treatment that combines local chemotherapy with efficient thermal therapy, to obtain a remarkably effective treatment due to the synergistic effects of the therapies. Biocompatibility and internalization capacity of IONPs in cells were examined, as well as the release and localization of the drug before and after thermal therapy. After hyperthermal treatment, this initial co-localization of DOX in lysosomes is lost. We can conclude that DOX-loaded IONPs are excellent agents for chemo-thermal therapy after an adequate functionalization of the IONPs.

## Acknowledgements

This work was supported by the Spanish Ministry of Economy and Competitiveness (SEV-2016-0686, RED2018102626-T, FPI SEV-2016-0686- 20-1, FPU18/02323, RYC2020-029282-I, PID2021-127033OB-C21), Comunidad de Madrid (2017-T2/IND5395 Talento, 2018-T1/IND-1005 Talento, 2018/NMT-4321), AECC (project Ideas Semilla 2019) and H2020 European project (No 685795. NoCanTher). Special thanks to BM23 (ESRF) beamline staff for the support during the experiments.

## References)

[1] Espinosa, A., et al. Adv. Funct. Mater. 28 (2018). (11), 1803660.
[2] Espinosa, A., et al. Nano Letters. 21 (2021). (1), 769-777.
[3] Lafuente-Gómez N. et al. Chem. Comm. 7 (2021). 13662–13677.

# Internalization of magnetic nanodisk in vortex state in cancer cells

*R. Zurbano[1]*, I.Solozabal[1], C. Redondo[1], M. D. Boyano[2,3] and R. Morales [4,5]*

*1. Department of Physical-Chemistry, University of the Basque Country UPV/EHU, 48940 Leioa, Spain.*

*2. Department of Cell Biology and Histology, University of the Basque Country UPV/EHU, 48940 Leioa, Spain. 3. Biocruces Health Research Institute, 48903 Barakaldo, Spain. 4. Department of Physical-Chemistry & BCMaterials, University of the Basque Country UPV/EHU, 48940 Leioa, Spain. 5. IKERBASQUE, Basque Foundation for Science, 48011 Bilbao, Spain.*

* raquel.zurbano@ehu.eus

Magnetic particles have attracted much attention in biomedicine by their physical properties to manipulate cells, release drugs at specific targets, develop novel diagnostics techniques, and envision promising therapies. In this field, magnetic nanodisks (MND) are very promising for novel applications. A new kind of particle with great potential in all these fields is a disk-shaped nanomagnet. These nanomagnets are fabricated by top-down lithography techniques and present remarkable properties for biomedical applications, e.g., large magnetic moments and anisotropic shapes appropriate for mechanical stimulations [1], which can lead to neuron signaling [2], or cell annihilation [3].

This work uses MND with diameters of 300 nm and 700 nm fabricated by interference lithography. The structures are made of Permalloy (Ni80Fe20) in a vortex state, a flux closed spin configuration that shows zero remanence in magnetization curves [4]. The internalization process of these particles was investigated in vitro assays with melanoma cells, melanocytes and macrophages. A magnetometry technique, superconducting quantum interference devices (SQUID), was used to quantify the number of nanodisks internalized by cells and determine the dynamics of the cellular uptakes. MND internalization and externalization times were evaluated and confirmed with other techniques such as confocal microscopy and transmission and scanning electron microscopy (figures 1).

We saw that the dynamics and behaviour of the disks with cells depends of the disk size and the characteristics of the cell type. Melanoma cells are capable of internalizing 300 and 700 nm disks, but only externalizing the smaller ones. While melanocytes are only capable of interacting with smaller disks, 300 nm, with a prolonged exposure time and without signs of externalization.

## Acknowledgements

Work supported by grant number PID2019-104604RB-C33 funded by MCIN/ AEI/10.13039/501100011033/, EU grant nuber 734801, GV IAT1491-22, and EU-Next Generation. Aid Predoctoral Training for Research Staff of the Basque Government 21-22 (PRE_2021_1_0080-A).

## References

[1] L. Peixoto et al., Appl. Phys. Rev. **7**, 011310 (2020). (DOI: 10.1063/1.5121702)
[2] C. Collier et al. Adv. Healthcare Mater**. 11**, 2101826 (2022). (DOI: 10.1002/adhm.202101826)
[3] D. Kim et al., Nature materials **9**, 165 (2010). (DOI: 10.1038/NMAT2591)
[4] B. Mora et al. ACS Appl. Mater. Interfaces **10**, 8165 (2018). (DOI: 10.1021/acsami.7b16779)

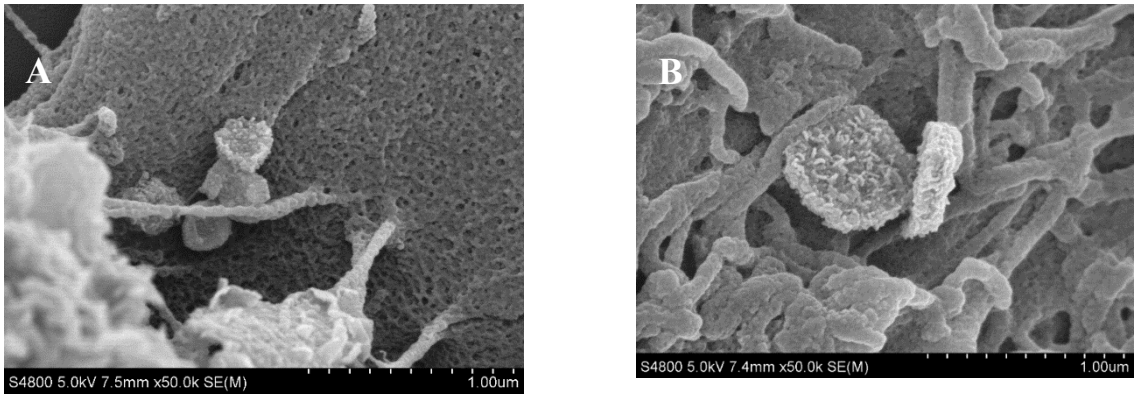



Figure 1: Scanning electron microscopy images of melanoma cells with figures A, 300 nm and B, 700 nm MND.

# Silk Fibroin Films With Embedded Magnetic Nanoparticles Promote The Osteogenic Differentiation Of Stem Cells

*L. Del Bianco[1], F. Spizzo[1]*, Y. Yang[2], G. Greco[3,4], M. L. Gatto[5], G. Barucca[5], N. M. Pugno[4,6], A. Motta[2]*

[1]*Department of Physics and Earth Science, University of Ferrara and INFN-Ferrara, Ferrara I-44122, Italy*
[2]*BIOtech Research Center, Department of Industrial Engineering, University of Trento, I-38123 Trento, Italy.*
[3]*Department of Anatomy, Physiology and Biochemistry, Swedish University of Agricultural Sciences, Uppsala, 750 07, Sweden*
[4] *Laboratory for Bioinspired, Bionic, Nano, Meta, Materials & Mechanics, Department of Civil,Environmental and Mechanical Engineering, University of Trento, Trento I-38123, Italy*
[5]*Department SIMAU, Università Politecnica delle Marche, I-60131 Ancona, Italy*
[6]*School of Engineering and Materials Science, Queen Mary University of London, London E1 4NS, UK*
*federico.spizzo@unife.it

The use of magnetic nanoparticles in regenerative medicine is a thriving research field aimed at remotely manipulating cells and/or conditioning their behavior [1]. On the other hand, tissue engineering using silk-based materials is gaining increasing interest. This is because the silk fibroin protein possesses high biocompatibility with low inflammatory and immunogenic responses, tunable biodegradability and mechanical strength, permeability to water and oxygen [2]. By combining these two elements, we have created a biomaterial in the form of film (~ 10 μm thick), for prospective applications as bioactive coating in regenerative medicine [3]. In fact, it consists of a silk fibroin matrix with embedded iron oxide superparamagnetic nanoparticles (mean size ~10 nm).

Films with different load of magnetic nanoparticles are produced (nanoparticles/silk fibroin nominal ratio = 5, 0.5 and 0 wt%) and the structural, mechanical and magnetic properties are studied. The nanoparticles form aggregates in the silk fibroin matrix and the film stiffness, as tested by nanoindentation, is spatially inhomogeneous, but the protein structure is not altered.

In vitro biological tests are carried out on human bone marrow-derived mesenchymal stem cells cultured on the films up to 21 days, with and without an applied static uniform magnetic field. The sample with the highest nanoparticles/silk fibroin ratio shows the best performance in terms of cell proliferation and adhesion. Moreover, it promotes a faster and better osteogenic differentiation, particularly under magnetic field, as indicated by the gene expression level of typical osteogenic markers.

These findings are explained in light of the results of the physical characterization, combined with numerical calculations. It is established that the applied magnetic field triggers a virtuous magneto-mechanical mechanism in which dipolar magnetic forces between the nanoparticle aggregates give rise to a spatial distribution of mechanical stresses in the silk fibroin matrix. The sample with the largest nanoparticle load, under cell culture conditions (i.e. in aqueous environment), undergoes matrix deformations large enough to be sensed by the seeded cells as mechanical stimuli that favor the osteogenic differentiation.

## Acknowledgements

L. Del Bianco was supported by the University of Ferrara under project ‘Fondo di Ateneo per la Ricerca 2022-FIRD’. F. Spizzo was supported by the University of Ferrara under project ‘Fondo per l’Incentivazione alla Ricerca’ (FIR–2021). G. Greco was supported by Caritro Foundation (prot. U1277.2020/SG.1130). N. M. Pugno was supported by the Italian Ministry of Education, University and Research (MIUR) under the ‘Departments of Excellence’ grant L. 232/2016 and the PRIN-20177TTP3S grant.

## References

[1] M. Matos, A. I. Gonçalves, A. J. El Haj, M. E. Gomes, Nanoscale Adv., 2020, 2, 140–148
[2] P. Bhattacharjee, B. Kundu, D. Naskar, H. W. Kim, T. K. Maiti, D. Bhattacharya and S. C. Kundu, Acta Biomater., 2017, 63, 1–17
[3] L. Del Bianco, F. Spizzo, Y. Yang, G. Greco, M. L. Gatto, G. Barucca, N. M. Pugno, A. Motta, Nanoscale, 2022, 14, 14558–14574

# Magnetic Nanoparticle Degradation and the Effect on their Heating Properties

*Yilian Fernández-Afonso[1,2], Laura Asín[1,3], Lilianne Beola[1], Raluca M. Fratila[1,3,4], Lucía Gutiérrez[1,2,3]*

*[1] Institute of Nanoscience and Materials of Aragon (INMA-CSIC/University of Zaragoza), Zaragoza, Spain. [2] Department of Analytical Chemistry University of Zaragoza, Zaragoza, Spain. [3] Centro de Investigación Biomédica en Red de Bioingeniería, Biomateriales y Nanomedicina (CIBER-BBN), Zaragoza, Spain. [4] Organic Chemistry Department, Universidad de Zaragoza, Zaragoza, Spain.*

*email: lu@unizar.es*

In the Magnetic Hyperthermia (MH) and Photothermal Therapy (PTT) applications, magnetic nanoparticles (MNPs) are used as an "antennae" able to capture energy (either from an alternating magnetic field (AMF) or a near infrared (NIR) light) and transform it into local heat. In the frame of this therapy design, it is fundamental to know how possible particle transformation would affect its performance over time.

In this work, we used MNPs with two different coatings (dimercaptosuccinic acid - DMSA-NPs) and poly(maleic anhydride-alt-1-octadecene) - PMAO-NPs) but with same magnetic core (≈13.5 nm) (Figure 1A). We evaluated how the MNP coating affects their degradation profile using a medium that simulates the lysosomal conditions and how this degradation affects their heating performance in the frame of both magnetic hyperthermia and photothermal treatments. The faster degradation of DMSA-NPs in comparison with PMAO-NPs was verified by transmission electron microscopy (TEM), magnetic and colorimetric measurements (Figure 1B).

To track how the transformations suffered by the particles along their degradation process affected their heating properties, magnetic hyperthermia and photothermal measurements were performed (Figure 1C). In both cases, the degradation process resulted in a decrease of the heating capacity of both types of materials. As a result of the faster degradation of DMSA-NPs, the reduction of the heating properties along time was increased for this material when compared to PMAO-NPs.

Thus, the less prone to degradation nanoparticles (PMAO-NPs) were selected for the *in vivo* analysis, to evaluate the degradation speed of this material in tumor tissues. In this study, although the number of particles decreased in the tumors along time after their administration, no transformations in the average particle size of particles occurred (Figure 1D).

**Acknowledgements**

This work was funded by the Ministerio de Ciencia, Innovación y Universidades (MCIU), the Agencia Estatal de Investigación (AEI) and Fondo Europeo de Desarrollo Regional (FEDER) through the MACBETH project (PGC2018-096016-B-I00 to R.M.F and L.G.) and the European Commission through the TBMED project (DT-NMBP-02-2018, ID: 814439 to R.M.F.).

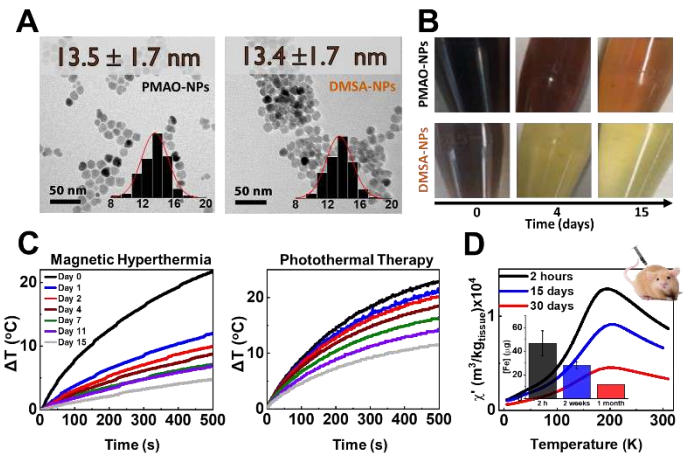



Figure 1. A) TEM images and particle size distribution of the nanoparticles used. B) Photos of the nanoparticle suspensions at different times during the degradation process. C) Magnetic Hyperthermia and Photothermal measurements of PMAO-NPs at different times of the degradation process. D) Temperature dependence of the AC magnetic susceptibility profiles of tumor tissues collected at different time points and iron concentration in the form of particles in the tumor calculated from the out-of-phase susceptibility data.

# Evaluation of Magnetic Hyperthermia Efficiency of Different Magnetic Particles in a Low-Cost Magnetic Hyperthermia Device

*T. Castelo-Grande[1], P. A. Augusto [2,3], J.C. Lobinho-Gomes[1], D. Barbosa[1]*

*(1) LEPABE – Laboratory for Process Engineering, Environment, Biotechnology and Energy, Faculty of Engineering, University of Porto, Rua Dr. Roberto Frias, 4200-465 Porto, Portugal*

*(2) Dep. Chemical Engineering, Faculty of Chemical Sciences, University of Salamanca, Spain.*

*(3) Institute of Molecular and Cellular Biology of Cancer, GIR Cytomic, Univ. Salamanca,Spain.*

*(4) Faculdade de Ciências Naturais, Engenharias Tecnologias, Univ. Lusófona do Porto, Portugal .*

* castelogrande@sapo.pt

This work presents the results of magnetically induced heat generation for different types of magnetic nanoparticles, to evaluate their application in Magnetic Hyperthermia. The study was done using a home-made low cost and portable device, which operates at frequencies between 150-350 kHz and generates magnetic fields in the range of 4-12 mT. The obtained results, which are quite promising, are justified having in consideration the heating mechanisms, the role of particle–particle interactions and the balance between generated and dissipated heat. The influence of particle concentration and their magnetic properties is studied for field strengths and frequencies within the range used in human treatment. Different methods for calculating the Specific Absorption Rate (SAR) are investigated and some possible sources of errors are pointed out. The synthesis of the magnetic particles used in this article will be also detailed as well as other practical aspects and limitations.

**Acknowledgements**

This research was funded by: (a) Project 2019/00057/001, “Application of Magnetic Methods and Magnetic Substances for Environmental Treatment and Biomedicine”, 26 RLD-PXIV, USAL, Spain. (b). Base Funding—UIDB/00511/2020 of the Laboratory for Process Engineering, Environment, <Biotechnology and Energy—LEPABE—Funded by national funds through the FCT/MCTES (PIDDAC); (c). Project POCI-01-0145-FEDER-028020—Funded by FEDER funds through COMPETE2020—Programa Operacional Competitividade e Internacionalização (POCI) and by national funds (PIDDAC) through FCT/MCTES; (d) Project 2SMART – engineered Smart materials for Smart citizens, with reference NORTE-01-0145-FEDER-000054, supported by Norte Portugal Regional Operational Programme (NORTE 2020), under the PORTUGAL 2020 Partnership Agreement, through the European Regional Development Fund (ERDF).

-

# Tunnel Magnetoresistance - Based Sensor for Detection and Quantification of Volume Distributed Magnetic Nanoparticles

*Crina Ghemes[1]*, Oana-Georgiana Dragos-Pinzaru[1], Mihai Tibu[1], Mihaela Lostun[1], Nicoleta Lupu[1] and Horia Chiriac[1]*

[1] *National Institute of Research and Development for Technical Physics, 700050 Iasi, Romania*

* cghemes@phys-iasi.ro

Since their first report in 1998 as sensors capable of detecting biological signals [1] magnetoresistive (MR) sensors have attracted the interest of the scientific community. Most of the recent research on biosensors involves attempts to develop sensors based on the MR effect to detect magnetic nanoparticles (MNPs). To be reliable in biosensing applications, a MR sensor must be able to detect low particle concentrations and quantify them in a linear range. So far, magnetorelaxometric imaging (MRX), magnetic resonance imaging (MRI), or magnetic particle imaging (MPI) have been used as methods to detect the magnetic response of MNPs [2,3], but there is still a need to improve the cost and efficiency of localizing MNPs, which is of great importance in medicine for use in magnetic hyperthermia. The detection of MNPs by scanning human tissue with a highly sensitive sensor could overcome the main problem of magnetic hyperthermia, heating only the diseased area without affecting the surrounding healthy tissue.

Among magnetoresistive sensors, which have special characteristics such as low-cost fabrication, small size, and low power consumption, those based on the tunnel magnetoresistance (TMR) effect offer better sensitivity and resolution due to their higher magnetoresistance ratio. Therefore, in this work we focused on improving the characteristics of TMR-based sensors to detect volume-distributed MNPs in real-time. The device consists of a TMR sensor with a multilayer structure: Ta (5 nm) / Ru (20 nm) / Ta (5 nm) / CoFe (2.5 nm) / IrMn (20 nm) / CoFe (2.5 nm) / Ru (0.85 nm) / CoFeB (3 nm) / MgO (1.5 nm) /CoFeB (3 nm) / Ta (10 nm) deposited on a $Si/SiO_2$ substrate (18 x 18 $mm^2$) using the ATC 2200 / AJA International deposition system. The surface of each component layer was analyzed and the roughness was minimized to increase the uniformity of the MgO layer, thus improving the properties of the TMR sensor. For the proposed TMR sensor, we measured a maximum magnetoresistance ratio of about 58 % and a sensitivity of 1.45 %/Oe.

To investigate the detection capability of the sensor, a human tissue "phantom" was fabricated from transparent epoxy resin (EPON 812) in which magnetic FeCrNbB nanoparticles were dispersed at various concentrations. After scanning this sample with the improved TMR-based sensor, we observed a linear change in the output signal as a function of nanoparticles concentration, demonstrating the capability to detect and quantify magnetic nanoparticles.

## Acknowledgements

This work was supported by a grant of the Ministry of Research, Innovation and Digitization, CCCDI - UEFISCDI, project number PN-III-P2-2.1-PED-2021-2739, within PNCDI III.

## References

[1] D. R. Baselt, G. U. Lee, M. Natesan, S. W. Metzger, P. E. Sheehan, R. J. Colton, *Biosens. Bioelectron.* **13** (1998).
[2] R. Weissleder, H.C. Cheng, A. Bogdanova, A. Bogdanov Jr., *J. Magn. Reson. Imaging* **7** (1997) 258–263.
[3] M. Liebl, U. Steinhoff, F. Wiekhorst, J. Haueisen, L. Trahms, *Phys. Med. Biol.* **59** (2014) 6607–6620.

# SPION trapping under liquid flow by Magnetophoresis

*Selin Su Karakol[1], Bahar Gungordu[1], Lorin Doğanturk[3], Nilay Gunduz Akdogan[2,3], Ozan Akdogan[1,3]**

[1]*Faculty of Engineering and Natural Sciences, Bahcesehir University, 34353, Istanbul, Turkey.*

[2] *Faculty of Engineering, Piri Reis University, 34940, Istanbul, Turkey.*

[3] *NANOTerial Technology Corporation, 34077, Istanbul, Turkey.*

*ozan.akdogan@eng.bau.edu.tr

Biosensors for the detection of various diseases, the ratios of constituents in blood content, and various biomedical applications have recently become diagnostic kits that science focuses on. Although there are many developments in this field, rapid results, and easy use have not been achieved. With continued advances in science, a magnetic gradient patch (MGP) is open to development, and nanoparticle/bio-entity capture capabilities of MGPs are promising.[1] MGPs have contributed greatly to developing biosensors that allow the identification of biological entities.[1] It was observed in the studies conducted by our group that the ferromagnetic nanoparticle passing through the microfluidic channels in MGP was captured with an efficiency of >90%.[2] It was also discovered in a preliminary study that it could capture RNA with an adequately regulated MGP.[3] Capture efficiencies ranged from 75% to 40% for epoxy and PDMS-based patches, respectively. (Figure 1 a) [3] This work is focused on the trapping efficiency of superparamagnetic iron oxide nanoparticles (SPIONs) under a liquid flow. In particular, $Fe_3O_4$ nanoparticles were synthesized and XRD analyses showed that their size was 7 nm. (Figure 1 b) The developed biosensor shows promise in the diagnosis of various diseases. In addition, thanks to early diagnosis, it offers the patients the opportunity to enter the fast treatment process for diseases.

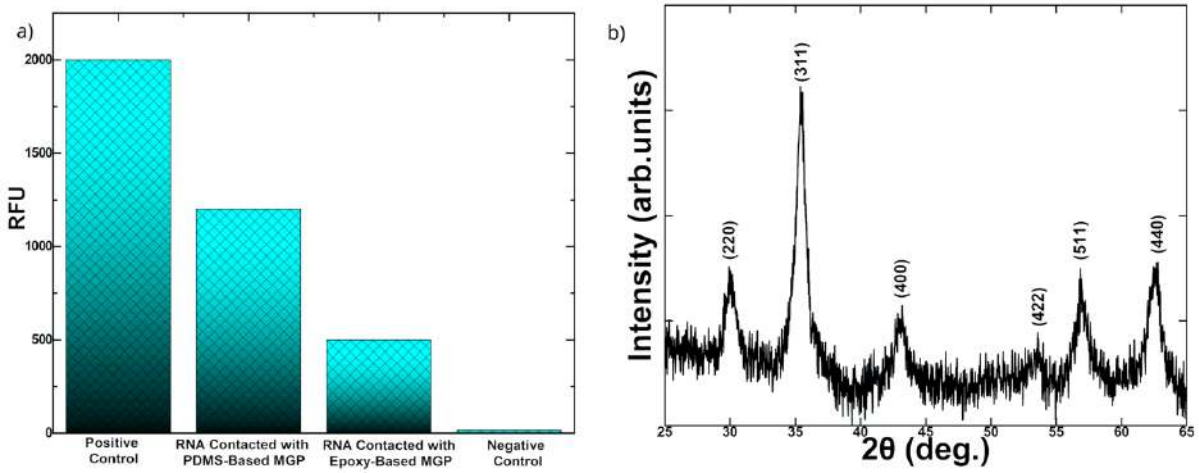



**Figure1.** a) The illustration of the process and the capturing ability of different polymer-based patches. b)XRD analysis graph of produced Fe3O4 nanoparticles.

## Acknowledgments

This work was supported by BAP.2022-02.15 (Bahcesehir University).

## References

[1] Deman, A.L., & Roy, D.L. (2019). Ch 7. Magnetophoresis in Bio-Devices, Engineering of Micro/Nano Biosystems.

**[2]** Ozunlu, S., Akdogan, N. G., Bozkurt, M. N., Doganturk, L., Alshammari, H. A., Le Roy, D., & Akdogan, O. (2021). Innovative technique for patterning Nd-Fe-B arrays and development of a microfluidic device with high trapping efficiency. Nanotechnology, 32(49), 10.1088/1361-6528/ac1dd6.

[3] Doganturk L, Akdogan NG, Polat O, Akdogan O..2022 “Magnetic Bead Free Trapping of Ribonucleic Acid by Magnetophoretic Based Reusable and Cost-Effective Patch (Under Consideration).

# Synthetic Limb, Implant and Prosthesis from Magnetic Microparticle Doped Hydrogel

*Ruya Sera Polat[1] , Ileyna Uvak[1], Nilay G.Akdogan [2,3], Ozan Akdogan[1,3*]*

[1]*Faculty of Engineering and Natural Sciences, Bahcesehir University, 34353 Istanbul, Turkey.*
[2]*Faculty of Engineering, Piri Reis University, 34940, Istanbul, Turkey*
[3]*NANOTerial Technology Corporation, 34077, Istanbul, Turkey*
* ozan.akdogan@eng.bau.edu.tr

The biomedical field is always seeking better and cheaper versions of a prosthesis. The most promising method for this is 3D printing which is developed to fit patients' demands and is designed to be more functional than existing prostheses/implants on the market. Nowadays, hydrogels are preferred for 3D printing bio inks due to their affordable, easy, and accessible production. Additionally, the biocompatibility of the hydrogel makes it a great option for the medical field [1]. Although simple hydrogels are available on the market, their formula is open to new additions/improvements which are then named composite hydrogels. Here, it is intended to create composite hydrogel with magnetic microparticles. A base hydrogel recipe whose biocompatibility is proven is selected for use. Based on their findings, the chosen hydrogel composition (gelatin-cellulose-alginate, 5:2:2%) and post-crosslinking process using $CaCl_2$ are suitable for enhancing the 3D printability and cytocompatibility of the material [2]. Different ferromagnetic powders such as $Fe_{12}O_{19}Sr$, NdFeB, Bismuth, Graphene, and Fe are used to give magnetic properties to the hydrogel. The mechanical characteristics of the composite materials are a combination of those of their constituent materials, and in certain situations, the composites have better mechanical properties than the separate materials [1]. This enriched formula aims to give new characteristics such as magnetism and solve the mechanical strength disadvantage of the hydrogel. Furthermore, the cytocompatibility of the magnetic hydrogels is going to be assessed on fibroblast cells using an MTT assay. The magneto-responsive hydrogel ink, which has the best biocompatibility and magnetic properties, is expected to be used in distinct designs using extrusion-based 3D printing (Figure 1). This method aims to broaden the technologies mainly in the biomedical industry.

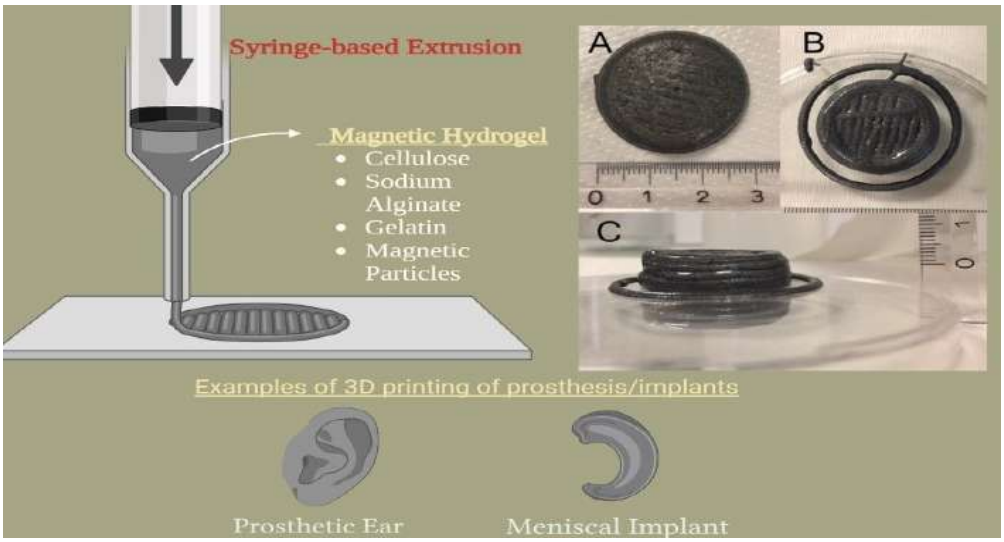



**Figure 1.** To facilitate 3D printing by extrusion, a hydrogel composite consisting of a gelatin/cellulose/alginate combination has been developed.

## Acknowledgments

This work is supported by BAP.2023-01.14 (Bahçeşehir University)

## References

[1] Liu, Y., He, W., Zhang, Z., & Lee, B. (2018). Recent Developments in Tough Hydrogels for Biomedical Applications. Gels, 4(2), 46. MDPI AG. Retrieved from http://dx.doi.org/10.3390/gels4020046

[2] Erkoc, P., Uvak, I., Nazeer, M. A., Batool, S. R., Odeh, Y. N., Akdogan, O., & Kizilel, S. (2020). 3D Printing of Cytocompatible Gelatin-Cellulose-Alginate Blend Hydrogels. Macromolecular bioscience, 20(10), e2000106. https://doi.org/10.1002/mabi.202000106

# Capturing Magnetic Nanoparticles with External Magnetic Fields in the Fluidic Flow

*S. Arjomandi[1,2], I. Dirba[1]*, S. Ener[1], A. Filatova[3], U. Nuber[3], and O. Gutfleisch[1]*

[1] *Functional Materials, Institute of Materials Science, Technical University of Darmstadt, 64287 Darmstadt, Germany*

[2] *Materials Engineering and Nanotechnology Department, Politecnico di Milano, Milan,20133, Italy*

[3] *Biology , Stem Cell and Developmental Biology, Technical University of Darmstadt, 64287 Darmstadt, Germany*

*imants.dirba@tu-darmstadt.de

Magnetic separation to extract material from solution using an external magnetic field enables higher concentrations for purposes such as separation of pathogens in blood samples for analysis. Magnetic drug targeting can guide medication to the precise location of the disease (e.g. cancer) by injecting magnetic particles into the bloodstream and capturing them at the target site under by an external magnetic field. To design devices for these applications, experimental *in situ* studies of forces on magnetic nanoparticles in fluid flow are needed.

In this study therefore, we investigate capturing behaviour of commercial magnetic nanoparticles in phosphate buffered saline solution upon application of external magnetic field. For this purpose, a modified optical microscope setup was used which is equipped with light polarization options and an AC/DC external magnetic field generator up to 500 mT. The external magnetic field was applied perpendicular to the flow direction to observe the capturing behaviour. Effect of fluid flow velocity, diameter of the magnetic nanoparticles and strength of the applied magnetic field on capturing was studied. In addition, the possibility to use alternative magnetic nanoparticles [1] with magnetization higher than the conventionally used iron oxides is considered.

Figure 1 shows the magnetic flux density required for capturing 130 nm and 250 nm iron oxide-based nanoparticles at different flow rates. The larger particles require lower magnetic fields due to their higher magnetic moment. The field necessary for capture increases continuously with the fluid flow because of the hydrodynamic drag force.

## Acknowledgements

We acknowledge funding by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation), Project ID Nos. 471878653, 405553726-TRR 270 and CRC 1487.

## References

[1] I. Dirba, C. K. Chandra, Y. Ablets, J. Kohout, T. Kmječ, O. Kaman and O. Gutfleisch, J. Phys. D: Appl. Phys. **56** (2023) 025001.

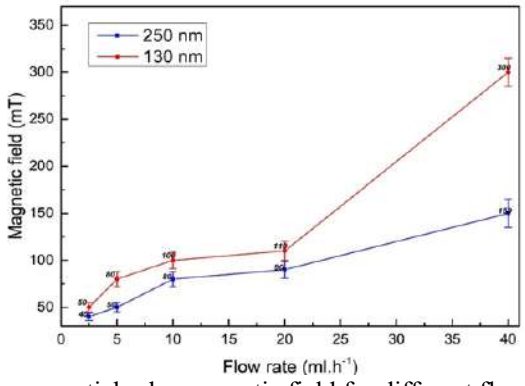



Figure 1: capture of 130 nm and 250 nm nanoparticles by magnetic field for different flow rates.

# Towards High Magnetization Nanoparticles for Biomedical Applications

*I. Dirba[1]*, Y. Ablets[1], K.Riahi[1], C. K. Chandra[1], T. Kmječ[2], L. Kubíčková[2], O. Kaman[2], and O. Gutfleisch[1]*

[1] *Functional Materials, Institute of Materials Science, Technical University of Darmstadt, 64287 Darmstadt, Germany*

[2] *FZU - Institute of Physics of the Czech Academy of Sciences, Cukrovarnická 10, 162 00 Praha 6, Czech Republic*

*imants.dirba@tu-darmstadt.de

Superparamagnetic iron oxide nanoparticles, e.g., γ-$Fe_2O_3$ (maghemite) or $Fe_3O_4$ (magnetite) are used in biomagnetic applications such as magnetic fluid hyperthermia (MFH), as they are inexpensive to produce, chemically stable, with low toxicity and their pathways of metabolism are known. However, due to their ferrimagnetic nature, the saturation magnetization remains moderate. This is particularly important for applications where the concentration is very low as for instance in the case of targeted hyperthermia. For this reason, a novel magnetic material with superior magnetic properties enabling enhanced heating power would be highly beneficial [1]. The heat power generated per particle unit mass should be as high as possible.

In this work we study iron nitride $Fe_3N$ nanoparticles as alternative to the conventional iron oxides and compare to core-shell iron/iron oxide nanoparticles syntesized under comparable conditions. Furthermore, a comparison in terms of heating ability with commercial Fe-O ferrofluids is performed. Bright-field TEM image in Fig. 1a shows that the synthesized iron nitride ε-$Fe_3N$ nanoparticles are monodisperse, exhibit rounded near spherical shapes with a narrow size distribution and an average particle size of 12.7 nm ± 1.2 nm. MFH heating rate measurements were conducted on nanoparticle dispersions in hexane with the concentration of 2 mg/mL. The results are shown in Fig. 1b. The step-like curve of the nitride sample in the beginning is caused by the rapid heating rate that exceeds the too-low signal processing rate for temperature readings of the experimental device. In agreement with the magnetization measurements, the ε-$Fe_3N$ nanoparticles significantly outperform comparable iron oxide nanoparticles, reaching SLP of 540 ± 54 W/g. Results from structural, magnetic, oxidation stability and MFH heating performance investigations will be presented.

**Acknowledgements**

We acknowledge funding by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation), Project ID Nos. 471878653, 405553726-TRR 270 and CRC 1487.

**References**

[1] I. Dirba, C. K. Chandra, Y. Ablets, J. Kohout, T. Kmječ, O. Kaman and O. Gutfleisch, J. Phys. D: Appl. Phys. **56** (2023) 025001.

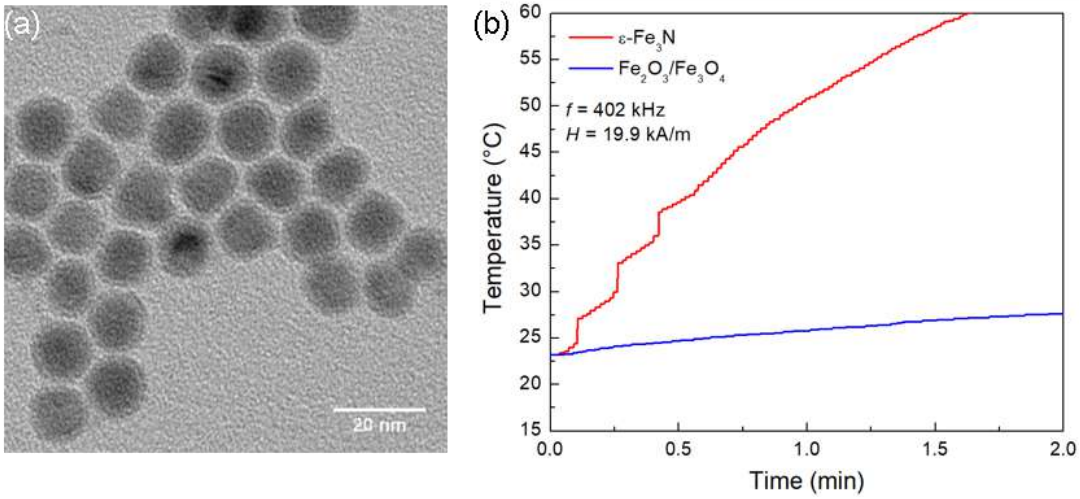



Figure 1: (a) bright-field TEM image of the ε-$Fe_3N$ nanoparticles and (b) heating rate measurements results for the ε-$Fe_3N$ and $Fe_2O_3/Fe_3O_4$ nanoparticles colloidal dispersions in hexane.

# *In silico* analysis of the role of magnetic field applicators in preclinical tests of magnetic hyperthermia

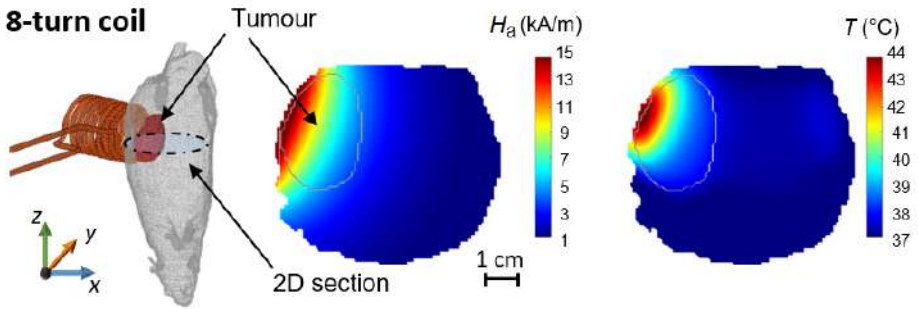
*M. Vicentini[1]*, R. Ferrero[1], A. Manzin[1]*

[1] *Istituto Nazionale di Ricerca Metrologica (INRIM), Strada delle Cacce 91, Torino, Italy*

* m.vicentini@inrim.it

Magnetic hyperthermia has shown great potential as an adjuvant to standard cancer therapies, such as radiotherapy or chemotherapy, enabling a selective heating of the diseased tissues with limited side effects in the healthy ones [1]. This technique is based on the administration of magnetic nanoparticles (MNPs) within a target region and on their activation with AC magnetic fields, with frequency between 50 kHz and 1 MHz [2]. The MNP activation leads to a release of heat that raises the temperature within the tumour, promoting the damage of cancer cells. Preclinical tests on murine models (mice, rats) are commonly used to evaluate the efficacy of this therapeutic technique [3], investigating how to maximize heat deposition and minimize side effects, connected to the occurrence of hotspots. To this aim, several factors have to be taken into account, including the fulfillment of biophysical constraints [4] when choosing magnetic field parameters (frequency and peak amplitude), the geometry of the field applicator and its placement with respect to the body, and the dependence of the MNP thermal efficiency on the experimental conditions.

In this framework, we performed *in silico* tests of magnetic hyperthermia treatment in high-resolution digital phantoms of two murine models (a 30 g mouse and a 500 g rat), to study the role of AC magnetic field sources. In the simulations, we compare different magnetic field applicators, varying their geometry (solenoid, pancake, Helmholtz-type) and the AC supply conditions (frequency and peak amplitude of the supply current). The analysis is performed by means of in-house finite element solvers [5, 6], which enable us to evaluate the magnetic field spatial distribution within the target region, the possible eddy current heating and the thermal effects due to MNP excitation, under different magnetic field application conditions.




The figure shows the results obtained when treating a tumour within the body of a 500 g rat, with iron oxide nanoparticles activated by a 300 kHz magnetic field, generated by a 8-turn coil. The MNPs are considered to be uniformly distributed within the tumor, with a local concentration of 5 mg/cm$^3$.

## Acknowledgements

This work was supported by Project 18HLT06 RaCHy that received funding from the European Metrology Programme for Innovation and Research and from the European Union's Horizon 2020 Programme.

## References

[1] X. Liu *et al*., *Theranostics* **10** (2020) 3793–3815.
[2] Q.A. Pankhurst, J. Connolly, S.K. Jones, J. Dobson, *J. Phys. D: Appl. Phys.* **36** (2003) R167–R181.
[3] H.F. Rodrigues, G. Capistrano, A.F. Bakuzis, *Int. J. Hyperthermia* **37** (2020) 76–99.
[4] R. Hergt, S. Dutz, *J. Magn. Magn. Mater.* **311(1)** (2007), 187–192.
[5] M. Vicentini et al., *Comput. Methods Programs Biomed.* **223** (2022) 106975.
[6] A. Manzin, R. Ferrero, M. Vicentini, *Adv. Theory Simul.* **4(5)** (2021), 2100013.

# Prediction of hyperthermia response in *in vivo* tests versus magnetic nanoparticles type

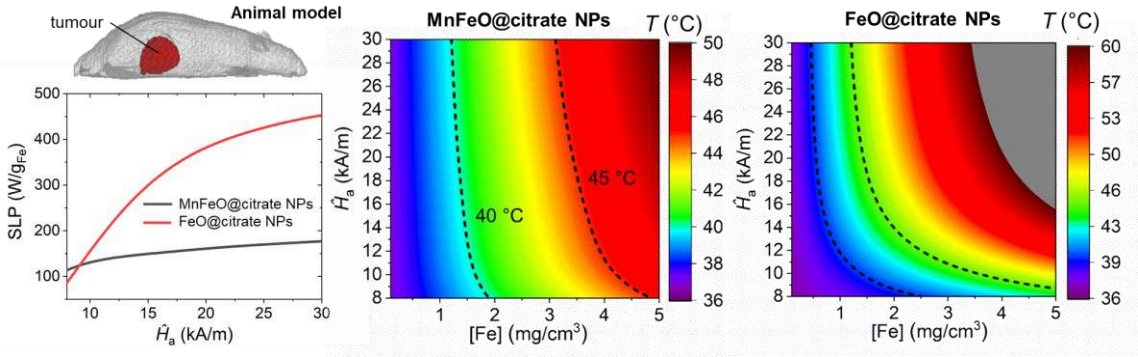
*M. Vicentini*[1] *, *R. Ferrero*[1], *A. Manzin*[1]

[1] *Istituto Nazionale di Ricerca Metrologica (INRIM), Strada delle cacce 91, Torino, Italy*

* m.vicentini@inrim.it

In magnetic hyperthermia, the temperature increase in diseased tissues is achieved after the administration and activation of magnetic nanoparticles (MNPs), which release heat when exposed to AC magnetic fields with frequencies between 50 kHz and 1 MHz [1]. The reaching of a target temperature in the tumour mass, within the range of 40-45 °C, contributes to cancer cell damage and increases cell sensitivity to other cancer therapies. The heating efficiency of MNPs is generally quantified by means of the specific loss power (SLP), which defines the power dissipated per unit mass of magnetic material. The SLP is affected by several factors, including MNPs' material composition, size, shape and surface coating [2, 3], as well as AC magnetic field parameters (peak amplitude and frequency), which have to fulfil biophysical constraints to limit eddy current effects. A crucial aspect when planning magnetic hyperthermia treatments is the monitoring of the temperature increase, considering that the target temperature range should be ideally reached in the entire tumour region and maintained for a sufficiently long time (from 20–30 min up to one hour). However, a complete knowledge of the temperature distribution within the region of interest is impossible during hyperthermia sessions.

In this context, we present a physics-based modelling approach to support *in vivo* tests of magnetic hyperthermia [4]. In particular, we have developed an *in silico* tool, which enables us to evaluate the spatial-temporal distribution of the temperature in the tumour region to be treated, versus the MNP heating properties (SLP), the AC magnetic field parameters, and the MNP local concentration. In the study, we test different MNPs [5] by conducting simulations on high-resolution digital phantoms of a rat and a mouse. The analysis is completed by integrating the results from thermal simulations with regression models, like multiple linear regression and Gaussian process regression.




Average temperature evaluated within the tumour of the mouse model for two different types of citrate-coated MNPs [5]: manganese ferrite (MnFeO) NPs, and iron oxide (FeO) NPs. Results obtained by considering a uniform magnetic field distribution and by varying the magnetic material concentration [Fe] and the field amplitude $\hat{H}_a$. The field frequency is fixed to 150 kHz and the SLP values of the MNPs are reported on the left as a function of the $\hat{H}_a$.

## Acknowledgements

This work was supported by Project 18HLT06 RaCHy that received funding from the European Metrology Programme for Innovation and Research and from the European Union's Horizon 2020 Programme.

## References

[1] E. A. Périgo et al., *Appl. Phys. Rev.* **2(4)** (2015) 041302.
[2] H. Gavilán et al. *Chem. Soc. Rev.* **50** (2021), 11614-11667.
[3] M. Vassallo et al. *ACS Omega* **8(2)** (2023), 2143–2154.
[4] A. Manzin, R. Ferrero, M. Vicentini, *Adv. Theory Simul.* **4(5)** (2021), 2100013.
[5] F. Soetaert, S.K. Kandala, A. Bakuzis, R. Ivkov, *Sci. Rep.* **7** (2017) 6661.

# Nanomagnetic force in action: towards magnetically guided nerve regeneration

*T. Nahar[1], M. Gates[1,2], E. Secret[3], J.M. Siaugue[3], J. Fresnais[3], M. Rotherham[1,4], A.J. El-Haj[4] and N. D. Telling[*]*

[1] *School of Pharmacy and Bioengineering, Keele University, Staffordshire, UK*

[2] *School of Medicine, Keele University, Staffordshire, UK*

[3] *Sorbonne Université, CNRS, Physico-Chimie des Électrolytes et Nanosystèmes Interfaciaux, PHENIX, 75005, Paris, France*

[4] *Healthcare Technology Institute, Institute of Translational Medicine, University of Birmingham, Birmingham, United Kingdom*

*corresponding author email address

Damage to the neural circuitry in the central nervous system (CNS) can be caused by acute trauma (for example spinal cord injury) or following the development of neurological disorders such as Parkinson's disease. Unfortunately, the ability of axons in the CNS to naturally regenerate is extremely limited, and so the functional deficits that result from damage to the brain or spinal cord can persist indefinitely. In this study magnetic nanoparticles were sequestered within intracellular compartments known as endosomes in neuronal cells. This accumulation of nanoparticles enabled the formation of micrometer sized aggregates within the cell (in effect forming magnetic endosomes). The magnetic forces effective on these endosomes are significantly larger than those for individual nanoparticles. We were able to manipulate this intracellular magnetic force by the application of suitable magnetic field gradients to the cells, influencing the direction of the neurites (projections) that grew from the cell bodies (Figure 1).

The average direction of neurite outgrowth in different cell regions was quantified using a 2D Fourier transform analysis, and showed excellent agreement with derived magnetic force vectors for the field configuration used. Significantly, the control of orientation was found to be effective over areas >1cm$^2$ using only modest forces of ~10 fN per endosome, apparently limited only by the local population density of cells. However, in regions where the force vectors converged, large (~100 µm) nanoparticle loaded neurospheres (ball-like collections of cells) were seen to form, connected by unusually thick linear neurite fibres. This suggests a magnetically driven enhancement of neurosphere growth, with the neurospheres themselves contributing to the local forces that direct outgrowth. Such structures, which have not been previously observed, could provide new insights into the development and possible enhancement of neural circuitry.

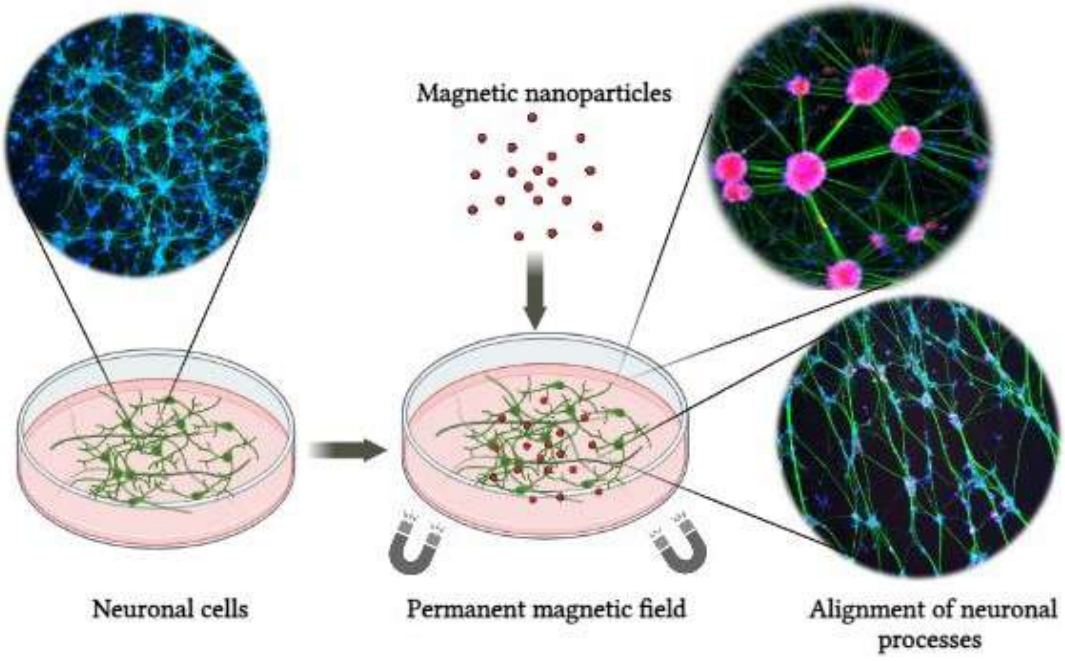



Figure 1: Nanomagnetic force methodology applied to neuronal cells. Insets show fluorescence microscopy images of neuronal cells with the cell bodies stained with green fluorescent dye and the cell nuclei with blue fluorescent dye. The nanoparticles are labelled with a red fluorophore and dense neurospheres which were heavily loaded with nanoparticles can be seen in pink (from the overlap of the blue and red fluorescence).

# Enhancing the Magnetic Response of Calcium Phosphate-Based Powders for Bone Tissue Regeneration

*T. S. S. Carvalho[1*], P.M.C. Torres[1], J. H. Belo[2*], J.C.C. Abrantes[1,3], D. Lopes[1], A. Kavaleuski[1], J. P. Araújo[2], M. Bañobre-Lopez[4], S.M. Olhero[1]*

[1] *Department of Materials and Ceramics Engineering (DEMaC), CICECO-Aveiro Institute of Materials, University of Aveiro, 3810-193 Aveiro, Portugal*

[2] *Institute of Physics of Advanced Materials, Nanotechnology and Photonics (IFIMUP), Departament of Physics and Astronomy, University of Porto, 4169-007 Porto, Portugal*

[3] *proMetheus, Polytechnic Institute of Viana do Castelo, 4900 Viana do Castelo, Portugal*

[4] *Diagnostic Tools & Methods/Advanced (Magnetic) Theranostic Nanostructures Lab, International Iberian Nanotechnology Laboratory (INL), Av. Mestre José Veiga s/n, 4715-330 Braga, Portugal.*

*tssc@ua.pt and jbelo@fc.up.pt

Characteristics such as versatility and low cost have catapulted magnetic materials such as iron oxides to a highlighted position in biomedical research, namely in bone tissue engineering. In the last decades, these type of materials have been developed as different structures like films, scaffolds, and implants, benefiting of their magnetic response by the application of a static external magnetic field to help improve bone tissue regeneration [1,2]. Recently, the doping of calcium phosphates (CaP) with iron ions has been shown a promising alternative for multifunctional applications for both bone restoration and cancer hyperthermia, encouraging the huge interest in such materials.

The main purpose of this research was the development of magnetic CaP-based powders doped with iron ions (FeCaP) by a wet chemical precipitation method, with the intention to improve the magnetic susceptibility of their undoped peers. The influence of thermal treatment on the crystalline structure and phases formed was evaluated and correlated with their magnetic and magneto-thermal properties. In contrast to other works reported in literature [3], the thermal treatment not only improved the crystallinity of the resultant FeCaP powders, but also improved their saturation magnetization and hyperthermic performance when in comparison with as-synthesized powders, exhibiting a ferromagnetic-like behavior for both conditions. In this work, magnetic CaP-based powders have been successfully obtained, with great potential for the development of new materials for hard tissue regeneration.

**Acknowledgements**

T. S. S. Carvalho (2021.06481.BD) wishes to thank Fundação para a Ciência e Tecnologia (FCT) for supporting their work. This work is also funded by FEDER funds through the COMPETE 2020 Program and National Funds through FCT under the projects 2BBone (PTDC/CTM-CER/29940/2017) and FlexMicroDerm (PTDC/BTM-MAT/29274/2017). The project CICECO-Aveiro Institute of Materials, UIDB/50011/2020, UIDP/50011/2020 & LA/P/0006/2020, financed by national funds through the FCT/MCTES (PIDDAC) is also acknowledged. P. M. C. Torres acknowledges FCT for CEECIND/01891/2017 contract. J H Belo thanks FCT for the projects PTDC/FISMAC/31302/2017, PTDC/EME-TED/3099/2020 and CERN/FISTEC/0003/2019, and for his contract DL57/2016 (SFRH-BPD-87430/2012).

## References

[1] J. Peng, J. Zhao, Y. Long, Y. Xie, J. Nie, and L.Chen, "Magnetic Materials in Promoting Bone Regeneration," Front. Mater., **6** (2019) 268

[2] D. Fan, Q. Wang, T. Zhu, H. Wang, B. Liu, Y. Wang, Z. Liu, X. Liu, D. Fan, X. Wang, Recent Advances of Magnetic Nanomaterials in Bone Tissue Repair, Front. Chem. **8** (2020) 745.

[3] A. Tampieri, T. D'Alessandro, M. Sandri, S. Sprio, E. Landi, L. Bertinetti, S. Panseri, G. Pepponi, J. Goettlicher, M. Bañobre-López, J. Rivas, Intrinsic magnetism and hyperthermia in bioactive Fe-doped hydroxyapatite, Acta Biomater. **8** (2012) 2.

# SYMPOSIUM 04.
## MAGNETISM FOR QUANTUM TECHNOLOGY.
## S4. INVITED ORAL PRESENTATIONS

# Coherent Manipulation of Spins in Diamond via Spin-Wave Mixing

*Toeno van der Sar*[1]*

[1] *Department of Quantum Nanoscience, Kavli Institute of Nanoscience, Delft University of Technology. Lorentzweg 1, 2628CJ Delft, The Netherlands*

*t.vandersar@tudelft.nl

Magnetic imaging based on nitrogen-vacancy (NV) spins in diamond enables probing condensed matter systems with nanoscale resolution[1]. In this talk, I will introduce NV magnetometry as a tool for imaging spin waves – the collective spin excitations of magnetic materials. Using the NV sensitivity to microwave magnetic fields, we can map coherent spin waves[2] and incoherent magnon gases[3] and provide insight into their interaction and damping underneath metals[4]. By using a single NV in a scanning diamond tip we gain access to spin-wave scattering at the nanoscale[5]. I will highlight the use of spin-wave mixing to generate frequency combs that enable high-fidelity, coherent control of the NV spins even when the microwave drive fields are far detuned from the NV spin resonance frequency[6] (see Figure). Our results form a basis for developing NV magnetometry into a tool for characterizing spin-wave devices and expand the control and sensing capabilities of NV spins.

## References

[1] F. Casola, T. van der Sar, and A. Yacoby, *Nat. Rev. Mater.* **3**, 17088, (2018).
[2] I. Bertelli *et al.*, *Sci. Adv.* **6**, eabd3556, (2020).
[3] B. G. Simon*, S. Kurdi* *et al.*, *Nano Lett.* **21**, 8213, (2021).
[4] I. Bertelli *et al.*, *Adv. Quantum Technol.* **4**, 2100094, (2021).
[5] B. G. Simon*, S. Kurdi*, J. J. Carmiggelt, M. Borst, A. J. Katan, and T. van der Sar, *Nano Lett.* **22**, 9198, (2022).
[6] J. J. Carmiggelt *et al.*, *Nat. Commun.* **14**, 490, (2023).

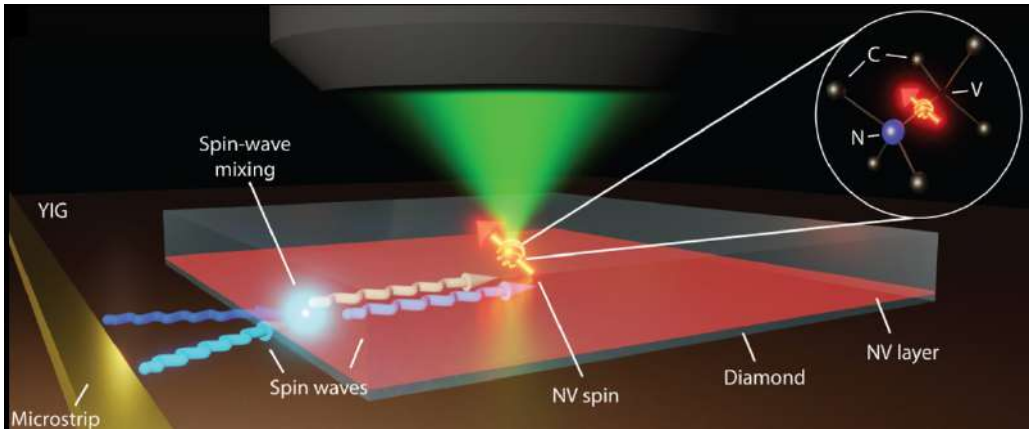



Figure: Using spin-wave mixing for coherent manipulation of spins in diamond [6].

# Improved Imaging with a Scanning Nitrogen-Vacancy Microscope

*William Huxter[1]*, Marius Palm[1], Miranda Davis[1], Martin Sarott[2], Morgan Trassin[2] and Christian Degen[1,3]*

[1] *Department of Physics, ETH Zürich, Zürich, Switzerland*
[2] *Department of Materials, ETH Zürich, Zürich, Switzerland*
[3] *Quantum Center, ETH Zürich, Zürich, Switzerland*
*whuxter@phys.ethz.ch

The continued development of spintronic and magnetoelectric devices benefits from a detailed understanding of the magnetic (and sometimes multiferroic) structure and behaviour of materials at surfaces and interfaces. In pursuit of such goals, we use a scanning probe technique with a single nitrogen-vacancy (NV) centre at the tip apex [1]. Scanning NV microscopy is a non-invasive, ambient-operating imaging technique capable of measuring magnetic fields with high spatial resolution [2]. This technique is commonly used to quantitatively image domain walls in thin ferromagnets, ferrimagnets, and antiferromagnets; however, DC sensitivity limitations hinder the detection of weaker stray fields [3].

We present a detection scheme that improves field sensitivity by at least one order of magnitude [4]. Our technique is based on the upconversion of a static magnetic field gradient into an effective AC signal via mechanical oscillation of the scanning probe. Through synchronisation of the probe's oscillation and coherent NV spin manipulations, we leverage AC detection protocols to improve our measurement sensitivity. With this gain in sensitivity, we are able to image weakly magnetic materials (with magnetisations on the order of 10 A/m) and resolve the nanotesla-sized fields from atomic steps on the antiferromagnetic $Cr_2O_3$(0001) surface. Additionally, our technique can be combined with regular DC measurements to produce superior DC field images.

As an extension of our detection scheme, we also image ferroelectric materials through their stray electric fields [5]. Electric field detection relies on the NV centre's Stark shift [6] and AC detection of the gradient signal mitigates the detrimental charge screening which prohibited previous imaging attempts. We extract ferroelectric domain distributions and gain information about the local charge environment on ferroelectric $PbZrTiO_3$ and $YMnO_3$.

By measuring magnetic and electric fields with improved sensitivity, we expand the scope of nanoscale phenomena accessible to scanning NV microscopy. In doing so, we establish a microscopy platform capable of imaging general magnetic, ferroelectric, and multiferroic materials.

**Acknowledgements**

This work was supported by the European Research Council through ERC CoG 817720 (IMAGINE), Swiss National Science Foundation (SNSF) project grants no. 200020_175600, no. 200021_188414, and Spark grant CRSK-2_196061, the National Centre of Competence in Research in Quantum Science and Technology (NCCR-QSIT), grant no. 51NF40-185902 and the ASTERIQS program of the European Commission, grant no. 820394.

**References**

[1] C. L. Degen, *Appl. Phys. Lett.* **92** (2008) 243111.
[2] L. Rondin et al., *Rep. Prog. Phys.* **77** (2014) 056503.
[3] E. Marchiori et al., *Nat. Rev. Phys.* **4** (2022) 49-60.
[4] W. S. Huxter et al., *Nat. Commun.* **13** (2022) 3761.
[5] W. S. Huxter et al., *Nat. Phys.* (2023).
[6] F. Dolde et al., *Nat. Phys.* **7** (2011) 459.

# SYMPOSIUM 04.
## MAGNETISM FOR QUANTUM TECHNOLOGY.
## S4. ORAL PRESENTATIONS

# Quantum Cavities Based On Magnonic Textures

*D. García-Pons, Carlos A. González, J. Pérez-Bailón, F. Luis, D. Zueco and María José Martínez-Pérez*

*Instituto de Nanociencia y Materiales de Aragón CSIC-Universidad de Zaragoza, Calle Pedro Cerbuna 12, 50009 Zaragoza, España*

*pemar@unizar.es

Solid state quantum computing and quantum sensing technologies are based on the strong coupling between qubits and a quantized field of excitations. Besides photons, the solid state offers a wide variety of bosonic excitations that can be emitted or absorbed such as, e.g., magnons, the quantum version of spin waves.

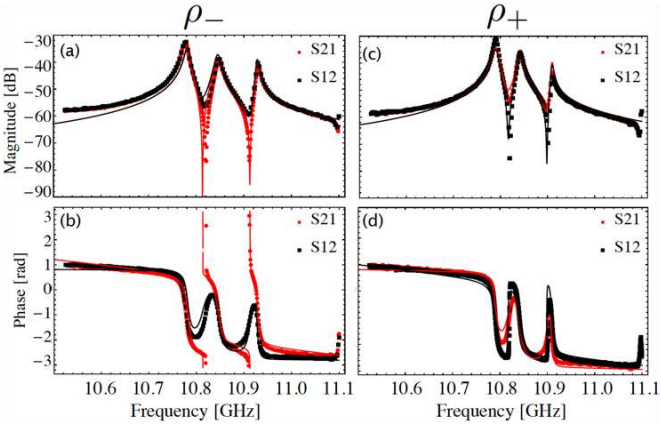
Magnonic cavities offer the advantage of operating at reduced wavelengths compared to electromagnetic resonators of the same frequency. Here, we investigate the integration of magnonic cavities based on topological magnetic solitons as, e.g., magnetic vortices. The latter are extremely stable magnetic textures exhibiting a very rich dynamical behavior in the sub-GHz to tens of GHz range. We focus on the coupling of individual spin qubits to vortex cavities for sensing and quantum computing applications.

# Generation of circulating cavity magnon polaritons

*Jeremy Bourhill[1], Weichao Yu[3], Vincent Vlaminck[3,4], Guillaume Bourcin[3,4], Gerrit E. W. Bauer[6,7] Giuseppe Ruoso[8] and Vincent Castel[3,4*]*

[1] *ARC Centre of Excellence for Engineered Quantum Systems and ARC Centre of Excellence for Dark Matter Particle Physics, University of Western Australia, 35 Stirling Highway, Australia*
[2] *State Key Laboratory of Surface Physics and Institute for Nanoelectronic Devices and Quantum Computing, Fudan University, Shanghai, 200433 China*
[3] *IMT Atlantique, Technopole Brest-Iroise, CS 83818, 29238 Brest Cedex 3, France*
[4] *Lab-STICC (UMR 6285), CNRS, Technopole Brest-Iroise, CS 83818, 29238 Brest Cedex 3, France*
[6] *WPI-AIMR and IMR, Tohoku University, Sendai 980-8577, Japan*
[7] *Zernike Institute for Advanced Materials, Groningen University, Groningen, Netherlands*
[8] *INFN, Laboratori Nazionali di Legnaro, Viale Dell'Universita 2, 35020 Legnaro, Padova, Italy*
*vincent.castel@imt-atlantique.fr

The study of cavity magnonics lead to the discovery of new and interesting regimes of the coupling dynamics between photons and magnons. The field attracted initial attention by the relative ease of achieving strong coupling, even at room temperature because of the large spin density in magnetic materials leads to a large coupling that easily exceeds cavity loss rates and thereby hybridized quasi-particles (magnon polaritons). Yu et al. [1] proposed generating non-reciprocity in a circular microwave cavity by breaking its time-reversal symmetry (TRS) with magnetic loads with preferential clockwise vs. counterclockwise spin and energy flows. The TRS breaking can be achieved by positioning magnets on special chiral lines in a microwave cavity on which the propagating photons are chiral, i.e. the sign of their circular polarization is locked to their linear momentum. This causes a strong direction-dependent coupling with the magnon excitations that can be controlled by applied magnetic fields.



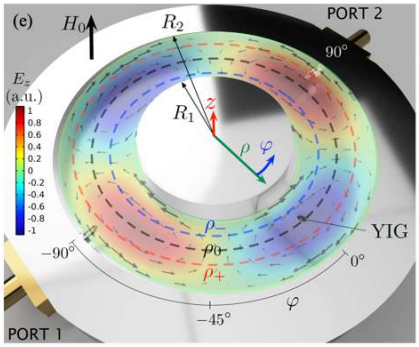




Figure: (a)-(d) Transmission spectra when the YIG sphere is located at $\rho_-$ and $\rho_+$ defined in (e). The thin red and black lines are the fit obtained by the model scattering matrix. (e) Torus-shaped cavity overlayed with a snapshot of the simulated electromagnetic fields of the m=2 TE cavity mode.

Here, we experimentally demonstrate circularly polarized, unidirectional magnon polaritons, thereby confirming theoretical predictions [1]. We place a YIG sphere inside a newly machined torus-shaped cavity on special positions and tune the magnetic resonance to a transverse electric (TE) cavity mode. We detect the coupling dynamics in the microwave scattering matrix as a function of an applied magnetic field. The experimental results agree with the simulations and the non-reciprocity of scattering parameters confirm the chiral nature of the hybrid modes.

## Acknowledgements

This work was funded by the Région Bretagne through the project OSCAR-SAD18024.

## References

[1] W. Yu, T. Yu, G. E. W. Bauer, *Phys. Rev. B* **102** (2020) 064416.

# Observation of the coupled magnon-phonon mode splitting in a surface acoustic wave cavity device

*Yunyoung Hwang [1, 2], Jorge Puebla [2]*, Kouta Kondou [2], Kei Yamamoto [3, 1], Sadamichi Maekawa [2, 4], Yoshichika Otani [1, 2]*

[1] *Institute for Solid State Physics, University of Tokyo, 5-1-5 Kashiwanoha, Kashiwa, Chiba 277-8581, Japan*

[2] *CEMS, RIKEN, 2-1 Hirosawa, Wako, Saitama 351-0198, Japan*

[3] *Advanced Science Research Center, Japan Atomic Energy Agency, Tokai 319-1195, Japan*

[4] *Kavli Institute for Theoretical Sciences, University of Chinese Academy of Sciences, Beijing 100049, People's Republic of China*

*jorgeluis.pueblanunez@riken.jp

The interaction between magnons and phonons has regained attention in the last few years. Of particular interest, it is to explore the ramifications of enhancing the strength of the interaction, towards the formation of a hybrid magnon-phonon quasiparticle in the strong coupling regime, with tantalizing perspectives [1]. The experimental signature of strong coupling is the mode splitting or band anticrossing at the intersection of two quasiparticles dispersions. In fact, magnon-phonon band anticrossing is being recently reported, for the case of a nanoscale magnet with phonons thermally excited by a pulse laser [2]; for the case of a YIG/GGG/YIG heterostructure, where the magnetic precession excited by a microwave antenna generates shear bulk phonons via magnetostriction [3]; and for the case of the layered antiferromagnet $FePS_2$, by studying the magnon band evolution by Raman spectroscopy at high magnetic fields [4]. In our work, we carefully engineer a planar surface acoustic wave (SAW) device in the presence of an acoustic cavity in the GHz frequency, having a CoFeB thin film as magnetic specimen within our acoustic cavity device. We characterize the resonant phonon absorption/transmission energy using a vector network analyser at room temperature, while applying external in-plane magnetic field. The device design and experimental details are similar to those in one of our previous studies [5]. In our study, we pay particular attention to minimise the magnetic damping of our CoFeB films and the losses of phonon energy in our acoustic cavity device. As a result, our experiments show an evident magnon-phonon mode splitting. By fitting, we estimated the coupling strength in the order of hundred-megahertz, being larger than both phonon and magnon relaxation rates in our device, therefore, achieving the requisites of the strong coupling regime. As we increase the thickness of the CoFeB, we notice a linear increase of the coupling strength, in contrast to the known square-root dependence of the quasiparticles in the interaction $\sqrt{N}$, the so-called "Jaynes-Cummings ladder". We argue that different from other recent reports of magnon-phonon band anticrossings, our SAW acoustic wave device offers larger flexibility for research and easier technological implementation, as exemplified by the resurgent interest of SAWs in spintronic research [6]. In analogy to the first implementation of exciton-polariton experiments in a semiconductor optical microcavity device in the 90's [7], we expect that the implementation of SAW acoustic devices accelerate the understanding of phenomena occurring in the magnon-phonon strong coupling regime, and facilitate technology developments.

## Acknowledgments

This work was supported by Grants-in-Aid for Scientific Research (S) (No. 19H05629) from the Ministry of Education, Culture, Sports, Science, and Technology of Japan.

## References

[1] Y. Li, W. Zhang, V. Tyberkevych, W.-K. Kwok, A. Hoffmann, V. Novosad, *J. Appl. Phys.* 128, (2020) 130902.
[2] C. Berk, M. Jaris, W. Yang, S. Dhuey, S. Cabrini, H. Schmidt, *Nat. Comm.* 10, (2019) 2652.
[3] K. An et al., *Phys. Rev. B* 101, (2020) 060407(R).
[4] S. Liu et al., Phys. Rev. Lett. 127, (2021) 097401.
[5] Y.Hwang, J. Puebla, M. Xu, A. Lagarrigue, K. Kondou and Y. Otani, *Appl. Phys. Lett.* 116, (2020) 252404.
[6] J. Puebla, Y. Hwang, S. Maekawa, Y. Otani, Appl. Phys. Lett. 120 (2022), 220502.
[7] C. Weisbuch, M. Nishioka, A. Ishikawa, and Y. Arakawa, Phys. Rev. Lett. 63 (1992) 23.

# Magnetoresistive sensors for measuring currents in quantum devices.

*Claude Fermon*[1], *Myriam Pannetier Lecoeur*[1] , *Aurélie Solignac*[1] *and Elodie Paul*[1]
[1] *SPEC, CEA, CNRS, Université Paris-Saclay, CEA Saclay 91191 Gif-sur-Yvette Cedex, France*
*Claude.fermon@cea.fr

Magnetoresistive sensors have a large sensitivity even at micron size. As they are mainly metallic systems, they are able to be cooled down to cryogenic temperatures without loosing their properties. This has been exploited to measure for the first time the gate voltage–dependent magnetization of a single graphene monolayer encapsulated between boron nitride crystals. The signal exhibits a strong diamagnetic peak at the Dirac point consistent with the theory [1].
More recently, paramagnetic singularities and additional diamagnetic singularities of the orbital magnetism in graphene with a moire potential have been measured in aligned graphene monolayer/HBN crystals [2].
We will present the path of optimization of Giant Magneto-Resistance 5GMR) sensors for this kind of applications which require highly sensitive sensors with small shapes and top surfaces compatible with graphene deposition and we will give the main properties of such devices.
We will discuss the signal to noise limit achievable at very low temperature and the interest of moving or not from GMR to Tunnel Magneto-Resistance (TMR) devices for this kind of measurement.
Finally we will present the residual stray field created by the GMR stacks, the way to reduce this effect and its possible influence for the use of magnetic sensors for probing quantum states of Qbits.

**References**

[1] Vallejo Bustamante, J., Wu, N. J., Fermon, C., Pannetier-Lecoeur, M., Wakamura, T., Watanabe, K., ... Bouchiat, H. Science, **374**(6573), 1399-1402. (2021).
[2] Vallejo Bustamante, J *et al*, Submitted.

# Spin coupling to electromagnetic fields through Jahn-Teller and spin-orbit interactions

*A.S. Miñarro*, B. Casals, G. Herranz**

*Institut de Ciència de Materials de Barcelona (ICMAB-CSIC), Campus UAB, 08193 Bellaterra, Catalonia, Spain*

*asanchez3@icmab.es, *gherranz@icmab.cat

Spin orbit interactions are key for spintronics, quantum computation and topological matter. They affect the transport dynamics in solids and through spin-dependent phenomena they enable the manipulation of spin degrees of freedom. On the other hand, the manipulation of solid-state electron spins is a promising route towards quantum computation, with interest in finding pathways to couple efficiently the electric field of photons to spins. Recently, our group has investigated how quantum spin states interact with electromagnetic fields, which may hold potential to tune their properties and reveal interesting physics. Using magneto-optical spectroscopy, we have uncovered a remarkably large gyrotropic signal in some Jahn-Teller manganites [1], which we attribute to the interaction of Jahn-Teller polarons with electromagnetic radiation [2]. By gyrotropy we mean a different response to light of opposite handedness, i.e., different response to left versus right circularly polarized light. Interestingly, the gyrotropic signal is directly related to a field-induced transition in which the spin of Jahn-Teller polarons is inverted. We have rigorously proved this observation, using a group-theoretic analysis of many-electronic quantum states in a multiconfiguration method [3]. Additionally, combining group-theoretical analysis and Green's functions formalism we have determined appropriate selection rules, which prove that the efficiency of the spin-photon coupling depends critically on the relative orientations of the propagation of light, spin quantization and Jahn-Teller distortions, which is consistent with our experimental optical spectra. Our work shows that electromagnetic interactions with spins in Jahn-Teller systems offer the opportunity to use optical wavelengths to entangle orbital and spin degrees of freedom. We believe that the realization of spin-photon coupling enabled by Jahn-Teller interactions can be extended to other transition metal compounds where Jahn-Teller physics is relevant (e.g., correlated manganites, cobaltites, Jahn-Teller molecules). In this regard, we envisage the use of electromagnetic fields to entangle spin and orbital degrees of freedom, which could allow studying quantum tunneling of vibronic states, which could form a so far unexplored basis for quantum states in quantum technologies.

## Acknowledgements

We acknowledge financial support from Projects No. PID2020-118479RBI00 and Severo Ochoa FUNFUTURE (No. CEX2019-000917-S) of the Spanish Ministry of Science and Innovation (Grant No.MCIN/AEI/10.13039/501100011033) and Generalitat de Catalunya 2021 SGR 00445.

## References

[1] B.Casals et al.,Phys. Rev. Lett. 117, 026401 (2016)

[2] J.M. Caicedo, J. Fontcuberta, and G. Herranz, Physical Review B, 89 045121 (2014))

[3] A.S. Miñarro and G. Herranz, Phys. Rev. B 106, 165108 (2022)

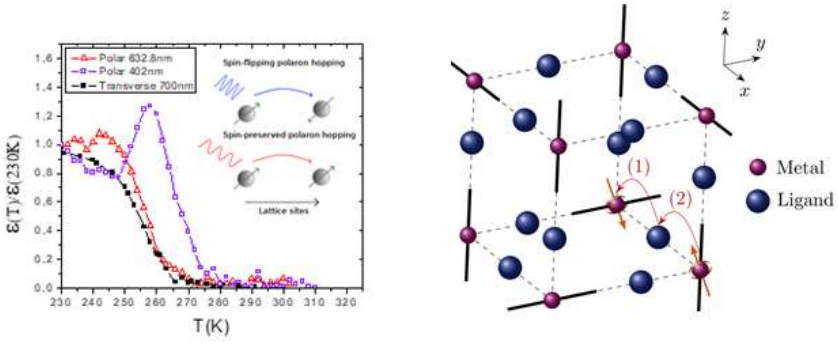



Figure: Figure: (Left) Temperature dependence of Kerr ellipticity (ε) of a $La_{2/3}Ca_{1/3}MnO_3$ film measured at different wavelengths. Around the Curie temperature, ε is strongly enhanced for blue-violet wavelengths. This enhancement is absent in the red region of the spectrum. The gyrotropic increase is due to spin-reversal photoinduced hopping driven by photons of large enough energy (inset and right panel).

# Theory of Fractionally-magnetized Quantum Ferromagnet

*Isao Maruyama[1]*, Shin Miyahara[2]*

[1] *Department of Information and Systems Engineering, Fukuoka Institute of Technology, 3-30-1 Wajiro-higashi, Higashi-ku, Fukuoka 811-0295, Japan*

[2] *Department of Applied Physics, Fukuoka University, 8-19-1 Nanakuma, Jonan-ku, Fukuoka 814-0180, Japan*

*i-maruyama@fit.ac.jp

A new kind of magnetism, such as L. Néel's ferrimagnetism and F. D. M. Haldane's antiferromagnetism, opens a new research field. For the latter, the entangled gapped quantum spin-liquid state[1], where fractionalized $1/2$ spins that form an entangled spin singlet on a bond in the valence-bond-solid picture, is impotant in quantum computer science.

Recently, we find a new ferromagnetism with quantum entanglement theoretically[2]. Our theory is based on rigorous correspondence between spin-1/2 model and spin-S model on a general lattice in any dimension. As a numerical validification, quantum phase diagram of spin-S bilinear biquadratic chains described by

$$H_\alpha = \cos\alpha \sum_i \boldsymbol{S}_i \cdot \boldsymbol{S}_{i+1} + \sin\alpha \sum_i (\boldsymbol{S}_i \cdot \boldsymbol{S}_{i+1})^2$$

has been calculated by the exact-diagonalization method. As a result, we obtain the phase transition point $\alpha_c$ from fully magnetized classical ferromagnetic phase to a new fractionally magnetized "quantum" ferromagnetic phase. The S=2 case is summarized in the Figure. At the rigorous eigenstate correspondence point $\alpha_r$, it is easy to understand that ferromagnetic ground state even after spontaneous magnetization can have quantum entanglement because corresponding antiferromagnetic ground state is known to have quantum entanglement. A key to realizing an entangled ferromagnetic state is to partly create an antiferromagnetic quantum state in a ferromagnetic classical background, that is, "spin liquefaction" of a ferromagnet. In the presentation, we will detail spin-1/2 liquefaction in spin-S bilinear biquadratic chains.

**Acknowledgements**

This work was supported by Japan Society for the Promotion of Science (JSPS) KAKENHI Grants No. 22H01171.

**References**

[1] F. D. M. Haldane, Nobel lecture: Topological quantum matter, Rev. Mod. Phys. 89, 040502 (2017).

[2] I. Maruyama, S. Miyahara, in preparation

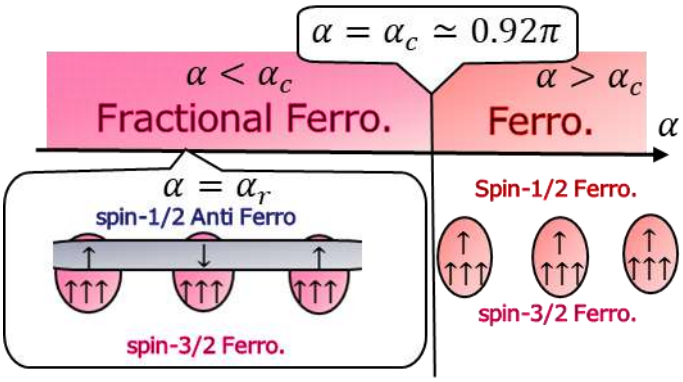



Figure: Phase diagram of S=2 BLBQ chain near the end point of classical Ferromagnetic phase

# SYMPOSIUM 04.
## MAGNETISM FOR QUANTUM TECHNOLOGY.
## S4. POSTERS

# Electrical two-qubit gates within a pair of clock-qubit magnetic molecules

*Alejandro Gaita-Ariño[1]*, Ziqi Hu,[1] Aman Ullah[1], Jesús Cerdá[1], Juan Aragó[1]*

[1] *ICMol, Universitat de València, c/cat José Beltrán, 2, 46980, Paterna, Spain*

*gaita@uv.es

Enhanced coherence in crystals containing $HoW_{10}$ molecular spin qubits was demonstrated by use of so-called clock transitions (CTs).[1] Later it was shown that, while operating at the CTs, it was possible to use an electrical field to selectively address subsets of $HoW_{10}$ molecules within a crystal that contains two kinds of identical but inversion-related molecules.[2] We recently theoretically explored the possibility of employing electric field pulses to effect entangling two-qubit quantum gates within a 2-qubit Hilbert space resulting from dipolar coupling of two neighbouring CT-protected $HoW_{10}$ molecules in a diluted crystal.[3] We lay out how to combine a sequence of microwave and electric field pulses to achieve coherent control within a switchable two-qubit operating space between symmetric and asymmetric qubit states that are protected both from spin-bath and from phonon-bath decoherence. This two-qubit gate approach presents an elegant correspondence between physical stimuli and logical operations, meanwhile avoiding any spontaneous unitary evolution of the qubit states. Current experimental efforts are ongoing to implement an extension of the proposed protocol.

**Acknowledgements**

This work is supported by the European Commission (FET-OPEN project FATMOLS (No. 862893)); the Spanish MICINN (grant CTQ2017-89993 and PGC2018-099568-B-I00 co-financed by FEDER, grant MAT2017-89528 and the Unit of excellence "María de Maeztu" CEX2019-000919-M); and the Generalitat Valenciana (CIDEGENT/2021/018 and PROMETEO/2019/066). J.A. is indebted to the MICINN for his "Ramón y Cajal" fellowship (RyC-2017-23500).

**References**

[1] M. Shiddiq, et al. *Nature* (2016) **531**, 348.
[2] J. Liu, et al. *Nat. Phys.* (2021) **17**, 1205.
[3] A. Ullah, et al. *npj quant. inf.* (2022) **8**, 133

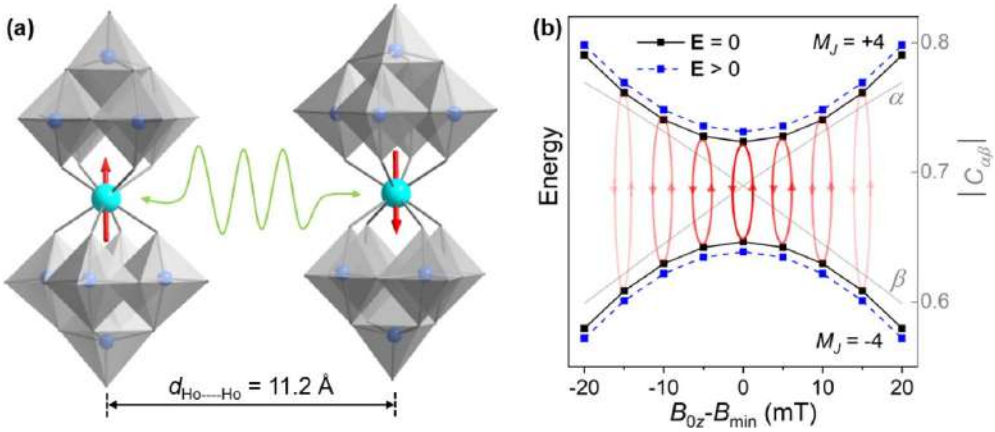



Figure: Nearest inversion-related HoW10 pair within the crystal, illustrating their dipolar interaction. Calculated energies (left axis) and mixing degree (right axis) of the eigenstates in terms of the $|MJ=\pm4>$ spin states of $HoW_{10}$ as a function of magnetic and electric fields.

# The Coupling Phase in Microwave Cavity Magnonics

*Alan Gardin[1,2,3]*, Jeremy Bourhill[2,4], Vincent Vlaminck[2,3], Christian Person[2,3], Christophe Fumeaux[5], Vincent Castel[2,3], Giuseppe Tettamanzi[1,6]*

[1] *School of Physics, The University of Adelaide, Adelaide SA 5005, Australia*
[2] *IMT Atlantique, Technopole Brest-Iroise, CS 83818, 29238 Brest Cedex 3, France*
[3] *Lab-STICC (UMR 6285), CNRS, Technopole Brest-Iroise, CS 83818, 29238 Brest Cedex 3, France*
[4] *ARC Centre of Excellence for Engineered Quantum Systems and ARC Centre of Excellence for Dark Matter Particle Physics, Department of Physics, University of Western Australia, 35 Stirling Highway, Crawley, Western Australia 6009, Australia*
[5] *School of Electrical and Electronic Engineering, The University of Adelaide, Adelaide SA 5005, Australia*
[6] *School of Chemical Engineering and Advanced Materials, The University of Adelaide, Adelaide SA 5005, Australia*
*alan.gardin@adelaide.edu.au

The coherent interaction between microwave photons and magnons is well understood and originates from the Zeeman coupling between spins and a magnetic field [1-2]. Interestingly, the magnon/photon interaction is accompanied by a phase factor which can usually be neglected [3]. However, under the rotating wave approximation, if two magnon modes simultaneously couple with two cavity resonances, this phase cannot be ignored as it changes the physics of the system. We consider two such systems, each differing by the sign of one of the magnon/photon coupling strengths. This simple difference, originating from the various coupling phases in the system, is shown to preserve, or destroy, two potential applications of hybrid photon/magnon systems, namely dark mode memories [4] and cavity-mediated coupling [5]. The observable consequences of the coupling phase in this system is akin to the manifestation of a discrete Pancharatnam–Berry phase [6], which may be useful for quantum information processing.

## Acknowledgements

We acknowledge financial support from Thales Australia and Thales Research and Technology.

## References

[1] B. Z. Rameshti, et al., Cavity magnonics, Physics Reports, Vol 979, Pages 1-61 (2022)
[2] Bourhill, Castel, Manchec, and Cochet, Universal characterization of cavity–magnon polariton coupling strength verified in modifiable microwave cavity, Journal of Applied Physics 128, 073904.
[3] Flower, Goryachev, Bourhill, and Tobar, Experimental implementations of cavity-magnon systems: from ultra strong coupling to applications in precision measurement, New Journal of Physics 21, 095004.
[4] Zhang et al., Magnon dark modes and gradient memory, Nature Communications 6, 10.1038/ncomms9914
[5] Grigoryan et al., Cavity-mediated dissipative spin-spin coupling, Physical Review B 100, 014415 (2019)
[6] Resta, Manifestations of berry's phase in molecules and condensed matter, Journal of Physics: Condensed Matter 12, R107 (2000).

# Strong to Ultra-Strong Coupling in a YIG/Cavity System

*Guillaume Bourcin[1,2]*, Jeremy Bourhill[3], Vincent Vlaminck[1,2], and Vincent Castel[1,2]*

[1] *IMT Atlantique, Technopole Brest-Iroise, CS 83818, 29238 Brest Cedex 3, France*

[2] *Lab-STICC (UMR 6285), CNRS, Technopole Brest-Iroise, CS 83818, 29238 Brest Cedex 3, France*

[3] *ARC Centre of Excellence for Engineered Quantum Systems and ARC Centre of Excellence for Dark Matter Particle Physics, University of Western Australia, 35 Stirling Highway, Australia*

*guillaume.bourcin@imt-atlantique.fr

Over the last decade, quantum systems offering new computational and sensing capabilities have emerged [1]. One of these promising hybrid systems involves the interaction between photons and magnons. This interaction is quantitatively known by the strength coupling g and furthermore by its ratio with the cavity frequency g/ω. It exists three different domains of this ratio: the Strong Coupling (SC) for $g/\omega < 0.1$; the Ultra-Strong Coupling (USC) for $0.1 < g/\omega < 1$; and the Deep-Strong Coupling (DSC) for $g/\omega > 1$. One of the objectives of this last decade is to achieve the USC, and to approach the DSC [2].

Here, we present an experimental study of the SC to USC regimes at room temperature (RT) in frequency-reconfigurable 3D re-entrant cavities coupled with a YIG slab, as show in Fig. 1(a). The observed coupling rate, defined as the ratio of the coupling strength (g) to the cavity frequency of interest ($\omega/2\pi$), ranges from 12% to 59%. One of the measurements is presented in Fig. 1(b), where is shown the S parameter according to a sweep on RF frequencies and the static H-field. We show that certain considerations must be considered when analyzing the polaritonic branches of a cavity spintronic device where the RF field is highly focused in the magnetic material. Our observations are fully confirmed by electromagnetic finite element simulations in the frequency domain.

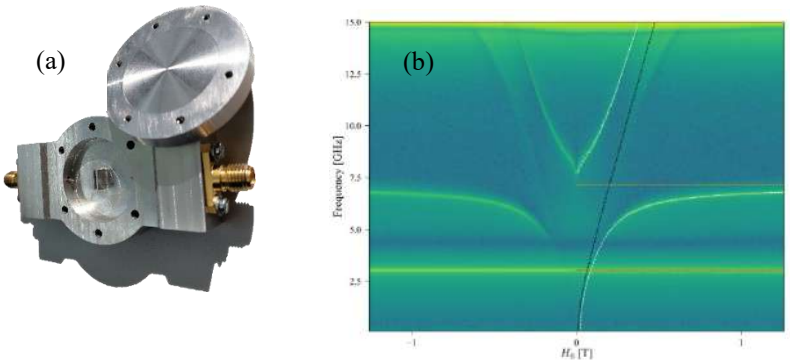



Figure1: (a) Double re-entrant, and (b) experimental transmission spectra versus frequency and applied magnetic field. Measurement at RT

We prove the ability of a double re-entrant cavity to reach the USC regime at RT and its tuneability to radically increase the coupling rate. The validation of the FEM model (including the FMR expression for a millimeter slab YIG sample) in the frequency domain allowed us to highlight the importance of the form factor on the understanding of CMPs in cavity spintronics.

## Acknowledgements

This work was funded by Brest Métropole through the CPER Project SOPHIE/STIC & Ondes, by the CPER SpaceTechDroneTech

## References

[1] D. Lachance-Quirion, Y. Tabuchi, A. Gloppe, *Appl. Phys. Exp.*, vol. **12** (2019), p. 070101.

[2] I.A. Golovchanskiy, N.N. Abramov, V.S. Stolyarov, *Phys. Rev. Applied.*, vol. **16** (2021), p. 034029.

# Scanning NV magnetometry of focused-electron-beam-grown cobalt micromagnets for spin qubit control

*Liza Žaper[1,2] *, Peter Rickhaus[2], Boris Gross[1], Marcus Wyss[3], Martino Poggio[1], and Floris Braakman[1]*
*[1]Department of Physics, University of Basel, Basel, Switzerland*
*[2]Qnami AG, Muttenz, Switzerland*
*[3]Swiss Nanoscience Institute, University of Basel, Basel, Switzerland*
* liza.zaper@unibas.ch, liza.zaper@qnami.ch

We fabricate highly magnetic cobalt nanostructures patterned in a single-step process using focused-electron-beam-induced deposition. We image the magnetic stray field of the micromagnets using scanning NV magnetometry and find good agreement with micromagnetic simulations and the TEM analysis of the structure. Our scanning probe measurements indicate the structure of the magnetic domains, extended halo particles, and the profile of the magnetic stray field. Furthermore, we estimate the effect of the disordered halo stray field on spin qubit decoherence. These measurements guide the use of single-step micromagnet patterning for future spin qubit devices.[1]

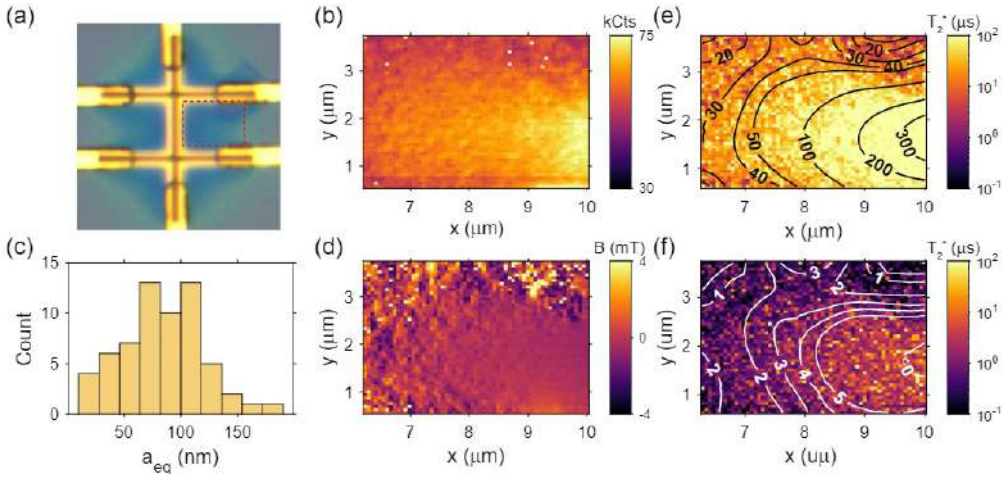



Figure 1. a) Optical micrograph of Co Hall bar, with halo distinguishable as dark shape. (b) Scanning NV fluorescence map of area outlined by red box in (a). (c) Histogram of equivalent square side of grains in SNVM data. (d) SNVM map of area outlined by red box in (a). (e, f) Maps of estimated T ∗2 (x, y) for spin qubit displacement amplitude of 10 pm and 1 nm, corresponding to displacement due to charge noise (e) and EDSR driving (f), respectively.

**References:**

[1] L. Žaper[1,2], P. Rickhaus[2], B. Gross[1], M. Wyss[3], M. Poggio[1], and F. Braakman[1], Scanning NV magnetometry of focused-electron-beam-grown cobalt micromagnets for spin qubit control, *publication in preparation*

# SYMPOSIUM 05.
## NEUROMORPHIC AND RESERVOIR COMPUTING.
## S5. INVITED ORAL PRESENTATIONS

# Pattern Recognition Using Nonlinear Magnons

*K. Schultheiss*[1]*

[1] *Helmholtz-Zentrum Dresden-Rossendorf, Dresden, Germany*

*k.schultheiss@hzdr.de

Within the last decade, spintronics and magnonics have demonstrated an impressive development in the experimental realization of Boolean logic gates. However, the exponential growth of data and the rise of the internet of things are pushing the deterministic Boolean computing of von-Neumann architectures to their limits or simply consume too much energy. Moreover, conventional Boolean computer architectures are likely to remain inefficient for certain cognitive tasks in which the human brain excels, such as pattern recognition, particularly when incomplete or noisy data are involved.

One of the most generic and abstract implementations of brain-inspired computing schemes is reservoir computing, which uses the nonlinearity and recurrence of a physical system to separate patterns of time series data into distinct manifolds of a higher dimensional output space. In this presentation, I will demonstrate the experimental realization of pattern recognition based on reservoir computing using magnons.

Recently, we reported on the nonlinear scattering of magnons in vortices in micron-sized NiFe discs [1] which we learned to control and stimulate by means of other magnons [2]. Now, we utilize these phenomena to employ magnons for pattern recognition without relying on magnon transport in real space [3]. I will present a comprehensive overview of experimental results and numerical simulations demonstrating the capabilities and advantages of magnon reservoir computing in reciprocal space. Additionally, I will elaborate on the potential of modifying nonlinear magnon scattering by the distortion of the magnetic vortex using static [4] and dynmic in-plane magnetic fields.

## Acknowledgements

The author acknowledges funding from the EU Research and Innovation Programme Horizon Europe under grant agreement No 101070290.

## References

[1] K. Schultheiss, R. Verba, F. Whermann, K. Wagner, L. Körber, T. Hula, T. Hache, A. Kákay, A. A. Awad, V. Tiberkevich, A. N. Slavin, J. Fassbender, and H. Schutlheiss, *Physical Review Letters* **122**, 097202 (2019).

[2] L. Körber, K. Schultheiss, T. Hula, R. Verba, J. Fassbender, A. Kákay, and H. Schutlheiss, *Physical Review Letters* **125**, 207203 (2020).

[3] L. Körber, C. Heins, T. Hula, J.-V. Kim, H. Schultheiss, J. Fassbender, K. Schultheiss, *arXiv* 2211.02328 (2022).

[4] L. Körber, C. Heins, I. Soldatov, R. Schäfer, A. Kákay, H. Schultheiss, and K. Schultheiss, Appl. Phys. Lett. 122, 092401 (2023).

# Hybrid Ferromagnetic And Superconducting Devices For Neuromorphic Computing

*Emilie Jué*[1, 2,*]

[1]*Associate of the National Institute of Standards and Technology, Boulder, Colorado 80305, USA*
[2] *Department of Physics, University of Colorado Boulder, Boulder, Colorado 80309, USA*
*emilie.jue@nist.gov

The performance of artificial intelligence (AI) technologies has improved significantly over the last decade in such a way that AI is now everywhere in our daily life via software neural networks. However, this continual growth in computational performance of these networks comes with large increases in the computational time and energy needed to train them. Developing AI at the hardware level has the potential to bend this curve and provide fast and lower energy computing. In this talk, I will present a new hybrid magnetic-superconducting device that can be used as an artificial synapse in neuromorphic circuits [1,2]. The device is a nanoclustered magnetic Josephson Junction (nMJJ) that consists of a barrier of magnetic nanoclusters between two Nb electrodes. The critical current of these junctions can be tuned by varying the magnetic order of the clusters, which can be used to perform synaptic weighting. I will describe the properties of the nMJJ and show that its synaptic properties can be obtained in different material systems [3] with an energy cost as low as $10^{-19}$J. Finally, I will present circuit simulations where MJJs are included in a neural network for image recognition operating at speeds over 100 GHz, and show some preliminary experimental validation of the simulations.

## References

[1] M. L. Schneider et al., "Ultralow power artificial synapses using nanotextured magnetic Josephson junctions," *Sci Adv*, vol. **4**, no. 1, p. e1701329, 2018.

[2] E. Jué, M. R. Pufall, I. W. Haygood, W. H. Rippard, and M. L. Schneider, "Perspectives on nanoclustered magnetic Josephson junctions as artificial synapses," *Appl Phys Lett*, vol. **121**, no. 24, Dec. 2022.

[3] E. Jué et al., "Artificial synapses based on Josephson junctions with Fe nanoclusters in the amorphous Ge barrier," *J Appl Phys*, vol. **131**, no. 7, p. 073902, Feb. 2022.

# SYMPOSIUM 05.
## NEUROMORPHIC AND RESERVOIR COMPUTING.
## S5. ORAL PRESENTATIONS

# Spintronic Extreme Learning Reservoirs

***Ian Vidamour*[1*], *Luca Manneschi*[1], *Matthew O A Ellis*[1], *Charles Swindells*[2], *Guru Venkat*[2], *Paul W Fry*[3], *Thomas J Hayward*[2], *and Eleni Vasilaki*[1]**

[1] *Department of Computer Science, The University of Sheffield, S1 4DP, UK*
[2] *Department of Materials Science and Engineering, The University of Sheffield, S1 3JD, UK*
[3]*Nanoscience and Technology Centre, the University of Sheffield, S3 7HQ, UK*
*Corresponding author - i.vidamour@sheffield.ac.uk

Physical computing is an exciting paradigm where the complex behaviours of material systems are exploited to perform computation. Spintronic platforms are especially promising due to their inherent nonlinear and hysteretic behaviours, proven nanofabrication routes, and ability to interface with existing electronic platforms. Many spintronic computational systems have been proposed under the reservoir computing (RC) paradigm [1-4], where input/reservoir connectivity weights are randomly generated and untrained, and the complexity of the underlying dynamic system provides the computational power [5]. However, experimental implementations often rely upon time-multiplexing techniques to construct networks of 'virtual' nodes from a single dynamical node. This limits both the throughput of data as well as the computational properties of the platform, with external delay lines often necessary to augment the system's dynamics to generate memory.

Another computational paradigm which similarly uses random input connectivities is the Extreme Learning Machine (ELM) [6]. In this paradigm, the network consists of a single hidden layer of unconnected neurons, and the outputs are trained via linear regression techniques similarly to RC. However, this technique is usually limited to time-invariant problems and has not seen widespread deployment for *in-materia* computing. In our work, we show how this paradigm can be extended to time-signal processing tasks with spintronic neurons (here, interconnected magnetic nanoring arrays), forming Extreme Learning Reservoirs (ELRs). We demonstrate the performance of in-memory computation without the need for connections between physical neurons- vastly improving the experimental viability of multi-neuron physical reservoirs. Using a neural-ODE model [7,8] to simulate the nanoring array's response, we show how computational capability can be maximised by ensuring the random input weights are appropriately scaled to drive the system into different dynamic regimes, then verify the results of these simulations with experiments in hardware. Finally, we show how the computational performance can be further enhanced by generating feed-forward networks of these ELRs with inter-layer connectivity, forming deep-ELRs capable of solving time series prediction tasks, surpassing the accuracies of other spintronic computing platforms (NRMSE 0.203 vs 0.367 [2]).

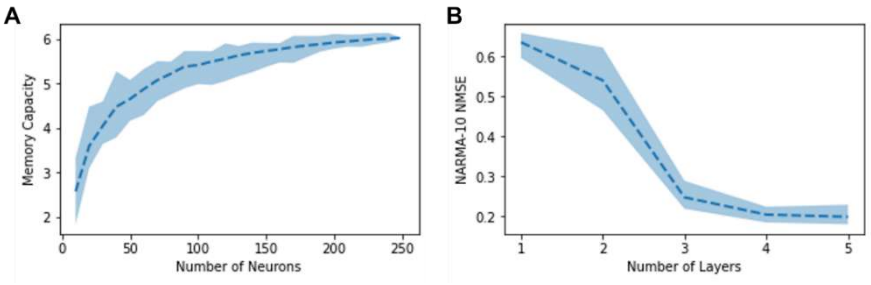



Figure: Figure 1-(a) Memory capacity vs number of neurons for single-layer ELR consisting of magnetic nanoring neurons. Dotted line shows the mean performance over 10 shuffles of randomly selected neurons, shaded region reflects minimum/maximum performance. (b) Normalised root-mean-squared error for NARMA-10 system modelling task versus number of layers, for layer width = 50 neurons. Dotted line shows mean performance over 10 repeats, shaded region reflects minimum/maximum performance.

## References

[1] J. Torrejon et al. Nature **547** (2017)
[2] I. Vidamour et al. (preprint at https://arxiv.org/abs/2206.04446)
[3] J. C. Gartside et al. Nature Nanotechnology **17** (2022)
[4] R.V. Ababei et al. Scientific Reports **11** (2021)
[5] H. Jaeger. GMD Technical report **148** (2001)
[6] G.B. Huang et al. Neurocomputing **70** (2006)
[7] R.T.Q. Chen et al. NeurIPS **31** (2018)
[8] X. Chen et al. Nature Communications **13** (2022)

# Multi-input Channel Skyrmion-Based Reservoir Computing

*Robin Msiska[1,3*], Jake Love[1], Jeroen Mulkers[2], Jonathan Leliaert[2], and Karin Everschor-Sitte[1,3]*
*[1]Faculty of Physics, University of Duisburg-Essen, 47057 Duisburg, Germany,*
*[2]Department of Solid State Sciences, Ghent University, 9000 Ghent, Belgium,*
*[3]Center for Nanointegration Duisburg-Essen (CENIDE), 47057 Duisburg, Germany*
*robin.msiska@uni-due.de

Reservoir computing (RC) is a type of computing framework based on recurrent neural networks that capitalizes on the inherent dynamics of complex systems. In RC, signals from a fixed input are introduced to a reservoir that is intentionally constructed to possess high-dimensional nonlinear dynamics. This reservoir then transforms the input patterns into a high-dimensional, linearly separable feature space, allowing for easy training of an output using uncomplicated techniques like linear regression. Physical reservoir computing (PRC) is a version of reservoir computing that utilizes a physical system as the reservoir rather than software-based implementation. PRCs leverage the unique properties of the physical system to perform computations in a way that would not be possible by conventional means.

We propose a multi-channel input skyrmion RC model made up of a magnetic thin-film hosting skyrmions in a random configuration, capable of nanosecond timescale pattern recognition. In contrast to single input channel RCs, multi-channel input systems provide the advantage of lower error rates and better power efficiency [1]. We demonstrate the outstanding multi-dimensional classification capabilities of the reservoir by subjecting it to a standard benchmark audio recognition test, where we classify isolated spoken English digits (from 0 to 9). During this process, audio samples are first converted into voltage signals with acoustic frequency bands as their parameters before being sent as inputs into the reservoir. The outputs states of the reservoir are then used to create a linear classification model, which can identify every spoken digit regardless of the speaker with an overall accuracy of 97.4%. The entire procedure is summarized in the figure below.

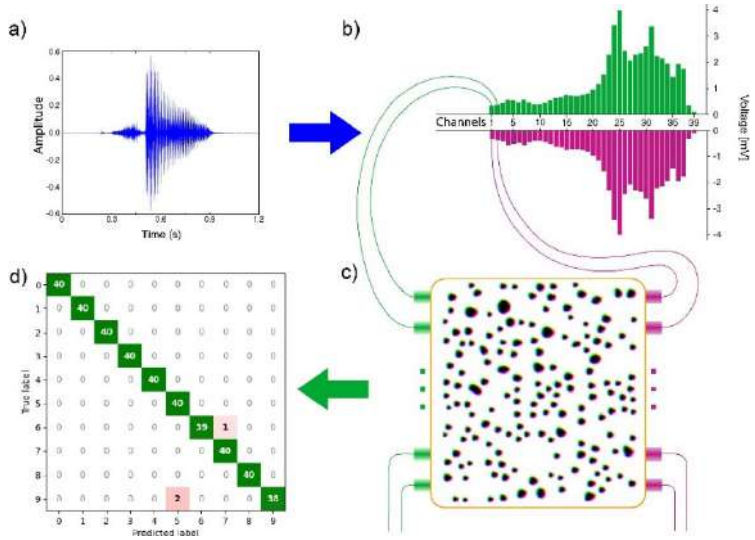



Figure: Skyrmion-based reservoir computing scheme used for audio recognition. (a) Audio waveforms are converted to (b) voltage signals which in turn are used as inputs for (c) a multi-input skyrmion reservoir. (d) Reservoir output is trained to classify the audio sample [2].

## Acknowledgements

We acknowledge funding from the Emergent AI Center funded by the Carl-Zeiss-Stiftung and the Deutsche Forschungsgemeinschaft Project No. 320163632. Jo.L. was supported by the Fonds Wetenschappelijk Onderzoek (FWO) with senior postdoctoral fellowship No12W7622N. Some computational resources were provided by the Flemish Supercomputer Center, funded by Ghent University, FWO and the Flemish Government – department EWI.

## References

[1] A. Katumba et al., *Cogn. Comput.* **9** (2017) 307.
[2] R. Msiska et al., *Adv. Intell. Syst.* (2023) in press.

# Spin Waves and Spin Orbit Torques: Efficient Interfacing With Magnetic Metamaterials For Reservoir Computing

*C. Swindells[1]*, I.T. Vidamour[1,2], G. Venkat[1], T.J. Hayward[1]*

[1] *Department of Materials Science and Engineering, University of Sheffield, UK*
[2] *Department of Computer Science, University of Sheffield*
*c.swindells@sheffield.ac.uk

Traditional neural networks based on CMOS architecture are notoriously inefficient, with substantial energy costs in both training weights between nodes and shuffling data between memory and processing units. Reservoir computing (RC) offers one potential solution by replacing conventional recurrent networks with a reservoir, composed of a dynamic system, where only output layers are trained. Additional efficiency is obtained moving from a von-Neuman system simulating a dynamical system to using material systems whose physics inherently provide the required properties. Two broad requirements for a material's suitability for RC are non-linear responses to input stimuli, and a 'fading memory', where the current state is dependent upon past states. Magnetic materials satisfy both of these criteria, making them ideally suited for in materio RC.

Several magnetic systems have been proposed for RC, notably spin-oscillators [1] and artificial spin-ice systems [2], each with their own benefits and drawbacks. Recently, the domain wall population of an array of interconnected magnetic rings has been observed to be an emergent property and proposed to be a possible vehicle for performing RC [3]. We have shown the capability of such an ensemble at performing a range of benchmark tasks [4] and reservoir configurations. However, a common problem with magnetic RC platforms are inefficient methods of data input/output. Typically, magnetic states require sufficient external magnetic driving fields to alter their configurations as input, while measuring the states of complex metamaterial systems in sufficient detail for a meaningful output is not trivial.

Here, we present the results of a series of studies which show the promise of the ring system as an on-chip RC platform through efficient interfacing. First, we show how the microwave spectra of ring systems provides insight into the internal states present in a uniform ensemble of interconnected permalloy rings (see figure), and how this information can be utilised as an output for RC. We then show how heterogeneous ring array systems with structural variations can fill this measurable space, providing a rich set of outputs to train on. Finally, we show how data input based on external magnetic driving fields can be replaced via spin orbit torques, through careful design of a Pt interface. The stochastic nature of domain walls at junctions allows for a large state space as a function of inputs in both the polar field and current amplitude applied (see figure), with the observed dynamics analogous to those measured with an external driving field. Together, these results illustrate the possibilities of creating powerful and efficient reservoir computers based on magnetic metamaterials.

## References

[1] D.Marković et al., *Appl. Phys. Lett.* **114** (2019) 012409.
[2] J.C.Gartside et al., *Nat. Nano.* **17** (2022) 460.
[3] R.W.Dawidek et al., *Adv. Funct. Mater.* **31** (2021) 2008389.
[4] I.T.Vidamour et al., *Nanotechnology* **33** (2022) 485203.

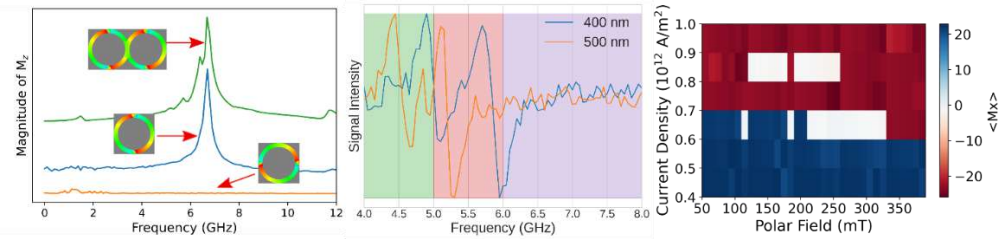



Figure: Simulated microwave spectra of single and bi-rings in onion states using Mumax3 (left). Measured microwave spectra of two arrays of different ring widths, used as an output for reservoir computing (middle). Average x-component of magnetisation in a simulated bi-ring / Pt layered system as a function of polar field and current density, for a fixed current direction (right).

# Task-Adaptive Physical Reservoir Computing Using Magnetic Skyrmions

*Oscar Lee[1*], Tianyi Wei[1], Kilian D. Stenning[2], Jack C. Gartside[2], Shinichiro Seki[3], Aisha Aqeel[4,5], Christian Back[4], Yoshinori Tokura[3,6,7], Will R. Branford[2,8], Hidekazu Kurebayashi[1,9,**]*

[1]*London Centre for Nanotechnology, University College London, London, United Kingdom*
[2]*Blackett Laboratory, Imperial College London, London, United Kingdom*
[3]*Department of Applied Physics, University of Tokyo, Tokyo Japan*
[4]*Physik-Department, Technische Universitat Munchen, Garching, Germany*
[5]*Munich Center for Quantum Science and Technology (MCQST), Munich, Germany*
[6]*RIKEN Center for Emergent Matter Science (CEMS), Wako, Japan*
[7]*Tokyo College, University of Tokyo, Tokyo, Japan*
[8]*London Centre for Nanotechnology, Imperial College London, London, United Kingdom*
[9]*Department of Electronic and Electrical Engineering, University College London, United Kingdom*
*zceesjl@ucl.ac.uk **h.kurebayashi@ucl.ac.uk

Physical reservoir computing (PRC) is a neuromorphic architecture potentially offering energy-efficient solutions for various machine learning (ML) tasks [1]. However, due to the rigidity of configuring the crucial hyperparameters required to maximise computational performance, physical reservoirs are typically constrained to execute a set of particular ML tasks. In this talk, we experimentally demonstrate a flexible task-adaptive PRC using the spectrum space of a single magnetic system with distinctive phase properties [2]. The reservoir is constructed with data-mapped collective spinwave excitations (Fig 1. a-b) of skyrmion and conical modes. We scrutinise the task-adaptive nature via trivial magnetic phase control in a chiral magnetic insulator $Cu_2OSeO_3$ as a *model* system and bridge the key reservoir properties with various magnetic phases. Our results highlight that skyrmions excel in forecasting chaotic signals, unlike the conical modes that are optimal for nonlinear transformative tasks with MSEs in orders of $10^{-3}$ and $10^{-7}$, respectively. Room-temperature demonstrations on FeGe and $Co_{8.5}Zn_{8.5}Mn_{8.5}$ confirms that our task-adaptive approach to PRC via magnetic phase control is transferable to other phase-rich systems, taking a step closer to energy-efficient computing.

**Acknowledgements**

O.L. and H.K. thank the Leverhulme Trust for financial support via RPG-2016-391.

**References**

[1] Nakajima, K. & Fischer, I. "Reservoir Computing" (Springer, 2021).
[2] Lee, O. et al. "Task-adaptive physical reservoir computing", arXiv preprint, arXiv: 2209.06962 (2022).

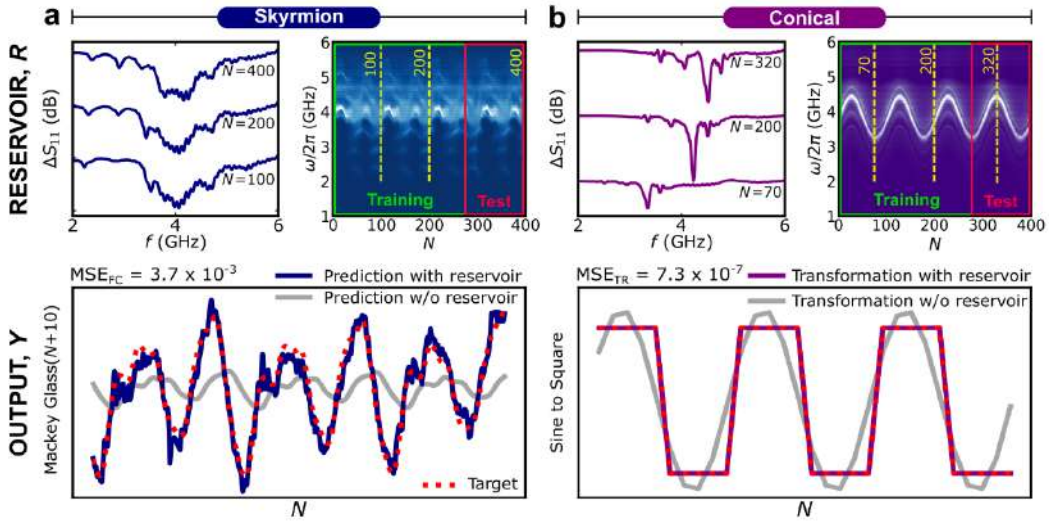



Figure 1. Visualisation of magnetic-phase reservoirs built with spectral spaces of (a) skyrmion and (b) conical modes incorporating a sinewave input. Skyrmion modes excel in future forecasting requiring high memory, whereas conical modes show excellent signal transformation performance requiring high nonlinearity.

# Global biasing using a Hardware-based artificial Zeeman term in Spinwave Ising Machines

*Victor H. González[1]*, Artem Litvinenko[1], Roman Khymyn[1], Johan Åkerman[1,2]*

[1] *University of Gothenburg, Origovägen 6B, 412 96 Gothenburg, Sweden*

[2] *Tohoku University, 2-1-1 Katahira, Aoba-ku, Sendai 980-8577, Japan*

*victor.gonzalez@physics.gu.se

A spinwave Ising machine (SWIM) [1] is a newly proposed type of time-multiplexed hardware solver for combinatorial optimization that employs feedback coupling and phase sensitive amplification to map an Ising Hamiltonian into phase-binarized propagating spin-wave RF pulses in an Yttrium-Iron-Garnet (YIG) film. In this work, we increase the mathematical complexity of the SWIM by adding a global Zeeman term to a 4-spin MAX-CUT Hamiltonian using a continuous external electrical signal with the same frequency as the spin pulses and phase locked with one of the two possible states.

For large field amplitudes, we are able to induce ferromagnetic ordering in both directions of the artificial spin phase space despite antiferromagnetic pairwise coupling and observe degeneracy in the solutions for intermediate amplitudes in the form of 3+1 spin states. These states arise due to non-linearity on the power transfer curve of the phase sensitive amplifier in the circuit that induces a shortening of the effective spin amplitude for heterogeneous electrical phases. We show that these suboptimal solutions can be supressed using digital feedback with a field programmable gate array (FPGA) or designing a custom amplifier with a smoother saturation curve. By introducing a magnetic field to a planar antiferromagnetic spin system, we can leverage its increased complexity [2] to unlock new degrees of computational power and explore the boundaries of spinwave-based hardware solvers.

**Acknowledgements**

This work has been conducted and presented with support from the University of Gothenburg and the European Research Council's Advanced Grant TOPSPIN.

**References**

[1] A. Litvinenko, R. Khymyn, V. H. González, A. A. Awad, V. Tyberkevych, A. Slavin, J. Åkerman, arXiv:2209.04291 [cond-mat.mes-hall] (2022)

[2] F. Barahona 1982 J. Phys. A: Math. Gen. 15 3241.

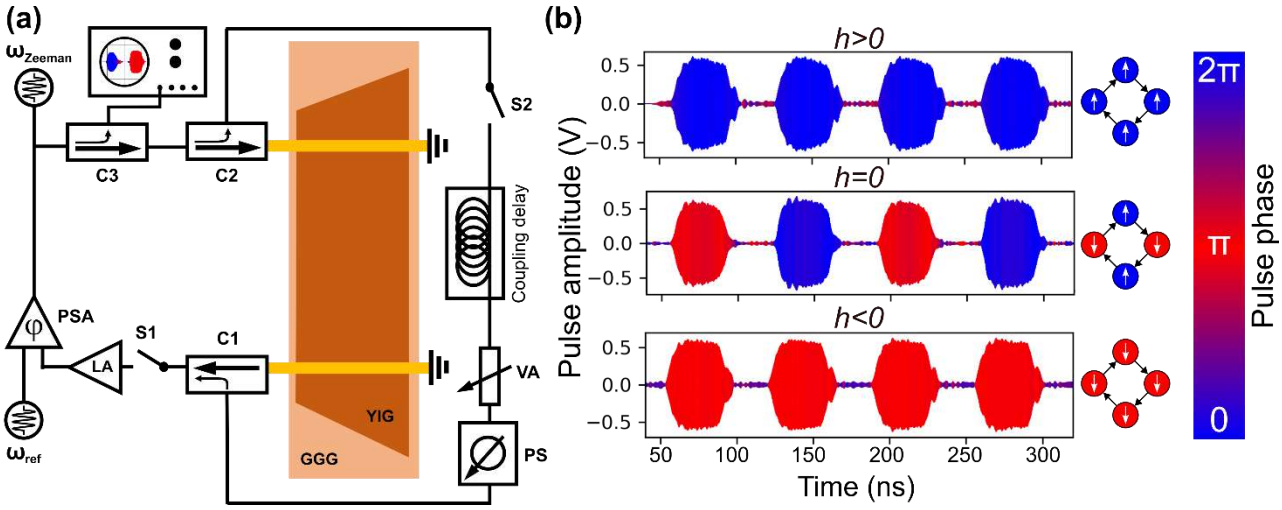



Figure: **(a)** Zeeman-biased spinwave Ising machine. PSA and LA stand for phase-sensitive and linear amplifiers, respectively. The propagating RF pulses have frequency of 3.125 GHz. The sign of the coupling is controlled by the total phase accumulation in the coupling delay 1 (C1) and the Zeeman field amplitude and sign is controlled by the amplitude and phase of the injected signal $\omega_{Zeeman}$. **(b)** Time traces of the RF pulses colored with their respective instantaneous phase for different signs of $h$.

# Helitronics for classical and unconventional computing

*Nicolai T. Bechler[1], Jan Masell[1,2]**

*[1]Institute for Theoretical Solid State Physics, Karlsruhe Institute of Technology (KIT), Karlsruhe, 76049, Germany*
*[2]Center of Emergent Matter Science (CEMS), RIKEN, Wako, 351-0198, Japan*
** jan.masell@kit.edu*

Magnetic textures are promising candidates for unconventional computing due to their non-linear dynamics. We propose to investigate the rich variety of seemingly trivial lamellar magnetic phases, e.g., helical, spiral, stripy phase, or other one-dimensional soliton lattices. These are the natural stray field-free ground states of almost every magnet. These states which can be viewed as a labyrinth of one-dimensional topological solitons, are coming back into the focus of spintronic research due to their unique symmetry. They have been shown to work as emergent inductors [1], lead to non-reciprocal charge and spin transport [2], or may actively pump spin and charge as Archimedean screws [3]. Moreover, they can serve as a non-trivial background for other topological excitations such as skyrmions or dislocations [4]. Only recently, it has been shown that the orientation vector q of this lamellar phase can be combed and steered by external magnetic fields and even electric currents [5].

We argue that the order parameters of these phases may be of potential interest for both classical and unconventional computing, which we refer to as *helitronics*. For the particular case of a chiral magnet and its helical phase, we use micromagnetic simulations to demonstrate the working principles of all-electrical (i) classical binary memory cells and (ii) memristor and neuron cells, based on the orientation of the helical stripes.

## References

[1] T. Yokouchi, F. Kagawa, M. Hirschberger, *et al. Nature* **586** (2020) 232–236.
[2] G. L. J. A. Rikken, J. Fölling, P. Wyder, *Phys. Rev. Lett.* **87** (2001) 236602.
[3] D. Kurebayashi, Y. Liu, J. Masell, N. Nagaosa, *Phys. Rev. B* **106** (2020) 205110.
[4] P. Schoenherr, J. Müller, L. Köhler, *et al. Nat. Phys.* **14** (2018) 465–468.
[5] J. Masell, X. Z. Yu, N. Kanazawa, *et al. Phys. Rev. B* **102** (2020) 180402.


Figure: Example of two distinct helitronic states 0 and 1 in a finite size system. Additionally, the transient state during switching is shown. Result of a simulation with spin transfer-torque. Color indicates the direction of the magnetization. Small gray arrows indicate the inplane component of the magnetization. The surrounding gray background is vacuum.

# Fully Parallel Spintronic Convolutional Layer with Frequency Interconnectivity

*Erwan Plouet[1]*, Dédalo Sanz Hernández[1], Pankaj Sethi[1], Nathan Leroux[1], Bruno Dlubak[1], Victor Zatko[1], Alice Mizrahi[1], Julie Grollier[1]*

[1] *Unité Mixte de Physique CNRS Thales, Université Paris-Saclay, Palaiseau, France*

*erwan.plouet@cnrs-thales.fr

Convolutional neural networks (CNNs) are state of the art algorithms for image processing. Despite a small number of synaptic weights, CNNs remain computationally costly to train in software due to the sequential application of the convolutional filter over the input. The field of neuromorphic spintronics offers the possibility of new promising parallel architectures that would perform a convolution in a single time step. Previously it has been demonstrated that the spin diode effect can be used to selectively apply a synaptic weight on a radiofrequency signal using frequency selectivity to address each device independently [1][2]. Here we go a step beyond, to show an experimental implementation of a convolutional layer able to calculate the result of a complete convolution in a single time step by exploiting the frequency domain as an additional dimension.

We design a compact architecture of 3 radio-frequency waveguides integrating each 3 spin-diodes connected in series. This architecture performs a padded convolution between a 3-pixels filter and a 5-pixels input. Inputs are represented by RF signals and are selectively processed by spin diodes at the matching frequency as represented by colors in Figure 1, the values of the filter are encoded by a small frequency detuning between the inputs and the spin-diodes resonance. Frequency selectivity enables us to produce each output with a single line geometry. Three strip lines in a crossbar configuration are implemented to write simultaneously the three weight values shared by the RF waveguides. This architecture exploits the intrinsic weight redundancy of convolutions to compute three outputs in parallel, instead of sequentially.

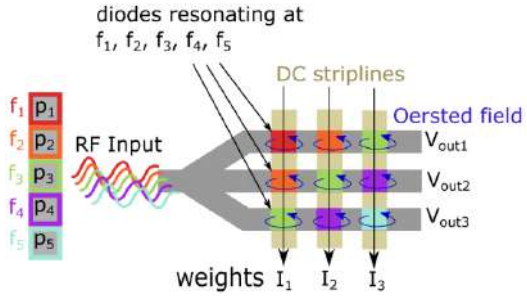



Figure 1 RF convolutional architecture

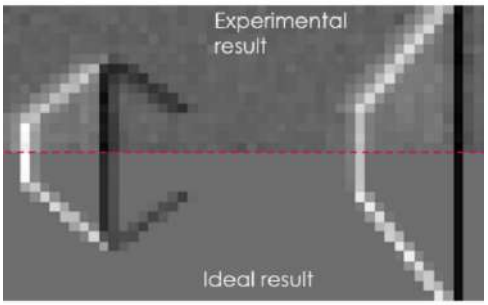



Figure 2 Convolution performed to enhance vertical edges

The proposed architecture enables us to both reduce the size and scaling of this hardware implementation while, at the same time, performing the convolution in one timestep contrary to previous time-multiplexed implementations. A potential decrease by one order of magnitude in energy consumption compared to current GPUs and two orders of magnitude in operating latency is envisioned upon scaling down of the technology [3]. This proof of concept of a spintronic CNN presented here, opens the path to the development of deep spintronic neural networks that can exploit the power of convolutional layers in a fully parallel, compact, and energy efficient way.

## Acknowledgements

This work was supported by the European Union's grant RadioSpin No 101017098.

## References

[1] Radio-Frequency Multiply-And-Accumulate Operations with Spintronic Synapses, Nathan Leroux *et al* *2021* Phys. Rev. Applied 15, 034067

[2] Hardware realization of the multiply and accumulate operation on radio-frequency signals with magnetic tunnel junctions, Nathan Leroux *et al* 2021 *Neuromorph. Comput. Eng.* 1 011001

[3] Convolutional neural networks with radio-frequency spintronic nano-devices, Nathan Leroux *et al* 2022 *Neuromorph. Comput. Eng.* 2 034002

# Neuromorphic Spintronics or How to Do More With Less!

*Flavio ABREU ARAUJO[1]*, Simon de WERGIFOSSE[1], Chloé CHOPIN[1], and Anatole MOUREAUX[1]*

[1] *Université catholique de Louvain, Place Croix du Sud 1, Louvain-la-Neuve, Belgium*

*flavio.abreuaraujo@uclouvain.be

Over the past decades, artificial intelligence (AI) has made significant technological advances with the prospect of increased computer capabilities (*e.g.*, automation in decision making and data processing) and acquired an increasingly important role in our everyday technological environment (Dall-E, Midjourney, ChatGPT, *etc.*). The main issue is that the digital silicon-based computing technologies are very energy-intensive while solving cognitive tasks such as speech or image recognition. Indeed, digital computers are designed to solve very complex numerical problems with high precision whereas the human brain performs many low-precision calculations in parallel while solving cognitive tasks like recognizing a familiar face.

In the framework of neuromorphic spintronics [1], we combine condensed matter physics and artificial intelligence to design nanoscale neuromorphic computing hardware. The physical building block considered in our research program to implement such bio-inspired hardware is the magnetic tunnel junction (MTJ). MTJs are made of two ferromagnetic (FM) layers separated by a non-magnetic insulating barrier (often MgO, see Fig. 1a). One of the FM layers is the spin polarizer and the other the free layer. At the nanoscale, the aspect ratio of the MTJs is such that the magnetic ground state of the free layer is the magnetic vortex. The vortex is a curling in-plane magnetization distribution with at its core a singularity where the magnetization is pointing out-of-plane. Here, we show how that kind of oscillators, the spin-torque vortex oscillators, can act as nano-neurons and nano-synapses for solving machine learning tasks using the physical reservoir computing (PRC) neural network concept [2, 3].

Using our new theoretical framework that speeds up the micromagnetic simulation by a factor 2.4 billion [4], we obtained a recognition rate of 98.8% for MNIST handwritten digits [5] (see Fig. 1b), *i.e*, state of the art for hardware-compatible implementations.

**Acknowledgements**

Computational resources have been provided by the Consortium des Équipement de Calcul Intensif, funded by the F.R.S.-FNRS. F.A.A is a Research Associate and S.d.W. is a FRIA grantee, both of the F.R.S.-FNRS.

**References**

[1] J. Grollier *et al.*, *Nature Electronics* **3** (2020) 360.
[2] J. Torrejon, M. Riou, F. Abreu Araujo *et al.*, *Nature* **547** (2017) 428.
[3] F. Abreu Araujo *et al.*, *Scientific Reports* **10** (2020) 328.
[4] F. Abreu Araujo, C. Chopin, S. de Wergifosse, *arXiv*:2206.13596 (2022).
[5] A. Moureaux, S. de Wergifosse, C. Chopin, J. Weber, and F. Abreu Araujo, *arXiv*:2301.11025 (2023).

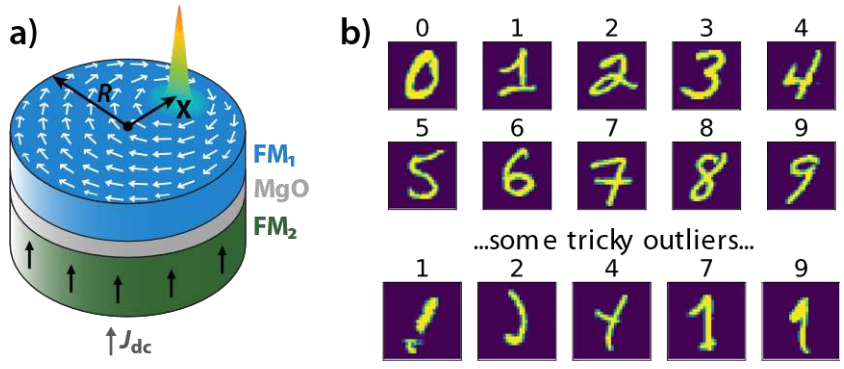



Figure 1: a) Schematic illustration of an MTJ-based spin-torque vortex oscillator. b) Subset of the MNIST handwritten digits dataset, which contains 60,000 images for training and 10,000 images for testing.

# Circular Memristive Nano-Gates on Spin Hall Nano Oscillators

*Maha Khademi[1,3]*, Akash Kumar[2], Mona Rajabali[3], Saroj P. Dash[1], and Johan Åkerman[2]*

[1] *Department of Microtechnology and Nanoscience, Chalmers University of Technology, 412 96, Gothenburg, Sweden*

[2] *Physics Department, University of Gothenburg, 412 96 Gothenburg, Sweden*

[3] *NanOsc AB, 164 40 Kista, Sweden*

*mahak@chalmers.se

Nano-constriction spin Hall nano oscillators (NC-SHNOs) are nano-meter-sized microwave signal-generating devices composed of a ferromagnetic layer adjacent to a heavy metal layer [1-2]. Their non-linearity, and synchronization in chains and arrays [3-5], make SHNOs great candidates for neuromorphic computing applications [4]. However, tuning the properties of individual SHNOs in large chains or arrays is a remaining challenge. Recently, Zahedinejad *et al.* demonstrated memristive control of mutual synchronization in SHNO chains and the results showed overall tunability of 60 MHz [6]. However, they used a broad bar-shaped gate over the NC, which required relatively thick oxide (>20 nm) [7], restricting its usefulness for larger arrays while reducing the frequency tunability.

In this work, we fabricate NC-SHNOs using W(5 nm)/CoFeB(1.4 nm)/$HfO_x$(6 nm) and compare circular memristive nano-gates positioned on top of, and just next to the nano-constriction SHNOs. For the case which the nano-gate is at the nano-constriction, we compared the SHNO auto-oscillation power spectral density (PSD), before and after activating the memristor, and discovered a 30 MHz irreversible downward shift. A plausible mechanism is a permanent change of the interfacial perpendicular magnetic anisotropy under the gate, which changes the operating frequency. By sweeping the gate voltage on the same device, we obtained very low tunability, indicating that placing the gate on top of the NC causes degradation and does not produce the desired frequency tunability. Furthermore, we studied the effects of a circular gate, placed 200 nm away from the NC. Memristive gating of this geometry showed frequency tunability of more than 200 MHz, which is almost three times higher than the previous reports, without changing the magneto-dynamic properties of the SHNOs. With nano-meter-sized gates, these results pave the path for efficient control of individual SHNO in larger chains or arrays which is crucial for their implementation for neuromorphic computing.

## References

[1] V. E. Demidov, *et al.*, *Nat. Mater.*, vol. 11, p. 1028, 2012.
[2] F. Moradi, *et al.*, *arXiv*, vol.1912.01347, 2019.
[3] A. A. Awad, *et al.*, *Nat. Phys.*, vol. 13, no. 3, pp. 292–299, 2016.
[4] M. Zahedinejad, *et al.*, *Nat. Nano.*, vol. 15, no. 1, pp. 47–52, 2020.
[5] A. Kumar, *et al.*, *arXiv*, vol. 2301.03859, 2023.
[6] M. Zahedinejad, *et al.*, *Nat. Mater.*, vol. 21, no. 1, pp. 81–87, 2022.
[7] A. Kumar, *et al.*, *Nanoscale*, vol. 14, pp. 1432–1439, 2022.

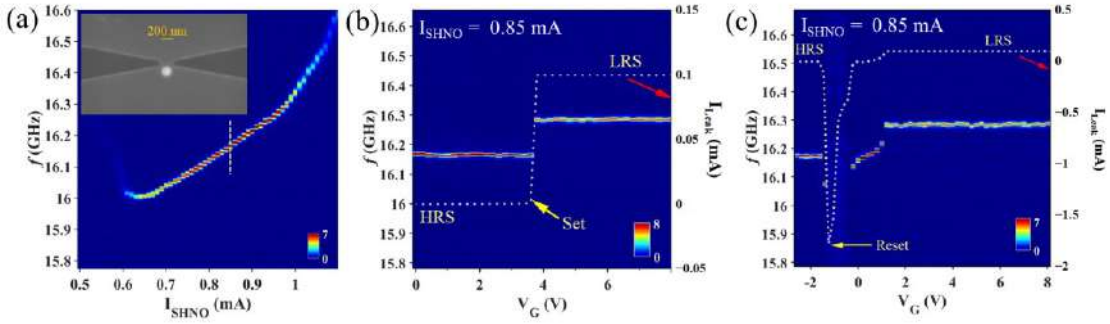



Figure: (a) The PSD plot versus $I_{SHNO}$ at 0.8 T magnetic field with 70° OOP and 20° IP angle for a 180 nm NC with a 200 nm circular gate separated by 200 nm. The dashed line indicates the current at which the gating is done. Inset: SEM image of a gated SHNO. The frequency and leakage current versus gate voltage from (b) zero to 8 V (forward sweep) and (c) 8 V to -3 V (reverse sweep).

# SYMPOSIUM 05.
## NEUROMORPHIC AND RESERVOIR COMPUTING.
## S5. POSTERS

# Artificial Synapses Based on Voltage-Controlled Magnetism via Nitrogen Ion Migration for Neuromorphic Computing

*P. Monalisha* [1]*, *Zheng Ma* [2], *Nicolau López-Pintó* [1], *Eva Pellicer* [1], *Enric Menéndez*[1], *and Jordi Sort* [1,2,*]

[1] *Departament de Física, Universitat Autònoma de Barcelona, E-08193 Cerdanyola del Vallès, Spain*

[2] *Institució Catalana de Recerca i Estudis Avançats (ICREA), Pg. Lluís Companys 23, E-08010 Barcelona, Spain*

*monalisha.peda@uab.cat, and jordi.sort@uab.cat

Synaptic devices with synchronized memory and processor are considered the core elements of neuromorphic computing (NC). So far, most synaptic devices are based on resistive memories, where the device resistance is tuned. However, the use of the electric current in such resistive devices causes significant power dissipation by Joule heating. Higher energy efficiency could be achieved in materials exhibiting voltage control of magnetism (VCM).[1] In particular, voltage-driven ion motion to modulate magnetism (magneto-ionics) is an emerging VCM mechanism that could offer new prospects for low-power implementation of NC. [2,3] In the present work, we exploit voltage-controlled nitrogen ion motion in transition metal nitrides (FeCoN) (i.e., nitrogen magneto-ionics) to emulate biological synapses. In the proposed device, we have realized multilevel non-volatile magnetic states for analog computing and high-density storage. Moreover, essential synaptic functionalities such as spike amplitude-dependent and duration-dependent plasticity, long term potentiation/depression of the human brain have been successfully mimicked. Furthermore, the device exhibits excellent retention and high endurance for real-life hardware implementation of NC. This research provides insight into the great potential of magneto-ionics-based synaptic devices for spin-based neuromorphic computing.

## Acknowledgements

The authors acknowledge financial support from the European Research Council (Grant Nº 101054687), the Spanish Government (PID2020-116844RB-C21 and PDC2021-121276-C31), and the Generalitat de Catalunya (2017-SGR-292).

## References

[1] M. Nichterwitz, S. Honnali, M. Kutuzau, S. Guo, J. Zehner, K. Nielsch, and K. Leistner, *APL Mater.* **9**, (2021) 030903.

[2] S. Martins, J. De Rojas, Z. Tan, M. Cialone, A. Lopeandia, J. Herrero-Martin, J.L. Costa-Kramer, E. Menendez, J. Sort, *Nanoscale* **14**, (2022) 842.

[3] R. Mishra, D. Kumar, H. Yang, *Phys. Rev. Appl.* **11**, (2019) 1.

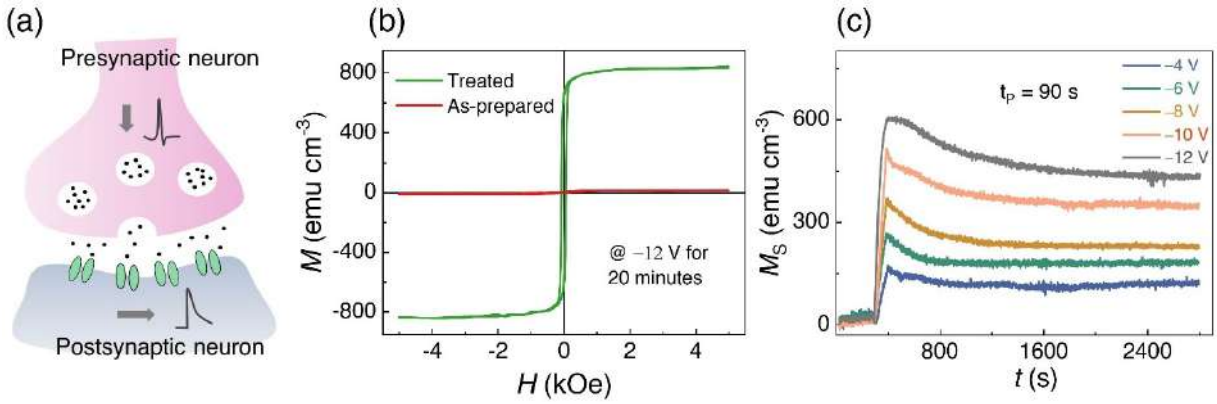



Figure: (a) Schematics of a biological synapse demonstrating signal transmission from presynaptic to postsynaptic neurons. (b) The *M-H* hysteresis loop of the CoFeN sample shows a paramagnetic to ferromagnetic switching with a voltage treatment of –12 V for 20 minutes. (a) Spike amplitude-dependent plasticity is emulated by firing spikes of different amplitudes (–4, –6, –8, –10, and –12 V) and of the same duration ($t_P$ = 90 s).

# Modelling of Reservoir Computing in Out-Of-Plane Artificial Spin Ice

*Jonathan Maes*[1]*, *Bartel Van Waeyenberge*[1]

[1] *Ghent University, Sint-Pietersnieuwstraat 25, 9000 Gent, Belgium*

*Jonathan.Maes@UGent.be

*Reservoir Computing* (RC) is a novel computational paradigm in which a nonlinear dynamical system (the 'reservoir') is exploited for computation. [1, 2] An input signal perturbs the reservoir, producing a complex dynamic response which facilitates classification. [1, 3, 4] By training a single linear readout on the output, the system can be used for machine learning purposes. [1, 3, 5] Since the reservoir is in general a 'nonlinear dynamical system', it is not limited to being a purely mathematical object. Many physical systems are highly nonlinear and exhibit fading memory, making them suitable to use as the reservoir. [3]

It has already been shown numerically that in-plane artificial spin ice (ASI) of various lattice geometries (e.g. Kagome, pinwheel, square) is suitable for RC. [1, 6, 7] We intend to explore the viability of using *out-of-plane ASI* as the reservoir, in particular the *square-lattice* geometry. This kind of system appears desirable as it allows input via spin-orbit-torque and readout through the anomalous Hall effect. A software tool 'Hotspice' was developed, to simulate the behavior of out-of-plane systems for a given combination of input and readout schemes. A simple Ising model approximation is used with a dipolar interaction present between all magnets along with an external field to apply input. Time evolution is performed either through Néel relaxation theory or Glauber dynamics. [8] Relevant metrics can then be calculated for a given system, e.g. the computing capacity $Q$ as shown in the accompanying figure, to optimize system parameters. [1]

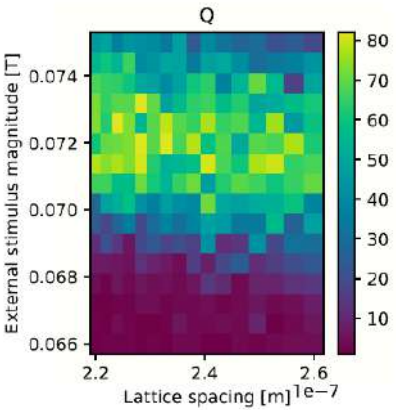



Figure: Computing capacity $Q$ for a 20×20 OOP Square spin ice with 10×10 checkerboard-masked readout and two-step checkerboard input scheme. For this system, the theoretical maximum value of $Q$ is 100. The input strength is varied along the vertical axis, the spacing between nearest neighbours along the horizontal axis.

The out-of-plane systems provide the challenge that they have fewer degrees of freedom as in-plane spin ices, as only one axis can be used to interact with the system. The two degenerate ground states of the system can not be distinguished by a uniform field, so a more intricate input scheme is required to get meaningful behavior in the system. Furthermore, it should be advantageous if domain walls can be moved by only a few lattice points per input value, to get a form of memory in the system. For these reasons, a two-step checkerboard-pattern input scheme was devised to promote one of the ground states over the other and move it by only a few lattice cells for each input value. As can be seen in the figure, this scheme gives decent values for $Q$.

## Acknowledgements

This project has received funding from the European Union's Horizon 2020 FET-Open program under Grant Agreement No. 861618 (SpinENGINE).

## References

[1] J. H. Jensen and G. Tufte, *Artificial Life Conference Proceedings* (2020) 376–383.
[2] D. Verstraeten, B. Schrauwen, M. D'Haene and D. Stroobandt, *Neural Networks* **20** (2007) 391–403.
[3] G. Tanaka , T. Yamane, J. B. Héroux, R. Nakane, N. Kanazawa, S. Takeda, H. Numata, D. Nakano and A. Hirose, *Neural Networks* **115** (2019) 100–123.
[4] L. Appeltant, M. C. Soriano, G. Van der Sande, J. Danckaert, S. Massar, J. Dambre, B. Schrauwen, C. R. Mirasso and I. Fischer, *Nature Communications* **2** (2011) 1–6.
[5] W. Maass, T. Natschläger and H. Markram, *Neural Computation* **14** (2002) 2531-2560.
[6] J. H. Jensen, E. Folven and G. Tufte, *Artificial Life Conference Proceedings* (2018) 15–22.
[7] K. Hon, Y. Kuwabiraki, M. Goto, R. Nakatani, Y. Suzuki and H. Nomura, *Applied Physics Express*, vol. **14** (2021) 033001.
[8] P. A. Flinn and G. M. McManus, *Physical Review* **124** (1961) 54.

# Development of A*d Hoc* Neuromorphic Computing Schemes for Energy-Efficient Machine Learning Using Spintronic Hardware

*Anatole MOUREAUX*[1]*, *Simon DE WERGIFOSSE*[1], *Chloé CHOPIN*[1], *and Flavio ABREU ARAUJO*[1]

[1] *Université catholique de Louvain, Place Croix du Sud 1, Louvain-la-Neuve, Belgium*

*anatole.moureaux@uclouvain.be

The rapidly growing use of artificial intelligence on a global scale since the last few years cannot be distinguished from ecological concerns regarding its energy consumption. In this regard, the development of beyond-CMOS computing systems has drawn a lot of attention in the scientific community. Among the proposed solutions, neuromorphic hardware offers to mimic the architecture of the human brain to build low-energy devices with the aim to solve machine learning tasks much more efficiently than conventional software neural networks (Fig. 1) [1]. The development of such devices can be performed using spin-torque vortex nano-oscillators (STVOs): magnetic tunnel junctions whose dynamics is known to be similar to that of biological neurons. They are also CMOS-compatible, low power, robust to noise and radiation, and can be densely integrated on a chip. Highly accurate automatic data classification such as speech recognition has already been demonstrated using STVOs [2, 3]. The development of such unconventional computing systems must also fully include the design of *ad hoc* algorithms suited to perform machine learning using neuromorphic spintronic hardware. Indeed, hardware neural networks cannot be trained using common methods such as error backpropagation and gradient descent due to the difficulty of tuning nanostructured systems repeatedly. Among the existing solutions, reservoir computing is a computing scheme that has already been successfully implemented on dedicated unconventional hardware [2, 3, 4].Recently, an ultra-fast STVO dynamics simulation framework called the data-driven Thiele equation approach (DD-TEA) was developed by Abreu Araujo *et al.* [5]. DD-TEA allowed to reach a speed-up of the STVO dynamics simulations of more than 2 billion compared to micromagnetic simulations. We used DD-TEA to carry extensive simulations of a STVO-based neural reservoir (Fig. 2) to assess its performance in various machine learning tasks [6]. Parametric studies allowed to assess the influence of the operating conditions such as the intensity of the input signal and the level of noise on the quality of the data classification. The high throughput of the simulations based on DD-TEA also allowed to perform image recognition and obtain state-of-the-art results on the MNIST dataset, a task which was initially not suited to reservoir computing. This suggests that STVO-based reservoir computing can be generalized to a bigger set of machine learning tasks, and that DD-TEA can be used to develop *ad hoc* computing schemes targeted on the hardware implementation of machine learning.

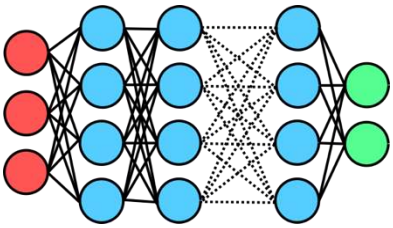


Fig. 1: A deep neural network (DNN) is usually implemented on conventional CMOS hardware.

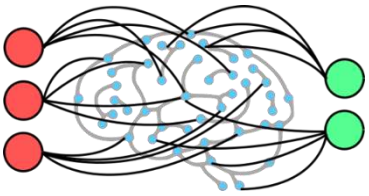


Fig. 2: A neural reservoir can be implemented using dedicated hardware, such as a STVO array.

## References

[1] J. Grollier, D. Querlioz, K. Y. Camsari *et al., Nature electronics* **3** (2020) 360.
[2] J. Torrejon, M. Riou, F. Abreu Araujo *et al., Nature* **547** (2017) 428.
[3] F. Abreu Araujo, M. Riou, J. Torrejon *et al., Scientific Reports* **10** (2020) 328.
[4] L. Larger, M. C. Soriano, D. Brunner *et al., Optics express* **20** (2012) 3241.
[5] F. Abreu Araujo, C. Chopin, S. de Wergifosse, *arXiv*:2206.13596 (2022).
[6] A. Moureaux, S. de Wergifosse, C. Chopin, J. Weber, and F. Abreu Araujo, *arXiv*:2301.11025 (2023).

# Coupled Resistive Switching based on Electrically triggered Metal-Insulator Phase transition in $La_{0.67}Sr_{0.33}MnO_3$ thin films

*Azminul Jaman[1]*, Anouk Goossens[1], Job van Rijn[2], & Tamalika Banerjee[1]*

[1] *Groningen Cognitive System and Materials Centre, University of Groningen, Groningen, Netherlands*

[2] *Zernike Institute for Advanced Materials, University of Groningen, Groningen, Netherlands*

*azminul.jaman@rug.nl

The development of in-memory computing hardware based on phase change resistive materials is an active research area. The resistive states are classified as either non-volatile or volatile and occur when changes to the material properties are triggered by an external stimulus such as temperature, current, voltage, or electric field. The volatile change of the resistance state arises due to the transition of the switching layer from an insulator to a metal. We have reported that an electric current-induced Joule heating triggers volatile resistive phase transition in a strained thin film of $La_{0.67}Sr_{0.33}MnO_3$ grown on a textured substrate of $LAlO_3$ [1]. The volatility of the two distinct resistance states has been utilized to establish a nanometer-thick relaxation oscillator. The two phases can be reversibly altered back and forth using a resistor and a capacitor in a simple electronic circuitry. The coupling of oscillators offer a new approach in realizing compact oscillatory neural networks. The anisotropic distribution of the LAO substrate's twin planes result in a local difference in the strain environment, resulting in a distribution of coexisting ferromagnetic metallic phases across the heterostructure. These local difference in the phases has been exploited to stabilize oscillators with a different oscillation frequencies in the same film. We also investigated the influence of the coupling capacitor and resistor on the coupled oscillation frequency along the different directions of the film.

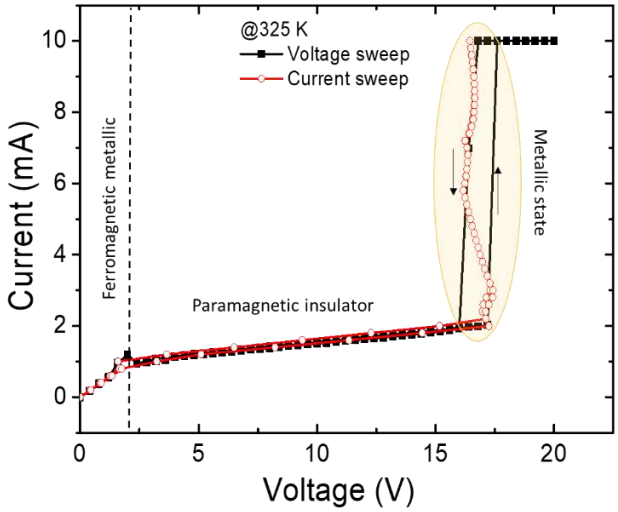



Figure: At 325 K, three distinct regimes of volatile resistive switching in voltage-controlled (black squares) and current-controlled mode (red open circles) is shown for LSMO thin films on LAO. In voltage-controlled mode, the change of slope at 2 V suggests a metal-to-insulator transition, and at higher voltage (16 V), it indicates an insulator to a metal transition by an abrupt current increase.

## References

[1] A. Jaman, A.S. Goossens, J.J. L van Rijn , L. Van der Zee and T. Banerjee, *Fron. Nano.* **5,** (2023) 6.

# Relaxation Time of Superparamagnetic Tunnel Junction at Various Temperatures

*Haruna Kaneko[1,2]*, Rikuto Ota[1,2], Keito Kobayashi[1,2], Shun Kanai[1-6], Hideo Ohno[1,5-7] and Shunsuke Fukami[1,2,5-8],*

[1]*Laboratory for Nanoelectronics and Spintronics, RIEC, Tohoku University, Sendai, Japan*

[2]*Graduate School of Engineering, Tohoku University, Sendai, Japan*

[3]*PRESTO, Japan Science and Technology Agency, Kawaguchi, Japan*

[4]*Division for the Establishment of Frontier Science, Tohoku University, Sendai, Japan*

[5]*Center for Science and Innovation in Spintronics, Tohoku University, Sendai, Japan*

[6]*WPI-Advanced Institute for Materials Research, Tohoku University, Sendai, Japan*

[7]*Center for Innovative Integrated Electronic Systems, Tohoku University, Sendai, Japan*

[8]*Inamori Research Institute for Science, Kyoto, Japan*

*haruna.kaneko.t3@dc.tohoku.ac.jp

Superparamagnetic tunnel junctions (s-MTJs) are gathering attention as a key element for spintronics-based probabilistic computers [1-4]. The temperature dependence of the s-MTJ properties is of importance because the stability of their bit states changes with the operation temperature, affecting the computation performance. In this work, we investigate the relaxation time and time-averaged response of the s-MTJs at different temperatures and discuss the factors that determine the temperature dependence of the properties.

A stack structure, buffer/ SyF/ Ta (0.2)/ CoFeB (1.0)/ MgO (1.1)/ CoFeB (1.5)/ cap (thickness in nm) is deposited by dc/rf magnetron sputtering, processed into circular MTJs with diameters of 10-30 nm by electron beam lithography and Ar ion milling, and annealed at 300°C for 1 hour. Both the CoFeB layers have a perpendicular easy axis. The magnetic moment of the bottom CoFeB is fixed by the synthetic ferrimagnet (SyF), while the moment of the top CoFeB layer is designed to fluctuate at room temperature. The typical tunnel magnetoresistance (TMR) ratio and the resistance area product are 77% and 11 Ωμm$^2$, respectively.

We measure rf transmitted voltage and the time-averaged (dc) resistance $\langle R \rangle$ of the s-MTJs at different temperatures $T$ of 20-130°C and perpendicular magnetic fields $H_z$. From the rf transmitted voltage, we determine the average relaxation time $\tau_{ave}$ [3]. Obtained $T$ dependence of $\tau_{ave}$ for four devices with different diameters $D$ is shown in Figure, indicating that $\tau_{ave}$ exponentially decreases with $1/T$ from ~100 ms to ~10 μs in the studied $T$ range and the data is deviated from the exponential fit (solid lines) at high $T$ region. The exponential relation agree with the Arrhenius model $\tau_{ave} = \tau_0 \exp(E/k_BT)$, where $\tau_0$, $E$, and $k_B$ denote attempt time, energy barrier, and the Boltzmann constant, respectively [5]. Combining the insights obtained from separate experiments [6], we find that the results shown in Figure can be explained with the temperature dependent effective anisotropy field, magnetic moment of the free layer, and $\tau_0$.

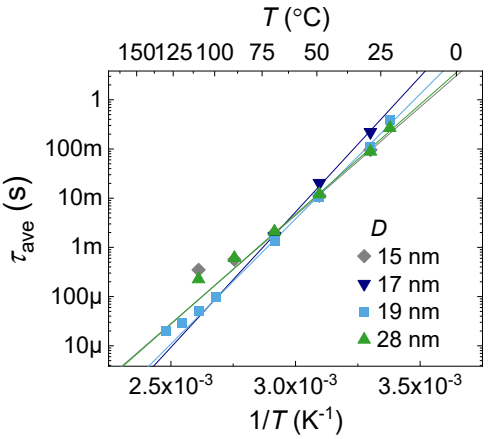



Figure: Inverse temperature $1/T$ dependence of relaxation time $\tau_{ave}$ with the fit based on the standard Arrhenius model $\tau_{ave} = \tau_0 \exp(E/k_BT)$.

**Acknowledgements**

This work was supported in part by the JST-CREST Grant No. JPMJCR19K3, the JST-PRESTO Grant No. JPMJPR21B2, JSPS Kakenhi Grant Nos. 19H05622, 19KK0130, and 20H02178, Shimadzu Research Foundation, Takano Research Foundation, and Cooperative Research Projects of RIEC.

**References**

[1] K. Y. Camsari *et al*., *Phys. Rev. X* **7**, 031014 (2017). [2] W. A. Borders *et al*., *Nature* **573**, 390 (2019).
[2] K. Hayakawa *et al*., *Phys. Rev. Lett.* **126**, 117202 (2021). [4] T. Funatsu *et al*., *Nat. Commun*. **13**, 4079 (2022).
[3] W. F. Brown, *Phys. Rev.* **130**, 1677 (1963). [6] K. Kobayashi *et al*., *Appl. Phys. Lett.* **119**, 132406 (2021).

# Lanthanide molecular nanomagnets as probabilistic bits for artificial stochastic neural networks

*Gerliz M. Gutiérrez-Finol*[1]*, *Silvia Giménez-Santamarina*[1], *Ziqi Hu*[1], *Lorena E. Rosaleny*[1],*Salvador Cardona-Serra*[1], *Alejandro Gaita-Ariño*[1]

[1] *ICMol, Universitat de València, c/cat José Beltrán, 2, 46980, Paterna, Spain*

*gerliz.gutierrez@uv.es

Over decades, the spin dynamics of a large set of lanthanide complexes have been explored [1,2]. Lanthanide-based molecular nanomagnets are bistable spin systems, generally conceptualized as classical bits, but many lanthanide complexes have also been presented as candidates for quantum bits (qubits). Here we offer a third alternative and model them as probabilistic bits (p-bits) [3], where their stochastic behavior constitutes a computational resource instead of a limitation. We employ STOSS [4], a modelling tool for molecular spin p-bits, to simulate bulk magnetic relaxation data and acexperiments and to simulate a minimal p-bit network under realistic conditions. Finally, we employ a recent systematic data gathering [5] and screen the best lanthanide complexes for p-bit behavior, lay out the performance of the different lanthanide ions and chemical families and offer some chemical design considerations. P-bits based on molecular nanomagnets constitute the low-size, high-frequency limit of Low Barrier Nanomagnets for Artificial Stochastic Neural Network.

**Acknowledgements**

We acknowledge funding to COST Project 15128, EU Horizon 2020 (ERC-2017-AdG-788222 “MOL2D” and FATMOLS project),GeneralitatValenciana CIDEGENT/2021/018 grant and GVA PROMETEO/2019/066, MICINN grants PID2020-117264GB-I00 and PID2020-117177GB-I00 (co-financed by FEDER funds), MICINN PRE2018-083350, CEX2019-000919-M and MINECO CTQ2017-89528-P.

**References**

[1] C. Bennelli, D. Gatteschi, *Introduction to Molecular Magnetism: From Transition Metals to Lanthanides*, Wiley-VCH Verlag GmbH & Co., Weinheim, 2015.
[2] Z. Zhu, M. Guo, X. Li, J.Tang. *Coordination Chemistry Reviews,* **378**(2019) 350.
[3] S. Chowdhury, et al. arXiv:2302.06457v2 [cs.ET].
[4] G. M.Gutiérrez-Finol, et al. *Under review at npj Computational Materials*, **arXiv:2301.08182**.
[5] Y. Duan, et al. *Nat. Commun*., **13** (2022) 7626.

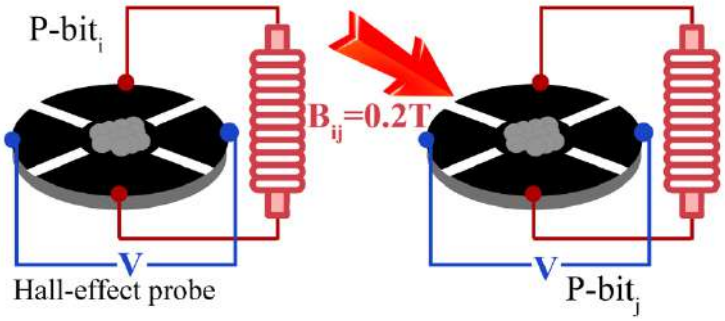



Figure: Proposed experimental setupfor a toy network scheme of correlated p-bits based on single molecules of $[(Cp^{iPr5})Dy(Cp^{*})]^{+}$ at T = 40 K and where the state of p-bit *i* controls a magnetic field B = 0.2 T acting on p-bit *j*.

# Weighted Spin Hall Nano-Oscillator-based Neuromorphic Computing System

*Mona Rajabali[1]*, Akash Kumar[2], Mohammad Zahedinejad[1], Saverio Ricci[3], Davide Bridarolli[3], Hooman Farkhani[4], Farshad Moradi[4], Daniele Ielmini[3], Johan Åkerman[2,5]*

[1] *NanOsc AB, Electrum 229, 164 40 Kista, Stockholm, Sweden*
[2] *University of Gothenburg, 412 96 Gothenburg, Sweden*
[3] *Politecnico di Milano and IU.NET, Piazza L. da Vinci 32, Milano 20133, Italy*
[4] *Aarhus University, 8200 Aarhus, Denmark*
[5] *Tohoku University, 2-1-1 Katahira, Aoba-ku, Sendai 980-8577, Japan*
*mona.rajabali@nanosc.se

Spintronics, which is privileged due to using the spin of the electron instead of its charge, has evolved into a prospective candidate to contribute to future high-density, high-speed, and energy-efficient brain-inspired neural networks. This branch of computing, also known as neuromorphic computing systems (NCSs), is shedding light on resolving the strict limitations of the widespread Von-Neumann architecture in terms of energy cost and computational speed [1]. In a novel approach, SpinAge, the Horizon 2020-funded EU project, portrays the idea of implementation of nonlinear oscillators as neurons, along with memory components as synapses [2,3], to mimic the behavior of the brain, especially for cognitive computing.

Nano-constriction Spin Hall nano-oscillators (NC-SHNOs) are nascent nonlinear oscillators that have aroused interest due to their easy fabrication, frequency tunability, direct optical access, and the potential for memristive control. [4,5]. Unlike STNOs, there has been no study on integrating SHNOs and separate memory elements as NCS. Hence here, a NC-SHNO combined with five memristors is being investigated as the building block of a complete neural network where the firing/non-firing state of the neurons can be controlled by weighted memristors (Fig.1(a)). The NCS benefits from these nonlinear high-frequency oscillators which can be simply utilized for any desired configurations of connected oscillators (neurons), interacting through programmable memory units (synapses), resulting in a hybrid energy-efficient brain-inspired architecture where memory and processing occur on a nano-scale device. Moreover, spintronic oscillators' capability of directly processing the Radio-Frequency (RF) data, in a wide frequency band, presents a potential solution to the high power consumption associated with digitizing RF inputs in conventional on-chip oscillators [6].

To build a single element of this network, a bow-tie-shaped NC-SHNO (inset in Fig.1(b)) fabricated on W(5nm) /NiFe(5nm)/ $Al_2O_3$(5nm) is sharing a contact pad with five separated memristors. The power spectral density (PSD) of an SHNO microwave signal, at 0.64 T external magnetic field, is shown in Fig.1(b). SHNO state (firing/non-firing) will be regulated by the current flowing to the system through the weighted memristors.

SpinAge envisions integrating all advanced technologies, including spintronic oscillators, memristors, on-chip laser technology, and CMOS, to implement an innovative NCS leading to at least 3-4 orders of magnitude better performance than the state-of-the-art CMOS-based NCSs.

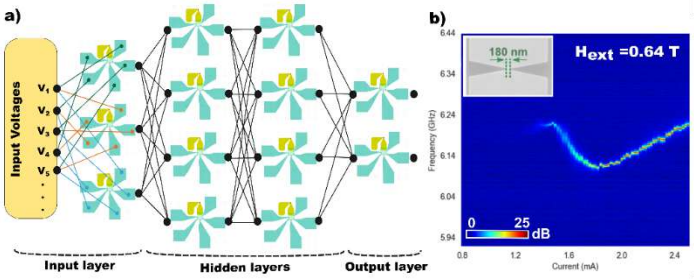



Figure 1: (a) The neural network diagram comprising of SHNOs (neurons) along with five memristors (synapses). (b) The PSD of the microwave signal generated by a single NC-SHNO. Inset shows a top-view SEM image of a bow-tie-shaped NC-SHNO.

## References

[1] I. L. Markov, *Nature* **512** (2014) 147.
[2] A. Milozzi, et al., *ESSCIRC 2022-IEEE 48th European Solid State Circuits Conference, IEEE* (2022).
[3] S. Ricci, et al., *Adv. Intell. Syst.* **4** (2022) 2200053.
[4] M. Zahedinejad, et al., *Nat. Mater.* **21** (2022) 81.
[5] M. Zahedinejad, et al., *Nat. Nano.* **15** (2020) 47.
[6] A., Ross, et al. arXiv preprint arXiv: 2211.03659 (2022).

# Switching of an Antiferromagnet with Neuromorphic Functionality

*Jan Zubáč[1, 2,*], Miloslav Surýnek[2], Filip Krizek[1], Vít Novák[1], Petr Němec[2], Tomáš Jungwirth[1, 3], Kamil Olejník[1]*

[1] *Institute of Physics of the Czech Academy of Sciences, Prague, Czech Republic*
[2] *Faculty of Mathematics and Physics, Charles University, Prague, Czech Republic*
[3] *School of Physics and Astronomy, University of Nottingham, United Kingdom*
*zubac@fzu.cz

Antiferromagnetic materials show their potential for next-generation technology due to their exceptional features: Fast terahertz magnetization dynamics, low power consumption and possible high integration density due to no cross-talk among adjacent devices are desirable for information storage and processing and for applications in unconventional computing.

Among antiferromagnets, tetragonal CuMnAs proved its uniqueness for possible ultimate scaling of its magnetic textures down to the atomic level [1], analogue multilevel storage capability and the possibility of writing using a single stimulus at a broad range of timescales – from a millisecond electrical pulse to a femtosecond laser pulse [2]. These characteristics, combined with the availability of conventional electrical readout of the magnetic state, make CuMnAs an ideal testbed material for exploring novel neuromorphic functionalities.

Here, we report on the optical switching of CuMnAs utilizing temporal correlations of two 100-fs laser pulses. We experimentally prove the readiness of the CuMnAs device to operate at ultrashort timescales as well as the ability to determine the relative timing of pulses, known as neuronal spike-time-dependent plasticity. Finally, we take advantage of these principles and demonstrate the practical usability of the device for processing digital information from an image. By bridging the gap between ultrafast optical switching and non-volatile memristors, our results open a pathway towards the design of novel computing architectures.

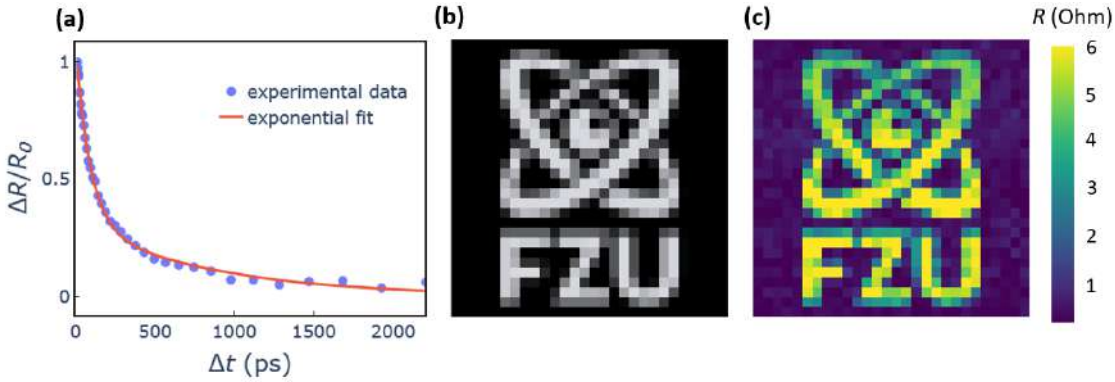



Figure: Image encoded using relative timing of two optical pulses: **(a)** Dependence of the resistive signal on the delay between optical pulses. **(b)** Original image. **(c)** Resulting resistive signal.

## Acknowledgements

This work was supported by Czech Science Foundation Grant No. 21-28876J.

## References

[1] F. Krizek, et al., *Sci. Adv.* **8.13** (2022) eabn3535.
[2] Z. Kašpar, et al., *Nat. Electron.* **4.1** (2021) 30-37.

# Towards smart sensing with a spintronic reservoir computer

*G. Venkat[1], I. Vidamour[2], M. O. A. Ellis[2], C. Swindells[1], L. Manneschi[2], E. Vasilaki[2], D. A. Allwood[1], T. J. Hayward[1]*

[1]*Department of Materials Science and Engineering, University of Sheffield, Sheffield, S1 3JD, UK*

[2]*Department of Computer Science, University of Sheffield, Sheffield, S1 4DP, UK*

*g.venkat@sheffield.ac.uk

Physical systems showing emergent behaviour are promising candidates for unconventional types of computing like *in-materia* processing of information [1]. In particular, 'reservoir computing' (RC), a highly efficient framework for dynamic signal processing, has been proposed as promising for edge computing and smart sensing. In this paradigm, the energy overheads of analog-to-digital conversion and powering discrete sensors can be significantly reduced if the reservoir is able to respond directly to the stimuli that will be sensed. We have recently demonstrated that the complex, probabilistic, emergent behaviour in interconnected $Ni_{80}Fe_{20}$ ring array metamaterials (Fig 1(a)) [2-4] is particularly useful for RC [5], with our devices exhibiting state-of-the-art performance in machine learning tasks with varying computational requirements [6]. In these demonstrations, data was encoded using a rotating magnetic field and the array response probed using anisotropic magnetoresistance (AMR). The device performed competitively in signal transformation (Fig. 1(b)), speech classification and the NARMA5/10 time series prediction tasks (Fig. 1(c)), the latter being the first such demonstrations using a spintronic reservoir without external feedback lines. Nanoring arrays are thus effective reservoir computers.

To explore the utility of the ring arrays as smart sensor, their response to temperature stimuli was assessed using a Peltier cell (Fig. 1 (d)). Field driven AMR amplitudes at different temperatures are shown in Fig. 1(e). The AMR response at a given temperature was strongly non-linear and showed a sharp transition at ~27 Oe (for 20 deg C), the onset of domain wall propagation [6]. Furthermore, the response exhibited a consistent, linear field shift with increasing temperature (Fig. 1 (f)). We then performed computation with input signals encoded in the device temperature, thus producing an RC-based smart temperature sensor. These results demonstrate the feasibility of creating reservoir computers that interact directly with environmental stimuli, laying the ground for smart sensors that both detect environmental changes and infer meaning from them.

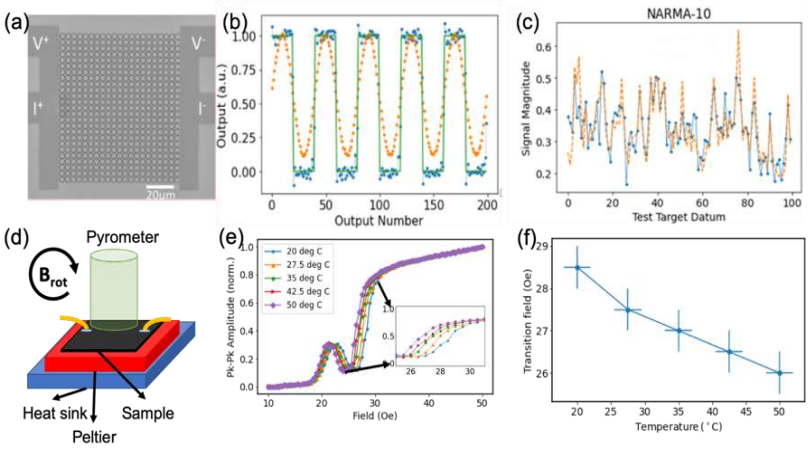



Figure 1:(a) An SEM micrograph of the ring array. (b) The reconstruction (blue) of a square wave agrees well with the target (green) while that without reservoir transformation does not agree. (c) The reconstruction (orange) of the NARMA-10 task agrees well with the target output (blue). (d) Experimental setup used for applying a temperature stimulus. The sample is mounted on a Peltier and a pyrometer measures the system temperature (e) The peak-to-peak amplitude of the AMR signal as a function of rotating field for different temperatures. The inset shows the shift in field response for various temperatures. (f) The variation of the transition field of the response with temperature shows a strongly linear relationship.

## References

[1] Miller et al. Evolutionary Intelligence 7.1, 49-67 (2014)
[2] Negoita et al., J. Appl. Phys. 114, 013904 (2013)
[3] Dawidek et al., Adv. Funct. Mater., 2008389 (2021)
[4] Vidamour et al. Nanotech. 33.48, 485203 (2022)
[5] Jensen et al., ALIFE 2018: The 2018 Conference on Artificial Life 15, (2018)
[6] Vidamour et al. arXiv preprint arXiv:2206.04446 (2022).

# Reading out a nanomagnetic reservoir by exploiting anisotropic magnetoresistance (AMR)

*Jonas J. Joos[1,*], Rolf Allenspach[2], Ian T. Vidamour[3], Guru Venkat[3], Thomas J. Hayward[3], Bartel Van Waeyenberge[1]*

[1] *Dynamat, Department of Solid State Sciences, Krijgslaan 281/S1, Ghent University, Belgium*
[2] *IBM Research–Zurich, Säumerstrasse 4, CH-8803 Rüschlikon,* Switzerland
[3] *Department of Materials Science and Engineering, University of Sheffield, United Kingdom*
*jonas.joos@ugent.be

It is well established that emergent dynamics can occur in complex nanomagnetic systems, opening a promising avenue for *materials computation* where the nanomagnetic structure forms a neural network, the so-called reservoir. Reservoir computing is then achieved by training the readout layer to map the output of the magnetic system to the targeted problem [1].

Recent research has shown that the desired complex behaviour can be found in the domain wall dynamics of arrays of permalloy rings which can be excited by external rotating magnetic fields. Different strategies have been explored to tune and optimize the computational capabilities of such arrays for classification tasks [2,3]. These encouraging results were obtained from microscopic details of the magnetic structure, however, these are not immediately accessible in real life. In practical reservoir computers, the read-out layer extracts a coarse-grained picture of the underlying microscopic reality.

In this talk, we report on an investigation on read-out layers that are based on electric resistance measurements. Due to the anisotropic magnetoresistance (AMR), domain walls have a direct impact on the electric resistance, enabling to capture their dynamics from resistance measurements. A model was developed where detailed information on electric currents in individual rings and junctions of rings, obtained from micromagnetic simulations [4], complemented with a finite-differences Poisson solver, can be extrapolated to study the current paths through a large-scale array of hundreds to thousands of rings. The magnetisation configurations of the array are created by an agent-based phenomenological solver, as systems of this size are inaccessible with micromagnetics [2,3]. Simulation results are compared to experiments.

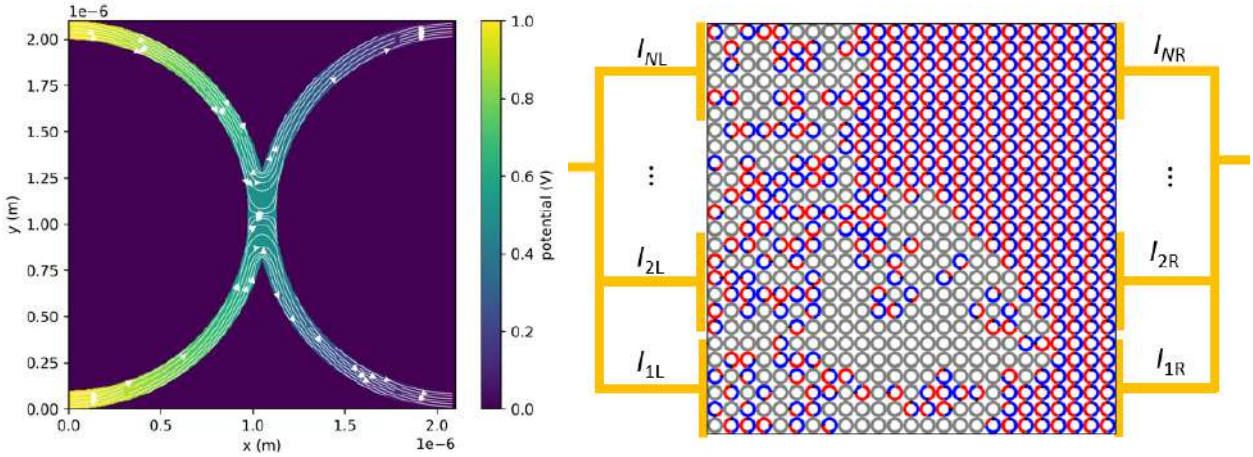



Figure: Electric current paths are simulated through individual rings and junctions, as well as through entire reservoirs.

**Acknowledgements**

We are grateful for funding from the European Union's Horizon 2020 FET-Open program under grant agreement No 861618 (SpinEngine).

**References**

[1] J. H. Jensen et al., *Artificial Life Conference Proceedings* (2018) 15-22.
[2] R. W. Dawidek et al., *Adv. Funct. Mater.* **31** (2021) 2009389.
[3] I. T. Vidamour et al., *Nanotechnology* **33** (2022) 485203.
[4] A. Vansteenkiste et al., *AIP Advances* **4** (2014) 107133.

# Nailed it: Reservoir Computing with a Rusty Iron Nail

*C. Swindells[1]*, I.T. Vidamour[1,2], C. Oh[1], G. Venkat[1], D.A. Allwood[1], T.J. Hayward[1]*

[1] *Department of Materials Science and Engineering, University of Sheffield, UK*

[2] *Department of Computer Science, University of Sheffield*

*c.swindells@sheffield.ac.uk

In recent years, the use of machine learning has grown significantly. However, on conventional CMOS based systems, this has resulted in massive increases in energy due to the von Neumann bottleneck, where data must be shuffled between memory and processing units. Additionally, in commonly used recurrent neural networks, this problem is amplified by costs to train weights of connections between nodes which have temporal interactions. One solution is Reservoir Computing (RC) where networks are replaced with a non-linear dynamical system, and only the output weights are trained. While this removes some of the energy overhead, on traditional CMOS systems the von Neumann bottleneck remains. This can be overcome by replacing simulated dynamic systems with an appropriate material platform. Two general requirements are non-linear responses to input stimuli, and 'fading memory', where current states are dependent upon past states, both of which are satisfied by most magnetic systems.

This has led to an arms race in magnetic platforms, with proposed RC devices based on spin oscillators [1], magnonic systems [2], artificial spin ices [3], and stochastic domain wall interactions [4] in nanoscale metamaterials [5,6], among others. Thus far, performance has typically been expressed in terms of benchmark tasks, such as speech or image recognition. However, it is not clear whether these tasks are indicative of a good RC platform, or indeed whether proposed materials are capable of performing well at other other tasks, which may require properties not present in the material's physics.

Here, we illustrate this problem with very competitive performance in several 'benchmarks', using a rusty nail. This reservoir is simple and has low energy requirements - needing less than 25 Oe as a driving field for input, and a simple pickup coil for output. As the nail is magnetic, different field scalings below the coercive field allow for different input transformations (see figure). With this framework we are able to achieve over 98% accuracy in a simple reservoir architecture in a speech recognition task and 0.35 NRMSE in NARMA 5. We show that in this task, a non-linear transform and good signal to noise is key (see figure), and that tasks may not be the most useful assessment of RC capability. Instead, task independent metrics of kernel rank, generalisation rank and memory capacity are more suitable.

## References

[1] D.Marković et al., *Appl. Phys. Lett.* **114** (2019) 012409.
[2] A.Papp et al., *Appl. Phys. Lett* **119** (2021) 112403.
[3] J.C.Gartside et al., *Nat. Nano.* **17** (2022) 460.
[4] A.Welbourne et al., *Appl. Phys. Lett.* **118** (2021) 202402.
[5] R.W.Dawidek et al., *Adv. Funct. Mater*. **31** (2021) 2008389.
[6] I.T.Vidamour et al., *Nanotechnology* **33** (2022) 485203.

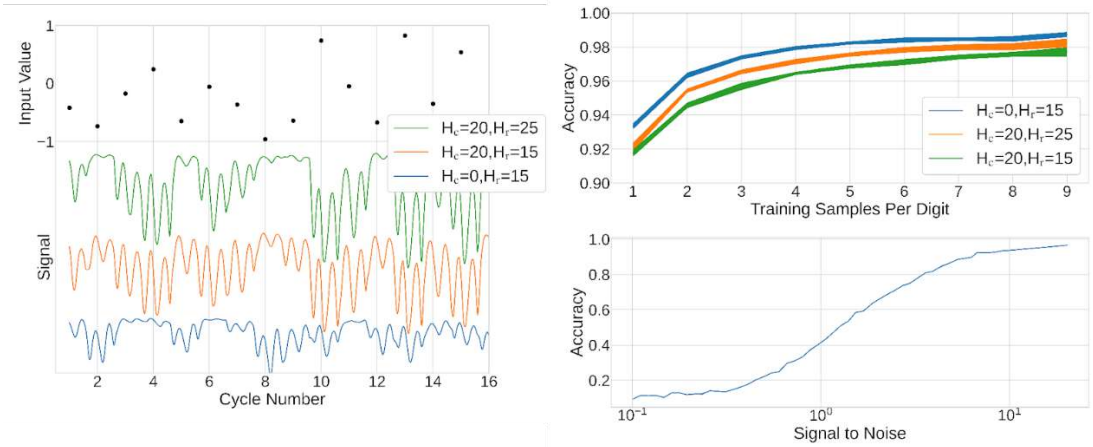



Figure: System response at three different input scaling parameters, for a given random input sequence (left). Accuracy of the nail at recognising spoken digits as a function of training iterations, for three input scalings. Final accuracy of the nail after 9 training iterations with additional noise from a gaussian distribution added (right).

# Nonlinear interactions between spin-wave modes in YIG microdisks

*G. Soares*[*1], A. Kolli[1],R. L. Seeger[1], I. Ngouagnia[1], T. Srivastava[1,2],M, Massouras[2], J.-V. Kim[2], H. Merbouche[3], S. Demokritov[3], V. Demidov[3], N. Beaulieu[4], J. Ben Youssef[4], M. Muñoz[5], S. Perna[6], M. D'Aquino[6], C. Serpico[6], A. Anane[7], G. De Loubens[1]*

[1]*SPEC, CEA, CNRS, Université Paris-Saclay, France*
[2]*Université Paris-Saclay, CNRS, Centre de Nanosciences et de Nanotechnologies, France*
[3]*Institute for Applied Physics, University of Muenster, Germany*
[4]*LabSTICC, CNRS, Université de Bretagne Occidentale, Brest, France*
[5]*Instituto De Tecnologías Físicas y de La Informacíon (CSIC), Madrid, Spain*
[6]*Department of Electrical Engineering and ICT, University of Naples Federico II, Italy*
[7]*Unité Mixte De Physique, CNRS, Thales, Université Paris-Saclay, France*
*gabriel.soares@cea.fr

Leveraging on nonlinear magnetization dynamics is promising for neuromorphic computing [1]. Recently, pattern recognition has been demonstrated using a magnon-scattering reservoir [2]. To proceed further from this stage, one should be able to train the neural network. In magnetic microstructures, spin-wave eigenmodes -- neurons -- are defined in the k-space. Mutual nonlinear couplings between these modes -- synaptic weights -- are predominantly determined by their amplitudes. We have previously demonstrated that parametric pumping allows the selective excitation of a large number of eigenmodes in YIG microdisks [3]. Here, we simultaneously excite pairs of modes by this mean to study their mutual nonlinear interactions. Two-tone MRFM spectroscopy demonstrates that each mode is coupled to all other modes, with enhanced or suppressed peaks, and the appearance of additional peaks in the spectrum (Fig.1). Full micromagnetic simulations and a description of the nonlinear magnetization dynamics in terms of normal modes [4] provide some insights into these nonlinear processes.

**Acknowledgements**

This work has received financial support from the Horizon 2020 Framework Programme of the European Commission under FET-Open grant agreement no. 899646 (k-NET).

**References**

[1] A. Papp, W. Porod, and G. Csaba, "Nanoscale neural network using non-linear spin-wave interference", Nature Commun. 12, 1–8 (2021).
[2] L. Korber, et al., "Pattern recognition with a magnon-scattering reservoir", 10.48550/arXiv.2211.02328.
[3] T. Srivastava, et al., "Complete identification of spin-wave eigenmodes excited by parametric pumping in YIG microdisks", 10.48550/arXiv.2301.13468.
[4] S. Perna et al., "Computational micromagnetics based on normal modes: bridging the gap between macrospin and full spatial discretization", J. Magn. Magn. Mater. 546, 168683 (2022).

# Synaptic Time Dynamics in Molecular $La_{0.7}Sr_{0.3}MnO_3$/Gaq3/AlOx/Co Spintronic Devices

*Alberto Riminucci[1]*, Andrei Shumilin[2], Prakriti Neha[1], Mattia Benini[1], Rajib Rakshit[1], Manju Singh[1], Luca Gnoli[1], Ilaria Bergenti[1] , Raimondo Cecchini[3]*

[1] *Institute for the Study of Nanostructured Materials, CNR, Via Gobetti 101, 40129 Bologna, Italy*
[2] *Ioffe Institute, Russian Academy of Sciences, 194021 St. Petersburg, Russia*
[3] *Institute for Microelectronics and Microsystems, CNR, Via Gobetti 101, 40129 Bologna, Italy*
*alberto.riminucci@cnr.it

The development of neuromorphic devices is a pivotal step in the development of low power artificial intelligence. We have studied the synaptic behaviour of molecular $La_{0.7}Sr_{0.3}MnO_3$/Gaq3/AlOx/Co spintronic devices[1], where the conductance plays the role of the synaptic weight[2]. We arranged the devices a in crossbar configuration, the most effective architecture for the purpose. We controlled the conductance of each cross point separately, with a maximum on/off ratio of $10^2$ and an average cross talk of 8%. The conductance was controlled by the application of voltage pulses. Positive voltage pulses were used for synaptic potentiation, i.e. conductance increase, while negative ones were used for synaptic depression, i.e. conductance decrease. When set in the high conductance state, the devices showed spin-valve magnetoresistance (see Figure 1), while in the low conductance state, no magnetoresistance was observed.

The time dependence of the resistive switching behavior is an important parameter for the synaptic behavior[3]. In order to study the time dynamics of the resistive switching after the voltages pulses, we conducted two different types of measurements. In the first type, we applied an initial reset voltage, followed by repeated, identical set voltage pulses of opposite polarity. The threshold voltage, whose magnitude must be exceeded in order to affect the conductance, was -1 V for depression and +1.75 V potentiation, regardless of pulse duration. Noise in the conductance after a voltage pulse above the threshold does not differ from the conductance noise after a voltage pulse below threshold. This means that the mechanism causing the noise was different from the one causing the resistive switching. The second type of pulsed measurements was carried out by applying set and reset voltages alternatively and by reading the resistive state as a function of time in the intervening time. In both types of measurements, we found a logarithmic relaxation of the conductance over time.

We put forward a theoretical model for this glassy logarithim dynamics. The transport was attributed to doping species, that in turn are affected by the set and reset voltages, if they exceed a threshold level. The activation energies to move these dopants had an exponential distribution, that can explain the galssy behavior.

## Acknowledgements

The authors would like to thank Federico Bona for his invaluable technical contribution.

## References

[1] A.Riminucci et al., ACS Appl. Mater. Interfaces **11** (2019) 8319-8326
[2] A.Riminucci and R.Legenstein , *arXive*: 1903.08624 (2019)
[3] D.V.Christensen et al., *Neuromorph. Comput. Eng.* **2** (2022) 022501

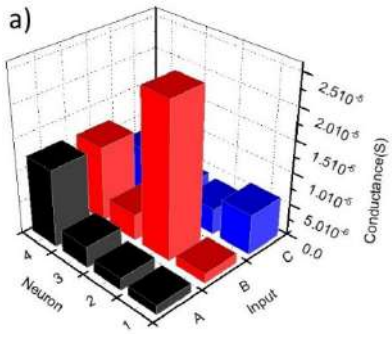



Figure 1: Conductance G at 100 K. a) G of devices arranged in a 4×3 crossbar, with Input 2, Neuron 2 set in high G b) Magnetoresistance of the Input 2, Neuron 2 device.

# Adaptive programmable networks for in-materia neuromorphic computing

**Kilian D. Stenning**[1,2], Jack C. Gartside[1,†], Luca Manneschi[3,†], Christopher T. S. Cheung[1] , Tony Chen[1] , Alex Vanstone[1], Jake Love[4,5], Holly H. Holder[1], Francesco Caravelli[6], Karin Everschor-Sitte[4], Eleni Vasilaki[3], and Will R. Branford[1]

[1]Blackett Laboratory, Imperial College London, London SW7 2AZ, United Kingdom
[2]London Centre for Nanotechnology, University College London, London WC1H 0AH, United Kingdom
[3]University of Sheffield, Sheffield S10 2TN, United Kingdom
[4]Faculty of Physics, University of Duisburg-Essen, 47057 Duisburg, Germany
[5]Institute of Physics, Johannes Gutenberg University Mainz, 55128 Mainz, Germany
[6]Theoretical Division (T4), Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA

Modern AI and machine-learning provide striking performance. However, this comes with rapidly spiralling energy costs[1] arising from growing network size and inefficiencies of the von Neumann architecture. 'Reservoir computing'[2] offers an energy-efficient alternative to large networks, fixing randomised weights for energetically cheap training. The massively parallel processing underpinning machine-learning is poorly catered for by CMOS, with in materia neuromorphic computing an attractive solution.
Nanomagnetic artificial spin-systems are ideal candidates for neuromorphic hardware. Their passive memory, state-dependent dynamics and nonlinear GHz spin-wave response provide powerful computation[3]. However, any single physical reservoir must trade-off between performance metrics including nonlinearity and memory-capacity[4], with the compromise typically hard-coded during nanofabrication.
Here, we present three artificial spin-systems: square artificial spin ice, square artificial spin-vortex ice and a disordered pinwheel artificial spin-vortex ice. We show how tuning system geometry and dynamics defines computing performance. We engineer networks where each node is a high-dimensional physical reservoir[5] implementing parallel, deep and multilayer physical neural network architectures[6]. This solves the issue of physical reservoir performance compromise, allowing a small suite of synergistic physical systems to address diverse tasks and provide a broad range of reprogrammable computationally distinct configurations. These networks outperform any single reservoir across a broad taskset. Crucially, we move beyond reservoir computing to present a method for reconfigurable programming of inter-layer network connections, enabling on-demand task optimised performance.

## References

[1] Andrae, A. S. & Edler, T. On global electricity usage of communication technology: trends to 2030. Challenges 6, 117–157 (2015)
[2] Tanaka, G. et al. Recent advances in physical reservoir computing: A review. Neural Networks 115, 100–123 (2019
[3] Stenning, Kilian D., Gartside, Jack C., Vanstone, Alex. et al. "Reconfigurable training and reservoir computing in an artificial spin-vortex ice via spin-wave fingerprinting." Nature Nanotechnology 17.5 (2022): 460-469.
[4] Inubushi, Masanobu, and Kazuyuki Yoshimura. "Reservoir computing beyond memory-nonlinearity trade-off." Scientific reports 7.1 (2017): 1-10.
[5] Manneschi, Luca, et al. "Exploiting multiple timescales in hierarchical echo state networks." Frontiers in Applied Mathematics and Statistics 6 (2021): 76.
[6] Stenning, Kilian D., et al. "Adaptive programmable networks for in materia neuromorphic computing." arXiv preprint arXiv:2211.06373 (2022).

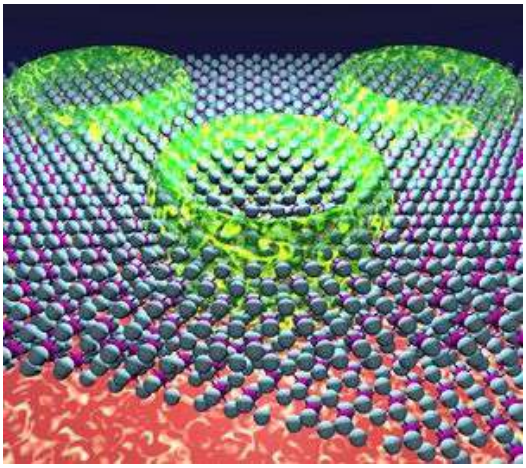
# SYMPOSIUM 06.
## NOVEL 2D MAGNETIC SYSTEMS AND HETEROSTRUCTURES: TOPOLOGICAL MATERIALS AND VALLEYTRONICS.
## S6. INVITED ORAL PRESENTATIONS

**Artificial van der Waals multiferroics with twisted two-dimensional materials**

Jose Lado
*Department of Applied Physics, Aalto University, Finland*

Twisted van der Waals materials have risen as a powerful platform to engineer artificial quantum matter. Artificial moire heterostructures, in general, display two length scales, the original lattice constant and the emergent moire length. Here we reveal a microscopic mechanism to engineer van der Waals multiferroics from the interplay of non-collinear magnetism and spin–orbit coupling, both in van der Waals monolayers [1] and twisted multilayers [2]. First, focusing on the recently isolated NiI2 multiferroic monolayer, we reveal the origin of the helimagnetic order, and the critical role of halide spin-orbit coupling in driving a ferroelectric distortion. We demonstrate that the electronic reconstruction accounting for the ferroelectric order emerges from the interplay of such a non-collinear magnetism and spin-orbit coupling. Second, we show the emergence of multiferroic order in twisted chromium trihalide bilayers, an order fully driven by the moiré pattern and absent in aligned multilayers. We show that a spin texture is generated in the moiré supercell of the twisted system as a consequence of the competition between stacking-dependent interlayer magnetic exchange and magnetic anisotropy. An electric polarization arises associated with such a non-collinear magnetic state due to the spin-orbit coupling, leading to the emergence of a local ferroelectric order following the moiré. Among the stochiometric trihalides, our results show that twisted CrBr3 bilayers give rise to the strongest multiferroic order. We further show the emergence of a strong magnetoelectric coupling, which allows the electric generation and control of magnetic skyrmions. Our results put forward van der Waals materials as a powerful platform to engineer artificial multiferroic order and electrically control exotic magnetic textures.

[1] Adolfo O Fumega and J L Lado 2022 2D Mater. 9 025010 (2022)
[2] Adolfo O. Fumega, Jose L. Lado, arXiv:2207.01416 (2022)



# Graphene Spacer in Topological Insulator $Bi_2Te_3$ /Graphene/NiFe Heterostructures for Efficient Spin-Orbit Torques

*V.Zatko*[1], T.Guillet[1], R.Galceran[1], M.Aoki[2], F.Bonell[3], G.Sauthier[1], MJ.Esplandiu[1], AI Figueroa[1], JF.Sierra[1] and SO.Valenzuela[1]*

[1] *Catalan Institute of Nanoscience and Nanotechnology (ICN2), Campus UAB, Barcelona 08193, Bellaterra, Spain*

[2] *Department of Electronic Science and Engineering, Kyoto University, Kyoto 615-8510, Kyoto, Japan*

[3] *Univ. Grenoble Alpes, CNRS, CEA Spintec, Grenoble 38000, Grenoble, France*

*corresponding author email address

Steady progress in two-dimensional materials (2DMs) offers new perspectives for downscaling and improving MRAM performance [1]. Their 2D nature and weak van-der-Waals interaction between layers enable to create atomically thin stacks with sharp interfaces, circumventing roughness and inter-diffusion between the films, which significantly degrade the spin properties of conventional materials [2]. Furthermore, known 2DMs cover a broad range of relevant properties for spintronics. Among them, graphene shows a very low SOC resulting in long spin diffusion length [3], being of great interest to avoid spin depolarization. The van der Waals materials family also includes topological insulators (TIs) that is an alternative of high SOC materials for charge-to-spin conversion (CSI) with unprecedented magnitudes that would be highly relevant for writing operation in MRAM technologies. Indeed, TIs exhibit conducting topological surface states (TSSs) where the spin of the carriers are locked to their momentum. This results in a highly-efficient generation of spin currents with a polarization that depends on the direction of the charge current.

However, materials disruptions at the interface, from band bending to alloying or air exposure, complicate this picture and the expected SOTs are not obtained.

This study aims in using the TI $Bi_2Te_3$ (BST) as a spin source in order to study its SOT properties. We first describe a step-by-step atmosphere-controlled graphene transfer on top of the TI BST. The coverage of the TI with graphene is checked by Raman spectroscopy and SEM. A complete XPS study is then showed comparing uncovered TI with graphene-protected TI after air exposure. Our data show that graphene preserves the BST chemical bondings that are known to be highly affected by air exposure. ARPES measurements are further confirming the preservation of the TSSs of the TI after graphene passivation that is required in order to maximize its SOT efficiency [4]. The stack is then covered with a NiFe layer forming a BST/graphene/NiFe heterostructure. XPS is carried out on this sample showing that the graphene avoids intermixing between the TI and the FM that is known to be detrimental for SOT. We finally share our measurements of electrical transport including second harmonics measurements of BST/Gr/NiFe stacks that show strong spin-orbit torques estimated to be one order magnitude higher than Co/Pt reference stacks. .

## References

[1] H.Yang, S. O.Valenzuela et al., Nature **606**, 663–673 (2022)

[2] F.Bonell et al., Nano Lett. **20**, 5893 (2020)

[3] B.Dlubak et al., Nature Phys **8**, 557 (2012)

[4] R.Galceran et al. Adv. Mater. Interfaces **9**, 2201997 (2022)

# Mixing disorder in the intrinsic magnetic topological insulators of the MBT/MST family

*Anna Isaeva[1]*, Laura T. Corredor[2], Jorge I. Facio[2,3], Abdul Tcakaev[4], Vladimir Hinkov[4]*

[1] *Institute of Physics, University of Amsterdam, 1098 XH Amsterdam, The Netherlands*
[2] *Leibniz IFW Dresden, D-01069 Dresden, Germany*
[3] *Centro Atomico Bariloche, 8400 Bariloche, Argentina*
[4] *Faculty of Physics and Astronomy, University of Würzburg, D-97074 Würzburg, Germany*
*a.isaeva@uva.nl

Magnetic topological materials (MTI) are a hotbed for exotic quantum phenomena such as the quantum anomalous Hall effect (QAHE), the topological magneto-electric effect, new topological states like axion insulators and magnetic Weyl semimetals. In reply to the high demand for optimized material systems, magnetic topological insulators made a decade-long journey from extrinsically doped $Bi_2Te_3$ and $(Cr,V)Bi_2(Se,Te)_3$ heterostructures, on which the QAHE was experimentally discovered in the mK range [1], to the intrinsically magnetic van der Waals material $MnBi_2Te_4$ [2]. The QAHE was observed in $MnBi_2Te_4$ thin films at notably higher temperature 6.5 K [3], pointing at a perspective pathway of materials optimization towards more robust quantum effects. Since $MnBi_2Te_4$ is an A-type antiferromagnet with $T_N$ = 25 K, the task of fabricating structurally similar ferri- or ferromagnets with an increasing $T_C$ is very pertinent.

In reply to these guidelines, I perform materials optimization in the family of the layered MTIs $(MnX_2Te_4)(X_2Te_3)_n$, $X$ = Sb or Bi, n = 0–4 (MBT/MST). I will discuss the Mn/$X$ site intermixing as a surprisingly powerful tool to tailor the magnetic ground state. Disorder is commonly perceived as detrimental to a material's properties, whereas in the present case it positively affects the magnetic order. The first example is that Mn/Bi site intermixing transforms the magnetic ground state from an antiferro- to a ferromagnet in $MnBi_6Te_{10}$ (Fig. 1, [4]). The second example is that the site mixing is capable of pushing the $T_C$ from 27 K in $Mn_{0.85}Sb_{2.15}Te_4$ to 58 K in $Mn_2Sb_{1.2}Te_2$ thanks to the Mn re-distribution in the same crystalline lattice and the formation of new magnetic exchange pathways. Further Mn-enrichment steered by the growth conditions leads to the formation of $Mn_{2.72}Sb_{1.28}Te_4$ with the highest so far $T_C$ of 73 K. This phase shows a notably higher coercive field than $Mn_{0.85}Sb_{2.15}Te_4$ hinting at a crossover towards more robust ferromagnetism. This enhancement comes at the cost of strong crystal-lattice transformation from a layered van der Waals structure to a 3D cubic framework. The effects on this transformation onto electronic band topology will be discussed.

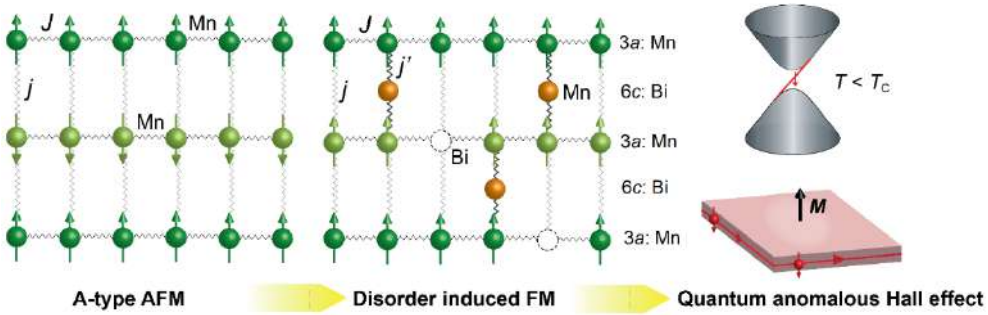



Figure 1: Scheme of the MBT layered structure motif with Mn and Bi atomic planes alternating along the stacking direction (vertical axis). It highlights various magnetic-exchange couplings ($J, j, j'$) possible in the Mn(II) lattice: (left) a non-mixed Mn(II) lattice with the A-type antiferromagnetism (AFM); (middle) an intermixed case with two Mn(II) sublattices and a ferrimagnetic order (FM). (Right) A scheme of the QAHE.

## References

[1] C.-Z. Chang et al. *Science* **340** (2013) 167.
[2] M. Otrokov, … A. Isaeva, E.V. Chulkov. *Nature* **576** (2019) 416.
[3] Y. Deng et al. *Science* **367** (2020) 895.
[4] A. Tcakaev, … A. Isaeva, V. Hinkov. *Adv. Sci.* (2023), doi:10.1002/advs.202203239.

# SYMPOSIUM 06.
## NOVEL 2D MAGNETIC SYSTEMS AND HETEROSTRUCTURES: TOPOLOGICAL MATERIALS AND VALLEYTRONICS.
## S6. ORAL PRESENTATIONS

# Scanning Tunneling Microscope Study of the van der Waals Ferromagnet $CrCl_3$

*Eugenio Gambari*, Sacha Guesne [1], Pascal David [1], François Debontridder[1], Laurent Limot[2], Fabrice Scheurer[2], Chrisophe Brun[1], Tristan Cren[1], Marie Hervé[1]*

[1] *Institut des Nanosciences de Paris, Sorbonne Université and CNRS-UMR7588, Paris*

[2] *Institut de Physique et de Chimie des Matériaux de Strasbourg, Strasbourg*

*eugenio.gambari@insp.upmc.fr

$CrCl_3$ is a van der Waals material particularly interesting for its magnetic properties. In each layer, the Cr atoms are coordinated in an octahedral configuration to the neighboring Cl atoms (Cr-Cl bonds are off-plane and the Cr atoms form a honeycomb lattice). The Cr atoms are coupled ferromagnetically via super-exchange and their magnetic moments lie in-plane. Its magnetic properties have been extensively studied in the bulk form [1], it is a weak ferromagnet with a curie temperature of 17 K. Recently, X-ray Magnetic Circular Dichroism was employed to show that it remains ferromagnetic down to the monolayer, with a Curie temperature very close to the one of the bulk material [2]. Motivated by these findings we have investigated the $CrCl_3$ monolayer on the Au (111) surface. In this communication I will show our Scanning Tunneling Microscopy (STM) investigation.

$CrCl_3$ was deposited by molecular beam epitaxy on the Au (111) surface, where it forms large monolayer islands of a width of several hundreds of nanometers. Our STM study shows that the lattice parameters of $CrCl_3$ are close to the ones of the free layer, indicating a weak structural interaction with the Au substrate. Upon cooling the sample to 4 K, we observed the appearance of a superstructure with a period of about 6 nm. One can see in *Figure 1* that the superstructure displays dislocations with a Burgers vector of 2. We found that the superstructure originates from a peculiar kind of moiré effect between the $CrCl_3$ monolayer and the Au substrate: a second order moiré. This effect has previously been reported only for systems with a large rotation between the two layers [3] and leads to some interesting properties, amongst them the fact that an edge dislocation with B=1 in the $CrCl_3$ crystalline lattice is transformed in a B=2 dislocation of the moiré super-lattice.

## References

[1] de Jongh, L. J. Magnetic properties of layered transition metal compounds; Springer Netherlands, 2012; Vol. 9.

[2] A. Bedoya-Pinto *et al.* "Intrinsic 2D-XY ferromagnetism in a van der Waals monolayer". **Science** - 28 Oct 2021 - Vol 374 - Issue 6567

[3] Emtsev KV, Speck F, Seyller T, Ley L and Riley JD, 2008. Interaction, growth, and ordering of epitaxial graphene on SiC{0001} surfaces: a comparative photoelectron spectroscopy study *Phys.Rev.* B77155303

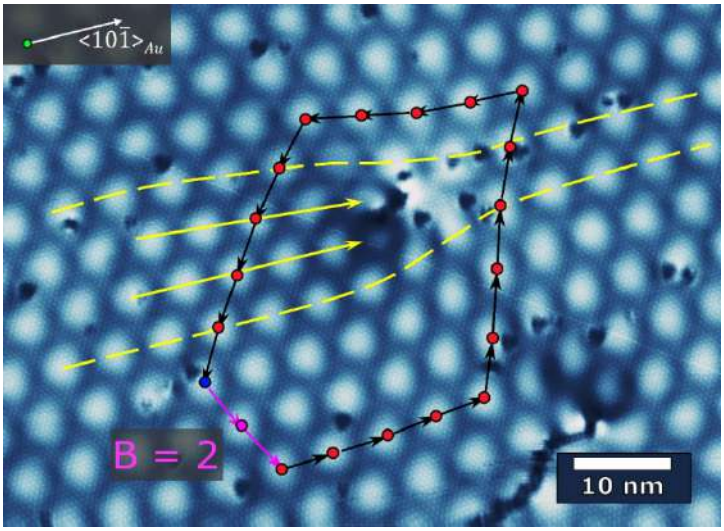



Figure 1: STM constant current topography of $CrCl_3$ monolayer island on Au.

# Hysteresis Opening In Twisted CrSBr Ferromagnetic Monolayers.

*Carla Boix-Constant[1]*, Samuel Mañas-Valero[1,2], Eugenio Coronado[1].*

[1] *Instituto de Ciencia Molecular (ICMol) - Universitat de València, Catedrático José Beltrán 2, Paterna 46980, Spain.*

[2] *Kavli Institute of Nanoscience, Delft University of Technology, 2628 CJ Delft, The Netherlands.*

*carla.boix@uv.es

2D magnetic materials offer unprecedented opportunities for fundamental and applied research in spintronics and magnonics. Of particular interest is the layered metamagnet CrSBr, a relatively air-stable semiconductor formed by antiferromagnetically-coupled ferromagnetic layers ($T_c$~150 K) that can be exfoliated down to the single-layer. It presents a complex magnetic behaviour with a dynamic magnetic crossover, exhibiting a low-temperature hidden order below $T^*$~40 K. In this work, the magneto-transport properties of CrSBr vertical heterostructures in the 2D limit are inspected. The results demonstrate the marked low-dimensional character of the ferromagnetic monolayer, with short-range correlations above $T_c$ and an Ising-type in-plane anisotropy, being the spins spontaneously aligned along the easy axis *b* below $T_c$. By applying moderate magnetic fields along *a* and *c* axes, a spin-reorientation occurs, leading to a magnetoresistance enhancement below $T^*$. In multilayers, a spin-valve behavior is observed, with negative magnetoresistance strongly enhanced along the three directions below $T^*$ [1]. Moreover, we fabricate an artificial magnet by twisting 90 degrees two CrSBr ferromagnetic monolayers, thus forming an 'orthogonally-twisted bilayer' [2]. The magneto-transport properties reveal multistep spin switching with a magnetic hysteresis opening, which is absent in the pristine case. By tuning the magnetic field, we modulate the remanent state and coercivity and select between hysteretic and non-hysteretic magneto-resistance scenariosThese results show that CrSBr monolayer/bilayer provides an ideal platform for studying and controlling field-induced phenomena in two-dimensions, offering new insights regarding 2D magnets and opening a fruitful playground for creating artificial magnetic symmetries and manipulating non-collinear magnetic textures their integration into vertical spintronic devices.

## References

[1] C.Boix-Constant, S.Mañas-Valero, A.M.Ruiz, A.Rybakov, K.A.Konieczny, S.Pillet, J.J.Baldoví, E.Coronado, *Adv. Mater.* **34** (2022) 2204940.

[2] C.Boix-Constant, S.Mañas-Valero, E.Coronado, 10.48550/arXiv.2301.05647.

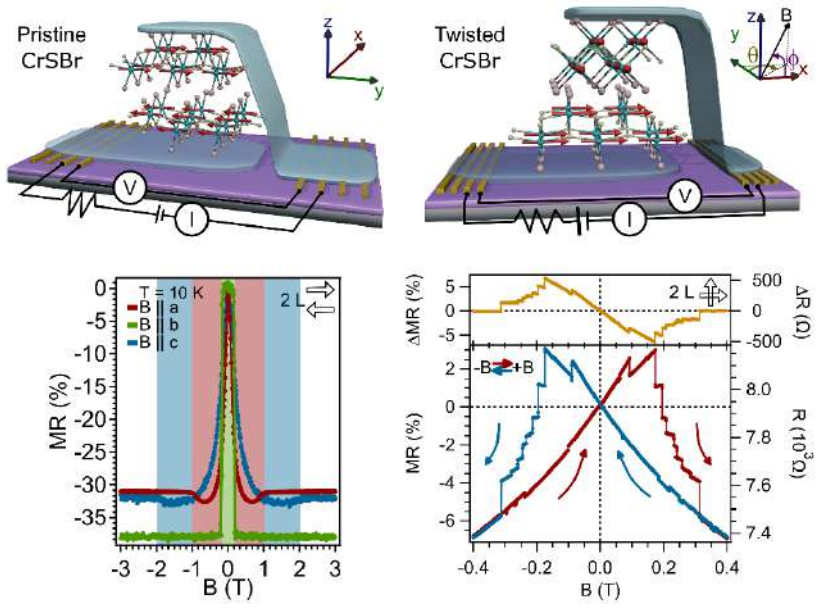



Figure. Left: Sketch of a van der Waals heterostructure with a pristine CrSBr bilayer and the field dependence of its magnetoresistance at 10 K. Right: Sketch of a van der Waals heterostructure with a twisted CrSBr bilayer and the field dependence of its magnetoresistance at 10 K.

# Superconducting Spin-Valve in a Two-Dimensional Bilayer Heterostructure

*Junhyeon Jo[1,*], Yuan Peisen[1], Haozhe Yang[1], Samuel Mañas-Valero[2], José J. Baldoví[2], Yao Lu[3], Eugenio Coronado[2], Fèlix Casanova[1,4], F. Sebastian Bergeret[3,5], Marco Gobbi[1,3,4,*], Luis E. Hueso[1,4,*]*

[1]*CIC nanoGUNE BRTA, Donostia-San Sebastian, Spain*
[2]*Instituto de Ciencia Molecular (ICMol), Universidad de Valencia, Paterna, Spain*
[3]*Centro de Física de Materiales CSIC-UPV/EHU, Donostia-San Sebastian, Spain*
[4]*IKERBASQUE, Basque Foundation for Science, Bilbao, Spain*
[5]*Donostia International Physics Center (DIPC), E-20018 Donostia–San Sebastián, Spain*

*j.jo@nanogune.eu; m.gobbi@nanogune.eu; l.hueso@nanogune.eu

The synergy between magnetism and superconductivity can boost development for diverse device platforms covering digital logic and quantum computing technologies. Introducing superconducting elements into spin-based devices, known as superconducting spintronics [1], would take great advantages of a zero resistance, a dissipationless current, and/or a spin-polarized supercurrent to maximize the figure of merit on magnetic switching devices. One of the representative features is superconducting magnetoresistance in a quasi spin-valve structure [2,3], which exploits the resistance difference between in a superconducting state and a normal state from a magnet/superconductor/magnet structure. Here, we reports superconducting magnetoresistance in a two-dimensional (2D) bilayer composed of a superconductor $NbSe_2$ layer and a transition magnet CrSBr layer. By utilizing unique magnetic switching of a CrSBr flake, between antiferromagnetic and ferromagnetic upon magnetic fields, we can manipulate an effective magnetic field to an adjacent $NbSe_2$ layer which generates superconducting switching in $NbSe_2$ with enormous magnetoresistance. Comprehensive experiment and simulation reveal that a localized stray field for the out-of-plane component at the edge of CrSBr plays a dominant role to induce superconducting switching with support of a proximity effect from the $NbSe_2$/CrSBr interface. The engineered effective field is further proved by means of nonreciprocal charge transport. The demonstration of superconducting magnetoresistance in a 2D bilayer system presents the potential for a superconductor/magnetic structure as a high-end device unit, and it further proposes a novel approach to utilize the unique properties of 2D materials.

## References

[1] J. Linder et al., Nat. Phys. 11, 307 (2015)
[2] B. Li, et al., Phys. Rev. Lett. 110, 097001 (2013)
[3] Y. Zhu, et al., Nat. Mater. 16, 195 (2017)

# Room Temperature Spin-Valve with van der Waals Ferromagnet $Fe_5GeTe_2$/Graphene Heterostructure

*Bing Zhao[1*], Roselle Ngaloy[1], Sukanya Ghosh[2], Soheil Ershadrad[2], Rahul Gupta[2,3], Khadiza Ali[1,4], Anamul Md. Hoque[1], Bogdan Karpiak[1], Dmitrii Khokhriakov[1], Craig Polley[4], Balasubramanian Thiagarajan[4], Alexei Kalaboukhov[1], Peter Svedlindh[3], Biplab Sanyal[2], Saroj P. Dash[1,5]*

[1]*Department of Microtechnology and Nanoscience, Chalmers University of Technology, SE-41296, Göteborg, Sweden.*
[2]*Department of Physics and Astronomy, Uppsala University, Box 516, SE-751 20, Uppsala, Sweden.*
[3]*Department of Materials Science and Engineering, Uppsala University, Box 35, SE-751 03 Uppsala, Sweden.*
[4]*MAX IV Laboratory, Lund University, Lund, Sweden.*
[5]*Graphene Center, Chalmers University of Technology, SE-41296, Göteborg, Sweden.*
*zbing@chalmers.se

The discovery of van der Waals (vdW) magnets opened a new paradigm for condensed matter physics and spintronic technologies. However, the operations of active spintronic devices with vdW ferromagnets are limited to cryogenic temperatures, inhibiting its broader practical applications. Here, we demonstrate the robust room-temperature operation of lateral spin-valve devices using the vdW itinerant ferromagnet $Fe_5GeTe_2$ in heterostructures with graphene. The room temperature spintronic properties of $Fe_5GeTe_2$ are measured at the interface with graphene with a negative spin polarization. Lateral spin-valve and spin precession measurements provide unique insights by probing the $Fe_5GeTe_2$/graphene interface spintronic properties via spin dynamics measurements, revealing multi-directional spin polarization. Density functional theory calculations in conjunction with Monte Carlo simulations reveal significantly canted Fe magnetic moments in $Fe_5GeTe_2$ along with the presence of negative spin polarization at the $Fe_5GeTe_2$/graphene interface. These findings open opportunities for vdW interface design and applications of vdW magnet-based spintronic devices at ambient temperatures [1].

## References

[1] B. Zhao, R. Ngaloy, S. Ghosh, S. Ershadrad, S. P. Dash, et al., Room Temperature Spin-Valve with van der Waals Ferromagnet $Fe_5GeTe_2$/Graphene Heterostructure. *Advanced Materials*, (2023) 2209113.

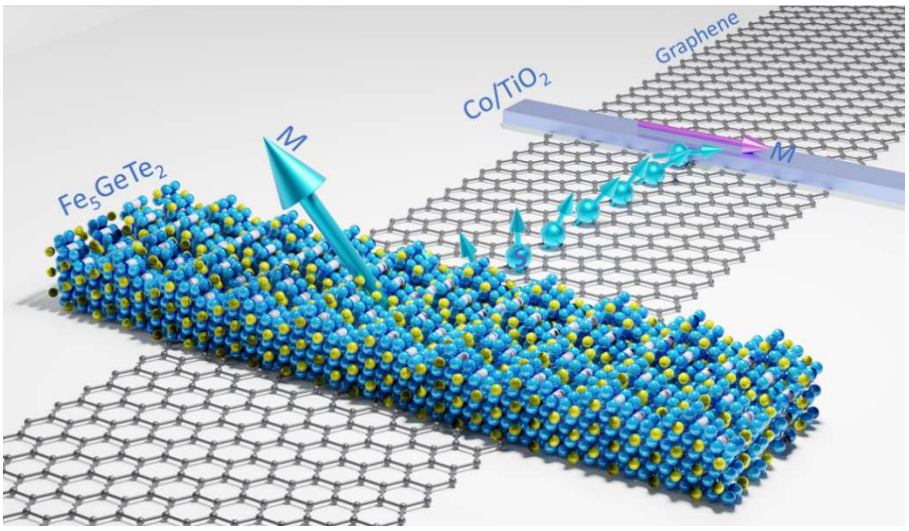



Figure: Schematic of a spin-valve device with $Fe_5GeTe_2$ (FGT) on a graphene (Gr) channel with reference $Co/TiO_2$ electrode. The top-left inset is a schematic illustration for spin injection from FGT into the graphene channel through the vdW gap, inducing a non-equilibrium spin accumulation $\Delta\mu$ in graphene.

# ARPES investigation of a Van der Waals ferromagnet $Fe_5GeTe_2$

Khadiza Ali[1*], Craig Polley[2], Dina Carbone[2], Bing Zhao[2], Frederik schiller[3], Paula A. Portugal[3], Tao Wang[3] Saroj P dash[1].

[1] *Chalmers university of technology, Chalmersplatsen 4, 412 96 Göteborg, Sweden*

[2] *MAXIV Laboratory , Fotongatan 2, 224 84 Lund, Sweden*

[3]*Centro de fisica de materiales, Paseo Manuel de Lardizabal, 5, E-20018 Donostia – San Sebastián (Gipuzkoa), Spain*

* khadiza.ali@maxiv.lu.se

Relation between Electron correlation and magnetism is explored in room temperature Van der Waals ferromagnet Fe5GeTe2 [1]. We observe a enhanced quasiparticle scattering occurring at the spin reorientation transition temperature of 100K. Using angle resolved Photoelectron spectroscopy we show a clear evidence of crossover of electronic states from coherent to incoherenet nature. Further our result unambiguously show that the crossover is directly related to the ordering of Fe1 atoms. It also explains the sign change of the Hall coefficient and the sharp decrease of the Seebeck coefficient due to drastic change in Fermi surface. Our result shows in layered metallic systems effective dimensions governs the electronic properties of the system at different temperature scales.

**Acknowledgements**

We acknowledge MAX IV Laboratory for time on Beamline Bloch under in-house Proposal.

**References**

1. Zhao, Bing, et al. "Room Temperature Spin-Valve with van der Waals Ferromagnet Fe5GeTe2/Graphene Heterostructure." *Advanced Materials* (2023): 2209113.

# Magnon polarons observed in $Fe_3GeTe_2$ by spin-polarized inelastic electron tunnelling spectroscopy

*Namrata Bansal[1], **Qili Li**[1]*, Paul Nufer[1], Hung-Hsiang Yang[1], Lichuan Zhang[2], Dongwook Go[2], Amir-Abbas Haghighirad[3], Yuriy Mokrousov[2,4], Wulf Wulfhekel[1, 3]*

[1] *Physikalisches Institut, Karlsruhe Institute of Technology, 76131 Karlsruhe, Germany*

[2] *Peter Grünberg Institut (PGI-1) and Institute for Advanced Simulation (IAS-1) Forschungszentrum Jülich GmbH, D-52425 Jülich*

[3] *Institute for Quantum Materials and Technologies, Karlsruhe Institute of Technology, 76131 Karlsruhe, Germany*

[4] *Institute of Physics, Johannes Gutenberg-University Mainz, 55099 Mainz, Germany*

*qili.li@kit.edu

Magnon and phonon are collective excitations that can both exist in magnetic materials. Magnon polarons are hybridized magnon-phonon quasiparticles due to magnon-phonon coupling [1-4]. $Fe_3GeTe_2$ is a ferromagnetic two-dimentional (2D) van der Waals material with a Curie temperature up to 230 K [5,6], which renders promises for spintronic devices [7]. Unraveling the exceptional properties in $Fe_3GeTe_2$ will promote its future applications. Here we report magnon polarons in Fe3GeTe2 by spin-polarized inelastic electron tunnelling spectroscopy (IETS). Three low energy excitations below 10 meV are observed in non-spin polarized IETS. They are further confirmed by spin-polarized IETS while presenting strong spin contrast across a domain wall. This demonstrates a spin-dependent character of these exciations. However, they could not stem from either phonon or magnon, because neither density of states (DOS) of phonon nor magnon has a peak below 10 meV by density functional theory (DFT) based calculations. Interstingly, there are low energy band crossings between magnon and phonon. Moreover, the energies of band crossings are consistent with our experimental results. The band crossings will result in van Hove singularities when strong magnon-phonon coupling exists. Therefore, the observed low energy exciations can be identified as magnon polarons. Our results shed light on the potential application for acoustic magnonics in $Fe_3GeTe_2$.

## Acknowledgements

The authors acknowledge funding from the Deutsche Forschungsgemeinschaft (DFG) with grant Wu 394/12-1 and through CRC TRR 288 - 422213477 “ElastoQMat,” project A07 and B06.

## References

[1] Berk, C. *et al.* Strongly coupled magnon–phonon dynamics in a single nanomagnet. Nature communications 10, 2652, doi:10.1038/s41467-019-10545-x (2019).

[2] Vaclavkova, D. et al. Magnon polarons in the van der Waals antiferromagnet $FePS_3$. Phys. Rev. B 104, 134437, doi:10.1103/PhysRevB.104.134437 (2021).

[3] Li, J. et al. Observation of Magnon Polarons in a Uniaxial Antiferromagnetic Insulator. Phys. Rev. Lett. 125, 217201, doi:10.1103/PhysRevLett.125.217201 (2020).

[4] Nambu, Y. et al. Observation of Magnon Polarization. Phys. Rev. Lett. 125, 027201, doi:10.1103/PhysRevLett.125.027201 (2020).

[5] Deiseroth, H. J., Aleksandrov, K., Reiner, C., Kienle, L. & Kremer, R. K. Fe3GeTe2 and Ni3GeTe2 – Two New Layered Transition-Metal Compounds: Crystal Structures, HRTEM Investigations, and Magnetic and Electrical Properties. European Journal of Inorganic Chemistry 2006, 1561-1567, doi:10.1002/ejic.200501020 (2006).

[6] Fei, Z. et al. Two-dimensional itinerant ferromagnetism in atomically thin Fe3GeTe2. Nature materials 17, 778-782, doi:10.1038/s41563-018-0149-7 (2018).

[7] Deng, Y. et al. Gate-tunable room-temperature ferromagnetism in two-dimensional Fe3GeTe2. Nature 563, 94-99, doi:10.1038/s41586-018-0626-9 (2018).

# Breaking through the Mermin-Wagner limit in 2D van der Waals magnets

*S. Jenkins,[1,2,3], L. Rózsa[4], U. Atxitia[5,6]*, R. F. L. Evans[1], K. S. Novoselov[7], E. J. G. Santos[8,9]*

[1] *Department of Physics, University of York, York, YO10 5DD, UK*

[2] *TWIST Group, Institute for Physics, Johannes Gutenberg University, 55128 Mainz, Germany*

[3] *TWIST Group, Institute for Physics, University of Duisburg-Essen, Campus Duisburg, 47057 Duisburg, Germany*

[4] *Department of Physics, University of Konstanz, DE-78457 Konstanz, Germany*

[5] *Dahlem Center for Complex Quantum Systems and Department of Physics, Freie Universität Berlin, 14195 Berlin, Germany*

[6] *Instituto de Ciencia de Materiales de Madrid, CSIC, Cantoblanco, 28049 Madrid, Spain*

[7] *Institute for Functional Intelligent Materials, National University of Singapore, Singapore, 117544, Singapore*

[8] *Institute for Condensed Matter Physics and Complex Systems, School of Physics and Astronomy, The University of Edinburgh, Edinburgh, EH9 3FD, United Kingdom*

[9] *Higgs Centre for Theoretical Physics, The University of Edinburgh, Edinburgh, EH9 3FD, United Kingdom*

*u.atxitia@csic.es

The Mermin-Wagner theorem states that long-range magnetic order does not exist in one- (1D) or two-dimensional (2D) isotropic magnets with short-ranged interactions [1]. Here we show that in finite-size 2D van der Waals magnets typically found in lab setups (within millimetres), short-range interactions can be large enough to allow the stabilisation of magnetic order at finite temperatures without any magnetic anisotropy [2]. We demonstrate that magnetic ordering can be created in 2D flakes independent of the lattice symmetry due to the intrinsic nature of the spin exchange interactions and finite-size effects (see Figure: (Left)). Surprisingly we find that the crossover temperature, where the intrinsic magnetisation changes from superparamagnetic to a completely disordered paramagnetic regime, is weakly dependent on the system length, requiring giant sizes (*e.g.*, of the order of the observable universe ~ $10^{26}$ m) to observe the vanishing of the magnetic order as expected from the Mermin-Wagner theorem (see Figure: (Right)). Our findings indicate exchange interactions as the main ingredient for 2D magnetism.

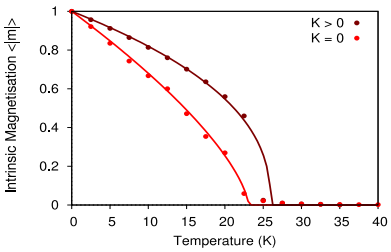
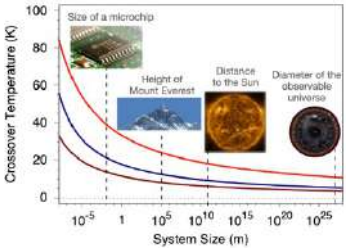



Figure: (Left) anisotropy in temperature lattice




Temperature-dependent intrinsic magnetisation with and without a 1 μm$^2$ flake size. (Right) Crossover as a function of system size for different symmetries on a log-scale.

## Acknowledgements

L.R. gratefully acknowledges funding by the National Research, Development and Innovation Office of Hungary via Project No. K131938. U.A. gratefully acknowledges funding by grants PID2021-122980OB-C55 and RYC-2020-030605-I funded by MCIN/AEI/10.13039/501100011033 and by "ERDF A way of making Europe" and "ESF Investing in your future". E.J.G.S. acknowledges computational resources through CIRRUS Tier-2 HPC Service (ec131 Cirrus Project) at EPCC funded by the University of Edinburgh and EPSRC (EP/P020267/1); ARCHER UK National Supercomputing Service (http://www.archer.ac.uk) *via* Project d429. K.S.N. is supported by the Ministry of Education, Singapore, under its Research Centre of Excellence award to the Institute for Functional Intelligent Materials (I-FIM, project No. EDUNC-33-18-279-V12) and by the Royal Society (UK, grant number RSRP\R\190000).

## References

[1] N. D. Mermin and H. Wagner, *Phys. Rev. Lett.* **17** (1966) 1133-1136.
[2] S. Jenkins, L. Rózsa, U. Atxitia, et al. *Nat. Comm.* **13** (2022) 6917.

# Ferromagnetism on an atom-thick & extended 2D-metal-organic framework

*Jorge Lobo-Checa[1]*, Leyre Hernández-López[1], Mikhail M. Otrokov[2], Ignacio Piquero-Zulaica[3], Adriana Candia[1], Pierluigi Gargiani[4], David Serrate[1], Manuel Valvidares[4], Jorge Cerdà[5], Andrés Arnau[2], and Fernando Bartolomé[1]*

[1]*Instituto de Nanociencia y Materiales de Aragón (INMA), CSIC-Universidad de Zaragoza, Zaragoza 50009, Spain.*

[2]*Centro de Física de Materiales CSIC/UPV-EHU-Materials Physics Center, Manuel Lardizabal 5, E-20018 San Sebastián, Spain.*

[3]*Physics Department E20, Technical University of Munich, 85748 Garching, Germany.*

[4]*ALBA Synchrotron Light Source, E-08290 Cerdanyola del Vallès, Spain.*

[5]*Instituto de Ciencia de Materiales de Madrid, CSIC, Cantoblanco, 28049 Madrid, Spain.*

*jorge.lobo@csic.es

Ferromagnetism is the collective alignment of atomic spins that retain a net magnetic moment below the Curie temperature, even in absence of external magnetic fields. The Mermin-Wagner theorem precludes this collective state mediated by short-range exchange interactions at finite temperatures when downscaling bulk materials into their two-dimensional (2D) magnetic isotropic form [1]. Single layer metal-organic frameworks grown on metallic supports were one of the earliest candidates for achieving such two-dimensional ferromagnetism [1], but remained unachieved till present.

We will show that extended, cooperative ferromagnetism in an atom thick two-dimensional metal-organic framework is feasible [3]. This is remarkable since ≈ 5% of a monolayer of Fe atoms produces an out-of-plane easy-axis square-like hysteresis loop with large coercive fields over 2 Tesla and an extraordinary magnetic anisotropy (see Figure). Such phenomena are driven by exchange interactions mainly through the molecular linkers, presenting a phase transition at $T_C \approx 35$ K. Our findings settle a two decade search for ferromagnetism in two-dimensional metal-organic frameworks and should trigger the research in this and related fields.

We acknowledge MCIN/AEI/10.13039/501100011033/PID2019-107338RB-C64

[1] N. D. Mermin and H. Wagner, Physical Review Letters 17 (1966) 1133.
[2] P. Gambardella et al., Nature Materials 8 (2009) 189.
[3] J. Lobo-Checa et al., arXiv:2209.14994.

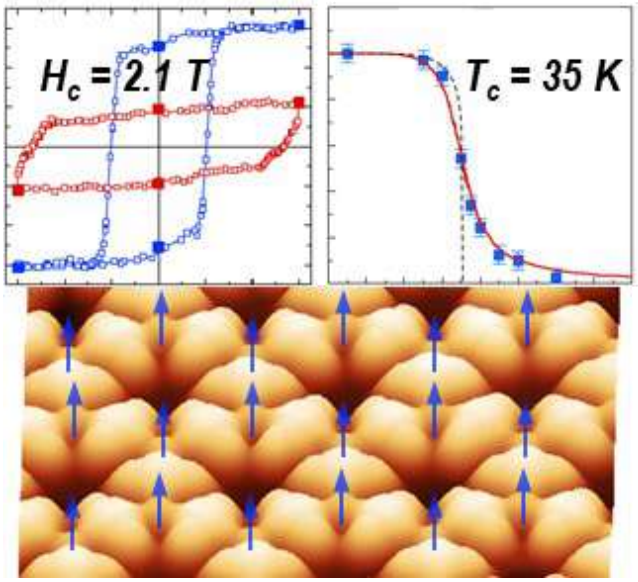



Figure : Ferromagnetic fingerprint of a two-dimensional metal-organic framework

# Modulated Kondo Screening Along Magnetic Mirror Twin Boundaries in Monolayer $MoS_2$ on Graphene

*Camiel van Efferen[1], Jeison Fischer[1], Theo A. Costi[2,3], Achim Rosch[4], Thomas Michely[1], and Wouter Jolie[1]**

[1] *II. Physikalisches Institut, University of Cologne, Germany*
[2] *Peter Grünberg Institut, Forschungszentrum Jülich, 52425 Jülich, Germany*
[3] *Institute for Advanced Simulation, Forschungszentrum Jülich, 52425 Jülich, Germany*
[4] *Institute for Theoretical Physics, University of Cologne, Germany*
*wjolie@ph2.uni-koeln.de

A many-body resonance emerges at the Fermi energy when an electron bath screens the magnetic moment of a half-filled impurity level. This Kondo effect, originally introduced to explain the abnormal resistivity behaviour in bulk magnetic alloys, has been realized in many quantum systems over the past decades. Here we describe a unique Kondo system which allows us to experimentally resolve the spectral function consisting of impurity levels and Kondo resonance in a large Kondo temperature range, as well as the spatial modulation of their wave functions using scanning tunnelling microscopy and spectroscopy [1]. Our experimental Kondo system, based on a discrete half-filled quantum confined state within a $MoS_2$ grain boundary, in conjunction with numerical renormalization group calculations, enables us to test the predictive power of the Anderson model which is the basis of the microscopic understanding of Kondo physics.

## References

[1] C. van Efferen et al., *arXiv:2210.09675*

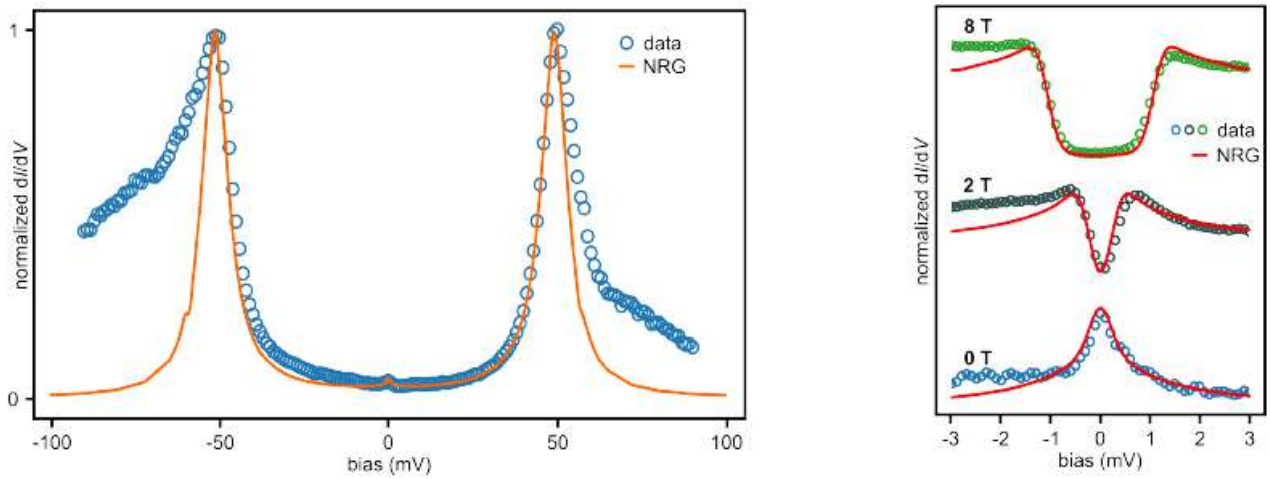



Figure: Kondo resonance within a magnetic MTB. (left) d*I*/d*V* spectrum of the half-filled quantum confined states (blue circles) measured on a mirror twin boundary, together with the corresponding NRG simulation (orange line). (right) d*I*/d*V* spectra showing the Kondo resonance at different magnetic fields (circles), together with the corresponding NRG simulations (lines).

# Interaction Effects in a 1D Flat Band at a Topological Crystalline Step Edge

*Souvik Das[1*], Felix Küster[1], Paolo Sessi[1], Stuart S.P.Parkin[1]*

*[1]Max Planck Institute of Microstructure Physics, Weinberg 2, Halle (Saale), Germany*
*souvdas@mpi-halle.mpg.de

Topological Crystalline Insulators (TCIs) are a class of materials in which the topological nature of the underlying electronic structure is protected by crystalline symmetries [1]. With the realization of a TCI phase in $Pb_{1-x}Sn_xSe$, it was perceived that step edges in TCIs can be viewed as predecessors of higher-order topology, as they embody one-dimensional (1D) edge channels embedded in an effective three-dimensional electronic vacuum emanating from the TCI. Here we use scanning tunneling microscopy and spectroscopy to investigate the behavior of these 1D step-edge channels under the influence of doping. By doping distinct 3d adatoms in $Pb_{1-x}Sn_xSe$ we observed that once the energy position of the step edge is brought close to the Fermi level, a new correlation gap starts to open. Our experimental findings are rationalized in terms of enhanced interaction effects since the electron density of states is collapsed to a 1D channel. This enables us to realize a unique system to study how topology and many-body electronic effects intertwine [2].

**Acknowledgments**
We acknowledge funding from the International Max Planck Research School (IMPRS).

**References**

[1] T. H. Hsieh, H. Lin, J. Liu, W. Duan, A. Bansil, L. Fu, *Nat. Commun.* **3** (2012) 982.
[2] S. Das, G. Wagner, J. Jung, A. Odobesko, F. Küster, F. Keller, J. Korczak, A. Szczerbakow, T. Story, S. S. P. Parkin, Ronny Thomale, Titus Neupert, Matthias Bode, Paolo Sessi, *Nano Lett.* (2023)

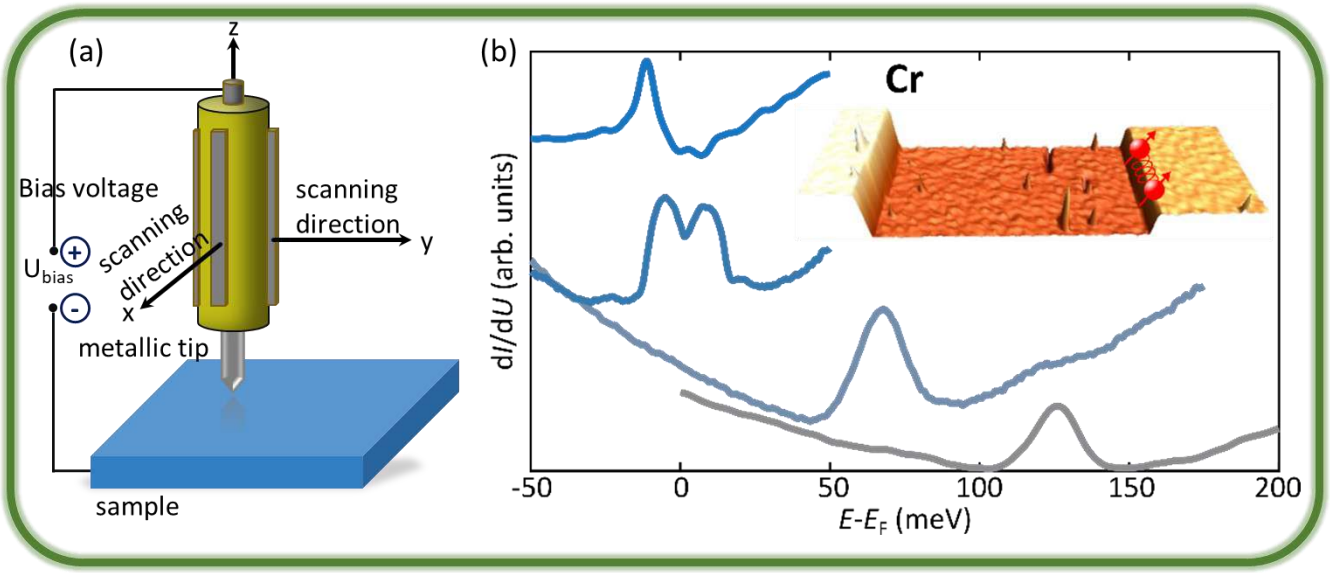



Figure: (a) Schematic of a Scanning Tunneling Microscope, (b) Scanning tunneling spectroscopy of the 1D flat band emerging at half-unit cell step edge as a function of the doping level. A splitting of the single peak in the local density of states into a double-peak structure is visible once the 1D flat band is energetically close to the Fermi level. Inset shows a 3D view of an STM topography of $Pb_{1-x}Sn_xSe$ with strongly interacting electrons at the half-unit cell step edge.

# Magnons, Magnon Bound Pairs, and Their Hybrid Spin-Multipolar Topology

*Alexander Mook[1,2,3], Rhea Hoyer[1,2]*, Jelena Klinovaja[2], and Daniel Loss[2]*

[1]*Institute of Physics, Johannes Gutenberg University Mainz, D-55128 Mainz, Germany*
[2]*Department of Physics, University of Basel, Klingelbergstrasse 82, CH-4056 Basel, Switzerland*
[3]*Department of Physics, Technical University of Munich, James-Franck-Straße 1, D-85748 Garching, Germany*
*rhhoyer@uni-mainz.de

Herein [1], we consider quantum condensed matter systems without particle-number conservation. Since the particle number is not a good quantum number, states belonging to different particle-number sectors can hybridize, which causes topological anticrossings in the spectrum. The resulting spectral gaps support chiral edge excitations whose wavefunction is a superposition of states in the two hybridized sectors. This situation is realized in fully saturated spin-anisotropic quantum magnets without spin conservation, in which single magnons hybridize with magnon bound pairs, i.e., two-magnon bound states. The resulting chiral edge excitations are exotic composites that carry mixed spin-multipolar character, inheriting spin-dipolar and spin-quadrupolar character from their single-particleness and two-particleness, respectively. In contrast to established topological magnons, the topological effects discussed here are of genuine quantum mechanical origin and vanish in the classical limit. We discuss implications for intrinsic anomalous Hall-type transport and estimate that the thermal Hall conductivity brought about by the hybridization of magnons and magnon bound pairs can be as large as that of magnons with other magnons. We conclude that fully polarized quantum magnets are a promising platform for topology caused by hybridizations between particle-number sectors, complementing the field of ultracold atoms working with a conserved number of particles.

## Acknowledgements

This work was supported by the Georg H. Endress Foundation, the Swiss National Science Foundation, NCCR QSIT, and NCCR SPIN. This project received funding from the European Union's Horizon 2020 research and innovation program (ERC Starting Grant, Grant No. 757725) and from the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation)–Project No. 504261060 (Emmy Noether Programme).

## References

[1] A. Mook, R. Hoyer, J. Klinovaja, and D. Loss, *Phys. Rev. B* **107** (2023) 064429 (Editors' Suggestion).

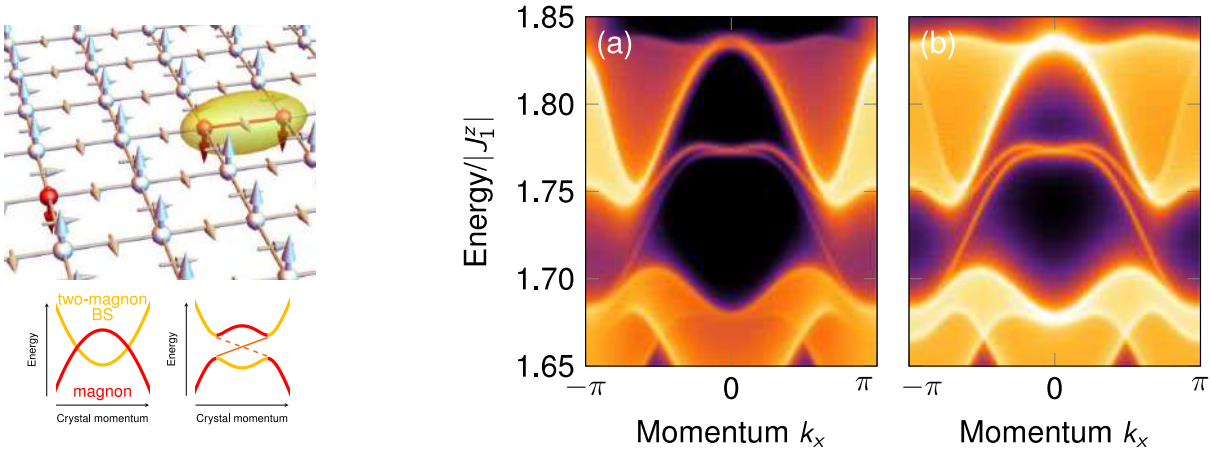



Figure: (Left) Sketch of a quantum condensed matter system with topological hybridizations of states belonging to different particle-number sectors [1]. (Right) Topological chiral edge spin excitations built from single magnons and two-magnon bound states in spin-1/2 square-lattice quantum magnets with strong Ising anisotropy [1].

# An all-proximitized-graphene spin valve with graphene/$Cr_2Ge_2Te_6$ van der Waals heterostructures

*Haozhe Yang[1], Marco Gobbi[1,2,3], Franz Herling[1], Van Tuong Pham[4], Francesco Calavalle[1], Beatriz Martín-García[1,2], Albert Fert[5,6], Luis E. Hueso[1,2], Fèlix Casanova[1,2] **

*1. CIC nanoGUNE, 20018 Donostia-San Sebastián, Basque Country, Spain*

*2. IKERBASQUE, Basque Foundation for Science, 48009 Bilbao, Basque Country, Spain*

*3. Centro de Física de Materiales (CSIC-EHU/UPV) and Materials Physics Center (MPC), 20018 Donostia-San Sebastian, Basque Country, Spain.*

*4. IMEC, Kapeldreef 75, Leuven, Belgium B-3001*

*5. Unité Mixte de Physique/CNRS Thales/Université Paris-Saclay, Palaiseau 91767, France*

*6. UPV/EHU and DIPC, 2018 Donostia-San Sebastian, Basque Country, Spain*

* f.casanova@nanogune.eu

The manipulation and transport of spins is a long-standing topic for spintronics. The generation or detection of spins usually require materials with strong spin-orbit coupling (SOC) or magnetic exchange coupling (MEC), while long-distance spin transport requires materials without these couplings in order to reduce spin relaxation. Optimizing the interfaces between different materials in a spintronic device is thus crucial and easily becomes the bottleneck. One interesting solution is to utilize the proximity effect [1], which can introduce new physical properties into a material system. Here, we design, fabricate, and test a novel van der Waals heterostructure spintronic device based on the proximity of graphene. Proximitized by the ferromagnetic semiconductor $Cr_2Ge_2Te_6$ (CGT), the graphene gains both MEC and SOC, giving rise to spin generation by spin injection and spin Hall effect, respectively. We can distinguish the two contributions by performing non-local spin precession measurements in graphene/CGT-based lateral spin valves. Based on this, we successfully demonstrated an all-proximity graphene spin valve with two separated proximitized regions (shown in Figure (a)) for spin generation and detection. Spin signal by switching the proximitized graphene individually can be seen in Figure (b). The measured signal is confirmed by spin precession shown in Figure (c). Additionally, the anomalous Hall effect of the graphene/CGT heterostructure is observed, as expected from the simultaneous presence of MEC and SOC. Our results show the full-proximity spintronic device is a potential candidate for magnetic memory technologies and spin logic applications.

**Acknowledgements**

We acknowledge funding by the "Valleytronics" Intel Science Technology Center, by the Spanish MICINN (Project No. PID2021-122511OB-I00 and "Maria de Maeztu" Units of Excellence Programme No. CEX2020-001038-M), and by the European Union H2020 under the Marie Sklodowska–Curie Actions (Project No. 0766025-QuESTech) .

**References**

[1] Žutić, I., Matos-Abiague, A., Scharf, B., Dery, H. & Belashchenko, K. *Mater. Today* **22** (2019) 85.

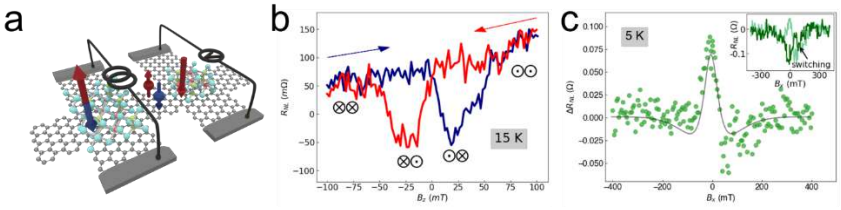



Figure: (a) Schematics of a full proximity lateral spin valve with two graphene/CGT heterostructures. (b) Non-local resistance measured at 15 K and Vg = 50 V by sweeping $B_z$. The blue/red curves represent the trace/retrace direction, showing the binary resistance state for parallel/antiparallel states. (c) Hanle spin precession signal measured at 5 K and Vg = 50 V as a $B_x$. The inset is the raw data with parallel ($R^{P}$ light green line) and antiparallel ($R^{AP}$ dark green line) configuration. The net spin signal calculated with $\Delta R_{NL} = (R^{P} - R^{AP})/2$ is shown in green circles.

# $TaCoTe_2$: A Candidate Magnetic Dirac System with a Large Intrinsic Nonlinear Hall Effect

*Federico Mazzola[1,2], Barun Ghosh[3], Jun Fujii[1], Gokul Acharya[4], Debashis Mondal[1], Giorgio Rossi[1,5], Arun Bansil[3], Daniel Farias[6], Jin Hu[4], Amit Agarwal[7], Antonio Politano[8], and Ivana Vobornik[1,*]*

[1] *Istituto Officina dei Materiali (IOM), CNR, 34149 Trieste, Italy*
[2] *Department of Molecular Sciences and Nanosystems, Ca' Foscari University, 30172 Venice, Italy*
[3] *Department of Physics, Northeastern University, Boston, Massachusetts 02115, United States*
[4] *Department of Physics, University of Arkansas, Fayetteville, Arkansas 72701, United States*
[5] *University of Milano, 20133 Milano, Italy*
[6] *Departamento de Física de la Materia Condensada and Instituto "Nicolás Cabrera" and Condensed Matter Physics Center (IFIMAC),Universidad Autónoma de Madrid, 28049 Madrid, Spain*
[7] *Department of Physics, Indian Institute of Technology Kanpur, Kanpur 208016, India*
[8] *Department of Physical and Chemical Sciences, University of L'Aquila, 67100 L'Aquila, Italy*
*vobornik@iom.cnr.it

In magnetic materials exhibiting topological Dirac fermions, the combined effect of the spin–orbit coupling and magnetic order enables the realization of novel topological phases with exotic transport properties, including the anomalous Hall effect and magneto-chiral phenomena. For this reason they are particularly attractive candidate materials for the developing field of spintronics. Here, via combination of angle-resolved photoelectron spectroscopy and first-principles density functional theory calculations, we report experimental signature of topological Dirac antiferromagnetism in $TaCoTe_2$ [1]. We find the existence of spin–orbit coupling-induced gaps at the Fermi level, consistent with the manifestation of a large intrinsic nonlinear Hall conductivity. Remarkably, we find that the latter is extremely sensitive to the orientation of the Néel vector, suggesting $TaCoTe_2$ as a suitable candidate for the realization of non-volatile spintronic devices with an unprecedented level of intrinsic tunability.

**References:**

[1] F. Mazzola et al., Nano Letters 2023, 23, 3, 902–907

# Evidence of large Dzyaloshinskii–Moriya interaction at the cobalt/hexagonal boron nitride interface

*Banan El-Kerdi* , Andre Thiaville , Stanislas Rohart , Sujit Panigrahy , Nuno Bras, João Sampaio, Alexandra Mougin*

*Université Paris-Saclay, CNRS, Laboratoire de Physique des Solides, 91405 Orsay, France*

*banan.kerdi @université-paris-saclay.fr

A large Dzyaloshinskii–Moriya interaction (DMI) has been predicted to occur at the interface of Co with hexagonal boron nitride (h-BN) [1], despite h-BN being a van der Waals insulator composed of light elements.

In this work, clean Co/h-BN interface were obtained by combining ultra-high vacuum growth and mechanical exfoliation. DMI and PMA were measured using Brillouin light scattering spectroscopy on series of samples of varying Co thickness grown on Pt or Au and covered either with h-BN or Cu. By comparing the h-BN-covered samples with their corresponding control Cu-covered samples, the effect of the Co/h-BN interface was extracted (Fig.1). We thus deduced a $D_S^{Co/hBN}$ = -0.89 pJ/m ($K_S^{Co/hBN}$= 0.84 mJ/m$^2$) and a $D_S^{Co/hBN}$ = -0.21 pJ/m ($K_S^{Co/hBN}$=0.85 mJ/m$^2$) from samples grown on Pt and Au respectively. This reveals that the Co/h-BN interface induces a large PMA and DMI comparable in strength to the largest known effects, such as those of the Pt/Co interface. The strong observed effect despite the weak spin-orbit interaction in h-BN supports a Rashba-like origin of DMI. Furthermore, the sign of DMI in Co/hBN is compatible with that of Pt/Co, resulting in an even larger DMI in Pt/Co/hBN. This enables the stabilisation of skyrmions in the Pt/Co/h-BN system, as was observed at room temperature with low magnetic fields [2], and demonstrates the interest of h-BN as functional material in the field of spintronics.

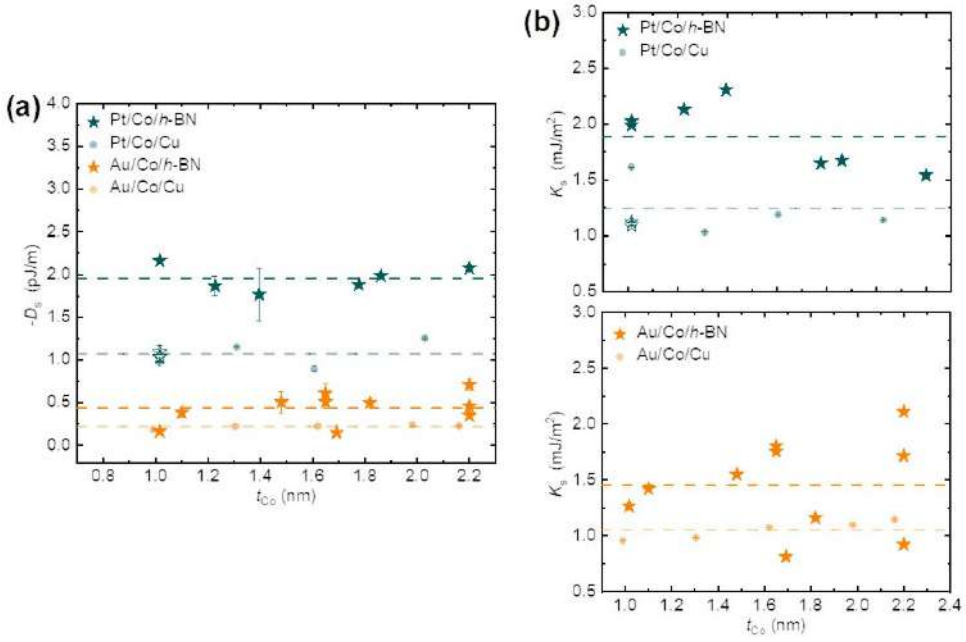



Figure 1: (a) Interfacial DMI strength $D_s$ and (b) interfacial anisotropy strength $K_s$ versus Co thickness ($t_{Co}$) for Pt/Co/h-BN, Pt/Co/Cu, Au/Co/h-BN and Au/Co/Cu. The dashed lines indicate the average values.

## Acknowledgements

This work was supported by the Agence Nationale de la Recherche (ANR) through the project ACAF (ANR-20-CE30-0027), and by the public grant overseen by the ANR as part of the 'Investissement d'Avenir' programme (LABEX NanoSaclay, ANR-10-LABX-0035 SPiCY and ANR-10-LABX-0035 CHIVAWAA).

## References

[1] Hallal et *al.,* Nano Letters 21, 17 7138 (2021)

[2] El-Kerdi et *al.,* arXiv:2212.04768v1, in press (2023)

# Rashba-like spin textures in Graphene promoted by ferromagnet-mediated Electronic-Hybridization with heavy metal

*A. Gudin[1], B. Muñiz[1], Jaime Sanchez-Barriga[1,2], Oliver J. Clark[2], I. Arnay[1], J.-M. Diez[1,3], A. Anadón[1], F. Ajejas[1], P. Olleros-Rodriguez[1], M. Jugovac[4], P. Le Fèvre[5], F. Bertrand[5], D. Mazhjoo[6], S. Blügel[6], G. Bihlmayer[6], M. Valvidares[7], J. Camarero[1,3], R. Miranda[1,3], M.-A. Valbuena[1], and P. Perna[1,*]*

*[1]IMDEA Nanociencia, c/ Faraday 9, Campus de Cantoblanco, 28049 Madrid, Spain*
*[2]Helmholtz-Zentrum Berlin fur Materialien und Energie, Albert-Einstein-Str. 15, 12489 Berlin, Germany*
*[3]DFMC, Instituto Nicolás Cabrera, and IFIMAC, Universidad Autónoma de Madrid, 28049 Madrid, Spain*
*[4]Elettra Sincrotrone Trieste, Strada Statale 14 km 163.5, 34149 Trieste, Italy*
*[5]Synchrotron SOLEIL, Saint-Aubin, Gif-sur-Yvette, 91192, France*
*[6]PGI and IAS, Forschungszentrum Jülich, Jülich, 52425, Germany*
*[7]ALBA synchrotron light source, Cerdanyola del Vallès, 08290 Barcelona, Spain*
* paolo.perna@imdea.org

Real world spintronics applications require the engineering of materials in which spin textures exist at room temperature, are protected from the atmosphere, and amenable to reading and writing and, if possible, integrated with a material than can sustain very long spin diffusion length over long timescale, such as graphene (Gr) [1].

In the last years, epitaxial Gr-based magnetic heterostructures have demonstrated to provide enhanced perpendicular magnetic anisotropy (PMA) and sizeable Dzyaloshinskii-Moriya interaction (DMI), both related to the interfacial Spin Orbit Coupling (SOC), which in turn allow for enhanced thermal stability and stabilization of chiral spin textures [2], enabling the realization of 2D materials spin-orbitronics.

A strong PMA in Gr/Co was first discovered in structures grown onto Ir-single crystals [3] and on both Pt and Ir buffers grown onto insulating oxides [4] and related to the strong anisotropy of the angular magnetic moment [2,5]. Instead, the existence of a unexpected SOC-induced DMI at Gr/FM interface is still under debate since the intrinsic negligible SOC of Gr. However, a DMI was theoretically predicted by M. Chsiev and coworkers [6] and experimentally observed by us [2]. Both works pointed to a Rashba origin of such DMI, but, in our knowledge, no experimental evidence has been provided yet.

Here, we analyze the element dependent and averaged surface/interface SOC induced magnetic properties of epitaxial Gr/FM stacks grown onto HM(Pt, Ir)/$Al_2O_3$(0001). In particular, by means of x-ray absorption, magnetic dichroism (XAS-XMCD) and spin-resolved photoemission spectroscopy (ARPES) experiments accompanied by DFT modelling, we have elucidated the nature of the induced SOC at Gr/Co interface on Ir. We have experimentally found that the interaction of the HM with the C atomic layer via hybridization with 2 ML thick FM is indeed the source of the SOC in the Gr layer. Furthermore, our studies reveal an energy splitting of in-plane spin polarized Gr π bands, consistent with an Rashba-SOC at the Gr/Co interface, which is thus either the fingerprint or the origin of the Dzyaloshinskii Moriya interaction [7]. This is then translated in a measurable induced magnetic moment in Gr. Interestingly, at larger Co thicknesses (~10 ML), neither in-plane or out-of-plane spin splitting is observed, indicating a Gr/Co interface decoupled from the Co/HM.

## Acknowledgements

We acknowledge support from the EU FLAG-ERA SOgraphMEM (Spain, AEI PCI2019-111867-2 and -111908-2, France, ANR-19-GRF1-0001-07, Germany DFG-SOgraphMEM), Community of Madrid (CM) Project P2018/NMT-4321, AEI/MICINN Projects RTI2018-097895-B-C42 and PID2021-122980OB-52, and CEX2020-001039-S.

## References

[1] W. Han, et al. Graphene Spintronics, Nat. Nanotech. 9, 794 (2014).
[2] F. Ajejas, et al., Nano Lett. 18(9), 5364-5372 (2018).
[3] H. Yang et al. Nano Lett. 16, 145 (2016).
[4] F. Ajejas, et al., ACS Appl. Mater. Interfaces, 12(3), 4088-4096 (2020).
[5] M. Blanco-Rey, et al. ACS Appl. Nano Mater. 4(5), 4398–4408 (2021).
[6] H. Yang, et al., Nat. Mater. 17, 605 (2018).
[7] B. Muñiz, A. Gudin et al. Submitted (2023), https://doi.org/10.48550/arXiv.2206.04351.

# Study of the quantum nonlinear Hall effect in $Pb_{1-x}Sn_xTe$ topological crystalline insulator

*M. Ahmad[1*], W. Wołkanowicz[2], S. Dad[2], P. Dziawa[2], T. Wojtowicz[1] and K. Dybko[1,2]*
[1] *1International Research Centre MagTop, Institute of Physics, Polish Academy of Sciences, Aleja Lotnikow 32/46, PL-02668 Warsaw, Poland*
[2] *Institute of Physics, Polish Academy of Sciences, Aleja Lotnikow 32/46, PL-02668 Warsaw, Poland)*
*mahmad@magtop.ifpan.edu.pl

The linear Hall conductivity occurs in the systems, which have broken time-reversal symmetry either by the intrinsic or external magnetic field. However, recently predicted nonlinear Hall effect (NLHE) preserves time-reversal symmetry but it breaks inversion symmetry [1]. NLHE appears from the anomalous velocity of the Bloch electrons due to Berry curvature. Therefore, topological crystalline insulators and the Weyl semimetals are the potential candidates for NLHE due to a Berry curvature dipole [1-3]. The surface of topological crystalline insulators hosts massless Dirac fermions protected by mirror symmetries [2]. At low temperatures, ferroelectric transition caused one of the mirror symmetries to be broken. In the case of thin film, the small mismatch of the lattice parameter can also break the mirror symmetry. The present study reports a NLHE in the $Pb_{1-x}Sn_xTe$ topological crystalline insulator thin films with varying Sn composition. The thin film samples of $Pb_{1-x}Sn_xTe$ with compositions x=0.35 and x=0.42 were grown by molecular beam epitaxy (MBE) on (100) oriented CdTe (4micro m) // GaAs substrates. The nonlinear Hall signal appeared only when the AC excitation was applied along the mirror axis i.e. [110] in our case. The measurement of a nonlinear Hall voltage follows the same geometry as used in ordinary Hall effect, but the transverse voltage is measured at double-frequency, $V_{2\omega}$ and zero-frequency (DC) $V_0$ by using lock-in amplifiers. Both voltages quadratically depend on the perpendicular driving current and decrease when reaching transition from topological to trivial band ordering with rising temperature. The observed phenomenon opens the possibility of exploring topological phase transition as a function of temperature, composition and hydrostatic pressure in topological crystalline insulators.

**Acknowledgements**

This work is supported by the Foundation for Polish Science through the IRA Programme

co-financed by EU within SG OP..

**References**

[1] Sodemann, Inti and Fu, Liang, Quantum Nonlinear Hall Effect Induced by Berry Curvature Dipole in Time-Reversal Invariant Materials, Phys. Rev. Lett. 115,216806 (2015).
[2] P. Dziawa, B. J. Kowalski, K. Dybko, R. Buczko, A. Szczerbakow, M. Szot, E. Łusakowska, T. Balasubramanian, B. M. Wojek, M. H. Berntsen, O. Tjernberg and T. Story, Topological crystalline insulator states in $Pb_{1-x}Sn_xTe$, Nature Materials volume 11, pages1023–1027 (2012)
[3] Kang, K., Li, T., Sohn, E. et al. Nonlinear anomalous Hall effect in few-layer $WTe_2$. Nat. Mater. 18, 324–328 (2019).

# SYMPOSIUM 06.
## NOVEL 2D MAGNETIC SYSTEMS AND HETEROSTRUCTURES: TOPOLOGICAL MATERIALS AND VALLEYTRONICS.
## S6. POSTERS

# Inducing Single Spin-Polarized Flat Bands in Monolayer Graphene

*Matteo Jugovac[1]*, Iulia Cojocariu[1,2], Jaime Sanchez-Barriga[3], Pierluigi Gargiani[4], Manuel Valvidares[4], Vitaliy Feyer[2], Stefan Blügel[5], Gustav Bihlmayer[5], Paolo Perna[3]*

[1*]*Elettra - Sincrotrone Trieste S.C.p.A., 34149 Trieste, Italy*
[2]*Peter Grünberg Institute (PGI-6), Forschungszentrum Jülich GmbH, 52425 Jülich, Germany*
[3]*IMDEA Nanociencia, c/ Faraday 9, Campus de Cantoblanco, 28049 Madrid, Spain*
[4]*ALBA Synchrotron Light Source, 08290 Barcelona, Spain.*
[5]*Peter Grünberg Institut and Institute for Advanced Simulation, FZ Jülich and JARA, 52425 Jülich, Germany*
*matteo.jugovac@elettra.eu

Due to the fundamental and technological implications in driving the appearance of non-trivial, exotic topological spin textures and emerging symmetry-broken phases, flat electronic bands in 2D materials, including graphene, are a hot topic in the field of spintronics. By means of spin-resolved angle-resolved photoemission spectroscopy (ARPES) experiments combined with density functional theory (DFT) calculations, we investigated the role of europium in modifying the spin-dependent electronic properties of monolayer Gr on Co(0001). Manifold effects can be revealed: i) an enhancement of the charge transfer into Gr via Eu doping (Fig. 1 central panel); ii) the existence of a spin-polarized Gr-Co hybrid state formed by positioning Eu on top or beneath the Gr monolayer, in both cases with a single spin (majority) character. While in the former case, the low-dispersive parabolic Gr-Co hybrid band is observed close to Fermi energy (Fig. 1 central panel), extending all over the surface Brillouin zone (SBZ), when Eu is intercalated, the π* band becomes flat (Fig. 1 right panel); iii) the large exchange coupling due to the presence of Eu induces the splitting of the π band that crosses the 4f states into minority and majority branches bending towards higher and lower binding energies respectively, accompanied by a bandgap opening at the Dirac point of about 0.36 eV. These findings demonstrate the relevance of using Eu for efficiently tuning the Gr electronic properties and driving the interlayer magnetic coupling.

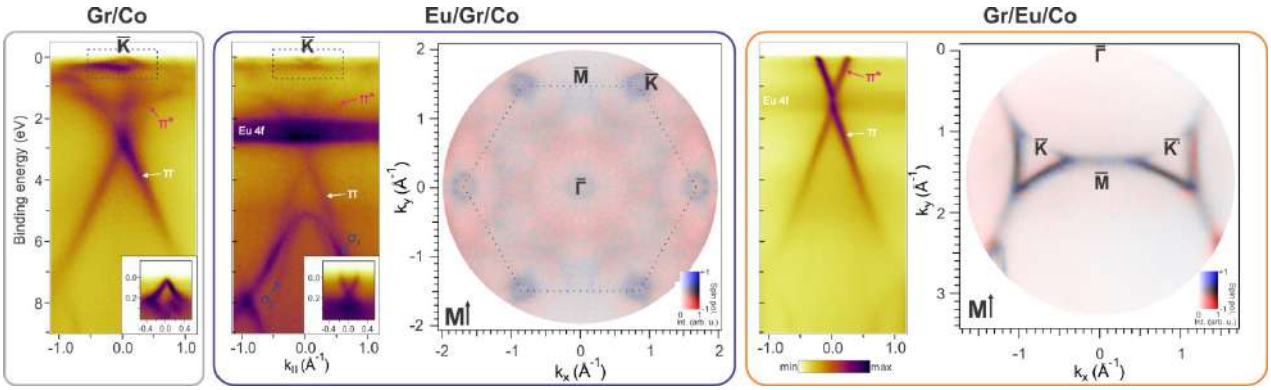



Figure 1: ARPES energy vs. momentum map acquired at the K point of the first Brillouin zone of the graphene/cobalt system (left panel). Energy vs. momentum map acquired at the K point and relative spin-resolved Fermi surface upon deposition of Eu on top of Gr/Co (central panel). Energy vs. momentum map acquired at the K point and relative spin-resolved Fermi surface upon intercalation of Eu at the Gr/Co interface (right panel).

## References

[1] M. Jugovac, I. Cojocariu, J. Sanchez-Barriga, P. Gargiani, M. Valvidares, V. Feyer, S. Blügel, G. Bihlmayer, P. Perna **2023**, *submitted*.

# Direct Magnetic Evidence, Functionalization and Low-Temperature Magneto-Electron Transport in Liquid-Phase Exfoliated $FePS_3$

***Lucía Martín-Pérez*[1]*, *Samara Medina Rivero*[1], *Manuel Vázquez Sulleiro*[1], *Alicia Naranjo*[1], *I. Jénnifer Gómez*[2], *Luisa Ruiz-Gonzalez*[3], *Andres Castellanos-Gomez*[4], *Mar García-Hernández*[4], *Emilio M. Pérez*[1] and *Enrique Burzuri*[1,5]**

[1]*IMDEA Nanociencia C/Faraday 9, Ciudad Universitaria de Cantoblanco, 28049 Madrid, Spain*
[2] *Department of Condensed Matter Physics, Masaryk University, Kotlářská 2, 61137 Brno, Czech Republic*
[3]*Departamento de Química Inorgánica, Universidad Complutense de Madrid, 28040 Madrid, Spain*
[4]*2D Foundry, Instituto de Ciencia de Materiales de Madrid (ICMM-CSIC), 28049 Madrid, Spain*
[5]*Departamento de Física de la Materia Condensada and Condensed Matter Physics Center (IFIMAC), Universidad Autónoma de Madrid, 28049 Madrid, Spain*
*lucia.martin@imdea.org

Magnetism and the existence of magnetic order in a material is determined by its dimensionality. In this regard, the recent emergence of magnetic layered van der Waals (vdW) materials provides a unique playground to explore the exotic magnetism arising in the two-dimensional (2D) limit. The magnetism of 2D flakes has been commonly studied by indirect methods like Raman spectroscopy. Here, taking advantage of liquid-phase exfoliation (LPE) method, we show a first direct magnetic evidence of the antiferromagnetic transition in $FePS_3$ few-layer flakes, performed with a SQUID magnetometer (Figure 1a) [1]. It is concomitant with a clear reduction of the Néel temperatures with the flake thickness, in contrast with previous Raman reports [2].

The quality of the LPE $FePS_3$ flakes allows the study of electron transport down to cryogenic temperatures in field-effect transistors where flakes are deterministically positioned between nanoscale electrodes by dielectrophoresis (DEP) (Figure 1b). The significant through-flake conductance is sensitive to the antiferromagnetic order transition (Figure 1c). Besides, an additional rich spectra of electron transport excitations, including secondary magnetic transitions and magnon states [3], appears at low temperatures (Figure 1d). Finally, we show that the LPE is additionally a good starting point for the mass covalent functionalization of 2D magnetic materials with functional molecules [4]. This technique is extensible to any vdW magnetic family.

## Acknowledgements

We acknowledge funds from Ministerio de Ciencia e Innovación in Spain (RTI2018-096075-A-C22, RYC2019-028429-I).

## References

[1] Martín-Pérez, L. et al., *ACS Nano,* **17** (3), 3007–3018 (2023).
[2] Lee, J.-U. et al., *Nano Lett.*, **16** (12), 7433–7438 (2016).
[3] Klein, D. R. et al., *Science*, **360** (6394), 1218-1222 (2018).
[4] Vázquez Sulleiro, M. et al., *Nat. Chem.*, **14** (6), 695-700 (2022).

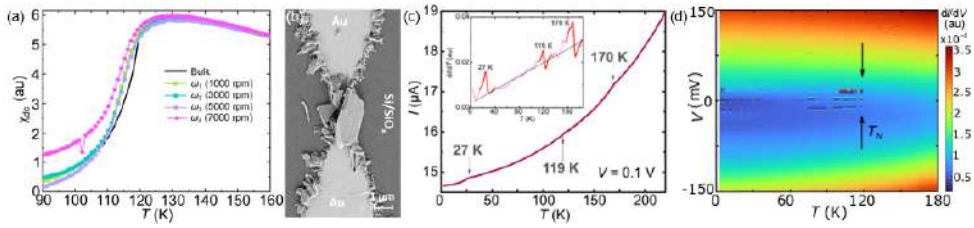



Figure 1: (a) Magnetic susceptibility (χ) measured as a function of temperature in a $FePS_3$ bulk reference sample and exfoliated samples $\omega_1$, $\omega_2$, $\omega_3$ and $\omega_4$. (b) Scanning Electron Microscopy (SEM) image of a representative electrode pair containing LPE $FePS_3$ flakes ($\omega_1$) trapped by dielectrophoresis (DEP). (c) Current $I$ – Temperature $T$ characteristics measured on a $FePS_3$ device at a fixed $V = 0.1$ V and its first derivative ($dI/dT$) (inset). (d) First derivative $dI/dV$ color plot as a function of $V$ and $T$. Low bias excitations are clearly observed at $V = 0$ V and symmetric ±18 mV, right below the Neel´ temperature (marked by the black arrows).

# Gate-tunable spin Hall effect in an all-light-element heterostructure: graphene with copper oxide

*Haozhe Yang[1], Zhendong Chi[1], Eoin Dolan[1], Josep Ingla-Aynés[1], C.K. Safeer[1], Franz Herling[1], Nerea Ontoso[1], Marco Gobbi[1,2,3], Beatriz Martín-García[1,2], Luis E. Hueso[1,2], Fèlix Casanova[1,2*]*

*1 CIC nanoGUNE BRTA, 20018 Donostia-San Sebastian, Basque Country, Spain.*

*2 IKERBASQUE, Basque Foundation for Science, 48009 Bilbao, Basque Country, Spain.*

*3 Centro de Física de Materiales (CSIC-EHU/UPV) and Materials Physics Center (MPC), 20018 Donostia-San Sebastian, Basque Country, Spain.*

* f.casanova@nanogune.eu

Graphene is a promising material for long distance spin transport, being an atomically light material, and thus having low spin-orbit coupling. This low spin-orbit coupling is simultaneously its main drawback where a large spin Hall effect is desired. Decoration by light atoms has been predicted to enhance the spin Hall angle in graphene while retaining a long spin diffusion length. Here, we combine a light-metal oxide (oxidized Cu) with graphene to induce the spin Hall effect and report the first unambiguous experimental observation of charge-to-spin conversion in graphene induced by a light-metal oxide. The $CuO_x$-covered graphene keeps its structural and electrical properties, while acquiring spin-orbit coupling from the oxidized Cu adlayer that leads to SHE up to room temperature, shown in Figure (a). After a careful estimation, considering the gate-dependent spin transport properties of the pristine graphene and the FM contacts, we find that the spin Hall angle of $CuO_x$/graphene is gate tunable, with a maximum value around the charge neutrality point, shown in Figure (b). Its efficiency, given by the product of the spin Hall angle and the spin diffusion length[1], exhibits a maximum ($1.8 \pm 0.6$ nm at 100 K) around the charge neutrality point. This all-light-element heterostructure shows a larger efficiency than conventional spin Hall materials. Our experimental demonstration provides an efficient spin-to-charge conversion system, free from heavy metals, and compatible with large scale fabrication.

**Acknowledgements**

We acknowledge funding by the "Valleytronics" Intel Science Technology Center, by the Spanish MICINN (Project No. PID2021-122511OB-I00 and "Maria de Maeztu" Units of Excellence Programme No. CEX2020-001038-M), by the European Union H2020 under the Marie Sklodowska–Curie Actions (Project Nos. 0766025-QuESTech and 955671-SPEAR) and by Diputación de Gipuzkoa (Project No. 2021-CIEN-000037-01), "Juan de la Cierva" Programme by the Spanish MICINN (grant No. FJC2021-047257-I).

**References**

[1] V. Pham, *et. al.* Nat. Electron. **3** (2020) 309.

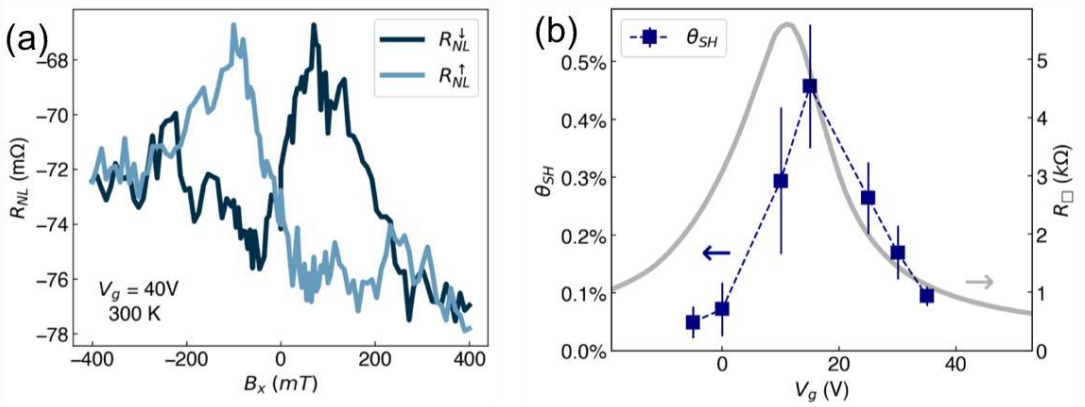



Figure: (a) Anti-symmetric spin precession of graphene/CuOx heterostructure measured with gate voltage of 40 V at room temperature; (b) Gate dependence of spin Hall angle measured at 100 K, indicating a peak around the charge neutrality point of graphene.

# Pure spin pumping in 2D van der Waals materials

*Karen Sobnath[1]*, Mehrdad Rahimi[1], Maria Luisa Della Rocca[1], Philippe Lafarge[1], Clément Barraud[1], François Mallet[1,2]*

[1] *Laboratoire Matériaux et Phénomènes Quantiques – Université Paris Cité*
*10 rue Alice Domon et Léonie Duquet, 75013 Paris, France*
[2] *Sorbonne Université, 4 Pl. Jussieu, 75005 Paris, France*
*karen.sobnath@u-paris.fr

One of the key challenges in spintronics is the efficient generation and detection of spin currents, paving the way for ultra-fast and low power consumption devices for data storage and data processing. While many of the first proposed approaches are suffering from conductance mismatch and power dissipation issues [1], spin pumping provides a powerful route to inject and control dissipationless spin currents without charge current. This phenomena relies on the magnetization dynamic of a ferromagnetic material (FM), brought to its ferromagnetic resonance (FMR) with an external magnetic field, leading to the transfer of spin angular momentum at the interface between the FM and a non magnetic material (NM) [2]. The understanding of the importance of interface effects in these systems, accelerated by the huge progress in nanofabrication techniques, has oriented spintronics towards bidimensional (2D) materials. Indeed, 2D materials can be combined with potentially defect-less interfaces, they cover nowadays all condensed matter phases (from metallic, to semiconducting, insulating, superconducting and even recently magnetic) and present a large sensitivity to external parameters such as proximity effect, electrostatic gating, strain, light, stacking and superlattice effects [3].

In this work, we have investigated spin pumping through different FM/NM material interfaces: Co/graphene/hBN and Co/$WSe_2$/hBN compared to reference samples of Co and Co/Pd, deposited on a 6x10 µm² area on a on-chip microstrip, generating microwaves with frequency ranging from 4 to 14 GHz. We have performed spin pumping ferromagnetic resonance (SP-FMR) measurements at room temperature, and we present the effect of the different interfaces on the magnetic damping of Co.

## References

[1] N. Tombros, C. Jozsa, M. Popinciuc, H. T. Jonkman, B. J. van Wees, *Nature* **448** (2007) 571–574.
[2] Y. Tserkovnyak, A. Brataas, and G. E. W. Bauer, *Phys. Rev. Lett.* **88**, (2002) 117601.
[3] Ethan C. Ahn, *npj 2D Materials and Applications* **4**, (2020) 17.

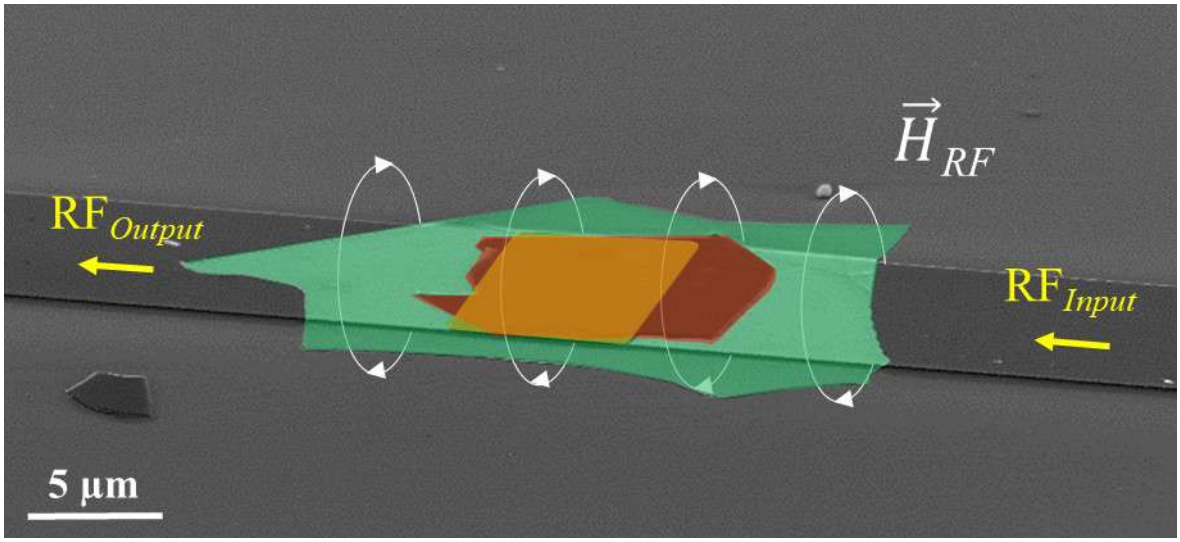



Figure: SEM image of a heterostructure of Co (orange)/$WSe_2$ (red)/hBN (green) deposited on a on-chip Au microstrip (from top to bottom) for SP-FMR measurements.

# Transmission X-ray vector microscopy of weak signal quasy-2D magnetic systems

*A.E. Herguedas-Alonso[1], V.V. Fernandez[2], J. Jurczyk[3] A. Sorrentino[1], E. Pereiro1, A. Fernández-Pacheco[3]J.I. Martín[2,4], M. Vélez[2,4], S. Ferrer1, C. Quirós[2,4], A. Hierro-Rodriguez[2,4]*
*[*1]ALBA Synchrotron Light Source, CELLS, Cerdanyola del Valles, 08290, Barcelona, Spain.*
*[2] Departamento de Física, Universidad de Oviedo, 33007, Oviedo, Spain.*
*[3] INMA (CSIC – Universidad de Zaragoza), 50009, Zaragoza, Spain.*
*[4]CINN (CSIC – Universidad de Oviedo), 33940, El Entrego, Spain.*
*hierroaurelio@uniovi.es

The ability of experimentally characterising the three-dimensional magnetisation field with high lateral resolution is of paramount importance to advance in our comprehension and exploitation of magnetic phenomena. In recent years, several applications of X-ray magnetic vector tomography/laminography have revealed the power of this methodology, allowing to visualise the three-dimensional magnetic textures present in systems from continuous films [1,2] to micro and nanostructures [3-5]. Although the method is devoted to volume resolve the magnetization field, it can be particularised to quasi-2D magnetic systems [6]. This makes it an excellent tool for Spintronics and 2D magnetic Van der Waals communities, where ultra-thin systems in heterostructure configuration are commonly used. However, a potential problem arising in the study of ultra-thin systems is associated to their intrinsic weak magnetic signal. In this poster we analyse the performance of soft X-ray magnetic vector microscopy in heterostructures of $NdCo_5/Ni_{80}Fe_{20}$ with different thicknesses and stacking positions of the permalloy (Py, $Ni_{80}Fe_{20}$) films. Different samples with Py layers of 10, 5 and 2 nm in thickness have been fabricated on top or within the middle of a total 80 nm thick NdCo5 film. The $NdCo_5$ alloy provides weak perpendicular magnetic anisotropy, supporting the formation of magnetic stripe domain patterns for 3D magnetic configurations. By measuring at the Fe $L_3$ edge, the effective material thickness probed corresponds to 2, 1 and 0.4 nm for each fabricated heterostructure. The measurements have been performed in the MISTRAL beamline of the ALBA synchrotron. Two differently oriented tilt series have been recorded allowing to reconstruct the three components of the magnetisation at top and central sample depths. The differences observed in the magnetic textures transferred from the $NdCo_5$ layer to the Py one will be shown. Finally, the influence of thickness and depth position, will be also discussed, including the weak signal effects associated to the low thicknesses of our probe Py system.

## Acknowledgements

Alba light source is funded by the Ministry of Research and Innovation of Spain, by the Generalitat de Catalunya and by European FEDER funds. This work has been supported by Spanish MICINN under grant PID2019-104604RB/AEI/10.13039/501100011033, and by Asturias FICYT under grant AYUD/2021/51185 with the support of FEDER funds.

## References

[1] A. Hierro-Rodriguez, C. Quiros, A. Sorrentino, L.M. Álvarez-Prado, J.I. Martín, J.M. Alameda, S. McVitie, E. Pereiro, M. Vélez and S. Ferrer, *Nature Communications* **11** (2020) 6382.
[2] J. Hermosa-Muñoz, A. Hierro-Rodriguez, A. Sorrentino, J.I. Martín, L.M. Álvarez-Prado, S. Rehbein, E. Pereiro, C. Quirós, M. Vélez and S. Ferrer, *Communications Physics* **5** (2022) 26.
[3] C. Donnelly, M. Guizar-Sicairos, V. Scagnoli, S. Gliga, M. Holler, J. Raabe and L.J. Heyderman, *Nature* **547** (2017) 328-331.
[4] C. Donnelly, K.L. Metlov, V. Scagnoli, M. Guizar-Sicairos, M. Holler, N.S. Bingham, J. Raabe, L.J. Heyderman, N.R. Cooper and S. Gliga, *Nature Physics* **17** (2021) 316-321.
[5] C. Donnelly, A. Hierro-Rodriguez, C. Abert, K. Witte, L. Skoric, D. Sanz-Hernández, S. Finizio, F. Meng, S. McVitie, J. Raabe, D. Suess, R. Cowburn and A. Fernández-Pacheco, *Nature Nanotechnology* **17** (2022) 136-142.
[6] A.E. Herguedas-Alonso, L. Aballe, J. Fullerton, M. Vélez, J.I. Martín, A. Sorrentino, E. Pereiro, S. Ferrer, C. Quirós and A. Hierro-Rodriguez, *Scientific Reports* Submitted.

# A Rigid Pyrimidyl Ligand to Construct of sql and hcp Two-Dimensional Magnets

*Chen-I Yangr*, Jun-Cheng Chen, Chun-Wei Liao*

*Department of Chemistry, Tunghai University, Taichung 407, Taiwan*

*ciyang@thu.edu.tw

The design and synthesis of new coordination polymers (CPs) with paramagnetic metal ions that function as molecule-based magnetic materials have attracted considerable attention and play important roles over recent years. The approach to construct such magnetic material is to utilize appropriate bridging ligands as effective magnetic couplers to link the spin carriers with magnetic anisotropy into the extended networks. Pyrimidyl ligands are receiving considerable attention in the construction of new magnetic CPs. The presence of two potential *N*-donor atoms in the pyrimidyl moiety enables the formation of polymeric assemblies, and combination with additional functional groups could be expected to increase this propensity. In previously, we demonstrated several 3D magnetic CPs with various structures and magnetic properties adapted by a flexible pyrimidyl ligand [1]. Herein, we present the syntheses, crystal structures and magnetic properties of two two-dimensional magnetic CPs by a rigid pyrimidyl ligands with **sql**- and **hcp**-network. The structural diversities, various magnetic and magneto-structural correlations of these CPs have been studied and these results will be reported.

**Acknowledgements**

We thank the Tungahi University and Ministry of Science and Technology, Taiwan for financial support.

**References**

[1] H.-Y. Tang, G.-H. Lee, K.-K. Ng, C.-I Yang, *Polymers*, *14* (2022) 1735.

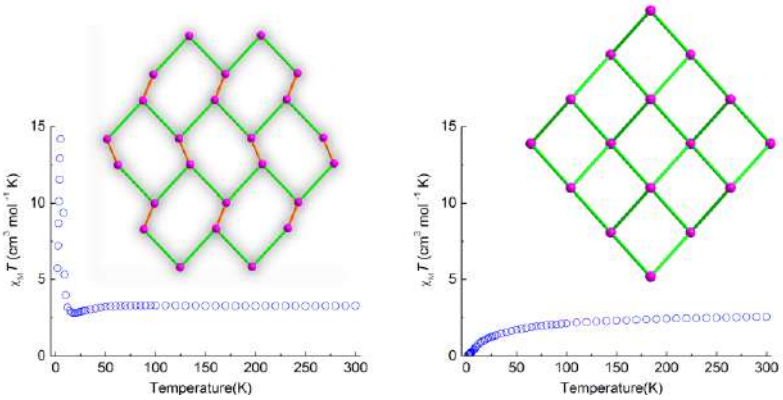



Figure: Temperature-dependence of CMT for 2D sql- and hcp-layed cp

# Magnetism and Excitations in Quasi-2D Vanadium Trihalides

*Karel Carva[1]*, Dávid Hovančík[1], Krishna K. Pokhrel[1], Alberto Marmodoro[2], Jiří Pospíšil[1]*

[1] *Charles University, Faculty of Mathematics and Physics, DCMP, Ke Karlovu 5, 121 16 Prague 2, Czech Republic*

[2] *Institute of Physics ASCR, Na Slovance 2, CZ-18221 Prague, Czech Republic*

*karel.carva@mff.cuni.cz

Systems where two-dimensional (2D) layers bound by weak van der Waals (vdW) interaction possess magnetic order are highly interesting for high-tech magnetic, magneto-electric and magneto-optic applications in nanostructures. While thermal fluctuations suppress magnetic order in reduced dimensions, in $CrI_3$ ferromagnetism has been to shown to exist even in an atomically thin layer. This finding has stimulated a lot work aimed at understanding the underlying physics, and initiated studies of similar transition metal trihalides. Here we focus on vanadium trihalides. $VI_3$ is a ferromagnet whose monolayer critical temperature is even slightly higher than that of bulk, $T_C = 60$ K. DFT calculations converge to two strikingly different solutions: either a ground state with quenched orbital momentum, typical for 3*d* transition metals, or a ground state with exceptionally high orbital momentum [2,3]. Predicted electronic configurations are compared to recent measurements based on the x-ray magnetic circular dichroism [4].

Furthermore we perform a complex investigation of lattice and magnetic excitations of $VI_3$ by employing the synergy of DFT calculations of phonon modes, infrared, terahertz, and Raman spectroscopies on bulk single crystals at low temperatures [5]. The transition to the long-range ferromagnetic order is accompanied by the observed variations of phonons frequencies induced by the strong magnetoelastic coupling. Two additional modes emerging below $T_C$ may be ascribed to magnons, and dramatically soften at temperatures where a second lattice distortion has been reported in the literature.

$VBr_3$ was originally believed to be a layered antiferromagnet, similarly to the intensively studied $CrI_3$. Our calculations predict a different ground state, zig-zag AFM, consistenly with high-field measurements. We also find how the relaxation of halogen atom positions affects the orbital occupation in the ground state, and in turn magnetic interactions and anisotropy. These findings suggest the possibility of controlling magnetic anisotropy in this system by selective occupation of specific lattice modes.

**Acknowledgements**

This work represents a part of Research Project GAČR 21-06083S which is financed by the Czech Science Foundation

**References**

[1] B. Huang, et al., *Nature* **546** (2017), 270

[2] L. M. Sandratskii and K. Carva, *Phys. Rev. B* **103** (2021), 214451

[3] K. Yang, F. Fan, H. Wang, D.I. Khomskii, H. Wu , *Phys. Rev. B* **101**, 100402 (2020).

[4] D. Hovančík et al., *Nano Lett.* **23** (2023) 1175

[5] D. Hovančík et al., *J. Phys. Chem. Lett.* **13** (2022) 11095

# Atomistic Spin Simulation of Magnon Dispersion Relations and Spin Transport in $CrI_3$

*Verena Brehm[1]*, Pawel Sobieszczyk [2] , Richard F.L. Evans[3], Elton J. G. Santos[4] and Alireza Qaiumzadeh[1]*

[1] *QuSpin, NTNU Trondheim, Norway*

[2]*Institute of Nuclear Physics, Polish Academy of Sciences, Krakow, Poland*

[3]*Department of Physics, University of York,, UK*

[4]*Institute for Condensed Matter Physics and Complex Systems, The University of Edinburgh, UK*

*verena.j.brehm@ ntnu.no

As a prominent example of a two-dimensional (2D) van der Waals (vdW) magnet, $CrI_3$ has been studied intensively since its isolation in 2017 [1]. While there is a general consent that this 2D material is a non-Heisenberg compound holding different types of spin interactions (e.g., biquadratic exchange or Kitaev interactions), the exact nature of its magnetic character is still unclear. The opening of a band gap at the Dirac point of the magnon dispersion indicates topological features explained via a variety of models [2,3]. Some groups contribute this topological gap to Kitaev interactions [4], while others assume Dzyaloshinskii–Moriya interactions (DMIs) are the main cause [5].

We simulate the magnon dispersion relation, topological edge modes and spin tranport of $CrI_3$ via atomistic spin dynamics methods [6] recently implemented in the Vampire code [7]. We consider a general spin Hamiltonian consisting of an isotropic Heisenberg exchange interaction, a DMI, a single ion magnetic anisotropy, and a biquadratic exchange. We study the system under different temperatures, magnetic fields, and sample orientations. We conclude that by changing the direction of the external magnetic field, we unveil the role of DMI and Kitaev interactions in the model.

## Acknowledgements

This project has been supported by the Norwegian Financial Mechanism Project No. 2019/34/H/ST3/00515, ``2Dtronics"; and partially by the Research Council of Norway through its Centres of Excellence funding scheme, Project No. 262633, ``QuSpin".

This project was undertaken on the Viking Cluster, which is a high performance compute facility provided by the University of York. We are grateful for computational support from the University of York High Performance Computing service, Viking and the Research Computing team.

This work was supported by the Polish National Science Centre with Grant Miniatura no. 2019/03/X/ST3/01968

## References

[1] Huang et al., Nature, 546:270 (2017)
[2] Chen et al., Phys. Rev. X, 8:041028 (2018)
[3] Chen et al., Phys. Rev. X, 11:031047 (2021)
[4] Lee et al., Phys. Rev. Lett., 124:017201 (2020)
[5] Kartsev et al., npj Comp. Mater., 6:150 (2020)
[6] Nowak, Wiley and Sons (2007)
[7] Evans et al., J. Phys. Condens. Matter, 26:103202

# Electronic, Magnetic, and Transport Properties in Rare-Earth Dichalcogenide

Mahmoud Zeer,[1, 2]* Dongwook Go,[1,3]† Tom G. Saunderson,[3] Mathias Kläui,[3] Jamal Ghabboun,[4] Wulf Wulfhekel,[3] Stefan Blügel,[1] and Yuriy Mokrousov[1, 3, ‡]

[1]Peter Grünberg Institute and Institute for Advanced Simulation, Forschungszentrum Jülich and JARA, 52425 Jülich, Germany

[2]Department of Physics, RWTH Aachen University, 52056 Aachen, Germany

[3]Institute of Physics, Johannes Gutenberg-University Mainz, 55099 Mainz, Germany

[4]Department of Physics, Bethlehem University, Bethlehem, Palestine

∗ m.zeer@fz-juelich.de ; † d.go@fz-juelich.de; ‡ y.mokrousov@fz-juelich.de

We use first-principles calculations to analyze the electronic, magnetic and transport properties of rare-earth dichalcogenides, with a monolayer of the H-phase $EuX_2$ and $GdX_2$ (X=S, Se, Te) taken as a representative [1]. We find that all H-phase of the $EuX_2$ and $GdX_2$ monolayer possess very high magnetic moments and wide bandgaps. Additionally, these systems display very large anomalous Hall conductivity (AHC), spin Hall conductivity (SHC), and orbital Hall conductivity (OHC). Moreover, we observe that the application of biaxial strain can significantly modify the electronic structure of these systems, leading to a significant enhancement of the associated Hall conductivities at the Fermi surface. Our results suggest that rare-earth dichalcogenides are a promising platform for topological spin-orbitronics.

**Acknowledgements**

This work was supported by the Federal Ministry of Education and Research of Germany in the framework of the Palestinian-German Science Bridge (BMBF Grant No. 01DH16027). We also gratefully acknowledge financial support from the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation).

**References**

[1] M. Zeer, D. Go, J. P. Carbone, Phys. Rev. Mater. 6 (2022) 074004.

# Angular Dependence of Spin-Orbit Torques in $Fe_3GeTe_2$

*Anna Liedtke[1]*, Tom G. Saunderson[1,4], Aga Shahee[2], Tanja Scholz[3], Dongwook Go[1,4], Haakon Thømt Siemensen[6], Martin Gradhand[1,5], Yuriy Mokrousov[1,4], Arne Brataas[6], Mathias Kläui[1,6]*

[1]*Institut für Physik, Johannes Gutenberg Universität Mainz, D55099 Mainz, Germany*
[2]*Center for Interdisciplinary Research and Innovation, University of Kashmir, Hazratbal, 190006 Srinagar, India*
[3]*Max Planck Institute for Solid State Research, Heisenbergstraße 1,70569 Stuttgart, Germany*
[4]*Peter Grünberg Institute and Institute for Advanced Simulation Jülich Research Center, 52425 Jülich, Germany*
[5]*University of Bristol, School of Physics, HH Wills Physics Laboratory, Tyndall Avenue, Bristol BS8 1TL, England*
[6]*Center for Quantum Spintronics, Norwegian University of Science and Technology, 7034 Trondheim, Norway*
*anliedtk@students.uni-mainz.de

The recent emergence of magnetic van der Waals (vdW) materials allows for the investigation of current-induced magnetization manipulation in two-dimensional (2D) spintronic devices with high-quality interfaces that do not rely on lattice matching [1],[2]. Magnetic vdW materials with strong spin-orbit torques are potentially very apt for current-induced magnetization manipulation, thus presenting themselves as effective building blocks for next-generation fast and low power spintronic devices [3]. In this family, the ferromagnetic metallic $Fe_3GeTe_2$ is a particularly interesting 2D magnetic material with a high Curie temperature (TC~220 K) [4],[5]. It has a centrosymmetric crystalline structure that allows for monolayer spin-orbit torques (SOTs) [6]. A site preference of Fe vacancies has been reported [6] and this leads to a non-centrosymmetric lattice, which enables bulk SOTs [7].

In this work the SOTs were mapped out as a function of the in-plane magnetic moment angle, $\phi$, using harmonic analysis [8]. We compare the experimental results to the theoretical expression from Johansen et al. [6] obtained by symmetry analysis and find a good agreement. We additionally extend our analysis with first principles calculations to gain a more fundamental insight into the underlying electronic structure. From the comparison between the experimental data and the theory we are able to determine the crystallographic orientation of the flake. We then subsequently address the deviations from theory by analyzing the dependence on current density and temperature. Our findings present a comprehensive scan of the transport properties of this material, bringing us closer to its use in next-generation spintronic devices.

## References

[1] B. Huang, G. Clark, E. Navarro-Moratalla, D. R. Klein, R. Cheng, K. L. Seyler, D. Zhong, E. Schmidgall, M. A. McGuire, D. H. Cobden, W. Yao, D. Xiao, P. Jarillo-Herrero, X. Xu, *Nature* **546** (2017) 270.
[2] C. Gong, L. Li, Z. Li, H. Ji, A. Stern, Y. Xia, T. Cao, W. Bao, C. Wang, Y. Wang, Z. Q. Qiu, R. J. Cava, S. G. Louie, J. Xia, X. Zhang, *Nature* **546** (2017) 265.
[3] A. Manchon, J. Železný, I. M. Miron, T. Jungwirth, J. Sinova, A. Thiaville, K. Garello, and P. Gambardella, *Rev. Mod. Phys.* **91** (2019) 35004.
[4] N. León-Brito, E. D. Bauer, F. Ronning, J. D. Thompson, R. Movshovich, *J. Appl. Phys.* **120** (2016) 83903.
[5] V. Y. Verchenko, A. A. Tsirlin, A. V. Sobolev, I. A. Presniakov, A. V. Shevelkov, *Inorg. Chem.* **54** (2015) 8598.
[6] Ø. Johansen, V. Risinggård, A. Sudbø, J. Linder, A. Brataas, *Phys. Rev. Lett.* 122 (2019) 217203.
[7] A. Chakraborty, A. K. Srivastava, A. K. Sharma, A. K. Gopi, K. Mohseni, A. Ernst, H. Deniz, B. K. Hazra, S. Das, P. Sessi, I. Kostanovskiy, T. Ma, H. L. Meyerheim, S. Parkin, *Adv. Mater.* 34 (2022) 2108637.
[8] F.Martin, K. Lee, M. Schmitt, A. Liedtke, A. Shahee, H. T. Siemensen, T. Scholz, T. G. Saunderson, D. Go, M. Gradhand, Y. Mokrousov, T. Denneulin, A. Kovács, B. Lotsch, A. Brataas, M. Kläui, *Mat. Res. Lett.* **11** (2023) 84.

# Proximity magnetization of intrinsic magnetic topological insulator $MnSb_2Te_4$ by a ferromagnetic insulator

*O. Rader[1]*, Chen Luo[1,2], F. Radu[1], E. Schierle[1], O. Caha[3], Jinjun Ding[4], V. Kalappattil[4], C. Back[2], Mingzhong Wu[4], and G. Springholz[5]*

[1] *Helmholtz-Zentrum Berlin für Materialien und Energie, 12489 Berlin, Germany*
[2] *Fakultät für Physik, Technische Universität München, 85748 Garching, Germany*
[3] *Department of Condensed Matter Physics, Masaryk University, Brno 61137, Czech Republic*
[4] *Department of Physics, Colorado State University, Fort Collins, CO 80523, USA*
[5] *Institut für Halbleiter- und Festkörperphysik, Johannes Kepler Universität, 4040 Linz, Austria*

*rader@helmholtz-berlin.de

The quantum anomalous Hall (QAH) effect was experimentally realized in $(Bi,Sb)_2Te_3$ epitaxial films doped by Cr [1]. As an alternative to magnetic doping, stoichiometric magnetic topological insulators have been considered. Mn doping leads to the formation of a natural heterostructure comprising quintuple $Bi_2Te_3$ and septuple $MnBi_2Te_4$ layers in which the magnetic gap can be directly observed spectroscopically [2-5]. Recently, a nearly quantized anomalous Hall effect has been observed up to 7 K for a $MnBi_2Te_4/Bi_2Te_3$ heterostructure [6]. While pure $MnBi_2Te_4$ is antiferromagnetic, the coupling between the septuple layers of $MnSb_2Te_4$ becomes ferromagnetic in polycrystalline samples [7] and in epitaxial films [8]. A slight Mn enrichment has been identified as the reason for this coupling as well as for the comparatively high Curie temperature $T_C$ of 50 K reached in this system [8].

In order to enhance $T_C$ and possibly the operating temperature of the QAH effect, magnetic topological insulators have been proximity magnetized by ferromagnetic insulators. Cr-doped $Sb_2Te_3$ can introduce long-range magnetic order in Dy-doped $Bi_2Te_3$ up to 17 K [9]. In multilayers of this system, also the $T_C$ of the Cr-doped $Sb_2Te_3$ was found to be increased by 60 K due to the magnetic proximity [10]. The ferrimagnetic insulator $Y_3Fe_5O_{12}$ provides a high $T_C$ of ~550 K and out-of-plane anisotropy. It has been used for proximity magnetization of Cr-doped $Bi_2Se_3$ which reached a $T_C$ of ~50 K, enhanced from a maximum $T_C$ of the topological insulator film of about 20 K [11].

Films of the ferrimagenetic insulator barium hexaferrite ($BaFe_{12}O_{19}$) have a large magnetocrystalline anisotropy and an effective perpendicular anisotropy field of about 22 kOe. A charge current in $Bi_2Se_3$ can switch the magnetization in an adjacent $BaFe_{12}O_{19}$ film [12]. In the present work, we investigate the proximity magnetization of $MnSb_2Te_4$ by $BaFe_{12}O_{19}$. We demonstrate by x-ray magnetic circular dichroism for 2 septuple layers (SL) a strong proximity magnetization at low temperature which persists at room temperature. The magnetic coupling is antiparallel. Data for 4 SL allow to determine the spacial extent of the proximity coupling. The results will be compared to density functional theory calculations.

**References**

[1] C.-Z. Chang, J. Zhang, X. Feng, J. Shen, Z. Zhang, M. Guo et al., *Science* **340**, 167 (2013).
[2] E. D. L. Rienks, S. Wimmer, J. Sánchez-Barriga, O. Caha et al., *Nature* **576**, 423 (2019).
[3] I. I. Klimovskikh, M. M. Otrokov, D. Estyunin et al., *npj Quantum Materials* **5**, 54 (2020).
[4] R. Lu, H. Sun, S. Kumar, Y. Wang, M. Gu, M. Zeng et al., *Phys. Rev. X* **11**, 011039 (2021).
[5] C. X. Trang, Q. Li, Y. Yin, J. Hwang, G. Akhgar, I. Di Bernardo et al., *ACS Nano* **15**, 13444 (2021).
[6] H. Deng, Z. Chen, A. Wolos, M. Konczykowski, K. Sobczak et al., *Nat. Phys.* **17**, 36 (2020).
[7] T. Murakami, Y. Nambu, T. Koretsune, Gu Xiangyu, T. Yamamoto et al., *Phys. Rev. B* **100**, 195103 (2019).
[8] S. Wimmer, J. Snchez-Barriga, P. Küppers, A. Ney et al., *Adv. Mater.* **42**, 2102935 (2021).
[9] L. B. Duffy, N.-J. Steinke, D. M. Burn, A. Frisk et al., *Phys. Rev. Materials* **2**, 054201 (2018).
[10] L. B. Duffy, N.-J. Steinke, D. M. Burn, A. Frisk et al., *Phys. Rev. B* **100**, 054402 (2019).
[11] Wenqing Liu, Liang He, Yongbing Xu, K. Murata, M. C. Onbasli et al., *Nano Lett.* **15**, 764769 (2015).
[12] Peng Li, J. Kally, Steven S.-L. Zhang, T. Pillsbury, Jinjun Ding et al., *Sci. Adv.* **5**, eaaw3415 (2019).

# Electronic, Magnetic, and Transport Properties of the Topological Insulator $MnBi_2Te_4$ through First-Principles Calculations

*Mirali Jafari[1]*, Martin Gmitra[2,3], Anna Dyrdał[1]*

[1] *Department of Mesoscopic Physics, ISQI, Faculty of Physics, Adam Mickiewicz University in Poznań, ul. Uniwersytetu Poznaskiego 2, 61-614 Poznań, Poland*

[2] *Institute of Physics, Faculty of Science, Pavol Jozef Šafárik University in Košice, Park Angelinum 9, 040 01 Košice, Slovakia*

[3] *Institute of Experimental Physics, Slovak Academy of Sciences, Watsonova 47, 040 01 Košice, Slovakia*

*mirali.jafari@amu.edu.pl

$MnBi_2Te_4$ is a unique material with non-trivial topological insulator properties and potential applications in spintronics, quantum computing, and other related fields. A combination of theoretical and experimental investigations has revealed that it exhibits several interesting electronic, magnetic, and transport properties.

Using the density functional theory (DFT), we investigated the electronic band structure and magnetic properties of bulk and surface states of $MnBi_2Te_4$. The results reveal that the material is a topological insulator with a significant bandgap of 0.57 eV in the bulk, and 0.1 eV in the Dirac points at the surface -- with and without spin-orbit coupling. The electronic structure exhibits a Dirac cone-like dispersion, with the Fermi level crossing the Dirac points. Moreover, the metallic surface states appear at the Dirac $\Gamma$ point, which is in good agreement with the pervious results [1-3].

The magnetic properties of $MnBi_2Te_4$ arise from the coupling between the magnetic moments of Mn atoms and the non-magnetic $Bi_2Te_4$ layers. This material exhibits a high magnetic anisotropy, with the easy-axis along the c-axis. The Curie temperature of $MnBi_2Te_4$ has been reported to be around 20 K, which is relatively low compared to other magnetic materials [4].

Transport properties of $MnBi_2Te_4$ have also been investigated experimentally, and theoretical calculations have provided insights into the mechanism of transport in this material. It has been shown that the electrical conductivity of $MnBi_2Te_4$ is highly anisotropic, with the conductivity along the c-axis much higher than that in the ab-plane. This anisotropy is attributed to the layered structure of the material and strong coupling between the Mn atoms and the $Bi_2Te_4$ layers. In addition, the application of an electric field to $MnBi_2Te_4$ has been found to induce a bandgap splitting in the electronic structure. This effect can be utilized in the design of spin-FETs and other spin-based devices [5].

However, the growth of high-quality $MnBi_2Te_4$ thin films and the optimization of their properties remain challenging tasks. Various methods, such as molecular beam epitaxy and pulsed laser deposition, have been employed to fabricate $MnBi_2Te_4$ thin films, but further research is needed to improve the quality and to optimize the growth conditions.

**Acknowledgements**

This work has been supported by the Norwegian Financial Mechanism 2014–2021 under the Polish-Norwegian Research Project NCN GRIEG “2Dtronics” no. 2019/34/H/ST3/00515.

**References**

[1] D. Zhang, et al., Phys. Rev. Lett. 122 (2019) 206401.
[2] M. M. Otrokov, et al., Nature 576 (2019) 416-422.
[3] J. Jiang, et al., Phys. Rev. Mater. 2 (2018) 024201.
[4] Y. Li, et al., Phys. Rev. B 100 (2019) 134438.
[5] S. Patel, et al., Phys. Rev. B 106 (2022) 035125.

# Topological transport in twisted graphene deposited on transition metal dichalcogenides TMDCs

*I.Wojciechowska*[1]*, *A.Dyrdał*[1]

[1] *Department of Mesoscopic Physics, ISQI, Faculty of Physics, Adam Mickiewicz University in Poznań, ul. Uniwersytetu Poznanskiego 2, 61-614 Poznan, Poland*

*izawoj6@st.amu.edu.pl

The discovery of unexpected phase transitions in magic angle bilayer graphene led to further investigation of angle-dependent properties of two-dimensional materials. The development of van-der-Waals structure engineering and advanced modelling of various proximity effects in multilayer hybrid structures (that may include TMDCs and graphene) allowed us to explore the dependence of proximity-induced spin-orbit coupling on angle in the case of graphene deposited on transition metal dichalcogenides.

We will present a theoretical study of topological transport properties of twisted graphene on $MoS_2$, $WS_2$, $MoSe_2$, and $WSe_2$ based on an effective modelled Hamiltonian derived from symmetry considerations and DFT study [1]. We analyse among others behaviour of Berry curvature as a function of angle and characteristic parameters defining the Hamiltonian and discuss possible phase transitions of proposed heterostructures. Moreover, we present detailed characteristics of intrinsic anomalous, spin and valley Hall effects [2] that may appear in specific phases.

## Acknowledgements

This work has been supported by the Norwegian Financial Mechanism 2014- 2021 under the Polish-Norwegian Research Project NCN GRIEG "2Dtronics" no. 2019/34/H/ST3/00515.

## References

[1] Thomas Naimer, Klaus Zollner, Martin Gmitra, JPhys. Rev. B 104, 195156 (2021)
[2] A.Dyrdał and J.Banaś, 2D Materials 4, 034003 (2017)

# Electronic and Topological Properties of a Thin Film of a Topological Insulator Sandwiched between Ferromagnets

*Piotr Pigoń*[1], *Bartłomiej Spisak*[1], *Anna Dyrdał*[2]

[1] *AGH University of Science and Technology, Faculty of Physics and Applied Computer Science, Mickiewicza 30, 30-059 Kraków, Poland*

[2] *Department of Mesoscopic Physics, Faculty of Physics, Adam Mickiewicz University in Poznań, ul. Uniwersytetu Poznańskiego 2, 61-614 Poznań, Poland*

*ppigon@agh.edu.pl

Two-dimensional electron systems, like surface states in a topological insulator, may play an important role in future low-dimensional spintronics, nano-electronics, and information technology. The importance of these materials results mainly from their unique electronic properties that open new possibilities for various quntum phanomena [1].

In this presentation we will show our results on transport properties (eg., anomalous Hall effect) in a thin film of a topological insulator sandwiched between two ferromagnetic layers. This configuration enables the control of proximity-induced magnetisation of the surface states and, therefore, control of changes in transport characteristics when the magnetisations of the two layers vary between ferro- and antiferromagnetic alignment. Additionally, we analyse the dependence of transport properties on the hybridisation of the surface states localised at the two surfaces, which appears when the layer of a topological insulator is sufficiently thin. The transport characteristics are determined in the framework of the Kubo formula. Moreover, we have also found Berry curvature and topological invariants.

**Acknowledgements**

This work has been supported by the Norwegian Financial Mechanism 2014–2021 under the Polish-Norwegian Research Project NCN GRIEG “2Dtronics” no. 2019/34/H/ST3/00515.

**References**

[1] A. G. Moghaddam, A. Qaiumzadeh , A. Dyrdał , and J. Berakdar, „Highly Tunable Spin-Orbit Torque and Anisotropic Magnetoresistance in a Topological Insulator Thin Film Attached to Ferromagnetic Layer”, Phys. Rev. Lett. 125, 196801 (2020)

# SYMPOSIUM 07.
## NOVEL 2D MAGNETIC NANOPARTICLES AND MULTIFUNCTIONAL MAGNETIC.
## S7. INVITED ORAL PRESENTATIONS

# From salt crystals to highly ordered $L1_0$ nanoparticles: an *in-situ* XAS study

*S. Laureti[1*], F. D'Acapito[2], P. Imperatori[1], E. Patrizi[1], G. Varvaro[1], A. Puri[2], C. Cannas[3] and A. Capobianchi[1]*

[1] *Istituto di Struttura della Materia, CNR, nM2-Lab, Monterotondo Scalo (Roma), 00015, Italy*
[2] *Istituto Officina dei Materiali -OGG , CNR c/o ESRF, LISA CRG, F-38043 Grenoble, France*
[3]*Department of Chemical and Geological Sciences, University of Cagliari, S.S. 554 Bivio per Sestu, 09042 Monserrato, Italy*
*sara.laureti@ism.cnr.it

Chemically ordered fct ($L1_0$) alloys belong to a class of materials whose peculiarity relies in the strict correlation between the atomic arrangement in the lattice and the resulting chemical and physical properties. As an examples, many applications of magnetic materials are based on the use of $L1_0$ equiatomic alloys (FePt, CoPt, FeNi, MnAl, MnPt) consisting of planes of pure atoms alternating along the c-axis of the face centered tetragonal (fct) unit cell. The higher the chemical order, the better the material performance.

Standard synthesis approaches are usually based on finding suitable conditions to favor a structural transition from the disordered phase (A1) to the ordered one ($L1_0$). However, in the last years innovative strategies have been proposed, aimed at promoting the order under increasingly mild conditions.

Among the successful approaches, an effective, single-step, easily scaled-up synthesis route, called Pre-ordered Precursor Reduction (PPR), exploits the intrinsic natural order of crystalline precursor salts resembling the alternate structure of the ordered alloy, to synthesize highly-ordered $L1_0$ nanoparticles by low-temperature treatment in reductive atmosphere [1].

A recent study allowed at highlighting, for the first time, the elemental and structural evolution during the PPR process thanks to a comprehensive *in-situ* X-ray Abosrption Spectroscopy analysis applied during the treatment on three different precursor compounds. Although not fully retained for all the investigated systems, the high atomic order naturally occurring in the initial precursor salt, is considered as a fundamental driving force to kinetically favor the formation of the ordered phase. For this reason, the *PPR* method is a potential synthesis approach that can be further extended, by properly selecting the precursor salt, to the synthesis under milder conditions than traditional thermal treatments of many other binary and ternary alloys, where the chemical order represents an indispensable property of the materials, as in the case of the $L1_0$-FeNi, $L1_0$-MnAl and $L1_0$-MnPt magnetic systems, which are of great interest for many applications including permanent magnets and spintronic devices.

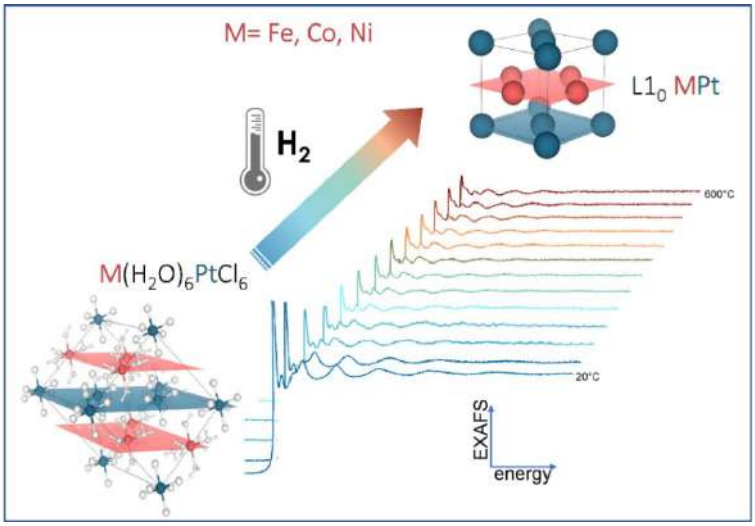



Figure: Sketch of the Pre-ordered Precursor Reduction Strategy for the synthesis of $L1_0$ MePt nanoparticles

## References

[1] G. Varvaro et al., *J. Alloys Compd.* **846** (2020) 156156.

# Structural, magnetic, and electronic characterization of nanoparticles at the atomic and nanoscale level

*A. Kleibert*

*Swiss Light Source, Paul Scherrer Institut, 5232 Villigen PSI, Switzerland*

armin.kleibert@psi.ch

Future technologies ranging from catalysis to spintronics and energy applications will depend on the functionalization of nanoparticle systems with tailored properties [1]. For this purpose, predicting and controlling the properties of nanoparticles is crucial. Immense progress in this area has been made possible by a better understanding of the role of particle size, shape, structure, and the interactions between the nanoparticles and the substrate. This knowledge has been achieved by significant developments in characterization techniques that enable one to probe the properties of individual nanoparticles down to the atomic scale.

In this contribution, we will present our recent investigations on *3d* transition metal and metal oxide nanoparticles that take advantage of the unique capabilities of soft x-ray spectromicroscopy combined with high resolution structural characterization to directly correlate magnetic, electronic and structural properties of individual nanoparticles. Using this approach, we have demonstrated that ferromagnetically ordered pure iron and cobalt nanoparticles can coexist in different states with distinct magnetic properties irrespective of their size, showing that simple scaling laws cannot predict the magnetic properties of these particles [2,3]. We have further studied the manipulation of these magnetic states in individual nanoparticles by means of single femtosecond laser pulse excitation [4]. In *in situ* experiments, we have further investigated the chemical reaction of individual cobalt nanoparticles with molecular oxygen and found that the early stage of oxidation occurs through the nucleation and growth of small oxide crystallites across the nanoparticle surface instead of the anticipated growth of a uniform oxide shell. This oxidation mechanism has implications for their chemical reactivity and the microstructural properties determining magnetic phenomena such as the exchange bias and for their application as nano-catalysts and in spintronics [5]. Finally, we will present new results on the magnetic structure in antiferromagnetically ordered *3d* transition metal oxide nanoparticles by employing a recently developed soft x-ray ptychography microscope. Our results provide an important benchmark for an improved understanding of the chemical, electronic, and magnetic properties of nanoparticles.

## References

[1] K. D. Gilroy et al., *Chem. Rev.* **45**, (2016) 10414.
[2] A. Balan et al., *Phys. Rev. Lett.* **112**, (2014) 107201.
[3] A. Kleibert et al., *Phys. Rev. B* **95**, (2017) 195404.
[4] T. M. Savchenko et al., *Phys Rev. B* **102**, (2020) 205418.
[5] J. Vijayakumar et al., *Nat. Commun.* **14**, (2023) 174.

# Tracking Transformations of Magnetic Nanoparticles

*Lucía Gutiérrez*[1]*

[1] *Dept. Química Analítica, Universidad de Zaragoza, Instituto de Nanociencia y Materiales de Aragón (UNIZAR-CSIC) and CIBER-BBN, C/Mariano Esquillor s/n, Campus Río Ebro, Zaragoza, Spain*

*lu@unizar.es

Magnetic nanoparticles (MNPs), especially those composed of iron oxides, have multitude of biomedical uses, such as contrast agents for Magnetic Resonance Imaging (MRI), drug delivery systems, and heat mediators for cancer treatment through magnetic hyperthermia. However, there is a need for techniques that can monitor the changes MNPs undergo during synthesis of these magnetic materials and after their administration in biological systems.

One powerful technique for characterizing MNPs is measuring the temperature dependence of the alternating current (AC) magnetic susceptibility. This method can track the transformations of MNPs and detect even small variations in parameters such as size, composition, or aggregation, which strongly affect the magnetic properties of the materials. AC magnetic susceptibility can thus be used to study different scenarios of MNP transformations, such as size variations during synthesis, aggregation effects, and degradation processes in biological environments.

Several series of samples characterized by AC magnetic susceptibility in which a single parameter (composition, size or aggregation) has been modified will be presented including: i) The synthesis of iron oxides with different sizes achieved using proteins as templates [1], ii) The effect of the aggregation in agar, cell and tissue samples [2], iii) The effect of Mn doping in magnetite nanoparticles critical to tune the contrast produced by these materials in the frame of MRI analysis [3] and iv) The degradation of nanoparticles in tissue samples and the formation of ferritin, the iron storage protein, over a period of 15 months [4].

Results from the characterization of all these series have allowed us to gain understanding on how these relevant parameters affect the magnetic properties of the nanoparticles and its impact in different biomedical applications.

**Acknowledgements**

The work presented corresponds to colaborations performed with the groups of Aitziber Cortajarena, Susana Carregal and Jesús Ruiz Cabello from CIC Biomagune, in Spain; Puerto Morales from the ICMM and Domingo Barber from the CNB.

**References**

[1] A. Aires, Y. Fernández-Afonso, G. Guedes, et al, *Chem Matter* **34** (2022) 10832–10841.
[2] Y. Fernández-Afonso, L. Asín, L.Beola, et al, *ACS App Bio Mat* **5** (2022) 1879-1889.
[3] S. Carregal-Romero, A.B. Miguel-Coello, et al, *Small* (2022) 2106570.
[4] Y. Portilla, Y. Fernández-Afonso, S. Pérez-Yagüe, et al, *J Nanobiotech* **20** (2022), 1-23.

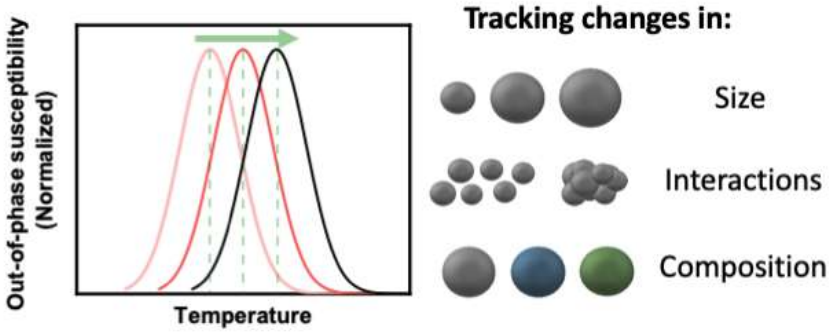



Figure: AC magnetic susceptibility measurements can be used to track changes in size, degree of dipolar interactions or composition in systems where magnetic nanoparticles evolve over time.

# SYMPOSIUM 07.
## MAGNETIC NANOPARTICLES AND MULTIFUNCTIONAL MAGNETIC MATERIALS.
## S7. ORAL PRESENTATIONS

# Domain Formation, Magnetization Reversal, and Magnetotransport in Self-Assembled 1D Magnetic Nanostructures

*Mehran Sedrpooshan,[1,2]*, Pau Ternero[1,2], Claudiu Bulbucan[3], Calle Preger[1,3,4], Adam Burke[1,2], Maria E Messing[1,2], Rasmus Westerström[1,2]*

[1]*NanoLund, Lund University, SE-221 00 Lund, Sweden*
[2]*Department of Physics, Lund University, SE-221 00 Lund, Sweden*
[3]*MAX IV Laboratory, Lund University, SE-221 00 Lund, Sweden*
[4]*Ergonomics and Aerosol Technology, Lund University, SE-221 00 Lund, Sweden*
*mehran.sedrpooshan@sljus.lu.se

Directed self-assembly of magnetic nanoparticles (NPs) using external fields is an efficient strategy for producing magnetic nanostructures. Recently, the generation of magnetic structures through the self-assembly of charged gas-phase particles using spark ablation has opened the way for further developments in this field [1]. In this method, the generation of nanostructures starts with the production of magnetic NPs in the gas phase as opposed to conventional chemical methods. For this, repetitive sparks between two metallic electrodes evaporate materials under an inert carrier gas flow. The metallic vapor nucleates and condensates to form agglomerates which are then compacted and size selected before being guided onto any substrate of choice. The resulting NPs have a transition-metal ratio almost identical to the seed material, allowing the nanoscale composition to be tuned by the composition of the electrodes [2]. To self-assemble the aerosol nanoparticles, an electric field is used to attract the charged particles to the substrate surface, and a magnetic field is utilized to arrange the incoming NPs into nanochains (NCs) via dipole-dipole interaction.

By this approach, a set of different nanostructures can be easily made as bundles, NCs, and nanowires (NWs) shown in Fig.1(a). The presented method provides a template-free, bottom-up technique for generating 1D magnetic nanostructures, which can be further developed into 2D and 3D structures. It is demonstrated that the composition of the produced structures can be tuned by forming alloys or segmented structures, as shown in Fig.1(b). Moreover, the 1D structures can be deposited onto most substrates, thus allowing for the direct integration of the NCs into devices, herein demonstrated by fabricating magnetoresistive devices.

In this work, we demonstrate the magnetic properties of single-component Co NCs and NWs as well as alloyed FeCo NCs and NWs, together with multi-component bundles, through magnetization study using SQUID magnetometry, X-ray microscopy and magnetotransport. The results are also complemented by micromagnetic simulations to further evaluate the magnetization reversal in the structures. For both Co and FeCo, the results show the introduction of a substantial magnetic shape anisotropy resulting in a ≈100% increase in the remanent magnetization when depositing the particles under an external magnetic field. Moreover, the transformation of NCs into NWs by post-annealing leads to a ≈50% increase in coercivity. On the other hand, single-NC X-ray microscopy (STXM-XMCD method) results, together with the simulations, suggest that the magnetocrystalline anisotropy of the particles and also local variations in the geometry of the system affect the magnetic equilibrium states (Fig.1(c)). Furthermore, magnetotransport measurements of the fabricated devices present a clear correlation between the electrical resistance and the magnetic states of the chains observed from the microscopy results and simulations, Fig.1(d).

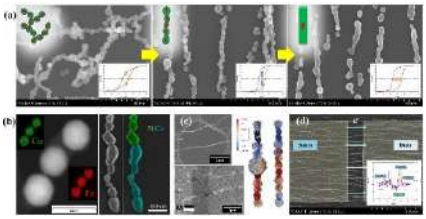


Fig.1: a) assembly of Co nanoparticles under no external magnetic field, nanochains produced under an in-plane magnetic field, and nanowires generated by the post-annealing of nanochains. b) tuning the composition of the structures by alloying and multi-component segmented structures, c) X-ray microscopy and domain formation studies in single nanochains, d) magnetoresistive device fabricated by direct integration of particles, together with the magnetotransport results.

## References

[1] Preger, Calle, *et.al., Nanotechnology* 32, 19 (2021) 195603.
[2] Preger, Calle, *et.al., The Journal of Physical Chemistry C* 123, 26 (2019) 16083.

# Microwave-assisted ultra-stable to oxidation nanopermalloys in aqueous media

*Antonio Santana-Otero[1*], Maria Eugenia Fortes-Brollo[2], Carlos Frade[1], María del Puerto Morales[3], Daniel Ortega[1,4,5]*

[1] *University of Cádiz, Condensed Matter Physics Dept., 11510 Puerto Real, Spain*
[2] *Department of Physics, Chalmers University, Fysikgränd 3, 412 96 Gothenburg, Sweden*
[3] *Institute of Materials Science of Madrid (ICMM-CSIC), Sor Juana Inés de la Cruz 3, 28049 Madrid, Spain*
[4] *Institute of Research and Innovation in Biomedical Sciences of Cádiz (INiBICA), 11002 Cádiz, Spain*
[5] *IMDEA Nanoscience, 28049 Madrid, Spain*

* santanaotero.a@gmail.com

Metal alloys with high magnetic permeability offer a wide range of applications in technology, such as electromagnetic shielding or energy storage. Down to the nanoscale, the number of applications of this type of materials grows exponentially, enabling magnetic inks for 3D direct printing, more capable catalysts, magnetic hyperthermia agents, or tags for cell labelling. The higher the permeability, the greater the response of the material to an applied magnetic field, being more effective. This would allow to reduce the amount of material that is required for a particular application. Metal alloys have difficulties in biological environments as a result of strong oxidising conditions. Some ways to solve this problem are to coat metals with different materials such as silica, organic polymers or even graphene to create a protective layer against oxidation [1,2].

We present a method to obtain oxidation resistant $Ni_xFe_{1-x}A_y$ (A = other transition metals) permalloys nanoclusters that feature high permeability and saturation magnetisation [3]. This protection is given by the 1,12-dodecanediol, which coats $Ni_xFe_{1-x}A_y$ nanoclusters and precludes corrosion due to its reducing character. The microwave reactor-assisted synthesis method consists of a 10-minute single step method in aqueous media, where the stoichiometry of the metal nanoparticles can be controlled by changing metal precursors.

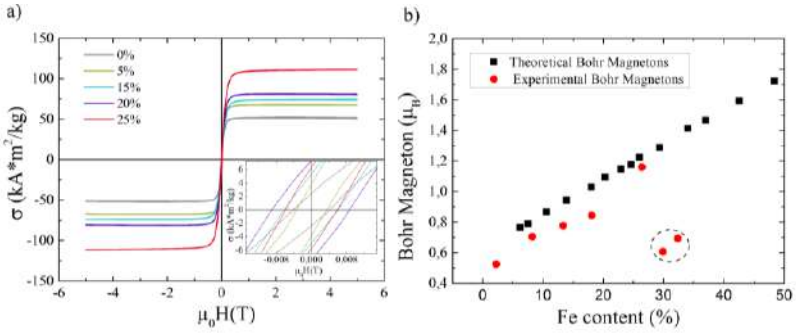



**Figure1** a) Hysteresis loops at 300 K of samples with 0, 5, 15, 20 and 25% Fe content. b) Variation of the experimental atomic magnetic moment ($M_{NiFe}$) values with the Fe content in the $Ni_xFe_{1-x}$ nanoclusters (red circles) and as described by the theoretical Slater-Pauling.

## Acknowledgements

We acknowledge support under grants PID2020-117544RB-I00, CEX2020-001039-S, RED2018-102626-T and MAT2017-85617-R funded by MCIN/AEI/ 10.13039/501100011033, and grant RYC2018-025253-I funded by MCIN/AEI/10.13039/501100011033 and by "FEDER A way of making Europe". This research was also funded by the CSIC Intramural project 201960E062 (MPM).

## References

[1] S. Chen, L. Brown, et al., *ACS Nano* **5** (2011) 1321.
[2] T. Nishimura, V. Raman, *Materials* **7** (2014) 4710.
[3] A. Santana-Otero, M. E. Fortes-Brollo, M. d. P. Morales, D. Ortega, *Nanoscale* **14** (2022) 16639.

# Nanostructured FeCo Films: Exceeding the Slater-Pauling Limit

*Chris Binns[1]*, Raúl López-Martín[1], José Angel de Toro[1], Benito Santos Burgos[1], Peter Normile[1], Andrew Pratt[2], Toby Bird2, Maha Alotaibi[2], Jack Pearce[2], David Hesp[2], Stuart Cavill[2], Sharon Louch[3], Steve Baker[4], Suart Tornton[4]*

[1] *IRICA, Universidad de Castilla-La Mancha, Ciudad Real, Spain*
[2] *School of Physics, Engineering and Technology, University of York, York, UK*
[3] *Smith and Nephew Ltd, Hull, UK*
[4] *Departmment of Physics and Astronomy, University of Leicester, UK*
*Christopher.binns@uclm.es

The most magnetic transition metal material currently available for applications is the alloy $Fe_{67}Co_{33}$ and this has been available since about 1912. A few years ago, it was demonstrated [1] and verified recently[2] that magnetic materials produced by co-depositing size-selected nanoparticles of Fe or Co with diameters around 2nm and an atomic vapour of the opposite material on a substrate have a magnetisation that is higher than conventional FeCo alloys with the same composition (see Fig. 1). This is the case until the nanoparticle volume fraction reaches the percolation threshold. For Co nanoparticles deposited with Fe matrices, that is at the Fe-rich end, the magnetisation reaches values higher (up to 3μB/atom) than the top of the conventional alloy magnetisation vs. composition curve (2.45 $\mu_B$/atom), the so-called Slater-Pauling limit. This limit, which has been in place for over a century, has represented a limit to performance in a number of applications, including electric machines and magnetic recording.

Following comprehensive studies by X-Ray Magnetic Circular Dichroism (XMCD), Extended X-Ray Absorption Fine Structure (EXAFS) and Transmission Electron Microscopy (TEM) it is now possible to have a clear understanding of why this material has such a high saturation magnetisation. The talk will present the latest results, how to optimise the magnetisation further and ideas for scaling up the production of the films for industrial applications.

[1] C. Binns et al, *J. Phys D:Appl. Phys.*. **38** (2005) R357.
[2] C. Binns et al, *Nanomaterials* (2023), manuscript submitted (see Fig. 1 for latest data)

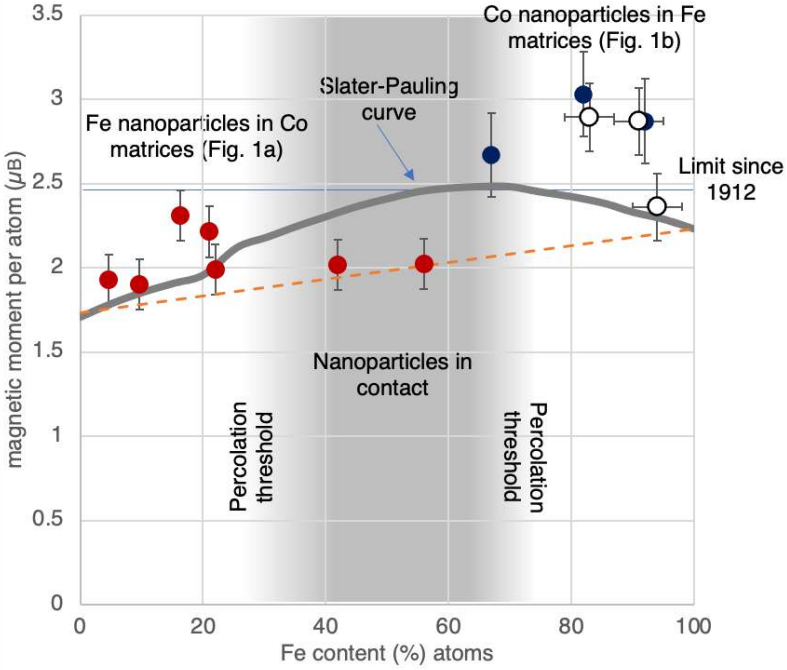



**Figure 1** magnetic moment/atom in nanoparticle-assembled films compared to the Slater-Pauling curve. The filled circles are the original published results[1] and the open circles are new data obtained in January this year[2]

# Spider Silk Threads Functionalized With Magnetostrictive FeCo Coating

*F. Spizzo[1]*, G. Greco[2,3], L. Del Bianco[1], M. Coisson[4], N. M. Pugno[3,5]*

[1]*Department of Physics and Earth Science, University of Ferrara and INFN-Ferrara, Ferrara I-44122, Italy*
[2]*Department of Anatomy, Physiology and Biochemistry, Swedish University of Agricultural Sciences, Uppsala, 750 07, Sweden*
[3]*Laboratory for Bioinspired, Bionic, Nano, Meta, Materials & Mechanics, Department of Civil,Environmental and Mechanical Engineering, University of Trento, Trento I-38123, Italy*
[4]*Nanoscience and Materials Division, INRIM, Torino I-10135, Italy*
[5]*School of Engineering and Materials Science, Queen Mary University of London, London E1 4NS, UK*
*federico.spizzo@unife.it

Flexible hybrid functional materials are attracting an increasing interest due to their technological potential [1]. In particular, these systems could display novel features, resulting from the combination, for instance, of electrical and magnetic properties, as often found in hard metallic materials, with stretchability and deformability typical of soft materials. Regarding the latters, silk is a remarkable soft material as, thanks to its excellent mechanical properties, it can be used in composites to provide compliance, stretchability and strength.

In this contribution, an original hybrid material is presented, consisting of spider silk threads coated with a metallic and magnetostrictive FeCo alloy [2]. The 100nm-thick FeCo layer was deposited using a dc magnetron sputtering apparatus, in Ar atmosphere. The durability and the homogeneity of the obtained fibers are validated, as well as the good mechanical properties. Due to the metallic coating, hybrid threads are electrically conductive, showing a ohmic behaviour; the resistance, even under strain, is of the order of ~ 10 kΩ.

The magnetic study, carried out by SQUID and MOKE magnetometry and Magnetic Force Microscopy, reveals that the magnetic behavior of the hybrid system is ruled by the silk thread–FeCo layer interaction, especially under mechanical stress. In fact, the application of a tensile strain to the thread changes the magnetic response, which therefore can be exploited to reveal the tensional state of the sample itself. In conclusion, the created hybrid system represents a solid proof of concept demonstrating the possibility of functionalizing spider silk and thus imparting electrical conductivity and stress-sensitive magnetic properties to this attractive base material. The obtained results pave the way to foreseen applications in flexible electronics and magnetic actuation.

## Acknowledgements

N.M.Pugno was supported by the Italian Ministry of Education, University and Research (MIUR) under the PRIN-20177TTP3S grant. G. Greco was supported by Caritro Foundation (prot. U1277.2020/SG.1130). F.Spizzo was supported by the University of Ferrara under project “Fondo per l’Incentivazione alla Ricerca” (FIR) – 2021.

## References

[1] J. Li, A. Wang, J. Qin, H. Zhang, Z. Ma, G. Zhang, Compos. Part A Appl. Sci. Manuf. 2021, 140, 106144.
[2] F. Spizzo, G. Greco, L. Del Bianco, M. Coïsson, N. M. Pugno, Adv. Funct. Mater. 2022, 32, 2207382

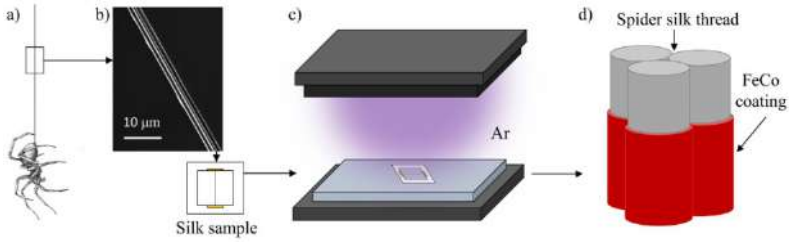



Figure: schematic of the sample preparation. a) The silk samples were collected from dragline threads naturally spun by the spider (*Cupiennius salei*). b) Scanning electron microscopy image of the native silk thread. c) The samples, mounted on a paper frame, were inserted in a dc magnetron sputtering apparatus and coated homogeneously with a 100nm-thick FeCo layer, as depicted in d).

# Magnetocaloric FeRh microstructures by laser-assisted direct writing

*Shabbir Tahir[1]*, Carlos Doñate-Buendía[1], David Koch[2], Joachim Landers[3], Soma Salamon[3], Heiko Wende[3], Bilal Gökce[1]*

[1]*Chair of Materials Science and Additive Manufacturing, University of Wuppertal, 42119 Wuppertal, Germany*

[2] *Institute of Materials Science, Technical University of Darmstadt, 64287 Darmstadt, Germany.*

[3] *Faculty of Physics and Center for Nanointegration Duisburg-Essen (CENIDE), University of Duisburg-Essen, 47057 Duisburg, Germany.*

*tahir@uni-wuppertal.de

Magnetocaloric refrigeration has the potential to replace compress gas-based refrigeration systems due to its higher energy efficiency and lower environmental impact. However, a prominent magnetocaloric effect is typically witnessed in materials containing rare and expensive elements. To reduce the amount of quantities required, the use of these materials in miniaturized applications such as magnetic microcoolers, thermal sensors, magnetic micropumps and actuators can be a feasible route.

Among magnetocaloric materials, near-to-equiatomic FeRh alloy is known to exhibit the largest magnetocaloric effect at the first order antiferromagnetic-ferromagnetic (AFM-FM) phase transition. In this contribution, it is proposed a stepwise process route to generate magnetocaloric FeRh microstructures from an equiatomic FeRh target. First, paramagnetic, near-to-equimolar FeRh nanoparticles were generated by ps-pulsed laser ablation of FeRh target in ethanol.[1] The nanoparticles were transformed into a 1. wt% ink, dispersed on PVP-coated glass substrates and patterned using a continuous wave laser (Fig. 1a).

Continuous wave direct writing leads to the sintering of the deposited nanoparticles into micrometric structures and simultaneously promotes the γ- to B2 FeRh phase transition required to achieve the FeRh magnetocaloric response. The partial (52%) phase transformation from γ- to B2-FeRh is achieved at the laser fluence of 242 J/cm$^2$, leading to an increase in magnetization of 35 Am$^2$/kg due to the field induced AFM-FM transition (Fig. 1b). Laser sintering has shown a sixfold increase in magnetization during the AFM-FM phase transition compared to furnace annealed FeRh ink [2]. Finally, 2D magnetocaloric custom structures were created, showing this route as a promising approach for fabricating micrometric cooling structures for miniaturized applications (Fig. 1c).

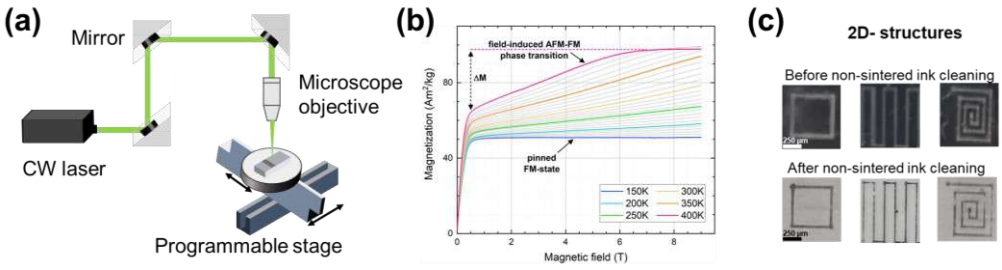



**Figure 1: (a)** Sintering by 532 nm continuous wave laser and a microscope objective (10X) providing the micrometric resolution required. **(b)** Magnetization of FeRh nanoparticles at a laser fluence of 246 J/cm$^2$ measured at 0 to 9 T at 150 up to 400 K across the AFM-to-FM phase transition. **(c)** Depiction of the custom structures produced through laser processing at a laser fluence of 246 J/cm².

## References

[1]. Nadarajah, R.; Tahir, S.; Landers, J.; Koch, D.; Semisalova, A. S.; Wiemeler, J.; El-Zoka, A.; Kim, S.-H.; Utzat, D.; Möller, R.; Gault, B.; Wende, H.; Farle, M.; Gökce, B., Nanomaterials 2020, 10 (12), 2362

[2]. Nadarajah, R.; Landers, J.; Salamon, S.; Koch, D.; Tahir, S.; Doñate-Buendía, C.; Zingsem, B.; Dunin-Borkowski, R. E.; Donner, W.; Farle, M.; Wende, H.; Gökce, B. Scientific Reports 2021, 11 (1), 13719.

# Magnetic Characterization on Single 120 nm Diameter Nanoplatelets through Photothermal Magnetic Circular Dichroism

*J. Li[1], S. Adhikari[2], Y. Wang[2], L. Ruijs[1], P. Spaeth[2], B. Koopmans[1], M. Orrit[2] & R. Lavrijsen[1]*
*1. Department of Applied Physics, Eindhoven University of Technology, 5600 MB Eindhoven, Netherlands*
*2. LION, Huygens Kamerlingh Onnes Laboratory, 2300 RA Leiden, The Netherlands*
*j.li6@tue.nl

Synthetic antiferromagnetic nanoplatelets (SAF NPs) with perpendicular magnetic anisotropy (PMA) are promising candidates for nano-torque-related applications [1]. To manipulate the NPs, a good understanding of the homogeneity in magnetic and structural properties is required. Most techniques measure these properties based on ensemble techniques [2], however, an efficient and easy magnetic characterization technique on single particle level, down to the nanometer range, is currently lacking.

Here, we present a novel characterization method based on photothermal magnetic circular dichroism (PT MCD), which measures the differential absorption of left and right circularly polarized light of an individual magnetic nanoparticle via to the polar Kerr effect [3]. This method allows us to address the spatial and temporal heterogeneity of the NPs at the single-particle level. We show that via PT MCD, we can measure the hysteresis loop on a single 120 nm diameter SAF NP (see Fig 1b). The loop exhibits the antiferromagnetic state at a zero applied field and a sharp magnetization switch at a large field which is comparable to the literature [4]. The statistics of the switching fields of 32 NPs are shown in Fig 1c where a difference in the distribution is observed for the $B_H$ and $B_L$ switch (see fig 1b for definition), which we attribute to a change in the dominant magnetic reversal mechanism of the NPs [5].

In this talk, we will discuss the PT MCD method, compare the results with conventional, ensemble-based SQUID magnetometry and present the statistics of individual SAF NP switching. Our results will pave the way to a deeper understanding of torque based applications using PMA-SAF NPs and introduce the PT MCD method to the magnetic-nanoparticle community.

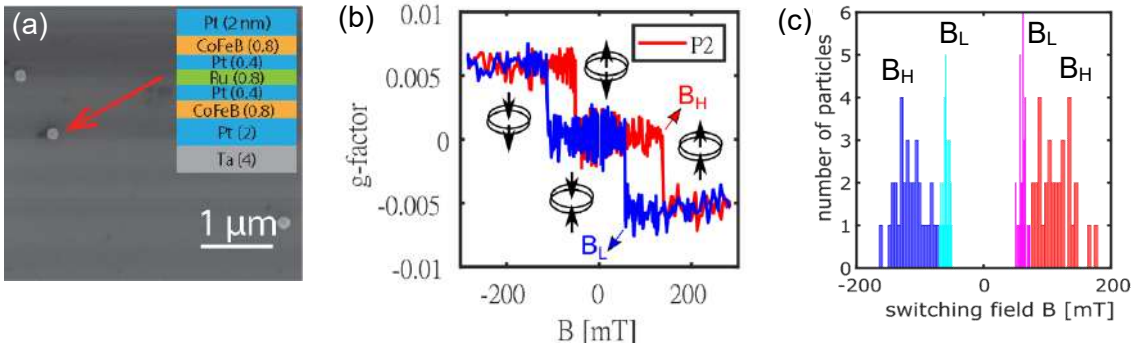



Fig 1. (a) SEM image of NPs used in the work. The insert shows the thin film stack. (b). Hysteresis loop on a single NP measured by PT MCD with the field applied along the out-of-plan direction. (Inset) Schematic representation of orientations of the magnetization of the two CoFeB layers. $B_H$ and $B_L$ are indicated by the arrows. (c) Histograms of the switching fields of 32 NPs measured by PT MCD. Blue: negative $B_H$; cyan: negative $B_L$; red: positive $B_H$; magenta: positive $B_L$.

## References

[1] Mansell, R., *et al*. Scientific reports 7.1 (2017): 1-7.
[2] Wasielewski, M. R., *et al*. Nature Reviews Chemistry 4.9 (2020): 490-504.
[3] Spaeth, Patrick, *et al*. Nano letters 22.9 (2022): 3645-3650.
[4] Li, J., *et al*. Applied Physics Letters 121.18 (2022): 182406.
[5] Thomson, T., *et al*. Physical review letters 96.25 (2006): 257204.

# Exchange bias in metal-doped single-phase ferrites induced by defect engineering of core@shell nanoparticles.

*Beatrice Muzzi,[1,2,3] Martin Albino,[3] Michele Petrecca,[3] Claudia Innocenti,[2,3] César de Julián Fernández,[4] Giovanni Bertoni,[5] M. Ricardo Ibarra, [6,7,8] Mogens Christensen,[9] Maxim Avdeed,[10] Clara Marquina, [7,8,*] Claudio Sangregorio[2,3,*]*

[1] *Dept. of Biotechnology, Chemistry and Pharmacy, University of Siena 1240, I-53100 Siena, Italy*
[2] *ICCOM - CNR, I-50019 Sesto Fiorentino (FI), Italy*
[3] *Dept. of Chemistry "U. Schiff", Uni. of Florence and INSTM, I-50019 Sesto Fiorentino (FI), Italy*
[4] *IMEM - CNR, I-43124 Parma, Italy*
[5] *CNR – Istituto Nanoscienze, I-41125 Modena, Italy*
[6] *Instituto de Nanociencia y Materiales de Aragón (INMA), Consejo Superior de Investigaciones Científicas (CSIC)-Universidad de Zaragoza, 50009-Zaragoza, Spain*
[7] *Dpto. de Física de la Materia Condensada, Universidad de Zaragoza, 50009-Zaragoza, Spain*
[8] *Laboratorio de Microscopias Avanzadas (LMA), Uni. de Zaragoza, 50018- Zaragoza, Spain*
[9] *Dept. of Chemistry and iNANO, Aarhus Uni. Langelandsgade 140, DK-8000 Aarhus C, Denmark*
[10] *Australian Nuclear Science and Technology Organisation (ANSTO), New Illawarra Road, Lucas Heights NSW 2234 Australia.*
*beatrice.muzzi@iccom.cnr.it

The crystal site occupancy of different divalent ions and the induction of lattice defects (vacancies, dislocations, stacking faults and antiphase boundaries) represent an additional tool for modifying the intrinsic magnetic properties (e.g. high coercive field and/or exchange bias) of spinel ferrites nanoparticles.[1] An efficient strategy to introduce defects into the ferrite spinel lattice is the controlled oxidation of core@shell AFM@F(i)M $Fe_{1-x}O@Fe_3O_4$ NPs. Interestingly, with this approach also defected cobalt ferrite with very large exchange bias are obtained. However, the final NPs displaied a magnetic behaviour strongly affected by the contribution of the antiferromagnetic component, due to the formation of a large magnetic disorder spinel ferrite sub-domains.[2] A possible strategy to overcome the drawback is to add, to the defected cobalt ferrite, a second divalent cation able to restore the magnetic order while retaining the lattice defects. Here we present an investigation aimed to rationalize the effect of $Co^{(II)}$ and $Ni^{(II)}$ on the lattice defects induced in spinel ferrite NPs by solvent mediated mild oxidation of AFM@FiM precursors, and on how they affect the magnetic properties. 20 nm core@shell $Fe_{0.95}O@Fe_3O_4$, $Co_{0.3}Fe_{0.7}O@Co_{0.8}Fe_{2.2}O_4$ and $Ni_{0.17}Co_{0.21}Fe_{0.62}O@Ni_{0.4}Co_{0.3}Fe_{2.3}O_4$ NPs synthetized by thermal decomposition method,[3] were oxidized to the single phase nanocrystals. As a result, the core@shell morphology is removed and transformed in a spinel-like nanoparticle, through a topotactic transformation. In addition, the appearance of crystal defects and antiphase boundaries improves the magnetic properties of the starting compounds and leads to the appearance of exchange bias at room temperature.

**Acknoledgments**

This work is supported by EU-H2020 AMPHIBIAN Project (n. 720853) and by European Union's Horizon 2020 research and innovation programme under grant agreement No 823717-ESTEEM3 and by the European Union - NextGenerationEU (National Sustainable Mobility Center CN00000023, Italian Ministry of University and Research Decree n. 1033 - 17/06/2022, Spoke 11 – "Innovative Materials & Lightweighting"; National Recovery and Resilience Plan (NRRP), PE0000021, Italian Ministry of University and Research Decree 1561 – 11/10/2022, "Network 4 Energy Sustainable Transition – NEST").

**References**

1 A. Lappas, G. Antonaropoulos, K. Brintakis, M. Vasilakaki, K. N. Trohidou, V. Iannotti, G. Ausanio, A. Kostopoulou, M. Abeykoon, I. K. Robinson and E. S. Bozin, *Phys. Rev. X*, 2019, **9**, 1–17.
2 A. López-Ortega, E. Lottini, G. Bertoni, C. De Julián Fernández and C. Sangregorio, *Chem. Mater.*, 2017, **29**, 1279–1289.
3 B. Muzzi, M. Albino, M. Petrecca, C. Innocenti, C. de J. Fernández, G. Bertoni, C. Marquina, M. R. Ibarra and C. Sangregorio, *Small*, 2022, **2107426**, 2107426.

# Unveiling new insights into bi-magnetic antiferromagnetic/ferrimagnetic Core/Shell nanoparticles: a multicharacterization approach

*M. Estrader[1*], A. G. Roca[2], A. López-Ortega[3], R. U. Ichikawa[4], I. Peral[5], X. Turrillas[6], D. del-Pozo-Bueno[7], M. Varela[8], F. Peiró[7], S. Estradé[7] and J. Nogués[2,9]*

*martaestrader@gmail.com

[1]Departament de Química Inorgànica i Orgànica, Universitat de Barcelona and Institute of Nanoscience and Nanotechnology of the University of Barcelona (IN2UB), E-08028 Barcelona, Spain,

[2]Catalan Institute of Nanoscience and Nanotechnology (ICN2), Campus UAB, Bellaterra, E-08193, Spain

[3]Departamento de Ciencias and Institute for Advanced Materials and Mathematics INAMAT, Universidad Pública de Navarra, 31006 Pamplona, Spain

[4]Nuclear and Energy Research Institute (IPEN/CNEN-SP), SP 05508-000 São Paulo, Brazil

[5]Department of Physics and Materials Science, University of Luxembourg, L-1511 Luxembourg

[6]Institut de Ciència de Materials de Barcelona- CSIC, UAB Campus, E-08193 Barcelona, Spain

[7]LENS-MIND, Departament Enginyeries Electrònica i Biomèdica and Institute of Nanoscience and Nanotechnology of the University of Barcelona (IN2UB), E-08028 Barcelona, Spain

[8]Departamento de Física de Materiales e Instituto Pluridisciplinar, Universidad Complutense de Madrid, 28040 Madrid, Spain

[9]ICREA, E-08010 Barcelona, Spain

$FeO/Fe_3O_4$ core/shell nanoparticles are a subject of considerable current interest both due to their appealing magnetic properties (e.g., tunable exchange bias or the presence of both a Néel and a Verwey transitions) and their potential applications (e.g., magnetic hyperthermia, magnetic bioassays, microwave absorbers, anode materials for Li-ion batteries, or solar hydrogen production via water splitting). In $FeO/Fe_3O_4$ core/shell nanoparticles both core and shell exhibit magnetic properties, hence, not only is the structural/morphological interface important but also the magnetic arrangement at the interface can play a crucial role in the properties and performance of the nanoparticles. In this work we have revealed two important features to understand the magnetic properties of bimagnetic $FeO/Fe_3O_4$ core/shell nanoparticles: i) the temporal evolution over four years of the oxidation front which leads to a final onion-like structure with a graded composition[1] and ii) the concomitant appearance of a graded magnetic structure[2]. We have demonstrated that the oxidation process reaches to a 'stand-by' state owing to the passivation character of the $Fe_3O_4$ shell and that the magnetic moment being largest at the surface decreases towards the inner part of the nanoparticle. The elucidation of these results has been addressed by a careful multicharacterization approach based on X-ray diffraction (Whole Powder Pattern Modeling-WPPM, Rietveld refinement, Pair Distribution Function-PDF), Electron Energy Loss Spectroscopy (EELS) and electron Magnetic Circular Dichroism (e-MCD) techniques. Finally, we have also unveiled that for the $Fe_xO_y/Fe_3O_4$ nanoparticulated system, for small nanoparticles (9 nm) the $Fe_xO_y$ core is highly non-stoichiometric and strained leading to the loss of its internal magnetic structure, namely, to the antiferromagnetic behaviour.

## Acknowledgements

M.E. and A. G. R acknowledges the grants RYC2018-024396-I and RYC2019-027449-I, respectively, funded by MCIN/AEI/ 10.13039/501100011033 and by "ESF Investing in your future'' and grants PID2019-106165GB-C22, PID2019-106165GB-C21 and RTI2018-095495-J-I00 funded by MCIN/AEI/ 10.13039/501100011033 and by "ERDF A way of making Europe".

## References

[1] R. U. Ichikawa, A. G. Roca, A. López-Ortega, M. Estrader, I. Peral, X. Turrillas, J. Nogués, *Small.* **14** (2018) 1800804.

[2] D. del-Pozo-Bueno, M. Varela, M. Estrader, A. López-Ortega, A. G. Roca, J. Nogués, F. Peiró, S. Estradé, *Nano Lett.* **21** (2021) 6923-6930.

# Widefield Magnetic Microscopy with NV Centers in Diamond for Monitoring of Magnetic Micro- and Nanoparticles

*N. Mathes[1], K. Everaert[2,3]*, R. Bleul[4], R. Sperling[4], J. Leliaert[3], P. Knittel[1], F. Wiekhorst[2], X. Vidal[1]*

[1] *Fraunhofer Institute for Applied Solid State Physics, Tullastraße 72, Freiburg, Germany*
[2] *Physikalisch-Technische Bundesanstalt, Abbestraße 2-12, Berlin, Germany*
[3] *Department of Solid State Sciences, Ghent University, Krijgslaan 281/S1, Ghent, Belgium*
[4] *Fraunhofer Institute for Microengineering and Microsystems, Carl-Zeiss-Str. 18, Mainz, Germany*
*katrijn.everaert@ptb.de / katrijn.everaert@ugent.be

Magnetic nanoparticles and magnetic microbeads are widely used in biomedical applications. Current research mainly focuses on the properties of magnetic micro- and nanoparticle ensembles as macroscopic samples, and the view of their magnetic properties on a microscopic scale is largely hidden in the ensemble averages. However, local information of single or few numbered magnetic entities, e.g. in cellular environments, will benefit the further optimization of their performance.

The traditional approach for high spatial resolution magnetic imaging involves a scanning probe that is moved across the sample surface step-by-step. Magnetic fields are then locally quantified by magnetic force measurements (MFM), nanoSQUIDs, or a Nitrogen-Vacancy (NV) defect in a diamond lattice on the scanning tip. These scanning processes are however delicate, time-consuming, or destructive, and they are not optimal for time-critical or technological applications.

Widefield magnetic imaging using NV centers in diamond allows the simultaneous monitoring of events over wider areas. The approach offers a unique compromise between resolution, sensitivity, and measurement time allowing for a relatively fast (sub-seconds to minutes) magnetic image acquisition with a large field of view and diffraction-limited resolution [1]. These combined properties make it an ideal measurement setup for the time-resolved monitoring of magnetic nano- and microparticles in fluids. Our approach to widefield NV-based sensing uses a diamond plate containing a near-surface high NV-concentration layer with a thickness of approximately 300 nm.

We present magnetic imaging of magnetic microbeads and nanoparticles distributed on the surface of the diamond as well as suspended in water. Optically detected magnetic resonance and longitudinal relaxation contrast imaging is performed, revealing information about the local magnetic field and GHz magnetic noise amplitudes respectively. Fig.1 shows particle clustering visualized by both modalities. Combined with conventional optical microscopy, complementary information on the position and movement of the particles driven by external magnetic field forces as well as the fluid flow is obtained. The experiments show the potential of widefield NV center microscopy as a real-time magnetic imaging system of magnetic micro- and nanoparticles for biomedical and microfluidic applications.

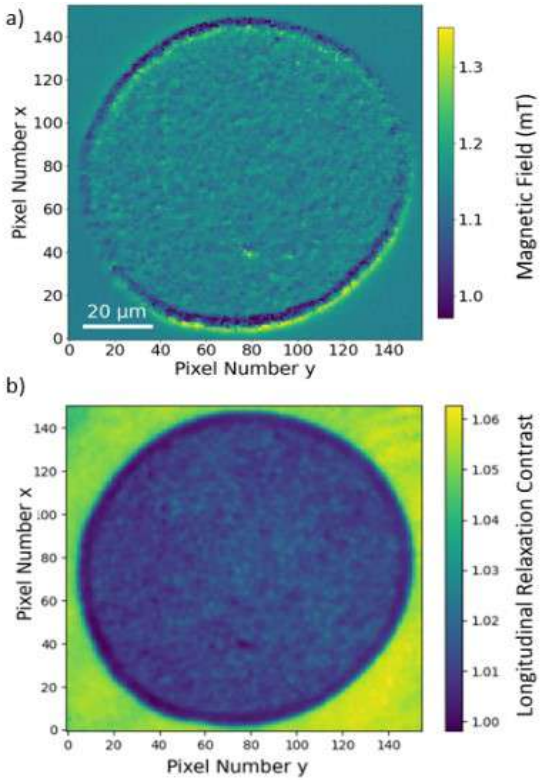



*Figure 1 a) Magnetic field component generated by magnetic nanoparticles reconstructed from the measured Zeeman splitting of the NV-center spin states.*
*b) Longitudinal relaxation contrast image of NV centers induced by MNP GHz noise*

## References

[1] E. V. Levine, et al., Principles and techniques of the quantum diamond microscope, Nanophotonics 8, 1945-1973 (2019).

# Monitoring in real time the aqueous coprecipitation synthesis of Fe-oxide nanoparticles with an AC susceptometer

*Maria Paz Fernández-García[1]*, Jose Miguel Teixeira[1], Maria González de la Vega[1],Clara Pereira[2], André M. Pereira[3], Jose Carlos B. Lopes[4], Cristina Freire[2], João Pedro E. Araújo[3]*

[1] *Department of Physics, University of Oviedo, 33007, Oviedo, Spain*

[2] *REQUIMTE, Department of Chemistry and Biochemistry, University of Oporto, Portugal*

[3] *Institute of Physics for Advanced Materials, Nanotechnology and Photonics, University of Oporto, Portugal*

[4] *Laboratory of Separation and Reaction Engineering, University of Oporto, Portugal*

*fernandezpaz@uniovi.es

We will present the potentiality of a simple and desktop AC susceptometer that we have designed and developed to monitor easily and in real time the coprecipitation synthesis of magnetic nanoparticles (NPs) obtained by aqueous coprecipitation route with both organic and inorganic bases. We will also show the paramount impact of rapid mixing by means of pitch-blade impellers into the physical and chemical properties of the final NPs. Besides, we will demonstrate that our set-up provide an *in situ* method to monitor the vital stages (nucleation, growth and diffusion) of the growing NPs by recording properly the in-phase and out-of-phase curves of the reaction mixture.

Our work put on view the potentiality of our AC susceptometer to explore and predict *in situ* and in less than 30 min the quality of the NPs (polydispersity, magnetic response, superparamagnetic character, etc) as time dependent in-phase and out-of-phase signals are fingerprints of the synthesized NPs. Therefore, this set-up might avoid long-term physicochemical characterizations usually done after NPs' synthesis.

**Acknowledgements**

The authors thank the mechanical and electrical work developed at the workshop of the Faculty of Sciences of the University of Porto. M.P. Fernández-García acknowledges financial support through the Spanish "Ministerio de Economía y Empresa" (MINECO) through research project RTI2018-094683-B-C52; Gobierno del Principado de Asturias and FICyT through research projects FC-GRUPINIDI/2018/000185, PAPI-18-GR-2010-0020, AYUD/2021/51822 and University of Oviedo for R&D project 2018/00061/008 in the competitive call PAPI-18-EMERG-8. J.P.A. thanks funding by projects POCI-01-0145-FEDER-032527 and ROTEIRO/0212/2013. M. González-de la Vega acknowledges funding through MCIU-19-RTI2018-094683-B-C52.

**References**

[1] M.P. Fernández-García, J.M.Teixeira, P. Machado, M.R.F.F. Oliveira, J.M. Maia, C. Pereira, A.M. Pereira, C. Freire, J.P Araújo, *Rev. Sci. Instruments* **86** (2015) 043904.

[2] M.P. Fernández-García, J.M.Teixeira, P. Machado, M. Enis Leblebici, J.C.B. Lopes, C. Freire, J.P Araújo, *Chem. Eng. Sci.* **138** (2015) 600-606.

*[3]* M.P. Fernández-García, J.M.Teixeira, M. González de la Vega, J.C. Moreira, J.P. Araújo, *Applied Physics A* **108** (2022) 209.

# Potential of AC Magnetometry to Display Protein Conformational Changes

*Alejandro Venegas-Gomez[1], Elena Sanz-de Diego[1], Sara H. Mejias[1], Jonathan Leon[2], Aitziber L. Cortajarena[1,2,3], Anna Solé-Porta[4], Anna Roig[4], Francisco J. Teran[1,5]*

*[1] iMdea Nanociencia, Ciudad Universitaria de Cantoblanco, 28049 Madrid, Spain*

*2 CIC biomaGUNE-BRTA, Paseo Miramón 194, 20014 Donostia-San Sebastian. Spain*

*3 Ikerbasque, Basque Foundation for Science, 48009 Bilbao, Spain.*

*4 ICMAB-CSIC, Campus de la UAB, 08193 Bellaterra, Spain*

*[5] Nanobiotecnología (iMdea-Nanociencia), Unidad Asociada al Centro Nacional de Biotecnología (CSIC), 28049 Madrid, Spain*

**alejandro.venegas@imdea.org*

Nowadays, magnetic nanoparticles (MNPs) are active agents in a broad range of research lines and biomedical applications, such as hyperthermia, drug delivery, imaging, or diagnosis. The latest application benefits from the variation of colloidal and magnetic features, after biomolecular recognition in liquids between target proteins and recognition ligands, which are present in bioconjugated MNPs (b-MNPs). AC magnetometry has shown to be a quick, reliable, and accurate technique to display variations of MNP magnetic relaxation processes, when proteins are attached onto MNP surface. Conformational changes of proteins alter their diffusion in liquids, altering MNP Brownian motion in liquids, which strongly influence AC magnetization cycles.

In this work, we report on the transduction potential of cobalt ferrite MNPs to detect protein conformational changes, based on variations of dynamic magnetization cycles in a field frequency range from 10 up to 100 kHz, for field intensities up to 24 kA/m. We studied two protein families covalently bonded onto commercial cobalt ferrite MNPs, with different chemical affinity and molecular mass. Proteins undergo conformational changes when exposed to temperature or denaturing agents (guanidine hydrochloride or urea) as corroborated by standard techniques (*i.e.* circular dichroism). Consequently, protein unfolding leads to conformational changes resulting in variation of MNP diffusion coefficient, quantified by nanoparticle tracking analysis and variations of the AC hysteresis area. The comparison of AC hysteresis area values, obtained from distinct controls subjected to similar denaturing conditions, allows probing reversible and irreversible changes depending on the employed denaturation method (temperature or chemical agents), in agreement with circular dichroism. This novel magnetic methodology is sensitive to protein conformational changes depending on their molecular mass, and affinity, requiring 20-fold less protein mass and providing quick and more accurate results (errors < 1%) than circular dichroism. Indeed, AC magnetometry has a novel technological biosensing potential beyond its use as a magnetic characterization technique.

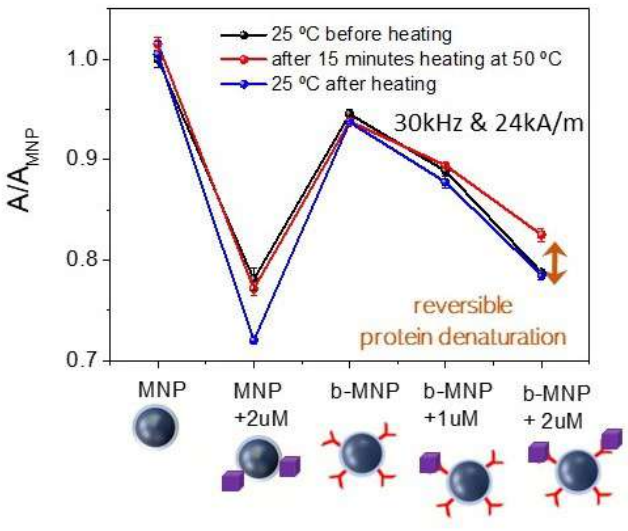



**Figure:** Normalized AC hysteresis area values obtained from different samples at 30 kHz & 24 kA/m at different temperatures.

**Acknowledgments**

Authors acknowledges financial support from PEJ-2017-AI/BMD-7517, M-ERANET 2018 (PCI2019-103600) and PID- 2020-117080RB-C53.

# Superparamagnetic particles for micro-inductor applications

*M. Zambach[1,*], M. Beleggia[2,3], Z. Ouyang[4], M. Knaapila[5] and C. Frandsen[1,§]*
*[1]DTU Physics, Technical University of Denmark, Kgs. Lyngby, Denmark*
*[2]DTU Nanolab, Technical University of Denmark, Kgs. Lyngby, Denmark*
*[3]Department of Physics, University of Modena and Reggio Emilia, Modena, Italy*
*[4]DTU Electro, Technical University of Denmark, Kgs. Lyngby, Denmark*
*[5]Department of Physics, Norwegian University of Science and Technology, Trondheim, Norway*
[§]fraca@fysik.dtu.dk

Magnetic components for power-electronics such as inductors and transformers are difficult to further miniaturize due to losses/heating in magnetic materials [1]. The reason is that miniaturized magnetic components operate at higher frequencies and magnetic losses generally increase strongly with increasing frequency [1]. Commercial high-frequency magnetic materials, e.g. Mn-Zn-ferrites, only offer acceptable losses below 1-2 MHz [2]. Nanocomposites, compromised of magnetic nanoparticles in a solid non-conducting matrix, have the potential to overcome this limitation by maintaining losses at an acceptable level above the 10 MHz range [3].

We have established a framework to guide the design and fabrication of magnetic devices containing magnetic single domain particles embedded in polymer materials. Theoretic and simulated results on DC and AC-susceptibility of dispersed superparamagnetic particles suggest that such systems may be usable as inductor core material in the MHz operation regime. This follows from the combination constant susceptibility in the MHz regime and moderate hysteresis losses, as illustrated by the in-phase and out-of-phase AC-susceptibility plot shown in Fig. 1. Based on the predictions and preliminary validating experiments, we present here a parameter-space overview of particle characteristics promising for micro-inductor cores used in power-converters. We focus on a minimal set of parameters; particle material, size, shape, and arrangement of particles to evaluate magnetic susceptibility and losses.

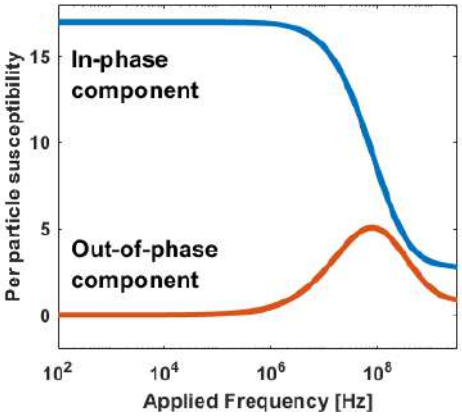



Figure 1: Calculation of dynamic susceptibility for randomly oriented 15±1 nm iron oxide particles.

Our theoretical and experimental results show that composites of certain superparamagnetic particles, embedded in a polymer matrix, are promising candidates for micro-inductor cores operating in the MHz range. The proposed materials offer high fabrication flexibility and enables tuning of magnetic susceptibility to fit inductor design optimum, allowing improved power-electronic efficiency.

## References

[1] M. Araghchini et al., IEEE Transactions on Power Electronics, 28, 4182–4201 (2013).
[2] M. Petrecca et al., Journal of Nanoscience and Nanotechnology, 19, 4974–4979 (2019).
[3] Michael P. Rowe et al., J. Mater. Chem. C, 3, 9789–9793 (2015).

# Conservation Laws For Interacting Magnetic Nanoparticles

Frederik Laust Durhuus[1] , Marco Beleggia[2,3] , Cathrine Frandsen[1,*]

[1] *DTU Physics, Technical University of Denmark, Denmark*
[2] *Department of Physics, University of Modena and Reggio Emilia, Italy*
[3] *DTU Nanolab, Technical University of Denmark, Denmark*

*Corresponding author, fraca@fysik.dtu.dk

Modelling of magnetic nanoparticle (MNP) dynamics is an active area of research, which is crucial for understanding aggregation and hysteresis behaviour at the nanoscale. However there are a number of subtleties in the conservation laws for MNP systems, which complicates model development and analysis. For instance it was recently found using angular momentum conservation for a single MNP that coupling between mechanical- and moment rotation leads to an extra term in the conventional LLG equation [1].

We have developed and implemented a general Langevin Dynamics model of interacting MNPs in liquid suspension at finite temperature. We further derived explicit formulas for the transfer of energy, linear- and angular momentum between the MNPs and to/from the environment. We demonstrate by numerical tests that all conserved quantities are fully accounted for, thus validating the model and the transfer expressions. Our work may be regarded as a generalisation of Ref. [2] to multiple particles at finite temperature, or a generalisation of Ref. [3] to include mechanical motion.

In addition to model development and verification, tracking the transfer of conserved quantities gives new insight on the physical system. Typically, energy dissipation is determined by the area of a hysteresis curve, however this is only possible for periodically driven systems in steady-state. Using our formulas, one can calculate the instantaneous power at each MNP and decompose into different loss channels. In particular magnetic losses from Gilbert damping heats the MNP itself, while mechanical losses from viscous damping heats the surrounding fluid. This gives a detailed, local perspective, applicable to non-equilibrium dynamics, which we demonstrate by analysing MNP collisions with and without an alternating, applied magnetic field. An example simulation is shown in Fig. 1.

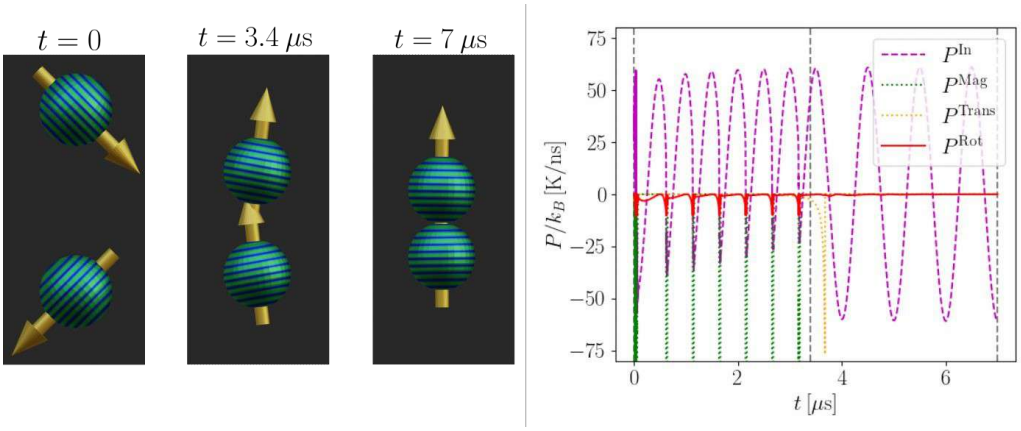



Figure 1: Simulation of 2, $R = 10$ nm, iron oxide MNPs at zero temperature, subject to a $B = 40$ mT sine-field oscillating at 1 MHz. **Left :** Snapshots of configuration. Arrows for moments. Anisotropy axes are perpen- dicular to stripes. **Right :** Gray lines indicate time-coordinates of snapshots. $P^{\mathrm{In}}$ is power exchanged between particles and B-field, $P^{\mathrm{Mag}}$ magnetic losses (Gilbert damping) and $P^{\mathrm{Trans}}$, $P^{\mathrm{rot}}$ translational and rotational vis- cous losses. Before the collision (orange spike) $P^{\mathrm{in}}$ is positive on average, indicating a net energy transfer from field to particles. Each green spike corresponds to a moment reversal, which dissipates energy. After colliding, the reversals cease and the net energy transfer is approximately 0.

## References

[1] Hedyeh Keshtgar et al. *Physical Review B*, **95**(2017), 134447.
[2] Santiago Helbig et al. *Physical Review B*, **107**(2023), 5.
[3] Jonathan Leliaert et al. *Nanoscale*, **13**(2021), 35.

# Dipolar Collective Magnetism: Anisotropy *vs* Interactions in Simple and Binary Particle Systems

*E. H. Sánchez,[1] M. Vasilakaki[2], S. S. Lee,[3] P. S. Normile,[1] M. S. Andersson,[4] R. Mathieu,[5] A. López-Ortega,[6] D. Peddis,[7] C. Binns,[1] K. Trohidou,[2] P. Nordblad,[5] J. Nogués,[8] J. A. De Toro[1*]*

[1]*Instituto Regional de Investigación Científica Aplicada (IRICA) and Departamento de Física Aplicada, Universidad de Castilla-La Mancha, 13071 Ciudad Real, Spain*

[2]*Institute of Nanoscience and Nanotechnology NCSR "Demokritos", 153 10 Agia Paraskevi, Greece*

[3]*Institute of Materials Research and Engineering, 31 Biopolis Way, #09-01, The Nanos, Singapore 138669*

[4]*Department of Chemistry, Ångström Laboratory, Uppsala University, 75121 Uppsala,Sweden*

[5]*Department of Materials Science and Engineering, Uppsala University, Box 35, 751 03, Uppsala, Sweden*

[6]*Departamento de Ciencias, Universidad Pública de Navarra, 31006 Pamplona, Spain*

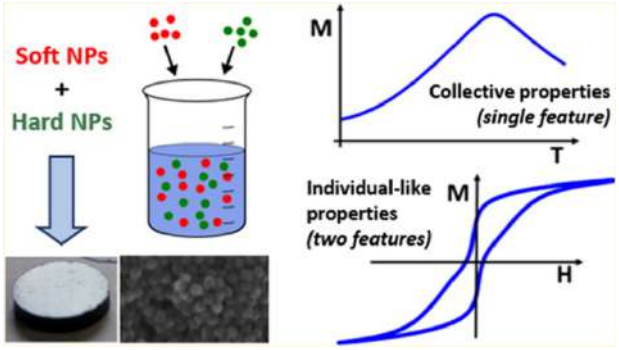
[7]*Università di Genova, Dip. di Chimica e Chimica Industriale, Via Dodecaneso 31, 1-16146 Genova, Italy*

[8]*Catalan Institute of Nanoscience and Nanotechnology (ICN2), CSIC and BIST, Campus UAB, Bellaterra, 08193 Barcelona, Spain*

* joseangel.toro@uclm.es

Dense systems of magnetic nanoparticles may exhibit dipolar collective behavior. However, two fundamental questions remain unsolved: (i) whether the transition temperature may be affected by the particles anisotropy or it is essentially determined by the intensity of the interparticle dipolar interactions, and (ii) what is the minimum ratio of dipole-dipole interaction ($E_{dd}$) to nanoparticle anisotropy ($K_{ef}V$) energies necessary to crossover from individual to collective behavior. We have studied a series of particle compacts (with packing fraction close to the random-close-packing limit) where dipolar interactions are similarly intense, but the nanoparticle anisotropy widely varies across the series [1]. This parameter has been tuned through different degrees of cobalt-doping in maghemite nanoparticles, resulting in a variation of nearly an order of magnitude. All the bare particle assemblies display collective behavior, except the one made with the highest anisotropy particles, which presents "marginal" features. Thus, a threshold of $K_{ef}V/E_{dd} \approx 130$ to suppress collective behavior is derived, in good agreement with Monte Carlo simulations. This translates into a crossover value of ≈1.7 for the easily accessible parameter $T_{MAX}$(interacting)/$T_{MAX}$(non-interacting) (temperature ratio of the maximum in the temperature dependent zero-field-cooled magnetization of interacting and dilute particle systems), which has been successfully tested against the literature to predict the individual-like/collective behavior of any given interacting particle assembly comprising relatively uniform particles.

Secondly, I will show the use of nanoscale-uniform binary random dense mixtures with different proportions of oxide magnetic nanoparticles with low/high anisotropy as a valuable tool to explore or, rather, define the dipolar-collective character of a given magnetic property as that resulting in the collapse of the individual features into a single one due to strong interactions (see Figure). Crucially, such collective character must, in general, be ascribed to specific properties and not to the system [2].




enough

## References

[1] E. H. Sánchez, M. Vasilakaki, S. S. Lee, P. S. Normile, M. S. Andersson, R. Mathieu, A. López-Ortega, D. Peddis, C. Binns, K. Trohidou, P. Nordblad, J. Nogués, J. A. De Toro, *Small* (2022) 2106762.

[2] E.H. Sánchez, M. Vasilakaki, S. S. Lee, P. S. Normile, G. Muscas, M. Murgia, M. S. Andersson, G. Singh, R. Mathieu, P. Nordblad, P. C. Ricci, D. Peddis, K. N. Trohidou, J. Nogués, J. A. De Toro, *Chem. Mater.* **32** (2020) 969.

# Effects of Pyrolysis on the Magnetic Structure and Dipolar Interactions in Iron Oxide Mesocrystals

*Leonhard Rochels[1,2]*, Bertold Rasche[1], Sabrina Disch[1,2]*

[1] *University of Cologne, Department of Chemistry, Cologne, Germany*
[2]*University of Duisburg-Essen, Institute of Inorganic Chemistry, Essen, Germany**
leonhard.rochels@uni-koeln.de

Self-organization of monodisperse nanoparticles into ordered mesostructures is a promising technique for exploration of novel functional materials [1,2]. The face-to-face orientation of nanocubes into mesocrystals [3-5] gives rise to a variety of arrangements with directionally anisotropic properties [6,7]. In response to rising demands for environmentally sustainable materials for Li-ion batteries, mesocrystals of iron oxide nanoparticles have been suggested as potential nanostructured electrode materials [8]. The self-assembly approach using nanoparticles as building blocks allows for tuning the magnetic and electrochemical behavior by variation of the nanoparticle size, morphology, composition, as well as the superlattice structure. In addition, iron oxide fulfills the criteria of cost-efficiency, abundance, and durability. Current challenges, however, arise from the low electric conductivity of the mesocrystals containing large amounts of organic material.

In this contribution, we highlight the potential of pyrolysis techniques to tune the magnetic interactions and enhance the electrochemical surface activity simultaneously. Pyrolysis transforms the organic ligand shell covering the particles while maintaining the overall mesocrystal habit, thus enhancing surface activity as well as the conductivity. Grazing-incidence SAXS and macroscopic magnetization measurements provide insights into the evolution of structural and magnetic properties as an effect of the pyrolysis conditions. A thorough understanding of pyrolysis-induced effects on the magnetic structure and dipolar interactions in iron oxide mesocrystals, of the decomposition pathways, and the electrochemical performance of the formed materials is crucial to optimize their fabrication and realize novel electrode materials with tailored properties.

**Acknowledgements**

The authors acknowledge the BMBF for financial support (Röntgen-Ångström Cluster project nPDFSAS, 05K20PKA).

**References**

[1] M. Boles, M. Engel, D. V. Talapin, *Chem. Rev*. **116** (2016) 11220-11289.
[2] E. V. Sturm, H. Cölfen, *Crystals* **7** (2017) 207.
[3] S. Disch, E. Wetterskog, *et al.*, *Nano Lett*. **11** (2011) 1651-1656.
[4] S. Disch, *et al.*, *Nanoscale* **5** (2013) 3969.
[5] E. Wetterskog, A. Klapper, S. Disch, *et al.*, *Nanoscale* **8** (2016) 15571.
[6] S. C. Glotzer, M. J. Solomon, *Nat. Mater*. **6** (2007) 557.
[7] S. Polarz, *Adv. Funct. Mater*. **21** (2011) 3214-3230.
[8] E. Uchaker, G. Cao, *Nano Today* **9** (2014) 499–524.

# Impact of surfactant-coat stripping on interparticle interactions in dense magnetic nanoparticle assemblies

*R. López-Martín[1*], P. Maltoni[2], E. H. Sánchez[1], B. Santos Burgos[1], S. S. Lee[3], M.S. Andersson[4], V. Håkonsen[5] D. Peddis[6] , C. Binns[1], R. Mathieu[2], J. A. De Toro[1], P. S. Normile[1]*

[1] *Instituto Regional de Investigación Científica Aplicada (IRICA) and Departamento de Física Aplicada, Universidad de Castilla-La Mancha 13071 Ciudad Real, Spain*

[2] *Department of Materials Science and Engineering, Uppsala University, Box 35, SE-751 03 Uppsala, Sweden*

[3] *Institute of Bioengineering and Nanotechnology, 31 Biopolis Way, The Nanos, Singapore 138669, Singapore*

[4]*Department of Chemistry, Ångström Laboratory, Uppsala University, Uppsala, 75121 Sweden*

[5]*NTNU Nanomechanical Lab, Department of Structural Engineering, Norwegian University of Science and Technology (NTNU), Trondheim 7491, Norway*

[6]*University of Genova, Department of Chemistry and Industrial Chemistry, Nanostructured Magnetic Materials Laboratory, Via Dodecaneso 31, 16146 Genova, Italy*

*raul.lopez@uclm.es

Interparticle interactions in systems of ferrimagnetic iron oxide nanoparticles are often studied by varying the particle concentration from a dilute to a dense regime. In the latter, complex (e.g., collective) behavior is commonly explained by interactions of a magnetic dipolar nature [1,2]. Here, we show how interparticle interactions can be significantly enhanced within the dense regime by a method involving the compaction of magnetic particles whose surfactant shell coating content is partially removed in a prior, wet-chemistry-based step. We obtain a series of dense assemblies in which the particles (maghemite of 5 nm diameter) go from being fully coated (29 % oleic acid, by mass) to increasingly barely coated (down to 8 % ). A modest increase in particle packing fraction (a relative increase of up to 70 %) is found along the series from density measurements and is also inferred from small-angle x-ray scattering data. Therefore, we expect a proportional, similarly moderate, rise in interparticle dipolar interaction strength [3]. In zero field cooled magnetization curves, however, we observe a huge enhancement in the peak temperature (by up to 800 % along the series), too large to be accounted for by the estimated rise in dipolar strength. Furthermore, at low temperature the magnetic field response hardens along the series, with the coercive field following the same trend as various interaction indicators. We suggest that superexchange between particles in direct contact, owing to the presence of uncoated surface regions, becomes the dominant interaction mechanism, accounting for the hardening and driving a collective (superspin glass) transition that emerges along the series. As the OA content decreases, we expect the number density of such contact points to increase within an assembly (as an increasingly greater fraction of each particle's surface is left uncoated), leading to an enhancement in the contribution from interparticle superexchange to nanoparticle magnetic stability.

## References

[1] J. A. De Toro et al., J. Phys. Chem. C, vol. 117, no. 19, pp. 10213–10219, May 2013
[2] E. H. Sánchez et al., Small, p. 2106762, Jun. 2022
[3] S. Mørup, Europhys. Lett., vol. 28, no. 9, p. 671, 1994.

# Simultaneous Coercivity and Size Determination of Magnetic Nanoparticles

*Annelies Coene*[1], *Jonathan Leliaert*[2*]

[1] Dept. of Electromechanical, Systems and Metal Engineering, Ghent University, Technologiepark 131, 9052 Zwijnaarde, Belgium

[2] *Dept. of Solid State Sciences, Ghent University, Krijgslaan 281/S1, Ghent, Belgium*

*jonathan.leliaert@ugent.be

Magnetic nanoparticles are increasingly employed in biomedical applications such as disease detection and tumor treatment. To ensure a safe and efficient operation of these applications, a noninvasive and accurate characterization of the particles is required. In this work[1], a magnetic characterization technique is presented in which the particles are excited by specific pulsed time-varying magnetic fields. This way, we can selectively excite nanoparticles of a given size so that the resulting measurement gives direct information on the size distribution without the need for any a priori assumptions or complex postprocessing procedures to decompose the measurement signal. This contrasts state-of-the-art magnetic characterization techniques. The possibility to selectively excite certain particle types opens up perspectives in "multicolor" particle imaging, where different particle types need to be imaged independently within one sample. Moreover, the presented methodology allows one to simultaneously determine the size-dependent coercivity of the particles. This is not only a valuable structure–property relation from a fundamental point of view, it is also practically relevant to optimize applications like magnetic particle hyperthermia. We numerically demonstrate that the novel characterization technique can accurately reconstruct several particle size distributions and is able to retrieve the coercivity–size relation of the particles. The developed technique advances current magnetic nanoparticle characterization possibilities and opens up exciting pathways for biomedical applications and particle imaging procedures.

## Reference

Coene, A.; Leliaert, J. Simultaneous Coercivity and Size Determination of Magnetic Nanoparticles. *Sensors* 2020, *20*, 3882.

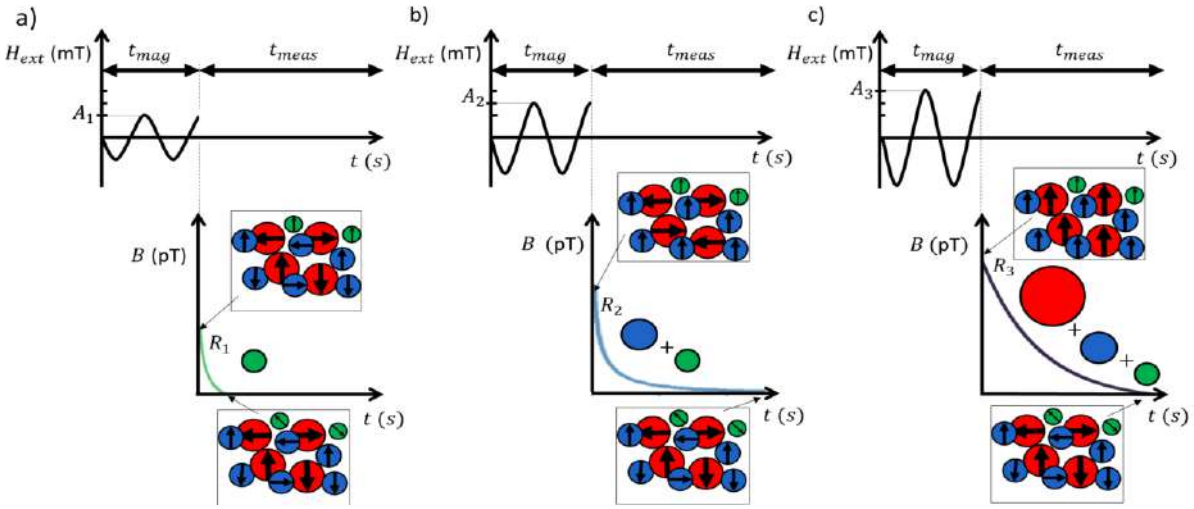



Figure: Overview of how the amplitude of an externally applied alternating magnetic field determines how particles with a) a small coercivity b) small and medium, and c) small, medium and large coercivity are aligned with the external field direction. In a second step, their size-dependent magnetic relaxation is measured, allowing to determine the relation between the particle size and coercivity.

# Magnetic Excitons, Spin Waves and Crystalline Electric Field of $RCu_2$ (R = Nd, Tb) Nanoparticles

*E. M. Jefremovas[1,2], M. de la Fuente Rodríguez[1], B. Fåk[3], J. A. Blanco[4], L. Fernández Barquín[1*]*

[1]*Dpto. CITIMAC, Universidad de Cantabria, 39005 Santander, Spain*
[2]*Institute of Physics, Johannes Gutenberg University Mainz, 55099 Mainz, Germany*
[3]*Institut Laue-Langevin, CS20156, 38042 Grenoble Cedex 9, France*
[4]*Dept. de Física, Universidad de Oviedo, 33007 Oviedo, Spain*

*luis.fernandez@unican.es

The atomic structure of magnetic nanoparticles has been anaysed thoroughly over three decades via X-ray diffraction and transmission electron microscopy (TEM). Their magnetic arrangement is much less known in detail at a microscopic level although studies using X-ray absorption techniques, Mössbauer spectroscopy and neutron diffraction have been largely employed, among others [1]. It is surpising though that the attention to magnetic collective excitations (spin waves  magnetic excitons) and crystalline field (CEF) studies is practically absent in the community, even if this basic knowledge is connected to potential applications. The main reason for this is that the only technique allowing to attain the required information is inelastic neutron scattering (INS). The access to this technique is limited, usally focused on single crystal analyses, and requires the use of a large mass of nanoparticles (around 10g) to obtain a good signal-to-noise ratio. Such a requirement is challenging for the common synthesis routes. Shall the experimental hindrances be overcome, the INS data analysis is also depedent on a profound knowledge of complex CEF and collective excitations. These are not easy to analyse, despite their crucial interest for condensed matter physics in general [2].

Since some years ago we have tackled this problem and started a research line which might be of interest in future Magnetism. For this we have selected the $RCu_2$ (approx. 10-20 nm) alloy nanoparticles ($R$ = Rare Earth), where a conventional antiferromagnetic order in the core coexists with a surface spin disorder, as revealed by macroscopic (DC-magnetisation and AC-susceptibility, specific heat) and microscopic analyses (TEM, X-ray, and neutron and small-angle neutron diffraction) [3-6]. Beamtime was granted in both IN4 and IN6 instruments located at the Institut Laue-Langevin facility (Grenoble, France) to measure $TbCu_2$ ($Tb^{3+}$, $J = 6$) and $NdCu_2$ ($Nd^{3+}$, $J = 9/2$) nanoparticle ensembles. Additional measurements on the non-magnetic $Tb_{0.1}Y_{0.9}Cu_2$ compound were performed to distinguish the CEF excitations from the magnetic ones. The analyses of the data, measured at both above and below $T_N$ (42 and 6 K, respectively) have reavealed in both $TbCu_2$ and $NdCu_2$ the key role of the surface in the propagation of collective excitations (spin waves and excitons), as there lower dimensionality and the reduced coordination of the surface magnetic moments act in favour of the magnetic exchange interactions. An overall appraisal of the results will be given in this communication together to figuring out intringuing ideas to unveil the dispersion of magnetic excitations and the connection of these ensembles of nanoparticles to systems where the magnetic structures and dynamics are complex [6, 7, 8].

## Acknowledgements

This work was supported in part by the Spanish MCIN/AEI under Projects MAT2017–83631–C3–R and PID2020–115704RB– C33. The work of E. M. Jefremovas was supported by the “Concepción Arenal Grant" (Gobierno de Cantabria). The authors would like to thank the ILL (international neutron facility) and staff for the allocation of beamtime and assistance during the experiments.

## References

[1] A. A. El-Gendy *et al.*, *Magnetic nanostructured materials* (Elsevier, 2018).
[2] S. N. Klausen *et al.*, Phys. Rev. B **70**, 214411 (2004).
[3] C. Echevarria-Bonet *et al.*, Phys. Rev. B **87**, 180407 (R) (2013).
[4] E. M. Jefremovas *et al.*, Phys. Rev. B **104**, 134404 (2021).
[5] E. M. Jefremovas *et al.*, Sci. Rep. **12**, 9733 (2022).
[6] A. Michels *et al.*, Phys. Rev. B **99**, 014426 (2019).
[7] M. Morin *et al.*, Nat. Comm. **10**, 1038 (2016).
[8] E. M. Jefremovas *et al.*, (submitted Comm. Mat, 2023).

# Energy-efficient control of magnetization switching in a biaxial nanoparticle

*Mohammad Badarneh*[1*], *Grzegorz Kwiatkowski*[1], *Pavel Bessarab*[1,2]
[1] *University of Iceland, Reykjavik, Iceland*
[2] *Department of Physics and Electrical Engineering, Linnaeus University, Kalmar, Sweden*
*mha5@hi.is

Control of magnetization switching is of critical importance for the development of novel technologies based on magnetic materials. Here we identify by means of optimal control theory energy-efficient protocols for magnetization switching in nanoparticles with uniaxial and biaxial anisotropy [1, 2], see Fig. A. Optimal control paths minimizing the energy cost of magnetization reversal are calculated as functions of the switching time and materials properties, and used to derive energy-efficient switching pulses of external magnetic field. Hard-axis anisotropy reduces the minimum energy cost of magnetization switching due to activation of the internal torque in the desired switching direction, see Fig. B. Analytical estimates quantifying this effect are obtained based on the perturbation theory. The optimal switching time providing a tradeoff between fast switching and energy efficiency is obtained. The energy cost of switching and the energy barrier between the stable states can be tuned independently in a biaxial nanomagnet. This provides a solution for the dilemma between energy-efficient writability and good thermal stability of magnetic memory elements.

## Acknowledgements

This work was supported by the Icelandic Research Fund (Grant Nos. 217813 and 184949), the University of Iceland Research Fund (Grant No. 15673), and the Swedish Research Council (Grant No. 2020-05110).

## References

[1] G. J. Kwiatkowski et al., Phys. Rev. Lett. **126**, 177206 (2021).

[2] M. H. Badarneh et al., *arXiv preprint arXiv:2210.13514*.

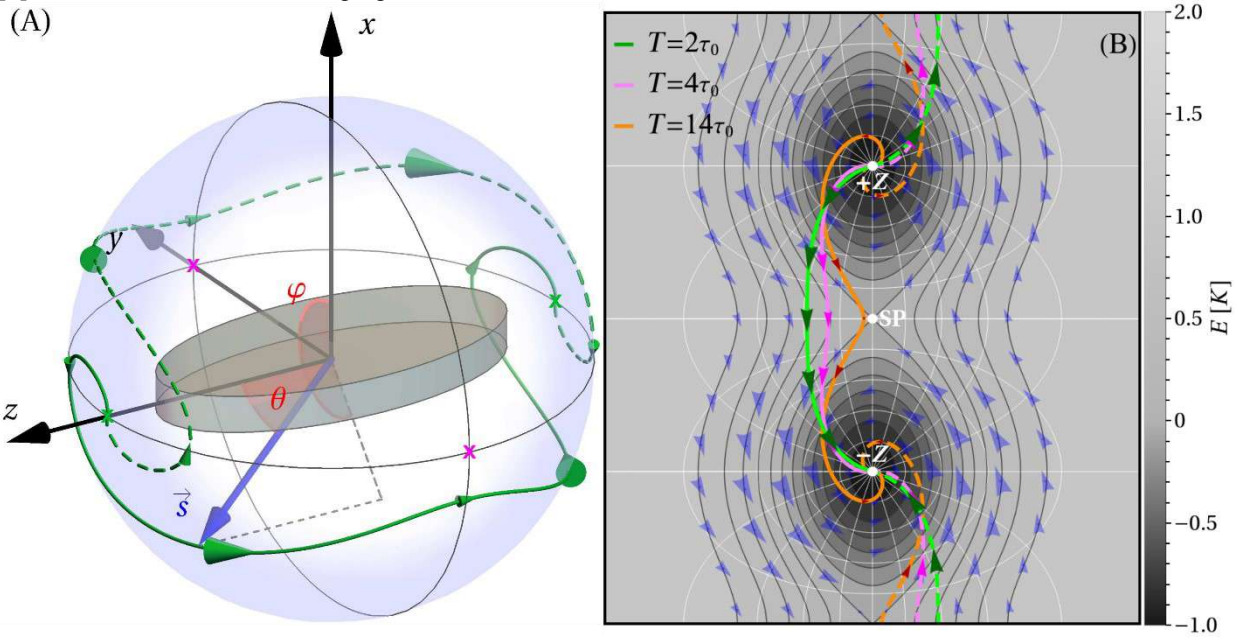



Fig. (A): Optimal switching of a flat elongated nanomagnet representing a biaxial anisotropy system. The single-domain nanoparticle is shown schematically by the thin flat elongated shape. The direction of the normalized magnetic moment is shown with the blue arrow. Orientations of the magnetic moment that correspond to the minima and the saddle points on the energy surface are marked with the green and magenta crosses, respectively. The calculated optimal control paths (OCPs) between the energy minima are shown with the solid and the dashed green lines. Fig. (B): Distribution of the torque, blue arrows, superimposed on the contour plot showing the energy surface of the system. The green, pink, and orange lines show the calculated OCPs between the energy minima at $+Z$ and $-Z$ for various switching time $T$ as depicted in the legend. The notation (SP) on the energy surface corresponds to the saddle point.

# Machine learning based identification of magnetization states in large-scale nano-ellipses arrays

*Hubert Brueckl[1]*, Astrit Shoshi[1], Thomas Schrefl[1], Michael J. Haslinger[2], Tina Mitteramskogler[2], Stefan Schrittwieser[3]*

[1] *Dept. for Integrated Sensor Systems, Danube University Krems, Wiener Neustadt, Austria*
[2] *PROFACTOR GmbH, Steyr-Gleink, Austria*
[3] *AIT Austrian Institute of Technology, Vienna, Austria*
*hubert.brueckl@donau-uni.ac.at

Magnetic nanoparticles operating in a magnetic vortex configuration are of interest for magnetic field sensors [1] and as magnetic nanoprobes for immunoassay diagnostics [2]. In both applications, vortex states prove to be more advantageous compared to single- and multi-domain magnetization states.

We use nanoimprint and thin film deposition to reliably fabricate and stabilize magnetic spin textures in large-scale nanoparticle arrays in a controlled manner. The periodically arranged nanoparticles are quasi-monodisperse elliptically shaped cylinders (400nm x 200nm with a periodicity of 600nm x 400nm) with highly uniform geometric and magnetic properties (figure). The size distribution is very narrow with a standard deviation of 3%. If the thickness of the $Ni_{80}Fe_{20}$ and $Co_{60}Fe_{20}B_{20}$ cylinders is increased from 5 to 60 nm, the magnetization transitions from a quasi-single domain state to a vortex state. The system is anisotropic.

The large-area arrays allow to measure first order reversal curves (FORC) in a vibrating sample magnetometer. The FORC plots show partially mixed magnetization states (figure on the right). To automatically extract the fraction of vortex and single-domain states, machine learning (ML) models are trained with both experimental and simulated data. The latter are computed from a kind of hysteron model. Results are presented on how well the different magnetization states can be identified and distinguished by the ML model. The quality of the hysteron model is demonstrated.

## References

[1] D. Suess, A. Bachleitner-Hofmann, A. Satz, H. Weitensfelder, C. Vogler, F. Bruckner, C. Abert, K. Prügl, J. Zimmer, C. Huber, S. Luber, W. Raberg, T. Schrefl, H. Brückl, Nature Electronics 1, 362–370 (2018)

[2] H. Brueckl, A. Shoshi, S. Schrittwieser, B. Schmid, P. Schneeweiss, T. Mitteramskogler, M.J. Haslinger, M. Muehlberger, J. Schotter, „Nanoimprinted multifunctional nanoprobes for a homogeneous immunoassay in a top-down fabrication approach", Sci. Rep. 11, 6039 (2021)

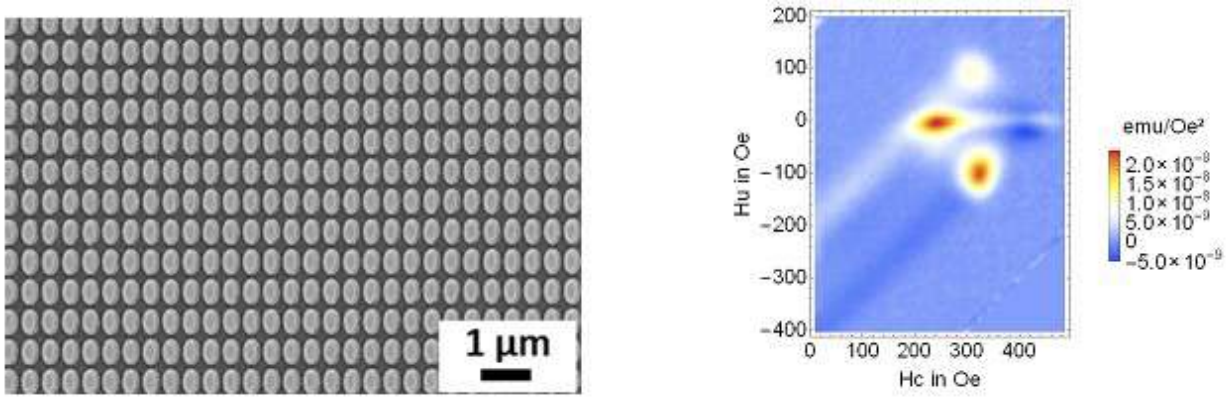



Figure: Left: Scanning electron microscopy image of a nano-imprinted array of elliptical cylinders. The total number of ellipses is more than $10^8$ on 1 $cm^2$. Right: FORC diagram of a mixed magnetization state with 40 nm thick NiFe cylinders.

# Room Temperature Spin-Phonon Coupling in $Cr_2O_3$ Nanocrystals

*M. Testa-Anta, Julia N. Majcherkiewicz, V. Salgueiriño*[1,2*]

[1] *Departamento de Física Aplicada, Universidade de Vigo, 36310 Vigo, Spain*

[2] *CINBIO, Universidade de Vigo, 36310 Vigo, Spain*

*vsalgue@uvigo.es

Nanocrystals of antiferromagnetic $Cr_2O_3$ are shown via Raman spectroscopy to display size-dependent lattice dynamics in terms of phonon softening and the occurrence of an exceptionally strong spin-phonon coupling. This effect, which is observed to persist well above the onset of the antiferromagnetic ordering temperature, is ascribed to locally correlated spin fluctuations due to the modulation of the magnetic exchange interactions as the chromium atoms oscillate about their equilibrium position. We find that the size-dependent spin-phonon coupling strength is governed by the competing antiferromagnetic and ferromagnetic interactions, where changes in the surface spin configuration can also play a crucial role. Overall, this work proves the interplay between the crystalline and magnetic structures in 3D antiferromagnets varying the surface-to-volume ratio, and helps establish the fundamentals for a spin-phonon coupling engineering at the nanoscale via a simple route in a very stable and easy to synthesize material. More importantly, it demonstrates the possibility of coupling phononic excitations with the magnetization dynamics at room temperature, offering a highly prospective nanomaterial for the design of novel magnonic devices. [1]

**Acknowledgements**

The authors acknowledge the financial support from, the Spanish Ministerio de Ciencia e Innovación under project PID2020-119242RB-I00 and the European Union H2020-MSCA-RISE-2019 PEPSA-MATE project.

**References**

[1] M. Testa-Anta, J. N. Majcherkiewicz, V. Salgueiriño (in preparation, 2023)

# Surface/Interface effects for size-selected FeRh nanomagnets deposited on perovskite oxide crystals

*F. Tournus[1]*, G. Herrera[1], S. Gonzalez[2], I. Canero-Infante[2], , P. Schoeffmann[3], P. Ohresser[3], E. Otero[3], M. Bugnet[4], A. Reyes[5], A. Vlad[3], A. Resta[3], A. Coati[3], X. Weng[6], G. Renaud[6], O. Boisron[1], A. Tamion[1], L. Bardotti[1], D. Le Roy[1] and V. Dupuis[1]*

[1] *Institut Lumière Matière, CNRS, Univ. Lyon 1, Villeurbanne, France*
[2] *Institut des Nanotechnologies de Lyon, CNRS, ECL INSA, Villeurbanne, France*
[3] *Synchrotron SOLEIL, L'Orme des Merisiers, Saint Aubin, Gif-sur-Yvette, France*
[4] *MATEIS, CNRS, Univ. Lyon 1, INSA Lyon, Villeurbanne, France*
[5] *Faculty of Science, Universidad Autonoma del Estado de México, Toluca, México*
[6] *Université Grenoble Alpes, CEA, IRIG-MEM, Grenoble, France*
*florent.tournus@univ-lyon1.fr

The major importance of surface atoms in small nanoparticles (NPs) offers the opportunity to tailor magnetic properties by playing with the interface between nanomagnet and its surrounding. FeRh alloy has attracted a lot of attention because the bulk material presents an antiferromagnetic to ferromagnetic order (AFM-FM) transition close to room temperature, for the chemically ordered B2 phase. Inspired by epitaxial FeRh film studies on perovskite oxide [1], and motivated by the possibility to obtain hybrid multiferroic nanosystems, we have studied the structural and magnetic properties of size-selected FeRh clusters (diameter < 10 nm) deposited on perovskite oxide surfaces. For this system, a strong interplay between surface configuration, morphology and magnetic state is taking place [2,3]. FeRh nanomagnets have been deposited on $BaTiO_3$ thin films and $SrTiO_3$ single crystals, using the mass-selected low energy cluster beam deposition technique (MS-LECBD) under ultra-high vacuum. Using synchrotron radiation, we have observed the chemical ordering of FeRh nanoparticles into the B2 crystalline phase upon annealing, which is also accompanied by a Fe magnetic moment evolution visible from X-ray magnetic circular dichroism (XMCD) measurements. The orientation dependence of x-ray diffraction FeRh peaks indicates that particles, despite their random deposition, are finally adopting preferential orientations. In addition to the usual epitaxy relationship met for thin films, a novel orientation is observed (corresponding to a 45° in-plane rotation), as well as other favorable coincidences for particles on $SrTiO_3$. At the same time, X-ray spectroscopy at iron edges reveals that NPs assemblies, systematically appear to be (partially) oxidized after transfer in air while FeRh nanoparticles can be reduced thanks to in situ vacuum annealing. Concerning magnetic behavior, as for previous FeRh NPs embedded in carbon matrix [2], no metamagnetic (AFM-FM) phase transition has been observed from XMCD measurements. This shows that despite the existence of well-defined atomic contact between FeRh NPs and $SrTiO_3$ substrate, epitaxial strain at the interface does not seem to be sufficient to counterbalance strong FM exchange interactions between Rh and Fe in nanoalloys.

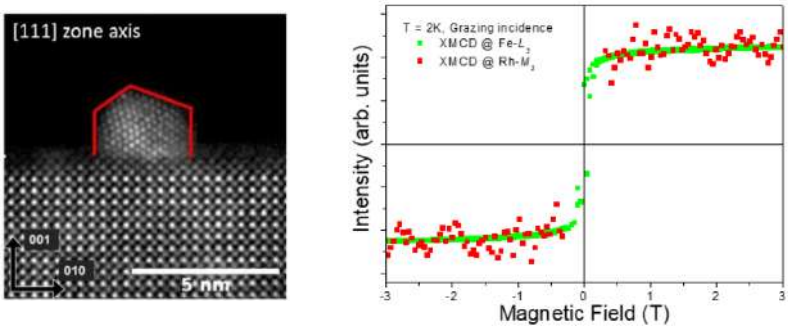



Figure: cross section image (STEM-HAADF) of a FeRh nano-particle on the $SrTiO_3$ surface (left) ; hysteresis loop at low temperature (2 K) from the XMCD signal at the Fe $L_3$ and Rh $M_3$ edges, showing that the particles are still displaying a FM order, with a ferromagnetic coupling between Fe and Rh.

## References

[1] Cherifi et al., *Nature Mater.* **13**, 345 (2014) ; Liu et al., *Nature Comm.* **7**, 11614 (2016).
[2] Hillion et al., *Phys. Rev. Lett.* **110**, 087207 (2013) ; Herrea et al. *Eur. Phys. J. Appl. Phys.* **97**, 32 (2022).
[3] Liu et al., *Euro. Phys. Lett.* **116**, 27006 (2016) ; Lewis et al., *J. Phys. D: Appl. Phys.* **49**, 323002 (2016) ;

# 3D-Printing of Magnetic Objects Using Composite Inks Based on Ferromagnetic Particles and Hydrogels

*Zaida Curbelo-Cano*, Ester M. Palmero, Cristina M. Montero, Javier de Vicente, and Alberto Bollero*

*Group of Permanent Magnets and Applications, IMDEA Nanoscience, 28049 Madrid, Spain*

* zaida.curbelo@imdea.org

Nowadays, additive manufacturing technologies based on composite materials are attracting much interest in many sectors (electronics, aerospace, medicine…) due to the possibility of fabricating functional objects with tailored properties, high-performance and complex designs. 3D-printing allows for efficient, fast and precise fabrication combined with a reduced waste generation compared to traditional methods [1]. Depending on the final application (i.e., required properties), the use of composites with different filler content - homogeneously dispersed along the matrix- will be required. Recent works using metallic/polymer composites have succesfully proven the potential of combining 3D-printing with the development of composites, while preserving the properties of the filler along the process [2,3]. On the other hand, hydrogel-based composites have been used in 3D-printing technology due to their interesting and tunable structural properties, combined with a broad range of available materials for applications like flexible and soft electronics or biomedicine [4,5].

In this work, Direct Ink Writing (DIW) 3D-printing has been used to fabricate objects with self-synthesized composite inks (Fig. 1a). Gas-atomized MnAlC particles (in particular, the so-called τ-MnAlC phase) have been used as filler and a hydrogel as matrix. By tuning and optimizing the necessary additives [4], it was possible to prepare highly loaded inks exceeding 50 wt.% of MnAlC particles (Fig. 1a). Magnetic characterization made by Vibrating Sample Magnetometry (VSM) showed that coercivity ($H_c$) remains constant after the composite ink synthesis in comparison to the starting particles and the magnetization scales according to the particles content (Fig. 1b). Objects with a variety of sizes and shapes have been succesfully printed by DIW (Fig. 1c). VSM measurements show that after 3D-printing and post-processing, the magnetic properties of the MnAlC particles are not deteriorated. The resulting 3D-printed objects show structural flexibility and mechanical strength. These results open the path to future developments of this additive manufacturing technology to be applied in high-tech sectors where parts with complex shapes and tuned properties made of magnetic/metallic composites are required.

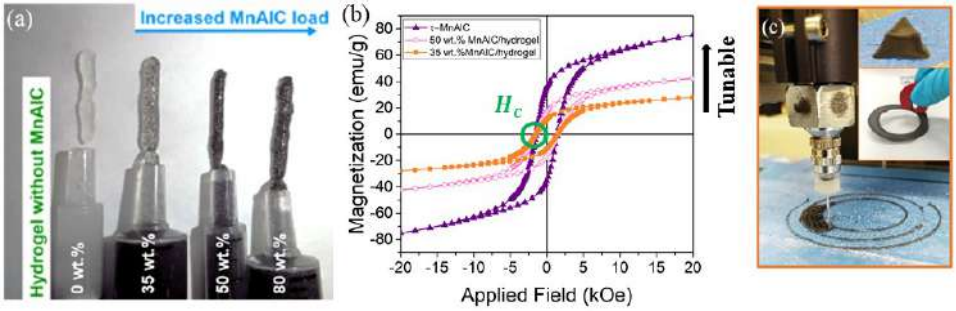



Figure 1: (a) Composite inks with different MnAlC particles load; (b) Room temperature hysteresis loops measured for MnAlC/hydrogel composites with 35 wt% and 50 wt% of MnAlC, compared to the hysteresis loop measured for the starting MnAlC powder; and (c) image of the DIW 3D-printing process with insets showing images of 3D-printed magnetic MnAlC (50 wt.%)/hydrogel pieces with different shapes.

## Acknowledgements

Authors acknowledge Höganäs AB (Sweden) for providing the gas-atomized MnAlC particles through an industrial IMDEA-Höganäs collaboration, and the financial support from MICINN by *NEXUS* project (PID2020-115215RB-C21). IMDEA Nanociencia thanks support from the ''Severo Ochoa'' Programme for Centers of Excellence in R&D (MICINN, Grant CEX2020-001039-S). E.M.P. acknowledges support from AEI by the JdC-I program (IJC2020-043011-I/MCIN/AEI/10.13039/501100011033) and EU by NextGenerationEU/PRTR.

## References

[1] L.E. Murr, *J. Mater. Sci. Technol.* **32** (2016) 987.
[2] E.M. Palmero et al.. *Compos. Part A Appl. Sci. Manuf.* **124** (2019) 105497.
[3] E.M. Palmero et al., *Sci. Technol. Adv. Mater.* **19** (2018) 465; *Addit. Manuf.* **33** (2020) 101179.
[4] D. Podstawczyk et al., *Addit. Manuf.* **34** (2020) 101275.
[5] E.M. Palmero and A. Bollero, 3D and 4D printing of functional and smart composite materials, in *"Encyclopedia of Materials: Composites" –Mater. Sci. Mater. Eng. ,* **Vol. 2**, pp. 402-419, Elsevier (2021).

# Locally Modulated Stiffness of Magneto-Active Composites Produced by Laser Powder Bed Fusion

*Kilian Schäfer[1,5]*, Matthias Lutzi,[1,5], Muhammad Bilal Khan[1,5], Sebastian Bruns[2,5], Iman Valizadeh[3,5], Oliver Weeger[2,5], Claas Hartmann[4,5], Mario Kupnik[4,5], Oliver Gutfleisch,[1,5]*

[1] *Functional Materials, TU Darmstadt, 64287, Germany*

[2] *Physical Metallurgy, TU Darmstadt, 64287, Germany*

[3] *Cyber-Physical Simulation Group, TU Darmstadt, 64293, Germany*

[4] *Measurement and Sensor Technology Group, TU Darmstadt, 64289, Germany*

[5] *Additive Manufacturing Center, TU Darmstadt, 64287, Germany*

*kilian.schaefer@tu-darmstadt.de

Mechanically soft sensors and actuators enable a compliant and safe interaction between humans and their environment [1]. Among many existing efforts to develop such devices, magnetic actuation mechanisms are well-known for their fast response, wireless operation and the possibility to operate in confined spaces [2]. With additive manufacturing, the production of magneto-active composites in complex and bioinspired shapes is possible. To mimic the properties of biological systems, the fabrication of composites with locally different mechanical properties is needed. Here, we present a method to locally tailor the stiffness of a magneto-active compound, consisting of 50 weight % hard magnetic $Nd_2Fe_{14}B$ particles in a thermoplastic polyurethane matrix with laser powder bed fusion. By utilizing different laser parameters at different locations during the process, the mechanical properties of the composite can be adjusted locally. The tailorable stiffness range is 2 to 22 MPa. The stiffness gradient within one sample is verified by line scans of Vickers indentations with a nanoindentation system. Then the actuation performance in a homogenous magnetic field is evaluated for samples with and without stiffness gradients. One example where the here proposed method can be utilized is a foldable cube where the hinges can be produced with lower laser power, as indicated in Fig.1 a). The actuation as a function of the applied field is shown in Fig.1 of the locally adjusted shape (a) in comparison to the cube where the composite has a homogenous stiffness (b). As can be seen, the locally adjusted cube reaches a larger degree of deformation at the same actuation field, which helps to reduce the energy consumption in an application.

**Acknowledgements**

This work was financially supported by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation), Project ID No. 405553726, TRR 270 and the RTG 2761 LokoAssist (Grant no. 450821862). The authors thank Denys Makarov for the fruitful discussions.

**References**

[1] G.M. Whitesides, Angew. Chem. Int. Ed Engl. **57** (2018) 4258–4273.
[2] Y. Kim, G.A. Parada, S. Liu, X. Zhao, Sci. Robot. **4** (2019).

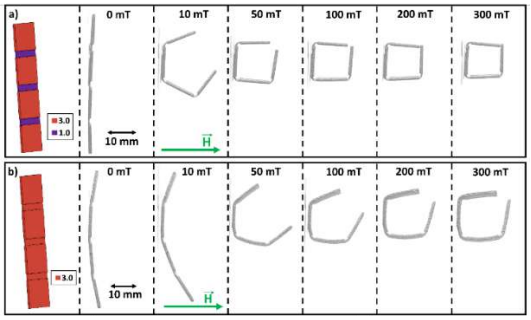



Figure 1: Comparison of magnetically actuated folding cubes at different magnetic actuation fields H. a) Cube with soft segments between the sides, which allows higher degree of folding at the same magnetic actuation fields in comparison to b) where the hinges are produced with the same process parameters.

# Additive Manufacturing Of Magnetocaloric 3D Structures: A Cost-Effective Way For Printing Cellulose-Based Metallic Structures

*Bosco Rodriguez-Crespo*[*1], *Daniel Salazar*[1], *Volodymyr Chernenko*[2]

[1] *BCMaterials, UPV/EHU Scientific Park, Leioa, Spain*
[2] *University of Basque Country, Leioa, Spain*

*bosco.rodriguez@bcmaterials.net

## Abstract

Solid-state refrigeration based on magnetocaloric effect is seen as a potential alternative to current less-efficient gas expansion-compression based conventional refrigeration. The heat exchanger, the functional element of this kind of refrigerator, is composed by the magnetocaloric alloy. Additive Manufacturing (AM) is a useful technique to build efficient heat exchangers, and since current AM techniques are expensive and energy consuming a cost-effective way has to be explored.

In this work, we developed original inks and implemented extrusion-based printing technique[1] to print 3D magnetocaloric structures that would act as the heat exchanger of a magnetic refrigerator, printing at room temperature in a green and a cost-effective way using various metallic powders (including magnetic and magnetocaloric powders), and cellulose as matrix with water as dissolvent (Fig. 1). The magnetocaloric ink containing 90-95 wt.% of powder was elaborated by achieving an optimal viscosity whereby high maximum number of layers (250 layers reached) with highest printing resolution (0.5mm wall thickness) was obtained. The elaborated technological route of the treatment of printed structures included: (i) special heat treatments to dry printed structures so the polymer was removed by calcination followed by a sintering to get entirely metallic structure, and (ii) electrodeposition of nickel to protect printed structure from any corrosion. We also demonstrated that any incorrectly printed workpiece can be recycled re-dissolving it in the water so material loss is reduced significantly making the printing more cost-efficient and environmentally friendly.

## References

[1] C. Xu, Q. Wu, G. L'Espérance, L.L. Lebel, D. Therriault, Environment-friendly and reusable ink for 3D printing of metallic structures, Mater. Des. 160 (2018) 262–269. https://doi.org/10.1016/j.matdes.2018.09.024.

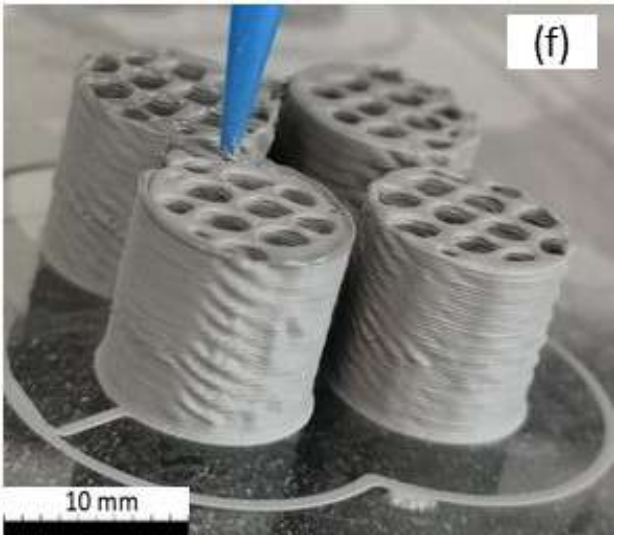



Figure 1: 3D printing magnetocaloric honeycomb-like structures

# Phase Transformations and Stability of $Fe_3N$ Nanoparticles at Elevated Temperatures

*Y. Ablets*, D. Koch, W. Donner, O. Gutfleisch, I. Dirba*

*Technical University of Darmstadt, Alarich-Weiss-Str. 16 64287, Darmstadt, Germany*

*yevhen.ablets@tu-darmstadt.de

Due to increased saturation magnetization values compared to the conventional $Fe_2O_3/Fe_3O_4$-based materials, Fe-$N_x$ and Fe-$C_y$ nanoparticles are promising candidates for magnetic fluid hyperthermia and other biomedical applications [1]. Crucial for applications, yet a so far less explored aspect for these materials, is their thermal and chemical stability. In this work, we study phase transformation and phase stability of $Fe_3N$ nanoparticles at elevated temperatures in oxidative and in inert conditions.

We have synthesized fine, monodisperse $Fe_3N$ nanoparticles with average size of 13.5 nm (Fig. 1a) via the thermal decomposition route. To investigate temperature stability and stability towards oxidation of the $Fe_3N$ nanoparticles, XRD measurements were carried out at elevated temperatures.

$Fe_3N$ particles which were exposed to air for 5 minutes show partial oxidation (Fig. 1e). At the same time, they demonstrate phase stability at 427°C (700 K) as shown in Fig 1d. $Fe_3N$ nanoparticles sealed in air-tight quartz capillary perform differently. Phase transformation from $Fe_3N$ to $Fe_5C_2$ takes place at ~400°C (Fig. 1f). Further increase of temperature leads to formation of $Fe_3C$ and finally formation of Fe at 600°C. Formation of $FeC_x$ nanomaterials happens due to presence of carbon containing surfactant on the surface of the nanoparticles.

To conclude, $Fe_3N$ nanoparticles can be protected from complete oxidation by a thin passivating $Fe_3O_4$ layer. $FeC_x$ phases are forming when heating $Fe_3N$ nanoparticles in absence of $O_2$.

## Acknowledgements

We acknowledge funding by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation), Project ID Nos. 471878653, 405553726-TRR 270 and CRC 1487.

## References

[1] I. Dirba, C. K. Chandra, Y. Ablets, J. Kohout, T. Kmječ, O. Kaman and O. Gutfleisch, J. Phys. D: Appl. Phys. **56** (2023) 025001.

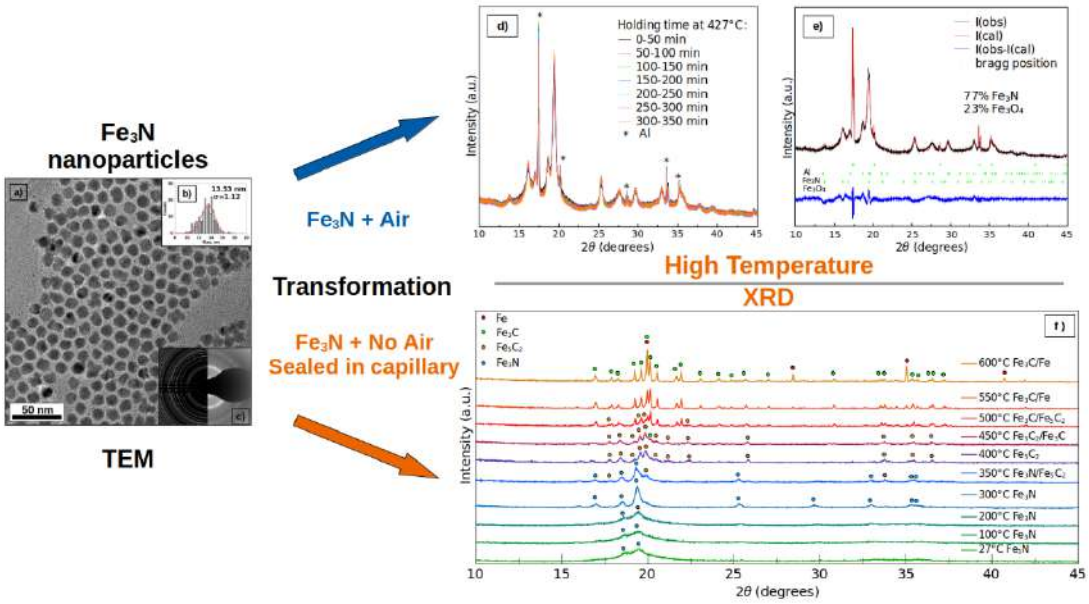



Figure 1: $Fe_3N$ nanoparticles - a) Transmission electron microscopy image of initial particles, b) Particle size distribution, c) Selected area electron diffraction, d) XRD of oxidized $Fe_3N$ holding up to 6 hours at 427°C, e) Rietveld refinement characterizing the oxidized particles, f) XRD of phase transformation at elevated temperatures.

# In-Situ Polymerisation of Ferrofluids Creating a Barium Hexaferrite Nanoparticle Composite Matrix

[1]*Ahmed, Y.*, [2]*Ward, B. D.*, [3]Mertelj, A., [4]Lisjak, D.,*and* [1*]*Zabek, D.*

[1]*School of Engineering, Cardiff University, CF24 3AA, Cardiff, United Kingdom.*

[2]*School of Chemistry, Cardiff University, CF10 3AT, Cardiff, United Kingdom.*

[3]*Department of Complex Matter, Jozef Stefan Institute, SI-1000 Ljubljana, Slovenia*

[4]*Department of Material Synthesis, Jozef Stefan Institute, SI-1000 Ljubljana, Slovenia*

*Zabekd@cardiff.ac.uk

Recently discovered barium hexaferrite ($BaFe_{12}O_{19}$) nano-platelets form a ferromagnetic-ferrofluid which exhibits interesting magnetic properties [1]. Manufacturing the barium hexaferrite nano-platelets via hydrothermal synthesis remains of fundamental interest and allows for optimisation of structural, magnetic, and morphological properties, which all impact on ferrofluid properties. We have developed a hexadecyltrimethylammonium bromide surfactant assisted manufacturing method for ethylene glycol based ferrofluids with barium hexaferrite. The ethylene glycol based ferromagnetic-ferrofluid can be polymerised in-situ via a condensation polymerisation reaction between the ethylene glycol based ferrofluid and succinic acid. The proposed in-situ method creates a homogeneous nano-platelets polyester polymer composite matrix exhibiting anisotropic magnetic properties. The polymer composite matrix was chemically characterised by Gas Chromatography (GPC) and magnetically characterised by Vibrating Sample Magnetometry (VSM). The proposed anisotropic magnetic polymer is highly stable with potential applications in thermal, electrical, medical, and analytical devices.

**Acknowledgements**

This research received funding from the Royal Academy of Engineering (RAEng) under the Research Fellowship Program Number RF\201819\18\202.

**References**

[1] M. Shuai, et *al*., *Nat. Com*. 7(1) (2016) 1-.8.

# On-Chip Funneling and Separation of Superparamagnetic Beads Using Engineered Magnetic Domains as Magnetophoretic Tracks

*Rico Huhnstock[1]*, Lukas Paetzold[1], Maximilian Merkel[1], Piotr Kuświk[2], Arno Ehresmann[1]*

[1] *Institute of Physics and Center for Interdisciplinary Nanostructure Science and Technology (CINSaT), University of Kassel, Heinrich-Plett-Str. 40, 34132 Kassel, Germany*

[2] *Institute of Molecular Physics, Polish Academy of Sciences, M. Smoluchowskiego 17, Poznań 60-179, Poland*

*rico.huhnstock@physik.uni-kassel.de

For the implementation of future Lab-on-a-chip (LOC) devices or micro total analysis systems (μTAS), the controlled actuation of magnetic nano- and microparticles is considered to be highly advantageous since the manipulation can be achieved via magnetic fields without disturbing occurring microfluidic or biological processes [1]. Detection of an analyte species in these systems typically requires appropriately surface-functionalized particles that enable specific binding of the analyte to catcher molecules [2]. It is hereby important to separate and identify particles based on their analyte interaction. As a prototypical methodology, we present in this work the usage of engineered magnetic domain patterns within topographically flat magnetic thin film systems and the dynamic transformation of the emerging magnetic stray field landscape (MFL) via external magnetic field pulses [3,4]. First, we demonstrate a controllable funneling of superparamagnetic beads (SPBs) within an aqueous medium using the MFL originating from parallel magnetic stripe domains of gradually decreasing length. After initiation of lateral bead transport by applying external magnetic field pulses, converging SPB motion trajectories were induced, forming distinct, closely packed particle clusters. As SPBs are brought into proximity to each other within these clusters, a potential route for analyte detection is opened up due to an inducible particle aggregation, with the analyte acting as molecular bridges between single particles [5]. Adjusting the frequency of the external pulse sequence, this magnetophoretic funnel can be jammed for incoming beads. When increasing the stripe domain length after funneling, the formed clusters were observed to reversibly decompose back to the initial state. Secondly, another domain pattern consisting of parallel stripe domains with gradually increasing width will be introduced for the separation of SPBs based on their magnetophoretic mobility. We show that SPBs can be immobilized at different locations above the chip substrate while applying a constant external magnetic field pulse sequence. Herein separated fractions of SPBs are relocatable by adjusting the magnitude of the externally applied magnetic field pulses. The investigated system holds promising potential for pre-sorting batches of commercially available SPBs in dependence on their size and magnetic content. For both investigated systems, it will be discussed how the observed behavior is determined by the acting forces on the SPBs.

## References

[1] M.A.M. Gijs, *Microfluidics and Nanofluidics* **1** (2004) 22-40.
[2] A. van Reenen, A.M. de Jong, J.M.J. den Toonder, M.W.J. Prins, *Lab on a Chip* **14** (2014) 1966-1986.
[3] D. Holzinger, I. Koch, S. Burgard, A. Ehresmann, *ACS Nano* **9** (2015) 7323-7331.
[4] R. Huhnstock, M. Reginka, A. Tomita, M. Merkel, K. Dingel, D. Holzinger, B. Sick, M. Vogel, A. Ehresmann, *Scientific Reports* **11** (2021) 21794.
[5] S. Rampini, P. Li, D. Gandhi, M. Mutas, Y.F. Ran, M. Carr, G.U. Lee, *Scientific Reports* **11** (2021) 5302.

# Magnetic Methods and Particles as Real Green and Sustainable Alternatives

*P. A. Augusto [1,2], A. Rodriguez[1], L. Hernandez[1], T. Castelo-Grande[3], D. Barbosa[3]*

*(1) Dep. Chemical Engineering, Faculty of Chemical Sciences, University of Salamanca, Spain.*
*(2) Institute of Molecular and Cellular Biology of Cancer, GIR Cytomic, Univ. Salamanca,Spain.*
*(3) LEPABE – Laboratory for Process Engineering, Environment, Biotechnology and Energy, Faculty of Engineering, University of Porto, Rua Dr. Roberto Frias, 4200-465 Porto, Portugal*
* pauloaugusto@usal.es

Many of the currently applied methods for waste, wastewater and leachates treatment are not sustainable enough and some of them are not even environmentally-friendly on their own. Magnetic methods are usually based on the application of magnetic forces over magnetic particles by separating them gently and effectively from the mainstream after they have performed the task they were required to do (e.g adsorption of contaminants, nutrients, catalysis of chemical reactions, etc.); therefore, several applications of these methods and particles have been studied and researched in the last decade, in order to allow the replacement of less sustainable and green technologies by them. Nonetheless, although several alternatives have been pointed out they have hardly reached real large-scale applications, this is mainly caused by the use of expensive and technologically complicated structured magnetic particles, and by the rarity of studies considering their efficient removal, regeneration and re-use (afterall some of their potentially associated main advantages). We have studied and successfully applied several methods to synthesize low-cost, easy-to-produce magnetic particles (maintaining a high enough efficiency) and also several regenerating methods, as well as cycles of their reuse, improving and determining their high-impact sustainability.

**Material and Methods – *Magnetic nano and microparticles synthesis***: The manufacture of magnetic particles was performed usually by co-precipitation methods, although some other methods have been applied. The magnetic particles obtained are all magnetite-core based with an organic shell coating in several cases. ***Applications***: the main applications studied were the removal or degradation of solids, organic matter and heavy metals from leachates, wastewaters and drinking water by adsorption, flocculation, (photo-) Fenton heterogeneous catalysis. ***Regeneration methods***: several magnetic particles regeneration methods were studied according to the application under question, e.g. watery based methods and chemical solvent methods (when required) when absorption was in order, milling-decantation and chemical destabilization in the case of flocculation, reduction of the magnetic catalyst in the case of Fenton reactions. ***Re-use***: after the application of particles cycles of re-use were studied allied with the regeneration when needed. ***Economical analysis***: In several of the cases the research study was accompanied by a full economical and technological viability analysis.

**Results and Discussion** [1-4]– Some results are presented in this section as examples and the full set presented in the congress. In Image 1 is presented the SEM analysis of a synthesized nanomagnetic particle, while in Table I are presented some efficiency results of the particles and processes in some of the applications tested, and in Table II some of the results obtained concerning cases of reuse (including some regeneration steps when required.

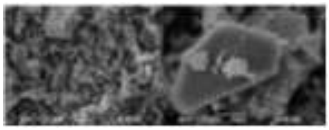


**Image 1.** Magnetic particles synthesized

**Tabla I.** Magnetic Processes Efficiency

| Process | Efficiency |
|---|---|
| Floculation Wastewater | 96 % (TSS); 91 % (COD) |
| Fenton Wastewater | Tirasol- 53% (Fent); 82% (Phot) TOC – 91,7 % (PhotoFenton) |
| Sorption Leachates | 45% (COD); 89% ($NH_4$+); 100% ($NO_3$-); 55% ($PO_4^{2-}$) |

**Tabla II.** Reusing cycles

| Process | Nr cycles | Min efficiency |
|---|---|---|
| Floculation Wastewater | 10 | 92% (TSS); 90% (COD) |
| Fenton Wastewater | 8 | 88% (COD) |
| Sorption Leachates | 8 | 40% (COD); 90% ($NO_3$-) |

**Conclusions –** We have developed and researched successfully the application of low-cost magnetic particles and methods in the treatment of water, wastewaters and leachates, using regeneration and re-use of the particles to efficiently improve their sustainability.

**Acknowledgements**
This research was funded by: (a) Project 2019/00057/001, “Application of Magnetic Methods and Magnetic Substances for Environmental Treatment and Biomedicine”, 26 RLD-PXIV, USAL, Spain. (b). Base Funding—UIDB/00511/2020 of the Laboratory for Process Engineering, Environment, <Biotechnology and Energy—LEPABE—Funded by national funds through the FCT/MCTES (PIDDAC); (c). Project POCI-01-0145-FEDER-028020—Funded by FEDER funds through COMPETE2020—Programa Operacional Competitividade e Internacionalização (POCI) and by national funds (PIDDAC) through FCT/MCTES; (d) Project 2SMART – engineered Smart materials for Smart citizens, with reference NORTE-01-0145-FEDER-000054, supported by Norte Portugal Regional Operational Programme (NORTE 2020), under the PORTUGAL 2020 Partnership Agreement, through the European Regional Development Fund (ERDF).

**References**

[1] - T. Castelo-Grande, P.A. Augusto et al., Journal of Environmental Management, 281, (2021) 111872. [2] - L. Hernández, P.A. Augusto et al., Journal of Environmental Management, 285, (2021) 112155. [3] - P.A. Augusto, T. Castelo-Grande et al., Science of the Total Environment, 648, (2018), p.636. [4] - P.A. Augusto, T. Castelo-Grande et al. Materials, 13 (10), (2020), 2219

# Room Temperature Ferroelectricity and Antiferromagnetism in γ-$BaFe_2O_4$

*Fabio Orlandi,[1*] Davide Delmonte,[2] Gianluca Calestani,[3] Enrico Cavalli,[3] Edmondo Gilioli,[2] Vladimir V. Shvartsman,[4] Patrizio Graziosi,[5] Stefano Rampino,[2] Giulia Spaggiari,[2,6] Chao Liu,[7,8] Wei Ren,[7] Silvia Picozzi,[8] Massimo Solzi,[6] Michele Casappa,[2,3] Francesco Mezzadri[2,3]*

[1] *ISIS neutron and muon facility, STFC, Rutherford Appleton Laboratory, Harwell Campus, Didcot, OX11 0QX, UK;*

[2] *IMEM-CNR, Parco Area delle Scienze 37/A, 43124, Parma, Italy;*

[3] *Department of Chemistry, Life Sciences and Environmental Sustainability, University of Parma, Parco Area delle Scienze 17/A, 43124, Parma, Italy;*

[4] *Institute for Materials Science and Center for Nanointegration Duisburg-Essen (CENIDE), University of Duisburg-Essen, Universitätsstrasse 15, 45141, Essen, Germany;*

[5] *CNR-ISMN, Via P. Gobetti 101, 40129, Bologna, Italy*

[6] *Department of Mathematical, Physical and Computer Sciences, University of Parma, Parco Area delle Scienze 7/A, Parma, Italy*

[7] *Physics Department, International Center of Quantum and Molecular Structures, Materials Genome Institute, State Key Laboratory of Advanced Special Steel, Shanghai Key Laboratory of High Temperature Superconductors, Shanghai University, Shanghai, 200444, China;*

[8] *CNR-SPIN, Unità di Ricerca presso Terzi c/o Università "G. D'Annunzio", 66100, Chieti, Italy;*

*fabio.orlandi@stfc.ac.uk

Room temperature magnetoelectric materials, showing the coexistence of magnetism and ferroelectricity, could lead to a revolution in multifunctional devices. However, only few examples are known, and bismuth ferrite is currently the most used material for multiferroics devices R&D. In this talk, we demonstrate that γ-$BaFe_2O_4$ shows ferroelectricity and antiferromagnetism at room temperature with high critical temperatures. [1]

γ-$BaFe_2O_4$ crystallize in the polar space group $Cmc2_1$ with a stuffed tridymite-type structure constituted by $FeO_4$ tetrahedra arranged in hexagonal rings with the tetrahedra apical oxygens placed in an up-up-up-up-up-down (UUUUUD) pattern. The cavities running along the [100] direction contain the $Ba^{2+}$ ions. Macroscopic measurements show an antiferromagnetic transition at $T_N = 890$ K whereas electrical measurements, confirmed by piezo force microscopy, show a switchable electrical polarization at room temperature. Powder neutron diffraction confirm both the antiferromagnetism and the polar structure. [1]

Symmetry analysis and density functional theory calculations, with respect to a latent parent structure with the *P6/mmm* symmetry, indicate that the observed polarization is improper in origin. It is indeed, induced by the presence of two non-polar distortions: the ordering of the $FeO_4$ apical oxygens with the UUUUUD pattern, and the in-plane rotation of the same units. The free energy coupling responsible for the appearance of ferroelectricity is bilinear-quadratic $F_{coup}=\rho\xi\zeta^2$, with the quadratic term ζ describing the UUUUUD ordering and the linear term describing the in-plane $FeO_4$ rotations ξ and the polarization ρ. The form of the coupling term, which is significantly different from the well know trilinear term in hybrid-improper ferroelectrics, indicate that the polarization switching path requires only the change of sign of the in-plane $FeO_4$ tetrahedra rotation, leaving the UUUUUD pattern unaltered.

Neutron diffraction measurements reveal that the long-range antiferromagnetic ordering is a G-type magnetic structure which does not allow any weak ferromagnetic moment. Nevertheless, symmetry analysis show that it is possible to construct a Lifshitz invariant similar to the one present in $BiFeO_3$ indicating an instability towards an incommensurate state. This state is not observed in γ-$BaFe_2O_4$ probably due to the competition with other interactions like single ion anisotropy or Dzyaloshinskii–Moriya interaction. [1]

Our study suggests not only γ-$BaFe_2O_4$ but the wider class of the stuffed tridymite materials as a new playground for the search of room temperature magnetoelectric materials.

## References

[1] Orlandi, F. et al. γ-$BaFe_2O_4$: a fresh playground for room temperature multiferroicity. *Nat. Commun.* (2022) **13**, 7968. https://doi.org/10.1038/s41467-022-35669-5

# High-Resolution Imaging of Cycloidal Antiferromagnetic Order in Bismuth Ferrite with Soft X-Ray Ptychography

*Tim A. Butcher[1]*, Nicholas W. Phillips[1], Simone Finizio[1], Manuel Langer[1], Michal Jambor[2], Elisabeth Müller[1], Ashna Bajpai[3,4], Carlos A. F. Vaz[1], Armin Kleibert[1] and Jörg Raabe[1]*

[1] *Paul Scherrer Institut, 5232 Villigen PSI, Switzerland*

[2] *Institute of Physics of Materials, Czech Academy of Sciences, Žižkova 22, 61600 Brno, Czech Republic*

[3] *Department of Physics, Indian Institute of Science Education and Research, Pune 411008, India*

[4] *Centre for Energy Science, Indian Institute of Science Education and Research, Pune 411008, India*

*tim.butcher@psi.ch

Bismuth ferrite possesses both ferroelectric and antiferromagnetic order at room temperature, which makes it the archetypal multiferroic material. A further peculiarity of bismuth ferrite is the arrangement of its antiferromagnetically aligned magnetic moments in the form of a cycloid with a period of approximately 62 nm [1]. Microscopic imaging of the cycloidal antiferromagnetic structure requires high sensitity and has hitherto been limited to scanning nitrogen-vacancy magnetometry [2]. Soft X-ray ptychography is an imaging technique, which relies on collecting coherent diffraction patterns from overlapping illumination spots of the sample and can provide a spatial resolution below 10 nm [3]. Ptychographic measurements in the soft X-ray regime, which encompasses the L-edges of the transition metals, benefit from strong X-ray magnetic circular dichroism (XMCD) and X-ray magnetic linear dichroism (XMLD) contrasts. Here we report the magnetic and ferroelectric structure of bismuth ferrite nanoparticles synthesised by the hydrothermal method and studied with the soft X-ray ptychography endstation at the Swiss Light Source [4]. Using this approach we are able to resolve the antiferromagnetic spin cycloid in the reconstructed real space images and uncover topological defects in the spin texture. Furthermore, circular dichroism in the resonant elastic X-ray scattering measured during a ptychographic scan yields information about the local spin chirality of the bismuth ferrite crystals.

## References

[1] S. R. Burns et al., *Adv. Mater.* **32** 2003711 (2020).
[2] I. Gross et al., *Nature* **549**, 252 (2017).
[3] D. Shapiro et al., *Nat. Photonics* **8** 765 (2014).
[4] M. Langer, et al., *Microsc. Microanal.* **24**, 54 (2018).

# Self-assembled epitaxial $BiFeO_3$ nanostructures toward multiferroic vertically aligned nanocomposites

*Wojciech Salamon[1]*, Łukasz Gondek[2], Jarosław Kanak[3], Marcin Sikora[1], Jakub Pawlak[1,2], Antoni Żywczak[1]*

[1] *Academic Centre for Materials and Nanotechnology, AGH University of Science and Technology*
[2] *Faculty of Physics and Applied Computer Science, AGH University of Science and Technology*
[3] *Institute of Electronics, AGH University of Science and Technology*
*salamon@agh.edu.pl

We present the spontaneous growth of self-assembled epitaxial nanostructures of $BiFeO_3$ (BFO) obtained by Pulsed Laser Deposition. The BFO phase architectures of squares, stripes and pyramids were grown on (001), (110) and (111) $SrTiO_3$ (STO) substrates, respectively. We determined the growth routes of BFO nanostructures and established a relationship between their morphology and structural properties. Such self-ordered crystalline nanostructures could be used as a tailored platform for the deposition of the ferromagnetic phase, thus providing an alternative method for the fabrication of BFO-based multiferroic vertically aligned nanocomposites [1] (VAN).

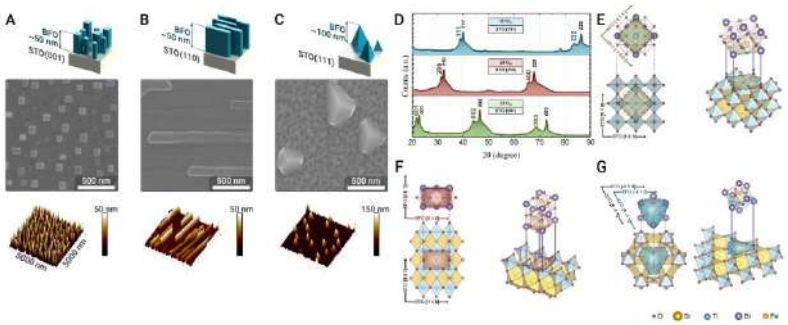



**Figure 1**: Morphology of self-assembled BFO phase architectures on A: (001), B: (110), C: (111) oriented STO substrates. D: X-ray Diffraction patterns for BFO nanostructures grown onto (001), (110) and (111)-oriented STO surfaces (subscripts denote: M−monoclinic, R-rhombohedral). STO-originated reflections are marked in black; Crystal matchings and suggested growth routes for: E) BFO/STO(001); F) BFO/STO(110) and G) BFO/STO(111)

Many VAN systems have been studied so far, and their magnetic, dielectric, and structural properties have been thoroughly investigated. However, there is a lack of alternative methods for VAN fabrication, which would enable the integration of any new VAN system that cannot be obtained using conventional method. Based on the research conducted to date, we present an alternative, two-step pathway to fabricate multiferroic VANs with a vertically aligned $BiFeO_3$ phase. As an example of our approach, we present the successful integration of BFO-nanostructured STO substrates with an epitaxial $CoFe_2O_4$ (CFO) phase. We have investigated the magnetic properties of the obtained nanocomposite. Thus, having a well-developed vertically aligned phase of BFO, it is possible to deposit another material as a matrix of the nanocomposite. The implementation of other new approaches in VANs engineering adds at least one degree of freedom to the design process of these types of nanomaterials. The technology of two-step VANs deposition still needs some optimization toward the growth control of nanopillars. Nevertheless, the success of this experiment gives hope for reengineering other existing multiferroic VAN systems using our alternative approach for ferroelectric:ferromagnetic VANs fabrication.

## Acknowledgments

The research was carried out thanks to the financial support of the National Science Centre, Poland (grant no. 2021/41/N/ST5/03009)

## References

1. Salamon, W. *et al.* Self-assembled epitaxial BiFeO3 nanostructures as a tailored platform for vertically aligned nanocomposites development. *Applied Surface Science* **607**, 154928 (2023).

# Assessing dielectric and ferroelectric properties in Lanthanum doped Bismuth Ferrite multiferroics

*Saba Aziz[1]*, Anna Grazia Monteduro[1], Silvia Rizzato[1], Angelo Leo[1], Chaitanya Lekshmi Indira[2], Giuseppe Maruccio[1]*

*[1] Omnics Research Group, Department of Mathematics and Physics - University of Salento, CNR-Institute of Nanotechnology, INFN Sezione di Lecce, Via per Monteroni, 73100, Lecce, Italy*

*[2] Materials Science/ Sensors & Nanoelectronics, CMR Institute of Technology, India*

*saba.aziz@unisalento.it

Multiferroic materials gained worldwide interest for their rich physics and potential use in multifunctional devices where data can be stored in ferroic domains and the response controlled by magnetic or electric fields [1]. However, accurately assessing their dielectric and ferroelectric properties requires significant attention due to the possible presence of spurious contributions and potential artifacts which can affect an appropriate evaluation [2, 3].

In this study, we report on methodological approaches and artifacts in assessing the dielectric and ferroelectric properties of lanthanum-doped bismuth ferrite (BFO) multiferroics. First, the synthesis and structural characterizazion of the $Bi_{(1-x)}La_{(x)}FeO_3$ (x = 0, 0.05, 0.10, 0.15, and 0.20) were carried out, showing the emergence of an impure $Bi_{2.1}La_{0.9}O_{4.54}$ and a $FeLaO_3$ phase along with a reduction of the Bi-rich ($Bi_{12.5}Fe_{0.5}O_{19.48}$) phase as lanthanum percentage increased. The presence of a $FeLaO_3$ phase also promotes a transition from a rhombohedral structure to an orthorhombic one as shown in Fig. 1(a) [4].

Then, to accurately evaluate the dielectric properties, we employed a statistical method and a fitting procedure to analyze as the capacitance scales linearly with the surface of the electrode in different samples [3]. This approach allows to separate the parasitic capacitive contributions from the intrinsic ones, as shown in Fig. 1(b-c) for pure BFO sample. By determining the dielectric properties of the pure and lanthanum-doped BFO pellets, our investigations reveal the highest permittivity values for the $Bi_{0.95}La_{0.05}FeO_3$ sample, while conductivity remained stable with respect to undoped BFO. However, in $Bi_{0.85}La_{0.15}FeO_3$ and $Bi_{0.8}La_{0.2}FeO_3$, we observed a reduction in conductivity due to the presence of the $LaFeO_3$ phase shown in Fig. 1(a).

For assessing the realistic ferroelectric responses, the PUND technique was employed since it allows to separate ferroelectric contributions from spurious ones due to leaky currents. A comparison of the P-E hysteresis loops evidences higher polarization for the $Bi_{0.95}La_{0.05}FeO_3$ samples, compared to un-doped and other doped BFO samples (Fig. 1(d-e)) because of the decrease leakage current resulting from La-doping. The findings of this study could have significant implications for the development of multifunctional materials for technological applications. By shedding light on the lanthanum doping effect on BFO, it could provide insight into the underlying mechanisms that govern the multifunctional properties of BFO and could pave the way for the development of new devices exploiting enhanced multifunctional properties, such as sensors, actuators, and energy harvesting devices.

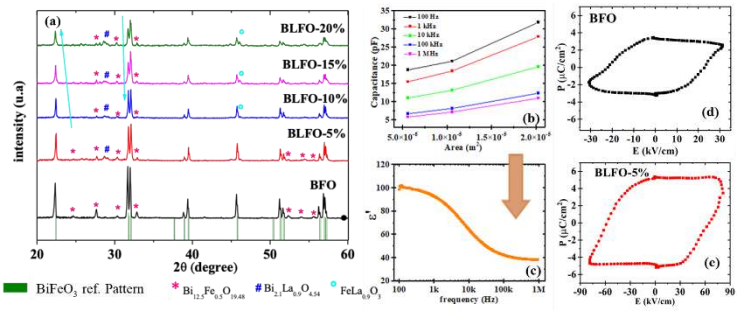



Figure 1**:(a)** XRD spectra of the analyzed BFO and BLFO samples, **(b)** capacitance as a function of the electrode area at certain frequencies, **(c)** the trend of the ε' of the pure BFO as a function of the frequency extracted by the procedure of decoupling the stary element from that specific to the sample, **(d-e)** P-E hysteresis of BFO and BLFO-5% samples

## References

[1] J. T. Heron *et al.*, Applied Physics Reviews **1** (2014).
[2] J. F. Scott, Journal of Physics: Condensed Matter **20**, 021001 (2008).
[3] A. G. Monteduro *et al.*, Journal of Materials Chemistry C **4**, 1080 (2016).
[4] N. Wang *et al.*, Nano-Micro Letters **12**, 81 (2020).

# Ferroelectricity in Ferrimagnetic Barium Hexaferrite Nanoplatelets

*Darja Lisjak[1]*, Peter Medle Rupnik[2], Alenka Mertelj[2], HanaUršič[3]*

[1] *Jožef Stefan Institute, Deprtment for Materials Synthesis, Jamova 39, Ljubljana, Slovenia*
[2] *Jožef Stefan Institute, Deprtment of Complex Matter, Jamova 39, Ljubljana, Slovenia*
[3] *Jožef Stefan Institute, Deprtment of Electronic Ceramics, Jamova 39, Ljubljana, Slovenia*
*darja.lisjak@ijs.si

Hexaferrites are a large family of ferrimagnetic oxides in the phase diagram $AO–Fe_2O_3–MO$, where A is a large cation (e.g., Ba or Sr) and M is a small cation (e.g., transition metal). The simplest structure is M-type, and the most studied M-type compound is barium hexaferrite ($BaFe_{12}O_{19}$). Barium hexaferrite is traditionally used for permanent magnet and microwave applications. A combination of the uniaxial magneto-crystalline anisotropy with the plate-like shape enables the coupling of the magnetic with other properties, such as optical, electric, and rheological [1]. Such coupling is enabled by the magnetic alignment of the barium hexaferrite nanoplatelets that are homogeneously dispersed in a medium. The intrinsic magneto-electric (ME) coupling of the magnetic and electric dipoles was observed in some hexaferrites of the Y- and Z-type with non-collinear magnetic spin structure (e.g., conical and helical) [2]. Some of these compounds also showed the multiferroic coupling between the ferrimagnetic and ferroelectric orders.

In this contribution, we present both ferroic orders in the $Sc^{3+}$-substituted M-type barium hexaferrite nanoplatelets at room temperature. The nanoplatelets were synthesized hydrothermally and dispersed in water. The dried nanoplatelets show typical hard-magnetic hysteresis with room-temperature saturation magnetization of around 35 $Am^2/kg$ (Figure 1, left-hand side). The nanoplatelets were deposited on the indium-doped tin-oxide (ITO) substrate for the piezo-response force microscopy (PFM) measurements. The results suggest the ferroelectric order in the nanoplatelets (Figure 1, right-hand side). Similar observations but well below the room temperature were reported for the $Sc^{3+}$-substituted $BaFe_{12}O_{19}$ bulk single crystals [3]. We will elucidate the effect of nano-size on the possible multiferroicity of M-type barium hexaferrites.

## Acknowledgements

The authors acknowledge funding from the Slovenian Research Agency, the research core programs P2-0089, P1-0192 and P2-0105.

## References

[1] D. Lisjak, A. Mertelj, *Prog. Mater. Sci.* **95** (2018) 286.
[2] R. Pullar, *Prog. Mater. Sci.* **57** (2011) 1191.
[3] Y. Tokunaga, et al., *Phys. Rev. Lett.* **105** (2010) 257201.

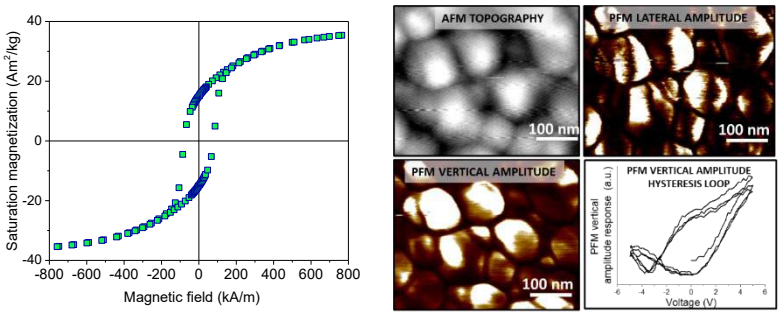



Figure 1: Magnetic hysteresis (left) and PFM experiment (right) of the $Sc^{3+}$-substituted barium hexaferrite nanoplatelets. The bright regions inside the nanoplatelets visible in the PFM amplitude images correspond to the enhanced piezoelectric response compared to the dark regions.

# Exchange Modes in Doped $BaFe_{12}O_{19}$ Spherical Nanoshells with Strong Uniaxial Magnetic Anisotropy

*María José Vázquez Bernárdez[1]*, Daniel Stoeffler[1], Christophe Lefevre[1], Nicolas Vukadinovic[2]*

[1]*Université de Strasbourg, CNRS, Institut de Physique et Chimie des Matériaux de Strasbourg, UMR 7504, 23 Rue du Loess Bâtiment 69, 67200 Strasbourg France*

[2]*Dassault Aviation, 78 quai Marcel Dassault, 92552 St-Cloud, France*

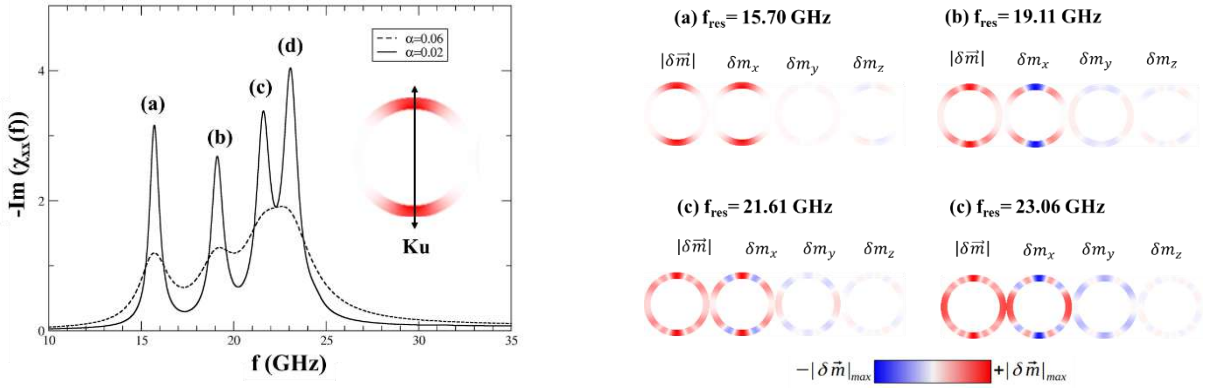
*contact & presenting author : mariajose.vazquez@ipcms.unistra.fr, +33 7 66 65 72 56

Composites made up of chemically substituted hexagonal ferrites have been proven to be promising electromagnetic wave absorbents with frequencies operating at Ku, K and Ka bands (from 12 to 40 GHz). In order to serve as performant materials, these are required to possess a low mass and a strong absorption in a broad frequency band. Whilst the dynamic susceptibility spectra of this family of compounds may be correlated to chemical composition, particle size and morphology [1], the effect of geometrical structuration has not yet been addressed.

Recently, curved magnetic 3D systems have been the focus of intense research due to the diversity of dynamic behaviours associated to surface and exchange magnetic contributions. Size-dependent resonance modes have been identified in Fe spherical nanoshells requiring a large applied magnetic field to reach saturation [2]. It has been demonstrated that, for low cubic-anisotropy ferromagnetic systems, there is a size range where exchange dominates over dipolar contributions. As a result, new resonance modes arise with amplitudes comparable to the main ferromagnetic resonance (FMR) mode. Nevertheless, no previous attention has been paid to the dynamic susceptibility response of such systems in the absence of an applied magnetic field.

This work aims to investigate zero-field dynamic susceptibility spectra of chemically substituted $BaFe_{12}O_{19}$ hollow spheres supporting single domain configurations at equilibrium by means of micromagnetic simulations (mumax3 software). Exchange resonance eigenmodes, corroborated by a spatially structured dynamic magnetic configuration at each eigenfrequency, have been obtained. Their eigenfrequencies are a function of the external diameter of the hollow sphere [2], their number is proportional to the thickness of the spherical shells and it increases when the latter decreases. Modulating the shell thickness of $BaFe_{12}O_{19}$ hollow spheres could be therefore proposed as a useful technique to generate new absorption peaks within the dynamic susceptibility spectra of K-band composites and consequently broaden the frequency absorption band.

The existence of such exchange modes in $BaFe_{12}O_{19}$ nanoshells has not been proven experimentally yet. In order to account for the effect of geometrical imperfections that could arise during fabrication, the preservation of such modes in nanoshells that deviate from ideal spherical geometry will be discussed.




(left) Zero-field susceptibility spectra of a doped $BaFe_{12}O_{19}$ hollow sphere of external diameter $D_e$=500 nm and internal diameter of $D_i$= 400 nm. (right) Pole-to-pole cross-section views of eigenmodes maps for the 4 identified eigenfrequencies.

## Acknowledgements

The authors acknowledge the joint research laboratory MOLIERE for supporting this work.

## References

1 D. Djuhana, D. C. Oktri, D. H. Kim, *Advanced Materials Research* (Vol. 896, pp. 414–417) (2014).

2 C. McKeever, F. Y. Ogrin, M. M. Aziz, *Phys. Rev. B*, **100(5)** (2019) 054425.

# Unveiling the Mechanism for Enhanced Ferromagnetism in Ru-doped $Ca_3Mn_2O_7$ Hybrid Improper Multiferroic

*G. Subías[1]*, V. Cuartero[1,2], J. Herrero-Martín[3], J. A. Rodriguez-Velamazán[4], S. Lafuerza[1], J. L. García-Muñoz[5], and J. Blasco[1]*

[1] *Instituto de Nanociencia y Materiales de Aragón, Dpto. de Física de la Materia Condensada, CSIC-Universidad de Zaragoza, C/ Pedro Cerbuna 12, Zaragoza, Spain*

[2] *Dpto. de Ciencia y Tecnología de Materiales y Fluidos, Universidad de Zaragoza, C/ María de Luna 3, Zaragoza, Spain*

[3] *ALBA Synchrotron Light Source, Carrer de la Llum 2-26, Cerdanyola del Vallès, Barcelona, Spain*

[4] *Institut Laue-Langevin, Grenoble Cedex 38042, France*

[5] *Institut de Ciència de Materials de Barcelona, ICMAB-CSIC, Campus UAB, Bellaterra, Spain*

*gloria@unizar.es

The search of multiferroic materials with magnetoelectric coupling near room temperature (RT) attracts enourmous research attention due to the promising applications for ultra-low-power electronics. Hybrid improper ferroelectrics (HIFs) are currently of great interest for the realization of RT multiferroelectricity with enough magnetoelectric coupling. The prototypical HIF n=2 Ruddlesden-Popper $Ca_3Mn_2O_7$ undergoes a structural phase transition at $T_S$=310 K from a high-temperature tetragonal to a low-temperature orthorhombic polar phase. However, only antiferromagnetic (G-type) order develops in this system at $T_N$=115 K [1]. In order to promote ferromagnetism, we have explored Ru-doped $Ca_3Mn_{2-x}Ru_xO_7$ compounds ($0<x<1$).

We have obtained a solid solution between Mn and Ru atoms in the these samples and they adopt the orthorhombic $Bb2_1m$ polar structure of the parent compound at RT [2]. Thus, the HIF mechanism is fully operational along the $Ca_3Mn_{2-x}Ru_xO_7$ series. Neutron powder diffraction only evidences distinct long-range antiferromagnetic (AFM) orders depending on the Ru content. In contrast with the macroscopic magnetic study, which also reveals the occurrence of spontaneous magnetization in the hysteresis loops. Moreover, at 5 K the observed remanence increases upon Ru doping ($0.3\leq x\leq 0.7$) [2].

We have investigated the origin of the enhanced ferromagnetic contributions upon Ru doping on the basis of the Mn and Ru local electronic and magnetic structures using X-ray absorption spectroscopy (XAS) and X-ray circular magnetic dichroism (XMCD) techniques. We confirm the presence of $Ru^{4+}$ and $Mn^{4+}$ valence states along the whole series, discarding the charge transfer from Ru to Mn as mechanism to enhance ferromagnetism in these samples. Furthermore, XMCD results indicate a small net Ru 4d magnetic moment antiferromagnetically coupled to the net Mn 3d magnetic moment. Therefore, the ferromagnetic enhancement observed in $Ca_3Mn_{2-x}Ru_xO_7$ with $x\geq 0.3$ is related to an intrinsic canting of the antiferromagnetically ordered $Mn^{4+}$ spins, which is favoured by Ru doping.

**Acknowledgements**

We acknowledge the Spanish Ministerio de Ciencia e Innovación for financial support (Projects PID2021-124734OB-C21 and PID2021-124734OB-C22) cofounded by ERDF from EU. We thank the ILL (experiment 5-31-2788); the ALBA synchrotron (Experiment 2020094610) and the ESRF synchrotron (Experiment HC-4500) for granting beam time.

**References**

[1] M. V. Lobanov *et al.*, *J. Phys.: Condens. Matter* **16** (2004) 5339.
[2] J. Blasco, *et al.*, *Phys. Rev. B* **106** (2022) 134403.

# Anisotropic Shaped Non-Magnetically Interacting Nano Crystallites as Precursors for Aligned Strontium Hexaferrite.

*Mogens Christensen[1]*, Harikrishnan Vijayan,[1,2], Jack Thomas-Hunt,[1]*

*[1] Department of Chemistry & iNANO, Aarhus University, Aarhus C-8000, Denmark*

*[2]Group of Permanent Magnets and Applications, IMDEA Nanoscience, 28049 Madrid, Spain*

*corresponding author email address: mch@chem.au.dk

Anisotropically shaped nanocrystallites of goethite (α-FeOOH), hematite (α-$Fe_2O_3$) and six-line ferrihydrite ($Fe_{0.86}OOH$) have been used in a simple dry-process to fabricate highly aligned M-type strontium hexaferrite ($SrFe_{12}O_{19}$). The simple dry mixing process of anisotropically shaped nanocrystallites with strontium carbonate ($SrCO_3$) and subsequent cold compaction and calcination alleviate the need for external magnetic fields in order to align the crystallites. The compacted precursor is calcined at elevated temperatures (>1000 °C) to form $SrFe_{12}O_{19}$ as a dense and mechanical stable pellet. An illustration of the process is shown in Figure 1.

Structural investigations of the prepared pellets reveal large crystallographic preferred orientation and high phase purity (>99 wt.%). In the case of α-FeOOH a $M_r/M_s$ = 0.83 could be obtained, while α-$Fe_2O_3$ resulted in $M_r/M_s$ = 0.70 and $Fe_{0.86}OOH$ revealed a $M_r/M_s$ of 0.66.[1,2] The coercivities for the corresponding samples were $H_c$(α-FeOOH) = 157 kA/m, $H_c$(α-$Fe_2O_3$) = 207 kA/m and $H_c$($Fe_{0.86}OOH$) = 150 kA/m, while $(BH)_{max}$ reached 22, 21 and 12 kJ/m$^3$, respectively. The texture was investigated using 2D synchrotron powder diffraction images, which allows extraction of the texture index, which can be compared to the $M_r/M_s$ confirming the strong alignment of the crystallites.

Similar samples have also been prepared using spark plasma sintering (SPS), here a low sintering temperature of only 750 °C was sufficient to transform $Fe_{0.86}OOH$ into highly aligned strontium hexaferrites ($SrFe_{12}O_{19}$) with the pressing direction and the magnetic easy axis coinciding. The best performing magnet had an $M_r/M_s$ = 0.93, $H_c$ = 247 kA/m and produced a $(BH)_{max}$ of 33 kJ/m$^3$.[3] The results reveal that the cold compaction and subsequent calcination method can still be improved to produce better performing magnets, however the method has the potential to make a new easy route for industrial preparation of sintered $SrFe_{12}O_{19}$ magnets.

## References

[1] J. Thomas-Hunt, A. Povlsen, H. Vijayan, C. G. Knudsen, F. H. Gjørup, M. Christensen, *Dalt. Trans.* 2022, 51, 3884–3893.

[2] H. Vijayan, A. Povlsen, J. Thomas-Hunt, M. I. Mørch, M. Christensen, *J. Alloys Compd.* 2022, 915, 165333.

[3] H. Vijayan, C. G. Knudsen, M. Mørch, M. Christensen, *Mater. Chem. Front.* 2021, 5, 3699–3709.

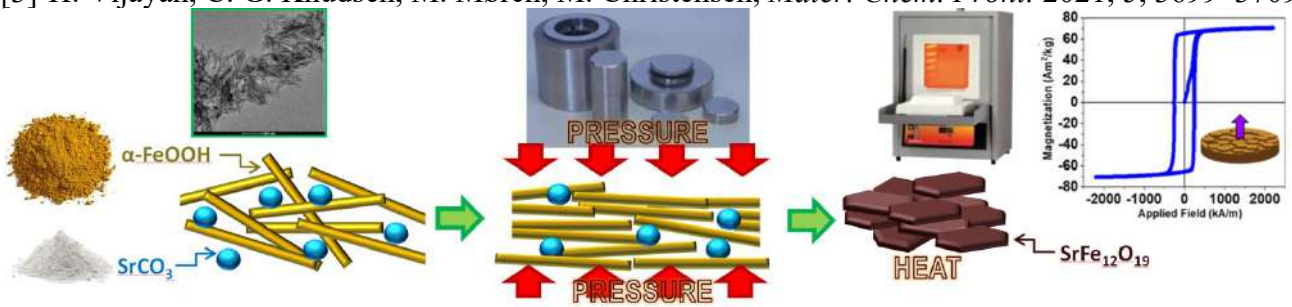



Figure 1: Conversion of goethite into aligned strontium hexaferrite through a simple compaction and subsequent calcination process.

# Towards Maximized Energy Product of Bicomponent Ferrite Permanent Magnets: Zn Substitution at Ni Ferrite

*S. Gallego[1]*, R. Rico[1], C. Tejera[1]*

[1] *Instituto de Ciencia de Materiales de Madrid, CSIC. c/ Sor Juana Inés de la Cruz, 3. 28049 Madrid. Spain.*

*sgallego@icmm.csic.es

Rare-earth free permanent magnets based on ferrites represent an abundant, cheap and non-critical alternative in the magnetic materials market. They are usually prepared in the form of nanostructured bicomponent systems that combine hard and soft materials. The hard component maximizes the coercivity, while the soft one enhances the magnetization. To be competitive as compared to other rare-earth free alternatives, improvement of the nanostructured ferrite magnets relies on the increase of the magnetization of the soft component. One of the most common ferrite soft magnets is Ni ferrite ($NiFe_2O_4$, NFO). Experimental efforts to enlarge its magnetization include the substitution of Ni by Zn, that is expected to occupy tetrahedral coordination sites displacing Fe, thus reducing the minority spin band contribution to the net magnetization. However, controversial results have been obtained following this procedure.

Our work explores in detail the properties of Zn substituted NFO based on ab initio calculations within density functional theory at the GGA+U level, providing a thorough characterization of the structural, electronic and magnetic properties for a wide range of Zn concentrations. We have paid special attention to the choice of U for the Zn d electrons, proving that it plays a crucial role in the energy balance that determines the ratio of Zn atoms at tetrahedral and octahedral coordination sites. This has relevant consequences on the magnetism of the system, particularly on the net magnetization. Further, we demonstrate that a similar situation emerges at the pure Zn cubic spinel ferrite, ZFO, where the value of U affects the relative stability of the inverse and direct forms.

The outcome of this study reveals a strong tendency of Zn to occupy tetrahedral coordination sites, more relevant the higher the Zn concentration in the spinel lattice. This is an important result to understand the abundant experimental evidence of ZFO samples adopting the inverse spinel structure, and explains the origin of measurements of anomalous ferromagnetism. Further, it explains the shortcomings of Zn substitution as a route to increase the magnetization over that of pure NFO. We will end discussing some open possibilities worth to explore in order to maximize the energy product of NFO bicomponent systems based on Zn substitution [1].

**Acknowledgements**

This work has been performed under grant TED2021-130957B-C53, financed by MCIN/AEI/10.13039/501100011033 and the EU NextGenerationEU/PRTR.

**References** C. Tejera, R. Rico, S. Gallego, *J. Mat. Sci.* (2023) , in press.

# Synthesis of Magnetic Nanoparticles by Recycling Residues from Industrial Steel Manufacturing

*Cristina M. Montero*[*1], Mónica Dhanjani[1], Alejandro Martín[1], Gorka Salas[1], Alberto Bollero[1]*
[1] *IMDEA Nanoscience, 28049 Madrid, Spain*
*cristinamaria.montero@imdea.org

Providing a second life to residues resulting from the fabrication of products has become an urgent matter to diminish the environmental impact and optimize an efficient use of the resources [1,2]. Steel industry is one of the sectors with a faster market growth and Europe is playing a major role on it. Development of a circular economy will guarantee a sustainable development of this key sector while opening new market possibilities. Magnetic nanoparticles (NPs) find many technological applications (energy, transport, medicine…) attracting a large interest, also in view of developing advanced fabrication techniques for tuning their magnetic properties.

This study shows results obtained from applying a novel process enabling the transformation of residues generated by the steel industry into iron oxide NPs. The precursor was provided by CELSA Group in the shape of powder with micrometer size particles in the framework of a collaboration project. In view of the potential industrial scalability of the process, a physical method has been used based on application of milling times as short as 5-25 min (IMDEA's self-developed "flash-milling" method [2,3]) followed by an optimized heat treatment. This route has enabled to move from a residue with a saturation magnetization of 23 emu/g to iron oxide NPs (25 nm mean particle size) with a saturation magnetization close to 130 emu/g accompanied by a coercivity of 200 Oe after milling for 15 min and annealing (Fig. 1). Further optimization of the process allows reducing the milling time to a total duration of 5 min, achieving an outstanding saturation magnetization close to 200 emu/g (and a coercivity below 100 Oe). These results open the path to exciting new uses for this industrial residue in the form of iron oxide NPs.

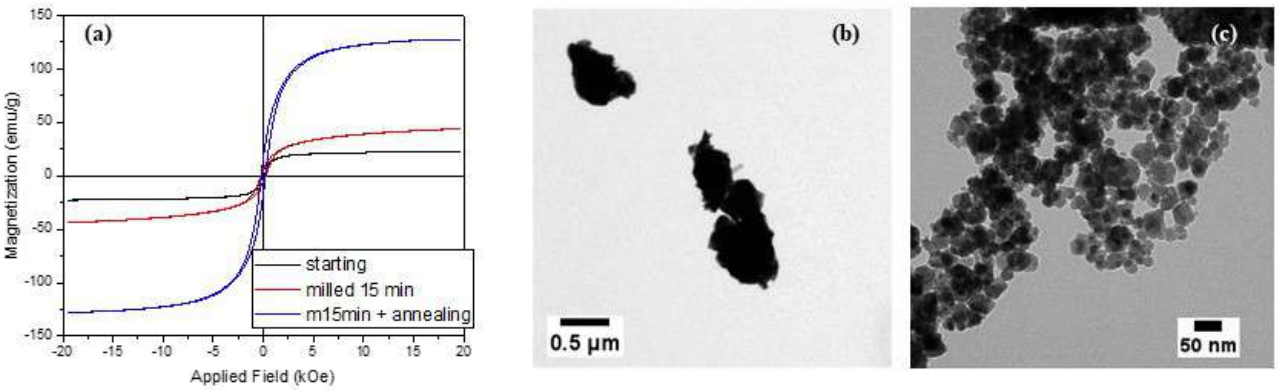



Figure 1: (a) Room temperature VSM hysteresis loops measured for the precursor powder and that after milling for 15 min before and after annealing. TEM images of (b) the starting powder and (c) the powder milled for 25 min.

## Acknowledgements

Authors acknowledge provision of the precursor material by CELSA Group and financial support through a CELSA-IMDEA Nanociencia collaboration project. IMDEA Nanociencia thanks support from the ''Severo Ochoa'' Programme for Centers of Excellence in R&D (MICINN, Grant CEX2020-001039-S) and Comunidad de Madrid through NANOMAGCOST project (Ref. S2018/NMT-4321).

## References

[1] K.W. Borth, C.W. Galdino, V.C. Teixeira, F.J. Anaissi. *Appl. Surf. Sci.,* **546** (2021) 149126.
[2] A. Bollero, J. Rial, M. Villanueva, K.M. Golasinski, A. Seoane, J. Almunia, and R. Altimira. *ACS Sustainable Chem. Eng.*, **5** (4) (2017) 3243.
[3] F.J. Pedrosa, J. Rial, K.M. Golasinski, M. Rodríguez-Osorio, G. Salas, D. Granados, J. Camarero, and A. Bollero. *RSC Adv.*, **6**, (2016) 87282.

# Scalable synthesis of hard ferromagnetic ε-$Fe_2O_3$ nanoparticles for 6G applications

*Naureen Khanam[1]*, Martí Gich[1]*

[1] *Institut de Ciència de Materials de Barcelona(ICMAB-CSIC), Bellaterra, Barcelona, 08193 Spain*

*nkhanam@icmab.es

Ferrites showing natural ferromagnetic resonances (NFMR) in millimeter wave frequencies, particularly in 95-220 GHz are interesting because of their potential applications in self-biased non-reciprocal devices for the next generations wireless communication (6G and beyond) [1, 2]. ε-$Fe_2O_3$ is an orthorhombic iron oxide phase with a sizeable magnetization ($M_S$ ~ 100 emu/cm$^3$) and a strong uniaxial magnetic anisotropy [3] reflected in an enormous coercivity ($H_C$ ~ 20 kOe) at room temperature [4] and NFMR at around 180 GHz [5]. The resonance frequency can be controlled by substitution of $Fe^{3+}$ with other trivalent metals which is interesting for the above mentioned application [5]. To be successfully used in self-biased non-reciprocal devices such as circulators, ε-$Fe_2O_3$ nanoparticles have to retain their remanent magnetization state. We have studied the relaxation of ε-$Fe_2O_3$ nanoparticles as a function of their size, finding that relaxation effects are largely supressed for particles above 25 nm. The current state of art allows preparing larger particle sizes but using techniques that are not compatible with a large scale production maintaining a sustainable approach [6]. In this contribution, we will discuss our newly developed method to obtain in one batch large amounts of ε-$Fe_2O_3$ particles which are not affected by relaxation processes, overcoming an important limitation for its application in 6G devices.

**Acknowledgements**

Funding is acknowledged from Severo Ochoa Programme for Centres of Excellence in R&D (FUNFUTURE CEX2019-000917-S) and European Research Council FeMiT project (ERC-CoG 819623). Judit Oró and Bernat Bozzo from the ICMAB scientific services are acknowledged for the analysis of materials by transmission electron microscopy and magnetic measurements respectively.

**References**

[1] T. S. Rappaport, Y. Xing, O. Kanhere, S. Ju, A. Madanayake, S. Mandal, A. Alkhateeb, G. C. Trichopoulos, *IEEE Acc.* 7(2019) 78729.

[2] I.F. Akyildiz, A. Kak, S. Nie, *IEEE Acc.* 8*(*2020) 133995.

[3] M. Gich, A. Roig, C. Frontera, E. Molins, J. Sort, M. Popovici, G. Chouteau, D. M. Marero, J. Nogués, *J of Appl. Phys.* 98 1(2005) 044307.

[4] J. Jin, S. I. Ohkoshi, K. Hashimoto, *Adv. Mater.* 16*(*2004) 48.

[5] A. Namai, S. Sakurai, M. Nakajima, T. Suemoto, K. Matsumoto, M. Goto, S. Sasaki, S. I. Ohkoshi, *J of the Amer. Chem. Soc.* 131 3(2009) 1170.

[6] S. I. Ohkoshi, S. Sakurai, J. Jin, K. Hashimoto, *J of Appl. Phys.* 97(2005) 10K312.

# Implementing tin-loaded magnetic nanoparticles in drinking water treatment practices

*K. Simeonidis*, K. Kalaitzidou, T. Asimakidou, N. Maniotis*

*Analytical Chemistry Laboratory, Department of Chemical Engineering, Aristotle; University of Thessaloniki, Thessaloniki, Greece*

*ksime@physics.auth.gr

The presence of hexavalent chromium Cr(VI) in drinking water resources is one of the major problems of environmental pollution that came to light during the last quarter-century. Long-term consumption of even small concentrations of Cr(VI) at values of a few decades of μg/L may be considered a serious threat to health, since recent findings that correlate exposure to Cr(VI) in drinking water with chronic disease, cancer, and overall decreased life expectancy. World Health Organization has issued guidelines for drinking water quality and a growing number of international policymakers have begun implementing strategies to reduce the risk. Suggestively, the California State follows a procedure to apply a maximum contaminant level of 10 μg/L by 2024, whereas European Union members agreed to set a limit of 25 μg/L by January 2036.

Adopting materials with high reducing potential has been proven as an efficient strategy to capture Cr(VI) from water oriented for drinking purposes by turning it into insoluble Cr(III) hydroxides. The objective of this work was to develop a novel class of Cr(VI) adsorbents, engineered in the nanoscale but realized in kilogram-scale production rates, based on nanocomposites consisting of a highly reducing phase, a tin oxy-hydroxide, and a magnetically-responding phase, iron oxide nanoparticles, which facilitates the post-treatment separation option. Such nanocomposite was prepared by the translation of nanoscale synthesis methods for nanoparticles into sequential continuous-flow reaction processes. Particularly, $Fe_3O_4$ nanoparticles were prepared by the oxidative precipitation of $FeSO_4$ in one line whereas the Sn chloro-oxyhydroxide (abhurite) was synthesized in parallel by the precipitation of $SnCl_2$ under acidic conditions. After obtaining the two products in the form of concentrated suspensions, they were homogenized in a high-energy homogenizer to obtain the nanocomposite. By this approach, $Fe_3O_4$ nanoparticles were successfully entrapped into the layered structure of the abhurite formations and that made a well homogenized nanocomposite with the ability of magnetic response to the whole volume.

Uptake experiments for low residual concentrations showed that the capacity of the nanocomposite for Cr(VI) when residual concentration of 25 μg/L was achieved, was 7 mg/g. The efficiency of the nanomaterials in a continuous flow pilot unit was evaluated by operating a contact reactor with mechanical stirring to mix nanoparticles' dispersions with the polluted water. To ensure complete recovery of the nanocomposite at the end of the process, a rotary magnetic separation setup based on the Halbach array of magnets was tested.

## Acknowledgements

The research project was supported by the Hellenic Foundation for Research and Innovation (H.F.R.I.) under the "2nd Call for H.F.R.I. Research Projects to support Post-Doctoral Researchers" (Project Number: 00046 MagnoSorb).

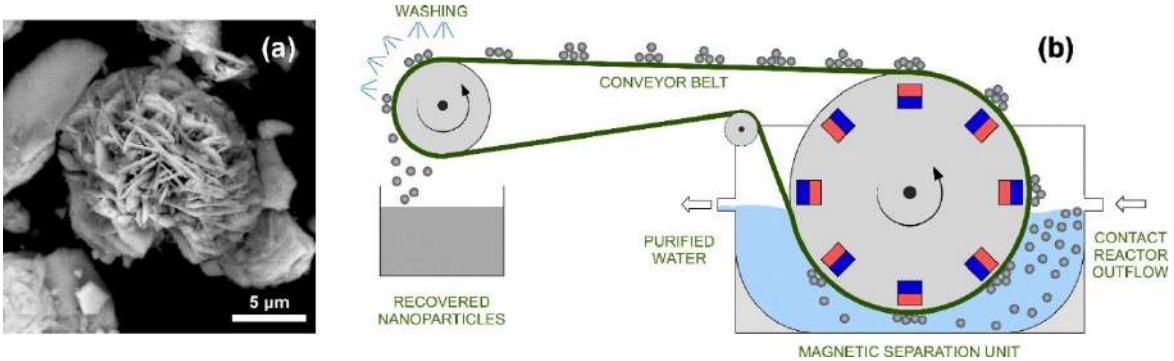



Figure: Scanning electron microscopy image of the $Fe_3O_4$/abhurite (a). Post-treatment rotary magnetic separator for the recovery of the nanocomposite.

# SYMPOSIUM 07.
## MAGNETIC NANOPARTICLES AND MULTIFUNCTIONAL MAGNETIC MATERIALS.
## S7. POSTERS

# Lithiation and Delithiation Induced Magnetism Variations in Iron Disilicide Nanoparticle Embedded Si/$SiO_x$

*Stefan Topolovec[1]*, Matthias Prasch[1], Roland Würschum[1]*

[1] *Institute of Materials Physics – Graz University of Technology, Petersgasse 16, 8010 Graz, Austria*

*stefan.topolovec@tugraz.at

For iron disilicide a metastable metallic (α-$FeSi_2$) and a stable semiconducting phase (β-$FeSi_2$) exist, which both are paramagnetic in the bulk state [1]. However, it was shown that ferromagnetism or superparamagnetism can occur in nanoparticles of these materials, which was attributed to chemical disorder and/or lattice distortions at the surface of the nanostructures [2-6]. In a recent theoretical work Zhandun et al. [7] have shown that beside lattice distortions, intercalation of light elements like Li can induce ferromagnetism in α-$FeSi_2$. Furthermore, Wu et al. [6] demonstrated that the surface ferromagnetism of β-$FeSi_2$ nanocubes embedded in a ferroelectric film can be modulated through the accumulation and depletion of charge carriers induced by the ferroelectric polarization reversal. Both of this makes iron disilicide nanostructures attractive for investigations with respect to electrochemically induced magnetism variations, for instance by electrochemical lithiation.

To obtain magnetic iron disilicide nanoparticles in a matrix which is sensitive to electrochemical charging, we followed a route of He et al. [8] to prepare Fe-Si nanoparticle embedded Si/$SiO_x$ Li-ion battery anodes by ball milling and heat treatment using industry-grade ferrosilicon as raw material. XRD measurements have shown that mainly α-$FeSi_2$ and only a small fraction of β-$FeSi_2$ are present in the Si/$SiO_x$ matrix before and after ball milling. The observed increase of the saturation magnetization with ball milling is consistent with the assumption that lattice distortions or chemical disorder are the main source of magnetism. Consecutive temperature treatment increases the fraction of the β-$FeSi_2$ phase, which becomes the dominant phase after temperature treatment at 1073 K. After high temperature annealing at 1273 K, the β phase was completely transformed into α-$FeSi_2$. This transition between the β and α phase was accompanied by a substantial increase of the saturation magnetization.

To study the influence of lithiation/delithiation of the $FeSi_2$ – Si/$SiO_x$ nanocomposites on the magnetic properties, we monitored the magnetic moment during charging/discharging using a special electrochemical cell enabling operando measurements in a SQUID magnetometer [9]. Independent of whether the α-$FeSi_2$ (after treatment at 1273 K) or β-$FeSi_2$ phase (after treatment at 1073 K) was the dominant one, an increase of the magnetic moment of up to 50% was observed during the first lithiation and the subsequent delithiation of the composite anode. In the following cycles a decrease of the magnetic moment by several percent occurred in the first stage of lithiation, followed by a more pronounced increase in the subsequent delithiation process. The trend of a general increase of the magnetic moment and the decreasing reversible fraction of the magnetism variation over multiple (de-)lithiation cycles indicate that the introduction of lattice defects/disorder by lithiation/delithiation are the main cause for the magnetism variation.

## Acknowledgements

The authors thank the Optics of Nano and Quantum Materials group of the Institute of Physics (University of Graz) for providing access to the SQUID magnetometer, as well as, K. Rumpf and P. Granitzer (Institute of Physics, University of Graz) for their help at the VSM measurements in the framework of the inter-university cooperation of TU Graz and Uni Graz on natural science (NAWI Graz). The authors thank I. Hanzu and A. Jodlbauer (Institute of Chemistry and Technology of Materials, TU Graz) for the anode slurry preparation.

## References

[1] X. Sun, X. Liu, X. Liu, Y. Meng, C. Song, Y. Yang, Y.-W. Li, X.-D. Wen, *J. Phys. Chem. C* **123** (2019) 11939.
[2] Y. Chen, P. Lee, C. Tsai, S. Neeleshwar, C. Wang, J. Ho, H. Hamdeh, *Appl. Phys. Lett.* **91** (2007) 251907.
[3] H. Hamdeh, M. Eltabey, J. Ho, P. Lee, K. Chen, Y. Chen, *J. Magn. Magn. Mater.* **322** (2010) 2227.
[4] M. Dascalu, F. Cesura, G. Levi, O. Diéguez, A. Kohn, I. Goldfarb, *Appl. Surf. Sci.* **476** (2019) 189.
[5] Z. He, S. Xiong, S. Wu, X. Zhu, M. Meng, X. Wu, *J. Am. Chem. Soc.* **137** (2015) 11419.
[6] S. Wu, A. Wang, Y. Shan, L. Liu, *Solid State Commun.* **35** (2022) 114954.
[7] V. Zhandun, N. Zamkova, P. Korzhavyi, I. Sandalov, *Phys. Chem. Chem. Phys.* **21** (2019) 13835.
[8] W. He, Y. Liang, H. Tian, S. Zhang, Z. Meng, W.-Q. Han, *Energy Storage Mater.* **8** (2017) 119.
[9] S. Topolovec. H. Kren, G. Klinser, S. Koller, H. Krenn, R. Würschum, *J. Solid State Electrochem.* **20** (2016) 1491.

# On the Magnetization Reduction in Iron Oxide Nanoparticles

*A. Feoktystov[1*], T. Köhler[1,2,3], O. Petracic[2], E. Kentzinger[2], T. Bhatnagar-Schöffmann[2,3,4], M. Feygenson[5,6], N. Nandakumaran[2,3], J. Landers[7], H. Wende[7], A. Cervellino[8], U. Rücker[2], A. Kovács[4], R.E. Dunin-Borkowski[4], Th. Brückel[2,3]*

[1] *Forschungszentrum Jülich GmbH, JCNS at MLZ, Garching, Germany*
[2] *Forschungszentrum Jülich GmbH, JCNS-2 and PGI-4, JARA-FIT, Jülich, Germany*
[3] *Lehrstuhl für Experimentalphysik IV C, RWTH Aachen University, Aachen, Germany*
[4] *Forschungszentrum Jülich GmbH, ER-C and PGI, Jülich, Germany*
[5] *Forschungszentrum Jülich GmbH, JCNS-1 and ICS-1, Jülich, Germany*
[6] *European Spallation Source ERIC, Lund, Sweden*
[7] *Faculty of Physics and Center for Nanointegration Duisburg-Essen (CENIDE), University of Duisburg-Essen, Duisburg, Germany*
[8] *Paul Scherrer Institut, Swiss Light Source, Villigen, Switzerland*
*a.feoktystov@fz-juelich.de

Iron oxide nanoparticles are presently considered as promising objects for various medical applications including targeted drug delivery and magnetic hyperthermia. The nanoparticle solution in water has to posses large enough saturation magnetization to react on external magnetic field. However, there remains several unsolved questions regarding the effect of size onto nanoparticle overall magnetic behavior. One aspect is the reduction of magnetization as compared to bulk samples. A detailed understanding of the underlying mechanisms of this reduction will improve the particle performance in the applications.

There are several proposed models for the spatial distribution of the magnetization, which include the presence of a magnetic core-shell structure, spin disorder around defects and a reduced magnetization in the core due to reversed moments and frustration. In this work we combine neutron and synchrotron X-ray scattering techniques with magnetometry, transmission electron microscopy (TEM), elemental analysis and Mössbauer spectroscopy to study nanoparticles of various sizes and to obtain as complete as possible picture of their properties [1]. We find that the nanoparticles possess a macroscopically reduced saturation magnetization, mostly due to the presence of antiphase boundaries (APBs) as observed with high-resolution TEM (HRTEM) and X-ray scattering [2] and to a lesser extent due to a small magnetically depleted surface layer and cation vacancies.

## References

[1] T. Köhler, A. Feoktystov, O. Petracic, E. Kentzinger, et al. *Nanoscale* **13** (2021) 6965.
[2] T. Köhler, A. Feoktystov, O. Petracic, N. Nandakumaran, et al. *J Appl Crystallogr* **54** (2021) 1719.

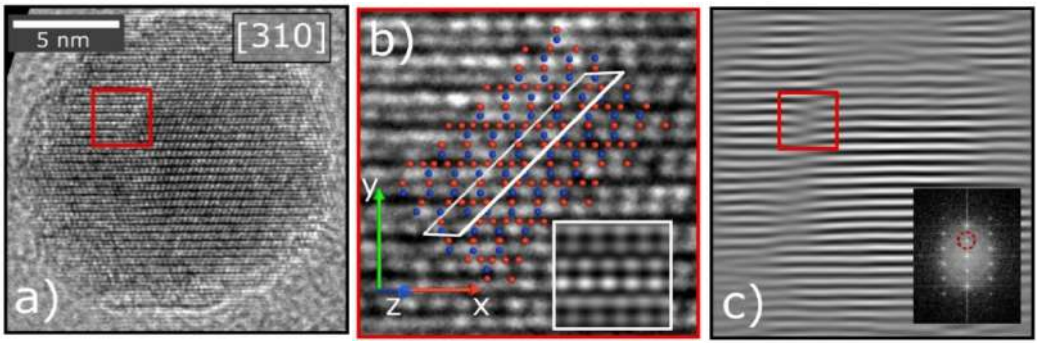



Figure: (a) HRTEM micrograph of an isolated nanoparticle viewed along [310]. A region with an APB is marked with the red square. (b) Marked region of (a) with a schematic of the crystal structure, the atom positions were verified by multislice TEM image simulations (inset). The lattice plane along which the translation occurs is indicated with the white rectangle. Red and blue dots represent iron atoms in octahedral and tetrahedral coordination, respectively. (c) The micrograph depicted in (a) after masking the 220 Bragg reflection (red circle in the inset) in the fast Fourier transform (FFT) and calculating the inverse FFT (IFFT). Now the lattice translation becomes clearly visible.

# Magnetically-activated 3D printable PLA/PCL/$Fe_3O_4$ composites for magnetic induction heating generation

*Itziar Galarreta-Rodriguez[1-2], Alberto Lopez-Ortega[1-2], Eneko Garayo[1-2*], Juan Jesús Beato-López[1-2], Paulo La Roca[1-2], Vicente Sanchez-Alarcos[1-2], Vicente Recarte[1-2], Cristina Gómez-Polo[1-2] and Jose Ignacio Pérez-Landazábal[1-2]*

[1]Departamento de Ciencias, Universidad Pública de Navarra, Campus de Arrosadía, 31006, Pamplona, Spain

[2]Institute for Advanced Materials and Mathematics (INAMAT2), Universidad Pública de Navarra, Campus de Arrosadía, 31006, Pamplona, Spain

*eneko.garayo@unavarra.es

Additive manufacturing technology has attracted the attention of industrial and technological sectors due to the versatility of the design and the easy manufacture of structural and functional elements based on composite materials [1]. The embedding of magnetic NPs in the polymeric matrix enables the development of an easy manufacturing process of low-cost magnetically-active novel polymeric composites. In this work, we report a series of magnetic composites prepared by solution casting method combining 5 to 60 wt.% of 90-180 nm commercial Fe3O4 nanoparticles (NPs), with a semi-crystalline, biocompatible and biodegradable polymeric blend made of Polylactic Acid (PLA) and Polycaprolactone (PCL). The composites were extruded, obtaining 1.5 mm diameter continuous and flexible 3D printable filaments for Fused Deposition Modelling. The chemical, magnetic and calorimetric properties of the obtained filaments were investigated by Differential Scanning Calorimetry (DSC), Thermogravimetric Analysis (TGA), magnetometry (SQUID), Scanning Electron Microscopy (SEM). Furthermore, taking advantage of the magnetic character of the filaments, their capability to generate heat under the application of low-frequency AC magnetic fields (magnetic induction heating) was analysed. The obtained results expose the versatility of these easy manufacturing and low-cost filaments, where selecting a desired composition, the heating capacity can be properly adjusted for those applications where magnetic induction plays a key role (i.e. magnetic hyperthermia, drug release, heterogeneous catalysis, water electrolysis, gas capture or materials synthesis).

## Acknowledgements

This work has been carried out with the financial support of the Navarra Government (project number PC017-018 AMELEC). The Spanish Government is acknowledged for the HIPERNANO research network (RED2018-102626-T).

## References

[1] Zhang C, Li X, Jiang L, Tang D, Xu H, Zhao P, Fu J, Zhou Q, Chen Y (2021) 54, Adv Funct Mater 3D Printing of Functional Magnetic Materials: From Design to Applications.

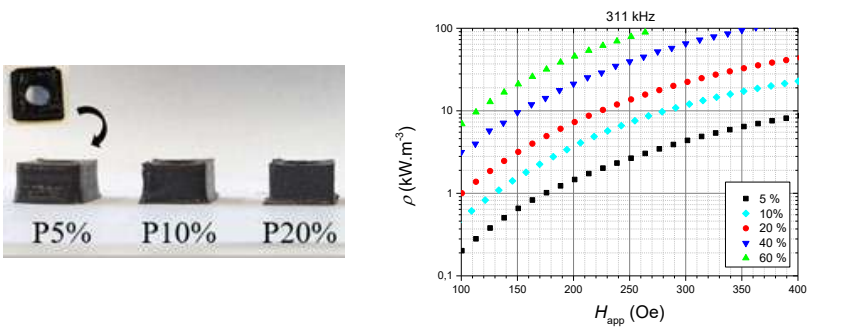



Figure: 3D printed manufactured magnetic composites with their heating power density, $\rho$, as a function of the applied magnetic field, $H_{app}$, for different magnetic NPs mass concentration (%) and the AC magnetic hysteresis loop for the 5% sample .

# Magnetism Of Polyhedral Antiferromagnetic Manganese Oxide Nanostructures

*Irene da Costa Barreiro[1,2]*, Beatriz Rivas Murias[3], Begoña Puértolas[2], Verónica Salgueiriño[1,2]*

[1] *Departamento de Física Aplicada, Universidade de Vigo, Vigo, Spain*
[2] *CINBIO, Universidade de Vigo, Vigo, Spain*
[3] *CACTI, Universidade de Vigo, Vigo, Spain*
*irene.costa@uvigo.es

Since the shape anisotropy of nanomaterials influences their properties [1], various modifications of the synthetic pathway to produce MnO nanocrystals with different final shapes were investigated.

In this context, we prepared octahedral or branched MnO nanoparticles by varying both the amount of oleic acid and the temperature of an intermediate step during the synthetic process [2]. The morphology of the resulting materials was examined by TEM and SEM, and the crystalline structure by X-Ray diffraction and Raman spectroscopy. The thermal stability of the nanostructures, studied through the increment of an applied power of the incident laser in Raman, showed that the branched sample evolves faster to the tetragonal $Mn_3O_4$ final phase than the octahedral-shaped sample. In addition, the magnetic characterization reflected interesting differences due to the presence of a thin $Mn_3O_4$ outer layer formed by the passivation of the as-synthesized particles in air and stabilized in a cubic symmetry, which could lead these nanocrystals to be very attractive for further technological applications such as catalysis [3].

**Acknowledgements**

The authors acknowledge the financial support from, the Spanish Ministerio de Ciencia e Innovación under project PID2020-119242RB-I00 and the European Union H2020-MSCA-RISE-2019 PEPSA-MATE project.

**References**

[1] E. Hao, G.C. Schatz, J.T. Hupp, *Journal of Fluorescence.* **14**, (2004) 331–341.
[2] B. Rivas-Murias, I. da Costa Barreiro, B. Puértolas, V. Salgueiriño (2023, in preparation).
[3] M.M. Najafpour, M. Hołyńska, S. Salimi, *CoordChemRev.* **285**, (2015) 65–75.

# Microscopic and Magnetic Analysis of Iron Oxide Core-Shell Nanoparticles with Variable Core/Shell Ratio

*Ullrich, A.[1]*, Kühn, M.[1], Albrecht, M.[1]*

[1] *Institute of Physics, University of Augsburg, , Universitätsstr. 1, 86159 Augsburg, Germany*

*aladin.ullrich@physik.uni-augsburg.de

Iron oxide nanoparticles of about 15 nm were prepared by thermal decomposition of an iron oleate precursor in 1-Octadecene. Sodium oleate was used as surfactant to produce cubic shaped particles [1].

From transmission electron microscopy (TEM) and electron energy loss sp ectroscopy (EELS) measurements a core-shell composition of the particles was found. The particles show a wustite like structure in the core and a spinel like structure in the shell that is indicated from signatures in the EELS spectra [2]. Figure 1a shows a peak shift of the Fe $L_3$ edge that indicates the oxidation state of the Fe, as well as a prepeak at the O-K edge that is an indicator for the spinel structure of magnetite or maghemite, respectively, and is absent in the case of wustite. Figure 1b shows a dark field scanning TEM image of the as-synthesized sample, while Fig. 1c shows a color coded chemical map revealing the homogeneity of the core-shell structure based on the peak shift of the Fe $L_3$ edge.

By successive oxidation of the particles under a controlled atmosphere, the core/shell ratio could be adjusted up to the complete disappearance of the core, as can be seen in Fig. 1d. The magnetic properties of this antiferromagnetic core - ferrimagnetic shell system were investigated for a sample series with core/shell ratios from about 0.3 to zero. The change of the magnetic properties like the blocking temperature, the coercivity, and the exchange bias field (Fig. 1e) was investigated.

Magnetic memory measurements following different protocols (intermittent field cooled as well as zero field cooled) were also performed at that series. These experiments indicate the presence of a super spin glass state in the samples.

## References

[1] A. Ullrich, M.M.Rahman, A. Azhar, M. Kühn,, M. Albrecht, *Nanoprticle Research* **24** (2022) 183.

[2] A. Ullrich, M.M. Rahman, P. Longo, S. Horn, *Scientific Reports* **9** (2019) 19264.

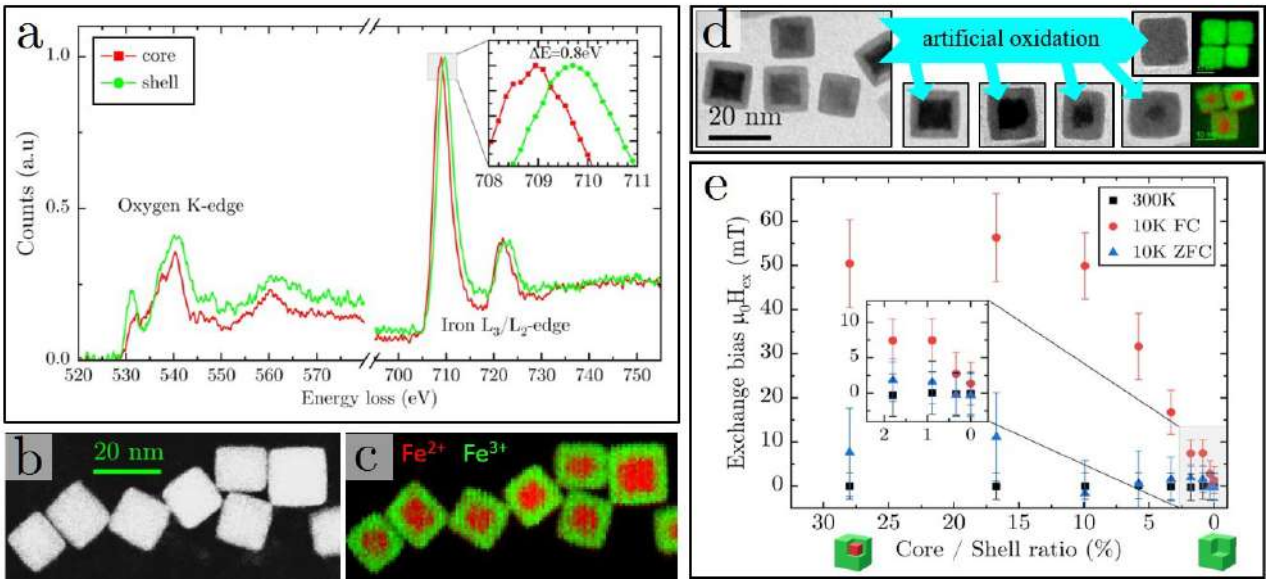



Figure: a) EELS specta of the core and shell region of the nanoparticles. b) Scanning TEM dark field image of the as-prepared samples. c) Chemical map based on the Fe $L_3$ peak position. d) Core/shell ratio change during the artificial oxidation process. e) Exchange bias field of the particles vs. core/shell ratio.

# Néel temperature and morphology tuning of CoO nanoparticles basing on polyol synthesis

*Miran Baričić[1]*, David Hrabovsky[2], Souad Ammar[1], Davide Peddis[3]*

[1] *ITODYS, UMR-CNRS 7086, Université Paris Cité, 15 Rue Jean Antoine de Baif, 75205 Paris, France*

[2] *IMPMC, UMR-CNRS 7590, Sorbonne Université, 6 Place Jussieu, 75005 Paris, France*

[3] *DCCI, Università degli Studi di Genova, Via Dodecaneso, 31, 16146 Genova, Italy*

*miran.baricic@u-paris.fr

Transition metal oxides (TMO) form an important class of functional materials. In the form of nanoparticles (NPs), their size distribution and aggregation state exert an strong influence onto their magnetic poperties [1] and – thus – their applicative potential. Despite the existence of several common synthesis methods, colloidal chemistry routes are the most appropriate when for having control on the NPs morphology, due to the possibility to strictly control their nucleation and growth (solute diffusion, primary particles aggregation…). Surfactant and organic solvents can drive the growth processes towards different outcomes, basing on their chemical and physical properties (polarity, viscosity, steric hindrance etc.), sometimes leading to interesting phenomena, such as oriented aggregation of nanocrystals to form mesocrystals [2], [3]. Modifying the synthesis to obtain different morphologies is an interesting perspective from the point of view of magnetic properties, and interesting correlations can be made between the use of a certain solvent or surfactant, its effect on the material's nanostructure, and the final magnetic properties. In this context, the growth of CoO NPs in polyol was studied focusing on the polyol chain length[4] and on the water amount effect on the morphology of the resulting NPs, thus allowing to easily tune NPs' aggregate size (from ~ 20 to ~ 150 nm), shape (octahedra, octahedral aggregates, and spherical/spheroidal aggregates) and crystalline size (~ 8 to ~ 26 nm). The crystalline size of the aggregates was found to have a regular influence on CoO's Néel temperature, thus providing an easy way to regulate it over ~ 80 K. Further, good synthetic conditions were found to avoid the formation of ferromagnetic layered hydroxide impurities[5], allowing us to unambiguously determine CoO's magnetic properties.

**Acknowledgements**

…

**References**

[1] C. Binns, *Nanomagnetism: Fundamentals and Applications*. Elsevier, 20142014.

[2] M. Niederberger en H. Cölfen, *Phys. Chem. Chem. Phys.*, **2006** , *8*, 3271–3287,

[3] A. P. Alivisatos, *Science (80-. ).*, **2000** , *289*, 736–737.

[4] T. Gaudisson *et al.*, *CrystEngComm*, **2021** , *23*, 1756–1764,

[5] L. Poul, N. Jouini, en F. Fiévet, *Chem. Mater.*, **2000** , *12*, 3123–3132.

# Functionalized Magnetic Iron Oxide/Glycine Imines Nanoparticles for Magnetic Hyperthermia Therapy

*Juliana Araújo,[1,2]*, Chrislaura Carmo[1], Laura C. J. Pereira[2], Abílio J. F. N. Sobral[1]*

[1] *Department of Chemistry, University of Coimbra, 3004-535 Coimbra, Portugal*
[2] *C2TN and Department of Nuclear Sciences and Engineering, Instituto Superior Técnico, Universidade de Lisboa, 2695-066 Bobadela LRS, Portugal*
*juliana.g.araujo@gmail.com

Cancer is one of the leading causes of death in the world, being characterized by the development of abnormal cells that divide uncontrollably and have the ability to spread to other parts of the body. Despite the recent advances in cancer diagnosis and treatment, the actual therapies show some limitations involving selectivity and effectiveness [1]. Magnetic Hyperthermia Therapy (MHT) is an alternative approach for cancer treatment based on the use of magnetic nanoparticles to generate heat when exposed to an alternating magnetic field, which is clinically applied in the treatment of various human solid cancers. The cancer cells are more sensitive to temperature changes than normal cells, and when they reached temperatures around 42-45°C can result in cancer cell death. The heat production will depend on the magnetic behaviour of the material used [2]. Iron oxide nanoparticles have strong magnetic properties and are biocompatible and non-toxic, showing some interest in medical applications, especially in magnetic hyperthermia. The surface of nanoparticles can be functionalized with a variety of organic and organometallic ligands [3a,3b].

In this study, we synthesized Schiff bases-functionalized iron oxide nanoparticles starting with the functionalization of $Fe_3O_4$ with glycine, to which the aldehydes were subsequently attached. The objective was to evaluate how the different coatings in the iron oxide nanoparticles can influence their magnetic behaviour when exposed to a magnetic field. Structural characterization by means of powder XRD, microscopy and thermal analysis was also performed. All the samples exhibited superparamagnetic behaviour at room temperature. As example, figure 1 shows the results for $Fe_3O_4$@(3,5-dihydroxybenzylidene(ethyl)amine), showing a saturation magnetization of 18.0 emu/g and a blocking temperature of 38.4K which reveals that the magnetic core still exhibits magnetic behaviour.

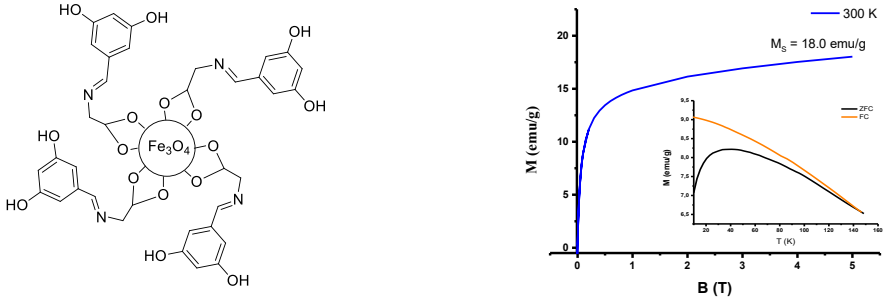



Figure 1: Magnetization curves for $Fe_3O_4$@(3,5-dihydroxybenzylidene(ethyl)amine).

## Acknowledgements

Thanks to ChemMat PhD program from the Fundação para a Ciência e Tecnologia (FCT, MEC, Portugal) for their general financial support (PD/BD/150440/2019) (SFRH/BPD/103103/2014), and to Centro de Química (UIDB/00313/2020) of University of Coimbra. Authors thank also C2TN, (UID/Multi/04349/2022, and LISBOA-01-0145-FEDER-022096) (IST, UL) and Trace Analysis and Imaging Laboratory (TAIL-UC) funded under QREN-Mais Centro Project ICT_2009_02_012_1890.

## References

[1] P. I. P. Soares, J. Romão, R. Matos, J. C. Silva, J. P. Borges, *Prog. Mater. Sci*. **116** (2021) 100742
[2] X. Liu, Y. Zhang, Y. Wang, W. Zhu, G. Li, X. Ma, Y. Zhang, S. Chen, S. Tiwari, K. Shi, S. Zhang, H. M. Fan, Y. X. Zhao, X.-J. Liang, Theranostics, **10** (2020), 3793-3815
[3] a) S.-H. Liao, C.-H. Liu, B. P. Bastakoti, N. Suzuki, Y. Chang, Y. Yamauchi, F.-H. Lin, K. Wu, Int. J. Nanomedicine, **10** (2015), 3315-3328; b) A. Ghorbani-Choghamarani, L. Shiri, G. Azadi, *Res Chem Intermed*, **42** (2016), 6049-6060

# Scrutinizing Magnetic Interactions in Co/Co-oxide Nanoparticles with Different Sizes and Oxide Morphology

*M. González de la Vega[1]*, M.P. Fernández García[1], N. Rinaldi-Montes[1], L. Marcano[1], A. M. Pérez-Mas[1], M. Sevilla[2], A.B. Fuertes[2], J.A. Blanco[1] and P. Gorria[1]*

[1] *Department of Physics, University of Oviedo, C/ Leopoldo Calvo Sotelo 18, 33007, Oviedo, Spain*
[2] *Carbon Science and Technology Institute (CSIC), C/ Francisco Pintado Fe 26, 33011, Oviedo, Spain*
*mariagonzalezv10@gmail.com

Co-based nanostructured systems have been extensively investigated due to their scientific interest related to nanoscale magnetism and their promising technological applications in our modern society. Regarding the magnetic behaviour, bulk metallic Co is ferromagnetic with a Curie temperature above 1350 K. Below a critical diameter of around 60 nm Co nanostructures behave as magnetic single domains exhibiting blocked or superparamagnetic regimes. Apart from that, cobalt nano-entities are prone to oxidise into CoO and $Co_3O_4$ which are instead antiferromagnets with Néel temperatures below those of their bulk counterparts (about 290 K and 40 K, respectively). Then, a typical core (metallic-Co)/shell (Co-oxide) morphology is formed in most of the cases when the size is reduced down to the nanometer length-scale, and the interaction between the magnetic moments in these core/shell nanoparticles (NPs) gives rise to the exchange bias (EB) phenomenon [1-3]. EB is of great relevance for multiple technological applications such as spintronics, magnetic recording media or permanent magnets. Thus, understanding the effect of the microstructural changes on the magnetic behaviour of these core/shell NPs is a crucial step to optimise their performance.

Joint complementary experiments revealed an intricate interplay between short- and long-range effects in NPs of different transition metals (Fe, Co, Ni, Cr, …) embedded in porous amorphous carbon matrices synthesized via a pyrolysis procedure at high temperature [4-8]. Herein, we report on the influence of the metallic Co NPs sizes', the morphology and the type of the oxide-shell in the magnetic properties that allow us to obtain a self-consistent picture that describes the magnetism of Co and its oxides at the nanoscale.

**Acknowledgements**

The authors thank financial support from Gobierno del Principado de Asturias and FICyT through research project AYUD/2021/51822.

**References**

[1] D. De, O. Iglesias, S. Majumdar and S. Giri, *Phys. Rev. B* **94** (2016) 184410.
[2] M. Ghoshani, M. Mozaffari, M. Acet, M. Hosseini and D. Vashaee, *Nanomaterials* **13** (2023) 880.
[3] J. Nogués, V. Skumryev, J. Sort, S. Stoyanov and D. Givord, *Phys. Rev. Lett*. **97** (2006) 157203.
[4] M. P. Fernández-García *et al.*, *Phys. Chem. Chem. Phys.* **13** (2011) 927.
[5] N. Rinaldi-Montes *et al.*, *J. Mater. Chem. C* **4** (2016) 2302.
[6] M. P. Fernandez-Garcia *et al.*, *Phys. Rev. B* **81** (2010), 094418
[7] N. Rinaldi-Montes *et al.*, *J. Mater. Chem. C* **3** (2015) 5674.
[8] N. Rinaldi-Montes, *et al.*, *J. Mater. Chem. C* **10** (2022) 1798.

# High Throughput Analysis of Surface-Functionalized Superparamagnetic Particles in Dynamic Magnetic Field Landscapes

*Yahya Shubbak[12]*, Rico Huhnstock[12], Kristina Dingel[23], Nikolai Weidt[12], Bernhard Sick[23] and A. Ehresmann[12]*

[1] *Institute of Physics & Center for Interdisciplinary Nanostructure Science and Technology (CINSaT), University of Kassel, D-34132 Kassel*

[2] *AIM-ED – Joint Lab Helmholtzzentrum für Materialien & Energie, D-14109 Berlin*

[3] *Intelligent Embedded Systems, University of Kassel, D-34121 Kassel*

*y.shubbak@physik.uni-kassel.de

In order to meet the growing need for rapid and affordable medical diagnostic tools [1, 2], lab-on-a-chip (LOC) devices are investigated as a potential solution by a large and growing scientific community [3–8]. From the perspective of a medical device, LOC systems may be seen as a scaled-down version of standard in-vitro diagnostics [9]. They can be mass-produced and thus allow, for example, the cheap and fast detection of diseases in developing countries [9]. A promising approach is hereby based on the microfluidic handling of micro- or nanometer-sized superparamagnetic particles (MPs) that are functionalized with capture molecules [3, 7, 10–12]. As an advantage, the magnetic particles are remotely controllable by external magnetic fields [7]. They can then induce specific binding events with analytes in the surrounding liquid, provoking a change in the particles' mobility or equilibrium distance relative to the substrate surface [7,10, 11, 13]. Here, we demonstrate a novel method harnessing AI-enhanced fully automated optical recognition algorithms [14] to analyze particles' 3D-motion behaviour changes to liquid-mediated surface-to-surface (particle-to-substrate) interactions, using video recordings captured by a high-speed camera via an optical microscope. Using the former, we demonstrate the different magnetophoretic mobilities of equally sized MPs (2 µm diameter) with two different chemical surface functionalization within a prototypical transport experiment due to their liquid mediated particle-surface to substrate surface interactions. Here, the MPs were directed within a dynamically transformed magnetic field landscape above a topographically flat but magnetically patterned substrate. Our analysis shows possible discrimination between the two MP species purely based on their different magnetophoretic mobilities. This proof-of-principle moves the concept further to life science applications where, e.g., bound viruses only induce a minuscule change in the MPs' surface properties and still allow for a separation of particles based on their surface functionalization.

## References

[1] A. Haleem, M. Javaid, R. P. Singh, R. Suman, and S. Rab, *Sensors International* **2** (2021), p. 100100.
[2] A. O. Oyewole, L. Barrass, E. G. Robertson, J. Woltmann, H. O'Keefe, H. Sarpal, K. Dangova, C. Richmond, and D. Craig, *Diagnostics* **11.2** (2021), p. 182.
[3] M. Reginka, H. Hoang, O. Efendi, M. Merkel, R. Huhnstock, D. Holzinger, K. Dingel, B. Sick, D. Bertinetti, F. W. Herberg, and A. Ehresmann, *Langmuir* **37**.28 (2021), pp. 8498– 8507.
[4] J. Knight, *Nature* 418.6897 ( 2002), pp. 474–475.
[5] L. J. Kricka, *Clinica Chimica Acta* **307**.1- 2 (2001), pp. 219–223.
[6] C. A. Holland and F. L. Kiechle, *Current Opinion in Microbiology* **8.5** (2005), pp. 504–509.
[7] A. Ehresmann, I. Koch, and D. Holzinger, *Sensors* **15.11** (2015), pp. 28854– 28888.
[8] W. Wang, Y. Wang, L. Tu, Y. Feng, T. Klein, and J.-P. Wang, *Scientific Reports* **4.1** (2014).
[9] M. Schönberger and M. Hoffstetter. *Future Trends in Medical Plastic Engineering and Manufacturing*. Elsevier Science & Technology Books, 2016, p. 302.
[10] S. Rampini, P. Li, D. Gandhi, M. Mutas, Y. F. Ran, M. Carr, and G. U. Lee, *Scientific Reports* **11.1** (2021), p. 5302.
[11] D. Gandhi, P. Li, S. Rampini, C. Parent, and G. U. Lee, *Scientific Reports* **10.1** (2020).
[12] A. Jarosz, D. Holzinger, M. Urbaniak, A. Ehresmann, and F. Stobiecki, *Journal of Applied Physics* **120.8** (2016), p. 084506.
[13] D. Issadore, Y. I. Park, H. Shao, C. Min, K. Lee, M. Liong, R. Weissleder, and H. Lee. "Magnetic sensing technology for molecular analyses". In: *Lab on a Chip* 14.14 (2014), p. 2385.
[14] K. Dingel, R. Huhnstock, A. Knie, A. Ehresmann, and B. Sick, *Computer Physics Communications* **262** (May 2021), p. 107859.

# Magnetic Morphology and Exchange-Coupling in Cobalt-Doped Iron Oxide Core-Shell Nanoparticles

*Nahal Rouzbeh,[1,2]*, Sabrina Disch[1,2]*

[1] *University of Cologne, Greinstraße 4-6, 50939 Cologne, Germany*
[2] *University Duisburg-Essen, Universitätsstraße 7, 45117 Essen, Germany*
*nahal.rouzbeh@uni-koeln.de

The exchange coupling in bimagnetic core-shell nanoparticles is a promising pathway to permanent magnetic materials [1]. For iron oxide core-shell nanoparticles, consisting of a wuestite-like particle core and a spinel-type shell, transition metal doping was recently shown to significantly enhance the magnetic anisotropy and exchange coupling [2]. Native iron oxide core-shell nanoparticles synthesized by thermal decomposition of iron oleate typically form as an intermediate through topotaxial oxidation of an initial wuestite phase towards highly defective maghemite [3]. We have recently reported how the combination of such native core-shell nanoparticles (with their alignment of core and shell phases) and cobalt doping leads to a significant enhancement of the exchange pinning between both phases, which is promising for a rational synthesis of nanoparticles with strong coercivity and exchange field. Using magnetic small-angle neutron scattering (SANS) [4, 5], we have unambiguously revealed a significant net magnetization even in the wüstite-type nanoparticle core that is commonly presumed antiferromagnetic or paramagnetic at room temperature [6].

In this contribution, we will present the systematic influence of a subtle variation in particle size on the exchange coupling within such native core-shell, cobalt-doped iron oxide nanoparticles. For freshly synthesized samples with a particle diameter ranging from 8.2 to 9.1 nm, a clear transition from exchange spring to exchange bias behavior is evident. We employ magnetic SANS to elucidate the intraparticle magnetization individually for the wuestite-like particle core and the spinel-type shell and to follow their coupling mechanism. We will moreover follow the effect of progressive topotaxial oxidation in these samples, revealing if the nature of the coupling changes with oxidation state between core and shell.

**Acknowledgements**

The ISIS Neutron and Muon Source is acknowledged for provision of beamtime at the ZOOM instrument under assistance of D. Honecker and D. Alba Venero. We acknowledge the DFG for financial support (Emmy Noether grant DI 1788/2-1).

**References**

[1] A. López-Ortega *et. al., Phys. Rep.* **553** (2015) 1−32.
[2] B. Muzzi *et. al., Small* **18** (2022) 2107426.
[3] E. Wetterskog *et al., ACS Nano* **7** (2013) 7132–7144.
[4] S. Mühlbauer, S. Disch, A. Michels *et al., Rev. Mod. Phys.* **91** (2019) 015004.
[5] D. Zákutná, S. Disch *et al., Phys. Rev. X* **10** (2020) 031019.
[6] D. Zákutná, N. Rouzbeh, S. Disch *et al., Chem. Mater.* (2023). https://doi.org/10.1021/acs.chemmater.2c02813

# Towards Quantitative Imaging of Magnetic Nanoparticles: A Comparison of MPI, MRXI and MRI

*Soudabeh Arsalani[1]*, Olaf Kosch[1], Norbert Löwa[1], Dirk Gutkelch[1], Patricia Radon[1], Alaor Augusto[2], Daniel Baumgarten[3] and Frank Wiekhorst[1]*

[1]*Physikalisch-Technische Bundesanstalt, Abbestrasse 2-12, D-10587 Berlin, Germany*

[2] *Institute for Diagnostic and Interventional Neuroradiology, University Hospital LMU Munich, Germany*

[3]*Institute of Electrical and Biomedical Engineering, UMIT TIROL-Private University for Health Sciences and Health Technology, Hall in Tirol, Austria*

* email:soudabeh.arsalani@ptb.de

Magnetic nanoparticles (MNPs) have shown excellent potential for cancer therapy and diagnosis. For therapy applications such as drug delivery and magnetic hyperthermia, it is necessary to localize and quantify the MNP distribution inside a specific body region (e.g., brain, breast, prostate etc.) before treatment. There are several techniques, such as magnetorelaxometry imaging (MRXI), magnetic particle imaging (MPI) and magnetic resonance imaging (MRI), that are capable to provide this information [1, 2]. All these imaging modalities have some pros and cons. Here, we aimed for a comparison of their quantitative imaging capability of MNPs in various 3D printed phantoms.

In our first experiment, we used MPI to detect and quantify a flow of MNPs in a brain vessel structure phantom. This phantom, which contains straight and curved sections of 8 cm total length, was derived from an MRI scan and constructed by silicone 3D printing. To provide flow, the phantom was connected to a syringe pump, see Figure 1a, and the syringe was filled with a mixture of glycerine (59.1%) and distilled deionised water (40.9%), to mimic the viscosity of blood. First, a bolus of about 300 µL of Synomag (Micromod Partikeltechnologie, Germany) with an iron concentration of 2 mmol/L was inserted into the phantom where a flow rate of 3 mL/min was maintained. The flow was imaged using a preclinical 3D-MPI system (Bruker BioSpin GmbH, Ettlingen, Germany) and a PTB made MRXI device. The geometry of the silicone phantom before and after the bolus experiments was investigated using a preclinical MRI scanner (ICON, 1 T magnet, Bruker, Germany). We could successfully visualise and reconstruct the MNP bolus movements in the different parts of the brain vessel structure phantom, see Figure 1b for an MPI image reconstruction. The evaluation of the quantitative localisation of MNPs in the phantom using the combination of MRXI, MPI, and MRI will be presented.

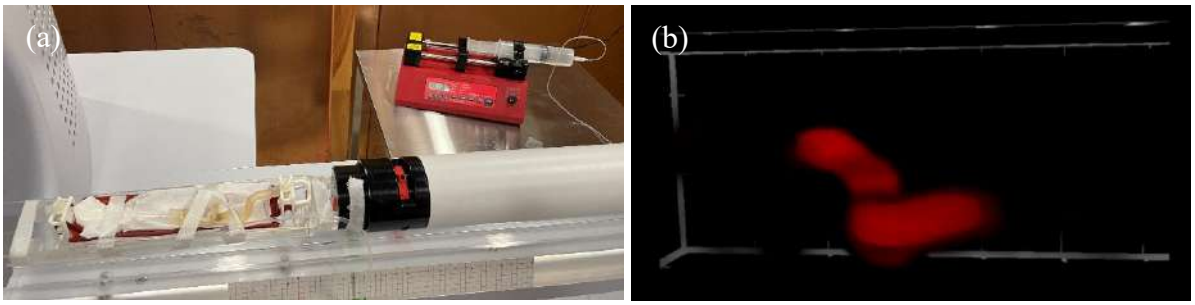



Figure 1: (a) Image of the pump, syringe and vessel structure phantom in the MPI sample holder. (b) MPI image reconstruction of MNP flow inside the phantom.

## Acknowledgements

Financial support by the German Science Foundation (DFG), project ‘quantMRX’, Grant 428329263, WI4230/4-1 and Austrian Science Fund (FWF), Grant number I 4357-B, is gratefully acknowledged.

## References

[1] S. Arsalani, P. Radon, et al., *Phys. Med. Biol*. 67, (2022), 225007.
[2] M. Liebl, F. Wiekhorst, et al., Biomed. Eng. Biomed. Tech. 60, (2015): 427–443.

# Magnetic Flocculation for Water Treatment

*P. A. Augusto [1,2], T. Castelo-Grande[3], D. Barbosa[3]*

*(1) Dep. Chemical Engineering, Faculty of Chemical Sciences, University of Salamanca, Spain.*
*(2) Institute of Molecular and Cellular Biology of Cancer, GIR Cytomic, Univ. Salamanca,Spain.*
*(3) LEPABE – Laboratory for Process Engineering, Environment, Biotechnology and Energy, Faculty of Engineering, University of Porto, Rua Dr. Roberto Frias, 4200-465 Porto, Portugal*
* pauloaugusto@usal.es

Many of the current wastewater and leachate treatment techniques have very high maintenance and operating costs (e.g inverse osmosis, etc.) and are also not as environmental-friendly or sustainable as desired. The treatment via coagulation-flocculation (CF) requires simple operating conditions that, associated with low operating costs make it one of the most popular options for surface water treatment with colloidal particles, and phosphorus and organic compounds. Furthermore, the use of magnetic flocculants is an important step forward towards quickness and sustainability, as these magnetic flocculants may reduce the operating time, increase efficiency of treatment and be easily recovered and then regenerated and reused. In this work we research on the application of this magnetic method.

***Methodology***: The main steps followed were: 1st – Magnetic nano and microparticles synthesis; 2nd – Preparation of magnetic flocculants; 3rd – Sample Treatment: a) Coagulation; b) Flocculation; c) Sedimentation; d) Magnetic decantation 4th – Recovery of magnetic particles and regeneration; 5th – Reuse; 6th – Effluent analysis and treatment efficiency determination. ***Magnetic nano and microparticles synthesis***: The manufacture of magnetic particles was performed usually by co-precipitation methods, although some other methods have been applied. The magnetic particles obtained are all magnetite-core based. ***Flocculation and Coagulation***: as a coagulant we have used aluminum sulfate (Al2(SO3)3), and as flocculant magnetite-polyacrylamide. ***Regeneration methods***: milling-decantation, US and chemical destabilization was used and then magnetic particles isolated and recovered by magnetic separation. ***Re-use***: after the application of particles cycles of re-use were studied allied with the regeneration when needed. ***Economical analysis***: A full economical and technological viability analysis was done.

**Main Results and Discussion** – Some results are presented in this section as examples and the full set presented in the congress. In Image 1 are presented photographs initial wastewater samples and the final status of these samples after application of the applied magnetic process. In Table I are presented some efficiency results of the applied process and in Table II some of the results obtained concerning cases of reuse (including some regeneration steps when required).

**ıage 1.** Wastewater Samples before (left) and after MCF process (right)

**Tabla I.** Samples content and Processes Efficiency

| Sample | Content | Efficiency |
|---|---|---|
| Wastewater | SST – 132 mg/L; COD – 363 $mgO_2/l$ | |
| After regular Coagulation-Flocculation | SST – 36 mg/L; COD – 146 $mgO_2/l$ | 73 % (TSS); 60 % (COD) |
| After Magnetic Coagulation-Flocculation | SST – 7.5mg/L; COD – 34 $mgO_2/l$ | 95 % (TSS); 91 % (COD) |

**Tabla II.** Reusing cycles

| Process | Nr cycles | Min efficiency |
|---|---|---|
| Magnetic Coagulation - Flocculation | 10 | 92% (TSS); 90% (COD) |

**Conclusions –** We have developed and researched successfully the application magnetic coagulation and flocculation in the treatment of wastewater, studying also with success the possibility of regenerating and re-using the magnetic particles to efficiently improve their sustainability.

**Acknowledgements**

This research was funded by: (a) Project 2019/00057/001, "Application of Magnetic Methods and Magnetic Substances for Environmental Treatment and Biomedicine", 26 RLD-PXIV, USAL, Spain. (b). Base Funding—UIDB/00511/2020 of the Laboratory for Process Engineering, Environment, <Biotechnology and Energy—LEPABE—Funded by national funds through the FCT/MCTES (PIDDAC); (c). Project POCI-01-

0145-FEDER-028020—Funded by FEDER funds through COMPETE2020—Programa Operacional Competitividade e Internacionalização (POCI) and by national funds (PIDDAC) through FCT/MCTES; (d) Project 2SMART – engineered Smart materials for Smart citizens, with reference NORTE-01-0145-FEDER-000054, supported by Norte Portugal Regional Operational Programme (NORTE 2020), under the PORTUGAL 2020 Partnership Agreement, through the European Regional Development Fund (ERDF). Lorenzo Hernández y A. Rodríguez (University of Salamanca).

**References**

[1] – M. S. Al-Rubaie, M. A. Dixon, T. R. Abbas, Desalinat and Water Treatment, 53 (8), (2015) p. 2086
[2] – C. Wang, Y. Wang, et al. Separation Science and Technology, 53(5), (2018), p. 814.

# Applications of Magnetic Fenton and PhotoFenton in Wastewater Treatment

*T. Castelo-Grande[1], P. A. Augusto [2,3], L. Hernández[3], A. Rodriguez[3], D. Barbosa[1]*

*(1) LEPABE – Laboratory for Process Engineering, Environment, Biotechnology and Energy, Faculty of Engineering, University of Porto, Rua Dr. Roberto Frias, 4200-465 Porto, Portugal*

*(2) Dep. Chemical Engineering, Faculty of Chemical Sciences, University of Salamanca, Spain.*

*(3) Institute of Molecular and Cellular Biology of Cancer, GIR Cytomic, Univ. Salamanca,Spain.*

*pauloaugusto@usal.es

Fenton and PhotoFenton processes have being intensively studied in the last years as new and potential advanced oxidation methods due to their high efficiency in the degradation of contaminants present in underground water, wastewater and leachates. Furthermore, the use of magnetic particle catalysts is an important step forward towards sustainability, as these magnetic substances may increase the efficiency of treatment and be easily recovered and then potentially regenerated and reused. Nonetheless the large majority of magnetic materials used in the process are hard to synthesize and costly magnetite-based substances, a fact that hinders their large-scale applications, and reduces the overall process to small lab-scale applications. Furthermore, the important and potential sustainability contemplated by the recovery, regeneration and reuse of these magnetic particles is usually neglected and not contemplated in most of the research studies performed so far. In this work we research the application of these magnetic methods to several watery media, and the possible regeneration and reuse of the magnetic substances.

**Methods** [1-2] – ***Magnetic nano and microparticles synthesis***: The manufacture of magnetic particles was performed usually by co-precipitation methods, obtaining magnetite or magnetite-chitosan particles and also nZVI particles, although some other methods have been applied. ***Fenton and Photofenton***: for the use in Fenton processes, magnetic particles prepared as previously described, were used together with a solution of $H_2O_2$, and in Photofenton the aid of two UV lamps was also required. ***Regeneration methods***: After the recovery and isolation of the used particles by magnetic separation, these were regenerated specially by using reducing agents and solvents. ***Re-use***: after the application of particles cycles of re-use were studied allied with the regeneration when needed. ***Economical analysis***: A full economical and technological viability analysis was done.

**Main Results and Discussion** – Some results are presented in this section as examples and the full set presented in the congress. In Image 1 are presented photographs initial wastewater samples and the final status of these samples after application of the applied magnetic process. In Table I are presented some efficiency results of the applied process and in Table II some of the results obtained concerning cases of reuse (including some regeneration steps when required).

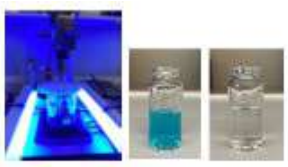


**Image.** Photofenton setup (left), textile contaminated water treatment: beginning sample (center) 24hours treated sample (right)

**Table I.** Processes Efficiency

| Sample | Efficiency |
|---|---|
| Textile simulated wastewater | 100% (MB); 99% (CV) |
| Wastewater | 73 % (TSS); 60 % (COD) |
| Tirasol | 82% (Fent); 92% (Phot)<br>TOC – 35 % (PhotoFenton) |

**Table II.** Reusing cycles

| Process | Nr cycles | Min efficiency |
|---|---|---|
| Fenton Wastewater | 8 | 88% (COD) |

**Conclusions** – We have developed and researched successfully the application magnetic fenton and photofenton to decontamination of WWTP wastewater, textile simulated wastewater, and fenol containing wastewater studying also with success the possibility of regenerating and re-using the magnetic particles to efficiently improve their sustainability.

**Acknowledgements**

This research was funded by: (a) Project 2019/00057/001, "Application of Magnetic Methods and Magnetic Substances for Environmental Treatment and Biomedicine", 26 RLD-PXIV, USAL, Spain. (b). Base Funding—UIDB/00511/2020 of the Laboratory for Process Engineering, Environment, <Biotechnology and Energy—LEPABE—Funded by national

funds through the FCT/MCTES (PIDDAC); (c). Project POCI-01-0145-FEDER-028020—Funded by FEDER funds through COMPETE2020—Programa Operacional Competitividade e Internacionalização (POCI) and by national funds (PIDDAC) through FCT/MCTES; (d) Project 2SMART – engineered Smart materials for Smart citizens, with reference NORTE-01-0145-FEDER-000054, supported by Norte Portugal Regional Operational Programme (NORTE 2020), under the PORTUGAL 2020 Partnership Agreement, through the European Regional Development Fund (ERDF).

## References

[1] - L. Hernández, P.A. Augusto et al., Journal of Environmental Management, 285, (2021) 112155. [2] - P.A. Augusto, T. Castelo-Grande et al. Materials, 13 (10), (2020), 2219

# Fate, Toxicity, Regulation and Management of Magnetic Nanoparticles.

*T. Castelo-Grande[1], P. A. Augusto [2,3], D. Barbosa[1]*

*(1) LEPABE – Laboratory for Process Engineering, Environment, Biotechnology and Energy, Faculty of Engineering, University of Porto, Rua Dr. Roberto Frias, 4200-465 Porto, Portugal*

*(2) Dep. Chemical Engineering, Faculty of Chemical Sciences, University of Salamanca, Spain.*

*(3) Institute of Molecular and Cellular Biology of Cancer, GIR Cytomic, Univ. Salamanca,Spain.*

*castelogrande@sapo.pt

Nanomaterials (NMs) exhibit unique properties due to their size, structure, physical-chemical properties, and therefore its range of applications has grown exponentially. Even though many benefits from this area are expected, some concerns with their potential effect in humans and in the environment has arisen. Several recent studies, analysis and assessments on hazardous risks of NMs have demonstrated some adverse effects on environment, human beings and ecosystems. This work discusses the recognized and potential nanomaterials effects in humans and in the environment, their fate, and the (in)existence of specific regulations for the production, handling and disposal of these potential harmful materials, even though absolute conclusions cannot be drawn [1-2].

As a first step of the analysis the different type of nanoparticles and production that exist are reviewed (two main groups: natural nanoparticles; engineered nanoparticles - which may be divided in many subclasses, but usually are divided into carbon-based ENPs and inorganic ENPs).

Then the development of Nanoscience - the EU 2020 and Horizon Europe strategy - will be detailed as for most scientists the arrival of nanotechnologies is the main turnoff point of the 21st century, concerning industrial development. Therefore, in the European Union (EU) 2020 and Horizon Europe Strategy, nanotechnology was and is considered a main emerging and enabling technology. Databases and some Regulation was established, and these will be reviewed: NanoProducts Data Bases – e.g. Nanotechnology Consumer Product Inventory, Nano Products and Technologies, Nanopartikel, etc.; Nanomaterials Regulation – e.g. EPA's Toxic Substances Control Act; Federal Insecticide, Fungicide, and Rodenticide Act; European Food Safety Authority (EFSA); etc. Nonetheless, no real clear specific regulation was developed so far, as entities are waiting on results coming from previous and currently running funded projects, such as e.g. REACH- Guidance for nanomaterials Registration, Evaluation, Authorization and restriction of Chemicals, FutureNanoNeeds, MARINA – Managing Risks of Nanomaterials, etc.

Risk Assessment (RA) has been proposed as a prime process to evaluate EHS risk and for decision making, where risk assessment experts expect to understand what can go wrong, and how to be expected to happen, and also the consequences of it happening. In this work focus will be given specially to Environmental Risk Assessment and Life Cycle Assessment.

The ENM Pathways in the Environmental Compartments will be detailed as intrinsic properties of ENMs, as well as the specific environment conditions, will determine their fate, and therefore the behavior and distribution in the different environmental compartment. A major knowledge gap, hindering the valid qualitative and quantitative estimation, is the current lack of understanding on how the novel physicochemical proprieties of ENMs influence the transformations process and hence ENM behavior in the environment. Three main compartments will be reviewed: Fate in Air; Fate in Soil ; Fate in Water.

An overview of Nanometrology methods will be given. In the last decade methods such as fractionation, light scattering, and spectroscopic for detection and characterization of nanomaterials have been developed mostly to determinate the dimension and description of nanomaterials. Current methods will be reviewed.

ENM Ecotoxicity scopes will be also briefly introduced as these are on different levels including microorganisms, plants, invertebrates and vertebrates, and standardized test systems, which can be found in protocols approved by OECD or ISO.

**Acknowledgements**

This research was funded by: (a) Project 2019/00057/001, "Application of Magnetic Methods and Magnetic Substances for Environmental Treatment and Biomedicine", 26 RLD-PXIV, USAL, Spain. (b). Base Funding—UIDB/00511/2020 of the Laboratory for Process Engineering, Environment, Biotechnology and Energy—LEPABE—Funded by national funds through the FCT/MCTES (PIDDAC); (c). Project POCI-01-0145-FEDER-028020—Funded by FEDER funds through COMPETE2020—Programa Operacional Competitividade e Internacionalização (POCI) and by national funds (PIDDAC) through FCT/MCTES; (d) Project 2SMART – engineered Smart materials for Smart citizens, with reference NORTE-01-0145-FEDER-000054, supported by Norte Portugal Regional Operational Programme (NORTE 2020), under the PORTUGAL 2020 Partnership Agreement, through the European Regional Development Fund (ERDF).

**References**

[1] - A.L. Dale, E. A. Casman,G.V et al. Environ. Sci. Technol., 49, 2015, p.2587.
[2] – S. Møller, P. F. Roel, U. Vogel et al. Environ. Health Persp., 125 (10), 2017, EHP424.

# Multifunctional platform for phothermal hyperthermia combined with luminescence nanothermometry probes.

*Manuel Horcajo Fernández[1*], Daniel Arranz[1,2], Luis García García-Arisco[1], Patricia de la Presa[1]*

[1] *Instituto de Magnetismo Aplicado Salvador Velayos (UCM-ADIF-CSIC), A6 km. 22.5 Las Rozas (Madrid).*

[2] *Dpto. de Óptica, Facultad de Ciencias Físicas, Universidad Complutense de Madrid, Avda. Complutense s/n, 28040 Madrid, Spain*

* mhorcajo@ucm.es

The design of multifunctional magnetic nanoparticles (NPs) that can generate and monitor heat in real-time during thermal therapy are a major challenge in nanomedicine. In this work, a trimodal system that combines magnetic hyperthermia (MH), photothermal therapy (PT) and luminescence nanothermometry (LT) properties has been set up in a single platform. Magnetite NPs were optimized focusing on MH and PT, then, the NPs have been coated with embedded $Nd^{3+}$ or $Er^{3+}$ cations in order to enhance the PT and also as LT probe.

Erbium has excitation lines at 790 nm and 980 nm, and luminescence around 500 nm, in the visible part of the spectrum, which makes it adequate for bioimaging since it is biocompatible. Neodymium is another interesting luminescent probe, with excitation around 800 nm and emission at the second biological window. Such hybrid system could act as heat mediator and imaging probe for in-vitro and in vivo experiments, since these wavelengths belong to the first biological window, as well as an in-situ thermometer during the photothermia application.

The samples was prepared by a modified hydrothermal method [1], the multifunctional system is composed of an iron oxide (IONPs) as core, then a coating shell which serve as insulator between the magnetic core and the luminescent probe, and an additional silica shell containing either $Nd^{3+}$ or $Er^{3+}$. The structure and morphology have been investigated by XRD and HRTEM, and the magnetic properties by SQUID magnetometry. The heating efficiency of this sytem has been measured under continuous laser irradiation of 808 nm [2]. Figure 1 shows the heating curves and the calculated specific heating rate (SAR) of the magnetic core, the core coated with a silica shell, and the final multifunctional system. It is observed the reduction of the SAR with the presence of silica, it is assumed that the silica, wich has a low thermal conductivity, is screening the thermal flow from the magnetic cores to the water suspension, decreasing the heating efficiency of the whole sample.

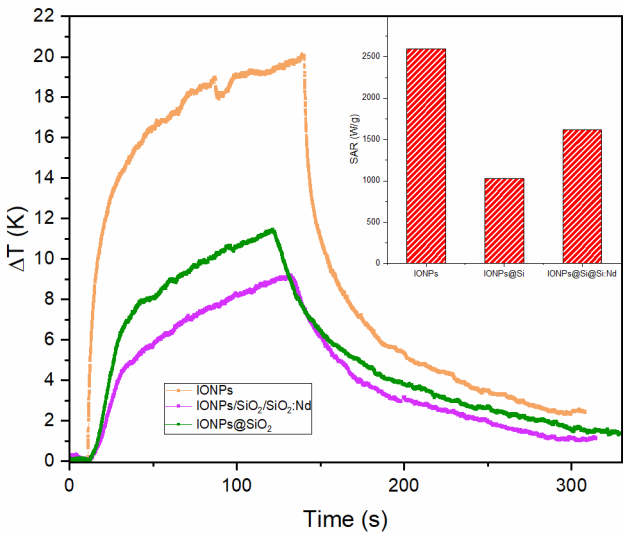



*Figure 1: Heating curves for IONPs, IONPs@$SiO_2$ and IONPs@$SiO_2$@ $SiO_2$:Nd. The inset shows the calculated SAR values.*

The luminescence properties of the multifunctional platform will be discussed along the work.

## Acknowledgments

This work was supported by PID2021-123112OB-C21. We also acknowledge the Sistema Nacional de Garantía Juvenil for the contract through the Comunidad Autónoma de Madrid (PEJ-2020-AI/IND-17711).

## References

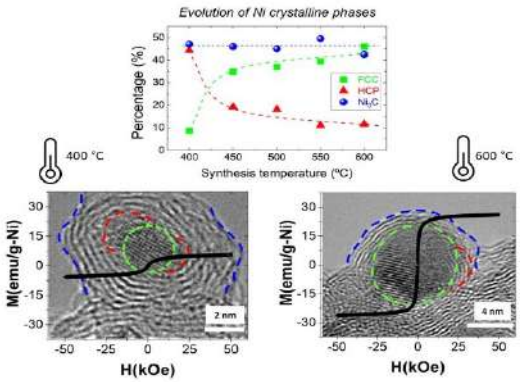
[1] Marcus Vinícius-Araújo, Navadeep Shrivastava, Ailton A. Sousa-Junior, Sebastiao A. Mendanha, Ricardo Costa De Santana, and Andris F. Bakuzis, ACS Appl. Nano Mater. 2021, 4, 2190−2210.

[2] Arranz D, Weigand R, de la Presa P. Towards the Standardization of Photothermal Measurements of Iron Oxide Nanoparticles in Two Biological Windows. Nanomaterials. 2023; 13(3):450.

# Untangling the role of carbon in the magnetic coupling of Ni@C nanoparticles with mixed FCC/HCP crystal structures

*Mona Fadel[1]*, Pablo Álvarez-Alonso[2], Jesús A. Blanco[1], Pedro Gorria[1]*

**Department of Physics, University of Oviedo, 33007, Oviedo, Spain*
*[1]Scientific-Technical Services, University of Oviedo, 33006 Oviedo, Spain*
*[2]National Institute of carbon CSIC, 33080, Oviedo, Spain*
*[Φ]IFIMUP-IN, Rúa do Campo Alegre, s/n, Porto, Portugal*
* Email:uo273017@uniovi.es




## Abstract

Ni@C nanostructured materials are currently attracting a lot of interest due to their many catalytic or magnetic properties. In this article, we discuss a study that looked at the correlation between the magnetic characteristics of Ni nanoparticles implanted in a carbon matrix and their microstructure.. The samples were synthesis of a nickel-imidazole-based metal-organic framework (MOF) in aqueous medium at moderate and carbonized at different temperatures between 400 and 600 ºC to obtain a carbon-supported hybrid material, containing Ni nanoparticles with "artichoke-like" morphology, where a Ni-FCC core is surrounded by "bracts" of Ni-HCP and $Ni_3C$. We demonstrate how the data from X-ray diffraction, electron microscopy, and the evolution of the magnetic behavior with carbonization temperature complement one another by presenting consistent structural and magnetic properties of the investigated Ni@C nanoparticles.

## Acknowledgements

Work at University of Oviedo was financially supported by research projects: RI2018-094683-B-C52 (MINECO, Spain), AYUD/2021/51822 (Gobierno del Principado de Asturias, Spain). Authors would like to acknowledge the technical support provided by Scientific-Technical Services, University of Oviedo, Spain.

## References

[1] Mona Fadel, Pablo Alvarez-Alonso, Jesus A Blanco, Pedro Gorria and etc..., Untangling the role of the carbon matrix in the magnetic coupling of Ni@C nanoparticles with mixed FCC/HCP crystal structures, ***Mater. Chem. C***, 2023, Accepted Manuscript , https://doi.org/10.1039/D3TC00257H

# Optimization and Harmonic Response of $Gd^{3+}$-doped $MnFe_2O_4$ Nanoparticles for MPI Applications

*T. Sakamoto[1], I. Kawaguchi[3], K. Ohara[1], K. Nakazawa[2], K. Nii[1], T. Moriwaki[2], Y. Fujita[1], H. Amano[3], S. Kobayashi[3], T. Shimohama[3], and Y. Ichiyanagi[1,3]**

[1]*Grad. Sch. of Sci. and Eng., Yokohama Nat. Univ.,*

[2] *Grad. Sch. of Environ. and Info., Yokohama Nat Univ.,*

[3] *Grad. Sch. of Sci. and Eng., Yokohama Nat. Univ.*

**yuko @ynu.ac.jp*

Superparamagnetic nanoparticles with high magnetization and low coercivity are expected for the applications in the biomedical field [1]. Magnetic particle imaging (MPI) is one of the new ideas for diagnostic method, which directly visualizes the magnetic response [2]. To obtain signals of magnetic particles, it is possible to control the harmonics of nanoparticles by applying a gradient magnetic field under the AC magnetic field. In this study, DC magnetic field was applied as the gradient magnetic field and field-free point (FFP) was provided as the measurement point. The behavior of the harmonic response of magnetic nanoparticles under an AC magnetic field was observed.

Gd-doped Mn-ferrite nanoparticles embedded in amorphous $SiO_2$ were prepared by our original wet-mixing method. The particle size($d$) was controlled by modulating the annealing temperature. ($d$ = 10.6, 14.0, 17.0, 21.2 nm), and the amount of Gd doping was unified to 6.5%, which was found to have a particularly high response to AC magnetic fields in our previous study. Magnetization measurements ($M$-$H$) were performed using a superconducting quantum interference device (SQUID) magnetometer to observe the differences in the coercive force ($Hc$) and initial permeability ($\mu$) of various size of the particles. The coercivity of the 10.6 nm and 14.0 nm particles was found to be small, while the initial permeability was large for the 14.0 nm and 17.0 nm particles. Based on the magnetic properties of the fabricated samples, harmonic measurements were performed. A sample was placed in a tube and inserted into the AC coil, and the third harmonic response was detected by a lock-in amplifier when an AC magnetic field was applied (Fig. 1). As a result, the largest response was observed for a 14.0 nm particle (Fig. 2). From these results, it can be said that the particles of 6.5% and 14.0 nm are the best particles for MPI applications because they have a small coercive force, can follow the

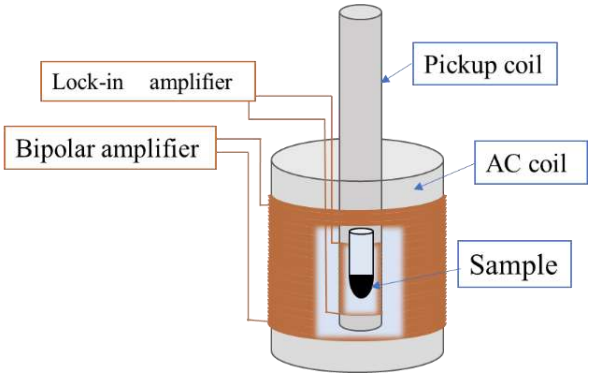



Fig. 1 Schematic diagram of harmonic measurement

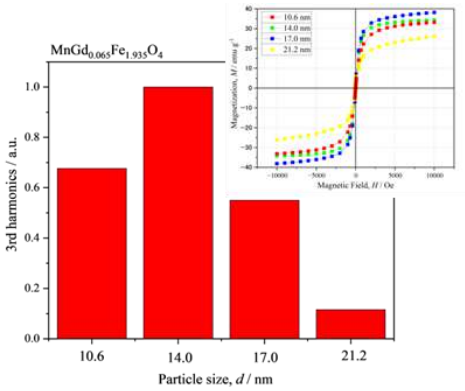



Fig. 2 response results (b) of the third harmonic measurement under AC magnetic field

external magnetic field quickly, and have the largest change in magnetization in the low-magnetic field region.

## Acknowledgements

Grant-in-Aid for Science Research (No. 17H02762, No. 20H00344) from the Japan Society for the Promotion of Science (JSPS), JST-Mirai Program No.JPMJMI17D7, Watanabe Foundation No,280118

## References

[1] A. Oshima, Y. Ichiyanagi et al., *Journal of Nanoscience and Nanotechnology*, 20, 7255-7262, (2020)

[2] J. Borgert et al., *Journal of Cardiovascular Computed Tomography*, 149-153 (2005)

# Ferrite-Based Nanocomposites as Random Field Magnets

*Jaume Calvo-de la Rosa[1,2], Joan Manel Hernàndez[1,2], Jose Maria Lopez-Villegas[2,3], Antoni García-Santiago[1,2,*], and Javier Tejada[1]*

[1] *Dept. de Física de la Matèria Condensada, Universitat de Barcelona, 08028 Barcelona, Spain*
[2] *Institut de Nanociència i Nanotecnologia (IN2UB), Universitat de Barcelona, 08028 Barcelona, Spain*
[3] *Dept. d'Enginyeria Electrònica i Biomèdica, Universitat de Barcelona, 08028 Barcelona, Spain*
*agarciasan@ub.edu

We present experimental evidence of the behavior of modified hexagonal ferrites as random field magnets. Firstly, we investigated the magnetic properties of ceramic nanocomposites generated by modifying a barium hexaferrite through the introduction of divalent cations into its structure. We measured the first magnetization curve and hysteresis cycles of different samples and found that the cation addition results in a considerable increase in the saturation magnetization and susceptibility, as well as in a remarkable drop in the coercivity and the area of the cycle. X-ray Diffraction and Scanning Electron Microscopy showed that the modified samples consisted in mixtures of clusters of two kinds, corresponding respectively to the hard-magnetic hexaferrite and the soft-magnetic divalent ferrite, between which an exchange interaction occurs [1, 2]. This prompted us to study the precise functional dependence of the first magnetization curve approaching saturation by fitting the experimental data to the theoretical model that describes random magnets [3]. Specifically, the behaviors at low and high magnetic fields followed respectively the $1/H^{1/2}$ and $1/H^2$ laws that are expected for such materials. Finally, according to recent theoretical developments, random field magnets have been postulated as strong microwave absorbers [4]. To check this out, we measured the complex permittivity and permeability in the GHz range and found remarkable changes in the theoretical reflection loss coefficient. These findings strongly support the consideration of our ceramic nanocomposites as random field magnets.

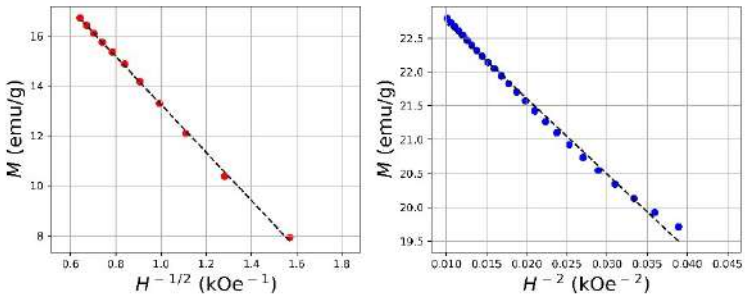



Figure. Low (left panel) and high (right panel) magnetic field dependence of the magnetization approaching saturation for a Mn-modified barium hexaferrite. The linear lines are the fits to the theoretical $1/H^{1/2}$ (left panel) and $1/H^2$ (right panel) laws.

**Acknowledgements**

This work has been supported by the Air Force Office of Scientific Research.

**References**

[1] T. Maeda *et al.*, *J. Magn. Magn. Mater.* **281**, 195 (2004); S. Tyagi *et al.*, *Ceram. Int.* **37**, 2631 (2011).
[2] R. Martínez García *et al.*, *J. Magn. Magn. Mater.* **574**, 168934 (2022).
[3] E. M. Chudnovsky, *J. Appl. Phys.* **64**, 5770 (1988); *J. Magn. Magn. Mater.* **79**, 127-130 (1989).
[4] D. A. Garanin and E. M. Chudnovsky, *Phys. Rev. B* **103**, 214414 (2021); *Phys. Rev. B* **105**, 064402 (2022).

# Low-temperature phase transition in Dy aluminoborate

*T. Zajarniuk[1*], A. Szewczyk[1], E. Lhotel[2], M. U. Gutowska[1], W. Szuszkiewicz[1], S. Petit[3], A. A. Prokhorov[4], R. Puzniak[1], H. Szymczak[1].*

[1] *Institute of Physics, Polish Academy of Sciences, al. Lotnikow 32/46, 02 668 Warsaw, Poland*

[2] *Institut Néel, CNRS & Université Grenoble Alpes, 25 rue des Martyrs BP 166, 38 042 Grenoble, France*

[3] *Laboratoire Leon Brillouin CEA-Saclay, Building 563, 911 91 Gif-sur-Yvette Cedex, France*

[4] *Institute of Physics, AS CR, str. Na Slovance 2, 182 21Praha, Czech Republic*

*zayar@ifpan.edu.pl

Aluminoborates are compounds showing a large magnetoelectric effect. Studies of a magnetic phase transition discovered below 1 kelvin in one of these compounds, $DyAl_3(BO_3)_4$, by means of specific heat, $C_B$, measurements are presented. It was found, that under influence of increasing external magnetic field, $B$, the temperature of the transition decreases, albeit the magnetization and neutron studies of the $DyAl_3(BO_3)_4$ compound showed that the magnetic order appearing below the transition point has a ferromagnetic character with magnetic moments directed along the threefold $c$ axis.

To analyze the results obtained, three contributions to $C_B$, i.e.: the lattice, nuclear, and magnetic ones, were considered. The magnetic contribution was determined by subtracting the estimated lattice and nuclear contributions from the total $C_B$. The lattice contribution, $C_l$, was determined by using two methods. In the first one, $C_l$ was approximated by the sum of the Debye and Einstein models [1] and in the second method, it was considered to be the sum of the two Debye contributions corresponding to two different Debye temperatures [2]. Both methods gave consistent, satisfying results.

Based on the specific heat and magnetization studies, the magnetic Grüneisen ratio, being the most informative parameter, diverging in the vicinity of quantum transitions, was calculated by using the following formula [3,4]:

$$\Gamma = -\frac{1}{T}\frac{\left(\frac{\partial S}{\partial B}\right)_T}{\left(\frac{\partial S}{\partial B}\right)_B} = -\frac{\left(\frac{\partial M}{\partial T}\right)_B}{C_B(T)} = \frac{1}{T}\left(\frac{\partial T}{\partial B}\right)_S$$

Analysis of the dependence of specific heat, magnetization, and the Γ parameter as a function of $T$ and $B$ near the transition point, that we performed, suggests these behaviors to be characteristic of the behaviors near the transition having a quantum nature, i.e. influenced by quantum fluctuations. The results were found to be consistent with those found for the $TbAl_3(BO_3)_4$ aluminoborate [1].

## Acknowledgements

This work was supported partially by the National Science Centre (NCN), Poland, under Project No. 2018/31/B/ST3/03289.

## References

[1] T. Zajarniuk, A. Szewczyk, P. Wisniewski, M. U. Gutowska, R. Puzniak, H. Szymczak, I. Gudim, V. A. Bedarev, and M. I. Pashchenko, Phys. Rev. B **105** (2022) 094418.

[2] J. A. Hofmann, A. Paskin, K. J. Tauer, and R. J. Weiss, J. Phys. Chem. Solid **1** (1956) 45.

[3] L. Zhu, M. Garst, A. Rosch, and Q. Si, Phys. Rev. Lett. **91** (2003) 066404.

[4] M. Garst and A. Rosch, Phys. Rev. B **72** (2005) 205129.

# Magnetic Cationic Effect depending on Microstructure in Copper Ferrite

*M A Cobos[1]*, J A Jiménez[2], I. Llorente[2] ,A. Hernando[14], P de la Presa [13,]*

[1] *Instituto de Magnetismo aplicado (UCM-ADIF-CSIC), A6 km.22'5 - Las Rozas – Madrid- Spain*
[2] *CENIM-CSIC, Av. de Gregorio del Amo, 8, 28040 Madrid – Spain*
[3] *Department of Material Physics, Complutense University of Madrid, 28040 Madrid, Spain*
[4] *Donostia International Physics Center, 20018 Donostia, Spain*

$CuFe_2O_4$ copper ferrite has been the subject of multiple studies in the last three decades due to both theoretical issues and technological applications in a wide variety of areas, such as hyperthermia[1], water treatment and catalysis. This interest is supported mainly by its magnetic properties and its dependence on the distribution of the constituent cations in the tetrahedral (A) and octahedral (B) sites of the spinel structure. Thus, the cation distribution can be written by $(Cu_{\delta}^{2+}\ Fe_{1-\delta}^{3+})^{A}[Cu_{1-\delta}^{2+}\ Fe_{1+\delta}^{3+}]^{B}$ where δ is the inversion parameter, and for this ferrite type full inversion corresponds at δ=0. Moreover, copper ferrite can be found in a body-centered tetragonal (space group I41/amd) and cubic close-packed (space group Fd3m) structures. As the tetragonal structure is strongly related to the Jahn–Teller[2] (JT) effect in the octahedral site, the gradual migration of $Cu^{2+}$ from B to A sites in a fully inverse spinel when the temperature increases give rise to the symmetry transition from tetragonal to cubic. This cation migration can be also induced by a mechanical deformation process, like high energy ball milling.

In this work, a stoichiometric copper ferrite was synthesized by the ceramic route. The magnetic properties of this material were tuned via cation redistribution through several processing routes. Initially, heat treatments were carried out at 600 and 1000 ºC to stabilize the cubic and tetragonal symmetry, respectively. The change of structural and magnetic properties due to the cation redistribution in samples with these two structures was also investigated in samples milled up to 40 hours in a high energy planetary mill. The structure and the inversion parameter were determined by the Rietveld refinement of their powder diffraction patterns (figure). These microstructural parameters were correlated with magnetic hysteresis loops recorded on a standard superconducting quantum interference device magnetometer with the maximum applied field of 5 T. The values of the saturation magnetization in the fully inverted samples showed a dominant antiferromagnetic interaction between the cations at the A and B sites, following the standard superexchange theory. On the other hand, a decrease in saturation magnetization is observed in the milled samples. This decrease could be explained by the partial inversion present in these samples, which would result in an exchange coupling $J_{BB}$ that could be AFM or FM depending on the bond angle cation-anion-cation.

## Acknowledgements

This research has been funded by Ministerio de Ciencia, Innovación y Universidades of Spain by means of the PID2021-123112OB-C21 and AFORMAR (PID2019-109334RB) projects. The support of the grants from the Community of Madrid number S2018 / NMT-4381-MAT4.0-CM is also recognized.

## References

[1] Jessyamma Kurian, B.B. Lahiri, M. Jacob Mathew, John Philip. Journal of Magnetism and Magnetic Materials 538 (2021) 168233

[2] M. V. Veidis, G. H. Schreiber T. E. Gough, Gus J. Palenik, Journal of the American Chemical Society 1 91:7 March 26, 1969

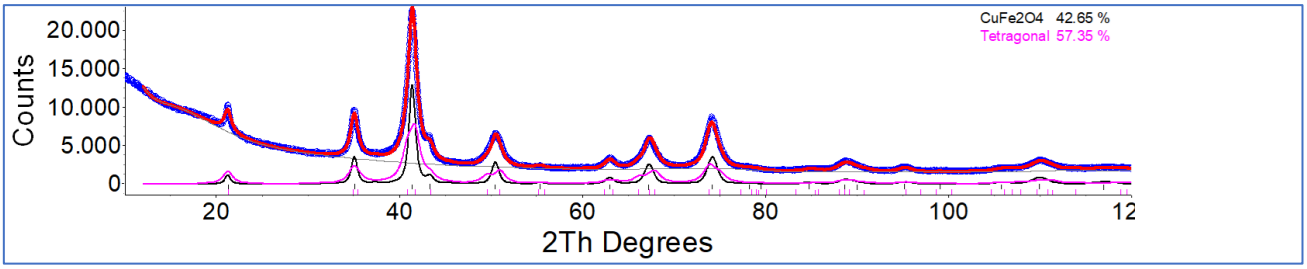



Figure: Rietveld analysis of Cu ferrite synthesized as starting cubic and its evolution in both cubic and tetragonal structure after 10 h milling (42,65% cubic and 57,35% tetragonal)

# Photothermal effects in iron oxide nanoparticles with different oxidation states.

*Daniel Arranz[1,2*], Rosa Weigand [2], Patricia de la Presa[1,3]*

*[1] Instituto de Magnetismo Aplicado Salvador Velayos (UCM-ADIF-CSIC), A6 km. 22.5 Las Rozas (Madrid). [2] Dpto. de Óptica, [3] Dpto. de Física de Materiales, Facultad de Ciencias Físicas, Universidad Complutense de Madrid, Avda. Complutense s/n, 28040 Madrid, Spain*

*daniarra@ucm.es

Photothermal effects on iron oxide nanoparticles (IONP) under laser irradiation are gaining the interest of the scientific community due to their potential biomedical application in cancer treatment [1]. One of this study's main challenges is knowing exactly the nanoparticle's heating mechanism to release heat. It is known that different intrinsic factors that could affect the heating of different samples are the size and oxidation state ($\gamma$-$Fe_2O_3$ or $Fe_3O_4$ for instance).

This work presents a facile hydrothermal method that allows the variation of the oxidation state of the IONP while maintaining the size constant. The synthesis consists of obtaining a $Fe_3O_4$ colloid, using a steel autoclave [2]. After obtaining $Fe_3O_4$ of d = 30 nm, the colloid is introduced again at the autoclave at different times. This second thermal treatment induces changes in the oxidation state, as can be observed with the naked eye by the change in color of the colloid, which changes from black to red-brown. The optimal time and oxygen volume in the autoclave space are key parameters and will be discussed. Typical IONP characterization has been done such as SQUID magnetization (figure 1), XRD, or HRTEM for structural characterization.

Additionally, photothermal studies have been performed in the different colloids with continuous lasers at 800 nm (first biological window) and 1053 nm (second biological window) [3]. The heating efficiency of the IONPs under laser irradiation is discussed as a function of Fe in the oxidation states.

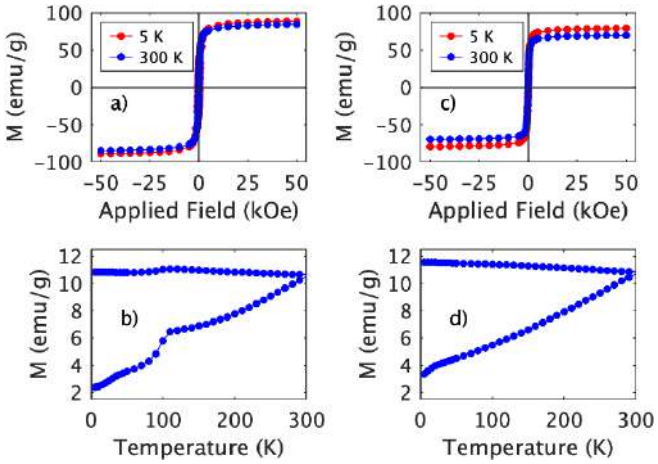



Figure 1: Hysteresis cycle and ZFC-FC of different samples. On the left, the typical hysteresis cycles for 5 K (red) and 300 K (blue), and ZFC-FC (100 Oe) curves for magnetite are shown. On the right, the hysteresis cycle and ZFC-FC curves for the same sample, but with a 6 h of second thermal treatment.

## Acknowledgments

This work was supported by Projects PID2021-123112OB-C21 (Ministerio de Ciencia, Innovación y Universidades) and S2018/NMT-4321. We also acknowledge the Sistema Nacional de Garantía Juvenil for the contract through the Comunidad Autónoma de Madrid (PEJ-2020-AI/IND-17711).

## References

[1] Estelrich J, Busquets MA. *Molecules*. **23(7),** (2018):1567.
[2] S. Ge, X. Shi, K. Sun, C. Li, C. Uher, J. R. Baker, M. M. Banaszak Holl, and B. G. Orr, *The Journal of Physical Chemistry* C **113**, (2009) 13593.
[3] Arranz D, Weigand R, de la Presa P. *Nanomaterials*. **13(3),** (2023) 450.

# Magneto-Amplification Of Visible-Light-Driven Total Photodegradation Of Noxious Dye Molecules By The Multifunctional $TiO_2$@$Fe_xO_y$ Core-Shell Nanoheterostructure

*Daniel Gherca, Adrian Iulian Borhan, Daniel Dumitru Herea, Marian Grigoras, George Stoian, Gabriel Ababei, Horia Chiriac, Nicoleta Lupu*

*National Institute of Research and Development for Technical Physics, 47 Mangeron Boulevard, 700050 Iasi, Romania*

*dgherca@phys-iasi.ro

Environmental pollution topics, particularly the presence of dangerous chemicals in water resources can have serious negative health effects [1], and thus is an urgent need for innovative and advanced treatment methods to address this pressing issues [2,3]. Among the techniques that can be used to decontaminate water and wastewater from pollutants, photocatalysis is a potentially promising approach that is based on the generation of reactive species upon UV-visible irradiation of $TiO_2$ photocatalyst [4]. However, the low photocatalytic activity of the $TiO_2$, particularly under visible illumination, limits its commercial application, therefore, research studies have been focused on developing multifunctional $TiO_2$ based core-shell nanoheterostructures with improved photocatalytic performance and light-response range [5,7]. Although the design of magnetic recoverable photocatalysts is widely analysed in the literature, the influence of external magnetic fields on photocatalysis has been scarcely addressed [5]. In particular, magnetic field effects on the photocatalytic response is usually ascribed to the contribution of Lorentz forces acting on the chemical radicals employing magnetic or non-magnetic (para or diamagnetic) photocatalysts [3,6]. Therefore, an in-depth study about the control of the charge separation in heterojunction photocatalysts under external magnetic field is currently beyond the state-of-art of the photocatalytic field. The main aim of the present study consist in the development of multifunctional $TiO_2$@$Fe_xO_y$ core-shell nanoheterostructures based on the synergistic combination of different type magnetic semiconductors (i.e. γ $Fe_2O_3$ ) to generate innovative titania-based nanocomposites with enhanced visible light response that can be tested in photodegradation processes to acquire of fundamental knowledge around photocatalysis under direct magnetic field. Our synthetic strategy highlights that the strong alkaline precipitation of $Fe_xO_y$ nanocompartmentalized $TiO_2$ material leading to core-shell heteronanostructure. Morphological, structural, magnetic and optical, properties of the as-prepared samples were in-depth investigated using a series of complementary characterization techniques, such as powder X-Ray Diffraction analysis (PXRD), Ultra-High Resolution Transmission Electron Microscopy (UHR-TEM), Field-Emission Scanning Electron Microscopy (FE-SEM), hard and soft X-ray absorption spectroscopy (XAS) in both total electron yield (TEY) and fluorescence yield (TFY), as well as VSM analysis. The visible-light-driven photodegradation tests were runned in an variable gap magnet system used to induce strong local magnetic flux. A dramatically improved photodegradation degree promoted by local magnetic field effects was observed for $TiO_2$@$Fe_xO_y$ core-shell nanoheterostructures. The present study offers an fesable opportunity to rationally design photocatalysts nanomaterials with magnetically enhanced photodegradation performance for water treatment applications.

**Acknowledgements**

This work was "Funded by the European Union - NextGenerationEU", Call # PNRR-III-C9-2022–I9, Contract No. 760013/27.01.2023.

## References

[1] P. Chowdhary, R. N. Bharagava, S. Mishra, N. Khan, *Environ. Concerns Sustain. Dev* **1** (2020) 235–256.
[2] M. Zhang, W. Chen, W. Choi, J. Yu, Y. Deng, X. Xie, Z. Lin, *ACS Sustainable Chem. Eng.* **10** (2022), 4741.
[3] C. Li, G. Liao, B. Fang, *Chem Catal.* **2** (2022) 238.
[4] Q. Wang, J. Cai, G. V. Biesold-McGee, J. Huang, Y. H. Ng, H. Sun, J. Wang, Y. Lai, Z. Lin, *Nano Energy* **78** (2020), 105313.
[5] H. Dong, G. Zeng, L. Tang, C. Fan, C. Zhang, X. He, Y. He, *Water Res.* **79** (2015) 128–146.
[6] W. Sittipol, C. Sronsri, K. U-yen, *J. Cleaner Prod.* **325** (2021) 129284.
[7] D.-E. Lee, V. Devthade, S. Moru, W.-K. Jo, S. Tonda, *J. Alloys Compd.* **902** (2022) 163612.

# Synthesis of FeO@Fe3O4@MgO@Fe3O4 onion-like magnetic nanoparticles

*Jorge M. Nuñez [1,2,3,5,6], Víctor Leboránn[4], Francisco Rivadulla[4], Gerardo. F. Goya,[5,6], Roberto D. Zysler[1,2], Elin L. Winkler[1,2], Myriam H. Aguirre[3,5]

[1] Resonancias Magnéticas, Centro Atómico Bariloche e Instituto de Nanociencia y Nanotecnología, CNEA/CONICET, S. C. Bariloche 8400, Río Negro, Argentina.

[2]Instituto Balseiro (UNCuyo, CNEA), Av. Bustillo 9500, S.C. de Bariloche 8400, Río Negro, Argentina.

[3]Instituto de Nanociencias y Materiales de Aragón-CSIC-Universidad de Zaragoza, C/ Mariano Esquillor s/n, Zaragoza 50018, Zaragoza, Spain

[4]Centro de Investigación en Química Biolóxica e Materiais Moleculares (CIQUS), Dpto. de Química Física, Universidad de Santiago de Compostela, 15782 Santiago de Compostela, Spain

[5]Laboratorio de Microscopías Avanzadas, Universidad de Zaragoza, Mariano Esquillor s/n, Zaragoza 50018, Zaragoza, Spain

[6]Dept. Física de la Materia Condensada, Universidad de Zaragoza, C/ Mariano Esquillor s/n, Zaragoza 50018, Zaragoza, Spain

*Corresponding author: jorge.nunez@ib.edu.ar

The Core/shell (CS) architecture make it possible to combine in the same nanoparticle (NP) different materials, increasing the degrees of freedom to design and manufacture new systems [1]. Recent studies in CS NPs show that, by systematically varying the shell composition, it is possible to fine-tune the magnetic and electrical transport properties of these systems [2]. In addition, new properties are observed in CS bimagnetic materials due the exchange interaction at the interface [3], as exchange-bias [4].Since the switching field in tunneling magnetoresistance (TMR) devices is determined by the anisoptropy of the magnetic materials [5], the design and fabrication of more complex and higher quality NPs is a key factor to develop new multifunctional nanoparticles for advanced applications. In this work we present that high quality onion-like NPs can be grown by adapting the seed-mediated growth method proposed by Sun et al [6].

FeO@Fe3O4@MgO@Fe3O4 monodispersed NPs were synthesized by thermal decomposition. Their structure and morphology were characterized by different techniques of transmission electron microscopy (TEM) and powder X-ray diffraction. By analyzing TEM images, we obtain a monodisperse size distribution with mean size of (24±4) nm. CSS structure can be confirmed by observing high angular annular dark field scanning transmission electron microscopy (HAADF STEM) images, that shown a dark annular contrast due to the presence of MgO in the inner shell, and from the elemental mapping performed by electron energy loss spectroscopy (EELS) the structure is corroborated. The magnetic properties were studied from magnetization measurements as a function of the applied field (MvsH) and temperature, in a range of ±2.5T and 5K-380K. In field cooling MvsH curves the presence of a bias field was observed below 125K. Magneto transport measurements were performed in devices of self-assembled NPs in a range of ±1T and 90K-300K. The results are analyzed in terms of the magnetic coupling between the soft Fe3O4 and hard FeO@Fe3O4 magnetic phases.

**References**

[1] A. López-Ortega, M. Estrader, G. Salazar-Alvarez, A.G. Roca, J. Nogués, J. Phys. Rep., 553 (2015).
[2] F. Fabris, E. Lima, C. Quinteros, L. Neñer, M. Granada, M. Sirena, R. Zysler, H. Troiani, V. Leborán, F. Rivadulla y E. Winkler, Physical Review Applied. 11 (2019).
[3] W.H. Meiklejohn, C.P. Bean, Phys. Rev., 102 (1956).
[4] J. Nogués, J. Sort, V. Langlais, V. Skumryev, S. Suriñach, J. Muñoz, M. Baró, Phys. Rep., 422 (2005).
[5]M. A. López-Quintela, L. E. Hueso, J. Rivas, F. Rivadulla, Nanotechnology 14, 212 (2003).
[6] S. Sun y H. Zeng, J. Am. Chem. 124 (2002).

# Crystal/Magnetic Structure, Nucleation Mechanism and Crystallization Dynamics of Spinel Ferrite Nanoparticles

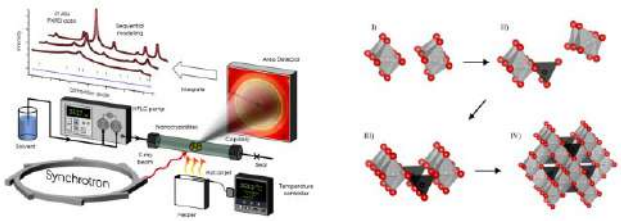
*Henrik Lyder Andersen,[1*] Cecilia Granados-Miralles,[2] Kirsten M. Ø. Jensen,[3] Matilde Saura-Múzquiz[1] and Mogens Christensen[4]*

[1] *Dept. of Materials Physics, Universidad Complutense de Madrid, Madrid, Spain*
[2] *Dept. de Electrocerámica, Instituto de Cerámica y Vidrio (ICV), CSIC, Madrid E-28049, Spain*
[3] *Dept. of Chemistry and Nanoscience Center, University of Copenhagen, Copenhagen, Denmark*
[4] *Dept. of Chemistry and iNANO, Aarhus University, Aarhus, Denmark*
*henrikla@ucm.es

The magnetic spinel-structured ferrite compounds ($M$Fe$_2$O$_4$, $M$=Mn, Fe, Co, Ni, Zn, *etc*.) are important materials, as their low cost, stability and versatile magnetic properties make them ideal for a variety of applications.[1] The performance of the spinel ferrite materials is determined by the complex interplay between their crystal-, magnetic-, and micro-structure of the nanoparticles.[2] Therefore, understanding their structure-property relationship and being able to target specific structural characteristics is key for optimizing the materials' performance.

The advent of high-flux synchrotron X-ray sources and development of custom-built reactors have facilitated the collection of time-resolved scattering data during nanoparticle nucleation and growth.[3] This has dramatically reduced the time needed to cover parameters space as well as provided unprecedented insight into reaction mechanisms all the way from pre-crystalline clusters in the precursor to their final crystalline states.[4] By studying the chemical reactions *in situ* we can observe the fundamental chemistry that takes place between inorganic species during nucleation and growth of nanoparticles, an aspect that is still relatively poorly understood compared to reactions in organic chemistry.

In the present study, the hydrothermal formation/growth mechanisms and crystal/magnetic structures of $MnFe_2O_4$, $CoFe_2O_4$, $NiFe_2O_4$ and $ZnFe_2O_4$ nanocrystallites prepared from co-precipitated transition metal (TM) hydroxide precursors treated at sub-, near- and super-critical conditions have been studied by a combination of *in situ* X-ray total scattering with pair distribution function (PDF) analysis, *in situ* synchrotron powder X-ray diffraction (PXRD) with Rietveld analysis, and joint structural modelling of *ex situ* PXRD and neutron powder diffraction (NPD) data. It is demonstrated how the crystal structure and crystallite sizes of the obtained nanoparticles can be controlled by varying the spinel ferrite composition (type of divalent cation, $M$) and/or through modifications to the reaction temperature and precursor composition/preparation route, thereby providing handles to tune the intrinsic and extrinsic magnetic properties.




**Acknowledgements**
The authors gratefully acknowledge the beamtime granted at MaxLab, Sweden; PETRA III, Germany; and Swiss Spallation Neutron Source (SINQ), Paul Scherrer Institute (PSI), Switzerland. Henrik Lyder Andersen gratefully acknowledges the financial support from the The Spanish Ministry of Universities (Ministerio de Universidades) and the European Union—NextGenerationEU through a Maria Zambrano—attraction of international talent fellowship grant.

**References**

[1] J. M. D. Coey, *J. Magn. Magn. Mater.*, 2002, **248**, 441-456.
[2] H. L. Andersen, *et al.*, *Nanoscale*, 2018, **10**, 14902-14914.
[3] H. L. Andersen, *et al.*, *J. Appl. Crystallogr.*, 2018, **51**, 526-540.
[4] E. D. Bøjesen, *et al.*, *CrystEngComm*, 2016, **18**, 8332-8353.

# Synthesis and characterization of Au-$Fe_3O_4$ nanohybrids

*Tatiana Smoliarova*, Marina Spasova, Michael Farle and Ulf Wiedwald*

*Faculty of Physics, University of Duisburg-Essen, Duisburg, 47057, Germany*

*tatiana.smoliarova@uni-due.de

Magnetic iron oxide nanoparticles ($Fe_3O_4$ NPs) are under extensive investigation for selective drug delivery in cancer treatment [1-2]. Covered with the hydrophilic, biocompatible and non-toxic polymer polyvinylpyrrolidone (PVP) $Fe_3O_4$ NPs are used for targeted drug delivery [3]. Furthermore, gold nanoparticles (Au NPs) decorating the surface $Fe_3O_4$ NPs of are used for controlled release of a drug by photothermal stimulation mediated by localized surface plasmonic resonance (LSPR). LSPR allows the Au NPs, as hot spots, efficiently trigger a drug release in the PVP-$Fe_3O_4$ composites under controlled conditions [4].

Herein, we report on the synthesis of Au-$Fe_3O_4$ nanohybrids via a chemical precipitation method with a chemisorbed Au NPs to the octahedral PVP-$Fe_3O_4$ NPs surface. The results of transmission electron microscopy (TEM) are presented in Fig. (a-c) and show Au NPs are attached to PVP covered $Fe_3O_4$ NPs at a number ratio of 25:1 (Fig. (e)). Fig. (d) shows size distribution of $Fe_3O_4$ NPs. Absorbance spectra of Au, $Fe_3O_4$, and Au- $Fe_3O_4$ NPs are shown in Fig. (f). Au NPs (red line) have the typical behavior with the plasmon peak at 520 nm. In comparison with the absorbance spectrum of $Fe_3O_4$ NPs (black line), the spectrum for Au $Fe_3O_4$ NPs (green line) demonstrates the shift of the light plasmon peak to 536 nm, which is an effect of Au-$Fe_3O_4$ nanohybrids.

## References

[1] Efremova, M. V. et al. Beilstein J. Nanotechnol 9, 2684 (2018).
[2] Efremova, M.V. et al. Sci Rep 8, 11295 (2018).
[3] Rose, P. Arsula, et al. Technology in cancer research & treatment 12(5), 463 (2013).
[4] Taheri-Ledari, Reza, et al. Communications Biology 5(1), 995 (2022).

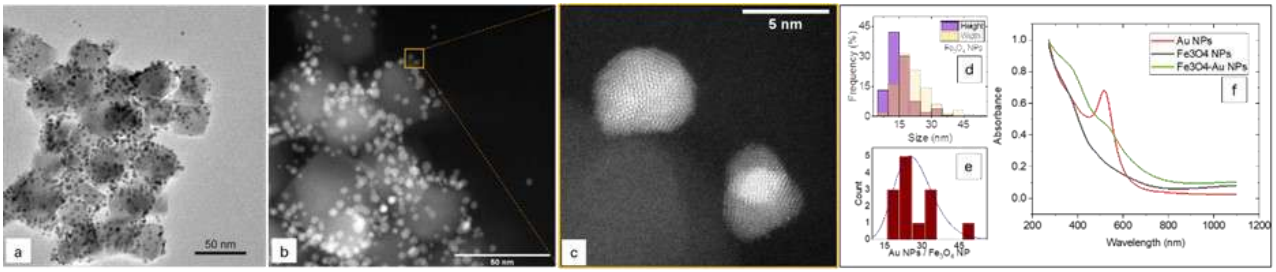



Figure: TEM (a) and HAADF (b-c) images of synthesized Au-Fe3O4 nanohybrids; size distribution of Fe3O4 NPs (d) and number of Au NPs per Fe3O4 NP (e); absorbance spectra of the synthesized NPs (f)

# Particle Size-Dependent Magnetic Behavior of $LaFeO_3$: Spin Canting and Spin Glass Exchange

*Dhoha R. Alshalawi [1]* José M. Alonso,[1,2] Angel R. Landa-Cánovas,[2] Patricia de la Presa.[1,3]*

*[1] Institute of Applied Magnetism, UCM-ADFI-CSIC, A6 22,500 km, 28230 Las Rozas, Spain*
*[2] Institute of Material Science of Madrid, CSIC, 28049 Madrid, Spain*
*[3] Department of Materials Physics, Complutense University of Madrid, 28040 Madrid, Spain*
*E-mail: dhohadho@ucm.es

This research explores the characteristics of $LaFeO_3$ at varying particle sizes. This study focuses on nanoparticle samples synthesized through the sol-gel method at various annealing temperatures 600 °C, 700 °C, 800 °C, and 900 °C. Including the Bulk sample has been synthesized at 1300 °C using solid-state method. A range of techniques were employed, including Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES), X-ray diffraction (XRD), and Thermogravimetric analysis (TGA). The crystal structure and morphology of the samples were determined from Transmission Electron Microscopy (TEM), Selected Area Electron Diffraction (SAED), and High-Resolution Electron Microscopy (HRTEM) images. The magnetic measurement was investigated at temperatures 300 K and 5 K by Zero Field Cooled and Field Cooled (ZFC-FC) curves, Field-cooled (FC) hysteresis cycles, and ac/dc magnetometry in different frequencies. The findings indicate the Bulk sample has antiferromagnetic canting spin due to the display of numerous twins in HRTEM. For nanoparticle samples, the spin canting effect ceases to exist for particles with a size of d ~125 nm. As nanoparticles with d < 60 nm exhibit a noteworthy exchange effect bias, which results from their core-shell phases. As the relation surface/volume (S/V) increases as particle size decreases, a strong magnetized disorder phase present on the surface, as seen previously [1]. The shell portion of the particles displays spin glass behavior, while the core component exhibits *G*-type antiferromagnetic order. The HRTEM images verified that the crystal structure of the nanoparticles is free of the core-shell, amorphous phase, or twins, showing a flawless, well-organized structure.

**Acknowledgements**

The authors thank the Agregaduría Cultural de Arabia Saudí, Spain (project code 4157592) for project funding.

**References**

[1] H. Aliyu, J. Alonso, P. de la Presa, W. Pottker, B. Ita, M. Garcia-Hernández, A. Hernando, *Chemistry of Materials*, **30** (2018) 7138-7145.

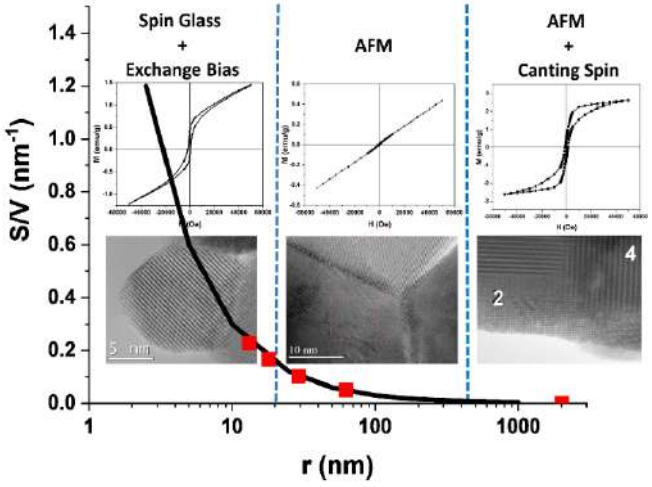



Figure 1: This is an illustration of the correlation between magnetic characteristics, hysteresis curves, particle morphology observed through HRTEM images, and the (S/V) ratio represented by the black curve and red data points.

# Seed mediated polyol synthesis of $CoFe_2O_4/Ni_xCo_{1-x}O$ exchange biased magnetic nanostructures

*Miran Baričić[1]*, Faris Flaifil[1], David Hrabovsky[2], Souad Ammar[1], Davide Peddis[3]*

[1] *ITODYS, UMR-CNRS 7086, Université Paris Cité, 15 Rue Jean Antoine de Baif, 75205 Paris, France*

[2] *IMPMC, UMR-CNRS 7590, Sorbonne Université, 6 Place Jussieu, 75005 Paris, France*

[3] *DCCI, Università degli Studi di Genova, Via Dodecaneso, 31, 16146 Genova, Italy*

*miran.baricic@u-paris.fr

The polyol method is a scalable and green wet chemistry synthesis for the production of nanostructured inorganic materials of any kind, including magnetic materials[1]. In the proper conditions, the synthesis is able to produce small nanoparticles (NP) aggregates with epitaxial relation, both in single and composite materials[2]–[4], with this feature being of paramount importance in the design of exchange coupled magnetic nanostructures. Even though CoO has already been exploited in this kind of studies, and despite its quite high Néel temperature ($T_N$ = 293 K[5]) – like many nanostructured materials – its $T_N$ meets a remarkable reduction when brought to the nanoscale (as we observed in our yet not published results on CoO NPs, but was already observed in other materials[6], [7]) making the exchange bias and its related effects impossible to observe at room temperature. However, NiO has a much higher $T_N$ than CoO (525 vs 291 K[5]), and the doping of CoO with $Ni^{2+}$ ions has already shown to be an effective strategy to increase its $T_N$[5].

Here, we present an exploitation of this strategy to synthesize polyol made magnetically contrasted $Ni_xCo_{1-x}O/CoFe_2O_4$ ($0.0 < x < 0.4$) to study exchange related magnetic phenomena and basic magnetic properties, observing the increase. Attention is dedicated to the effect of the interfacial coupling to the blocking temperature of the spinel material, to the structural features of the interface and to the effect of the synthesis parameters (such as water quantity, temperature and others) on the morphological, structural and magnetic properties of the resulting material.

**Acknowledgements**

…

**References**

[1] F. Fiévet *et al.*, *Chem. Soc. Rev.*, **2018** , *47*, 5187–5233,
[2] T. Gaudisson *et al.*, *CrystEngComm*, **2021** , *23*, 1756–1764,
[3] T. Gaudisson *et al.*, *CrystEngComm*, **2016** , *18*, 3799–3807,
[4] N. Flores-Martinez *et al.*, *Part. Part. Syst. Charact.*, **2018** , *35*, 1–8,
[5] A. Mandziak *et al.*, *Sci. Rep.*, **2019** , 1–8,
[6] B. Pan, X. Meng, Y. Xia, en H. Lu, *Phys. Chem. Chem. Phys.*, **2020** , *22*, 5735–5739,
[7] Z. X. Tang, C. M. Sorensen, K. J. Klabunde, en G. C. Hadjipanays, *Phys. Rev. Lett.*, **1991** , *67*, 3602–3605.

# Magnetic Relaxation and MR Measurements of Zn-doped $MgFe_2O_4$ Nanoparticles

*Noboru Kataoka [1], Sota Hamada [1], Akihito Usui [2], Yuko Ichiyanagi [1]**

[1] *Department of Phyics, Graduate School of Engineering Science, Yokohama National University, 79-5 Tokiwadai, Hodogaya, Yokohama, 240-8501 Japan*

[2] *Institution Graduate School of Medicine, Tohoku University, 2-1 Seoryo-cho, Aoba-ku, Sendai, 980-8575 Japan*

*yuko@ynu.ac.jp

Since these several years, we have reported on the biomedical applications of magnetic nanoparticles [1]. Magnetic resonance imaging (MRI) is currently one of the most popular diagnostic techniques. The clinical use of gadolinium (Gd) complexes contrast media began in 1988; Gd was used because of its high degree of relaxation. However, the use of chelate complex Gd contrast agents has been curtailed in recent years due to their safety concerns. Therefore, there is an urgent need to develop chemically stable and biocompatible contrast agents.

In this study, biocompatible $Mg_{0.8}Zn_{0.2}Fe_2O_4$ nanoparticles encapsulated in amorphous $SiO_2$ with various particle sizes were prepared, and magnetic relaxation phenomena were investigated. Magnetic resonance (MR) effect and the relationship between magnetic properties was also discussed.

Magnetization measurements were performed and superparamagnetic behaviour was observed. AC magnetic susceptibilities were measured to analyze magnetic relaxation phenomena. $T_1$ and $T_2$ relaxation curves were measured in magnetic field of 0.3 T, resonance frequency of 13 MHz of MRI system. The $T_2$ relaxation signals were carefully measured thrice while the echo time ($TE$) was increased. A longer echo time results in a lower signal-to-noise (S/N) ratio. As a result, compared to agarose as the background, the obtained particles exhibited a significant $T_2$ shortening effect for all the samples (Fig.1). Figure 2 summarizes the relaxation rate $R_2$, taking the inverse of $T_2$. These samples exhibited large $R_2$ values compared to $\gamma$-$Fe_2O_3$, which are conventional materials for MRI. The magnetic moments of the magnetic nanoparticles cause dipole interactions with protons, which may result in energy transfer and promote $T_2$ relaxation.

On the other hand, these Zn-doped Mg-ferrite nanoparticles exhibited a significant heat dissipation effect in AC magnetic fields and has been confirmed to be effective in hyperthermia treatment of cancer cells [3]. These particles greatly enhanced both $T_2$ relaxation in MR measurement and hyperthermia effect, and can be expected to be theranostic agent.

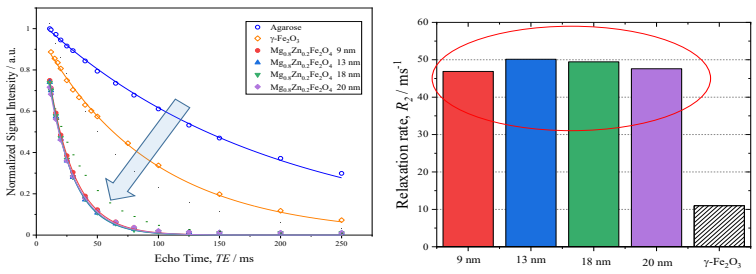
## Acknowledgements

JST-Mirai Program No.JPMJMI17D7, Grant-in-Aid for Science Research No.25286041 and Watanabe foundation No. 280118.

## References

[1] T. Kondo, K. Mori, M. Hachisu, T. Yamazaki, D. Okamoto, M. Watanabe, K. Gonda, H. Tada, Y. Hamada, M. Takano, N. Ohuchi, and Y. Ichiyanagi, *J. Appl. Phys.*, **117** (2015) 17D157.

[2] S. Hamada, K. Aoki, K. Kodama, K. Nashimoto, Y. Ichiyanag, *J. Magn. Magn. Mater.* **559** (2022) 169536.




Figures: $T_2$ relaxation curves of $Mg_{0.8}Zn_{0.2}Fe_2O_4$ nanoparticles (left) and summary of the relaxation rate $R_2$ (right).

# Photothermal Effects in $MFe_2O_4$ (M= Fe, Co, Ni, Cu, Zn) Nanoparticles

*Luis García García-Arisco[1*], Daniel Arranz[1,2], Manuel Horcajo Fernández[1], Patricia de la Presa[1]*

*[1] Instituto de Magnetismo Aplicado Salvador Velayos (UCM-ADIF-CSIC), A6 km. 22.5 Las Rozas (Madrid).*

*[2] Dpto. de Óptica, Facultad de Ciencias Físicas, Universidad Complutense de Madrid, Avda. Complutense s/n, 28040 Madrid, Spain*

*luisga19@ucm.es

The photothermal effects on iron oxide nanoparticles under laser irradiation have awaked the interest of the scientific community because the interest of understanding the physical mechanism behind this effect [1]. In this work, the heating efficiency of different ferrite nanoparticles $MFe_2O_4$ (M= Fe, Co, Ni, Cu, Zn) is studied in order to understand the role played by the cation M in the heating efficiency.

All the samples have been synthesized by a facile hydrothermal method, briefly, the molar ratio of $MCl_2$ was dissolved in water and then, $NH_3 \cdot H_2O$ was suddenly added to cause a fast precipitation. This mixture was then autoclaved in a sealed vessel at 134 °C for 3h. Thereafter, the samples were washed with water and centrifuged several times. The structural characterization has been done by XRD using a Co source ($\lambda = 1.79$ Å) and HRTEM in a JEOL JEM100 (100 kV). The XRD results show that only $Fe_3O_4$, $CuFe_2O_4$ and $ZnFe_2O_4$ are single phases, whereas the other ones show impurity phases that can be indexed with their corresponding cationic oxides. The particle sizes determined by Scherrer formula are 33 nm for $Fe_3O_4$, 15 nm $ZnFe_2O_4$ and 7 nm $CuFe_2O_4$, these values match pretty well the with the sizes determined by HRTEM. The magnetic characterizations show almost superparamagnetic behaviour at room temperatures for all the samples, except for $CoFe_2O_4$, which shows a coercive field around 560 Oe.

The Specific Absorption Rate (SAR) has been investigated in the different colloids with continuous laser irradiation at 808 nm (first biological window) and the procedures described in Ref. [2]**.** As can be seen in Fig. 1, the heating efficiency is around 600 to 800 W/g for M = Mn, Fe, Ni, Zn, it raises to ~1,000 W/g for M = Co and decreases to ~300 W/g for M = Cu. It is expected that particle size plays an additional effect on the photothermal efficiency that will be discussed in the present work.

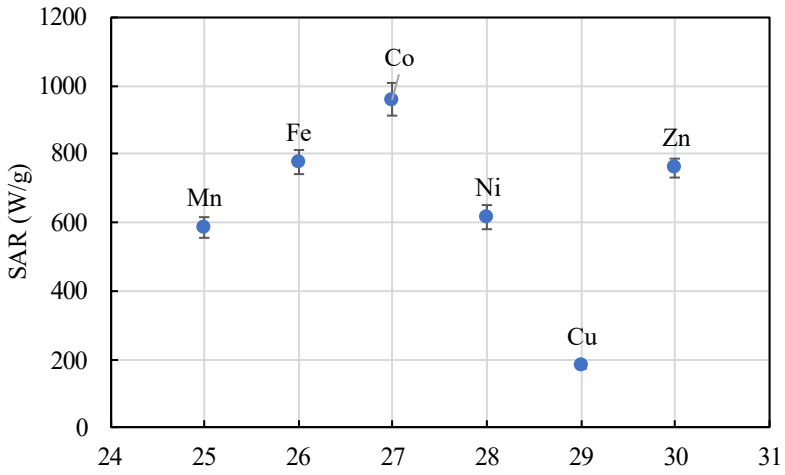



*Figure: SAR values as a function of the atomic numbers of $MFe_2O_4$.*

## Acknowledgments

This work was supported by Projects PID2021-123112OB-C21. We also acknowledge the Sistema Nacional de Garantía Juvenil for the contract through the Comunidad Autónoma de Madrid (PEJ-2020-AI/IND-17711).

## References

[1] Lozano-Pedraza, C.; Plaza-Mayoral, E.; Espinosa, A.; Sot, B.; Serrano, A.; Salas, G.; Blanco-Andujar, C.; Cotin, G.; Felder-Flesch, D.; Begin-Colin, S.; et al. Assessing the parameters modulating optical losses of iron oxide nanoparticles under near infrared irradiation. Nanoscale Adv. 2021, 3, 6490–6502.

[2] Arranz D, Weigand R, de la Presa P. Towards the Standardization of Photothermal Measurements of Iron Oxide Nanoparticles in Two Biological Windows. Nanomaterials. 2023; 13(3):450.

# Heusler Micro- and Nanowires

*L. Galdun[1*], M. Hennel[1,2], M. Varga[1,2], V. Huntosova[3], P. Szabo[4], V. Vega[5], V. M. Prida[5], R. Varga[1]*

*[1]CPM-TIP, UPJS, Tr. SNP1, 04 001 Kosice, Slovakia*

*[2]Department of Condensed Matter Physics, UPJS, Park Angelinum 9, 041 54 Kosice*

*[3]CIB-TIP, UPJS, Jesenna 5, 04 101, Kosice, Slovakia*

*[4]Institute of Experimental Physics, Slovak Academy of Sciences, 04001 Kosice, Slovakia*

*[5]Departamento de Física, Facultad de Ciencias, Universidad de Oviedo, C/ Federico Garcia Lorca nº 18, 33007-Oviedo, Asturias, Spain*

*ladislav.galdun@upjs.sk

Heusler alloys are known for various properties related to their chemical composition, stoichiometry or structure. Different physical phenomena such as spin polarization, magnetocaloric effect, shape memory effect or superconductivity have been successfully observed in the case of bulk samples or ribbons. New research, especially at the nanoscale [1], revealed that their properties should be preserved despite the reduction of their size. Thanks to this, new possibilities have been opened up in various application areas dealing with spintronics, magnetocalorics, sensors or actuators [2].

While the fabrication of Heusler alloys in the form of glass-coated microwires is already known [2], producing homogeneous Heusler nanoparticles is a relatively new topic [1]. The electrochemical deposition of nanowires in a nanoporous aluminium oxide membrane offers new possibilities in the production of multi-element alloys such as Heusler alloys ($X_2YZ$). Using the mentioned approach, it is possible to prepare a large amount of homogeneous nanowires ($10^9$ nanowires/cm$^2$) within a few minutes.

One of the most significant advantages of Heusler alloys is their multifunctionality and the possibility of tuning their features by simple adjustments in their chemical composition. Therefore, with an electrochemical deposition, it is possible to create functional Heusler nanowires, which can be tuned based on the requirements of the selected applications like actuators with sensoric properties or materials for nanorobotics or bioapplications.

**Acknowledgements**

This work was supported by Slovak Grant Agency VEGA 1/0053/19, VEGA 1/0180/23, Slovak Grant Agency grant number APVV-16-0079, VVGS-PF-2020–1420 and VVGS-2022-2408.

**References**

[1] M. Varga, L. Galdun, P. Diko, K. Saksl, R. Varga, *J. Alloys Compd.* **944** (2023) 169196.
[2] L. Galdun, R. Vidyasagar, M. Hennel, R. Varga, *J. Phys. D* **55** (2022) 045303.

Figure: Nanowire arrays prepared by electrochemical deposition

# Interactions and grain-interface effects on the magnetoresistance of single cobalt nanowires during magnetization reversal

*Lukas Ingenbleek[1], Julia Andrews[1], Mahmoud Deeb[1], Thomas Heinzel[1], Mihai Cerchez[1]*, Shibesh Dutta[2], Anshul Gupta[2], Shreya Kundu[2], Zsolt Tőkei[2], Florin Ciubotaru[2], Christoph Adelmann[2]*

*[1]Condensed matter physics laboratory, Heinrich-Heine-University, 40204 Düsseldorf, Germany*

*[2]Imec, 3001 Leuven, Belgium*

*Mihai.Cerchez@HHU.de

Cobalt nanowires gather continuous interest due to downscaling of electronics and possible use in various applications from liners to spintronic devices. At few-nanometer scale, the resistance and magnetoresistance (MR) of Co wires are dominated by surface and grain boundaries effects that overcome the bulk transport properties. Here we present measurements on single cobalt wires produced by self-aligned ion beam etching (Figs. 1). The MR is measured at low temperature (50 mK – 10 K) for various orientations of the magnetic field (Figs. 2). Both positive and negative fluctuations in MR, associated respectively with intergrain tunneling magnetoresistance (TMR) [1] and anisotropic magnetoresistance (AMR) [2] are explained in terms of interplay between magnetic interactions and anisotropies during magnetic reversal. Neither of the

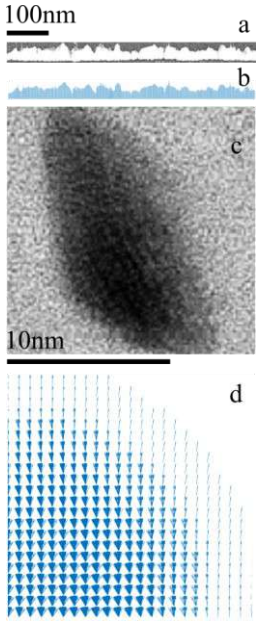



Fig. 1 a. and c. Longitudinal and transversal TEM imaging; b. and d. Binarization of the images a. and c., used for modelling.

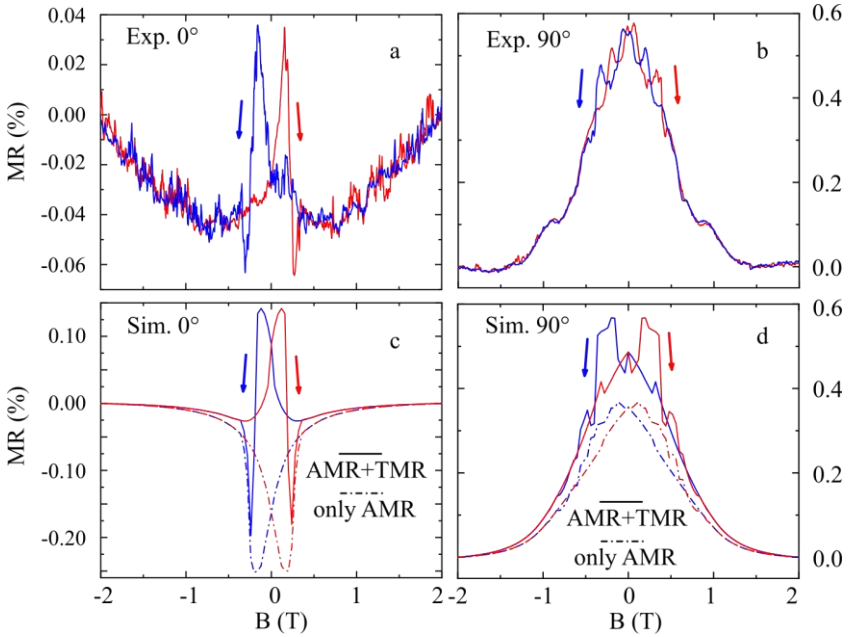



Fig. 2 a. and b. MR for magnetic field parallel and respectively perpendicular to electric current direction; both positive and negative features are present in MR, indicating the presence of two mechanisms. c. and d. OOMMF simulations of the cases b. and d. using AMR and/or TMR.

mechanisms alone can explain the observations. The interpretation of the experimental results is aided by detailed simulations of the nanowires, with realistic geometry following the binarization of the TEM longitudinal and transverse cross-sections of the wire (Figs. 1b and 1d). The geometry is implemented in OOMMF [3], and the magnetic moment vector direction for each cell is determined while varying the magnetic field, such that the AMR effect can be calculated. The TMR is considered at the grain interface. Some simulation results are shown in Figs 2c and 2d, switching off and on the TMR.

## References

[1] H. Y. Hwang, S-W. Cheong, N. P. Ong, and B. Batlogg *Phys. Rev. Lett*. 77 (1996) 2041.
[2] T. McGuire and R. Potter, *IEEE Transactions on Magnetics*, 11 (1975) 1018.
[3] M. J. Donahue, OOMMF User's Guide, Version 1.0 (1999).

# Magnetic Thin Films with Tuneable Characteristics via Combination of Different Nanoparticle Building Blocks

*Marion Görke*[1]*, *Georg Garnweitner*[2]

[1] *Technische Universität Braunschweig, Institute for Particle Technology, Volkmaroder Str. 5, 38104 Braunschweig, Germany*

[2] *Technische Universität Braunschweig, Laboratory for Emerging Nanometrology, Langer Kamp 6A, 38106 Braunschweig, Germany*

*corresponding author m.goerke@tu-braunschweig.de

To realize thin films with highly versatile magnetic properties, a modular system of building blocks, consisting of nanoparticles from different materials, was created. The building blocks were combined to form nanocomposite thin films via wet processing and were broadly varied in terms of materials and compositions as well as particle size to gain a deeper understanding on the possibilities and the underlying structure-property relations.

The nanoparticle building blocks were prepared by a non-aqueous synthesis route with different added ligands. For each synthesis the respective metal acetylacetonates, a reducing agent, solvent and the desired ligand system, consisting of an organic acid and the corresponding amine, were utilized. After the synthesis process, followed by a washing and drying step to remove excess organics, the particles were easily dispersible in non-polar solvents like hexane or chloroform. This synthesis procedure is experimentally simple and can be readily adapted to new ligand systems or different metal precursors to prepare nanoparticles of different materials.

The created library comprises magnetite, cobalt ferrite and iron platinum nanoparticles, all of them showing narrow size distribution and spherical shape. Due to the synthesis in the presence of organic ligands, the nanoparticles feature an organic shell. As ligands, combinations of carboxylic acids and the corresponding amines with specific chain length were used, to control the size and crystallinity of the particles and also the dispersibility in different solvents. The interaction of particles and ligands of various chain length gives the flexibility to combine different materials in any desired ratio while simultaneously adjusting the particle-particle distances. Thin films can be formed by mixing the dispersions of the desired compounds and then using a drop casting method, depositing the nanoparticle dispersion on any kind of planar substrate. Depending on the used building blocks the resulting thin films show controllable magnetic properties, like saturation magnetization.

First results from this approach, established in our research group, were presented by Preller et al., combining iron oxide with iron platinum nanoparticles in thin films[1]. By changing the ratio of both materials, the thin films showed increasing saturation magnetizations with increasing content of iron oxide. After annealing of the thin films an exchange-coupling effect between soft and hard magnetic phases could be observed.

To develop this approach even further, the synthesis of new particle systems was established. Also the emphasis was subsequently put on the systematic investigation of the combination of further building blocks, as well as on the usage of prepatterned substrates. In both strategies the particle-particle interactions, and therefore the magnetic properties, can be tuned by choosing different building blocks from the modular system. In a future prospect this modular system of particles and substrates can enable, for example, the easy and versatile fabrication of magnetic data storage devices via printing processes.

**Acknowledgements**

Marion Görke acknowledges support from the Hannover School for Nanotechnology.

**References**

[1] T. Preller, S. Knickmeier, D. Menzel, B. Temel, G. Garnweitner, *Langmuir*, 36 (2020) 2093-2101.

# Investigation of $Co_2FeGa$ Heusler Alloy Nanostructures Supported on SBA-15 Mesoporous Silica

*R. D. Domingos[1], A. C. Krohling[1], E. M. B. Sousa[1], K. R. Pirota[2], W. A. A. Macedo[1*]*
[1]*Centro de Desenvolvimento da Tecnologia Nuclear, CDTN, 31270-901 Belo Horizonte, MG, Brazil*
[2]*Instituto de Física Gleb Wataghin, Universidade Estadual de Campinas, 13083-859 Campinas, SP, Brazil*
*wmacedo@cdtn.br

$Co_2FeGa$ Heusler alloys are half-metallic ferromagnets that present very interesting properties for spintronic devices and other applications [1-3], and challenges for the preparation in the nanoscale. In this work, the synthesis of $Co_2FeGa$ nanostructures was explored using a wet impregnation technique supported on SBA-15 mesoporous sílica templates [4, 5], aiming to obtain an ordered material, with $L2_1$ structure, mainly seeking the formation of nanowires, for further exploration of the magneto-electrical properties of individual elements. The influence of variations in the molar ratios of the starting Co, Fe, and Ga salts, and heat treatment times in $H_2$ atmosphere on the phase composition and structural order of the obtained composite nanostructures, and, after dissolution of the sílica templates, of the metallic nanoparticles and nanowires was investigated by multiple techniques: X-ray diffraction (XRD), scanning and transmission electron microscopy, $^{57}$Fe Mössbauer spectroscopy (MS) and vibrating sample magnetometry. Our study shows that the applied non-stoichiometric synthesis, in conditions as already reported in the literature, results in significant ordering of the $Co_2FeGa$ nanostructures, for all syntheses, but also in a significant fraction of α-Fe in the samples. MS results allowed also to rule out the presence of DO3 structural disorder in the $Co_2FeGa$ nanostructures. In this system, the phase separation only by conventional XRD is not trivial. The use of Mössbauer spectroscopy is of fundamental importance for the correct characterization of the samples, to determine if the formation of pure $Co_2FeGa$ is obtained in this system via the applied synthesis conditions.

**Acknowledgements**
The financial support from the Brazilian Agencies CNPq, FAPEMIG, and FAPESP is gratefully acknowledged.

**References**

[1] C. Felser, L. Wollmann, S. Chadov, G. H. Fecher, and S. S. P. Parkin, *APL Mater.* **3** (2015) 041518.
[2] J. Hu, S. Granville, and H. Yu, *Annalen der Physik* **532** (2020) 1900456.
[3] S. Chatterjee, S. Chatterjee, S. Giri, and S. Majumdar, *J. Phys.: Condens. Matter.* **34** (2022) 013001.
[4] P. Simmon, D. Wolf, C. Wang, A. A. Levin, A. Lubk, S. Sturm, H. Lichte, G. H. Fecher, and C. Felser, *Nano Lett.* **16** (2016) 114.
[5] P. Nehla, C. Ulrich, and R. S. Dhaka, *J. Alloys Compd.* **776** (2019) 379.

# Hardening of magnetic nanoparticles by solvent-mediated local strain release

*A. López-Ortega[1,2,*], B. Muzzi,[3,4] E. Lottini,[4] N. Yaacoub,[5] D. Peddis,[6] G. Bertoni,[7] C. de Julián Fernández,[7] C. Sangregorio,[3,4]*

[1]*Dept. de Ciencias, Universidad Pública de Navarra, E-31006 Pamplona, Spain*

[2]*Institute for Advanced Materials and Mathematics, Universidad Pública de Navarra, E-31006 Pamplona, Spain*

[3]*ICCOM – CNR, I-50019 Sesto Fiorentino (FI), Italy*

[4]*Dept. of Chemistry "U. Schiff", University of Florence and INSTM, I-50019 Sesto Fiorentino (FI), Italy*

[5]*IMMM, Université du Mans, CNRS UMR-6283, F-72085 Le Mans, France*

[6]*Dept. of Chemistry and Industrial chemistry, University of Genoa, I-16146 Genova, Italy*

[7]*IMEM – CNR, I-43124 Parma, Italy*

*lopezortega.alberto@gmail.com

In this work, we demonstrate that a solvent mediated annealing process in mild condition is an effective post-treatment tool for improving the magnetic properties of chemically synthesized nanoparticles.[1] To this aim, non-stoichiometric cobalt ferrite particles of average size of 32(8) nm (see Fig. 1a) were synthesized by thermal decomposition [1] and further subjected to solvent mediated annealing at variable temperatures between 150 °C and 320 ºC in inert atmosphere. The post-synthesis treatment produces a 50% increase of the coercive field (see Fig. 1b), without affecting neither the remanence ratio nor the spontaneous magnetization. As a consequence, the energy product and the magnetic energy storage capability, key features for applications as permanent magnets and magnetic hyperthermia, can be increased by ca. 70%.

A deep structural, morphological, chemical, and magnetic characterization reveals that the mechanism governing the coercive field improvement is the reduction of the concomitant internal stresses induced by the mild annealing post-synthesis treatment (see Fig. 1c). Furthermore, we show that the medium where the mild annealing process occurs is essential to control the final properties of the nanoparticles, since classical annealing procedure performed on a dried powder does not allow the release of the lattice stress, leading to the reduction of the initial coercive field (see Fig. 1c). The strategy here proposed, therefore, constitutes a new method to improve the magnetic properties of nanoparticles, which can be particularly appealing for those materials, as is the case of cobalt ferrite, currently investigated as building blocks for the development of novel rare-earth free permanent magnets. [1-3]

**References**

[1] B. Muzzi *et al.* ACS Appl. Nano Mater. 5, (2022), 14871.
[2] A. Lopez-Ortega *et al.*, Chem. Mater. 27, (2015) 4048.
[3] E. Lottini *et al.*, Chem. Mater. 28, (2016) 4214.
[4] A. López-Ortega *et al.*, Chem. Mater. 29, (2017) 1279.

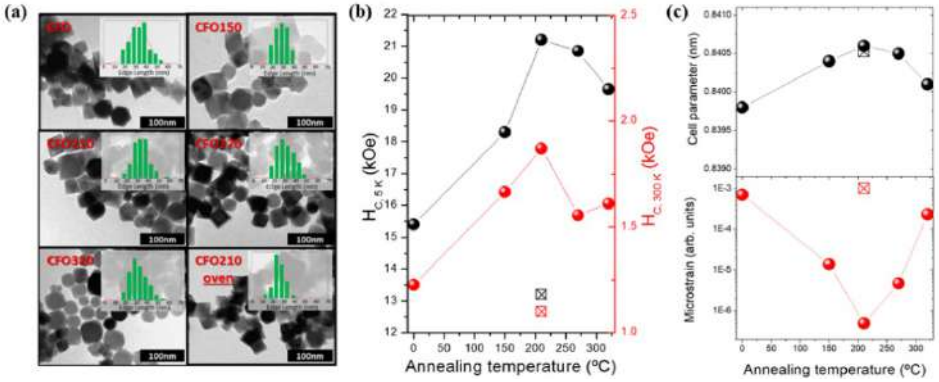



Figure 1. (a) Selected TEM images and corresponding particle size histograms for as-prepared and annealed nanoparticles. (b) Coercive field (HC) dependence measured at 5 K and 300 K as a function of the annealing temperature (empty dots refer to the oven annealed sample). (c) Cell parameter and microstrain dependence on the annealing temperature (empty dots refer to the oven annealed sample).

# Multifunctional Ni-Mn-Ga, Ni-Mn-Cu-Ga and Ni-Mn-Co-In Heusler Particles Towards the Nanoscale by Milling Techniques

*M. Solzi*[1,2,*], *G. Cavazzini*[1,2], *F. Cugini*[1,2], *D. Delmonte*[2], *G. Trevisi*[2], *L. Nasi*[2], *S. Fabbrici*[2], *L. Righi*[3], *F. Puglielli*[4], *S. Ener*[5], *L. Pfeuffer*[5], *D. Koch*[5], *O. Gutfleisch*[5], *F. Albertini*[2]

[1] *SMFI Department, University of Parma, Parco Area delle Scienze 7/A I-43124 Parma, Italy*
[2] *IMEM-CNR Institute, Parco Area delle Scienze 37/A, I-43124 Parma, Italy*
[3] *SCVSA Department, University of Parma, Parco Area delle Scienze 11/A I-43124 Parma, Italy*
[4] *MUSP, Machine Tools and Production Systems Lab., Str. Torre d. Razza 29122 Piacenza, Italy*
[5] *Institute of Material Science, Technical University of Darmstadt, Alarich-Weiss-Str. 16, 64287 Darmstadt, Germany*
*massimo.solzi@unipr.it

Microparticles of $Ni_{50}Mn_{30}Ga_{20}$, $Ni_{50}Mn_{18.5}Cu_{6.5}Ga_{25}$ and $Ni_{45.7}Mn_{36.6}In_{13.5}Co_{4.2}$ have been prepared by means of different milling methods (hand-grinding, cryo-milling, vibration and planetary ball-milling) followed by annealing treatments in order to recover the original martensitic and magnetic properties. A rapid reduction of particle size and a high dislocation density have been obtained after few hours of milling, even under low energy input, as it results from mechanical and morphological analyses. Milling temperature, time, and medium strongly impact on the degree of induced atomic disorder and agglomeration, significantly changing the morphology, crystal structure and magnetic properties.

In the case of NiMnGa and NiMnCuGa particles, we can tune the transformation temperatures or even improve the magnetic properties, in terms of phase homogeneity, thermal hysteresis and saturation magnetization, by decreasing the particles' size. Moreover, a fast recovery of the Heusler and martensitic phases is observed after a suitable annealing treatment [1].

A high-temperature annealing treatment (T> 1000 K) is required for recovering both magnetic and magneto-structural phase transitions in the case of NiMnInCo particles (Figure). Time and temperature of the treatment have been optimized in relation to the disorder introduced by the milling process, which depends on its energy and duration. However, the complex interaction between recrystallization, chemical homogenization and atomic ordering, prevents the full restoration of the magneto-structural properties of bulk [2].

Our results show a strong variability of the functional properties of this metamagnetic compound as a function of size and disorder that imposes a careful selection and tuning of the preparation route.

## References

[1] G. Cavazzini, F. Cugini, G. Trevisi, D. Delmonte, S. Ener, D. Koch, L. Righi, M. Solzi, O. Gutfleisch, and F. Albertini, *J. Alloys Compd.* **872** (2021) 159747.
[2] G. Cavazzini, F. Cugini, F. Puglielli, S. Fabbrici, D. Delmonte, G. Trevisi, L. Nasi, L. Righi, S. Ener, L. Pfeuffer, D. Koch, O. Gutfleisch, F. Albertini and M. Solzi, *J. Alloys Compd.* **906** (2022) 164377.

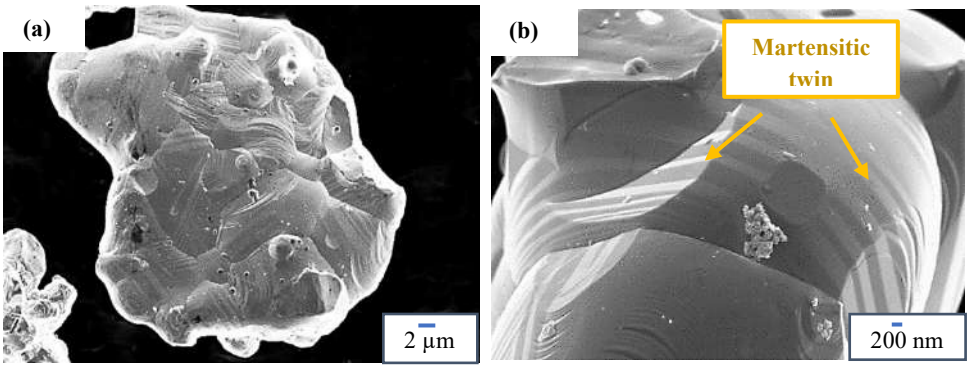



Figure: SEM micrographs of the cryo-milled (for 2 h) powders annealed for 24 h at 1100 K.

# SPIONs And Flexible Films A Prospective Study Of A New Material

*Daniel Alves Barcelos [1,2,4]*, Laura C. J. Pereira [3,4], M. Clara Gonçalves [1,2]*

*[1] Chemistry Engineering Department, Institute Superior Technical, University of Lisbon, Av. Rovisco Pais,1049-001 Lisbon, Portugal*

*[2] Center of Structural Chemistry, Av. Rovisco Pais, 1049-001 Lisbon, Portugal*

*[3] Department of Engineering and Nuclear Science, Institute Superior Technical, University of Lisbon, 2685-066 Bobadela LRS, Portugal;*

*[4] Center of Science and Nuclear Technology, Institute Superior Technical, Universidade de Lisboa, 2685-066 Bobadela LRS, Portugal*

* danielbarcelos@tecnico.ulisboa.pt

Magnetic glasses are an essential class of materials that is investigated by many different researchers [1]. Superparamagnetic Iron Oxide Nanoparticles (SPIONs) are outstanding magnetic nanoparticles that retain an important characteristic at room temperature, the superparamagnetic behavior. [1]. Corning® Willow® Glass is a cut-edge technological glass that merges the outstanding properties of glasses with flexibility that can be useful in many areas, semiconductors, and solar cells, for example [1]. Nevertheless, both materials are obtained at high temperatures, by fusion processing, and therefore, it is a highly costly production.

For that matter, our group has been conducting an investigation on the use of sol-gel (films) and co-precipitation (SPIONs) methods to check if by using these room temperature methods it would be possible to obtain such materials with a much more environmental friendly and less expensive pathway. On the behalf of the films the strategy used to obtain flexible glassy films was to use Silicon precursors ((3-Glycidyloxypropyl)trimethoxysilane (GPTMS), Triethoxy-methyl-silane (MTES), and Triethoxy-vinyl-silane (VTES)) which possess one non-hydrolyzable ligand that will be present at the final structure and accordingly this would provide a more flexible (plastic) behavior for the final product. Moreover, 3 different kinds of tapes (FCC – polyimide tape, FCT – polypropylene tape, and FC – common tape) were tested as substrates to grew-up the films in an attempt that such support would help the films to acquire more flexibility while it provides an easier way to obtain the film by itself. The first results showing the flexibility of the obtained films are shown in figure 1(a).

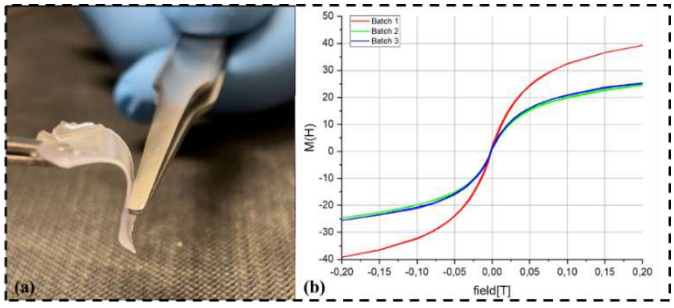



Figure 1: (a) Flexible films obtained by sol-gel method (GPTMS). (b) Superparamagnetic Behavior of SPIONs at 300 K.

SPIONs with good magnetic behavior were obtained by the co-precipitation method [2] and the superparamagnetic behavior is observed in figure 1 (b). Efforts to homogeneously spread the nanoparticles over the films are been made and will be reported at JEMS 2023.

## Acknowledgments

Authors aknowledge the finatial support from FCT (Fundação para a Ciência e Tecnologia) through projects UIDB/00100/2020, UIDP/00100/2020, PD/BD/150417/2019, UID/MULTI/04349/2022, and LISBOA-01-0145-FEDER-022096.

## References

1. Barcelos, D.A.; Leitao, D.C.; Pereira, L.C.J.; Gonçalves, M.C. *Materials 2021, Vol. 14, Page 2926* **2021**, *14*, 2926, doi:10.3390/MA14112926.
2. Matos, J.C.; Clara Gonçalves, M.; Pereira, L.C.J.; Vieira, B.J.C.; Waerenborgh, J.C. *Nanomaterials* **2019**, *9*, doi:10.3390/nano9070943.

# Magnetic anisotropy of chemically ordered CoPt and FePt nanoparticles, why is it so different?

*F. Tournus[1]*, A. Tamion[1], A. Rogalev[2], F. Wilhelm[2], J. Gutiérrez Valdés[3], L. E. Díaz-Sánchez[3], G. M. Pastor[4] and V. Dupuis[1]*

[1] *Institut Lumière Matière, CNRS, Univ. Lyon 1, Villeurbanne, France*
[2] *European Synchrotron Radiation Facility, Grenoble, France*
[3] *Universidad Autonoma del Estado de Mexico, Faculty of Sciences, Toluca, Mexico*
[4] *Institut für Theoretische Physik, Universität Kassel, Kassel, Germany*
*florent.tournus@univ-lyon1.fr

The CoPt and FePt alloys, when chemically ordered in the $L1_0$ phase [1], are among the magnetic materials displaying the highest magnetic anisotropy constant. Therefore, they are perfect candidates for ultra-high density magnetic storage applications, provided nanoparticles can be prepared in such a high anisotropy phase. In the bulk $L1_0$ phase both alloys exhibit close magnetic and structural properties. Indeed, the tetragonalization of the unit cell, measured by the c/a ratio is almost the same and equal to 0.97 for both alloys while their anisotropy constant is equal to 5 and 7 $MJ.m^{-3}$ respectively for CoPt and FePt [1]. In the present work [2], we first wanted to determine and compare the intrinsic magnetic anisotropy constant of CoPt and FePt $L1_0$ nanomagnets. For this purpose, we have prepared mass-selected nanoparticles (around 3 nm diameter) diluted in an inert carbon matrix to avoid coalescence and to make magnetic interactions negligible. To promote the chemically $L1_0$ ordered phase the samples have to be annealed at 600°C during two hours. The chemical order is detected by extended x-ray absorption fine structure (EXAFS) measurements (local environment around absorbing atoms) as well as directly visible from transmission electron microscopy observations. Nevertheless, both for CoPt and FePt particles, we observe a coexistence of single $L1_0$ domain and multi-$L1_0$ domains particles (in particular for multi-twinned decahedral particles [3]). The magnetic properties were then determined using SQUID magnetometer and a global fitting procedure [4] of the entire set of magnetic measurements (see Figure) [2]. We find that the magnetic anisotropy of CoPt and FePt nanoparticles differ greatly. To understand the origin of such a difference, we discuss the atomic relaxation as measured with EXAFS and investigated with density functional theory (DFT) calculations on truncated-octahedral particles. Even with a perfect chemical order, finite size effects result in a relaxation which is of particular importance in the case of CoPt. We conclude that the very high magnetic anisotropy measured in FePt nanoparticles (see Figure) is related to the weak atomic relaxation in this alloy, while for chemically ordered CoPt nanoparticles the strong atomic relaxation in the Co plans [5] seems to reduce drastically the magnetic anisotropy.

## References

[1] P. Andreazza et al., *Surf. Sci. Rep.* **70**, 188 (2015).
[2] A. Tamion et al., *Phys. Rev. B* **106**, 104420 (2022).
[3] F. Tournus et al., *Phys. Rev. Lett.* **110**, 055501 (2013).
[4] A. Tamion et al., *Phys. Rev. B* **85**, 134430 (2012) ; A. Hillion et al., *Phys. Rev. B* **88**, 094419 (2013).
[5] N. Blanc et al., *Phys. Rev. B* **87**, 155412 (2013).

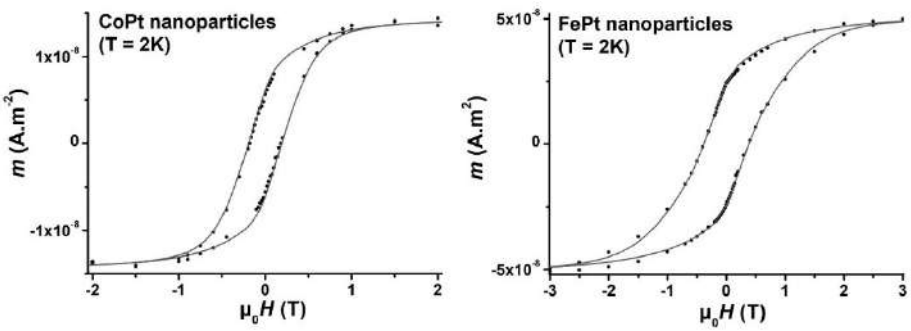



Figure: low temperature (2 K) hysteresis loop measured for a diluted assembly of chemically ordered CoPt (left) or FePt (right) particles. The solid lines are numerical fits of the experimental data (dots).

# Innovative Recycling Method of Soft Ferrite Cores by Ball Milling to Obtain Printable Composites

*Uxua Jiménez-Blasco.*[1-2*], *Eneko Garayo*[1-2], *Juan Jesús Beato-López*[1-2], *Alberto Lopez-Ortega*[1-2], *Antonio Urbina* [1-2], *Vicente Sanchez-Alarcos*[1-2], *Vicente Recarte*[1-2], *Cristina Gómez-Polo*[1-2] *and Jose Ignacio Pérez-Landazábal*[1-2]

[1]Departamento de Ciencias, Universidad Pública de Navarra, Campus de Arrosadía, 31006, Pamplona, Spain

[2]Institute for Advanced Materials and Mathematics (INAMAT2), Universidad Pública de Navarra, Campus de Arrosadía, 31006, Pamplona, Spain

*uxua.jimenez@unavarra.es

Soft ferrites are the most widely traded magnetic material in the world [1] and are mainly used as protective mitigators of electromagnetic noise. Every year a large amount of waste is generated that requires a considerable economic investment for its treatment. To solve this problem, an innovative ferrite recycling method is proposed, based on the production of 3D printer polymeric filaments with ferrite particles inside. The recycling process begins by grinding the ferrite cores in a ball mill to obtain particles with a size below 100 µm. Different grinding conditions are studied to produce recycled ferrite particles (RFPs) [2]. Its structural and magnetic characteristics are characterized by VSM, SQUID and XRD. Once the RFPs are obtained, a composite synthesis is performed starting from a solution of polymer and RFP. To obtain the printable filament, the synthesized compound is extruded and characterized by SEM in order to obtain an adequate dispersion of the RFPs (Figure 1). Finally, the extruded filaments are fed into a 3D printer to obtain simple, ecological and economical devices. The goal is that once the devices are printed they have suitable properties for different applications.

## Acknowledgements

This work has been carried out with the financial support of the Navarra Government (project number PC003-04 3D-MAGNET).

## References

[1] «Ferrite Market». https://www.futuremarketinsights.com/reports/ferrite-market (accedido 9 de marzo de 2023).

[2] M. J. Molaei, A. Ataie, S. Raygan, y S. J. Picken, «Role of intensive milling in the processing of barium ferrite/magnetite/iron hybrid magnetic nano-composites via partial reduction of barium ferrite», *Mater. Charact.*, vol. 101, pp. 78-82, mar. 2015, doi: 10.1016/j.matchar.2015.01.006.

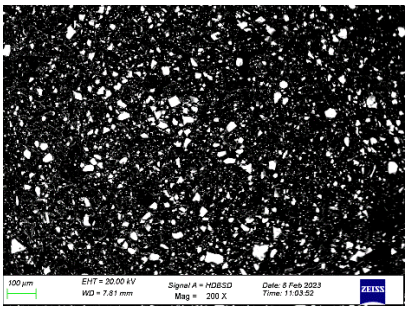



Figure 1. SEM image of RFPs dispersion into synthesized RFP-PCL composite filament.

# Using Magnetic Nanofibers To Control The Magnetic Anisotropy Of A Thin Permalloy Film

*M. Volmer[1]*, M. David[1], M. Avram[2], M. Florescu[1], A. Bezergheanu[1], C. Mușuroi[1], M. Oproiu[1]*

[1] *Transilvania University of Brasov, Eroilor 29 Blvd, 500036 Brasov, Romania*

[2] *National Institute for Research and Development in Microtechnologies, Erou Iancu Nicolae 32B Street, 72996 Bucharest, Romania*

* volmerm@unitbv.ro

One-dimensional magnetic structures, such as magnetic nanofibers, are of high interest since the strong shape anisotropy reveals new magnetization reversal modes and possible applications in cancer theranostics, energy harvesting, electromagnetic shielding and others. One possibility to create magnetic nanofibers in a relatively simple way is offered by electrospinning them from polymer solutions with incorporated magnetic nanoparticles [1,2].

In this study we chose to deposit polymeric nanofibers, doped with $Fe_2O_3$/NiO nanoparticles, on thin Permalloy disks, 1 mm diameter and 10 nm thick, Fig. 1(a). For this geometry there is no uniaxial anisotropy axis. The polymer was a mix of Chitosan (CS) and Polyvinyl alcohol (PVA). After mixing these two components and homogenization, magnetic nanoparticles of $Fe_2O_3$/NiO with a mean diameter lower than 50 nm, at a concentration of 0.1 % (10 mg/mL), were added. For electrospinning was used the E-FIBER EF100 machine. The deposition lasted 40 mins at an injection flow rate of 10µL/h, and a voltage of 35 kV between the metallic needle and the substrate. A magnetic field of 200 Oe was applied during the deposition in order to obtain well align nanofibers on the Permalloy disks, Fig.1 (a). Through SEM measurements we found well aligned nanofibers with lengths ranging from 10 to 20 µm and a mean diameter of about 197 nm.

To see the influence of the magnetic nanofibers on the Permalloy disks, were performed galvanomagnetic measurements using a planar Hall effect (PHE) setup. The applied magnetic field, H, can be parallel or perpendicular to the main direction of nanofibers, Fig. 1 (b and c). This direction will be assumed as an induced easy axis of magnetization. Figures 1(d) and 1(e) present the field dependences of the measured signal when H is applied parallel and perpendicular to the main direction of the nanofibers. These characteristics show the effect of the magnetostatic coupling between magnetic nanofibers and the Permalloy disks. Other orientations of the magnetic field are used to complete this study. By running micromagnetic simulations, will be explained the obtained experimental results like, for example, the sharp peaks from Fig. 1(e). Through this study, we show that magnetic nanofibers can be used to successfully control the detection characteristics of magnetic sensors, by inducing in their layer a well-defined anisotropy axis.

**Acknowledgements**

This work was supported by a grant of the Ministry of Research, Innovation and Digitization, CCCDI - UEFISCDI, project number PN-III-P2-2.1-PED-2021-3112, 597PED/2022, within PNCDI III.

**References**

[1] Y. Liu, X. Zhang, Y. Xia, H. Yang, *Adv Mater.* **22** (2010) 2454.

[2] T. Blachowicz, A. Ehrmann, *Journal of Engineered Fibers and Fabrics,* **15** (2020) 1.

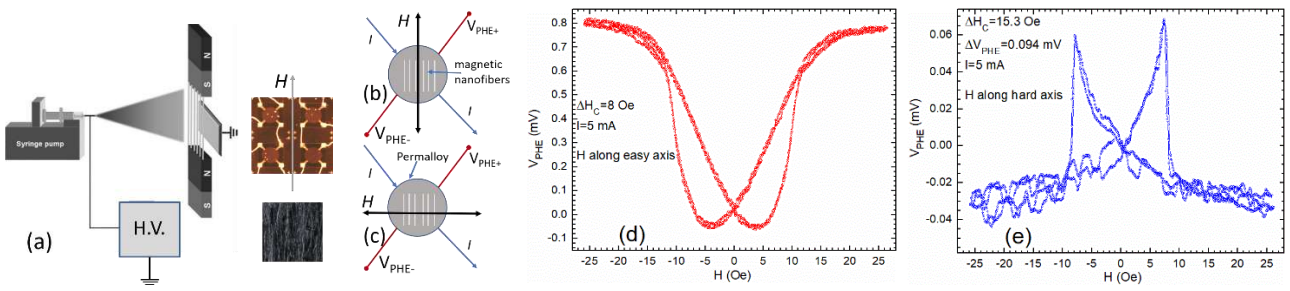



Figure 1: (a) Simplified schematics of magnetic nanofiber deposition setup based on electrospinning method, showing the chip with four Permalloy disks, (b and c) measurement configurations and results for (d) magnetic field applied parallel with nanofibers and (e) perpendicular on nanofibers.

# High Anisotropy Magnetic Oxides with mm-Wave Natural Ferromagnetic Resonances

*Vinod V K Thalakkatukalathil, Naureen Khanam, Nico Dix*, Marti Gich*

*Institut de Ciència de Materials de Barcelona (ICMAB-CSIC), Campus UAB, Bellaterra 08193, Spain*

*ndix@icmab.es

The advances in communication technology and the forthcoming internet of things (IoT) era, place millimeter wave frequencies in the focus for a variety of applications including sensors, imaging, wireless communications, radars and advanced driver assistance systems. Millimeter wave frequencies, particularly in the 95-300 GHz range will become important in the next generation of wireless transmission applications 6G and beyond [1],[2]. Ferrites are the functional materials in non-reciprocal devices such as circulators commonly used in telecommmunications. Their operating frequency is dependent on the saturation magnetization and anisotropy field of the ferrite, which determines the ferromagnetic resonance frequency in an external applied field. High anisotropy magnetic oxides like $Sr_{0.67}Ca_{0.33}Al_4Fe_8O_{19}$ hexaferrite [4] and ε-$Fe_2O_3$ [4] present natural ferromagnetic resonances (NFMR) at millimeter wave frequencies. Such high-frequency NFMR makes these materials appealing for implementing 6G non-reciprocal devices that would not require the use of external magnetic fields. Here, we investigate sub-micron sized powder samples of these two high anisotropy ferrites prepared by chemical methods. We detail briefly the structural features and then compare magnetic properties and millimetre wave absorption of both compounds. The samples were analysed with a quasi optical measurements system with Vector network analyser, to determine the absorption and permittivity in these complex oxides at frequencies ups to 220 GHz. Due to the strong magnetic anisotropy of these ferrites they show a natural zero field resonance around 180 GHz. We conclude that ε-$Fe_2O_3$ may yield advantages due to a much stronger absorbtion and sharper absorption line than the hexaferrite.

## Acknowledgements

Funding is acknowledged from Severo Ochoa Programme for Centres of Excellence in R&D (FUNFUTURE CEX2019-000917-S) and European Research Council (FeMiT project ERC-CoG 819623). Judit Oró and Bernat Bozzo from the ICMAB scientific services are acknowledged for the analysis of materials by transmission electron microscopy and magnetic measurements respectively.

## References

[1] D T. S. Rappaport *et al.*, IEEE Access, vol. 7, 78729–78757, 2019.
[2] I. F. Akyildiz *et al.*, IEEE Access, vol. 8, 133995–134030, 2020.
[3] M. Popov *et al.*, IEEE Trans. Magn., vol. 47, no. 2, 289–294, 2011.
[4] M. Gich *et al.*, J. Appl. Phys., 98, 044307, 2005.

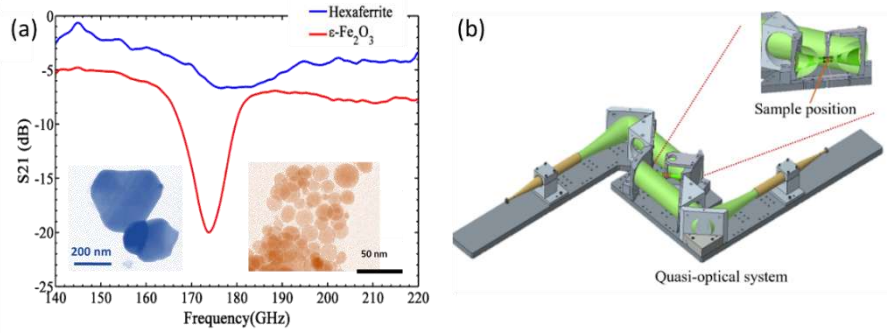



Figure: (a) Millimeter wave absorption (natural ferromagnetic resonance, NFMR). Hexaferrite (blue line) and ε-Fe2O3 (red line). Insets show the corresponding TEM images for the measured sub-micron particles. (b) Schematic quasi optical measurement setup [©Thomas Keating Ltd.].

# Semi-hard Magnetic Properties of FeCo-Alloy/Co-Ferrite Core/Shell Nanoparticles with Spring Exchange Embeeded in a Carbon Matrix.

*Alberto Castellano-Soria[1,2],*, Jesús López-Sánchez[3,4], Aida Serrano[5], Giulio Gorni[6,7], María Varela[2,8], Antonio Hernando[1,2], Pilar Marín,[1,2] and Elena Navarro[1,2]*

[1] *Instituto de Magnetismo Aplicado (IMA-UCM-ADIF), 28230 Madrid, Spain*

[2] *Departamento de Física de Materiales, Facultad de Físicas, Universidad Complutense de Madrid (UCM), 28040 Madrid, Spain*

[3] *SpLine, Spanish CRG BM25 Beamline, ESRF-The European Synchrotron, 38000 Grenoble, France*

[4] *Instituto de Ciencia de Materiales de Madrid (ICMM-CSIC), 28049 Madrid, Spain*

[5] *Instituto de Cerámica y Vidrio (CSIC), 28049 Madrid, Spain*

[6] *CELLS-ALBA Synchrotron Light Facility, E-08290 Cerdanyola del Vallès, Spain*

[7] *Instituto de Óptica (IO-CSIC), 28006, Madrid, Spain*

[8] *Instituto Pluridisciplinar, Universidad Complutense de Madrid (UCM), 28040 Madrid, Spain*

*albcas04@ucm.es

The body-centered cubic (bcc) Fe-Co alloy is known to be a ferromagnetic compound with an elevated curie temperature, a large magnetic permeability, and the highest saturation magnetization among the magnetic materials. Such excellent magnetic properties have resulted in many nano-applications of technological interest as in biomedicine as highly sensitive tracers for magnetic-particle-imaging, magnetic fluid hyperthermia, photothermal drug delivery; as supporting media for enhanced catalysis, electrocatalysis, and magneto-assisted catalysis, microwave absorption, and spintronics. Among the different synthesis methodologies, the chemical methods have constituted a solid alternative against the physical approaches combining both, high control of the alloys, and morphology of nanosized materials, along with a high scalability and reproducibility desirable for industrial up-scaling.

We present a synthesis approach based on a sol-gel macromolecule-surfactant method, for a fine tuning of the magnetic properties of Fe-Co alloyed/Co-ferrite core/shell nanoparticles embedded in a carbon matrix. We explore the versatility of the synthesis by advanced materials characterization techniques, finding a precise control of the Fe-Co alloyed nanoparticles and obtaining a modulation of their magnetic properties. The magnetic nanoparticle-carbon composites obtained display an elevated saturation magnetization and an enhanced coercive field. Such magnetic hardening is induced by a spring-exchange effect between the soft core and semi-hard shell. The thickness of Co-ferrite shells is much lower when it is compared with other investigations [1,2], indicating a very effective exchange coupling.

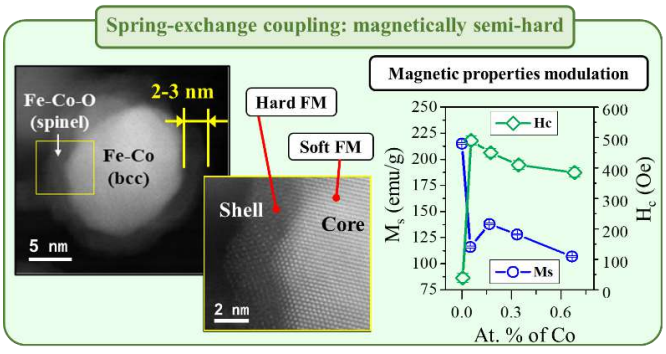



Figure: Fe-Co alloyed/Co-ferrite core/shell nanoparticles and its magnetic properties.

**Acknowledgements**

Comunidad de Madrid through the project S2018/NMT-4321 NANOMAGCOST.

**References**

[1] Dias, D. R. C., *et. al. Materials Research-Ibero-American Journal of Materials*, 22(3), (2019).

[2] Karipoth P., *et. al*. *Journal of Applied Physics,* 120, 123906 (2016).

# Dependence of Exchange Bias on Interparticle Interactions in Co/CoO Core/Shell Nanostructures

*Suchandra Goswami[1], Pushpendra Gupta[2], Sagarika Nayak[2], Subhankar Bedanta[2], Òscar Iglesias[3]*, Manashi Chakraborty[1], Debajyoti De[1,4]*

[1] *Material Science Research Lab., The Neotia University, Sarisha, West Bengal, India*

[2] *School of Physical Sciences, National Institute of Science Education and Research (NISER), Jatni, India*

[3] *Dpt. Física de la Matèria Condensada and IN2UB, Univ. de Barcelona, Barcelona, Spain*

[4] *Dpt. of Physics, Sukumar Sengupta Mahavidyalaya, Keshpur, India*

*oscariglesias@ub.edu

We report the dependence of exchange bias (EB) effect on interparticle interactions in nanocrystalline Co/CoO core/shell structures, synthesized using the conventional sol-gel technique [1]. Analysis via powder X-Ray diffraction (PXRD) studies and transmission electron microscope (TEM) images confirm the presence of crystalline phases of core/shell Co/CoO with average particle size ≈ 18 nm. Volume fraction (ϕ) is varied (from 20% to 1%) by the introduction of a stoichiometric amount of non-magnetic amorphous silica matrix ($SiO_2$) which leads to a change in interparticle interaction (separation). The influence of exchange and dipolar interactions on the EB effect [1], caused by the variation in interparticle interaction (separation) is studied for a series of Co/CoO core/shell nanoparticle systems [2]. Studies of thermal variation of magnetization (M- T) and magnetic hysteresis loops (M - H) for the series point towards strong dependence of magnetic properties on dipolar interaction in concentrated assemblies whereas individual nanoparticle response is dominant in isolated nanoparticle systems. The analysis of the EB effect reveals a monotonic increase of coercivity ($H_C$) and EB field ($H_E$) with increasing volume fraction. When the nanoparticles are close enough and the interparticle interaction is significant, collective behavior leads to an increase in the effective antiferromagnetic (AFM) CoO shell thickness which results in high $H_C$ and $H_E$ [2]. Moreover, in concentrated assemblies, the dipolar field superposes to the local exchange field and enhances the EB effect contributing as an additional source of unidirectional anisotropy.

**Acknowledgements**

S.G., M.C. and D.D. thank SERB Project EMR/2017/001195.S. Bedanta thanks DAE of Government of India for financial support. M.C. and D.D. acknowledge financial support from UGC-DAE through CRS project numbers CRS/2021-22/02/503 and CRS/2021-22/02/492. Ò. I. thanks Spanish MINECO (PGC2018-097789-B-I00, PID2019-109514RJ-I00), Catalan DURSI (2017SGR0598) and EU FEDER funds (Una manera de hacer Europa).

**References**

[1] D. De, Ò. Iglesias, S. Majumdar, S. Giri, *Phys. Rev. B* **94** (2022) 184410

[2] S. Goswami et al., *Nanomaterials* **12** (2022) 3159

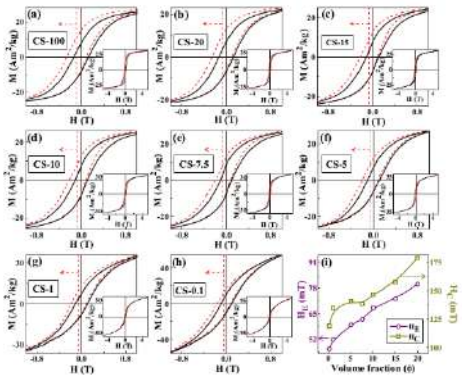



Figure: **a**–**h**) ZFC (solid line) and FC (under $H_{cool}$= 1 T, dashed line) hysteresis loops at 4 K for different increasing sample concentrations (**i**) Variation of coercivity ($H_C$) and EB field ($H_E$) with change in volume fraction (ϕ).

# Phase transitions, magnetic structures and frustration in ε-$Fe_2O_3$ nanoparticles

*Naureen Khanam[1], Arnau Romaguera[1], Javier Herrero[2], Oscar Fabelo[3], José Luis García-Muñoz, Martí Gich[1]**

[1]*Institut de Ciència de Materials de Barcelona, Consejo Superior de Investigaciones Científicas,Spain*
[2]*CELLS-ALBA Synchrotron, Cerdanyola del Vallès, Barcelona, 08290 Spain*
[3]*Institut Laue-Langevin Synchrotron, Grenoble Cédex, 38042, France*
*mgich@icmab.es

ε-$Fe_2O_3$ is typically stabilized in form of nanoparticles. It presents interesting properties and is receiving increasing attention for its prospective application potential. In particular, it displays a huge coercive field (up to 20 kOe at room temperature), millimeter-wave ferromagnetic resonance, magneto-electric coupling, and room-temperature ferroelectricity [1-3].

The complex noncentrosymmetric structure of ε-$Fe_2O_3$, with three distinct octahedral and one tetrahedral environments for Fe sites, and a geometrical frustration, plays a key role in its rich phase diagram: two consecutive collinear ferrimagnetic orders FM1 and FM2 [4] between its paramagnetic state at about 850 K and 150 K where and incommensurate structure is developed upon cooling. Combining neutron and synchrotron X-ray diffraction and X-ray circular dichroism, we present new insights on the magnetic phase diagram of ε-$Fe_2O_3$ and on the interplay between magnetic anisotropy, frustration and the stabilization of the super-hard ferrimagnetic FM2 phase in the 150-500 K interval. Structural changes observed at the onset of the super-hard FM2 magnetic phase reveal the connection between frustration and magnetocrystalline anisotropy. We also studied the stability of the incommensurate magnetic order under applied magnetic field by neutron diffraction at different temperatures, finding a field-dependent transition to the collinear phase.

**Acknowledgements**

Funding is acknowledged from Severo Ochoa Programme for Centres of Excellence in R&D (FUNFUTURE CEX2019-000917-S), the Spanish Ministerio de Ciencia e Innovación (MAGMUF project PID2021-124734OB-C22) and European Research Council (FeMiT project ERC-CoG 819623). We are grateful to ALBA synchrotron and the Institut Laue Langevin for providing access to beamtime.

**References**

[1] B. Jin, S. Ohkoshi, K. Hashimoto, *Adv. Mater.* **16** (2004) 48.
[2] A. Namai, M.Yoshikiyo, K. Yamada, S. Sakurai, T. Goto, T. Yoshida, T. Miyazaki, M. Nakajima, T. Suemoto, H. Tokoro, S. Ohkoshi, *Nature Commun.* **3** (2012) 1035.
[3] M. Gich, M.; Fina, A. Morelli, F. Sánchez, M. Alexe, M.; J. Gàzquez, J. Fontcuberta, A. Roig, *Adv. Mater.* **26** (2014) 4645.
[4] J. L. García-Muñoz, A. Romaguera, F. Fauth, J. Nogués, M. Gich, *Chem. Mater.* **29** (2017) 9705.

# Neutron Diffraction Study of High-Coercivity Al-Substituted Hexaferrite Magnetic Nanoparticles

*Matilde Saura-Múzquiz[1]*, Anna Zink Eikeland[2], Marian Stingaciu[2], Henrik Lyder Andersen,[1] Maxim Avdeev[3] and Mogens Christensen[2]*

[1] *Dept. of Materials Physics, Universidad Complutense de Madrid, Madrid, Spain*
[2] *Dept. of Chemistry and iNANO, Aarhus University, Aarhus, Denmark*
[3] *Australian Nuclear Science and Technology Organisation, Lucas Heights, NSW, Australia*
*matsaura@ucm.es

The M-type strontium hexaferrite, $SrFe_{12}O_{19}$, is a highly used rare-earth free permanent magnetic material due to its relatively low cost, high chemical stability, good magnetic properties, and high Curie temperature ($T_C$ = 737 K).[1] Given its widespread use, even slight improvements to its performance may have significant socioeconomical impact. In our previous work, we have demonstrated how nanostructuring methods can be effectively used to tune and enhance the magnetic properties of hexaferrites.[2-4] A complementary approach to further optimize the material's performance is modifying the intrinsic properties at the atomic level by chemical substitution. So far, the most significant enhancement of coercivity has been achieved by substituting Ca and Al into the hexaferrite structure, although it is always accompanied by a drastic reduction in magnetic saturation.[5]

In this study, we investigate the structure and magnetic properties of aluminium-substituted strontium hexaferrite nanoparticles, $SrFe_{12-x}Al_xO_{19}$, with doping levels $x$ = 0-3, synthesized *via* three different methods: hydrothermal autoclave synthesis, citrate sol-gel synthesis and solid-salt matrix sol-gel synthesis. The nuclear and magnetic structure, site occupation of Al within the structure, and its influence on the macroscopic magnetic performance have been investigated by means of powder X-ray diffraction (PXRD), neutron powder diffraction (NPD) and vibration sample magnetometry (VSM). Joint Rietveld analysis of the NPD and PXRD data reveals a clear affinity of Al for specific crystallographic sites within the structure, and how it evolves as a function of increasing Al doping. Furthermore, the intrinsic saturation magnetization obtained from the magnetic structure refinement of the NPD data is in remarkable agreement with the observed macroscopic magnetic properties as well as with theoretical calculations,[6] confirming the robustness and accuracy of the model. This is, to the best of our knowledge, the first experimental study to show evidence of Al site distribution within $SrFe_{12}O_{19}$ and to provide direct experimental correlation between atomic structure and macroscopic magnetic performance.

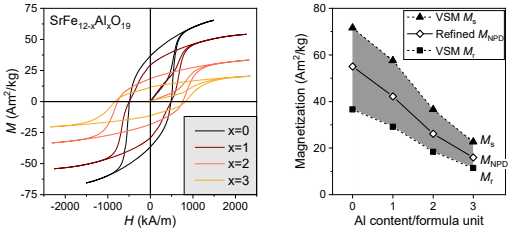



Figure 1. Left: Hysteresis curves of differently doped $SrFe_{12-x}Al_xO_{19}$ samples. Right: Saturation and remanent magnetization ($M_s$, $M_r$) obtained by VSM, and intrinsic RT magnetization obtained by refinement of NPD data ($M_{NPD}$).

## Acknowledgements

The authors gratefully acknowledge the beamtime granted by the Australian Centre for Neutron Scattering, Australian Nuclear Science and Technology Organisation, Australia. Matilde Saura-Múzquiz gratefully acknowledges the financial support from the Comunidad de Madrid, Spain, through an "Atracción de Talento Investigador" fellowship (2020-T2/IND-20581).

## References

[1] R. C. Pullar, *Prog Mater Sci,* **57**, (2012) 1191-1334.
[2] M. Saura-Múzquiz *et al.*, *Nanoscale,* **8**, (2016) 2857-2866.
[3] M. Saura-Múzquiz *et al.*, *ACS Appl Nano Mater,* **1**, (2018) 6938-6949.
[4] M. Saura-Múzquiz *et al.*, *Nanoscale,* **12**, (2020) 9481-9494.
[5] E. A. Gorbachev *et al.*, *Materials Today,* **32**, (2020) 13-18.
[6] V. Dixit *et al.*, *J Appl Phys,* **117**, (2015) 243904.

# SYMPOSIUM 08.
## MAGNETISM IN MOLECULAR, IONIC, ORGANIC BASED SYSTEMS.
## S8. INVITED ORAL PRESENTATIONS

# Devices for Quantum Computing: Towards a Measurement of on-Surface Qubit Phase Memory Time

*Guillem Gabarró-Riera,*[1,2] *E. Carolina Sañudo*[1,2*]

[1] *Institut de Nanociència i Nanotecnologia UB (IN2UB). C/Martí i Franqués 1 -11, 08028 Barcelona, Spain;)*

[2] *Departament de Química Inorgànica i Orgànica, Facultat de Química, Universitat de Barcelona. C/Martí i Franqués 1 -11, 08028 Barcelona, Spain*

* esanudo@ub.edu

Applications of quantum computation are key for the new Quantum Revolution. So far, most of these applications rely on Josephson Junctions or defects doped on diamond as qubits. However, molecular qubits offer a wide variety of advantages towards other qubit options: molecules are all prepared equal, there is no size distribution, they can be functionalized at will with desired properties by ligand modeification, they will require small voltages so that a moecular device will consume small amount of energy... The possible applications of molecules as qubits or for spintronics applications relies on the surface deposition of said molecules over large areas of a conductive material.[1] In this work, we show how we can achieve chemisorption of molecular systems over functionalized Silicon wafers.[2] Among the deposited molecules, we have chosen several examples of known qubits with the main objective of measuring the decoherence (or phase memory) time of a monolayer of qubits.

## References

[1] Guillem Gabarró-Riera, Guillem Aromí, E. Carolina Sañudo. Coord. Chem. Rev. **2023**, *475*, 214858

[2] Guillem Gabarró-Riera, Jesús Jover, Juan Rubio Zuazo, Elena Bartolomé, E. Carolina Sañudo. *Inorg. Chem. Front.*, **2022**, *9,* 4160

# SYMPOSIUM 08.
## MAGNETISM IN MOLECULAR, IONIC, ORGANIC BASED SYSTEMS.
## S8. ORAL PRESENTATIONS

# STOSS: A stochastical spin simulator for molecular nanomagnets

*Gerliz M. Gutiérrez-Finol[1]*, Silvia Giménez-Santamarina[1], Ziqi Hu[1], Lorena E. Rosaleny[1],Salvador Cardona-Serra[1], Alejandro Gaita-Ariño[1]*

[1] *ICMol, Universitat de València, c/cat José Beltrán, 2, 46980, Paterna, Spain*

*gerliz.gutierrez@uv.es

Initial studies on molecular nanomagnets assumed that a simple Orbach process was the main relaxation mechanism at high temperatures, thus the focus was set on modelling the effective barrier (Ueff), [1] with a Quantum Tunneling of the Magnetisation (QTM) model being responsible for spin relaxation at low temperatures; [2] later the role of Raman and the direct process was also recognized. Combining these mechanisms with the idea of probabilistic bits (p-bits), we present STOSS (STOchastic Spin Simulator), a microscopic simulator code for spin p-bits based on molecular nanomagnets. STOSS [3] is able to reproduce the most characteristic macroscopic magnetic dynamics of molecular nanomagnets, namely magnetization relaxation, and in-phase, out-of-phase susceptometry, by simulating the individual states in a collective of spin p-bits. Furthermore, we employed the code to simulate the thermal- and field- dependence of hysteresis in a novel endohedral metallofullerene. There is a lack of standards in SIM characterization, with competing reports of hysteresis temperatures at different sweep rates, temperature for a tau=100s relaxation exponent, ac susceptometry etc. STOSS is a tool that can take parameters extracted from a kind of measurement and allows estimating the expected result of a different experiment and thus will be useful to compare the performance of compounds being subjected to dissimilar studies.

**Acknowledgements**

We acknowledge funding to COST Project 15128, EU Horizon 2020 (ERC-2017-AdG-788222 “MOL2D” and FATMOLS project),Generalitat Valenciana CIDEGENT/2021/018 grant and GVA PROMETEO/2019/066, MICINN grants PID2020-117264GB-I00 and PID2020-117177GB-I00 (co-financed by FEDER funds), MICINN PRE2018-083350, CEX2019-000919-M and MINECO CTQ2017-89528-P.

**References**

[1] S. Takamatsu, T. Ishikawa, S. Y. Koshihara, N. Ishikawa,*Inorganic Chemistry*, **46** (2007) 7250.
[2] J. Liu, E. Del Barco, S. Hill, *Physical ReviewB*, **85** (2012) 012406.
[3] https://github.com/gerlizg/STOSS

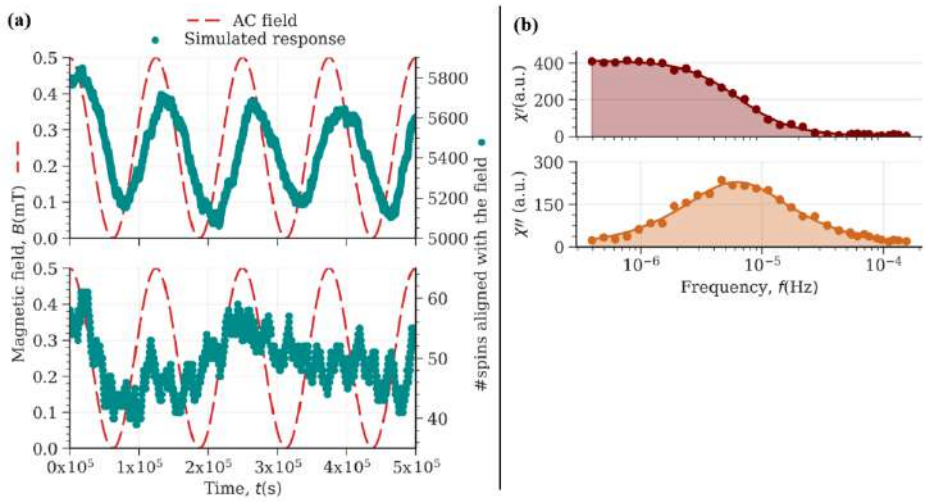



Figure: Periodically driven p-bits behave exactly as spins in ac susceptometry: (a) calculated time evolution of N = $10^4$ (up) or N = $10^2$ (down) stochastic spins described by the parameters corresponding to $[(Cp^{iPr5})Dy(Cp^*)]^+$, at 20 mK (b) simulated χ′, χ″; spin dynamics is governed by QTM. Solid lines indicate the fits using a generalized Debye model.

# Spin filtering effects at graphene/molecules interfaces

*L. Chen[3,1], A. Droghetti[2] , B. Dlubak[4], P. Martin[3] , R. Mattana[4], P. Seneor[4] and C. Barraud[1],*

[1] *MPQ, Physics Dept., Université Paris Cité, Paris, France*

[2] *School of Physics and CRANN, Trinity College, Dublin, Ireland*

[3] *ITODYS, Chemistry Dept., Université Paris Cité, Paris, France*

[4] *Unité Mixte de Physique CNRS/Thales, palaiseau, France*

*linsai.chen@etu.u-paris.fr

We present a bias-controlled spin-filtering mechanism in spin-valves including a hybrid organic chain/graphene interface [1]. Wet growth conditions of oligomeric molecular chains would usually lead, during standard CMOS-compatiable fabrication processes, to the quenching of spintronics properties of metallic spin source due to oxidation. We demonstrate by X-ray photoelectron spectroscopy that the use of a protective graphene layer fully preserves the metallic character of the ferromagnetic surface and thus its capabillity to deliver spin polarized currents. We focus here on a small aromatic chian of controllable lengths, formed by nitrobenzene monomers and derived from the commercial 4-nitrobenzene diazonium tetrafluoroborate, covalently attached to the graphene passivated spin sources thanks to electroreduction. A unique bias dependent switch of the spin signal is then observed in complete spin valve devices, from minority to majority spin 84 ABSTRACTS carriers filtering. First-principles calculations are used to highlight the key role played by the spin-dependent hybridization of electronic states present at the different interface [2].

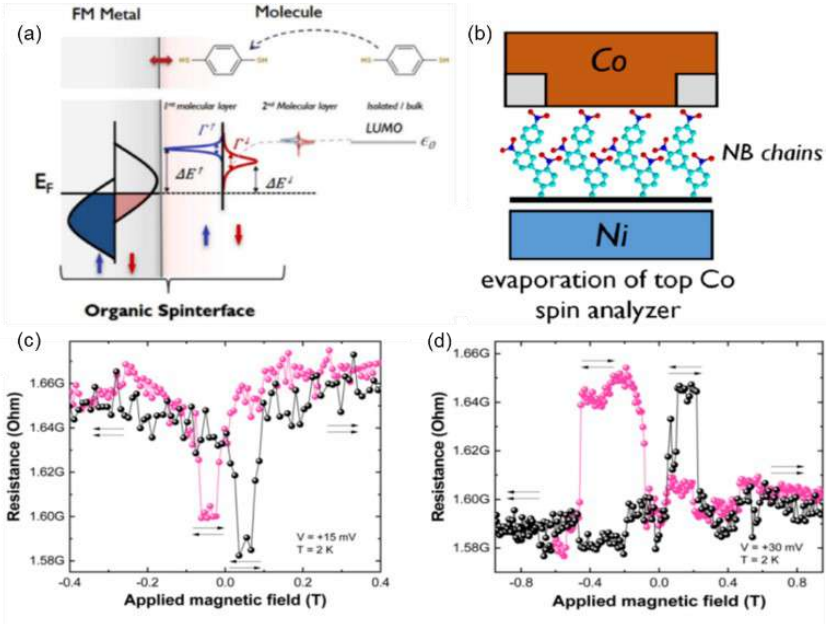



Figure: a). Schematic explaining the hybridization between a ferromagnetic surface and a molecular orbital. Extracted from Delprat *et al*., *J. Phys. D: App. Phy.* **51** 473001 (2018) b). Schematics of the molecular magnetic junction. c) and d). Magnetoresistive signals measured in Ni/graphene/molecule/Co magnetic junctions.

## References

[1] P. Martin et al., *Nanoscale* **14** (2022) 12692-12702.

[2] P. Martin et al., *Adv. Quant. Tech* **5** (2022) 2100166.

# Magnetic anisotropy energy of 3d adatoms and 3d–O molecules on the bilayer of MgO

*Sufyan Shehada,[1,2,3]*, Manuel dos Santos Dias[4], Muayad Abusaa[3], and Samir Lounis[1,5]**

[1] *Peter Grünberg Institute (PGI), FZJ, 42425 Julich, Germany*
[2]*Department of Physics, RWTH Aachen University, 52056 Aachen, Germany*
[3]*Department of Physics, Arab American University, Jenin, Palestine*
[4]*Scientific Computing Department, STFC Daresbury Laboratory, Warrington WA4 4AD, United Kingdom*
[5] *Faculty of Physics, University of Duisburg-Essen, 47053 Duisburg, Germany*
* s.shehada@fz-juelich.de, s.lounis@fz-juelich.de

Designing systems with large magnetic anisotropy energy [MAE] is desirable and critical for nanoscale magnetic devices. Thus far, the MAE per atom in single-molecule magnets and ferromagnetic films remains typically one to two orders of magnitude below the theoretical limit imposed by the atomic spin-orbit interaction. Experimentally Rau et al. realized the maximum MAE for a 3d transition metal atom by coordinating a single Co atom to the O site of a MgO(100) surface [1].

Theoretically, simple density functional theory (DFT) calculations do not recover the large MAE of this system. Here, including a Hubbard U correction and spin-orbit coupling, we reproduce the large MAE of an individual Co adatom on a MgO (001) surface. More importantly, we take one step further by investigating the possibility of enhancing the MAE of 3d transition metal adatoms by considering various structural geometries of 3d-O molecules deposited on MgO. In one of the structures, where the molecules are perpendicular to the surface, the MAE can be enhanced while reducing the interaction with the substrate, which should minimize spin fluctuations and enhance the magnetic stability. Moreover, we evidence the ability to substantially modify the MAE by atomic control of the location of the 3d–O molecules on the substrate.

**Acknowledgements**

This work was supported by the Federal Ministry of Education and Research of Germany in the framework of the Palestinian-German Science Bridge (BMBF grant number 01DH16027).

**References**

[1] Rau et al., *Science.* **344** 988 (2014).

# Tuning the blocking temperature distribution of the AF grains by organic molecules

*Luca Gnoli[1]*, Mattia Benini[1], Alberto Riminucci[1], Rajib Kumar[1] Manju Singh[1], Alessio Mezzi[2], Ko-Wei Lin[3], Valentin Dediu[1] and Ilaria Bergenti[1]*

[1] *Institute of Nanostructured Materials ISMN-CNR Via Gobetti 101 40129 Bologna, Italy*
[2] *Institute of Nanostructured Materials ISMN-CNR Via Salaria km 29.300 00015 Monterotondo Scalo, Italy*
[2] *NCHU Department of Materials Science and Engineering, Taichung City 402, Taiwan*
*luca.gnoli@ismn.cnr.it

The interface between organic molecules and a magnetic layer has been designated as the key element in defining the the spin functionality of hybrid molecular spintronic devices. The origin of such effects is related to the formation of the so-called "Spinterface" that can affect the spin properties of both interface components as for example the generation of a spin polarization in the molecular layer or a change of the magnetic behaviour of the ferromagnetic layer (FM)[1]. Hybridization effects have been widely investigated both theoretically and experimentally considering FM layers[2] while the adsorption and coupling of molecules on surfaces with Antiferromagnetic (AFM) order has not a clear clue. In this view, because of the insensitivity of AFM to the external magnetic fields, with the aim of evidencing the effect of coupling between AFM and organics, we have considered the AFM layer also coupled to a FM featuring thus exchange bias effect.

We deposited polycrystalline thin films of Co and we formed a bilayer with a single interface between Co and CoO by exposing the sample to a controlled oxygen atmosphere ($10^4$L), generating an oxide thickness of 2.5 nm. On the top of CoO a thick molecular layer (25nm) of C60 (buckminsterfullerene) was deposited. The magnetic properties of the Co/CoO/C60 stacks were determined with a MOKE apparatus. Temperature dependent of Co hysteresis loops of the samples were acquired after cooling down to 80 K after appropriate training. The presence of the molecular layer onto CoO determines a hardening of the Co layer and an increase of the blocking temperature related to the exchange bias effect.

We interpret our data considering that the hybridization with molecular layer generates an imbalance of Energy barrier for AFM reversal in CoO[3]

**Acknowledgements**

This work is supported by European Union Horzon 2020 Research and iNnovation Programme under grant agreement n. *964396* FET-Open *SINFONIA*.

**References**

[1] I. Bergenti and V. Dediu, V, Nanomaterials science **1** (2019) 149.
[2] M. Cinchetti, V. Dediu, L.E. Hueso, Nat. Mater., **16** (2017) 507
[3]K. O'Grady et al., J. Magn. Magn. Mater., **322** (2010) 883

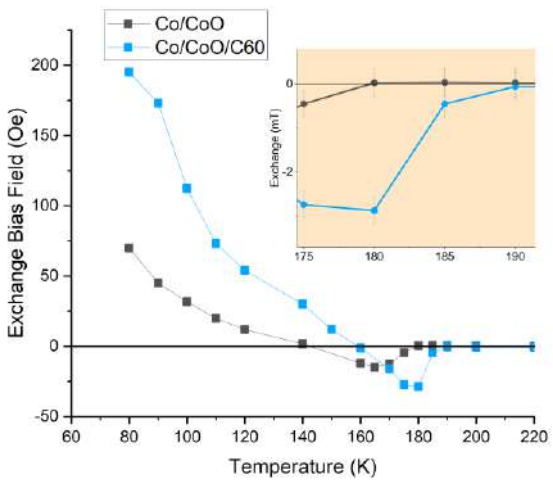



Figure: Temperature dependent exchange bias field of Co/CoO samples with and without C60 overlayer. Samples were cooled at 80K in a negative magnetic field of 750 Oe

# Electric field control of RKKY coupling through solid-state ionics

*Maria Ameziane[1], Roy Rosenkamp[1], Lukáš Flajšman[1], Sebastiaan van Dijken[1], Rhodri Mansell[1]**

[1] *NanoSpin, Department of Applied Physics, Aalto University School of Science, P.O. Box 15100, FI-00076 Aalto, Finland*

*rhodri.mansell@aalto.fi

Placing a suitable spacer layer between two magnetic layers can lead to a thickness dependent interaction between the magnetic layers known as Ruderman–Kittel–Kasuya–Yosida (RKKY) coupling. Controlling RKKY coupling through applied voltages, particularly the ability to switch between ferromagnetic and antiferromagnetic coupling, would lead to ability to create new magnetic data storage devices [1]. Here we combine a solid-state Li ion storage layer and electrolyte used in battery technology with an out-of-plane magnetized Co/Pt-based stack coupled through a Ru interlayer (Fig. 1a,b,c,d). Through electrical cycling, which introduces Li ions into the metallic stack (Fig. 1e) we investigate the effects of the insertion of Li ions on the RKKY coupling (Fig. 1f,g). The insertion of Li ions mainly reduces the amplitude of the RKKY coupling but for Ru interlayer thicknesses around ferromagnetic to antiferromagnetic crossings the amplitude can increase. This indicates that the phase of the RKKY coupling can also be controlled by voltage. This leads to the ability to switch the RKKY coupling between ferromagnetic and antiferromagnetic with applied voltages (Fig. 1h) [2], opening a path for novel forms of digital data storage devices.

**Acknowledgements**

This work was funded by the Academy of Finland under project numbers 316857 and 295269. We acknowledge the provision of facilities by Aalto University at OtaNano - Micronova Nanofabrication Centre.

**References**

[1] A. E. Kossak, *et al*., Science Advances, **9**, eadd054 (2023).

[2] M. Ameziane, *et al.* arXiv:2301.11626 (2023)

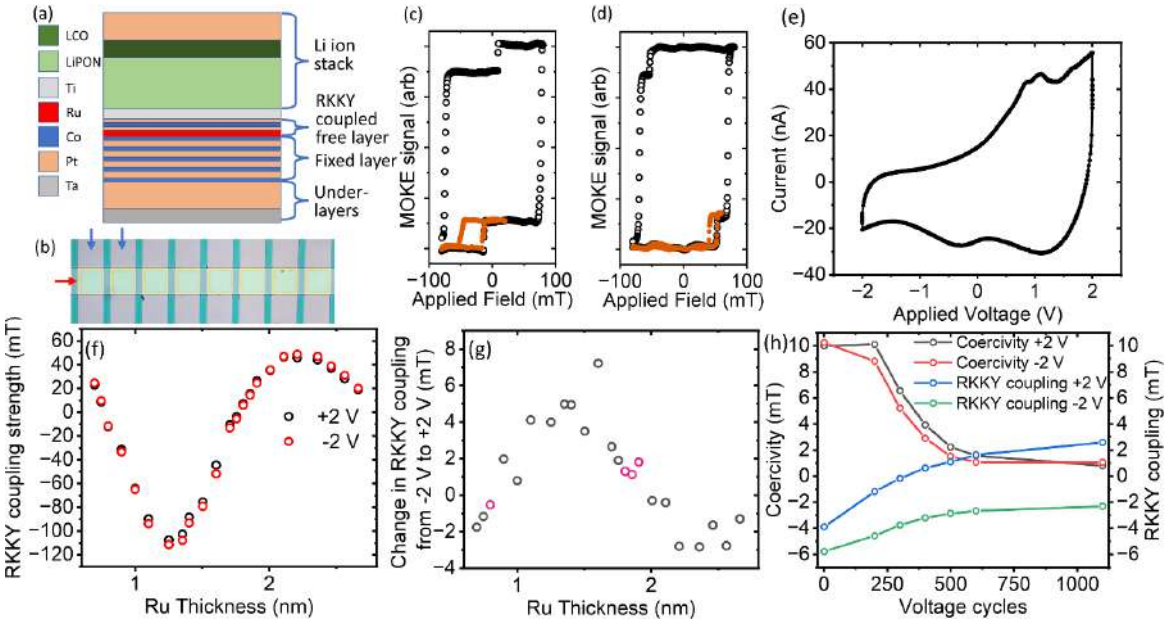



Figure 1: (a) Cross-sectional schematic of a junction. (b) Optical microscopy image of the device consisting of vertical bottom electrodes (two of which are indicated by blue arrows) and a horizontal top electrode (red arrow). The electrodes are 100 µm across. The thickness of the Ru interlayer increases from left to right. (c) Major hysteresis loop (black) and minor loop (orange) of the junction with 1.0 nm Ru at 0 V showing antiferromagnetic coupling between the free and fixed magnetic layer. (d) Major hysteresis loop (black) and minor loop (orange) of the junction with 2.4 nm Ru at 0 V showing ferromagnetic coupling. (e) Cyclic voltammogram of the junction with 1.9 nm Ru interlayer thickness taken at 50 mV/s. (f) RKKY coupling strength as a function of Ru interlayer thickness for +2 V and -2 V. (g) Difference in RKKY coupling strength for +2 V and -2 V as a function of Ru interlayer thickness. Red dots indicate junctions where the magnitude of the coupling increased with +2 V applied. (h) Changes in the coercivity and RKKY coupling of the junction with 1.75 nm Ru interlayer at -2 V and +2 V as a function of the number of voltages cycles.

# Brain-inspired magneto-ionics in transition metal nitrides

*Zhengwei Tan[1]*, Aitor Arredondo López[1], Eva Pellicer[1], Enric Menéndez [1], Jordi Sort[1,2]*

[1]*Departament de Física, Universitat Autònoma de Barcelona, E-08193 Cerdanyola del Vallès, Spain*

[2]*Institució Catalana de Recerca i Estudis Avançats (ICREA), Pg. Lluís Companys 23, E-08010 Barcelona, Spain*

*Zhengwei.Tan@uab.cat

Magneto-ionics is an emerging technology that enables precise modulation of magnetic properties through voltage-driven ion migration. This approach is of great appeal for reliable information storage and it has been recently proposed for neuromorphic computing applications due to its low-power and non-volatile operation [1,2]. Here, we demonstrate that nitrogen magneto-ionics in CoN films can mimic several neuromorphic functionalities, such as potentiation/depression, learning through spike-rate-dependent plasticity, memory retention, forgetting, and self-evolution by maturity, which refers to tunable updating of information even after voltage has been switched off, in a controllable manner. In particular, the post-stimulated behavior of magneto-ionic systems, presented as their spontaneous evolution after the removal of electric stimuli, is of great importance to ensure the non-volality of the recorded information. Yet the manipulation of this behavior is challenging. In this work, we show a voltage-controllable N ion accumulation effect on the outer surface of CoN films immersed in a liquid electrolyte, which is achieved through the voltage-driven transport of $N^{3-}$ ions with response time of $10^{-2}$ s. This ion accumulation effect causes a post-stimulated increase of magnetization (i.e., once electrical voltage has been switched off). This is only possible by properly tuning the CoN film thickness (which determines ion motion rates) and the pulse frequency. Specifically, the ion accumulation effect and subsequent increase of magnetization is observed in thin films (thickness < 50 nm) since they exhibit sufficiently high voltage-driven ion motion rates when actuated with either DC or pulsed voltage at frequencies $f$ = 1, 10 and 100 Hz. At low frequencies, relaxation effects during the "off" periods of the voltage pulses dominate over the aforementioned interfacial ion accumulation effect. Besides boosting energy efficiency, our approach offers an important additional logical function: after voltage has been applied, the device can be either programmed to learn or forget without any further energy input, thus mimicking synaptic functions under deep sleep, when processing of information can continue without any external signal input. This novel effect may open opportunities, at the materials level, for advanced brain-inspired computing paradigms [3].

## Acknowledgements

Financial support by the European Research Council (REMINDS 2021-Advanced Grant, Agreement No. 101054687), the European Union's Horizon 2020 research and innovation programme (European Training Network, BeMAGIC ETN/ITN Marie Skłodowska-Curie grant No. 861145, and the Spanish Research Agency (PID2020-116844RB-C21 and PDC2021-121276-C31) is acknowledged.

## References

[1] J. de Rojas, A. Quintana, A. Lopeandia, J. Salguero, B. Muñiz, F. Ibrahim, M. Chshiev, A. Nicolenco, M. O. Liedke, M. Butterling, A. Wagner, V. Sireus, L. Abad, C. J. Jensen, K. Liu, J. Nogués, J. L. Costa-Krämer, E. Menéndez and J. Sort, *Nat. Commun*. **11** (2020) 5871.

[2] Z. Tan, S. Martins, M. Escobar, J. De Rojas, F. Ibrahim, M. Chshiev, A. Quintana, A. Lopeandia, J. L. Costa-Krämer, E. Menéndez and J. Sort, *ACS Appl. Mater. Interfaces* **14** (2022) 44581.

[3] Z. Tan, J. De Rojas, S. Martins, A. Lopeandia, A. Quintana, M. Cialone, J. Herrero-Martín, J. Meersschaut, A. Vantomme, J. L. Costa-Krämer, J. Sort and E. Menéndez, *Mater. Horiz.* **10** (2023) 88.

# Oxygen Magneto-Ionics, without Oxidation

*T. Bhatnagar-Schöffmann[1], P. Schöffmann[2], A. Lamperti[3] , A. Kovàcs[4], D. Ourdani[5], M.-A. Syskaki[6], Y. Roussigné, S. Ono[7], J. Langer5, R. E. Dunin-Borkowski[4], D. Ravelosona[1], M. Belmeguenai3, A. Solignac[8], and L. Herrera Diez[1*]*

[1] *Centre de Nanosciences et de Nanotechnologies, CNRS-Université Paris-Saclay, Palaiseau, France*
[2] *Synchrotron SOLEIL, L'Orme des Merisiers, 91190 Saint-Aubin, France*
[3] *CNR-IMM, Agrate Unit, Via C. Olivetti 2, 20864 Agrate Brianza (MB), Italy*
[4] *Forschungszentrum Jülich GmbH, Ernst Ruska-Centre for Microscopy and Spectroscopy with Electrons and Peter Grünberg Institute (PGI-5), 52425 Jülich, Germany*
[5] *Laboratoire des Sciences des Procédés et des Matériaux, CNRS-UPR 3407, Université Sorbonne Paris Nord, 93430 Villetaneuse, France*
[6] *Singulus Technologies AG, Hanauer Landstrasse 103, 63796 Kahl am Main, Germany*
[8] *Central Research Institute of Electric Power Industry, Yokosuka, Kanagawa 240-0196, Japan*
[8] *SPEC, CEA, CNRS, Université Paris-Saclay, CEA Saclay, 91191 Gif-sur-Yvette Cedex, France*
* liza.herrera-diez@c2n.upsaclay.fr

In this study we show that a Pt dusting layer of a nominal thicknes of 0.09nm inserted at the interface between CoFeB and MgO in a Ta/CoFeB/MgO/$HfO_2$ induces in-plane magnetic anisotropy in the stack, which can be switched to perpendicular magnetic anisotropy (PMA) through magneto-ionic gating (see figure). The polycristalline structure of the MgO layer observed by transmission electron microscopy in the as-grown state is preserved after gating [1], indicating a potential magneto-ionic mechanism mediated by oxygen motion through grain boundaries.

Interestingly, this system does not show evidence of the typical oxidation involved in magneto-ionics relying on voltage-induced migration of oxygen species [2]. XAS measurements show a strong and reversible change in the oxygen edge upon gate voltage exposure, showing that the magneto-ionic process is mediated by the incorporation and release of oxygen species. However, XMCD measurements show that the voltage-gate induced spin-reorientation transition in the CoFeB/Pt/MgO stacks is not accompanied by a significant change in the magnetic moment of neither Fe nor Co.

This magneto-ionic mechanism is attributed on the one hand to the crystallinity of the MgO barrier, and on a likely change in the oxidation potential of the Co and Fe atoms at the oxide interface through the interaction with the Pt atoms in the dusting layer. This shows that the fine-tuning of the chemistry at the interfaces of magneto-ionic stacks is important for the design of magneto-ionics materials and devices.

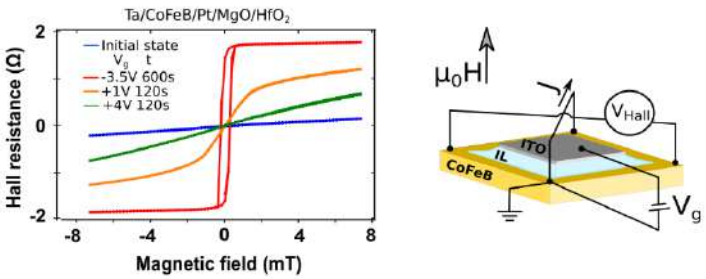



Figure: Anomalous Hall effect signal at different ionic-liquid gate voltages in Ta/CoFeB/Pt/MgO/HfO2 (left), and a cartoon of the device geometry (right).

## Acknowledgements

We acknowledge financial support from the European Union H2020 Program: grant MSCA ITN 860060 and Grant No. 823717, project "ESTEEM3" for access to TEM experiments. We also thank the French National Research Agency (JCJC SplaSy and Labex NanoSaclay, reference: ANR-10-LABX-0035).

## References

[1] T. Bhatnagar-Schöffmann, et al., *Appl. Phys. Lett.* 122, 042402 (2023).
[2] L. Herrera Diez et al. *Phys Rev. Appl.* 12, 034005 (2019).

# Stimuli-responsive van der Waals heterostructures triggered by a spin crossover metal-organic framework

*S. Mañas-Valero,*[1,2] *C. Boix-Constant,*[1] *Víctor García-López*[1], *E. Navarro-Moratalla,*[1] *M. Clemente-León,*[1] *F. Guinea,*[3] *E. Coronado*[1]

[1]*Instituto de Ciencia Molecular (ICMol), Universitat de València, Paterna, 46980, Spain.*

[2] *Kavli Institute of Nanoscience, Delft University of Technology, 2628 CJ Delft, The Netherlands.*

[3] *Instituto Madrileño de Estudios Avanzados en Nanociencia (IMDEA - Nanociencia), Madrid, 28049, Spain.*

*samuel.manas@uv.es, S.ManasValero@tudelft.nl

Van der Waals heterostructures (vdWHs) provide the possibility of engineering new materials with emergent functionalities that are not accessible in another way. These heterostructures are formed by assembling layers of different materials used as building blocks. Beyond inorganic 2D crystals, layered molecular materials remain still rather unexplored. In this work,1 the family of van der Waals heterostructures is enlarged by introducing a molecular building block able to produce strain: the spin-crossover (SCO). In these metal–organic materials, a spin transition can be induced by applying external stimuli. In particular, smart vdWHs are prepared in which the electronic and optical properties of the 2D material (graphene, $WSe_2$) are switched by the strain caused by the spin transition. These hybrid vdWHs represent the deterministic incorporation of bistable molecular layers with other 2D crystals in the emergent fields of straintronics and band engineering in low-dimensional materials.

## References

[1] Adv. Mater. 34, 11, 2110027 (2022).

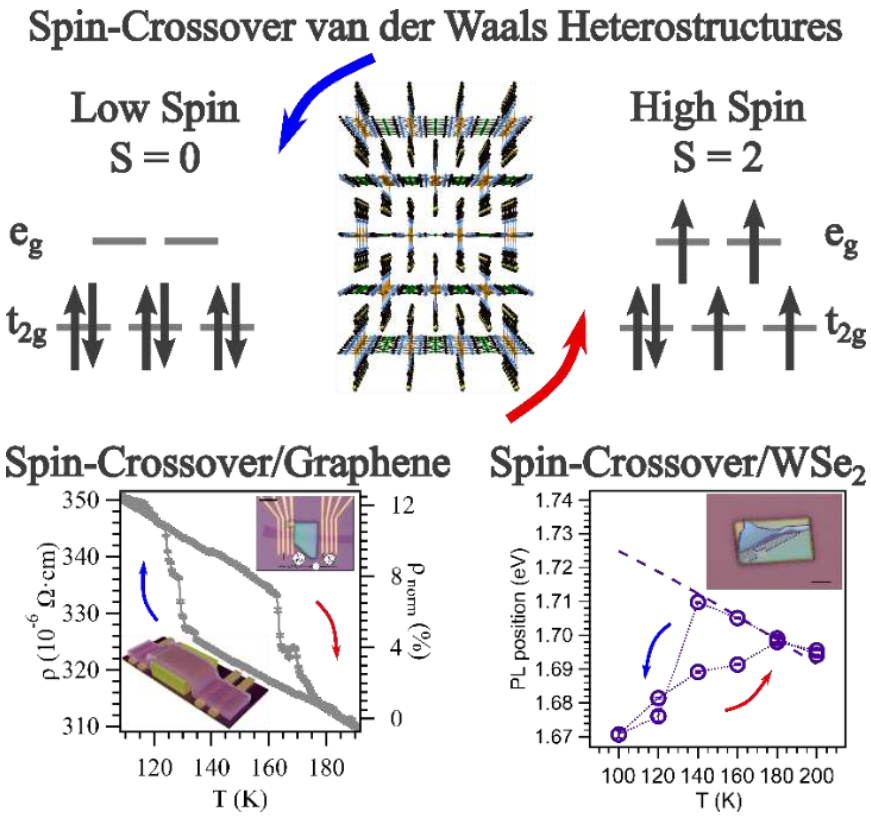



Figure: Upper figure: Scheme of the spin-crossover phenomenon for a $d^6$ ($Fe^{2+}$) metal complex. Bottom left: Thermal dependence of the resistivity for a vdWH. Bottom right: Thermal dependence of the PL position for the vdWH and a reference $WSe_2$ monolayer.

# Engineering of a 2D Metal-organic Network Featuring a Large Unquenched Orbital Magnetic Moment

*Sofia O. Parreiras[1]*, Cristina Martín-Fuentes[1], José I. Urgel[1], Victor Rubio-Giménez[2], Beatriz Muñiz-Cano[1], Daniel Moreno[1], Koen Lauwaet[1], Manuel Valvidares[3], Miguel A. Valbuena[1], Pierluigi Gargiani[3], Wolfgang Kuch,[4] Julio Camarero[1,5], José M. Gallego[1,6], Rodolfo Miranda[1,5], José I. Martínez[6], Carlos Martí-Gastaldo[2], David Écija[1]*

[1] *IMDEA Nanoscience, Madrid, Spain*
[2] *ICMol, Universitat de València, Paterna, Spain*
[3] *ALBA Synchrotron Light Source, Barcelona, Spain*
[4] *Institut für Experimentalphysik, Freie Universität, Berlin, Germany*
[5] *IFIMAC, Universidad Autónoma de Madrid, Madrid, Spain*
[6] *ICMM-CSIC, Madrid, Spain*

*sofia.oliveira@imdea.org

Free transition metal atoms have high spin and orbital magnetic moments. However, in solids, the orbital moment is partial or totally quenched by crystal field effects [1]. This quenching reduces drastically the magnetic anisotropy, that is associated to the orbital moment anisotropy. The reduction of the coordination of transition metals has been proposed as a route to achieve an unquenched orbital moment [1,2]. 2D metal-organic networks on surfaces are promising candidates to reduce the coordination and stabilize a large orbital moment.

On the other side, π-conjugated metal-organic networks have been attracting great attention since they can present exotic quantum phases of matter [3,4]. The conjugation also enhances the coupling between magnetic moments and can lead to antiferromagnetic ground-states [5]. The absence of stray fields and the magnetic robustness of antiferromagnetic materials make them promising candidates to replace ferromagnets in the next generation of spintronic devices [6].

In this work, we have investigated the structural, electronic and magnetic properties of a π-conjugated 2D metal-organic network consisting of 2,3,6,7,10,11-hexahydroxytriphenylene (HOTP) molecules coordinated with Co atoms on Au(111). We have performed a multidisciplinary study, combining scanning probe microscopy and spectroscopy, X-ray absorption spectroscopy, X-ray linear and X-ray circular magnetic dichroism [7]. Our results reveal a network based on three-fold $Co^{+2}$ coordination displaying a large unquenched orbital magnetic moment with an orbital to effective spin moment ratio of 0.8, as also a large magnetic anisotropy with an in-plane easy axis. Furthermore, density functional theory complemented by a Hubbard model (DFT+U) predicts an antiferromagnetic ground-state, that is compatible with our experimental results.

## References

[1] R. A. Guirado-López, et al., *Phys. Rev. Lett*. **90** (2003) 226402.
[2] P. Gambardella et al., *Science* **300** (2003) 1130-1133.
[3] X. Huang et al., *Angew. Chem. Int. Ed*. **57** (2018) 146.
[4] L. Dong et al., *Phys. Rev. Lett*. **116** (2016) 096601.
[5] M. Hua et al., *J. Phys. Chem. Lett*. **12** (2021) 3733.
[6] T. Jungwirth et al., *Nat. Nanotechnol*. **11** (2016) 231.
[7] C. Martín-Fuentes et al., *J. Am. Chem. Soc*. **144** (2022) 16034.

# Multiferroic Metal-Organic Heterostructures with Memristive Properties

*Nicusor Iacob[1], Cristina Chirila[1], Anda E. Stanciu[1], Andrei Kuncser[1], Marcela Socol[1], Catalin C. Negrila[1], Lucian Trupina[1], Cristina Bartha[1], Claudiu Locovei[1,2], Gabriel Schinteie[1], Aurelian C. Galca[1], Lucian Pintilie[1], Victor Kuncser[1], Bogdana Borca[1,*]*

[1] *National Institute of Materials Physics, Atomistilor 405A, 077125 Magurele, Ilfov, Romania*
[2] *Faculty of Physics, University of Bucharest, Atomistilor 405, 077125, Magurele,Ilfov, Romania*
*bogdana.borca@infim.ro

The memristive properties of multiferroic tunnel junctions are determined by the possibility to modulate the junction's electrical resistance by independently controlling the magnetization orientation of the ferromagnetic electrodes and the ferroelectric polarization of the tunnel barrier [1]. The coupling of these magneto-electric properties is important for electronic applications with low power consumption from nonvolative memory, to elements in logic circuits, sensing devices, biological synapses models in the emerging area of neuromorphic computing and artificial intelligence. Realizing these multifunctional electronic elements using organic materials and/or biomolecules is presenting various advantages related to their low cost, versatile synthesis, flexibility, lightweight and biocompatibility that are actually of major scientific and technological interest for applications in the fields of spintronics, molecular electronics, transient electronics, bioelectronics, and therapeutic techniques. Moreover, the lightweight elements in their composition have a small spin-orbit coupling and thus, favour a longer spin lifetime and spin transport length of the charge carriers than in standard inorganic materials.

Herein, we demonstrate the non-volatile memristive spintronic behavior of multiferroic junctions that can be manipulated with small external magnetic and electric fields. These memristors have two terminals and are composed of organic biomolecular ferroelectric films [2] of a DNA component, the guanine nucleobase, sandwiched between two different ferromagnetic electrodes of Co and $Co_xCr_{1-x}$ films that have an in-plane easy axes of magnetization and different coercive fields. At 100 K a difference of about 370% is achieved between the magnetoresistive hysteresis loops in the low-resistance and the high-resistance states (Figure). The guanine film ensures a very long spin transport length of up to 200 nm. The guanine films show typical ferroelectric polarization–electric-field hysteresis loops with large electrical polarization and pyroelectric signal at low temperatures of up to 200 K. Above this transition temperature, at which different properties are affected, the guanine films have a preponderant paraelectric phase containing residual or locally induced nanoscopic ferroelectric domains, as observed by piezoresponse force microscopy at room temperature.

## Acknowledgements

The authors thank the Core Program of the National Institute of Materials Physics under the Project PC2-PN23080202 and the PN-III-P2-2.1-PED-2021-0378 (contract no. 575PED/2022) project founded by the Romanian Ministry of Research, Innovation and Digitalization.

## References

[1] D. Pantel, S. Goetze, D. Hesse, M. Alexe, *Nature Mater* **11** (2012) 289.
[2] M. Socol, L. Trupina, A.-C. Galca, C. Chirila, G. E Stan, A.-M. Vlaicu, A. E. Stanciu, A. G. Boni, M. Botea, A. Stanculescu, L. Pintilie, B. Borca, *Nanotechnology* **32** (2021) 415702.

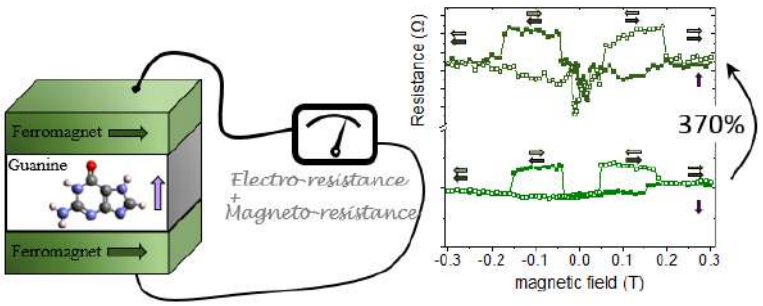



Figure: Metal-organic memristive multiferroic junctions.

# Magnetic Properties Control in Layered Hybrid Organic-inorganic Metal-halide Perovskites by Composition and Perovskite Phase Selection

*Yaiza Asensio[a]*, Sergio Marras,[b] Davide Spirito,[c] Marco Gobbi,[a,d,e] Mihail Ipatov,[f] Fèlix Casanova,[a,e] Aurelio Mateo-Alonso,[g,e] Luis E. Hueso[a,e] and Beatriz Martín-García[a,e]*

*[a]CIC nanoGUNE BRTA, Tolosa Hiribidea 76, 20018, Donostia-San Sebastián, Spain*

*[b]Istituto Italiano di Tecnologia - Materials Characterization Facility, Genova 16163, Italy*

*[c]IHP–Leibniz-Institut für innovative Mikroelektronik, Im Technologiepark 25, 15236 Frankfurt (Oder), Germany*

*[d]Materials Physics Center CSIC-UPV/EHU, 20018 Donostia-San Sebastián, Spain*

*[e]IKERBASQUE, Basque Foundation for Science, 48009 Bilbao, Spain*

*[f] SGIker Medidas Magnéticas Gipuzkoa, UPV/EHU, 20018 Donostia-San Sebastián, Spain*

*[g]POLYMAT, University of the Basque Country UPV/EHU, Avenida de Tolosa 72, 20018 Donostia-San Sebastián, Spain*

* y.asensio@nanogune.eu

Research interest in magnetic layered (2D) materials has grown significantly since the isolation of intrinsically magnetic monolayers, leading to the expansion of the library of magnetic 2D materials. Achieving control over the magnetic properties of these materials is crucial for their successful integration into devices.[1]. In this regard, layered hybrid organic-inorganic metal halide perovskites (HOIPs) are an ideal platform for magnetic tunability due to their chemical and structural versatility[2-3]. In this work, we conducted a comprehensive study to understand how the transition metal ($Cu^{2+}$, $Mn^{2+}$ and $Co^{2+}$), organic spacer (alkyl- and aryl-ammonium), and perovskite phase (Ruddlesden-Popper and Dion-Jacobson) impact on their magnetic properties. Our results show that an increase in the in-plane anisotropy together with a decrease in the interlayer distance of $Cu^{2+}$ HOIPs lead to a change in their magnetic behavior from a 2D ferromagnet to a quasi-3D antiferromagnet (Figure 1). On the other hand, the magnetism of $Mn^{2+}$ HOIPs is characterized intrinsically by antiferromagnetic intralayer interactions. Finally, $Co^{2+}$ crystals with a non-perovskite structure display a dominant paramagnetic behavior. Therefore, our work demonstrates the potential of using HOIPs for developing new layered magnetic materials with tailored magnetic properties[4].

## Acknowledgements

Funding from Spanish MCIN/AEI: Projects PID2019-108153GA-I00, RTI2018-094861-B-I00, PID2021-128004NB-C21, and under the María de Maeztu Units of Excellence Programme (CEX2020-001038-M).

## References

[1] S. Zhang, R. Xu, N. Luo, X. Zou, *Nanoscale*, **13**, 1398-1424 (2021).
[2] A. Privitera, M. Righetto, F. Cacialli, M. K. Riede, *Advanced Optical Materials*, **9**, 2100215 (2021).
[3] M. Bochalya, G. V. Prakash, S. Kumar, *Journal of Solid State Chemistry*, **273**, 32-36 (2019).
[4] Y. Asensio, S. Marras, D. Spirito, M. Gobbi, M. Ipatov, F. Casanova, A. Mateo-Alonso, L. E. Hueso, B. Martín-García, *Advanced Functional Materials*, 2207988 (2022).

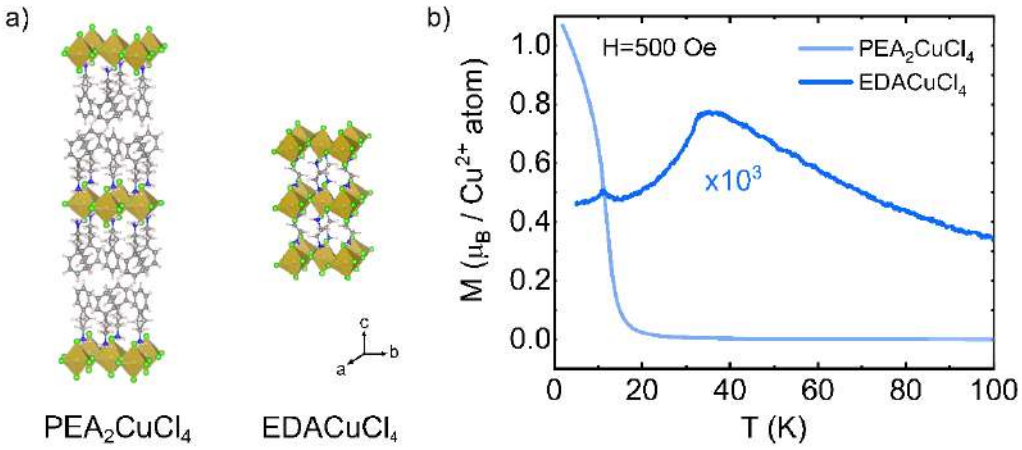



Figure 1: a) Crystal structures of $PEA_2CuCl_4$ (CCDC 751 958) and $EDACuCl_4$ (CCDC 1 148 696) drawn using VESTA software and b) their magnetization (M) versus temperature (T) parallel to the $[CuCl_6]^{4-}$ octahedra layers.

# SYMPOSIUM 08.
## MAGNETISM IN MOLECULAR, IONIC, ORGANIC BASED SYSTEMS.
## S8. POSTERS

# Magneto-elastic Interaction in κ-[(BEDT-TTF)$_{1-x}$(BEDT-STF)$_x$]$_2$Cu$_2$(CN)$_3$ Quantum Spin Liquid Candidate

*Petr Doležal[1,2]*, Rikumaru Saito[3], Atsushi Kawamoto[3], Andrej Pustogow[2]*

[1] *Department of Condensed Matter Physics, Faculty of Mathematics and Physics, Charles University, Ke Karlovu 5, 121 16 Prague, The Czech Republic*

[2] *Institute of Solid State Physics, Technical University Vienna, Wiedner Hauptstraße 8-10, 1040 Vienna, Austria*

[3] *Department of Physics, Graduate School of Science, Hokkaido University, Kita 10, Nisi 8, Kita-ku, 060-0810 Sapporo, Japan*

* Petr.Dolezal@karlov.mff.cuni.cz

κ-[(BEDT-TTF)$_{1-x}$(BEDT-STF)$_x$]$_2$Cu$_2$(CN)$_3$ series are 2D organic salts realizing a paradigmatic Mott-metal insulator transition. The localization of conduction electrons is driven by strong electronic correlations which can be tuned by hydrostatic pressure or by substitution with BEDT-STF cations as in the present study. The parent κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ compound remains without magnetic ordering down to the lowest temperatures. To fully understand its magnetic properties it is necessary to assess its crystal structure in detail. The (BEDT-TTF) cations are separated by the layers of Cu$_2$(CN)$_3$ anions within in the ***bc*** plane, see figure below. If one looks on the cations along ***a*** crystallographic axis it is possible to see the triangular arrangement of (BEDT-TTF)$_2$ dimers, see panel (b) below. The triangular lattice is close to ideal, $t/t' = 0.83$, providing ideal conditions for geometrical frustration [1]. Each dimer is carrying one electron with spin S = ½. The interaction between spins is antiferromagnetic [2]. These all are necessary conditions for the formation of quantum spin liquid (QSL) sate and also the reason why this compound was the prime QSL candidate. The observation of $T^* = 6$ K anomaly in thermal expansion brought the question about the proper ground state, which seems to be rather a valence bond solid (VBS) than QSL [3]. This leads to the more general question whether geometrical frustration on its own can fully suppress antiferromagnetic order and whether the systems can truly remain stable towards magneto-structural instabilities, like VBS phases, down to absolute zero.

Our presented contribution is focused on low temperature X-ray diffraction study of a single crystalline sample κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$, showing pronounced anomaly of lattice parameters around $T^* = 6$ K. The structural results are supplemented by a microscopic study of quadrupolar interaction using nuclear magnetic resonance (NMR). It also maps the evolution of the $T^*$ anomaly across the series.

## Acknowledgements

This work was supported by the Czech Science Foundation under Grant No. 23-06810O.

## References

[1] H. C. Kandpal, I. Opahle, Y.-Z. Zhang et al., *Phys. Rev. Lett.* **103** (2009) 67004.
[2] Y. Shimizu, K. Miyagawa, K. Kanoda et al., *Phys. Rev. Lett.* **91** (2003) 107001.
[3] B. Miksch, A. Pustogow, M. J. Rahim et al., *Science* **372** (2021) 276-279.
[4] A. Pustogow, Solids **3** (2022) 93-110.

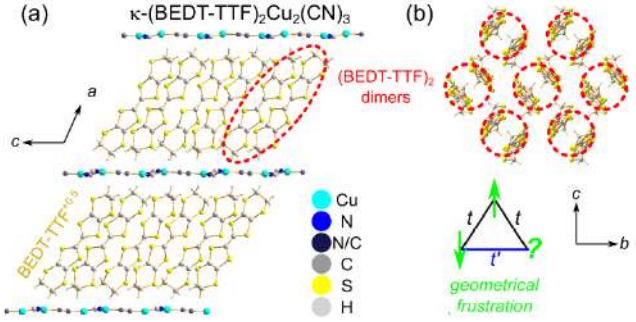



Figure: Crystal structure of κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ 2D organic salt. a) View along ***b*** axis. b) The arrangement of dimers within ***bc*** plane. Taken from [4].

# Charge Transfer-Induced Spin Transition in Fe/Co Prussian Blue Analogues in Ultrafast Regime

*Serhiy Demeshko*[1*], *Jennifer Zimara*[2], *Hendrik Stevens*[1], *Rainer Oswald*[3], *Sebastian Dechert*[1], *Ricardo A. Mata*[3], *Franc Meyer*[1], *Dirk Schwarzer*[2]

[1]*University of Göttingen, Institute for Inorganic Chemistry, D-37077 Göttingen, Germany*
[2]*Max-Planck-Institute for Biophysical Chemistry, D-37077 Göttingen, Germany*
[3]*University of Göttingen, Institute for Physical Chemistry, D-37077 Göttingen, Germany*
*sdemesc@gwdg.de

Fe/Co Prussian Blue analogues (PBA) are known for their switchable spin and redox states, making them particularly interesting for information processing at a molecular level and as nanoscale electronic components. An attractive feature of some Fe/Co PBA compounds is their photomagnetic behavior where intramolecular metal-to-metal electron transfer is coupled to a spin-crossover (SCO) process at the cobalt ion, viz., a cyanide-bridged diamagnetic $Fe^{II}_{LS}$-(μ-CN)-$Co^{III}_{LS}$ entity transforms into a paramagnetic $Fe^{III}_{LS}$-(μ-CN)-$Co^{II}_{HS}$ state. This phenomenon has been termed charge transfer induced spin transitions (CTIST) or electron-transfer-coupled spin transition (ETCST).[1]
In this work, we investigated the ultrafast dynamics following intervalence charge transfer (IVCT) excitation of the known[2] square-type molecular $Fe_2Co_2$ PBA **1**, in both its high and low temperature phases, and of the new dinuclear FeCo PBA **2**, using pump−probe transient IR and UV/vis absorption spectroscopy of the complexes in MeCN solution. The results indicate that the photoinduced one-electron transfer of **1** is restricted to one edge of the $Fe_2Co_2$ square and does not trigger a second (cooperative) charge transfer step at the opposite edge. Complex **2** and the high temperature form of PBA **1** show similar photophysical behaviour after IVCT excitation. Different relaxation mechanisms dependent on excitation wave lengths are discussed.[3]

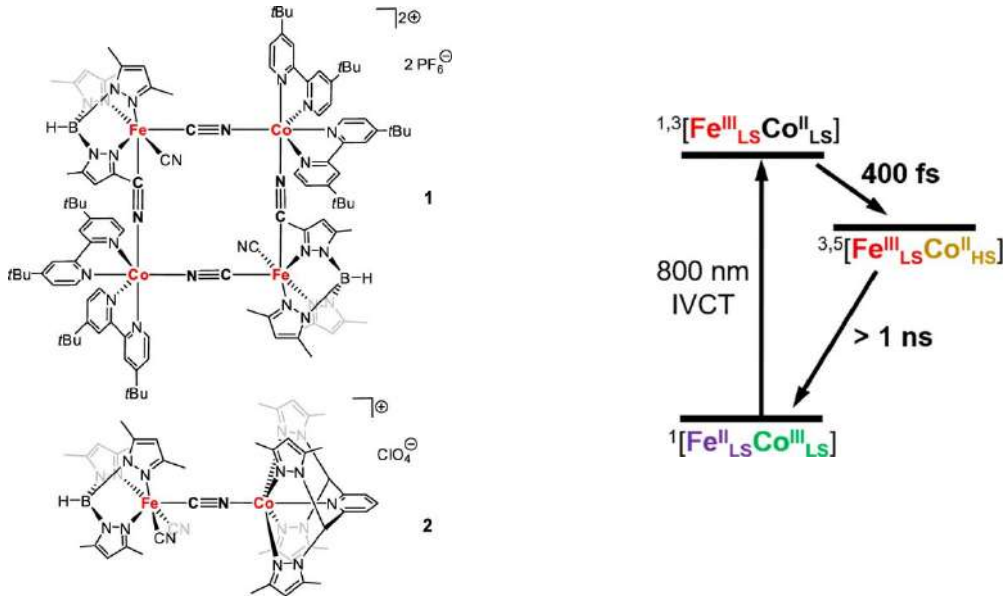



**Figure 1:** Complexes **1** and **2** investigated in this work (left) and light-induced dynamics in one Fe−Co pair of the low temperature phase of square PBA **1**.[3]

## References:

[1] N. Shimamoto, S. Ohkoshi, O. Sato, K. Hashimoto *Inorg. Chem.* **41** (2002) 678.
[2] M. Nihei, Y. Sekine, N. Suganami, K. Nakazawa, A. Nakao, H. Nakao, Y. Murakami, H. Oshio *J. Am. Chem. Soc.* **133** (2011) 3592.
[3] J. Zimara, H. Stevens, R. Oswald, S. Demeshko, S. Dechert, R. A. Mata, F. Meyer, and D. Schwarzer *Inorg. Chem.* **60** (2021) 449.

# Mechanoelastic Simulations of Monolayer Lattices of Spin Crossover Molecules on a Substrate

*Anastasia Railean[1*], Cristian Enachescu[1], Laurentiu Stoleriu[1], Amandine Bellec[2], Vincent Repain[2], Marie-Laure Boillot[3]*

[1] *Faculty of Physics, Alexandru Ioan Cuza University, Iasi 700506, Romania*
[2] *Université Paris Cité, CNRS, Laboratoire Matériaux et Phénomènes Quantiques, F-75013 Paris, France*
[3] *Institut de Chimie Moléculaire et des Matériaux d'Orsay, Université Paris-Saclay, CNRS, UMR 8182, 91400 Orsay, France*
* anastasia.railean@student.uaic.ro

Here we discuss in the framework of a mechanoelastic model [1] the electronic and mechanical behaviour of layers of spin crossover (SC) molecules self-organized on a substrate. We consider the molecules situated in a face centred cubic structure interacting in between and with sites in the substrate by the way of connecting springs with given elastic constants (Fig. 1a).

The main experimental results are reproduced, such as the typical incompleteness of the hysteresis loop, residual fractions after low temperature relaxations, cooperativity or kinetic features [2]. However, we prove that the simple model, implying fixed neighbours on the substrate for every SC molecule, leads in some cases to unphysical situations, corresponding to unexpected large curvatures of the SC layer (Fig. 1b).

Therefore, in a further approach we let every SC molecule to change its neighbour sites in the substrate, by connecting at every moment to the closest molecules on the substrate. This approach, corroborated with the use of different densities of the sites on the substrate, allows us to simulate further experimental observations, such as the appearance of cracks inside the layer (Fig. 1c) or periodic arrangements of heights of SC molecules on the layer leading to moiré patterns (Fig. 1d), for which corresponding experimental data are also provided [3].

## References

[1] L. Stoleriu, M. Nishino, S. Miyashita, A. Stancu, A. Hauser, C. Enachescu, *Phys. Rev. B* 96, 064115 (2017) ; A. I. Popa, L. Stoleriu, C. Enachescu, *J. Appl. Phys.* 129, 131101 (2021)
[2] C. Fourmental et al., *J. Phys. Chem. Lett.* 10, 4103 (2019) ; M. Kelai et al., *J. Phys. Chem. Lett.* 12, 6152 (2021) ;
[3] A. Railean, M. Kelai, A. Bellec, V. Repain, M.-L. Boillot, T. Mallah, L. Stoleriu, C. Enachescu, *Phys. Rev. B* 107, 014304 (2023)

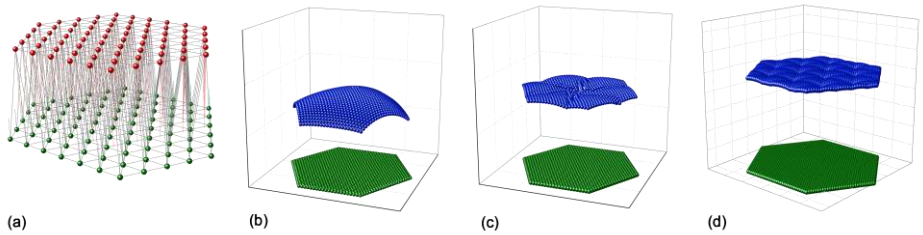



**Figure 1:** (a) First layer of SC molecules (red circles) connected to substrate molecules (green circles). (b) A system showing large distortions in its full LS state after relaxation in the case of fixed links between SC molecules (blue circle) and the substrate. (c) A system with large local distortions leading to cracks in the case of flexible links (self-adjustable) interactions between SC molecules (blue circles) and the substrate. (d) A system showing periodic arrangements of heights of SC molecules in the case of self-adjustable interactions with a larger density of substrate sites.

# Reversible magneto-Ionic effect in crystallized W-CoFeB-MgO-HfO2 ultra-thin films with perpendicular anisotropy

*Song Chen[1], Elmer Monteblanco[1], Rohit Pachat[2], Liza Herrera Diez[2], Dafiné Ravelosona[1,2]*

*[1]Spin-Ion Technologies, 10 Boulevard Thomas Gobert, 91120 Palaiseau, France*
*[2]Centre de Nanosciences et de Nanotechnologies, CNRS, Université Paris-Saclay, 10 Boulevard Thomas Gobert, 91120 Palaiseau, France*

**Abstract**

Voltage control of magnetic properties such as interface magneto-crystalline anisotropy have been widely studied in ferromagnetic metal (FM)/metal oxide (MO) heterostructures due to their potential to reduce power consumption in spintronic devices. The underlying physical phenomena is based on several magneto-electric mechanisms, including carrier-meditated electrostatic effect[1] or magneto-ionics response[2]. The later effect where the electric field can modify the oxidation state of the FM material, has drawn intense interest recently since it provides non-volatility and much larger change of magnetic properties with respect to pure charge effects. In particular, the influence of the FM/oxide interface state on the MI effect have been widely studied showing complex behavior in term of modulation of magnetic properties, reversibility and non-volatility.

Here, we have investigated electric field induced modulation of perpendicular magnetic anisotropy (PMA) in both amorphous and crystalline W/CoFeB/MgO/HfO2 ultra-thin films. We find that in the amorphous state, the electric field effect is volatile and reversible, which is consistent with the conventional electrostatic effect through charge accumulation. In the crystallized system annealed at 370°C, we find that in addition to the electrostatic response the voltage induced effect on PMA is non-volatile and reversible. We discuss these results in terms of higher oxygen mobility at the crystallized CoFeB-MgO interface, which induces a non-volatile magneto-ionic response. Modulating PMA in crystallized W-CoFeB-MgO materials through ionic migration opens the path to integrating magneto-ionics in full magnetic tunnel junctions.

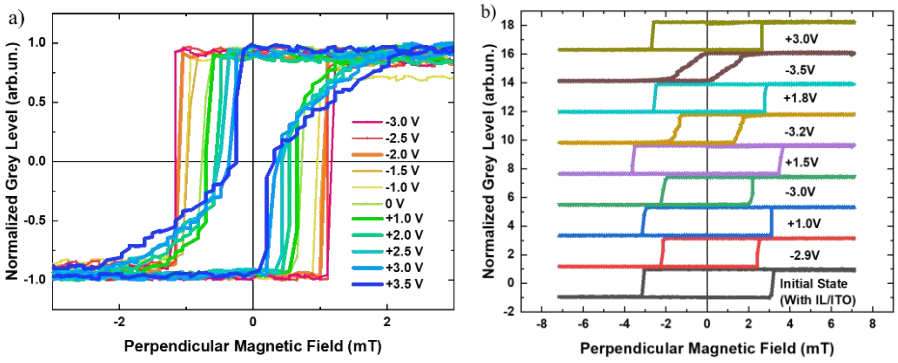



***Figure 1:*** *Hysteresis loop under perpendicular field for different voltages in (a) non annealed and (b) annealed W (5nm)-CoFeB (1nm)-MgO (2nm)-HfO2 (1nm) structures. An ionic liquid) [EMI][TFSI] has been used to provide large E-fields with a counter ITO electrode. In the amorphous (crystalline) structure, the E-Field is volatile (non-volatile) respectively.*

**References**

[1] Weisheit M, Fähler S, Marty A, Souche Y, Poinsignon C and Givord D 2007 Electric Field-Induced Modification of Magnetism in Thin-Film Ferromagnets *Science* 315 349–51
[2] Bauer U, Yao L, Tan A J, Agrawal P, Emori S, Tuller H L, Dijken S van and Beach G S D 2015 Magneto-ionic control of interfacial magnetism *Nat Mater* 14 174–81

# Simplifying the First Order Reversal Curves method for molecular magnets by using a calorimetric approach

*Diana Plesca[1*], Cristian Enachescu[1], Radu Tanasa[1], Alexandru Stancu[1], Denis Morineau[2], Marie-Laure Boillot[3]*

*[1]Faculty of Physics, "Alexandru Ioan Cuza" University, 700506, Iasi, Romania;*

*[2]Institut de Physique de Rennes, CNRS, Université de Rennes 1, UMR 6251, 35042 Rennes, France*

*[3]Institut de Chimie Moléculaire et des Matériaux d'Orsay, Université Paris-Saclay, CNRS, 8182, Orsay, France;*

*[*]Correspondence: diana.plesca@student.uaic.ro*

The First-Order Reversal Curve (FORC) diagram method is one of the most successful characterization techniques for complex hysteretic phenomena. The FORCs are a specific class of minor hysteresis loops, for which the sweep of the input parameter is reversed once starting from specific points on the major hysteresis loop [1]. The main problem in the numerical calculation of the FORC diagrams is that the second-order derivative of a function with discrete noise-contaminated data increases the noise that is inevitably present in the measurements [2].

In order to minimize the drawbacks of the standard method for obtaining FORC distribution, we propose here a method based on the calorimetric measurements: as the heat capacity already corresponds to the first derivative of the magnetization with the temperature, only a single derivative is necessary in order to obtain the FORC distributions [1]. In this way, the calorimetric method provides a unique alternative to the classical FORC diagram approach.

As model compounds to check the applicability of this method, we have considered the spin crossover molecular magnets, which are bistable systems with two electronic states in thermodynamic competition: the diamagnetic low spin state (LS), stable at low temperatures and the paramagnetic high spin state (HS), stable at high temperatures. Specific experiments have been performed on the coordination polymer $[Fe(btr)_2(NCS)_2]\cdot H_2O$ (btr=4,4'-bis(1,2,4-triazole)], which presents a large thermal hysteresis loop centered around 130 K [3].

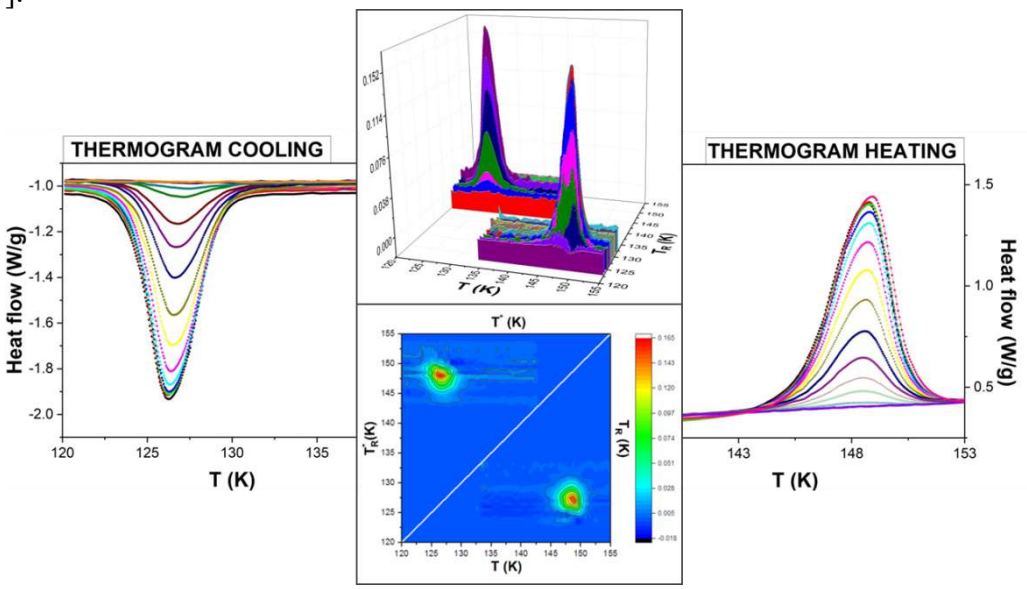



Figure: FORC thermograms for $Fe(btr)_2(NCS)_2\cdot H_2O$ microparticles in the cooling and heating mode for calorimetric measurements and corresponding diagrams.

## References

[1] R. Tanasa, C. Enachescu, J. Laisney, D. Morineau and A. Stancu, *J. Phys. Chem*. C, **123** (2019) 10120−10129.

[2] D. Cimpoesu, I. Dumitru and A. Stancu, *J. Appl. Phys*., **125** (2019) 023906.

[3] J. Martin, J. Zarembowitch, A. Dwerkin, J.G. Haasnoot and E. Codjovi, *Inorg. Chem.*, **33** (1994) 2617-2623.

# Plasma Treatment - The Unconventional Method of Tailoring Magnetic Properties in Molecular Magnets

*Dominik Czernia[1]*, Piotr Konieczny[1], Dawid Pinkowicz[2]*

[1] *Institute of Nuclear Physics PAN, ul. Radzikowskiego 152, 31-342 Cracow, Poland*

[2] *Jagiellonian University, Faculty of Chemistry, ul. Gronostajowa 2, 30-087 Cracow Poland*

*dominik.czernia@ifj.edu.pl

Molecular magnets are a class of metal-organic materials exhibiting unique physical properties that can potentially play an essential role in developing modern technology, including spintronics, quantum computing, magnetic cooling, or bistable magnetic switches. On the one hand, one of the most apparent approaches to discovering new molecular compounds is a consistent synthesis that involves various building blocks. On the other hand, molecular magnets are often sensitive to external stimuli such as temperature, pressure, light, or the presence of guest molecules, which opens new ways of modification of magnetic properties.

Here, we report results obtained for the three-dimensional molecular magnet **$NbMn_2$** based on the Nb (IV) and Mn(II) ions subjected to plasma irradiation under various conditions. The antiferromagnetic exchange interaction between Nb(IV) and Mn(II) ions leads to the long-range ferrimagnetic order observed below $T_C$ = 47.5 K and confirmed by the saturation magnetization of 9 Bohr magnetrons per **$NbMn_2$** formula unit (for two spins $S$ = 1/2 and one $S$ = 5/2).

Numerous samples of **$NbMn_2$** were ground down and scattered on the adhesive tape to ensure a large surface-to-volume ratio of exposed crystallites. Then, the samples were irradiated with plasma made of air, nitrogen, oxygen, and argon. Two other parameters were tested: plasma power (low, medium, high) and irradiation time (2 – 30 min). The magnetic properties of the irradiated samples of **$NbMn_2$** show a clear appearance of the second magnetic phase with $T_C$ around 70 K. However, the changes in magnetic signal are observed only for samples irradiated with at least 10 minutes and medium power, indicating the presence of threshold effects. The most extensive modification of the magnetic properties was observed for argon, then oxygen, nitrogen, and air (Fig. 1).

X-ray powder diffraction studies revealed that the samples after plasma irradiation have the same diffraction pattern but with broader peaks that are shifted towards higher 2θ angles. It indicates that crystal lattice parameters become smaller. Therefore, the distance between magnetic centers is reduced, and thus, exchange interactions between Nb(IV) and Mn(II) may be enhanced, explaining the appearance of the second magnetic phase with higher Curie temperature. The most probable cause of structural changes is related to the dehydration process of $H_2O$ coordination and crystallization molecules present in the system.

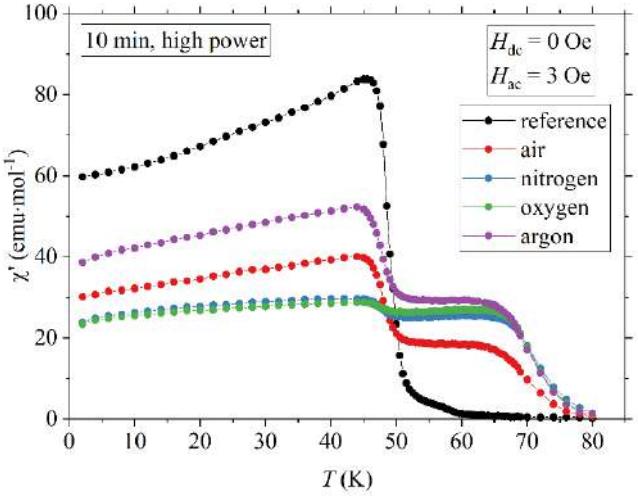



Figure 1. The real part of the AC magnetic susceptibility of **$NbMn_2$** measured for samples irradiated for 10 minutes with high power air, nitrogen, oxygen, and argon plasma.

# Magnetic and electronic properties of hybrid metal–organic interfaces on rare earth-gold surface compounds

L. Fernández[1], R. Castrillo-Bodero[1], M. Blanco-Rey[2,3], K. Ali[1], J. E. Ortega,[1,2,3], F. Schiller[1,3]
[1]Centro de Física de Materiales CSIC-UPV/EHU, 20018 San Sebastián, Spain
[2] Universidad del País Vasco UPV/EHU, 20018 San Sebastián, Spain
[3]Donostia International Physics Center, 20018 Donostia-San Sebastián, Spain
*lauraisabel.fernandez@ehu.eus

Fundamental knowledge of magnetic and electronic interaction phenomena that takes place at the hybrid metalorganic interface is decisive to explore the possibility to create highly spin-polarized spinterfaces to enhance the performance of organic spintronic devices [1]. To this end, tailoring magnetic properties on structurally robust substrates is crucial. Rare-earth/noble metal monolayer alloys, grown on gold surfaces, possess a big potential as nanostructured magnetic templates with structural stability. Different elements of the lanthanide series have been observed to form surface-confined alloys in Au(111) [2], characterized by a high crystal quality and nanoscale periodic corrugation. We have prepared hybrid metal–organic interfaces by the adsorption of copper phthalocyanine CuPc on $REAu_2$ monolayers (RE = Gd, Ho and Yb) and studied their growth, electrostatics, electronic structure and magnetic properties. We observe a significant effect of the RE valence of the substrate on the carrier injection barrier of the hybrid metal–organic interface. CuPc adsorbed on trivalent RE-based surfaces ($HoAu_2$ and $GdAu_2$) exhibits molecular level energies that may allow injection carriers significantly closer to an ambipolar injection behavior than in the divalent case ($YbAu_2$) [3] (see Fig.1). Moreover, the magnetic exchange coupling between the ferromagnetic surfaces ($HoAu_2$ and $GdAu_2$) and the CuPc layer is explored, studing the effect of the RE atom on the magnetic behavior of the hybrid metal-organic interface.

## Acknowledgements

The authors acknowlege Spanish Ministerio de Ciencia e Innovación (grants No. MAT-2017-88374-P, PID2020-116093RB-C44, PID2019-103910GB-I00 and PTA2019-018134-I and the Basque Government (grants No. IT-1591-22, IT-152722), the EU Horizon 2020 through the Marie Skłodowska-Curie Grant MagicFACE No. 797109.

## References

[1] S. M. Urdampilleta et al. Nat. Mater. **10**, 502 (2011).
[2] M. Corso et al. ACS Nano **4**, 1603 (2010); M. Ormaza et al. Phys. Rev. B **88**, 125405 (2013).
[3] R. Castrillo-Bodero et al. Nanoscale, 2023,**15**, 4090 (2023).

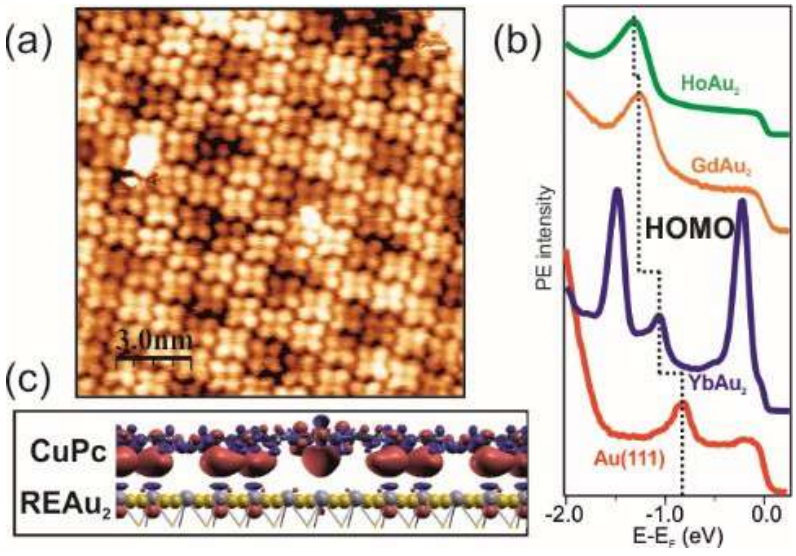



Figure 1: 1ML of CuPc on $REAu_2$ surface:(a) STM of 1ML CuPc on a $HoAu_2$ surface. (b) Valence bands measured on CuPc ML grown on $GdAu_2$, $HoAu_2$ and $YbAu_2$. For comparison the valence band of 1 ML of CuPc/Au(111) is given (c) Side view of the carrier density difference $\Delta\rho(\mathbf{r})$ in the CuPc/$GdAu_2$/Au(111)-2 ML interface with Cu at the Gd atop site.

# Ionic Liquid Gating of Magnetic Multilayers with Structure Inversion Asymmetry

*Md Golam Hafiz[1]*, Philippa Mary Shepley[1], Craig Knox[2], Matthew Rogers[1], Mannan Ali[1], Rohit Pachat[3], Liza Herrera Diez[3], Gavin Burnell[1]*

[1] *School of Physics and Astronomy, University of Leeds, Leeds LS2 9JT, UK*
[2] *School of Electronic and Electrical Engineering, University of Leeds, Leeds LS2 9JT, UK*
[3] *C2N, Université Paris-Saclay, 91120 Palaiseau , France*
*pymgh@leeds.ac.uk

In recent years, magnetic multilayers with structure inversion asymmetry have been widely studied for spintronics applications, due to their strong perpendicular magnetic anisotropy (PMA), strong spin-orbit coupling (SOC), and high Dzyaloshinskii-Moriya Interaction (DMI). In such a film, PMA, DMI and SOC can stabilize the Néel domain walls and complex chiral magnetic textures such as skyrmions [1]. These properties are highly dependent on the interfaces between the ferromagnet (FM) layer and the adjacent non-ferromagnetic layers. In this work we have been investigating the possibilities for controlled, non-volatile, local modification of these interfaces through electrically driven oxygen migration through the film structure. Ionic liquid gating has potential for this due to its ability to modulate magnetic properties such as perpendicular magnetic anisotropy (PMA) via ion migration [2-3]. With a small applied voltage (of order -2 V) oxygen ions migrating from an adjacent oxide layer towards the magnetic layer can change the magnetization direction. The technique is potentially fully reversible allowing the possibility of field programmable skyrmion devices. We are developing a material system based on Ta/Pt/CoB/Ir/Pt multilayer and applying e-field through liquid ionic gating across an $HfO_2$ gate dielectric. This type of structure has been overlooked due to the distance between the oxygen ions and magnetic layers, although, as we show here, it can be used to remotely manipulate magnetization. This experimental study investigates the magneto-ionic effect on fundamental material properties, such as effective anisotropy constant (Keff). We have observed that positive gate voltages significantly decreases the domain nucleation field, whilst negative voltage creates domain wall pinning near saturation. Moreover, we also found that oxygen species can be driven through up to 8 Å of Pt and 2 Å of Ir, we have also demonstrated that these effects are reversible. Our results show that through application of modest positive and negative voltages, we can control the oxygen migration through the upper Pt/Ir layer in our devices with a potential application in a reconfigurable spintronic device.

## References

[1] A. Hrabec, N. A. Porter, A. Wells, M. J. Benitez, G. Burnell, S. McVitie, D. McGrouther, T. A. Moore, and C. H. Marrows, Phys. Rev. B 90, 020402(2014)
[2] L. Herrera Diez, Y. Liu, and D. Gilbert, Physical Review Applied 12, 034005 (2019)
[3] U. Bauer, S. Emori, and G. S. D. Beach, Nature Nanotechnology 8, 411 (2013)

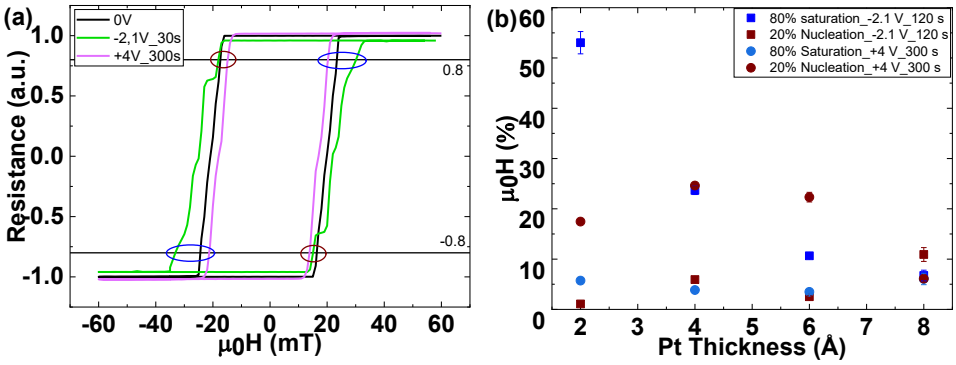



Figure: figures shows the oxygen driving possibilities. a) hysteresis loop changes significantly with applied voltage, b) 20% nucleation and 80 % saturation at the negative gate voltage.

# The correlated random anisotropy model of the Co-organic composite films

*V.V.Kabanov[1]*, A.V. Shumilin[1], M. Benini[2], T. Mertelj[1], V.A. Dediu[2]*

[1] *Jozef Stefan Institute, Jamova cesta 39, SI-1000 Ljubljana, Slovenia*

[2] *CNR-ISMN, Via Piero Gobetti 101, 40129, Bologna, Italy*

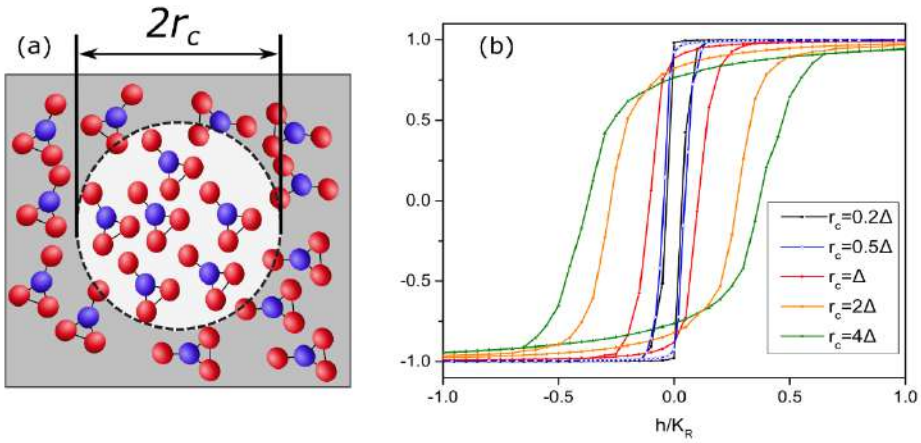
* viktor.kabanov@ijs.si

We propose the correlated random anisotropy model that describes thin ferromagnetic films hybridized with organic molecular layers. The asymmetry of the molecules leads to the random in-plane anisotropy induced at the surface of the magnetic film. We show that this strongly modifies the magnetic anisotropy of the whole cobalt layer which magnitude critically depends on the correlation radius of random anisotropy (fig. a). When this radius is small even strong induced anisotropy can be neglected. However, with the increase of correlation radius, the effect of molecules starts to dominate the magnetic properties. It results in the colossal increase of the coercive field, modification of the hysteresis loop shape (fig. b), and breaking of the Raleigh law at low fields.

Physically, the effect results from the modification of pseudoground states in the magnetic film. At a low correlation radius the state can be described by standard domain wall picture, while at larger correlation radiuses a correlated spin glass state emerges.

**Acknowledgements**

We acknowledge the support of the EC projects INTERFAST (H2020-FET-OPEN-965046).




(a) The correlation radius $r_c$ of molecule-induced anisotropy. (b) Hysteresis loops for different values of $r_c$.

# .Strong Modifications of Magnetic Properties in Thin Co Films by Molecular Chemisorbtion

*Mattia Benini[1]*, Giuseppe Allodi[2], Andrei V. Shumilin[3], Rajib Kumar Rakshit[1], Manju Singh[1], Alberto Riminucci[1], Patrizio Graziosi[2], Viktor V. Kabanov[3], Tomaž Mertelj[3], Ilaria Bergenti[1], V. Alek Dediu[1]*

[1] *ISMN-CNR, Via Piero Gobetti 101, Bologna, Italy*

[2] *Department of Mathematical, Physical and Informatics sciences, University of Parma, Viale Università 12, Parma, Italy*

[3] *Jožef Stefan Institute, Jamova cesta 39, 1000 Ljubljana, Slovenia*

*mattia.benini@ismn.cnr.it

The chemisorbtion of organic molecules on surfaces of *3d*-ferromagnetic thin films alters drastically such key magnetic parameters as the magnetic anisotropy, magnetization and others [1-3]. This is due to the hybridization of *3d* metallic and $\pi$ molecular orbitals and represents one of the most promising routes for tailoring of the components in magnetic and spintronic devices [1].

In this work we present the results obtained for thin polycrystalline Co layers (with thicknesses of 3-7 nm) interfaced with two different molecules, namely $C_{60}$ and $Gaq_3$. A wide range of magnetic characterization techniques were employed, where the dynamics of the magnetic state was investigated by exciting it at different timescales, from nanoseconds to minutes, and studying how the local modifications on the magnetic anisotropy affect the magnetic response of the whole FM layer. MOKE magnetometry and AMR measurements showed a molecule-dependent increase of the in-plane coercivity that is colossaly enhanced at low temperatures. Moreover, minor loops measurements at 150 K indicate a critical deviation (see Figure (b) ) from the universal power law behaviour derived for coercive fields from Rayleigh law [4]. Additionally, $^{59}$Co Zero-Field NMR characterizazion indicated an increase of the magnetic stiffness of the whole FM layer, demonstrating that the interfacial effects propagate several nm into the bulk of the FM material[5]. All the observed molecule-induced effects suggest the establishment of a conceptually new magnetization dynamics and can be reasonable well explained on the basis of a new phenomenological model based on correlated random anisotropy approach.

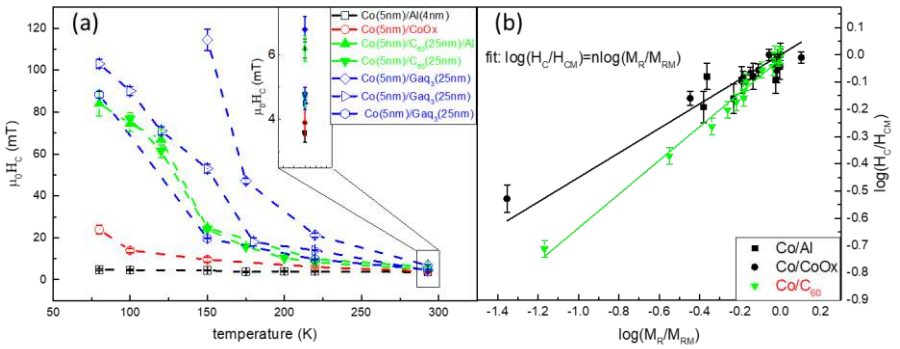



Figure: (a) Coercive fields as a function of temperature for $Co/Gaq_3$ and $Co/C_{60}$ systems, compared with reference Co/Al and Co/CoOx bilayers showing a colossal enhancement of the coercive fields at low temperatures. (b) logarithmic coercive field ratio (minor loops divided by saturated loop) vs remanence anisotropy ratio, showing a clear change in the slope between reference samples (black points) and $Co/C_{60}$ (green points).

## Acknowledgements

The authors acknowledge the support of the EC projects INTERFAST (H2020-FET-OPEN-965046).

## References

[1] M. Cinchetti, V. A. Dediu, L. E. Hueso, Nat. Mater. 2017, 16, 507.
[2] K. V. Raman, A. M. Kamerbeek, A. Mukherjee, *et al.*, Nature 2013, 493, 509.
[3] K. Bairagi, A. Bellec, V. Repain, *et al.*, Phys. Rev. B 2018, 98, 085432.
[4] Takahashi S, Kobayashi S. Philosophical Magazine. 2011, 91(17) 2216-26.
[5] M. Benini, G. Allodi, A. Surpi *et al.*, Adv. Mater. Interfaces 2022, 9, 2201394.

# Electrically Driven Singlet-Triplet Transition in Triangulene Spin-1 Chains

*Gabriel Martínez-Carracedo[1,2], László Oroszlány[3,4], Amador García-Fuente[1,2], László Szunyogh[5,6*], and Jaime Ferrer[1,2],*

[1] *Departamento de Física, Universidad de Oviedo, 33007 Oviedo, Spain*
[2] *Centro de Investigación en Nanomateriales y Nanotecnología, Universidad de Oviedo–CSIC, 33940 El Entrego, Spain*
[3] *Department of Physics of Complex Systems, Eötvös Loránd University, 1117 Budapest, Hungary*
[4] *ELKH-BME Lendület Topology and Correlation Research Group, Budapest University of Technology and Economics, Műegyetem rkp. 3., H-1111 Budapest, Hungary*
[5] *Department of Theoretical Physics, Institute of Physics, Budapest University of Technology and Economics, Műegyetem rkp. 3, H-1111 Budapest, Hungary*
[6] *ELKH-BME Condensed Matter Research Group, Budapest University of Technology and Economics, Műegyetem rkp. 3, H-1111 Budapest, Hungary*
*szunyogh.laszlo@ttk.bme.hu

Simple spin models have played a key role in the formulation and comprehension of the basic principles of magnetism and statistical mechanics since the early days of quantum theory. The interest in these models and in the systems realizing them persists today due to their connection to many topological properties of matter, as well as their potential to become the building blocks of viable and robust quantum computers.

Recently, graphene triangulene (GT) chains have been synthesized and their magnetic response has been analyzed by STM methods by Mishra and coworkers [1]. Motivated by this study, we determine the exchange bilinear and biquadratic constants of the triangulene chains by calculating two-spin rotations in the spirit of the magnetic force theorem. We then analyze open-ended, odd-numbered chains, whose edge states pair up forming a triplet ground state (see Figure). We propose three experimental approaches that enable us to trigger and control a singlet-triplet spin transition. Two of these methods are based on applying a mechanical distortion to the chain. We finally show that the transition can be controlled efficiently by the application of an electric field. [2]

## Acknowledgements

G.M.-C., A.G.-F., and J.F. have been funded by Ministerio de Ciencia, Innovación y Universidades, Agencia Estatal de Investigación, Fondo Europeo de Desarrollo Regional, via Grant No. PGC2018-094783. G.M.-C. has also been supported by Programa "Severo Ochoa" de Ayudas para la investigación y docencia del Principado de Asturias. This research is supported by the NRDI Office under projects no. K131938, K142179, and FK124723 and within the Quantum Information National Laboratory of Hungary by the Ministry of Culture and Innovation and the National Research, Development and Innovation Office via Grant No. 2022-2.1.1- NL-2022-00004. L.O. also acknowledges the support of the NRDI Office of Hungary and the Hungarian Academy of Sciences through the Bolyai and Bolyai+ scholarships.)

## References

[1] S. Mishra *et al*., *Nature* (London) **598** (2021) 287.
[2] G. Martínez-Carracedo *et al., Phys. Rev. B* **107** (2022) 035432.

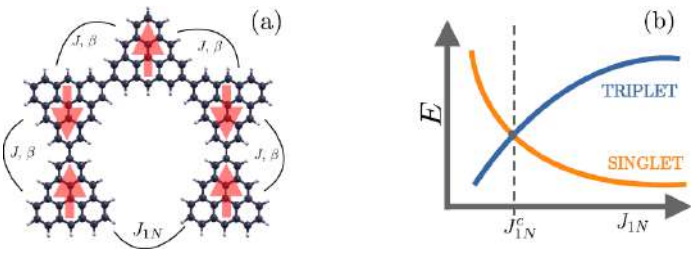



Figure: (a) Sketch of a horseshoelike N = 5 GT chain, where the spins at each triangulene pair are coupled by the same exchange constants $J$ and $\beta$, and where the two end GT spins are coupled by a smaller exchange constant $J_{1N}$ . (b) A singlet-triplet crossing occurs at a finite value of $J_{1N}/J$ smaller than 1.

# Magnetic Behaviour of Layered 2-haloethylammonium Tetrahalocuprates(II)

*P. Šenjug[1]*, D. Barišić[1], E. Topić[2], M. Rubčić[2], D. Pajić[1]*

[1] *Department of Physics, Faculty of Science, University of Zagreb, Bijenička c. 32, Zagreb, Croatia*
[2] *Department of Chemistry, Faculty of Science, University of Zagreb, Horvatovac 102a, Zagreb, Croatia*
*psenjug@phy.hr

Layered metal-organic perovskites are a class of materials with the general chemical formula $A_2BX_4$, also known as 2D hybrid organic-inorganic perovskites. The inorganic layers are separated by two layers of organic cations that are coupled by hydrogen bonds to the inorganic layers, usually via the $NH_3$ group. The structure allows great freedom in the choice of organic cations, allowing very different distances between the metal layers and greater tilting of the octahedron, compared to their 3D counterparts.

In this work, we studied the magnetic properties of a series of layered 2-haloethylammonium tetrahalocuprates(II), based on a well-known ethylammonium tetrachlorocuprate(II), $EA_2CuCl_4$ on which the existence of the magnetic and electric order was observed. Investigated 2D layered metal-organic perovskite materials consisted of 2-chloroethylammonium tetrachlorocuprate ($ClEA_2CuCl_4$), 2-bromoethylammonium tetrachlorocuprate ($BrEA_2CuCl_4$), 2-chloroethylammonium tetrabromocuprate ($ClEA_2CuBr_4$) and 2-bromoethylammonium tetrabromocuprate ($BrEA_2CuBr_4$). All the compounds have the same stoichiometry as the starting $EA_2CuCl_4$ but differ in the additional halogen atom attached to the ethylammonium ion and the halide ion that forms the inorganic backbone. The magnetic DC measurements were performed using an MPMS3 and MPMS5 SQUID magnetometer, in the temperature range from 2K to 300K, and fields up to 5T. Ultra-low field (ULF) option of MPMS3, which enables magnetization measurements in precise low magnetic fields, was used to measure the temperature dependence of magnetization in fields smaller than 5 Oe. The study showed that small changes in the organic cation can give significantly different magnetic properties and that the substitution of $Cl^-$ with $Br^-$ ions in the inorganic layers in addition to changing the strength of the interaction within the plane and the transition temperature, also causes the appearance of hysteresis and a significant coercive field. Compounds with 2-chloroethylammonium organic cations, similar to the starting $EA_2CuCl_4$, show an antiferromagnetic ground state, with a metamagnetic transition, transition to the ferromagnetic state, already for small applied magnetic fields. Replacement of $Cl^-$ with $Br^-$ ion in the organic cations, leads to a completely different magnetic ground state and, in the case of 2-bromoethylammonium cations, it is ferromagnetic. Although in the compounds with tetrabromocuprate(II) layers the distances between neighbouring Cu ions are larger, higher polarizability of bromine ions leads to stronger interactions within the inorganic and thus higher temperatures of magnetic phase transitions. Therefore, only small changes in the organic ions or the change of the halogen atom in the inorganic layers can lead to very different magnetic behaviour. As the structure of the layered metal-organic perovskites offers a wide choice of organic cations, it could be possible to easily design the desired magnetic properties.

**Acknowledgements**

We acknowledge the support of projects CeNIKS, CIuK and QuantiXLie co-financed by the Croatian Government and the European Union through the European Regional Development Fund - Competitiveness and Cohesion Operational Programme (Grants KK.01.1.1.02.0013. and KK.01.1.1.02.0016. and KK.01.1.1.01.0004).

# Hybrid Organic-Inorganic Layered Halocuprates as Quasi-two-dimensional Long-Range Ordered Ferro- and Antiferromagnets

*Damir Pajić [1]*, Pavla Šenjug [1], Dario Barišić [1], Edi Topić [2], Mirta Rubčić [2]*

[1] *Department of Physics, Faculty of Science, University of Zagreb, Bijenička c. 32, Zagreb, Croatia*
[2] *Department of Chemistry, Faculty of Science, University of Zagreb, Horvatovac 102a, Zagreb, Croatia*

*dpajic@phy.hr

Two-dimensional magnetic systems can be realized as the single- or multi-layered metal-nonmetal inorganic compounds and increasing interest in them is driven with the fundamental questions about magnetic interactions and ordering as well as with possible appearance of the electronic excitations leading to applications such as magnonics, spintronics, light-induced ultra-fast magnetic switching, etc.

Our research is focussed on hybrid organic-inorganic layered halometallates which have a perovskite three-dimensional crystal structure. Layered systems allow large freedom in choosing the organic cations, leading to the possibility of designing multifunctional materials with additional ferroelectric, luminiscent, photonic, semiconducting, or other properties beside the magnetic ones. Our investigated systems consist of halometalate layers, where metal ions are chosen to be magnetic and halogen bridges serve for efficient super-exchange interaction leading to strong magnetic correlations.

Magnetic properties of our investigated tetra-chloro and -bromo cuprates are quite interesting in their diversity. They show quasi-two-dimensional magnetic behaviour, but also the long-range magnetic order and phase transitions. Namely, within the cuprate planes usually the moderate super-exchange interaction between the neighbouring spins is present (order of 10 – 100 K) leading to the ferromagnetic planes, and much smaller interactions between the planes trigger the magnetic phase transition (usually at temperatures 5 – 30 K). Other local symetry breaking can also help the ordering due to cutting out the fluctuations. The magnetic ground state can be ferromagnetic as well as antiferromagnetic, that might depend on competition of these smaller interactions originating from the magnetic dipolar energies, super-exchange over the organic layers, and other interactions transferred through secondary bonds. Especially interesting is the appearance of metamagnetism with relatively small critical fields.

Magnetism in several series of tetrahalocuprates will be presented and discussed in a way to show their diversity dependending on chemical and structural peculiarities.

**Acknowledgements**

We acknowledge the support of projects CeNIKS and CIuK co-financed by the Croatian Government and the European Union through the European Regional Development Fund - Competitiveness and Cohesion Operational Programme (Grants KK.01.1.1.02.0013. and KK.01.1.1.02.0016.).

# SYMPOSIUM 09.
## STRONGLY CORRELATED SYSTEMS, SUPERCONDUCTIVITY AND FRUSTRATED AND DISORDERED MAGNETISM.
## S9. INVITED ORAL PRESENTATIONS

# Emergent Gauge Fields and Quasi-Particles in Spin Liquids

*Ludovic D.C. Jaubert* [1]*

[1] *CNRS, LOMA, University of Bordeaux, 33400 Talence, France*

*ludovic.jaubert@cnrs.fr

Spin liquids are malleable magnetic textures obeying their own microscopic rules. These rules, due to frustrated constraints, can take the form of emergent gauge fields and topological phases able to support quasi-particles such as magnetic monopoles, Majorana fermions and fractons, to cite but a few.

In this talk we will give a pedagogical presentation of some of the most fascinating phenomena supported by spin liquids, in particular how to move from the microscopic Hamiltonian to the gauge-field description and how to confirm the presence of spin liquids and quasi-particles in experiments. We will illustrate these ideas through a variety of models and materials, such as (quantum) spin ice, Kitaev systems, higher-rank gauge fields ...

## Acknowledgements

We acknowledge financial support from CNRS PICS (MEFLS) and from the "Agence Nationale de la Recherche" under Grant No. ANR-18-CE30-0011-01.

## References

1. Anjana M. Samarakoon, S. A. Grigera, D. Alan Tennant, Alexander Kirste, Bastian Klemke, Peter Strehlow, Michael Meissner, Jonathan N. Hallen, Ludovic Jaubert, Claudio Castelnovo & Roderich Moessner, PNAS **119** (2022) e2117453119.
2. H. Yan, O. Benton, L.D.C. Jaubert & N. Shannon, Phys. Rev. Lett. **124** (2020) 127203.

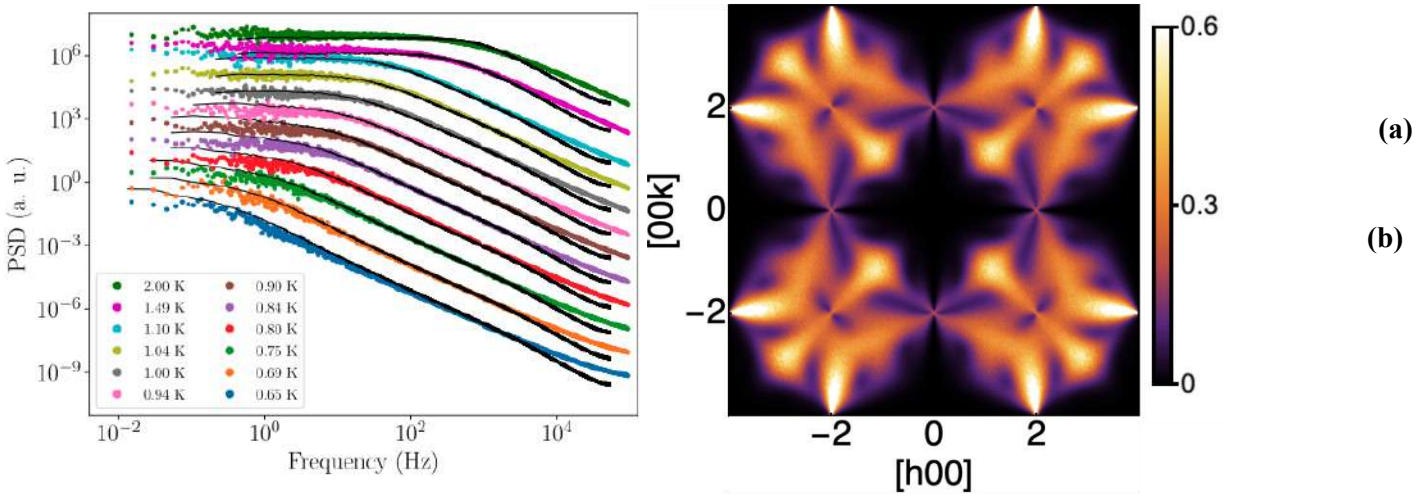



Figure: **(a)** Power Spectral Density of the spin-ice material Dy2Ti2O7 measured by ultra sensitive SQUID (symbols) and Monte-Carlo simulations (black line) [1]. **(b)** Structure factor of a rank-2 U(1) gauge field obtained from simulations on the breathing pyrochlore lattice, with its characteristic four-fold pinch points [2].

# Magnetic Order in Nanoscale Gyroid Networks

*Naëmi Leo*[1]*

[1] *Instituto de Nanosciencia y Materiales de Aragón, CSIC—University of Zaragoza, 50009 Zaragoza, Spain*

* naemi.leo@unizar.es

Three-dimensional magnetic metamaterials feature interesting phenomena that arise from a delicate interplay of material properties, local anisotropy, curvature, and connectivity. A particularly interesting magnetic lattice that combines these aspects is that of nanoscale gyroids shown in Fig. 1a. Gyroid lattices with periodicities in the order of a few tens of nanometres can be grown by polymer self-assembly and feature a highly-interconnected chiral network with local three-connectivity reminiscent of 2D artificial spin ices.

In this talk, I will present recent results on finite-element micromagnetic simulations on field-driven and relaxed spin configurations in nanoscale nickel gyroids [1]. These feature complex magnetic states, as shown in Fig. 1b, with spin chiral effects arising from the non-trivial local magnetic anisotropy. Especially in comparison to planar devices, the 3D geometry and connectivity, truly 3D spin order in response to 3D fields, and the multitude of parallel conduction channels of the regular gyroid network, as shown in Fig. 1c, result in an extensive manifold of frustrated magnetic states as well as gives many possible choices for magnetotransport geometries. This vast phase space therefore is ideal to be explored for future three-dimensional spintronic applications, such as non-reciprocal transport as well as for probabilistic and neuromorphic computing

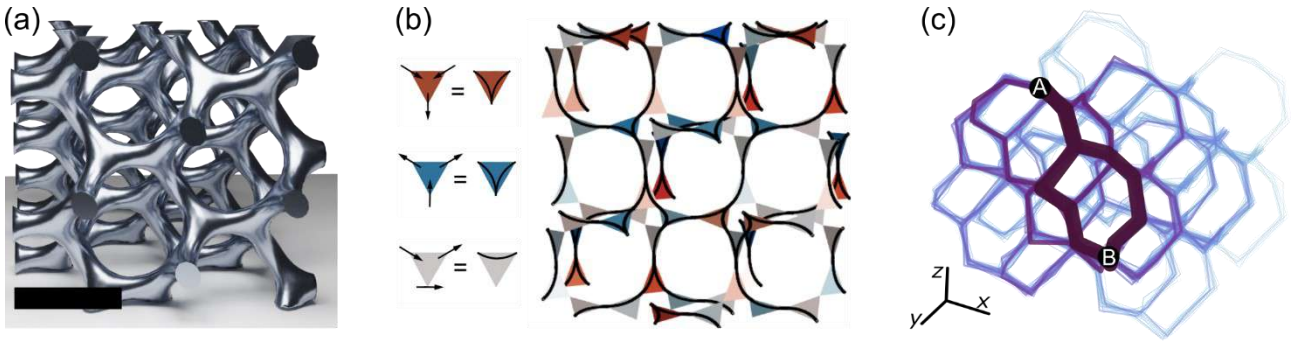



Fig. 1: (a) Gyroid structure used for micromagnetic simulations. The scale bar measures 50 nm. (b) The magnetic order (here at remanence) can be represented on a network of highly-connected triangular plaquettes, with effective macrospins located at each corner. Black lines mark pair-wise in-out macrospin configurations, and the colour scale denotes two-in-one-out (red) or one-in-two-out (blue) ice-like correlations. (c) Possible paths $A \rightarrow B$ connecting nodes $A$ and $B$ through the gyroid network, with different path length indicated by line colour and width. The multitude of connecting paths and magnetic order influence decidedly the anisotropic magnetoresistance.

platforms.

**Acknowledgements**

This work was supported by funding from the European Research Council (ERC) under European Union's Horizon 2020 research and innovation programme under the Marie Sklodowska Curie Grant Agreement No. 844304 (LICONAMCO), as well as support by the European Community under the Horizon 2020 Program, Contract No. 101001290 (3DNANOMAG).

**References**

[1] A. Koshikawa, J. Llandro, M. Ohzeki, S. Fukami, H. Ohno and N. Leo., arXiv:2303.05816 (2023).

# Control of Spin Polarization in Oxide Tunnel Junctions

*V. Rouco[1]*, F. Gallego[1,2], D. Hernandez-Martin[1], D. Sanchez-Manzano[1], J. Tornos[1], J.I. Beltran[1,3], M. Cabero[4], F. Cuellar[1], D. Arias[1], G. Sanchez-Santolino[1,3], F.J. Mompean[2,5], M. Garcia-Hernandez[2,5], A. Rivera-Calzada[1,5], M. Varela[1,3], M.C. Muñoz[5,6], C. Leon[1,5], Z. Sefrioui[1,5] and J. Santamaría[1,5]*

[1] *GFMC, Universidad Complutense de Madrid, 28040 Madrid, Spain*

[2] *2D-Foundry Group, Instituto de Ciencia de Materiales de Madrid ICMM-CSIC, 28049 Madrid, Spain*

[3] *Instituto Pluridisciplinar, Universidad Complutense de Madrid, 28040 Madrid, Spain*

[4] *Centro Nacional de Microscopia Electronica, Universidad Complutense, 28040 Madrid, Spain*

[5] *Unidad Asociada UCM/CSIC, "Laboratorio de Heteroestructuras con Aplicación en Spintrónica", 28040 Madrid, Spain*

[6] *Instituto de Ciencia de Materiales de Madrid ICMM-CSIC, 28049 Madrid, Spain*

*vrouco@ucm.es

The resistance control of a device through the magnetic properties of its counterparts is one of the major achievements in the last decades for the advances in electronics and memory devices. From this perspective, different architectures have been proposed convining ferromagnets with non-magnetic and/or insulating materials. For instance, magnetoelectric coupling may exist in interfaces between an oxide ferroelectric barrier and two ferromagnetic oxide electrodes in a tunnel junction. Here, the inversion of the ferroelectric polarization produces large changes in the value of the tunnelling magnetoresistance. Moreover, the controlled motion of oxygen vacancies in the device through external electric fields allows an additional knob for the modulation of the magnetoresistive effects.

Here we will show that the concurrence of interface oxidation and ferroelectric switching allows for the controlled inversion of the interface spin polarization. We will moreover show that the controlled oxidation of the interface allows for a continuum of non-volatile resistance states in the tunneling magnetoresistance. These results points towards new perspectives in memory and neuromorphic computing devices where the magnetoelectronic responses are modulated by electrochemical degrees of freedom.

**Acknowledgements**

This work was supported by Spanish MINECO through Grant No. MAT2017-87134-C02. Authors acknowledge received funding from the project To2Dox of FlagERA ERA-NET Cofund in Quantum Technologies implemented within the European Union's Horizon 2020 Program. Work (JS, CL, FM , M G-H) supported by Spanish AEI through grants, PID2020-118078RB-I00 and by Regional Government of Madrid CAM through SINERGICO project Y2020/NMT-6661 CAIRO-CM. G.S.-S. acknowledges financial support from Spanish MCI Grant Nos. RTI2018-099054-J-I00 (MCI/AEI/FEDER, UE) and IJC2018-038164-I. Electron microscopy observations were carried out at the Centro Nacional de Microscopia Electrónica, CNME-UCM. VR acknowledges financial support from the Madrid Government (Comunidad de Madrid Spain) under the Multiannual Agreement with Universidad Complutense de Madrid in the line Research Incentive for Young PhDs, in the context of the V PRICIT (Regional Programme of Research and Technological Innovation.

# Curved electronics: geometry-induced effects at the nanoscale

*Paola Gentile[1,2,*], Mario Cuoco[1,2], Oleksii M. Volkov[3], Zu-Jian Ying[4], Ivan J. Vera-Marun[5,6], Denys Makarov[3] and Carmine Ortix[2]*
*[1]CNR-SPIN, I-84084 Fisciano (Salerno), Italy*
*[2]Dipartimento di Fisica "E. R. Caianiello", Università di Salerno, I-84084 Fisciano (Salerno), Italy*
*[3]Helmholtz-Zentrum Dresden-Rossendorf, Institute of Ion Beam Physics and Materials Research, Dresden, Germany*
*[4]School of Physical Science and Technology, Lanzhou University, Lanzhou, China*
*[5]Department of Physics and Astronomy, University of Manchester, Manchester, UK*
*[6]National Graphene Institute, University of Manchester, Manchester, UK*
*paola.gentile@cnr.spin.it

Over the past thirty years, Rashba spin-orbit coupling (RSOC) has been at the basis of the predictions and discoveries of new classes of topological materials and novel phenomena, particularly attractive within the context of spintronics, such as non-standard magnetic textures, spin Hall and topological spin Hall, Edelstein effects, etc [1]. These progresses renewed the interest in the development of new inversion asymmetric structures, like for instance the interfaces between complex oxides, where the inversion symmetry breaking promotes and allows to tune the formation of unusual interfacial electronic phases which are absent in the constituent materials. Furthermore, the multi-orbital character of the quasi-two-dimensional electron gas at the interface between oxide materials, combined with the inversion symmetry breaking, can give rise to an exotic spin-triplet superconductivity, which is spatially isotropic and topologically non trivial [2], and exhibits an unconventional nodal structure with unique tunable features [3,4], responsible for the occurrence of an anomalous Josephson coupling and a dominance of high-harmonics in the current phase relation [5].

A new frontier of exploration within the context of low dimensional structures has been recently opened by the demonstration of the possibility to create flexible semiconductor nanomaterials bent into curved, deformable objects ranging from semiconductor nanotubes, to nanohelices, etc. The consequences of the nanowire bending on the electronic quantum properties have been demonstrated to become of particular importance in systems with structure inversion asymmetry, where the interplay between nanoscale deformations and Rashba spin-orbit coupling (RSOC) [6] allows an all-geometrical and electrical control of electronic spin textures and spin transport properties [7,8], including the possibility to induce topological nontrivial phases [9]. The exciting developments in the discovery and exploitation of novel effects induced by curvature at the nanoscale allow ultimately to define a completely new field – curved nanoelectronics [10]. We here examine in details the origin of curvature effects at the nanoscale and illustrate their potential applications in innovative electronic [6,8,9], spintronic [11], and in particular in superconducting devices [12].

**Acknowledgements**

EU-FET OPEN project "CNTQC", grant agreement N. 618083 (http://www.nano2qc.eu/)

**References**

[1] A. Manchon, H. C. Koo, J. Nitta, and R. A. Duine, *Nature Materials* **14** (2015) 871
[2] Y. Fukaya, S. Tamura, K. Yada, Y. Tanaka, P. Gentile, and M. Cuoco, *Phys. Rev. B* **97**, 174522 (2018) ;
[3] Y. Fukaya, S. Tamura, K. Yada, Y. Tanaka, P. Gentile, and M. Cuoco, *Phys. Rev. B* **100**, 104524 (2019)
[4] Y. Fukaya, K. Yada, Y. Tanaka, P. Gentile, M. Cuoco, *Phys. Rev. B* **102**, 144512 (2020).
[5] Y. Fukaya, Y. Tanaka, P. Gentile, K. Yada, M.Cuoco, *npj Quantum Materials*, vol. 7, **99** (2022).
[6] P. Gentile, M. Cuoco, C. Ortix, *SPIN* **3**, 1340002 (2013).
[7] Z.-J. Ying, P. Gentile, C. Ortix, M. Cuoco, *Phys. Rev. B* **94**, 081406(R) (2016).
[8] G. Francica, P. Gentile, M. Cuoco, *EPL* **127**, 30001 (2019).
[9] P. Gentile, M. Cuoco, C. Ortix, *Phys. Rev. Lett.* **115**, 256801 (2015); S. Pandey, N. Scopigno, P. Gentile, M. Cuoco, C. Ortix, *Phys. Rev. B* **97**, 241103(R) (2018).

[10] P. Gentile, M. Cuoco, O. M. Volkov, Z.-J. Ying, I. J. Vera-Marun, D. Makarov and C. Ortix, *Nature Electronics* **5**, 551–563 (2022).
[11] K. S. Das, D. Makarov, P. Gentile, M. Cuoco, B. J. van Wees, C. Ortix, I. J. Vera-Marun, *Nano Lett.* **19**, 6839–6844 (2019).
[12] Z.-J. Ying, M. Cuoco, C. Ortix, P. Gentile, *Phys. Rev. B* **96**, 100506(R) (2017); G. Francica, M. Cuoco, P. Gentile, *Phys. Rev. B* **101**, 094504 (2020).

# SYMPOSIUM 09.
## STRONGLY CORRELATED SYSTEMS, SUPERCONDUCTIVITY AND FRUSTRATED AND DISORDERED MAGNETISM.
## S9. ORAL PRESENTATIONS

# From spin liquid to magnetic ordering in the anisotropic kagome Y-Kapellasite $Y_3Cu_9(OH)_{19}Cl_8$: a single crystal study

*Dipranjan Chatterjee[1*], Pascal Puphal[2], Edwin Kermarrec[1], Philippe Mendels[1], Fabrice Bert[1]*
*[1] Laboratoire de Physique des Solides, Université Paris-Saclay, France*
*[2] Max Planck Institute for Solid State Research: Stuttgart, Germany*
*dipranjan.chatterjee@universite-paris-saclay.fr

Quantum spin liquids are novel magnetic states of matter characterized by the absence of phase transition down to T=0 K, macroscopic entanglement and emergent fractionalized excitations. The quest for materials realizing such states recently yielded a new family of interesting kagome compounds $YCu_3(OH)_{6+x}Cl_{3-x}$ (x = 0, 1/3) resulting from the substitution of $Zn^{2+}$ by $Y^{3+}$ in the archetypal herbertsmithite $ZnCu_3(OH)_6Cl_2$. Here we present a detailed study of the x=1/3 variant [1], $Y_3Cu_9(OH)_{19}Cl_8$, also coined Y-kapellasite, a distorted kagome materializing a new frustrated model with a rich magnetic phase diagram [2]. We studied large phase-pure single crystals, obtained via an external gradient method, by susceptibility, specific heat, thermal expansion, neutron scattering and local μSR and NMR techniques.

In contrast with previous polycrystalline and powder sample studies [3], this single crystals investigation gives evidence for subtle structural instabilities at ~ 33 K and ~ 13 K which preserve the global symmetry of the system and thus the magnetic model. Further, our results provide clear evidence for a magnetic transition at ~ 2.1 K from complementary experimental methods, which is in line with the theoretical prediction of a (1/3,1/3) long range ordering. However our analysis of the spin wave excitations yields magnetic interactions which locate the compound closer to the phase boundary to a classical jammed spin liquid phase. Likely due to large quantum fluctuations at this boundary, the ordered moment of $Cu^{2+}$ is strongly reduced, undetected with neutron diffraction, but revealed with μSR and NMR.

**Acknowledgement:**
The authors acknowledge the support of the French Agence Nationale de la Recherche, under Grant No. ANR-18-CE30-0022 ”LINK”, and funding from Deutsche Forschungsgemeinschaft (DFG) through TRR 288-422213477 (project A03).

**References:**
[1] D.Chatterjee et al , accepted in Physical Review B.[arXiv preprint arXiv:2211.16624 (2022)]
[2] M.Hering et al , npj Comput Mater **8**, 10 (2022).
[3] Q.Barthelemy et al , Phys. Rev. Materials, **3** (7), 074401 (2019)

# Magnetism and Conductivity in Newly Synthesized $A_2Ir_2O_7$ Single Crystals – Phase Diagram and Anisotropy

*Milan Klicpera*[1]*, *Daniel Staško*[2], *Kristina Vlášková*[1], *Ross H. Colman*[1], *Jiří Kaštil*[2]

[1] *Charles University, Faculty of Mathematics and Physics, Department of Condensed Matter Physics, Ke Karlovu 5, 121 16 Prague 2, Czech Republic*

[2] *Institute of Physics of the Czech Academy of Sciences, Na Slovance 1999/2, 182 00 Prague 8, Czech Republic*

*mi.klicpera@seznam.cz

Diversity of complex ground-states in rare-earth $A_2Ir_2O_7$ oxides originates primarily from the geometrical frustration of magnetic moments on pyrochlore lattice (*F d -3 m*, no. 227), and from the confluence and delicate balance between exchange, dipolar and spin-orbit interactions (SOI) in this system. The 3D-geometrically frustrated lattice significantly affects the antiferromagnetically and ferromagnetically coupled moments, leading to, e.g., spin-ice and spin-liquid states [1,2,3]. SOI and intermediate electron Coloumb correlations in iridates generate topologically insulating phases, e.g., topological Mott insulator or topological band insulator, or Weyl semimetal state with Fermi-arc surface states [4].

The iridium sublattice orders magnetically at intermediately-low temperatures in all so far studied iridates, except $Pr_2Ir_2O_7$ [4]. Based on neutron diffraction experiments, the Ir magnetic order has been determined as a long-range all-in-all-out (AIAO) order [3,5]. Simultaneously, semimetallic/nonmetallic $A_2Ir_2O_7$ become insulators below ordering temperature [6]. The rare-earth moments are subjected to the Ir molecular field through *d-f* coupling, which might result in induced AIAO magnetic order of the *A* sublattice [7] and/or phase competition at low temperature [3]. Fragmentation of magnetic moments [5,8] has been reported as well.

Our most recent work focuses on magnetic and conducting properties related primarily to the Ir-sublattice studied on ***newly synthesized single crystals of $A_2Ir_2O_7$*** with *A* = La, Nd, Er, Tm, and Lu (edge of a bipyramid of up to 1 mm long). The iridium sublattice magnetic phase diagram has been completed for the whole series. AIAO magnetic structure was confirmed in new member *A* = Tm by neutron diffraction studies. Single crystals enabled to study, inter alia, the anisotropy of magnetic properties. Hypothesis on Ir antiferromagnetic domains and ferromagnetic interfaces across the $A_2Ir_2O_7$ series, pathing a way towards new type of spintronics, has been supported by measurements on single crystals. Rise of insulating state in heavy *A* members was studied and described by the Slater mechanism, relating it to antiferromagnetic ordering of Ir sublattice. Our studies have been supplemented by inelastic neutron scattering, synchrotron radiation scattering, and muon spin rotation experiments.

## Acknowledgements

We acknowledge the MGML (mgml.eu), which is supported within the program of Czech Research Infrastructures (project no. LM2018096); and the grant agency of the Charles University, project no. 148622.

## References

[1] E. Lefrancois, V. Simonet, R. Ballou, E. Lhotel, A. Hadj-Azzem, S. Kodjikian, P. Lejay, P. Manuel, D. Khalyavin, L.C. Chapon, *Phys. Rev. B* **114** (2015) 247202.

[2] K. Matsuhira, M. Tokunaga, M. Wakeshima, Y. Hinatsu, S. Takagi, *J. Phys. Soc. Japan* **82** (2013) 023706.

[3] H. Jacobsen, C.D. Dashwood, E. Lhotel, D. Khalyavin, P. Manuel, R. Stewart, D. Prabhakaran, D.F. McMorrow, A.T. Boothroyd, *Phys. Rev. B* **101** (2020) 104404.

[4] J.S. Gardner, M.J.P. Gingras, J.E. Greedan, *Rev. Modern Phys.* **82** (2010) 53.

[5] E. Lefrancois, V. Cathelin, E. Lhotel, J. Robert, P. Lejay, C. V. Colin, B. Canals, F. Damay, J. Ollivier, B. Fåk, L. C. Chapon, R. Ballou, V. Simonet, *Nat. Commun.* **8** (2017) 209.

[6] K. Matsuhira, M. Wakeshima, Y. Hinantsu, S. Takagi, *J. Phys. Soc. Japan* **80** (2011) 094701.

[7] H. Guo, C. Ritter, A.C. Komarek, *Phys. Rev. B* **96** (2017) 144415.

[8] V. Cathelin, E. Lefrançois, J. Robert, P. C. Guruciaga, C. Paulsen, D. Prabhakaran, P. Lejay, F. Damay, J. Ollivier, B. Fåk, L. C. Chapon, R. Ballou, V. Simonet, P. C. W. Holdsworth, E. Lhotel, *Phys. Rev. Res.* **2** (2020) 032073.

# $K_2ReCl_6$: an unconventional Jahn-Teller system?

*A. Bertin[1*], T. Dey[1], D. Brüning[1], D. Gorkov[1,2], K. Jenni[1], A. Krause[1], P. Becker[3], L. Bohatý[3], D. Khomskii[1], T. Lorenz[1], and M. Braden[1]*

[1] *II. Physikalisches Institut, Universität zu Köln, Zülpicher Straße 77, D-50937 Köln, Germany*
[2] *Heinz Maier-Leibnitz Zentrum, Technische Universität München, D-85748 Garching, Germany*
*3Abteilung Kristallographie, Institut für Geologie und Mineralogie, Zülpicher Straße 49b, D-50674 Köln, Germany*
*bertin@ph2.uni-koeln.de

Many recent developments in condensed matter physics arise from the interplay between spin-orbit coupling (SOC) and electronic correlations. Much of the effort have been focused on the *4d* or *5d* transition metals, as for instance the honeycomb iridates, potential candidates for the realization of the Kitaev model. The impact of strong SOC on other *5d* electronic configuration has been much less studied. Although, it could lead to completely different ground states as well. With this purpose, other material for novel spin-orbit driven phenomena are being synthesized. Antifluorite compounds of chemical formula $K_2MX_6$ (where *M* is a 4d/5d transition metal and *X*=Cl,Br) can exhibit various crystallographic phase transitions, often understood by the softening of rotary phonon modes of the ligand octahedra surrounding the central transition metal. Among this family, $K_2ReCl_6$ is a promising material among the *5d* Mott insulators. It exhibits on cooling four distinct structural phases and may constitute a playground to investigate the interplay between spin-orbit coupling and Jahn-Teller (JT) effect. Indeed, the $5d^3$ electronic ground state in the weak SOC limit (Russel-Saunders scheme) does not show any orbital degeneracy. On the other hand, in the strong SOC limit (*jj*-coupling scheme), the $j_{eff}$=3/2 ground state is JT active [1]. The question whether one of the crystallographic phase transitions is JT driven will be tackled by presenting a detailed temperature dependent structural study of $K_2ReCl_6$ and of its non magnetic counterpart $K_2SnCl_6$ by means of powder and single crystal X-ray diffraction. With high resolution single crystal neutron diffraction experiments and considering the monoclinic symmetry of the low temperature phase the magnetic structure was solved. Frustration is only partially lifted by the structural distortions and the magnetic order causes further symmetry reduction. Finally, the strong magneto-elastic effect observed by thermal expansion measurements will be discussed with the key ingredients (domain re-orientation and weak ferromagnetism) provided by these structural studies [2].

## References

[1] S. V. Streltsov and D. I. Khomskii, Phys. Rev. X 10, 031043 (2020)
[2] A. Bertin *et al.*, arXiv:2207.11101

# Long-range order, re-entrant spin glass and spin liquid correlations in anion disordered $Gd_2Hf_2O_7$

*Jianhui Xu[1,2,3,4,*], Vivek Anand[4, 5], Ena Osmic[6], Sumanta Chattopadhyay[6], Thomas Herrmannsdörfer[6], Clemens Ritter[7] Andrew Wildes[7] and Bella Lake[4,8]*

[1]*Institut für Kristallographie, RWTH Aachen University, Jägerstr. 17-19, 52056 Aachen, Germany*

[2]*Jülich Centre for Neutron Science (JCNS) at Heinz Maier-Leibnitz Zentrum (MLZ), Forschungszentrum Jülich GmbH, Lichtenbergstr. 1, 85747 Garching, Germany*

[3]*Heinz Maier-Leibnitz Zentrum (MLZ), Technische Universität München, 85748 Garching, Germany*

[4]*Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, Hahn-Meitner-Platz 1, D-14109 Berlin, Germany*

[5]*Department of Physics, SRM University, AP-Amaravati, 522502 Andhra Pradesh, India*

[6]*Helmholtz-Zentrum Dresden-Rossendorf, Bautzner Landstrasse 400, D-01328 Dresden, Germany*

[7]*Institut Laue-Langevin, Boite Postale 156, 38042 Grenoble Cedex, France*

[8]*Institut für Festkörperphysik, Technische Universität Berlin, Hardenbergstraße 36, D-10623 Berlin, Germany*

*jianhui.xu@frm2.tum.de

Pyrochlore antiferromagnets (AFM) $Gd_2T_2O_7$ ($T$: tetravalent metal elements) are prototypical materials for realizing classical spin liquid states. However, all of them have been observed to show long-range magnetic order [1-3]. Previous specific heat data of $Gd_2Hf_2O_7$ show a tiny sharp peak on the top of a large broad maximum indicating a long-range AFM order [4]. However, our sample does not show that sharp peak in specific heat, but the ac susceptibility evidences an ordering transition followed by a spin-glass transition. Using neutron diffraction, we found that the sample has oxygen Frankel defects and undetectable Gd/Hf anti-site defects. The polarized neutron diffuse scattering pattern shows liquid-like scattering without any magnetic Bragg peaks. The subtle long-range order and re-entrant spin glass are attributed to bond disorder due to oxygen anion disorder.

## References

[1] J. S. Gardner, et al., *Reviews of Modern Physics* 82, 53 (2010).
[2] X. Li et al., *Phys. Rev. B* 94, 214429 (2016).
[3] A. M. Hallas et al., *Phys. Rev. B* 91, 104417 (2015).
[4] M. D. Alice et al., *J. Phys: Conden Matter* 20, 235208 (2008).

# Complex magnetic phases in dhcp Nd: self-induced spin glass and thermally induced order.

## (Times New Roman, Bold, 14 points, centred)

*Anders Bergman*[1]

[1] *Department of Physics and Astronomy, Uppsala University, Box 516, SE-751 20 Uppsala, Sweden*

*anders.bergman@physics.uu.se

Among the wide variety of magnetic orders, one of the most intriguing phases is manifested by the spin glass state where the magnetization exhibit glassy dynamics, including ageing and memory effects. The peculiar dynamics of spin glass materials can be explained from an energy landscape with several local minima combined with larger energy barriers which result in non-ergodic behavior. Such energy landscapes have historically been associated with disorder and spin glass dynamics have been expected to occur only in chemically disordered materials.

Recent theoretical and experimental findings [1] have indicated the existence of glassy dynamics in thick films of elemental crystalline Nd, i.e in a material with limited chemical disorder. Bulk Nd crystallizes in the dhcp structure and earlier experimental studies have reported the existence of several non-collinear states across its phase diagram. Here, we present our interpretation that the low temperature state of dhcp Nd can in fact be described as a self-induced spin glass. First-principles DFT calculations of magnetic exchange interactions combined with atomistic spin dynamics simulations show that there is an intrinsic frustration of the exchange interactions between Nd atoms at the cubic and the hexagonal sites in the dhcp structure. The competitions between the exchange interactions causes an energy landscape that can explain the glassy dynamics.

Furthermore, we show that the exchange interaction competition between the different sites in the dhcp structure has a temperature dependence that leads to a surprising phase transition from the self-induced spin glass phase to an ordered multi-Q phase[2] with increasing temperature.

**References**

[1] U. Kamber, A. Bergman, A. Eich, D. Iuşan, M. Steinbrecher, N. Hauptmann, L. Nordström, M. I. Katsnelson, D. Wegner, O. Eriksson, and A. A. Khajetoorians, Science 6757 **368** (2020)

[2] B. Verlhac, L. Niggli, A. Bergman, U. Kamber, A. Bagrov, D. Iuşan, L. Nordström, M. I. Katsnelson, D. Wegner, O. Eriksson & A. A. Khajetoorians, Nature Physics 905 **18** (2022)

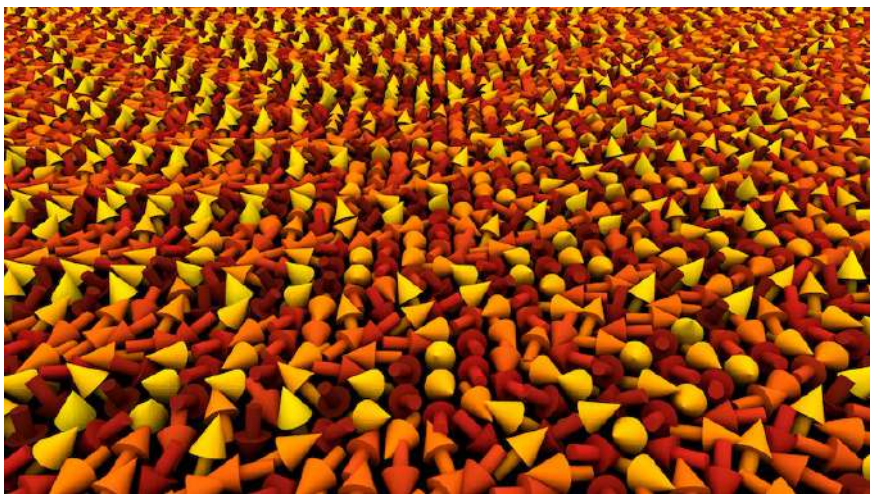


Figure: Simulated magnetic structure in the self-induced spin glass phase of dhcp Nd

# Topological Spintronics in Magnetically Frustrated Systems

*Ricardo Zarzuela*

*Institut für Physik, Johannes Gutenberg Universität Mainz, D-55099 Mainz, Germany*

rzarzuela@uni-mainz.de

Frustrated magnets, understood as magnetic systems with frustrated interactions dominated by isotropic exchange, have resurged in condensed matter due to their striking spin transport properties [1,2] and ability to host three-dimensional magnetic solitons such as Shankar skyrmions [3] and Anderson-Toulouse vortices [4]. These topological textures are encoded in the corresponding SO(3)-order parameter, which describes the spin-spin correlations of these magnetic platforms, and also coexist with the more conventional magnetic solitons emerging in the magnetization field, e.g. domain walls and baby skyrmions. In this talk, I will discuss recent advances in the transport of charge and spin in magnetically frustrated conductors [5,6], with special emphasis on the topological aspects. In particular, I will consider the spin-transfer and the topological Hall effects mediated by the SO(3) solitons and magnetic disclinations (namely, the topological singularities of the order parameter) that emerge in frustrated magnets, which find no counterparts in collinear magnetism. I will also present a collective variable framework well suited to describe the current-driven dynamics of SO(3) solitons in magnetically frustrated platforms [7]. Furthermore, experimental setups for the measurement of the aforementioned phenomena will be discussed.

**Acknowledgements**

This work has been supported by the Transregional Collaborative Research Center (SFB/TRR) 173 SPIN+X, the skyrmionics SPP: ZA1194/2-1, SI1720/12-1, Grant Agency of the Czech Republic Grant No. 19-28375X, and the Dynamics and Topology Centre funded by the State of Rhineland Palatinate.

**References**

[1] N.L. Nair, E. Maniv, C. John, S. Doyle, J. Orenstein, J.G. Analytis, *Nat. Mater.* **19** (2020) 153.
[2] A. Little, C. Lee, C. John, S. Doyle, E. Maniv, N.L. Nair, W. Chen, D. Rees, J.W.F. Venderbos, R.F. Fernandes, J.G. Analytis, J. Orenstein, *Nat. Mater.* **19** (2020) 1062.
[3] R. Shankar, *J. Physique* **38** (1977) 1405.
[4] P.W. Anderson, G. Toulouse, *Phys. Rev. Lett.* **38** (1976), 508.
[5] R. Zarzuela, J. Sinova, *Phys. Rev. B* **105** (2021) 024423.
[6] R. Zarzuela, J. Sinova, arXiv:2112.06680 (2021).
[7] R. Zarzuela, J. Sinova, in preparation (2023).

# Stepwise Domain Growth in Artificial Spin Ice

*Ida Breivik*[1]*, *Johannes Høydahl Jensen*[2], *Anders Strømberg*[1], *Arthur Penty*[2], *Miguel Angel Nino*[3], *Michael Foerster*[3], *Gunnar Tufte*[2] *and Erik Folven*[1]

[1] *Department of Electronic Systems, Norwegian University of Science and Technology, Trondheim, Norway*

[2] *Department of Computer Science, Norwegian University of Science and Technology, Trondheim, Norway*

[3] *ALBA Synchrotron Light Facility, Cerdanyola del Vallès, Barcelona, Spain*

*ida.breivik@ntnu.no

Artificial Spin Ices (ASIs) are interacting arrays of bistable nanomagnets which show potential in a wide range of fields, from fundamental physics to unconventional computing. The 45 degree pinwheel ASI exhibits a long-range ferromagnetic ordering [1] and the two sublattices of nanomagnets, rotated 90 degrees with respect to each other, allow for four possible ferromagnetic domains oriented in the cardinal directions. Using the open-source simulation software flatspin [2], we show that these ASIs display interesting behaviour in response to global, in-plane magnetic field protocols. By exploiting the asymmetric Stoner-Wohlfarth switching astroids of the stadium shaped nanomagnets, we are able to tailor a global field protocol to grow ferromagnetic domains from the polarized state, in a controlled, stepwise manner. This stepwise domain growth is fundamentally different from avalanche switching typically observed in such systems, and provide a new level of control over domain states in pinwheel ASIs. We also show that by a modification to the field protocol, we can reverse the domain growth. Domain growth and reversal by global field protocols is also shown experimentally, using XMCD-PEEM and MOKE microscopy.

## References

[1] R. Macêdo et al. *Phys. Rev. B* **98** (2018) 014437.
[2] J. Jensen et al. *Phys. Rev. B* **106** (2022) 064408.

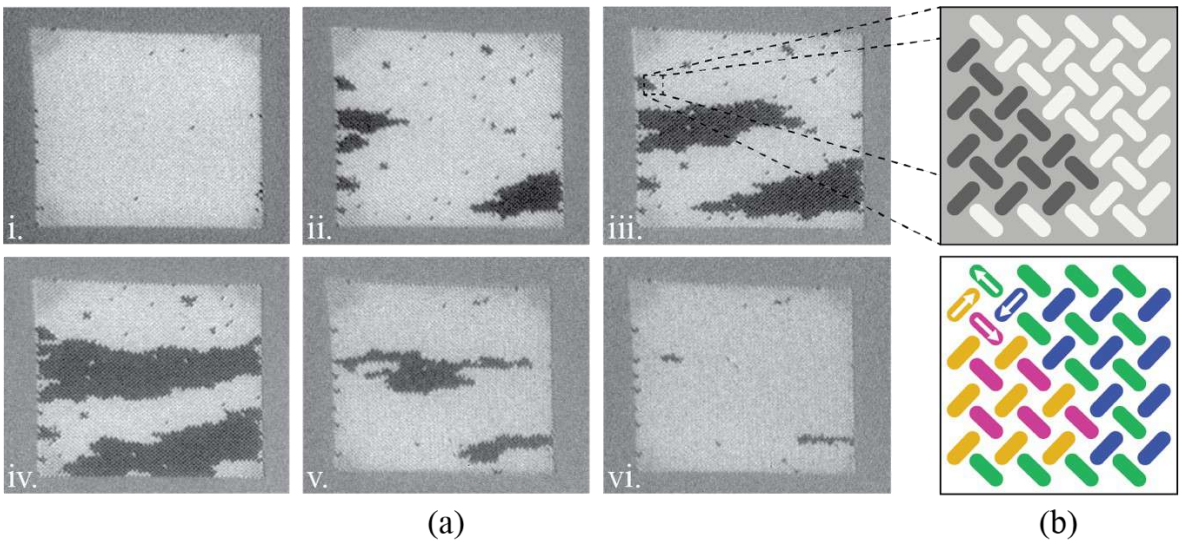



**Figure 1** (a) XMCD-PEEM micrographs of the pinwheel ASI. Dark domains are grown from the light polarized state (i.-iv.) by a global field protocol, which can be modified to reverse the domain growth through a different set of domain states (v.-vi.). (b) Close-up schematic of part of the magnetic state shown in iii., as seen in the experimental XMCD-PEEM micrographs and with the magnetization of all components.

# Topological Effects and Frustration in Artificial Spin Ices to Low Dimensional Effects in Superconducting Nanostructures

*Ana Parente[1]*, M. Abuín[2], A. Rivelles[2], R. Guedas[2], I. Figueruelo[3], G. Caballero[3], M. Menghini[3], A. Muñoz Noval[1,3] and E. M. González[1,3]*

[1] *Department of Material Physics, Facultad CC. Fisicas UCM, 28040 Madrid, Spain*
[2] *ISOM, Universidad Politécnica de Madrid, Avda. Complutense 30, 28040 Madrid, Spain*
[3] *IMDEA Nanociencia, Calle Faraday 9, Campus de Cantoblanco, 28049 Madrid, Spain*
*aparente@ucm.es

On one hand, artificial spin ices (ASIs) are examples of magnetic interacting nanostructures which have opened a way to study topological phenomena such as frustration, emergent magnetic monopoles and phase transitions. [1] Geometric properties of the ASI are key to determine the dynamics of the magnetic charges and the possible energetic configurations, which can also have an influence on the magnetic textures present in these systems. One the other hand, due to the miniaturization of electronic devices, quantum mechanical effects become important. A fundamental question is how the collective properties of these devices, such as superconductivity, are affected when the systems dimensions are reduced.[2] In this context, low dimensional effects in superconducting nanostructures have been studied.

The main goal of this work is to characterize the magnetic properties, spin textures and frustration in ASIs with different geometries and its influence in the formation and ordering of magnetic features. ASIs with different geometries and types of topological protection will be presented. Furthermore, this work study how the fabrication process of superconducting nanostructures based on ASIs influences the superconducting properties of these nanostructures.

Different geometries of ASI systems and superconducting nanostructures had been fabricated by combining nanolithographic techniques (optical and electronic lithography) and DC magnetron sputtering. The fabricated nanostructures were characterized using different microscopy techniques including SEM, AFM and MFM (Figure). The energetically most favorable magnetic structures in ASIs with different geometries (and following different magnetic field protocols) were studied by micromagnetic simulations. Transport measurements of the superconducting nanostructures at low temperature allows the distinction of two different regimes due to lateral confinement: nanowire regime and thin film regime.

## References

[1] O. Ambriz et. al. *Rev. Mod. Phys*, **91**, 041003 (2019)
[2] L. Chen et al. , *ACS Nano,* **14** (2020)

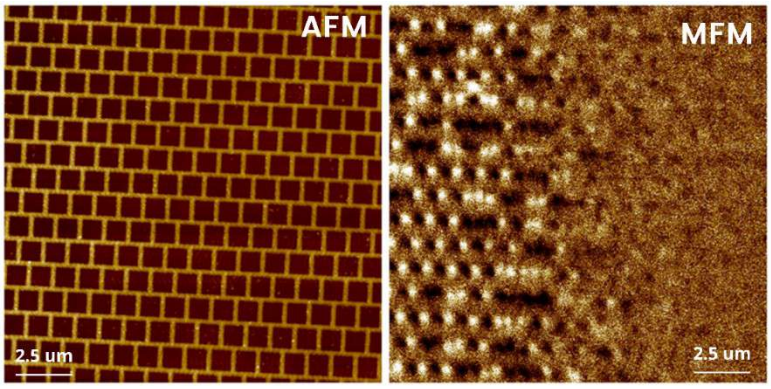



Figure: AFM/MFM image of an artificial spin ice with a decreasing gradient in thickness.

# Multipolar exchange interactions, ordered phases and magnetic excitations in spin-orbit double perovskites

*Leonid V. Pourovskii [1,2,*], Dario Fiore Mosca[1,2,3], Cesare Franchini [3,4]*

[1] *CPHT, CNRS, Ecole polytechnique, Institut Polytechnique de Paris, 91120 Palaiseau, France*
[2] *Collège de France, 11 place Marcelin Berthelot, 75005 Paris, France*
[3] *Faculty of Physics and Center for Computational Materials Science, University of Vienna, Vienna 1090, Austria*
[4] *Department of Physics and Astronomy "Augusto Righi", Alma Mater Studiorum - Università di Bologna, Bologna 40127, Italy*
*leonid@cpht.polytechnique.fr

The double perovskites (DP) of the type $ABB'O_6$ (where $B'$ is a 4$d$ or 5$d$ magnetic ion, while $A$ and $B$ are non-magnetic cations) exhibit a range of unusual low-temperature ground states and peculiar magnetic excitation spectra. These properties are belived to stem from a large value of spin-orbit coupling for partially filled $d$ shells on $B'$ ions leading in many cases to spin-entangled atomic ground states. In the case of $d^2$ occupancy the ground state has an effective anglular momentum $J$=2 and a rich space of possible order parameters. In particular, a multipolar state has been suggested for the spin-orbit coupled $d^2$ DPs $Ba_2BOsO_6$ ($B$=Ca, Mg, Zn). However, whether their ground state is a Janh-Teller–distorted order of quadrupoles or an exotic octupolar order remains debated. We directly calculate all-rank intersite exchange interactions for this double-perovskite series by means of a many-body ab initio force theorem [1]. The ground state order and inelastic neutron scattering cross section are subsequently derived from this ab initio effective Hamiltonian. Our calculations predict the ground state ferro-ordered "$xyz$" octupoles coupled by superexchange interactions within the ground-state doublet [2]. Minuscule distortions of the parent cubic structure are shown to qualitatively modify the structure of gaped magnetic excitations. In $d^3$ DPs, the Hund's rule coupling induces a spin-3/2 orbital-singlet ground state. The spin-orbit interaction is not expected to qualitatively impact low-energy degrees of freedom in such systems. Indeed, $d^3$ DPs of heavy transition metals are believed to exhibit conventional collinear magnetic orders. However, their inelastic neutron scattering spectra feature large gaps of unclear origin. We derive first-principles low-energy Hamiltonians for the cubic DP $Ba_2YB'O_6$ ($B'$= Os, Ru) and show that they include significant multipolar – dipole-octupolar – intesite exchange terms [3]. These terms break continuous symmetry of the spin-3/2 Hamiltonian opening an excitation gap. The dipole-octupolar intersite exchange is induced due to excited states of the $t_{2g}^3$ manifold that are admixed by spin-orbit into the spin-3/2 ground state.

## References

[1] L. V. Pourovskii, *Phys. Rev. B* **94** (2016) 115117.
[2] L. V. Pourovskii, D. Fiore Mosca, C. Franchini, *Phys. Rev. Lett.* **127** (2021) 237201.
[3] L. V. Pourovskii, *arXiv*:2212.03358.

# Scanning SQUID Microscopy Study of the Suppression of the Ferromagnetism in $LaMnO_3$ by Metallic Overlayers

*Carlos M. M. Rosário[1,2]*, Bart Folkers[1], Thijs Roskamp[1], Thies Jansen[1], Hans Hilgenkamp[1]*

[1]*Faculty of Science and Technology and MESA+ Institute for Nanotechnology, University of Twente, Drienerlolaan 5 7522 NB, Enschede, The Netherlands*

[2] *International Iberian Nanotechnology Laboratory, Avenida Mestre José Veiga s/n 4715-330, Braga, Portugal*

*carlos.rosario@inl.int

While $LaMnO_3$ (LMO) is an antiferromagnet in the bulk, thin films deposited on $SrTiO_3$ (STO) usually exhibit ferromagnetism with a Curie temperature of approximately 115 K, accompanied by electrically insulating behaviour [1]. The thickness of the LMO plays a significant role in the emergence of the ferromagnetic behaviour in LMO/STO heterostructures [2]. A critical minimum thickness for ferromagnetic behaviour of 6 unit cells was observed. This effect was clearly visible using Scanning SQUID Microscopy (SSM), where a SQUID chip is scanned along the surface of a sample. This is a powerful tool to image the ferromagnetism with a micrometer scale resolution [3].

We report the use of SSM to study the suppression of the ferromagnetism in metal/LMO/STO structures obtained by pulsed laser deposition of LMO on STO and sputtering of the metals. By only partially covering the LMO surface with the metallic layers, the SSM is capable of showing in the same image both the covered and uncovered regions of the LMO, and thus a clear picture of the effect of adding the metal/LMO interface. While Au alone does not influence the ferromagnetic order of the underlying LMO film, a thin oxygen-scavenging Ti layer induces a suppression of the stray magnetic field picked up by the scanning SQUID. A 4 nm-thick Ti layer completely suppresses the magnetic signal stemming from the underlying LMO thin film with 20 unit cells (Figure 1a). On the other hand, Ti films with the same thickness on top of 50 unit cells of LMO do not completely suppress the ferromagnetism. However, we observe for the latter case that the level of suppresion of the measured magnetic signal increases over time on a timescale of several days.

By depositing Ti on top of LMO and leaving some micrometer-sized uncovered areas, using conventional photolithography, we could show effective patterning of the ferromagnetism on the LMO films down to the microscale. Circular uncovered regions with a diameter of 5 µm exhibit a behaviour typical of single ferromagnetic domains with in-plane orientation, evidencing the possibility of occurrence of large ferromagnetic domains in LMO (Figure 1b). These results could enable a local control of the ferromagnetism in LMO for potential applications in oxide electronics and spintronics.

## References

[1] A. Gupta et al., *Appl. Phys. Lett.* **67** (1995) 3494.
[2] X. Renshaw Wang et al., *Science* **349** (2015) 716.
[3] P. Reith, X. Renshaw Wang, H. Hilgenkamp, *Rev. Sci. Instrum.* **88** (2017) 123706.

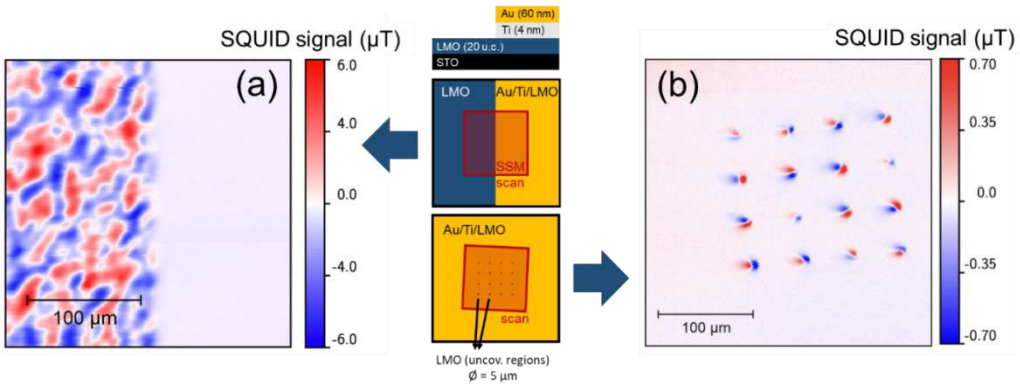



Figure 1: Scanning SQUID microscopy images of 20 u.c. LMO thin films partially covered by 4 nm-thick Ti: (a) half covered sample. (b) Array of circular holes in the Ti layer (uncovered LMO) with a diameter of 5 µm.

# Magnetocrystalline Anisotropy in Ferromagnetic Fe-Sn Kagome Magnets

*L. Prodan[1]*, M. Hemmida[1], H-A. Krug von Nidda[1], L. Chioncel[2], V. Tsurkan[1,3], I. Kézsmarki[1]*

[1] *Experimental physics V, Institute of Physics, Augsburg University, D-86135 Augsburg, Germany*

[2] *Theoretical physics III, Institute of Physics, Augsburg University, D-86135 Augsburg, Germany*

[3] *Institute of Applied Physics, MD 2028, Chisinau, R. Moldova*

*lilian.prodan@physik.uni-augsburg.de

Magnetic materails with kagome-lattice arrangement of atoms have recently attracted much attention due to their unusual magnetic and electronic properties related to the specific topology of their electronic band structures. It was theoreticaly predicted and experimetally proved that flat bands, nodal points and nodal lines appearing close to the Fermi energy significantly affect magnetotransport and magneto-optical properties of kagome materials and provide an ideal platform to study electronic band topology and its interplay with magnetic spin and orbital effects [1].

Recently, particular attention was paid to the Fe-Sn itinerant kagome magnets with different stacking sequences of kagome layers. Depending on the stacking these compounds realize various magnetic ground states and exhibit nontrivial features like large anomalous Hall and Nernst effects and room-temperature skyrmion bubbles being of high interest for both fundamental research and application [2-4].

Here we report detailed static-magnetometry and electron-spin resonance studies of the magnetocrystalline anisotropy performed on high-quality bulk single crystals of $Fe_3Sn$ and $Fe_3Sn_2$, complemented with DFT calculations of the spin and orbital moments as well as anisotropy parameters. In $Fe_3Sn$ with one kagome bilayer structure a strong easy-plane anisotropy was found with the first anisotropy constant $K_1 = -1.3 \times 10^6$ J/m$^3$ at 2 K [5]. For $Fe_3Sn_2$ with two kagome bilayers structure, the uniaxial anisotropy constant shows a sign change at about 85 K, which corresponds to a second order spin-reorientation transition from easy-plane (at 2 K) to easy-axis (at 400 K) anisotropy. The self-consistent DFT computations for the components of the spin/orbital moments indicate that the small difference between the saturation magnetisations measured along and perpendicular to the kagome layers results from the subtle balance between the Fe and Sn spin/orbital moments on the different sites.

**Acknowledgements**

This work was supported by the DFG through TRR 80, and by the project ANCD 20.80009.5007.19.

**References**

[1] H. Zhang, H. Feng, X. Xu, W. Hao, and Y. Du, Advanced Quantum Technologies **4**, 2100073 (2021).
[2] L. Ye, M. Kang, J. Liu, *et al.,* Nature **555**, 638 (2018).
[3] T. Chen, S. Minami, A. Sakai, *et al*., Science Advances **8**, eabk1480 (2022).
[4] M. Altthaler, E. Lysne, E. Roede, *et al*., Phys. Rev. Research **3**, 043191 (2021).
[5] L. Prodan, D. M. Evans, S. M. Griffin, *et al*., arXiv:2302.13810 (2023).

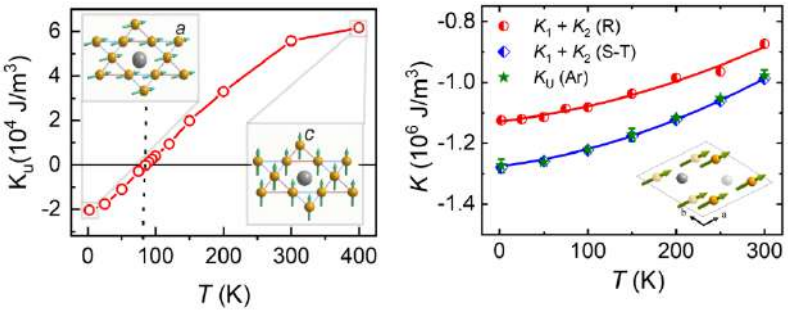



Figure: The anisotropy constants for $Fe_3Sn_2$ (left panel) and $Fe_3Sn$ (right panel). The insets show the magnetic spin arrangement of Fe atoms.

# Investigating The Electronic Charge And Magnetic Spin Dynamics In $HgCr_2Se_4$ Using Resistance Fluctuation (Noise) Spectroscopy

*Charu Garg[1]*, Zhilin Li[2], Youguo Shi[2], Jens Müller[1]*

[1] *Institute of Physics, Goethe University Frankfurt, 60438 Frankfurt (Main), Germany*

[2] *Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China*

* c.garg@physik.uni-frankfurt.de

The n-type $HgCr_2Se_4$ has been reported to exhibit a pronounced semiconductor-to-metal transition below and a colossal magnetoresistance (CMR) effect at the ferromagnetic transition temperatura at $T_C$ = 107 K. Our recent study of the charge carrier dynamics using resistance fluctuation spectroscopy [1] suggests that isolated magnetic polarons form at $T > 2T_C$, which coalesce at $T_C$. Below this temperature, the trapped carriers in magnetic polarons are unbound in the presence of a magnetic field. The peculiar slow dynamics around this percolation transition has been discussed using a model where the effective radius of the polarons is strongly influenced by the spin correlation length close to $T_C$. In this talk, we discuss new results highlighting the strong correlation between the magnetic and electronic degrees of freedom that can lead to complex exchange pathways. Likely due to competing antiferromagnetic and ferromagnetic interactions, we observe a distinctly slow decrease in resistance below the CMR transition. The striking dynamics of distinct two-level fluctuations superimposed on 1/f-type noise corroborates a slowing down of charge carrier and/or magnetic dynamics. Further, below 20 K, a strong upturn in resistance and simultaneously in resistance noise down to 500 mK is observed and is speculated to be linked to the emergence of spiral type magnetic order. Our results demonstrate that the presence of pronounced electron-spin correlations plays a key role in the unconventional temperature dependence of resistance and CMR effect in this spinel.

## References

[1] Mitschek *et al*., Phys. Rev B 105, 064404 (2022).
[2] Lin *et al*., Phys. Rev B 94, 224404 (2016).

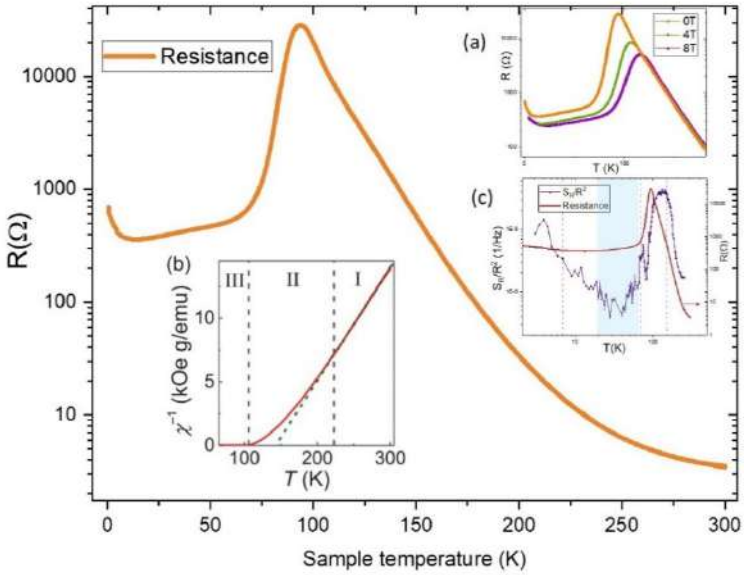



Figure: Temperature dependent resistance of $HgCr_2Se_4$. Below the CMR and ferromagnetic transition ($T_c$ = 107K), there is a gradual change in resistance below 65K and a strong upturn below 20K down to 500mK. The inset shows (a) magnetic field dependence of resistance, (c) temperature dependence of the inverse susceptibility ($\chi^{-1}$) is shown for an applied magnetic field of 0.01T [2]. The strong deviation from Curie-Weiss law starting ~T= 220K ($T > 2T_C$) strongly suggests the presence of isolated magnetic polarons, and (c) the change in resistance (red symbols), and normalized resistance noise (PSD) $S_R/R^2$(f = 1 Hz) (purple) of the 1/ f type (and Lorentzian noise) with temperature.

# Magnetoelastic Properties of $MnSc_2S_4$

*M. Doerr[1]*,M. Deeb[1], J. Grumbach[1], M. Rotter[2], V. Tsurkan[3,4], A. Miyata[5], S. Zherlitsyn[5], J. Wosnitza[1,5]*

[1] *Institut für Festkörper- und Materialphysik, TU Dresden, 01062 Dresden, Germany.*
[2]*McPhase-project, 01159 Dresden, Germany.*
[3]*Experimental Physics V, University of Augsburg, 86135 Augsburg, Germany.*
[4]*Institute of Applied Physics, Academy of Sciences Moldova, MD-2028 Chisinau, Republic of Moldova.*
[5]*Hochfeld-Magnetlabor Dresden (HLD-EMFL) and Würzburg-Dresden Cluster of Excellence ct.qmat, Helmholtz-Zentrum Dresden-Rossendorf, 01328 Dresden, Germany.*
*mathias.doerr@tu-dresden.de

Magnetic skyrmions, which are topologically stabilized spin configurations induced by anisotropic exchange interactions or crystallographic frustrations, are currently attracting attention due to their rich fundamental physics and their high potential for applications in quantum technology. The cubic spinel $MnSc_2S_4$ where corner-sharing tetrahedra form a frustrated lattice is known to exhibit an antiferromagnetic skyrmion structure in magnetic field [1,2].

Due to the strong magnetoelastic coupling, this interesting phenomenon is closely related to lattice effects. Here, we discuss lattice changes of $MnSc_2S_4$ single crystals, measured by thermal expansion and magnetostriction as function of temperature and magnetic field using high-resolution dilatometry. The variations of the lattice parameters in the order of $10^{-5}$ show that especially the skyrmion phase between 4 and 8 T induces significant lattice-structural effects. Based on the new data, we confirmed and refined the magnetic (*H,T*) phase diagram. The experimental results are complemented by a mean-field simulation using the program package *McPhase*. On the basis of a hamiltonian containing an exchange, a Zeeman, and a magnetoelastic term, we reproduced the magnetic (*H,T*) phase diagram reasonably well, as well as various magnetostriction curves via the correlation function. In addition to the antiferromagnetic skyrmion phase, the theoretical analysis also reflects the intermediate spin order in the range between 1.5 and 2.0 K.

**Acknowledgements**

We acknowledge support from Helmholtz-Zentrum Dresden-Rossendorf (HZDR), especially the Hochfeld-Magnetlabor Dresden at HZDR, member of the European Magnetic Field Laboratory (EMFL). This research was supported by the DFG through SFB1143 (Project-id 247310070).

**References**

[1] S. Gao *et al.*, *Nature* 586 (2020) 37.
[2] Y. Iqbal *et al.*, *Phys. Rev. B* 98 (2018) 064427.

# Spin pumping in YIG/s-wave superconductor hybrids

*S. J. Carreira[1]*, H. Hassan[1], S. Menouni[1], C. Carretero[1], S. Collin[1], H, Jaffres[1], M. Anane[1], J. E. Villegas[1]*

[1] *Unité Mixte de Physique, CNRS, Thales, Université Paris-Saclay, 91767, Palaiseau, France*

*scarreir@ucm.es

Spin pumping into superconductors (SC) constitutes a very active research topic within the nascent field of superconducting spintronics, aiming at expanding spintronic functionalities by exploiting the dissipationless electron transport and quantum coherence characteristic of superconductivity [1]–[4]. In this context we analyzed the spin absorption efficiency of s-wave superconductors (Nb and $Mo_{80}Si_{20}$) in ferromagnetic resonance (FMR) and inverse spin Hall effect (ISHE) measurements, using as spin source the ferrimagnetic insulator Yttrium Iron Garnet $Y_3Fe_5O_{12}$ (YIG).

The spin injection analyzed in terms of FMR experiments shows a drop in the magnetic damping at the critical temperature Tc due to the opening of the superconducting gap. However, the ISHE signal reveals an exponential increment (a peak) at the critical temperature, which can be associated to the coherence peak of the quasiparticle's density-of-states at the FM/SC interface [5], [6]. We studied the thickness dependence of the ISHE peak and found that the signal drops significantly above a threshold SC thickness, revealing the central role played by the spin duffusion length in the system.

## Acknowledgements

Work supported French ANR grant ANR-15-CE24 0008-01 "SUPERFAST", COST action "SUPERQUMAP" and EU H2020 Marie Curie Actions Nº 890964 "SUPERMAGNONICS".

## References

[1] F. S. Bergeret, A. F. Volkov, and K. B. Efetov, *Phys. Rev. Lett*. **86** (2001) 4096–4099.
[2] M. G. Blamire and J. W. A. Robinson, *J. Phys.: Condens. Matter*. **26** (2014) 453201.
[3] J. Linder and J. W. A. Robinson, *Nat. Phys.* **11** (2015) 307–315.
[4] M. Eschrig, *Rep. Prog. Phys*. **78** (1015) 104501.
[5] F. S. Bergeret, M. Silaev, P. Virtanen, and T. T. Heikkilä. *Reviews of Modern Physics*, **90** (2018) 41001.
[6] K.-R. Jeon, J.-C. Jeon, X. Zhou, A. Migliorini, J. Yoon, and S. S. P. Parkin, *ACS Nano* **14** (2020) 15874–15883.

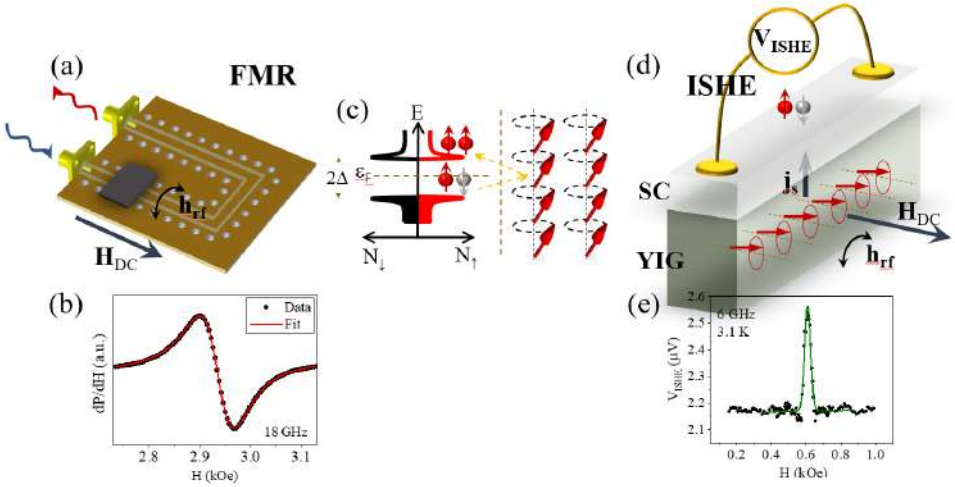



Figure: (a) Complanar waveguide, (b) typical FMR lineshape, (c) SC density of states and qusiparticles excitation above the Fermi energy, (d) Sketch of the ISHE measurement geometry and (e) Typical ISHE voltage obtained on the SC across the resonance of the YIG.

# Universal stripe order as a precursor of the superconducting phase in $BaFe_2Se_3$ spin ladder

*V. Balédent[1]*, W. Zheng[1], D. Colson[2], A. Forget[2], C. Colin[3], J.-P. Rueff[4], P. Foury-Leylekian[1]*
*[1]Université Paris-Saclay, CNRS, Laboratoire de Physique des Solides, 91405, Orsay, France*
*[2]SPEC, CEA, CNRS-UMR3680, Université Paris-Saclay, Gif-sur-Yvette, France*
*[3]Institut Néel, Université Grenoble Alpes, CNRS, Grenoble, F-38042, France*
*[4]Synchrotron SOLEIL, L'Orme des Merisiers, BP 48 St Aubin, 91192 Gif-sur-Yvette, France*

In the last years, superconductivity has been observed in iron based one dimensional compounds, $BaFe_2X_3$ X=S,Se [1-3]. They have a centrosymmetric average space group (Cmcm and Pnma [4] respectively) at room temperature, in which iron atoms form two distinct ladders along the b-axis. Upon pressure, a metalization is observed and superconductivity develops below 14 K above 10 GPa. A significant difference between the two systems is their magnetic ground state at low pressure. The magnetic order of $BaFe_2Se_3$ takes the form of staggered ferromagnetic blocs along the ladder [6], while for $BaFe_2S_3$ a conventional antiferromagnetic order is formed along the ladder. These different magnetic symmetries raise the question of the role of magnetism in the mechanism leading to superconductivity. One would indeed expect a similar magnetic order for both compounds allowing a common and universal mechanism.

Our recent neutron scattering results on the compounds $BaFe_2Se_3$ revealed a new picture of the magnetic properties from ambient pressure up to the superconducting critical pressure [5,6]. Using single crystal and powder neutron diffraction on $BaFe_2Se_3$, we refined the magnetic structure at ambient pressure and discovered a new magnetic order similar to the $BaFe_2S_3$ stripe phase near the superconducting dome. This magnetic phase thus appear to be universal for this family, giving a strong indication of its role in the origin of superconductivity.

**References**

[1] H. Takahashi et al., Nature Materials 14, 1008 (2015).
[2] J. Ying et al., Physical Review B 24, 241109 (2017).
[3] J.M. Caron et al., Physical Review B 18, 180409 (2011)
[4] W. Zheng, V. Balédent et al., Rapid Communication Physical Review B 101, 020101(R) (2020)
[5] W. Zheng, V. Balédent et al. Nature Communication Physics 5, 183 (2022)
[6] W. G. Zheng, V. Balédent et al. Physical Review B 107, 024423 (2023)

# Charge Order and Time-reversal Symmetry in Kagome Superconductors

*Pietro Bonfà[1]*, Jonathan Frassineti[2], Giuseppe Allodi[1], Erick Garcia[3], Rong Cong[3], Brenden R. Ortiz[4], Andrea Capa Salinas[4], Stephen D. Wilson[4], Roberto De Renzi[1], Vesna F. Mitrovìc[3], Samuele Sanna[2]*

[1] *Department of Mathematical, Physical and Computer Sciences, University of Parma, Parco Area delle Scienze 7/A, 43124 Parma, Italy*

[2] *Department of Physics and Astronomy "A. Righi", University of Bologna and INFN Sezione di Bologna, via Berti Pichat 6/2, 40127 Bologna, Italy*

[3] *Department of Physics, Brown University, Providence, Rhode Island 02912, USA)*

[4] *Materials Department and California Nanosystems Institute, University of California Santa Barbara, Santa Barbara, California 93106, USA)*

*pietro.bonfa@posteo.net

Kagome compounds with formula $AV_3Sb_5$ (A = Cs, Rb, K) [1] are nonmagnetic layered materials that display two electronic transitions: a charge-density wave (CDW) order followed by superconductivity at lower temperature. The interplay between the two phenomena has been the subject of intense research, but the nature of the CDW phase and its relation to superconductivity remains elusive. Here we present a study which combines Nuclear Magnetic Resonance (NMR) measurements and Density Functional Theory calculations to unveil the structure of the CDW of the $RbV_3Sb_5$ [2] and $KV_3Sb_5$ compounds. The results unambiguously identify the lattice and the electronic structure in the CDW phase. We finally extend the study to the analysis of the low temperature behaviour where the internal field width sensed by the muon ensemble in muon spin spectroscopy experiments and the line widths probed by nuclear magnetic resonance have been previously reported to be strongly enhanced, even in the superconducting phase [3,4].

## References

[1]B. R. Ortiz et al., Phys. Rev. Materials 3 (2019), 094407
[2]J. Frassineti et al., Phys. Rev. Research 5 (2023) L012017
[3]Y. M. Oey et al., Phys. Rev. Materials 6 (2022), L041801
[4]J. Frassineti et al., in preparation

# Spin effects on the nonequilibrium thermoelectrics of a correlated quantum dot

*Anand Manaparambil*[1]*, *Ireneusz Weymann*[1]

[1] *Institute of Spintronics and Quantum Information, Adam Mickiewicz University, Uniwersytetu Poznanskiego 2, Poznan, Poland*

*anaman@amu.edu.pl

Thermoelectric properties of nanostructures with strong correlations have been a focus of intensive studies. This interest originates primarily from the prospect of using zero-dimensional materials such as quantum dots or molecular junctions as a part of efficient heat engines. Moreover, the thermoelectric properties of quantum dot systems have been shown to contain the signatures of Kondo correlations in the system. Such systems have been thoroughly studied in the linear response regime, but extension to the nonlinear regime remains rather less explored. In this work, we study the nonlinear thermoelectric properties of a quantum dot asymmetrically coupled to metallic/magnetic leads. Using perturbation theory on the weak coupling and solving the strongly coupled subsystem using Numerical Renormalization Group (NRG) as it was in equilibrium, one can define the current through the system at any arbitrary nonequilibrium potential and/or thermal bias setting [1]. In particular, we explore how the exchange field induced by the ferromagnetic lead affect the thermoelectric signatures of the Kondo effect and how the configuration of the metallic/magnetic leads can affect the transport and the ability of the system to work as a heat engine.

**Acknowledgements**

This work was supported by the Polish National Science Centre from funds awarded through the decision Nos. 2017/27/B/ST3/00621 and No. 2021/41/N/ST3/02098. We also acknowledge the computing time at the Poznan Supercomputing and Networking Center.

**References**

[1] A. Manaparambil, I. Weymann, Phys. Rev. B **107**, 085404 (2023).

# Theoretical study of the magnetic properties of the $CoCu_2O_3$ compound

*Julien Lévêque* [1,2], *Elisa Rebolini* [3], *Marie-Bernadette Lepetit* [2,3*], *and Andrés Saúl* [1†]

[1] *Aix Marseille Univ, CNRS, CINAM, Marseille, France*

[2] *Univ. Grenoble Alpes, CNRS, Grenoble INP, Institut Néel, 38000 Grenoble, France*

[3] *Institut Laue Langevin, Grenoble, France*

* marie-bernadette.lepetit@neel.cnrs.fr

† andres.saul@cnrs.fr

Over the last decades cuprates have attracted a lot of attention following the discovery of high temperature superconductivity. Apart from superconductivity, a large number of compounds of the cuprate family have been studied for their low-dimensional quantum magnetism. Indeed, the S = 1/2 character of the $Cu^{2+}$ ion, and the directionality of the associated 3d magnetic orbital, are responsible for a tendency to form one- or two-dimensional magnetic systems with quantum character. This leads to high exchange integrals and thus to a high magnetic ordering temperature.

Among them, the $ACu_2O_3$ family (A = Ca, Mg, Co) presents a 3D magnetic order associated with high transition temperatures. Although this family shows very similar structures characterized by puckered layers, the magnetic order and the associated transition temperature changes with the element A [1,2]. In particular, the compound $CoCu_2O_3$ presents the highest magnetic transition temperature of this family (215 K) [3].

In this presentation, we study the magnetic properties of this compound using ab-initio and classical Monte-Carlo calculation methods. The exchange integrals have been calculated with a MRCI method allowing to treat exactly the electronic correlation [4]. We find that the Co atom plays a preponderant role in the magnetic properties of this compound. Indeed, as a magnetic ion, it creates additional magnetic interactions. This results in a magnetic pattern based on coupled three-leg ladders, different from the two-leg structural ladders. Moreover, we show that the observed high transition temperature is related to its single ion anisotropy [5].

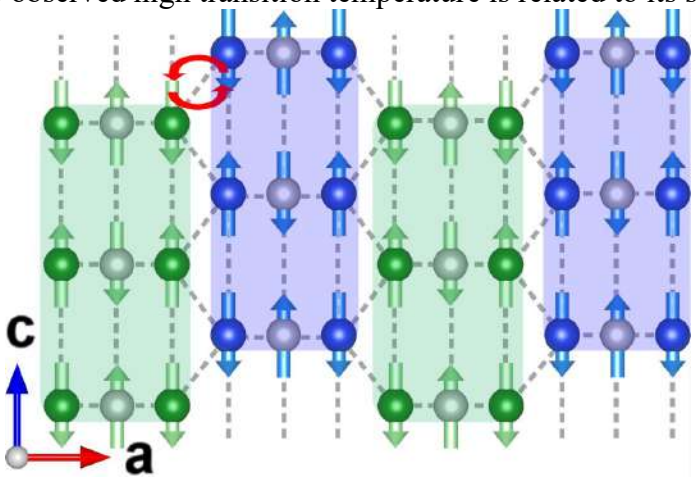



Figure: Schematic of the magnetic order of $CoCu_2O_3$ on the puckered magnetic layers.

**Acknowledgements**

We acknowledge financial support from the ANR HTHPCM. This work was performed using computer resources obtained from GENCI-IDRIS and CRIANN under Projects No. 91842 and No. 2007013.

**References**

[1] M. Winkelmannet al., PRB 49, 310 (1994).
[2] V. Kiryukhin et al., PRB 63, 144418 (2001).
[3] C. Sekar et al., JMMM 323, 3033 (2011)
[4] A. Gellé et al., Europhys. Lett. 88, 37003 (2009)
[5] J. Lévêque et al., PRB 106, 224402 (2022)

# SYMPOSIUM 09.
## STRONGLY CORRELATED SYSTEMS, SUPERCONDUCTIVITY AND FRUSTRATED AND DISORDERED MAGNETISM.
## S9. POSTERS

# NMR observation of short-range charge order fluctuations in kagome materials $AV_3Sb_5$

*Xin Zhang[1,2], Yongkai Li[1], Xuekui Xi[*2], Yong-Chang Lau[2], Zhiwei Wang[*1], Wenhong Wang[3]*

*[1] Department of physics, Beijing Institute of Technology, Beijing 100190, China*

*[2]Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, CAS Beijing 100190, China*

*[3]Tiangong University, Tianjin 300387, China*

3120215767@bit.edu.cn

In kagome materials $AV_3Sb_5$(A = Rb, Cs), we observed a signature of short-range order charge density fluctuations [1] even at well above long-range order, i.e., charge density wave (CDW) formation temperature through solid state $^{51}V$ nuclear magnetic resonance (NMR) spectroscopy. The emergence of short-range charge order fluctuations not only changes the electronic density of states near the Fermi surface, as revealed by dynamic crossover of NMR shifts, but also corresponds to the observed in-plane translational symmetry breaking of the lattice. Such short-range order charge fluctuations can also be found in other two-dimensional materials, indicating it should be a common feature for CDW phase transitions. It is worth noting that a similar crossover behavior was observed by NMR at temperatures above the so-called nematic transition point, suggesting that the nematic phase also embraces short-range order fluctuations.

## Acknowledgements

This work is financially supported by the National Key R&D Program of China (No. 2021YFB3501402) and National Natural Science Foundation of China (No. 51831003).

## References

[1] Q. Chen, D. Chen, W. Schnelle, C. Felser, and B. D. Gaulin, Charge Density Wave Order and Fluctuations above T-CDW and below Superconducting T-c in the Kagome Metal CsV3Sb5, Phys. Rev. Lett. **129**, 056401 (2022).

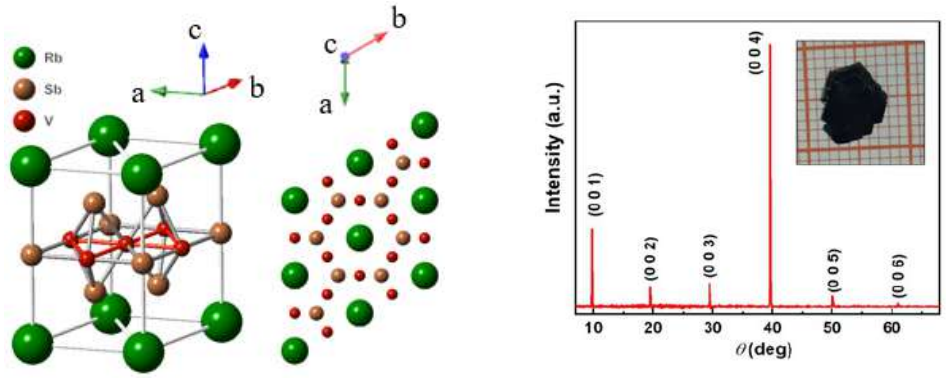



Figure: (Left) Structure of $RbV_3Sb_5$. (Right) X-ray diffraction and (inset) a photo of $RbV_3Sb_5$ single crystal.

# Theory of Absorption in the Shastry-Sutherland Model for $SrCu_2(BO_3)_2$

*Shin_Miyahara**

*Department of Applied Physics, Fukuoka University, 8-19-1 Nanakuma, Jonan-ku, Fukuoka, Japan*

*smiyahara@fukuoka-u.ac.jp

The magnetic properties of quasi two-dimensional material $SrCu_2(BO_3)_2$, *e.g.* spin gap excitations and magnetization plateaus, can be explained well by the Shastry-Sutherland model, of which the ground state is rigorously described as a direct product of dimer singlet states, and have been studied both from experimental and theoretical point of view [1]. Spin excitations have been investigated by ESR and far-infrared spectroscopy and one- and two-triplet excitations have been observed. However, the comprehensive understanding of the origin of the excitations, the mechanism, and the selection rule has not yet been established.

We introduce the magnetoelectric couplings, which can excite an electromagnon, *i.e.*, an electro active magnon in multiferroics materials [2], to the Shastry-Sutherland model with Dzyaloshinskii-Moriya interactions for $SrCu_2(BO_3)_2$ and successfully explain the results of ESR and far-infrared spectroscopy.

**Acknowledgements**

This work was supported by Japan Society for the Promotion of Science (JSPS) KAKENHI Grants No. 22H01171.

**References**

[1] S. Miyahara and K. Ueda, J. Phys.:Condens. Matter 15, R327 (2003).
[2] H. Katsura, A.V. Balatsky, and N. Nagaosa, Phys. Rev. Lett. 98, 027203 (2007).

# Magnetic Phase Diagram and Metamagnetism of Layered Borides $(Fe_{1-x}Mn_x)_2AlB_2$ Single Crystals

*T. Shiotani[1]*, T. Waki[1], Y. Tabata[1], and H. Nakamura[1]*

[1] *Department of Materials Science and Engineering, Kyoto University, 606-8501, Kyoto, Japan*

*shiotani.taiki.48e@st.kyoto-u.ac.jp

Layered materials exhibit a wide variety of novel physical properties originating from its two-dimensional (2D) structure, having potential to significantly improve device performance. However, for practical application, they also need environmental stability such as heat resistance and high hardness.

$M_2AlB_2$, crystalizing in the orthorhombic *Cmmm* structure, has an alternating stacking of Al layer and $(MB)_2$ slab along *b* axis. This structural motif leads both anisotropic conductivity and environmental stability [1]. In addition, in the case of *M* = Fe and Mn, $M_2AlB_2$ exhibits itinerant magnetism of 3*d* electrons in the $(MB)_2$ slab. $Fe_2AlB_2$ is a strong ferromagnet with the Curie temperature $T_C$ = 273 K and easy magnetization axis along *a* axis [2], whereas $Mn_2AlB_2$ is an antiferromagnet with the Néel temperature $T_N$ = 313 K and spins aligned along *b* axis [3]. A previous report on neutron diffraction measurements of solid solution $(Fe_{1-x}Mn_x)_2AlB_2$ suggests the coexistence of ferromagnetic and antiferromagnetic correlations in the range of $x$ = 0.1–0.5 [3]. Therefore, $(Fe_{1-x}Mn_x)_2AlB_2$ is expected to show novel magnetism originating from competition of the magnetic correlation or magnetic anisotropy.

We have succeeded for the first time in synthesizing single crystals in the entire composition range of $(Fe_{1-x}Mn_x)_2AlB_2$ and performed magnetic measurements to obtain its magnetic phase diagram. The ferromagnetic correlation of $Fe_2AlB_2$ becomes weakened with increasing Mn concentration and ferromagnetic ordering disappeared at $x$ = 0.53, whereas the Néel temperature has a maximum at $x$ = 0.65. The spin directions vary significantly by Mn concentration, indicating competition of the magnetic anisotropy. In the range of $x$ = 0.3–0.5, two-step metamagnetic behaviour along *a* axis was observed between $T_N$ and $T_C$. Between the ferromagnetic and antiferromagnetic phases, another phase suggesting the coexistence of both correlations was found. On the other hand, at $x$ = 0.65 and 0.74, one-step metamagnetic transition was observed below $T_N$. In comparison with $x$ = 0.3–0.5, the transition field is insensitive to temperature. The magnetic structure of $(Fe_{1-x}Mn_x)_2AlB_2$ is expected to be sensitive to the composition, temperature and field.

## References

[1] S. Kota, M. Sokol, M. W. Barsoum, *Int. Mater. Rev.* **65** (2020) 226.
[2] T. N. Lamichhane, L. Xiang, Q. Lin, T. Pandey, D. S. Parker, T.-H. Kim, L. Zhou, M. J. Kramer, S. L. Bud'ko, P. C. Canfield, *Phys. Rev. Mater.* **2** (2018) 084408.
[3] T. N. Lamichhane, K. Rana, Q. Lin, S. L. Bud'ko, Y. Furukawa, P. C. Canfield, *Phys. Rev. Mater.* **3** (2019) 064415.
[4] D. Potashnikov, E. N. Caspi, A. Pesach, S. Kota, M. Sokol, L. A. Hanner, M. W. Barsoum, H. A. Evans, A. Eyal, A. Keren, O. Rivin, *Phys. Rev. Mater.* **4** (2020) 084404.

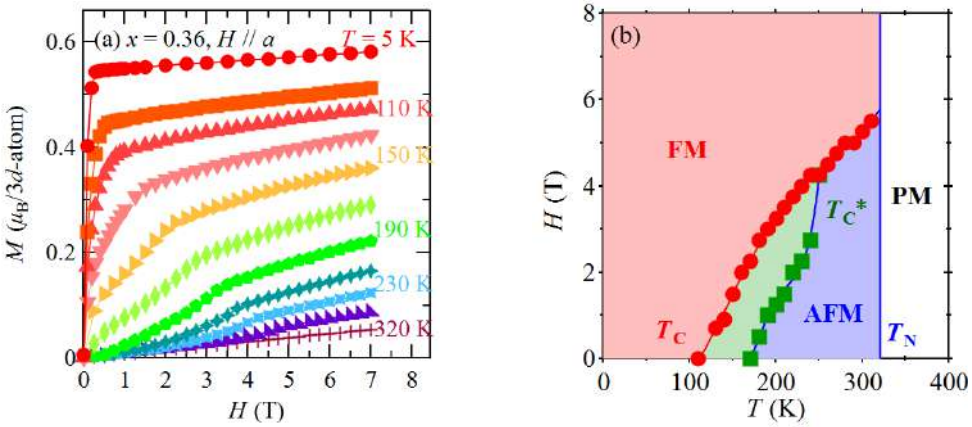



Figure: (a) Field-dependent magnetization and (b) magnetic phase diagram of $(Fe_{0.64}Mn_{0.36})_2AlB_2$ when $H$ // $a$.

# Interplay of Frustration, Strain and Phase Co-existence in the Mixed Valent Hexagonal Iridate $Ba_3NaIr_2O_9$

*Charu Garg[1]*, Antonio Cervellino[2], Sunil Nair[3]*

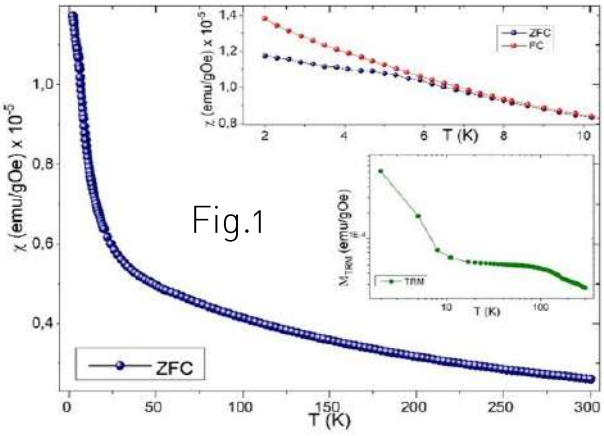
[1] *Institute of Physics, Goethe University Frankfurt, 60438 Frankfurt (Main), Germany*
[2] *Swiss Light Source, Paul Scherrer Institute, CH-5232 Villigen, Switzerland*
[3] *Department of Physics, Indian Institute of Science Education and Research, Dr. Homi Bhabha Road, Pune 411008, India*

* c.garg@physik.uni-frankfurt.de

Geometrically frustrated magnets- where triangular lattice antiferromagnets (TLAFs) are considered to be an archetype remain at the forefront of contemporary condensed matter [1]. Of particular interest in recent years have been a number of Ruthenium and Iridium based perovskite [2,3]. In this talk, we will discuss the structure-property relationship in the mixed valent geometrically frustrated triple perovskite iridate $Ba_3NaIr_2O_9$ using detailed synchrotron diffraction, magnetization, thermodynamic and transport measurements [4]. In contrast to what is expected from purely structural considerations, this system stabilizes in a high symmetry hexagonal symmetry at room temperatures. On reducing the temperature, the lattice prefers to be strained rather than distort to a low symmetry phase, as is the norm in this family of materials. Though a low symmetry orthorhombic phase is nucleated below 50 K, this conversion is only partial and the high symmetry hexagonal structure remains the dominant one down to the lowest measured temperatures. Magnetic measurements indicate an extended co-operative paramagnetic regime, which finally freezes to a cluster glass-like phase at very low temperatures. This makes an interesting addition to the family of triple perovskite iridates where complex interplay between lattice strain and structural phase co-existence arises as a consequence of a number of competing energy scales.

## References

[1] Jason S Gardner, *J. Phys.: Condens. Matter* **23** 160301 (2011)
[2] Charu Garg *et al*., Phys. Rev B 103, 014437 (2021).
[3] Katharine E. Stitzer et al., J. Am. Chem. Soc., 124, 46, 13877–13885 (2002)
[4] Charu Garg *et al., J. Phys.: Condens. Matter* **34** 285602 (2022).



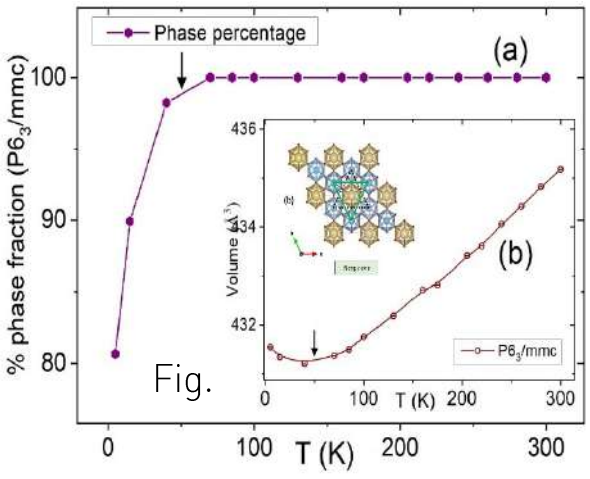




Fig.1: Temperature dependent magnetization at 500 Oe for $Ba_3NaIr_2O_9$. Inset (top): The ZFC-FC splitting close to 6K corresponds to the cluster glass transition. Inset (bottom): TRM measured at 1 kOe with two systematic jumps corresponding to the onset of the co-operative paramagnetic regime, and the cluster glass state respectively
Fig.2 (a): Temperature dependent variation of the phase fraction of the hexagonal ($P6_3/mmc$) phase and Fig.2 (b) volume has been shown. As the temperature decreases, the hexagonal phase gradually transforms to the orthorhombic phase with almost 80% change in the total volume at 5K.

# Coexistence of Two Spin Frustration Pathways in the Quantum Spin Liquid $Ca_{10}Cr_7O_{28}$

*D. Alshalawi[1], M. Pardo-Sáenz[2], K. Ohishi[3], A Hoshikawa[3], J. Campo[2], A. Landa[4], J. M. Alonso[1,4], P. de la Presa[1]**

[1] *Institute of Applied Magnetism, Complutense University of Madrid, 28230 Las Rozas, Spain*
[2] *Aragón Nanoscience and Materials Institute, (CSIC-University of Zaragoza),50009, Zaragoza, Spain*
[3] *Neutron Science and Technology Center, Comprehensive Research Organization for Science and Society (CROSS), 319-1106, Tokai, Japan*
[4] *Material Science Institute of Madrid, 28049 Madrid, Spain*
*pmpresa@ucm.es

Previous studies of $Ca_{10}Cr_7O_{28}$ material demonstrated that it exhibits a range of intriguing properties, including broad temperature-dependent of Quantum Spin Liquid (QSL). It has been proposed that the crystal structure of $Ca_{10}Cr_7O_{28}$ have distorted Kagome bilayers that leads to the high frustration of the spin moments [1,2].

In this work, a detailed investigation of the structural, composition and magnetic properties is presented [3]. The characterizations have been performed with XRD, HRTEM, TGA, calorimetry, neutron diffraction and magnetometry. By means of Rietveld refinements of XRD and neutron diffraction patterns it has been determined that, besides the distorted Kagome bilayers, there exists also a zigzag ladders connecting the Kagome layers. These zigzag ladders could also act as an additional frustration pathway [4]. Additionally, the TGA characterization show that there are six $Cr^{5+}$ and one $Cr^{6}$ in the structure, and these values are in agreement with ToF neutron diffraction analysis, where the oxidation states are calculated with the Bond Valence Sum.

The HRTEM images and analysis reveals that the structure is highly stable, without vacancies, twins or defects, which is surprising considering the large amount of atoms in the unit cells. The magnetic susceptibility show that the whole system could have ferromagnetic interactions [3].

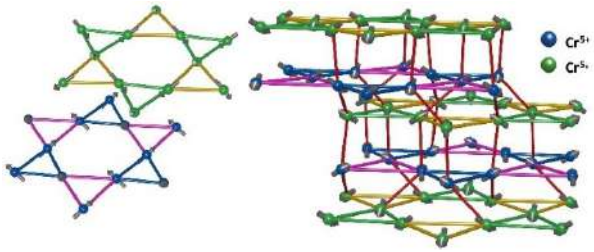



**Fig. 1:** *Kagome layers (left) and the zigzag ladders (red) between magnetic Kagome layers of $Cr^{5+}$(1) and $Cr^{5+}$ (right).*

## Acknowledgements

This work was supported by the Spanish Ministery of Science and Innovtation PID2021-123112OB-C21. We also acknowledge the Spanish Cultural Attaché Office of Saudi Arabia for financial support

## References

1. Balz et al. Nature Physics 12, 942 (2016)
2. Pohle et al. Physical Review B 104, 024426 (2021).
3. Alshalawi et al., Inorganic Chemistry 61, 16228 (2023).
4. Cabra et al. The European Physical Journal B 13, 55 (2000)

# Elastic features in the vicinity of critical end point $UTe_2$

*T.Haidamak[1]*, A.Cabala[1], J.Prokleška[1], A.Miyata[2], M.Vališka[1], V.Sechovský[1]*

[1] *Charles University, Faculty of Mathematics and Physics, Department of Condensed Matter Physics, Ke Karlovu 5, Prague 2, 121 16, Czech Republic*

[2] *Hochfeld-Magnetlabor Dresden (HLD-EMFL) and Würzburg-Dresden Cluster of Excellence ct.qmat, Helmholtz-Zentrum Dresden-Rossendorf, 01328 Dresden, Germany*

*t.haidamak@mag.mff.cuni.cz

Superconductivity (SC) was very recently discovered in the heavy-fermion paramagnet $UTe_2$ [1] which is superconducting in temperature range $T_{sc} = 1.6$ K up to 2K depending on the sample quality[2]. SC is dramatically suppressed by a first-order metamagnetic transition above $H_m = 35$ T for fields applied along the *b*- axis at very low temperatures. This termination of the SC state is associated with a huge increase in electrical resistivity in the field-induced polarized paramagnetic state [3-5]. The metamagnetic field $H_m$ remains almost constant with increasing temperature up to the so-called critical endpoint (CEP) at 11 K, where the first order character changes in the crossover regime [6]. Further increasing of temperature causes a decrease in the $H_m$ value until it finally vanishes around 30 K. It can be extrapolated toward zero magnetic field limit to the value of $T_{max} = 35$ K, the temperature of magnetic susceptibility maximum for field applies along *b*-axis ($H \| b$).

We were able to prepare and characterize well-defined $UTe_2$ single crystals (with dimensions of 2-5 mm along *a*-axis and 0.5mm along c-axis).

We have observed and investigated the magnetoelastic interaction magnetic field applied $H \| b$, in temperature range around critical end point T=7-15 K more in detail. Unexpected sharp anomaly at 11.4 K for 3 geometries which were investigated ($C_{44}$,$C_{55}$,$C_{66}$). All the geometries are transversal which means **q**-elastic wave vector and **u**-displacement vectors are orthogonal to each other. (Geometries are $C_{44}$ qllc ullb, $C_{55}$ qllc ulla, $C_{66}$ qlla ullb, magnetic field was applied along $H \| b$ for all geometries.)

## References

[1] S. Ran et al., *Science* **365** (2019) 684.

[2] D. Aoki et al., *Journal of the Physical Society of Japan* **88** (2019) 043702.

[3] W. Knafo, M. Vališka, D. Braithwaite, G. Lapertot, G. Knebel, A. Pourret, J.-P. Brison, J. Flouquet, and D. Aoki, *Journal of the Physical Society of Japan* **88** (2019) 063705.

[4] S. Ran et al., *Nature Physics* **15**, (2019) 1250.

[5] G. Knebel et al., *Journal of the Physical Society of Japan* **88** (2019) 063707.

[6] A. Miyake et al., *Journal of the Physical Society of Japan* **88** (2019) 063706.

# Impact of the Rare Earth Ions on Magnetic Properties of $RCr_3(BO_3)_4$ Crystals with $La^{3+}$, $Gd^{3+}$ and $Tb^{3+}$ Ions

*Yu. Savina [1], A. Bludov [1], V. Khrustalyov [1], V. Pashchenko [1], A. Lynnyk [2], T. Zajarniuk [2], M.U. Gutowska [2], A. Szewczyk [2], I. Kolodiy [3]*

[1] *B. Verkin Institute for Low Temperature Physics and Engineering of the NAS of Ukraine, 47 Nauky Ave., 61103 Kharkiv, Ukraine*

[2] *Institute of Physics, Polish Academy of Sciences, al. Lotników 32/46, 02-668 Warsaw, Poland*

[3] *National Science Center «Kharkov Institute of Physics and Technology», 1 Akademichna St., 61000 Kharkiv, Ukraine*

savina@ilt.kharkov.ua

Compounds with the general formula $RM_3(BO_3)_4$ ($R$ = Y or rare-earth elements; $M$ = Al, Ga, Fe, Cr, and Sc) attract considerable attention of scientists due to their optical, magnetic and ferroic properties.

The majority of crystals of this family have the huntite mineral structure, which crystallizes in the trigonal crystal system with space group $R32$. The crystal structure consists of spiral chains of sharing edges $MO_6$ octahedra along the $c$ axis. The $RO_6$ prisms and the planar triangular $BO_3$ groups combine the chains into 3D framework. Several other structural polytypes with space groups $C2/c$ and $C2$ and similar motif (chains of $MO_6$ octahedra) are known for the family.

Crystal structure characterization carried out by using X-ray powder diffraction technique show that $GdCr_3(BO_3)_4$ and $TbCr_3(BO_3)_4$ crystals are single phase with $R32$ space group, while samples with La turned out to be biphasic ($R32$ – 40wt.% and $C2/c$ – 60wt.%). Magnetic and heat capacity measurements of the studied samples have been performed by the SQUID magnetometer MPMS-XL7 and the PPMS system, respectively. Magnetic resonance investigations have been carried out by means of a handmade multifrequency spectrometer. Investigations of electrical polarization have been done by means of a homemade setup in pulsed magnetic field.

Above 50 K, susceptibility of the crystals with La and Gd obey the Curie-Weiss law with following parameters: for $LaCr_3(BO_3)_4$ effective magnetic moment $\mu_{eff}$ = 3.86 $\mu_B/Cr^{3+}$ and Curie–Weiss temperature $\theta$ = –23 K, for $GdCr_3(BO_3)_4$ - $\mu_{eff}$ = 10.6 ± 0.1 $\mu_B$ per formula unit and $\theta$ = 7 ± 1 K. Rough evaluation of values of the intrachain ($J/k$ = 4 ± 1 K) and interchain ($J'/k$ = –0.5 ± 0.2 K) Cr-Cr exchange interactions does not allow to find out how that parameters depends on the rare earth ions.

It was found that both structural modifications of lanthanum chromium borate undergo into antiferromagnetically ordered state at 6.5 K and 8.5 K for $C2/c$ and $R32$ phases, respectively. Crystals $GdCr_3(BO_3)_4$ and $TbCr_3(BO_3)_4$ become antiferromagnets at 7.15 K and 8.8 K respectively. Compounds with La and Gd demonstrate the easy-plane magnetic anisotropy in temperature range from 2 K up to $T_N$ but for $TbCr_3(BO_3)_4$ spontaneous spin-reorientation phase transition from the easy-plane (EP) antiferromagnetic state to the easy-axis (EA) one takes place in the vicinity of 5 K. The EA state exists in a limited range of temperatures (below 5 K) and magnetic fields (below 0.5 T for $H||c$).

Metamagnetic phase transition has been detected below $T_N$ for all studied crystals and field of the transition strongly depends on the rare earth ion.

The $H$-$T$ phase diagrams for studied compounds have been constructed.

# Structural studies of $Nd_{0.5}Sr_{0.5}MnO_3$ under variable magnetic field and temperature

*AM Milinda Abeykoon[1]*, Sandun Amarasinghe[2], Trevor A. Tyson[2]**

[1] *NSLS-II, Brookhaven National Laboratory,* [2]*New Jersey Institute of Technology*

*aabeykoon@bnl.gov, tyson@njit.edu

When the crystal structures of materials are closely coupled to their magnetic structures, changes occur in both when they undergo phase transitions due to external stimuli, such as, the temperature and/or the magnetic field. Structural studies on multiple length-scales are necessary to fully understand the mechanism behind these phase transitions. Such studies can lead to the discovery of novel materials with technologically desirable properties. The pair distribution function (PDF) beamline (28-ID-1) of National Synchrotron Light Source-II at the Brookhaven National Laboratory offers an unprecedented combination of techniques and equipment in a single experimental end-station for this purpose. The PDF, Wide angle X-ray Scattering (WAXS), and Small Angle X-ray Scattering (SAXS) under variable magnetic field and/or temperature enable structural studies of the formation of structural/magnetic domains with short coherent length-scales as well as the evolution of long range ordered features extending to several thousands of Angstroms when materials undergo phase transitions. In addition, these methods are currently being utilized at 28-ID-1 to study magnetostriction in complex materials.

$Nd_{0.5}Sr_{0.5}MnO_3$ (NSMO) is a strongly correlated system that has a rich magnetic field-temperature phase diagram in which several structural transitions strongly overlap with magnetic transitions. Phase transitions in this system can be induced by changing temperature at zero field, or by applying an external magnetic field isothermally staying close to the transition temperatures. Evolution of the powder diffraction Bragg peaks of NSMO seems to be independent of the driving stimuli. However, it is not clear if the nature of the local structural response to the magnetic field, vs temperature is the same as the signal is hidden in the diffuse background. We use PDF, WAXS and SAXS techniques to probe the crystal structure on multiple length-scales when NSMO crosses over several phase transitions. We will show the evolution of two structural phases that seems to be strongly linked to the magnetic behavior of the system.

**Acknowledgements**

This research used resources [or insert beamline if applicable] of the National Synchrotron Light Source II, a U.S. Department of Energy (DOE) Office of Science User Facility operated for the DOE Office of Science by Brookhaven National Laboratory under Contract No. DE-SC0012704. This work is supported in part by NSF Grant No. DMR-1809931.

# Complex magnetic orders and the emergent topological Hall effect in the kagome metal $RMn_6Sn_6$

*Yishui Zhou[1]*, Changjiang Yi[2], Claudia Felser[2], Sabreen Hammouda[1], Fabio Orlandi[3], Dmitry Khalyavin[3], Pascal Manuel[3], Thomas Brückel[4], Yixi Su[1]*

[1]*Jülich Centre for Neutron Science JCNS-MLZ, Forschungszentrum Jülich GmbH, Garching*
[2]*Max Planck Institute for Chemical Physics of Solids, Dresden*
[3]*ISIS Neutron & Muon Facility, STFC, Rutherford Appleton Laboratory, Didcot, UK*
[4]*JCNS-2 & PGI-4, Forschungszentrum Jülich GmbH, Jülich*
*yi.zhou@fz-juelich.de

Following the discovery of a quantum-limit magnetic Chern phase in TbMn6Sn6 [1], and the observation of a large topological Hall effect (THE) related to the field-induced magnetic phases in YMn6Sn6 [2], the magnetic topological metal series $RMn_6Sn_6$ (R=Gd-Yb, and Y, Lu, etc.) [3], which possess an ideal kagome lattice of Mn, have emerged as a new platform to explore exotic states and novel functionalities. We have recently carried out the growth of high-quality single crystals of the magnetic kagome metal ErMn6Sn6 via the flux method, and the physical properties characterizations via the magnetic susceptibility, heat capacity, and Hall conductivity measurements. We have also undertaken comprehensive neutron diffraction experiments on both single-crystal and powder samples at the WISH diffractometer at ISIS. A number of distinct magnetic ordered phases, including the spiral and k = 0 magnetic orders, have been identified in cooling to low temperatures. Furthermore, we have also observed a range of complex field-induced magnetic phases, including the multi-k non-coplanar magnetic orders, via field-dependent single-crystal neutron diffraction at WISH. We have found that these complex field-induced magnetic phases are directly associated with our observed THE over a wide phase space of field and temperature in this compound. Our study has clearly hinted at a fascinating interplay between topologically non-trivial electronic band structures, magnetism and electronic correlations in $ErMn_6Sn_6$.

## References

[1] J. X. Yin, et al., Nature **583**, 533 (2020).
[2] N. J. Ghimire, et al., Sci. Adv. **6**, eabe2680 (2020).
[3] W. Ma, et al., Phys. Rev. Lett. **126**, 246602 (2021).

# Pseudogap and excess conductivity of $YBa_2Cu_3O_{7-\delta}$ single crystals in the course of long-term aging

*L. V. Bludova[1], A. L. Solovjov[1,2], R. V. Vovk[2]*

[1]*B. Verkin Institute for Low Temperatures Physics and Engineering of the National Academy of Sciences of Ukraine, Kharkiv 61103, Ukraine*

[2]*The Faculty of Physics, V. N. Karazin Kharkiv National University, Kharkiv 61022, Ukraine*

*omelchenko@ilt.kharkov.ua

The discovery of high-temperature superconductors (HTSCs) is undoubtedly one of the landmark events in modern solid state physics. However, despite the efforts of numerous scientific groups and an extraordinary number of publications on HTSCs, the mechanism of superconducting (SC) pairing, which makes it possible to obtain real Cooper pairs at $T >> 100$ K [1], is still not clear. It is believed that the answer to this and other questions concerning HTSC can be obtained by studying such an unusual phenomenon as the pseudogap (PG), which opens in underdoped cuprates at $T^* >> T_c$ [1, 2].

The temperature dependences of both fluctuation conductivity (FLC) $\sigma'(T)$ and pseudogap (PG) $\Delta^*(T)$ derived from measurements of resistivity $\rho(T)$ of an optimally doped (OD) $YBa_2Cu_3O_{7-\delta}$ single crystal subjected to a long-term storage have been studied. The as-grown sample S1 exibits characteristics typical of OD $YBa_2Cu_3O_{7-\delta}$ single crystals containing twins and twin boundaries (TBs). Analysis of both FLC and PG showed an unexpected improvement in all characteristics of the sample after 6 years of storage (sample S2), indicating that the effect of TBs is somehow limited. After 17 years of storage, all characteristics of the sample changed dramatically, which indicates a strong influence of internal defects formed during the aging process. For the first time, the temperature dependences of both FLC and PG were obtained after 17 years of storage. In contrast to these results, the values of $\sigma'(T)$ and $\Delta^*(T)$ obtained after 17 years of storage (sample S3) changed dramatically. Resistivity $\rho(300K)$, $\rho(100K)$ and their linear slope $a = d\rho/dT$ increased by more than 3, 1.6 and 4 times, respectively. The resistive transition of S3 became very wide ($\Delta T_c \approx 7$ K) and pointed out the appearance of a second low-temperature phase with $T_c(\rho=0) \approx 84$K. Thus, the total $\Delta T_c \approx 16$ K. This form of $\rho(T)$ differs markedly from the "classical" behavior of $\rho(T)$ with a similar slope and $\rho(300K)$, which is obtained by reducing the charge carrier density $n_f$ in YBCO with a decrease in the oxygen doping level [2]. Correspondingly, the shape of both σ'(T) and Δ*(T) has also changed greatly. It was found that the fluctuation contribution of 2D-MT is almost completely suppressed. In addition, S3 exhibits the largest scaling factor $C_{3D} = 2.1$ and an unexpectedly increased distance between conducting $CuO_2$ planes $d_{01} = 6.7$ Å, which is about 1.6 times greater than that estimated for YBCO. All this data indicate the presence of a large number of defects leading to significant structural distortions in the crystal. This conclusion is confirmed by the results of the analysis of the pseudogap. A rather peculiar $\Delta^*(T)$ was found in this case, which does not find a direct analogue in our data bank $\Delta^*(T)$.

**References**:

[1] A. L. Solovjov, L. V. Omelchenko, V. B. Stepanov, R. V. Vovk, H.-U. Habermeier, H. Lochmajer, P. Przysłupski, and K. Rogacki, "Specific temperature dependence of pseudogap in $YBa_2Cu_3O_{7-\delta}$ nanolayers," Phys. Rev. B 94, 224505 (2016).

[2] R.V. Vovk and A.L. Solovjov, Electric transport and the pseudogap in the 1-2-3 HTSC system, under all-around compression (Review Article). Low. Temp. Phys. 44, 81-113 (2018).

# Size induced changes on the thermal and magnetic properties of Ferromagnetic $YbNi_2$

*D. P.Rojas[1]*, J. I. Espeso[2], L. Fernández Barquín[2]*

[1] Dpto Estructuras y Física. ETSAM. Universidad Politecnica de Madrid , *Avenida Juan Herrera 4. 28040, Madrid, Spain*

[2] *Dpto CITIMAC.Facultad de Ciencias.Universidad de Cantabria, Avenida Los Castros s/n. 39005, Santander, Spain*

*d.rojas@upm.es

The study of $YbNi_2$ nanoparticles, with sizes ranging from 10 nm to 18 m, obtained by high-energy ball milling revealed interesting properties related with a coexistence of Ferromagnetism and Spin Glass behaviour [1]. In the present work, the influence of the size reduction on the magnetic and thermal properties of the Heavy-Fermion ferromagnetic $YbNi_2$ alloy is further explored. The specific heat ($c_p$) on nanosized samples show striking differences respect to that of the bulk alloy. For temperatures above 150 K, an excess contribution respect to the bulk sample is observed, whereas for intermediate temperatures, between 20 K and 150 K, the opposite situation is found. This behaviour can be explained by negligible Crystalline Electric Field effects in the nanometric samples and, also, because of the softening of the phonon modes at the surface of the nanoparticles [1,2]. Thus, within this framework, it seems reasonable to analyse the $c_p$ data, for temperatures between 20 K and 300 K, only using the electronic and phonon terms. In addition, a simple model that considers a modification in the phonon spectra of surface atoms respect to the core of the nanoparticles (two different values of the Debye temperature) can be also applied [2]. As result, the Sommerfeld coefficient is found to decrease from 73 $mJ/molK^2$ (18 nm nanoparticles) to 37 $mJ/molK^2$ (10 nm nanoparticles). On the other hand, the obtained Debye temperature for the core is around 350 K, but the ones obtained for the surface atoms range from 114 K (18 nm) to 142 K (10 nm).

At low temperatures, below 5 K, the $c_p/T$ curves exhibit marked upturns following a -lnT dependence, characteristic of a Non-Fermi liquid behaviour in the presence of disorder and within a ferromagnetic behaviour [3]. This last feature is consistent with the DC magnetic susceptibility results, which show divergencies of the $M/H \sim T^{-1+\lambda}$ kind, with $\lambda = 0.8$, a value near to that reported for bulk ferromagnetic systems [3].

The results are explained by the influence of disorder on the alloys enhanced by the milling process in the ensemble of nanoparticles.

## Acknowledgements

This work has been supported by the Spanish MAT2017-83631-C3-R Project.

## References

[1] D.P. Rojas *et.al*, *J. Magn. Magn. Mater*. 475 (2019) 264
[2] D. P. Rojas *et.al*, *Nanotechnology* **21** (2010) 445702.
[3] M. Nicklas *et.al*, *Phys. Rev. Letters* 82 (1999) 4268.

# Superconducting State Properties of Intercalated $Li_x(C_2H_8N_2)(Fe_ySe_zS_{1-z})$ Systems

*Artem Lynnyk[1]*, Anna Krzton-Maziopa[2], Edyta Pesko[2], and Roman Puzniak[1]*

[1] *Institute of Physics, Polish Academy of Sciences, Aleja Lotników 32/46, PL-02-668 Warsaw, Poland*

[2] *Faculty of Chemistry, Warsaw University of Technology, Noakowskiego 3, PL-00-664 Warsaw, Poland*

* artem.lynnyk1@gmail.com

The intercalated iron selenides FeSe are capable of exhibiting a transition to the superconducting state over a wide range of temperatures. The character of the transition and the parameters of the resulting superconducting state are strongly dependent on the type of intercalant, intercalation method, and stoichiometry of the obtained compounds. The unique combination of a high content of magnetic inhomogeneities and the presence of a superconducting phase has led to the need for determining their respective thermodynamic parameters. This study presents the results of SQUID magnetometry investigations of the superconducting properties of intercalated $Li_x(C_2H_8N_2)(Fe_ySe_zS_{1-z})$ chalcogenides. The investigated materials were synthesized by means of a solvothermal method. Both dc and ac susceptibility measurement techniques were utilized to define the superconducting state parameters, including the upper critical field $H_{c2}$, lower critical field $H_{c1}$, and related coherence length $\xi$ and field penetration depth $\lambda$. The general nature of the obtained phase diagrams indicates a consistent tendency for the magnetic phases to obscure the response of the superconducting state, since all of the probed materials exhibited the developed magnetization hysteresis over the investigated range of temperatures.

# Spin Pumping in Non-Centrosymmetric-Superconductor/Ferromagnet Trilayers

*Antoni García-Santiago[2,3,*], Carla Cirillo[1], Marc Rovirola[2,3], Carla González[2,3], Blai Casals[2,3], Joan Manel Hernàndez[2,3], Ferran Macià[2,3], and Carmine Attanasio[4]*

[1] *CNR-SPIN, c/o Università degli Studi di Salerno, I-84084 Fisciano (Sa), Italy*
[2] *Dept. Física de la Matèria Condensada, Universitat de Barcelona, 08028 Barcelona, Spain*
[3] *Institut de Nanociència i Nanotecnologia (IN2UB), Universitat de Barcelona, 08028 Barcelona, Spain*
[4] *Dipartimento di Fisica "E.R. Caianiello", Università degli Studi di Salerno, I-84084 Fisciano (Sa), Italy*
*agarciasan@ub.edu

NbRe is a non-centrosymmetric superconductor (NCS) whose superconducting order parameter may consist of a mixture of spin-singlet and spin-triplet components, although this is still a debated issue [1-5]. To shed light on this question, spin pumping across a superconductor/ferromagnet (SC/FM) interface is probed by measuring ferromagnetic resonance (FMR) spectroscopy in NbRe/Co/NbRe trilayers. The microwave transmission coefficient is acquired as a function of dc magnetic field, frequency, and temperature. The linewidth of the FMR spectra ($\mu_0\Delta H$) follows a linear dependence with frequency ($f$) that allows to determine the Gilbert damping parameter ($\alpha$) associated with spin pumping. As temperature decreases and the NbRe layers become superconductor, $\alpha$ remains mostly invariant through the transition, suggesting that spin pumping would still be effective in the superconducting state. Additionally, $\alpha$ is studied as a function of the thickness of the NbRe layers (see the enclosed figure) to determine the values of the spin mixing conductance at the SC/FM interface (18−21 nm$^{-2}$) and the spin diffusion length in the SC layer (7.1−12.5 nm). These results may indicate that spin-polarized supercurrents would occur in the SC layer, suggesting that spin-triplet would be the dominant pairing mechanism in NbRe [6, 7].

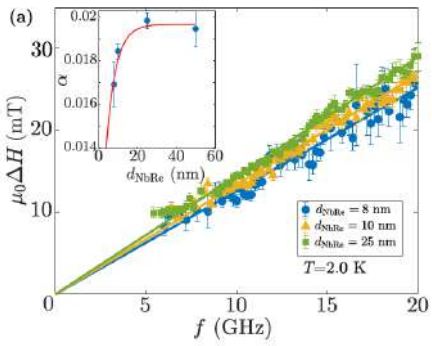



Figure. Frequency dependence of the linewidth of the FMR peaks measured at 2.0 K for different NbRe/Co/NbRe trilayers. The inset shows the NbRe-thickness dependence of the Gilbert damping parameter extracted from the linear fits shown in the main panel.

**References**

[1] C. Cirillo *et al.*, *Phys. Rev. B* **91**, 134508 (2015); *Phys. Rev. B* **94**, 104512 (2016).
[2] T. Shang *et al.*, *Phys. Rev. Lett.* **121**, 257002 (2018).
[3] K.-R. Jeon *et al.*, *Nat. Mater.* **17**, 499 (2018).
[4] S. Sundar *et al.*, *Supercond. Sci. Technol.* **32**, 1 (2019).
[5] C. Cirillo *et al.*, *Thin Solid Films* **758**, 139450 (2022).
[6] A. B. Vorontsov, I. Vekhter, and M. Eschrig, *Phys. Rev. Lett.* **101**, 127003 (2008).
[7] C.-K. Lu and S. Yip, *Phys. Rev. B* **82**, 104501 (2010).

# Spin pumping in LSMO/YBCO heterostructures

*H. Hassan[1], S. J. Carreira[1]*, J. Santamaría[2], M. Anane[1], J. E. Villegas[1]*

[1] *Unité Mixte de Physique, CNRS, Thales, Université Paris-Saclay, 91767, Palaiseau, France*

[2] *GFMC, Dept. Fisica de Materiales, Facultad de Fisica, Universidad Complutense, 28040 Madrid, Spain*

*hadi.hassan@cnrs-thales.fr

Spin-pumping across d-wave superconductors such as $YBa_2Cu_3O_{7-d}$ (YBCO) in high quality ferromagnet/superconductor interfaces provides a new playground for the study of spin polarized supercurrents. Here we use wideband ferromagnetic resonance to study spin-pumping in bilayers that combine the half metallic ferromagnet $La0.7Sr_{0.3}MnO_3$ (LSMO) and YBCO. These bilayers were epitaxially grown on $NdGaO_3$ (NGO) substrates with two different crystallographic orientations $(110)_o$ and $(100)_o$ (Fig.1a-b).

We evaluated the spin conductance at the LSMO/YBCO interface by analyzing the magnetization dynamics in LSMO. We found that the Gilbert damping shows an upturn followed by a drop as the heterostructures are cooled across the normal-superconducting transition when the ab-plane of the YBCO is parallel to the interface (grown on NGO $(110)_o$) (Fig.1c), and a drop followed by an upturn when the ab-plane is tilted 45° with respect to the interface (grown on NGO $(100)_o$) (Fig.1d). The latter case is reminiscent to the phenomena we recently reported for d-wave superconductors, where the opening of the superconducting gap reduces the spin injection efficiency and leads to a drop in the damping, followed by an upturn due to spin resolved Andreev bound states [1]. However, the upturn observed in the damping for samples grown on NGO $(110)_o$ reflects an increment in the spin injection efficiency which could be ascribed to long range spin pumping mediated by spin triplets. This possibility is supported by the generation of spin triplets in c-axis oriented LSMO/YBCO interfaces [2]. These findings put in evidence the anisotropic character of the superconducting gap in YBCO and the potential of this interfaces in the field of superconducting spintronics.

## Acknowledgements

Work supported by French ANR grant “SUPERFAST”, COST action “SUPERQUMAP” and EU H2020 Marie Curie Actions Nº 890964 “SUPERMAGNONICS”.

## References

[1] S. J. Carreira, D. Sanchez-Manzano, M.-W. Yoo, K. Seurre, V. Rouco, A. Sander, J. Santamaría, A. Anane, and J. E. Villegas, *Phys. Rev. B* **104** (2021) 144428.

[2] D. Sanchez-Manzano, S. Mesoraca, F.A. Cuellar *et al.*, *Nat. Mater.* **21** (2022) 188–194.

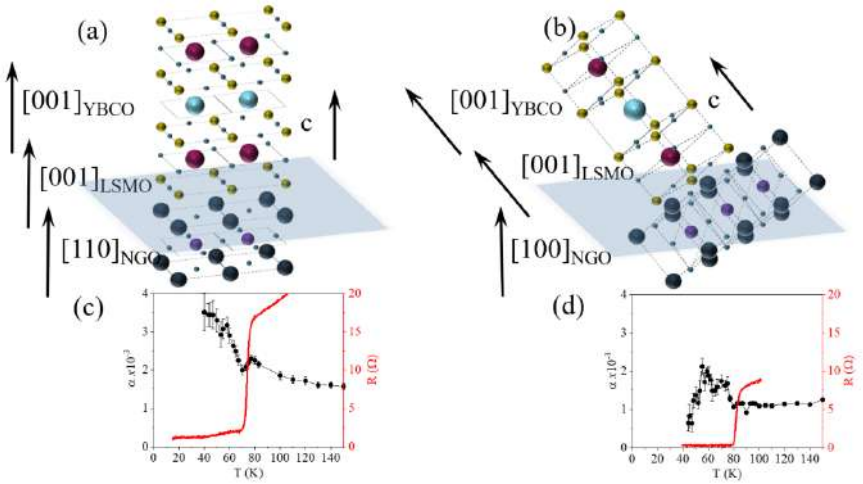



Figure: Scheme of the crystalline structure of the LSMO/YBCO interface for (a) NGO (110) and (b) NGO (100). (c-d) Damping (left axis) and resistance (right axis) vs temperature for (c) YBCO/LSMO//NGO(110) and (b) YBCO/LSMO//NGO(100).

# Effect of the crystal structure on the superconducting properties of a Co monolayer on Ru(0001)

*P.M. Buhl[1,2], L. Mougel[3], Q. Li[3], A. Müller[3], H.H. Yang[3], M. Verstraete[2], P. Simon[4], W. Wulfhekel[3] and B. Dupé*[2,4]*

[1] *Institute of Physics, Johannes Gutenberg University Mainz, 55099 Mainz, Germany*
[2] *Nanomat/Qmat/CESAM, Université de Liège, 4000 Sart-Tilman, Belgium*
[3] *Physikalisches Institut, Karlsruhe Institute of Technology, 76131 Karlsruhe, Germany*
[4] *Laboratoire de Physique des Solides, UMR 8502, CNRS, Université Paris-Sud, Université Paris-Saclay, 91405 Orsay, France*
[5] *Fonds de la Recherche Scientifique (FNRS-FRS), 1000 Bruxelles, Belgium*
*bertrand.dupe@uliege.be

Skyrmions are non-collinear, localized magnetic states. Their chiral properties do not only result in their stabilization but also create an emergent magnetic field that exhibits a non-zero Chern number. When this emergent field is coupled to a s-type superconductor, Majorana states may emerge in the superconductor [1,2]. In that case, the presence of Majorana states in combination with the high skyrmion mobility may allow quantum computing operations.

Here, we use density functional theory (DFT) calculations to explore the interplay between a magnetic Co monolayer on a superconducting substrate Ru substrate where isolated skyrmions were observed under magnetic field [3]. We project the DFT band structure on Wannier functions that we use to parametrize a tight-binding Hamiltonian. Then, the superconducting properties are obtained by solving the Bogoliubov-De Gennes equations. Using this methodology, we explore the influence of the magnetic and cristal properties of the Co monolayer on the proximity effect with a bulk Ru(0001) superconductor substrate [4].

**Acknowledgements**

W. Wulfhekel acknowledges funding by the Deutsche Forschungsgemeinschaft (DFG) under Grant Nos. WU 349/15-1 and WU 349/16-1. H. S. Yang acknowledges funding by the Alexander-von-Humboldt Foundation. B. Dupé and P. Buhl acknowledge funding by the DFG under Grant No. DU 1489/3-1.

**References**

[1] G. Yang, P. Stano, J. Klinovaja, and D. Loss *Phys. Rev. B* **93**, 224505 (2016).
[2] M. Garnier, A. Mesaros, and P. Simon *Communications Physics* **2**, 126 (2019).
[3] M. Hervé, B. Dupé, R. Lopes, M. Böttcher, M. D. Martins, T. Balashov, L. Gerhard, J. Sinova, and W. Wulfhekel *Nat. Commun.* **9**, 1015 (2018).
[4] L. Mougel, P. M. Buhl, Q. Li, A. Müller, H.-H. Yang, M. J. Verstraete, P. Simon, B. Dupé, and W. Wulfhekel *Appl. Phys. Lett.* **121**, 231605 (2022).

# Strain Dependent Electroresistance Loops in $NdNiO_3$-$BaTiO_3$ Ferroelectric Tunnel Junctions

*Isabel Barbero[1]*, Víctor Rouco[1], Carlos León[1], Lucas Pérez [1, 2], Javier Tornos[1], Jacobo Santamaría[1], Miguel Romera[1]*

[1] *Dept. Física de Materiales, Fac. CC Físicas, Universidad Complutense de Madrid,28040 Madrid, Spain*

[2] *Instituto Madrileño de Estudios Avanzados – IMDEA Nanociencia, Campus Cantoblanco, 28049 Madrid, Spain*

*maibarbe@ucm.es

Oxide-based ferroelectric tunnel junctions (FTJs) are promising devices for neuromorphic computing since they exhibit both ferroelectric [1] and ferroionic memristive behavior [2,3]. Understanding the influence and timescale of the different parameters governing transport is crucial for designing neuromorphic response [4].

In the present work, we have fabricated $NdNiO_3$-$BaTiO_3$-Au micrometric-sized FTJs on two different substrates with different degrees of mismatch strain: $SrTiO_3$ (001) (tensile strain) and $LaAlO_3$ (001) (compressive strain). Both $NdNiO_3$-$BaTiO_3$ based FTJs show very large tunnel electroresistance (TER) in the range $10^4$-$10^5$ % at low temperatures and $10^3$ % close to room temperature (Figure 1). Changing epitaxial strain yields different temperature dependences of the high and low resistance states and of the switching voltages of the electroresistance loops, indicating that transport mechanisms are differently affected by the ferroelectric groundstate of the barrier and by ferroionic effects induced by electromigration of oxygen atoms. These results constitute an important step towards neuromorphic computing devices based on oxide-based FTJs.

## Acknowledgements

This work was supported by the Spanish MIC, AEI and FEDER under Grant No. PID2020-116181RB-C33 (MCI/AEI/FEDER, UE) and by Comunidad de Madrid (Atracción de Talento) under Grant No. 2018-T1/IND-11935. Isabel Barbero was funded by the Spanish Ministry of Science and Innovation (MCINN) through the FPU Fellowship No. FPU21/05565.

## References

[1] Vicent Garcia and Manuel Bibes. *Nature Communications*, **5**, (2014) 4289.
[2] V. Rouco et al. *APL Materials*, **9**, (2021) 031110.
[3] D. Hernandez-Martin et al. *Physical Review Letters*, **125**, (2020) 266802.
[4] P. Nukala et al. *Science*, **372**, (2021) 6542.

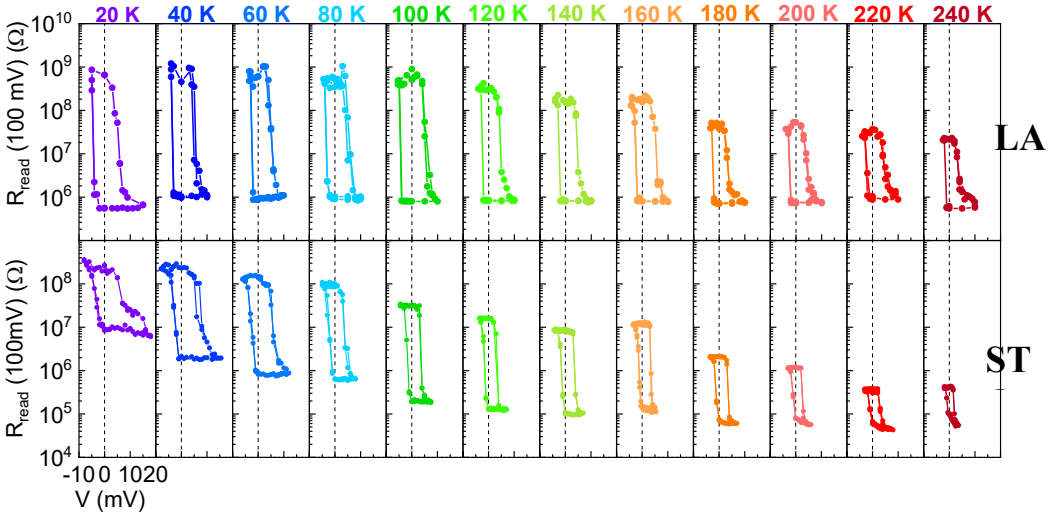



Figure 1: Electroresistance loops of the FTJs grown on $LaAlO_3$ (top) and $SrTiO_3$ (bottom).

# Asymmetric Charge Transport in a Superconducting and Magnetic Heterostructure

*A. Mehrnejat[1†], M. C. Hatnean[2], M. C. Rosamond[3] N. Banerjee[1], G. Balakrishnan[2] and F. K. Dejene[1†]*
*[1] Department of Physics, Loughborough University, Loughborough LE11 3TU, UK.*
*[2] Department of Physics, University of Warwick, Coventry CV4 7AL, UK.*
*[3] School of Electronic and Electrical Engineering, University of Leeds, Leeds, LS2 9JT, UK.*
†Corresponding authors: a.mehrnejat@lboro.ac.uk and f.dejene@lboro.ac.uk

**Abstract:**
Bilayers comprising superconducting and magnetic layers provide a platform for investigating a plethora of physical interactions in such heterostructures. In type II superconductors, the behaviour of the Abrikosov vortices can be manipulated by the magnetic domains of the magnetic layer and their transformations. Here, we explore further a reported flux pinning observed in van der Waals heterostructures of two-dimensional (2D) materials $NbSe_2$ and $CrGeTe_3$ (CGT) as the superconducting and the magnetic layers, respectively[1]. We then report on the observation of the characterised asymmetric charge transport in such devices and how asymmetric magnetic interfaces and the transformation of the magnetic domains of CGT from stripe phase to bubbles[2] can play a role in this observation.

**References:**

[1] D. Nutting and F. Withers, *Phys. C Supercond. its Appl.*, vol. 581, p. 1353803, Feb. 2021.
[2] M. G. Han *et al.*, *Nano Lett.*, vol. 19, no. 11, pp. 7859–7865, Nov. 2019.

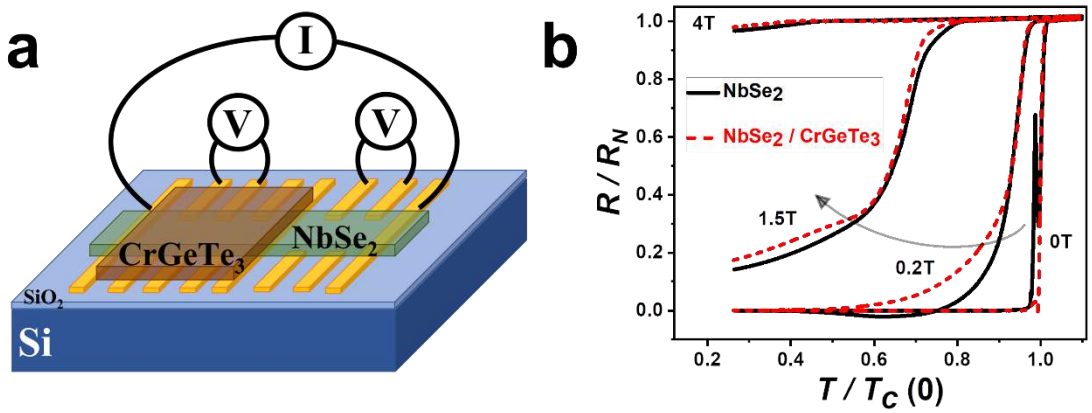



Figure 1: a) An exemplary schematic of the fabricated devices. b) Normalised temperature dependence of the resistance for the $NbSe_2$ and the $NbSe_2$/CGT heterostructure at different applied fields.

# Multipolar super-exchange interactions in f-electrons systems and their impact on the magnetic order and the thermal expansion.

*S. Khmelevskyi[1]* and L. P. Pourovskii[2]*

[1 4] *Research Center for Computational Materials Science and Engineering, Vienna University of Technology, Karlsplatz 13, 1060 Vienna, Austria*

[2] *CPHT, Ecole Polytechnique, CNRS, Université Paris-Saclay, Route de Saclay, 91128 Palaiseau, France*

*sk@cms.tuwien.ac.at

The nature of order in low-temperature phases of some materials is not directly seen by experiment. Such "hidden orders" (HOs) may inspire decades of research to identify the mechanism underlying those exotic states of matter. In insulators, HO phases originate in degenerate many-electron states on localized f - shells that may harbor high-rank multipole moments. We show [1] how the ground-state order and magnetic excitations of a prototypical HO system $NpO_2$, can be fully described by a low-energy Hamiltonian derived by a many-body ab initio force theorem method. A primary non collinear order of time odd rank 5 (triakontadipolar) moments has been predicted. We show also that exotic non-chiral magnetic order in $PrO_2$ is a results of a strong high order multipolar interactions within whole |JM> ground state multiplet. The problem of magnetic order in $PrO_2$ cannot be reduced to the interactions within a ground state Kramer's doublet. Sumarizing, including also our earlier studies of super-exchange interactions also in $UO_2$ [2], we conclude that the higher order multipolar interactions cannot be ignored even in the case of the "normal" collinear magnetic order. We also demonstrate the impact of the multipolar exchange on the magnovolume properties and describe the phenomena of the multipolar exhcnage striction.

## References

[1] L. V. Pourovskii, and S. Khmelevskyi. *Proc Natl Acad Sci USA (PNAS)*, **118** (2021) e2025317118.
[2] L..V. Pourovskii, and S. Khmelevskyi*, Phys. Rev* B **99** (2019), 094439.

# Hyperfine Interactions in Magnetite around Verwey Temperature

*Ondřej KRČMÁŘ[1]*, Tomáš KMJEČ[1], Denisa KUBÁNIOVÁ[1], Helena ŠTĚPÁNKOVÁ[1], Jaroslav KOHOUT[1]*

[1] *Faculty of Mathematics and Physics, Charles University, V Holešovičkách 2, 180 40 Praha 8, Czech Republic*

*ondrej.krcmar@matfyz.cuni.cz

Magnetite, as a typical representative of ferrimagnetic iron oxides, is a material of mixed valence. Its cubic crystal lattice of the Fd-3m space group contains 8 formula units in the elementary cell. The formal valence composition of magnetite can be written as $(Fe^{3+})_A[Fe^{3+}\ Fe^{2+}]_BO_4$, in which A denotes the tetrahedral and B the octahedral sublattices of the spinel structure. An anomaly in the temperature dependence of specific heat [1, 2], a jump in magnetization [2, 3], and a jump of two orders of electrical conductivity magnitude [2, 4, 5] around the temperature $T_V \sim 125$ K was ascribed to the phase transition named Verwey transition. When studying magnetite above $T_V$ using $^{57}$Fe NMR [6], no difference was observed between $Fe^{3+}$ and $Fe^{2+}$ ions in the B-sites of the crystal lattice, which indicates a delocalized character of charge carriers and is in agreement with previous NMR experiments [7] and Mössbauer spectroscopy [8, 9].

We acquired $^{57}$Fe NMR and Mössbauer spectra of a monocrystalline plate of magnetite with a diameter of ~3 mm and thickness of ~40 µm at liquid helium temperature. In the Mössbauer spectroscopy experiments, the sample was cooled at zero external magnetic field and in the field of 6 T. After cooling without the applied magnetic field, the magnetization is perpendicular to the plane of the sample, i.e., parallel to the direction of the γ-rays, which results in the suppression of the intensity of the 2$^{nd}$ and 5$^{th}$ line of the sextets. The NMR spectra provide higher resolution and enable the determination of the hyperfine fields of all inequivalent iron crystallographic sites of magnetite in the Cc structure below the Verwey transition (8 A and 16 B-sites) [10]. These values were used in the analysis of the Mössbauer spectra as fixed parameters, which allowed us to determine the isomer shift and quadrupole shifts of the previously ambiguous three sextets with the lowest hyperfine fields.

## Acknowledgements

This work was supported by Charles University, project GA UK No 410622.

## References

[1] G. S. Parks and K. K. Kelly, The Heat Capacities of Some Metallic Oxides The Journal of Physical Chemistry. 30 (1926), 47

[2] T.Okamura: Sci. Rep. Tohoku Imp. Univ. 21 (1932), 231

[3] P. Weiss, R. Forrer, La saturation absolue des ferromagnétiques et les lois d'approche en fonction du champ et de la température, Ann. Phys. 12 (1929), 279

[4] E. J. W. Verwey, E. Electronic Conduction of Magnetite (Fe3O4) and its Transition Point at Low Temperatures. Nature 144 (1939), 327

[5] E. J. W. Verwey, P. W. Haayman, Electronic conductivity and transition point of magnetite ("Fe3O4"), Physica, Volume 8, Issue 9 (1941), 979

[6] P. Novák, H. Štěpánková, J. Englich, J. Kohout , VAM Brabers, NMR in Magnetite below and around the Verwey Transition, PHYSICAL REVIEW B 61 (2000) 1256-1260

[7] N.M.Kovtun and A.A.Shamyakov: Solid State Communications 13 (1973), 1345

[8] F. van der Woude, G. A. Sawatzky, A. H. Morrish, Relation between the Hyperfine Magnetic Fields and Sublattice Magnetizations in Fe3O4, Phys. Roar. 167 (1968), 533

[9] Mössbauer experiments in magnetite are reviewed in REVanderberghe and E. de Grave: Mössbauer Spectroscopes Applied to Inorganic Chemistry, vol. 3 (1989), 59

[10] P. Novák, H. Štěpánková, J. Englich, J. Kohout , VAMBrabers, Temperature Dependence of NMR in Magnetite, Proceedings of International Conference on Ferrites, Kyoto 2000, 131-134

# Magneto-transport properties and anomalous Hall effect in ferromagnet/nanostructured superconductor hybrid systems

*Gabriel Caballero[1,2], Ana Parente[2], Ignacio Figueruelo[1,2], Yako Irusta[2], Manuel R. Osorio[1], Iciar Arnay[1], Álvaro Muñoz-Noval[1,2], Paolo Perna[1], Elvira M. Gonzalez[1,2], Mariela Menghini[1]*

[1] *IMDEA Nanociencia, C/Faraday 9, Madrid, Spain*

[2]*Facultad Ciencias Físicas, Universidad Complutense de Madrid, Plaza de Ciencias 1, Madrid, Spain*

mariela.menghini@imdea.org

Ferromagnetism and superconductivity are still attracting a lot of attention in the condensed matter physics community not only due to the rich physical phenomena they embrace but also due to their great potential in different novel applications (memories, sensors, quantum devices…). Although they are considered antagonist effects, the interplay between them can lead to a wide variety of exotic phenomena. Some examples are superconducting vortex pinning induced by magnetic nanostructures [1], domain wall superconductivity [2], triplet superconductivity [3] and superconducting vortex – magnetic skyrmions interactions [4,5].

Here, we study magneto-transport properties of a ferromagnetic (FM) multilayer with perpendicular magnetic anisotropy (PMA) in contact with a nanostructured superconductor (SC). The magneto-resistance and Hall resistance of the FM change below the transition temperature of the nanostructured SC even when an insulating layer is placed between both materials. The symmetry of the Hall response indicates that these changes are not due to topological effects but possibly caused by modifications of magnetic structures or proximity effects.

## References

[1] M. Vélez et al., J. Magn. Magn. Mater., **320**, 2547 (2008)

[2] Z. Yang et al., Nature Materials, **3**, 793 (2004)

[3] T. S. Khaire et al., Phys. Rev. Lett., **104**, 137002 (2010)

[4] V. L. Vadimov et al., Appl. Phys. Lett., **113**, 032402 (2018)

[5] J. Baumard et al., Phys. Rev. B, **99**, 014511 (2019)

# Thermoelectric Response of a Quantum Dot coupled to Ferromagnetic Electrode and Unconventional Superconductor Lead

*Vrishali Sonar*[1] *and Piotr Trocha*[1]

[1] *Institute of Spintronics and Quantum Information, Faculty of Physics, Adam Mickiewicz University, Poznań 61-614, Poland*

*vrison@amu.edu.pl

The BCS theory of conventional (s wave) superconductors present the Cooper pair as a net spin zero particle. In turn, unconventional superconductors[1] of triplet type like p wave superconductors form Cooper pair with S=1 spin. These have been theoretically modelled and seen experimentally[2] though origin of interaction is not identified singularly precisely. Here, we investigate spin-dependent thermoelectric response of a quantum dot hybrid structure coupled to triplet superconductor and ferromagnetic lead. We present the local and non-local transport coefficients like electrical and thermal conductances, thermopower and its spin counterparts. These quantities are calculated with the help of non-equilibrium Green's function approach. Moreover, we also investigate nonequilibrium case, in which the system can work as heat engine or refrigerator. To characterize the effectiveness of such device we calculate the efficiency at maximum power or the coefficient of performance (COP) depending on the way the system works. Apart from that, the triplet superconductor might also exhibit the breaking of both time and inversion symmetry, and thus, we also analyse the influence of this phenomena on transport properties of the considered system.

**References**

[1] Manfred Sigrist , "Introduction to Unconventional Superconductivity", AIP Conference Proceedings 789, 165-243 (2005).

[2]Catherine Kallin and John Berlinsky 2016 Rep. Prog. Phys. 79 054502

# SYMPOSIUM 10.
## MAGNETISM OF ALLOYS, INTERMETALLICS, THIN FILMS AND MULTILAYERS.
## S10. INVITED ORAL PRESENTATIONS

# Control of chiral domain walls in [Pt/CoFeB/Ir]$_2$ using $BaTiO_3$ domains

*K.N. Alshammari[1,2], R.G. Hunt[1], M. Ali[1], P.S. Keatley[3] and T.A. Moore[1]**

[1] *School of Physics and Astronomy, University of Leeds, Leeds LS2 9JT, U.K.*

[2] *Physics Department, College of Science, Jouf University, P.O. Box 2014, Sakaka 42421, Saudi Arabia*

[3] *Department of Physics and Astronomy, University of Exeter, Exeter EX4 4QL, U.K.*

*T.A.Moore@leeds.ac.uk

Magnetic multilayers with perpendicular magnetic anisotropy (PMA) and an interfacial Dzyaloshinskii-Moriya interaction (iDMI) are actively researched, and the chiral spin textures within them are not only of fundamental interest but are also considered as potential information carriers in devices [1]. To operate any of these devices in an energy-efficient way, it is increasingly recognised that alternatives to magnetic field or electric current must be used, such as an electric field to modify the exchange coupling or magnetic anisotropy. Strain applied via a voltage-controlled piezoelectric, for example, modifies the magnetic anisotropy and has the advantage of allowing reversible control of magnetic properties of a PMA multilayer [2].

Here we deposit Pt(2.3nm)/$Co_{68}Fe_{22}B_{10}$(0.7nm)/Ir(0.5nm) multilayers with PMA and iDMI by sputtering onto $BaTiO_3$(001) (BTO) single crystal substrates. By direct imaging of the demagnetised multilayer at room temperature in a wide-field Kerr microscope, we find that the strain pattern from the ferroelectric BTO domains is imprinted onto the magnetic domains. In contrast to the maze domain pattern in an identical multilayer deposited on $Si/SiO_2$ (Fig 1(a)), the domains in the [Pt/CoFeB/Ir]$_2$ on BTO are stripes aligned with the polarisation in the BTO domains (Fig 1(b)). To demonstrate that this is due to strain generated by in-plane lattice elongation in the BTO, we simulate the effect of an in-plane uniaxial anisotropy using micromagnetic parameters obtained from experiment. Measuring a negative magnetostriction (-22 ppm), we deduce that the magnetic easy axis is orthogonal to the BTO polarisation and that the magnetic moment at the centre of the chiral Néel walls is aligned with the easy axis, orienting the stripe domains parallel to the BTO polarisation.

When the temperature of the BTO is varied in the range 13-320 K, it undergoes phase transitions from rhombohedral (R) to orthorhombic (O) to tetragonal (T) structure, with abrupt increases in the lattice parameter at the transition temperatures ~180 K and ~280 K. We find that the stripe domain period increases abruptly at the transition temperatures too (Fig. 1(c)). This amounts to strain control of the domain period, and thus the domain wall energy. By independently measuring the saturation magnetization and PMA as a function of temperature, we find that both the PMA and iDMI undergo strain-driven changes at the R-O transition.

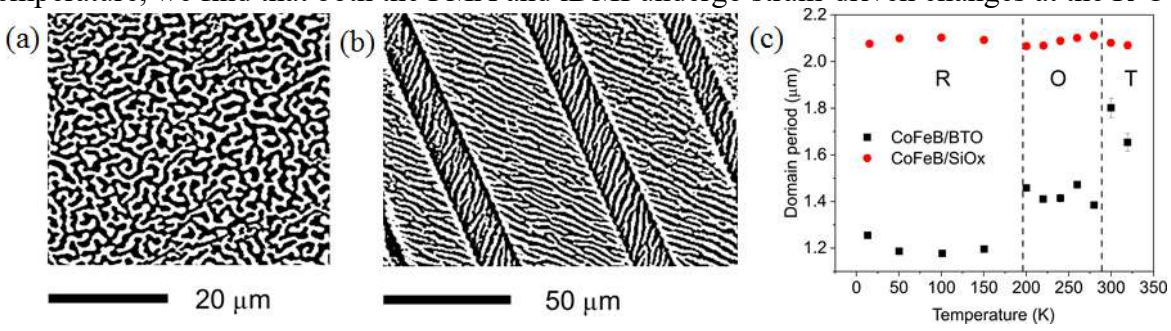



Figure 1: Room temperature Kerr micrograph of demagnetised domains in a [Pt(2.3nm)/CoFeB(0.7nm)/Ir(0.5nm)]$_2$ multilayer deposited on (a) $Si/SiO_2$ and (b) $BaTiO_3$(001). (c) Magnetic domain period vs. temperature for the sample in (b); R,O,T correspond to the rhombohedral, orthorhombic and tetragonal structural phases of the $BaTiO_3$ substrate.

**Acknowledgements**

K.N.A is supported by Jouf University. R.G.H. acknowledges the support of a studentship from EPSRC (UK). Variable-temperature, wide-field Kerr microscopy was performed at the Exeter Time-Resolved Magnetism Facility (EPSRC Grant Reference EP/R008809/1 and EP/V054112/1).

**References**

[1] C.H. Marrows and K. Zeissler, *Appl. Phys. Lett.* 119, 250502 (2021).

[2] P.M. Shepley, A.W. Rushforth, M. Wang, G. Burnell and T.A. Moore, *Sci. Rep.* 5, 7921 (2015).

# Microstructure as a Key to Control the Properties of Magnetic Shape Memory Films

*F. Casoli[1]*, M. Takhsha Ghahfarokhi[1], F. Albertini[1], J. A. Arregi[2], V. Uhlíř[2], O. Gutfleisch[3], P. Tiberto[4]*

[1] *IMEM – CNR, Parco Area delle Scienze 37/A, Parma, Italy*

[2] *CEITEC BUT, Brno University of Technology, Purkyňova 123, Brno, Czech Republic*

[3] *Functional Materials, TU-Darmstadt, Alarich-Weiss-Str. 16, Germany*

[4] *Advanced Materials Metrology and Life Sciences (INRiM), Strada delle Cacce 91, Torino, Italy*

*francesca.casoli@imem.cnr.it

Magnetic shape memory compounds are among the most promising classes of materials for multiple-stimuli actuation and energy harvesting, thanks to the multifunctionality arising from their distinctive magnetostructural transformation. $Ni_2MnGa$ is a model system within this class of compounds; it shows a martensitic phase transformation from a cubic phase (austenite) to a lower symmetry phase (martensite) by decreasing temperature. We grow Ni-Mn-Ga films with thickness up to 400 nm by sputtering on MgO(100) or Cr/MgO(100). The $L2_1$ austenitic phase grows epitaxial at high temperature and transforms to the 7M monoclinic martensitic phase when samples are cooled down to room temperature. The martensitic phase shows a complex twin microstructure, with characteristics that we can engineer by growth parameters, external stimuli (temperature, magnetic field, stress) and patterning on the micron scale [1 and references therein]. Thanks to electron and scanning probe microscopies and to magnetic characterization, we have deepened the correlation between microstructure and magnetic/magnetothermal properties, also imaging the films while varying temperature and applying a magnetic field.

Each twin microstructure imposes a specific geometrical arrangement of the easy-magnetization axes, giving rise to distinctive magnetization patterns and magnetization processes. Consequently, Ni-Mn-Ga films display a unique versatility of the magnetic properties, which can be modified by choosing a specific twin microstructure (X- or Y-type), combining the two microstructures, or modifying their spatial arrangement on the scale of tens of microns. The support of a micromagnetic model, built on the film microstructure and experimental characteristics, has been essential for modifying the magnetic properties by microstructure engineering [2]. Each twin microstructure also shows characteristic magnetothermal properties, i.e., thermal hysteresis and sharpness, which can be improved by choosing a specific microstructure. Finally, we have investigated the magnetothermal properties in Ni-Mn-Ga micro- and nano-structures, and the local impact of a variable force applied by nanoindentation, which increases the transformation temperature up to 5 K [3].

## Acknowledgements

FC and FA acknowledge financial support from PNRR MUR project ECS_00000033_ECOSISTER.

## References

[1] M. Takhsha Ghahfarokhi, J.A. Arregi, et al., *Appl. Mater. Today* **23** (2021) 101058.
[2] F. Casoli, G. Varvaro, et al., *J. Phys. Mater.* **3** (2020) 045003.
[3] M. Takhsha Ghahfarokhi, F. Casoli, et al., *Acta Mater.* **245** (2023) 118603.

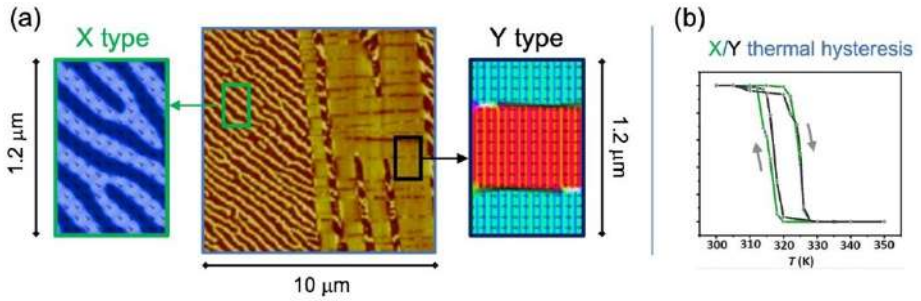



Figure: a) MFM image of a Ni-Mn-Ga film and corresponding micromagnetic configurations of X/Y microstructures; b) thermal hysteresis of X/Y microstructures across the martensitic transformation

# Growth, optimization, and electrical manipulation of rare-earth iron garnets for efficient spintronics

*Can Onur Avci*

*Institut de Ciència de Materials de Barcelona (ICMAB-CSIC), Campus de la UAB, 08193 Bellaterra, Spain*

cavci@icmab.es

Spintronics, the concept of harnessing electron spin as an active variable in electronic circuits, has evolved into a broad and interdisciplinary research field at the intersection of physics, materials science, and nanotechnology. Our improved understanding of transport phenomena and magnetic interactions in solid-state, along with the discoveries of new materials and experimental techniques, enabled rapid progress. Current-induced control of magnetization lies at the heart of the research efforts as it will ultimately lead to efficient nonvolatile solid-state memory, logic, and signal transmission devices. These devices can boost the capabilities of the contemporary CMOS technologies and potentially offer beyond-CMOS concepts leading to paradigm shifts in the microelectronic industry [1].

In the first part of the talk, we will discuss current-induced magnetic manipulation and detection experiments in insulating magnetic garnet systems mainly enabled by spin-orbit torques, spin Hall magnetoresistance, and other emerging spin transport phenomena. We will first present the fundamentals and symmetry of spin-orbit torques [2] and how they are accurately characterized magnetic insulator/platinum bilayers [3] and used to switch magnetization with high efficiencies [4]. We will then show how interfacial chiral magnetism stabilizes homochiral Neel domain walls in some perpendicularly magnetized iron garnets, which can be propelled as fast as 800 m/s with moderate current densities of the order of $10^8$ A/cm$^2$ [5].

In the second part of the talk, we will present recent advances in the optimization of various rare-earth iron garnets by magnetron sputtering and pulsed laser deposition methods for efficient spintronics applications. We will show how their properties can be conveniently engineered for different experimental purposes [6]. We will conclude by providing some suture directions in spintronics that may benefit from the research progress in perpendicular ferrimagnetic insulators.

**Acknowledgements**

The author acknowledges funding from the European Research Council (ERC) under the European Union's Horizon 2020 research and innovation programme (project MAGNEPIC, grant agreement No. 949052) and from the Spanish Ministry of Science and Innovation through grant reference No. PID2021-125973OA-I00

**References**

[1] Dieny et al., *Nat. Electron.* 3, 446 (2020).
[2] Garello, Avci, et al., *Nat. Nanotech.* **8**, 587 (2013).
[3] Testa et al., arXiv:2302.01141 (2023)
[4] Avci et al., Nat. Mater. 16, 309 (2017).
[5] Avci et al., Nat. Nanotech. 14, 561 (2019).
[6] Damerio & Avci, J. Appl. Phys. 133, 073902 (2023).

# Spin dynamics and spin-glass states in electron-doped $Cr_2Ge_2Te_6$

*S. Khan[1], E. Aw[1], L. Nagle-Cocco[2], M. B. K. Subhan[1], D. Sagkovits[1], A. G. Llorente[3], I. A. Verzhbitskiy[4], C. Howard[1], G. Eda[4] and H. Kurebayashi[1,]**

[1] *University College London, London, WC1H 0AH, UK*
[2] *University of Cambridge, JJ Thomson Ave, Cambridge CB3 0HE, UK*
[3] *Universidad Rey Juan Carlos, 28933 Móstoles, Madrid, Spain*
[4] *National University of Singapore, 21 Lower Kent Ridge Rd, 119077, Singapore*
*h.kurebayashi@ucl.ac.uk

Charge carrier injection into materials are a powerful way to enhance targeted material properties as well as to discover novel, hidden quantum phases. For van der Waals (vdW) materials, it is possible to inject charge carriers into nano-meter thick exfoliated flakes by solid-state and electrolyte gating, and into bulk flakes by chemical doping within vdW gaps, so-called intercalation. This approach is very effective in controlling magnetism, e.g. a dramatic change of Curie temperature of vdW magnet such as $Fe_3GeTe_2$ and $Cr_2Ge_2Te_6$ been observed by electrostatic carrier injection [1, 2] and intercalation [3]. Here, we present our work on spin dynamics characterisation of Na-intercalated, electron-doped $Cr_2Ge_2Te_6$. We have successfully intercalated Na atoms into the vdW gap of pristine $Cr_2Ge_2Te_6$ and observed clear enhancement of its conductivity due to electron doping and an increase of Curie temperature from 65 K (undoped case) to 240 K (doped-system).

We characterised the spin dynamics properties by ferromagnetic resonance techniques. Together with the Curie temperature enhancement as well as the easy-hard axis switch of uniaxial anisotropy previously observed [2, 3], its magnetic relaxation properties is significantly changed due to electron doping, i.e. the Gilbert damping to be in the order of $10^{-4}$ at higher temperatures, which is at least an order of magnitude smaller compared to spin dynamics response in other vdWs magnetic systems [4]. Using ac susceptibility experiments, we also find that the doped $Cr_2Ge_2Te_6$ exhibits a spin-glass like behaviour at low temperatures, where the Mydosh parameter and emipirical scaling down law suggest a cluster spin glass state, and Cole-Cole analysis reveals that the system consist of a distribution of clusters that cannot be explained by a single characteristic relexation time of clusters instead a distribution of relaxation times in the perspective generalised Debye picture.

**Acknowledgements**

We acknowledge supports from the Engineering and Physical Sciences Research Council through project EP/T006749/1 and the Ministry of Education (MOE), Singapore, under AcRF Tier 2 (MOE2017-T2-1-134).

**References**

[1] Y. Deng et al., Nature 563, 94–99 (2018).
[2] I. A. Verzhbitskiy et al., Nat. Electron. 3, 460 (2020).
[3] N. Wang et al., J. Am. Chem. Soc. 141, 17166–17173 (2019).
[4] L. Alahmed, et al. 2D Materials 8.4 (2021): 045030.

# SYMPOSIUM 10.
## MAGNETISM OF ALLOYS, INTERMETALLICS, THIN FILMS AND MULTILAYERS.
## S10. ORAL PRESENTATIONS

# A 2D metalorganic network as a template for magnetic nanoparticulate films with enhanced anisotropy

*L. Hernández López[1,2], I. Piquero-Zulaica[3], D. Serrate[1,2,4], J. Lobo-Checa[1,2,4], F. Bartolomé[1,2]**

[1] *Instituto de Nanociencia y Materiales de Aragón, CSIC-Universidad de Zaragoza, E50009 Zaragoza,Spain*

[2] *Departamento de Física de la Materia Condensada, Universidad de Zaragoza, E50009 Zaragoza, Spain*

[3] *Physics Department E20, Technical University of Munich, 85748 Garching, Germany*

*4 Laboratorio de Microscopías Avanzadas, Universidad de Zaragoza, E50018, Zaragoza, Spain*

*fernando.bartolome@csic.es

Exciting quantum effects emerge whenever atomic clusters undergo an ultimate reduction of their spatial dimensions. Nanostructured magnetic materials characterized by atoms with a lower average coordination number, present enhanced orbital magnetic moment and anisotropy. In Material Science it is always a challenge to discover new processes that can lead to tunable, size selective and self-assembled nanostructures with demon- strated stability up to room temperature. We have achieved this for Fe atoms that form well-defined nanoclusters by reducing their mobility on the Cu(111) substrate mediated by the presence of a metal-organic network [1-3]. When Fe is evaporated onto this molecular array, mostly single atomic height nanodots are formed, instead of the usual triangular bi-layer islands formed on a clean metallic substrate. We find that both, substrate temperature as well as degree of coverage, affect the average nanodot size, demonstrating that a measure of control can be exerted during fabrication. Moreover, X-ray Magnetic Circular Dichroism experiments show that the Fe nanostructures, which are stable at room temperature, present a significantly enhanced orbital moment and magnetic anisotropy compared to the case when the molecular layer is missing. This new approach of using organic templates for the growth of well-defined, metallic nanodots could be extended to other elements with the prospect of exhibiting exciting functionalities.

We have used an extended metal-organic network to nanostructure Fe into nanodots with sizes ranging between 1 and 6 $nm^2$. In this new fabrication method, we find that we can slightly tune the average particle size by simply changing the deposition temperature and the overall metal coverage. Importantly, all cases yield stable structures up to room temperature. We explain this nanoparticle nucleation from the reduced kinetics of Fe atoms upon presence of the organic network, that is significantly reduced when compared to the Fe mobility upon the bare substrate. Correspondingly, we obtain enhanced perpendicular magnetic anisotropy of the nanodots by XMCD without apparent interparticle interactions due to proximity effects. Particularly, while $\mu_S$ is practically insensitive to the particle size, $\mu_L$, $H_A$, and $H_C$ reveal clear surface effects in the form of enhanced values for the smaller particles due to their higher number of average broken bonds per Fe atom. The here presented new growth methodology that uses extended molecular organic networks as base for Fe deposition, might be key for obtaining unprece- dented low-dimensional functionalities in cost-effective ways.

## Acknowledgements

We acknowledge the ESRF for synchrotron beamtime at ID32. We also gratefully acknowledge financial support from PID2019-107338RB-C64, PID2020-115159GB-I00, MCIN/AEI/10.13039/501100011033, by ERDF A way of making Europe and European Union NextGenerationEU/PRTR. Also financing from the European Regional Development Fund (ERDF) under the program Interreg V-A España-Francia-Andorra (Contract No. EFA 194/16 TNSI) is acknowledged. We also thank the Aragonese Project RASMIA E12-23R and NANOMIDAS E13-23R, co-funded by Fondo Social Europeo.

## References

[1] G. Pawin, K. L. Wong, D. Kim, D. Sun, L. Bartels, S. Hong, T. S. Rahman, R. Carp, and M. Marsella, *Angewandte Chemie - International Edition* 47 (2008) 8442

[2] J. Zhang, A. Shchyrba, S. Nowakowska, E. Meyer, T. A. Jung, and M. Muntwiler, *Chemical Communications* 50 (2014) 12289

[3] L. Hernández-López, I. Piquero-Zulaica, C. A. Downing, M. Piantek, J. Fujii, D. Serrate, J. Enrique Ortega, F. Bartolomé, J. Lobo-Checa, *Nanoscale* (2021) 13 5216-5223.

# Perpendicular spin-valve systems on flexible substrates: Direct deposition vs transfer-and-bonding approaches

*Mariam Hassan[1,2]*, Sara Laureti[1], Christian Rinaldi[3], Federico Fagiani[3], Gianni Barucca[4], Annamaria Gerardino[5], Nataliia Schmidt[2], Mario Fix[2], Manfred Albrecht[2], Gaspare Varvaro[1]*

[1]*Institute of Structure of Matter, National Research Council, nM2-Lab, 00015 Monterotondo Scalo, Italy*
[2]*Institute of Physics, University of Augsburg, 86159 Augsburg, Germany*
[3]*Department of Physics, Politecnico di Milano, c/o via G. Colombo 81, 20133 Milano, Italy*
[4]*Department SIMAU, University Politecnica delle Marche, 60131 Ancona, Italy & UdR INSTM Ancona Italy*
[5]*Institute of Photonics and Nanotechnologies, National Research Council, 00156 Roma, Italy*
*mariam.hassan@uni-a.de

Flexible spintronics is an emerging field of research that has received great attention over the past few years for fundamental studies and applications in many technological fields including automotive, wearable electronics, soft robotics, and bio-integrated electronics. The ability to bend a device or to adjust its shape, along with the lower weight and cost, makes flexible electronics more advantageous than conventional rigid systems in many contexts [1-3]. In this work, different strategies for the fabrication of spintronic devices with perpendicular magnetic anisotropy are compared, i.e., transfer-and-bonding approaches exploiting wet and dry lift-off methods on the one side, and direct deposition on flexible substrates on the other side. To evaluate the potential of the proposed strategies, perpendicular magnetized [Co/Pd]-based giant magneto-resistive spin-valves, consisting of a fully compensated $[Co/Pd]_N/Ru/[Co/Pd]_N$ synthetic antiferromagnet used as reference electrode and a $[Co/Pd]_N$ free layer, were investigated. Such stacks represent a demanding model system, owing to the sharpness and the number of interfaces whose quality strongly affects the overall magnetic and electric performance. Advantages and drawbacks of the different strategies are discussed to provide crucial indications for the development of flexible spintronic devices with complex architectures.

**References**

[1] M. Hassan, S. Laureti, C. Rinaldi, F. Fagiani, S. Varotto, G. Barucca, N.Y. Schmidt, G. Varvaro, M. Albrecht, Nanoscale Adv. **3**, 3076 (2021).

[2] P. Makushko, E.S. Oliveros Mata, G.S. Cañón Bermúdez, M. Hassan, S. Laureti, C. Rinaldi, F. Fagiani, G. Barucca, N. Schmidt, Y. Zabila, T. Kosub, R. Illing, O. Volkov, I. Vladymyrskyi, J. Fassbender, M. Albrecht, G. Varvaro, D. Makarov, Adv. Funct. Mater. **31**, 2101089 (2021).

[3] M. Hassan, S. Laureti, C. Rinaldi, F. Fagiani, G. Barucca, F. Casoli, A. Mezzi, E. Bolli, S. Kaciulis, M. Fix, A. Ullrich, M. Albrecht, G. Varvaro, ACS Appl. Mater. Interfaces, **14**, 51496 (2022).

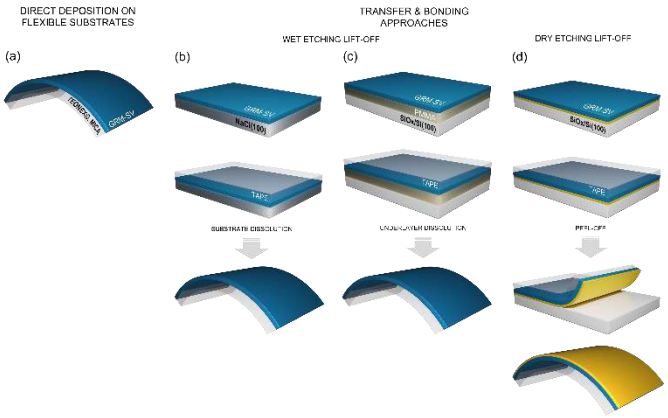



Figure: Schematics of the different strategies explored to obtain PMA flexible spintronic heterostructures. a) Direct deposition on a flexible substrate (i.e., Teonex® tapes, freshly cleaved MICA foils). b, c) Transfer-and-bonding / wet etching lift-off using b) freshly cleaved KBr(100) sheets and c) PMMA underlayers; after depositing the film stack, the whole structure is transferred on a flexible tape glued on top by dissolving KBr and PMMA in water and polar solvents, respectively. d) Transfer-and-bonding / dry etching lift-off using an Au underlayer; after depositing the film stack, the low adhesion between Au and SiOx/Si(100) is exploited to transfer the whole structure on a flexible tape glued on top by mechanical peel-off.

# Growth-induced Changes in the Magnetic Properties of Co/Gd-based Synthetic Ferrimagnets for Magneto-Photonic Integration

*Thomas J. Kools[1]*, Julian Hintermayr[1], Bert Koopmans[1], Reinoud Lavrijsen[1]*

[1] *Eindhoven University of Technology, De Groene Loper 19, Eindhoven, The Netherlands*

*t.j.kools@tue.nl

Recent work has demonstrated Co/Gd (double) bilayers to combine all-optical switching of the magnetization with efficient current-driven manipulation of the magnetic order in one material system [1]. This makes them interesting candidates for bridging the gap between spintronics and photonics [2]. Typically, these synthetic ferrimagnets are fabricated through DC magnetron sputtering. Therefore, for the ultrathin synthetic ferrimagnets with PMA the study of the impact of the sputtering process on basic magnetostatic properties like the net moment and the effective out-of-plane anisotropy is highly relevant.

To this end, we systematically study the simplest Co/Gd-based synthetic ferrimagnet with PMA, the Pt/Co/Gd system [3]. Experimentally, we perform polar MOKE measurements on double wedge samples (Fig. 1) to investigate the transition of the net moment from Co- (red) to Gd- dominated (dark blue), and between non-magnetic, out-of-plane, and in-plane magnetization. For high Gd thickness (~ >3 nm) we would expect that the magnetic properties do not change anymore as the Gd is only magnetized close to the Co interface [3]. Strikingly we observe the opposite, where the magnetic properties keep on changing monotonically up to at least 6 nm of Gd. We hypothesis that sputter-induced intermixing plays a role here. To test this we grow the films with different working gas pressures, changing the kinetics of the sputtered atoms, and indeed a large dependence is observed as shown in Fig 2.

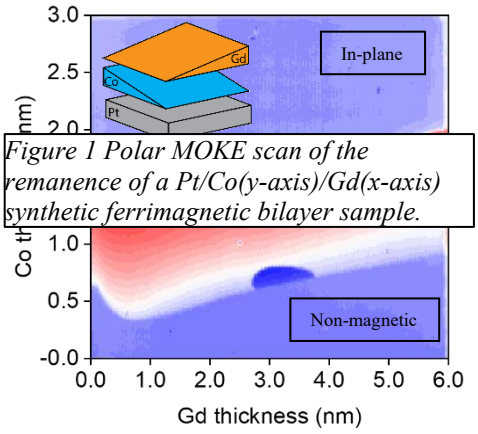



*Figure 1 Polar MOKE scan of the remanence of a Pt/Co(y-axis)/Gd(x-axis) synthetic ferrimagnetic bilayer sample.*

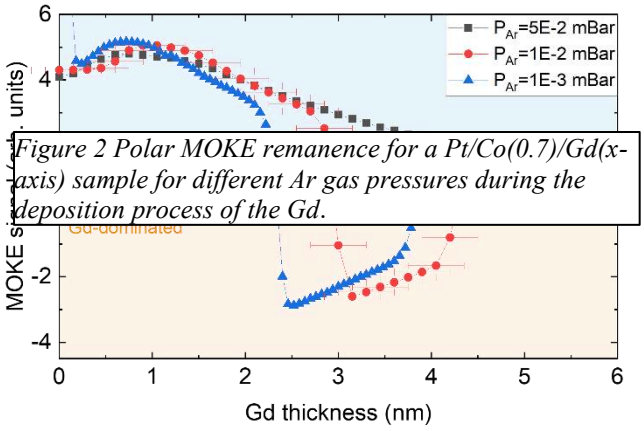



*Figure 2 Polar MOKE remanence for a Pt/Co(0.7)/Gd(x-axis) sample for different Ar gas pressures during the deposition process of the Gd.*

To corroborate this observation we performed numerical simulations of the intermixing process of the Pt/Co/Gd system. We simulate our DC sputtering process in Tridyn [4], and use the extracted atomic profiles in conjunction with a layered Weiss model [5] to calculate the net moment and effective anisotropy. We find that key features in the experimental phase diagram (Fig. 1) can be reproduced, further suggesting that sputter-induced intermixing between Co and Gd in these systems cannot be neglected when describing their magnetic properties.

In conclusion, our work provides highly relevant insight on the impact sputtering has on fundamental magnetic properties of Co/Gd bilayers and paves the way towards a better understanding of synthetic ferrimagnetic systems for spintronics.

## References

[1] Li, P., Kools, T. J., *et al.* (2023), *Adv. Electron. Mater.* 9, 2200613.
[2] Lalieu, M.L.M., *et al.,* (2019). *Nat Commun* 10, 110
[3] Lalieu, M. L. M., *et al.* (2017). Phys. Rev. B, 96, 220411.
[4] Möller, W., & Eckstein, W. (1984). Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 2(1), 814–818.
[5] Beens, M., *et al.,* (2019). *Physical Review B*, *100*(22), 220409.

# Double Exchange Bias and Ultraslow Magnetization Relaxation in TbFe-based Bilayers

*Johannes Seyd*[1]*, *Manfred Albrecht*[1]

[1] *Institute of Physics, University of Augsburg, Universitätsstraße 1, 86159 Augsburg, Germany*

*johannes.seyd@uni-a.de

Exchange bias (EB) is a phenomenon that manifests itself in the horizontal shift of a hysteresis loop. This usually occurs in systems with coupled ferromagnetic-antiferromagnetic or ferromagnetic-ferrimagnetic interfaces. We report on the magnetic reversal characteristics of exchange-biased heterostructures consisting of two ferrimagnetic $Tb_xFe_{100-x}$ layers whose respective magnetizations are Fe-dominated (layer A) and Tb-dominated (layer B) due to their different compositions. Both layers are amorphous and display perpendicular magnetic anisotropy. An exemplary measurement of double exchange bias, revealing giant EB shifts of several tens of kOe, can be seen in Fig. 1a.

Based on previously published results [1], we expanded the range of investigated layer compositions of $Tb_xFe_{100-x}$ bilayers and layer structures. Since not a sharp interface, but a gradual change of composition over a region of ~2.5 nm thickness between the two layers was found in the bilayer structure, we investigated the influence of different additional interlayers on the magnetic reversal behaviour at low temperatures after field cooling (FC).

While investigating these samples, another phenomenon was seen: ultraslow magnetization relaxation. This manifests itself in an overcrossing of the hysteresis branches in the first and third quarter of the M-H-diagram, as shown in Fig. 1b: coming from negative saturation, layer A first switches to an antiparallel alignment in region 1. After the second switching event at point 2, where layer B switches, the two layers are nominally in a parallel alignment. Here, at point 3, a magnetization just below the saturation value is reached, but surprisingly the magnetization subsequently relaxes again to an antiparallel alignment and thus a lower magnetization value. This happens even under increase of the applied magnetic field. The parallel alignment is only gradually regained in region 4, where the field is strong enough to switch layer A again.

Usually, the slowest process in magnetization dynamics is domain wall motion with a timescale between a few ns to hundreds of µs [2]. The process observed here, however, takes in the order of a few tens to hundreds of seconds to complete, as can be seen in Fig. 1c. This ultraslow magnetic relaxation might be attributed to the interlayer region, which is near compensation in the observed temperature range and thus magnetically very hard.

## References:

[1] B. Hebler, P. Reinhardt, G. L. Katona, O. Hellwig, and M. Albrecht, *Phys. Rev. B* **95**, 104410 (2017)

[2] A. Barman and A. Haldar, *Solid State Physics* **65**, pp. 1-108 (2014)

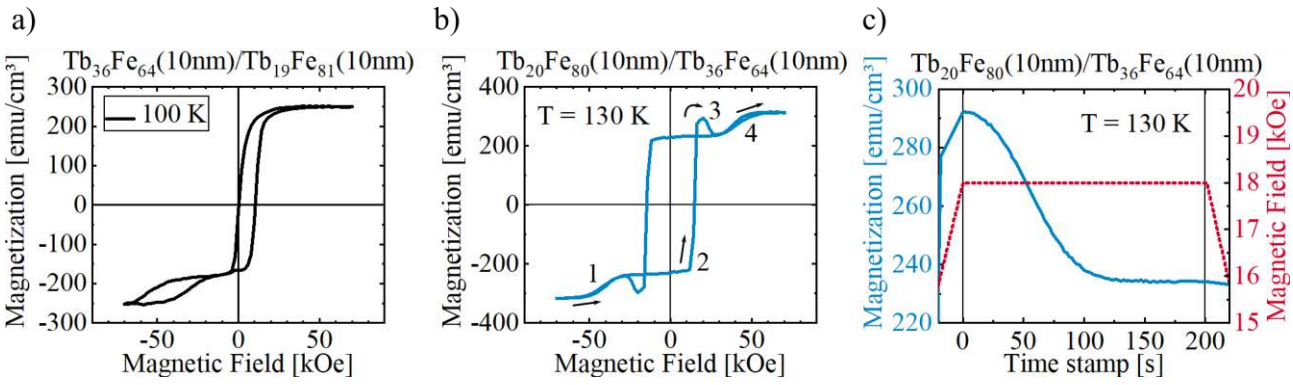



Figure 1: a) Image of the double exchange bias phenomenon in a TbFe-based bilayer at 100 K. b) Demonstration of the overcrossing behaviour in the hysteresis loop. The arrows show the course of the magnetization curve coming from negative saturation. c) Demonstration of ultraslow relaxation at a temperature of 130 K. The hysteresis loop was measured until layer B has completely switched (18 kOe, region 3) and held there for 200 s, while the magnetization slowly decreased without field change, reaching the antiparallel alignment of both layers after approximately 100 s.

# Enhancement of perpendicular magnetic anisotropy of ultrathin Co films by a monolayer Pd capping

Benito Santos Burgos[1,2], Raúl López-Martín[1,2], José A. de Toro[1,2], Chris Binns[1,2], Andreas K. Schmid[3] and Juan de la Figuera[4]

[1] *Instituto Regional de Investigación Científica Aplicada (IRICA), 13005 Ciudad Real, Spain.*
[2] *Dept. Física Aplicada, Universidad de Castilla-La Mancha, 13071 Ciudad Real, Spain.*
[3] *Lawrence Berkeley National Laboratory, Berkeley California 94720, USA.*
[4] *Instituto de Química-Física "Rocasolano", CSIC, Madrid 28006 , Spain.*
*benito.santos@uclm.es

Due to its ability to absorb hydrogen, Pd is one of the most used materials in gas sensor technology. The tensile stress due to the β-phase can crack the Pd crystal. This may be relieved in some cases by growing Pd thin films on appropriate substrates. However, in the limit of very thin films the hydrogen solubility seems to be limited or blocked due the clamping of the Pd in-plane lattice parameter by the substrate. Co/Pd is a very interesting system for its industrial applications, not only in the gas sensor technology but also in the magnetic recording industry. It shows very large perpendicular magnetic ansitropy (PMA) and Kerr rotation. In addition, the magnetization easy axis is controlled by the film thickness. Upon hydrogenation the system presents an enhancement of the perpendicular magnetic anisotropy. When thin Pd films are grown on magnetic substrates such as Ni, Co, or Fe, their magnetic properties can change due to electronic effects, which are also modulated by hydrogen absorption [1].

Here we present the magnetic characterization of ultrathin Co films (2-5 AL) on Ru(0001), before and after capping with a single atomic layer of palladium. The growth of the films was studied in real space by LEEM and the magnetic characterization was carried out in SPLEEM, which registers in real time the evolution of the magnetization easy-axis. Co/Ru(0001) shows out-of-plane anisotropy for 2 AL films, switching to in-plane in thicker films [2]. The addition of a single Pd atomic layer enhances the Co perpendicular anisotropy, pushing the out- to in-plane anisotropy transition up to 5 atomic layers. Finally, motivated by previous results where we reported the reversible spin reorientation transition of Co bilayers upon hydrogen exposure[3], we have investigated if the same phenomenology occurs in Pd/Co/Ru films by exposing them to both molecular and atomic hydrogen atmospheres.

## References

[1] T. Hübert, L. Boon-Brett, G. Black, and U. Banach, "Hydrogen sensors – A review," Sensors Actuators B Chem., vol. 157, no. 2, pp. 329–352, 2011

[2] F. El Gabaly et al., "Imaging Spin-Reorientation Transitions in Consecutive Atomic Co Layers on Ru(0001)," Phys. Rev. Lett., vol. 96, no. 14

[3] B. Santos et al. "Hydrogen-induced reversible spin-reorientation transition and magnetic stripe domain phase in bilayer Co on Ru(0001)," Physical Review B - Condensed Matter and Materials Physics 85 (2012)

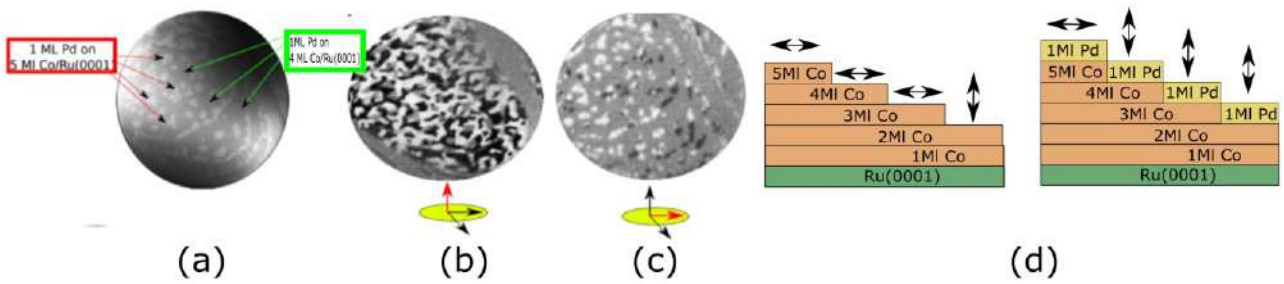



Figure 1. (a) LEEM recorded after growing 1 ML of Pd on 4 and 5 ML thick Co/Ru(0001). (b-c) SPLEEM image acquired with the electron beam spin polarization direction out-of-plane and in-plane, respectively. The FOV is 8 μm. The beam energy is 5.2 eV. (d) Schematic representation showing the magnetization easy-axis before and after depositing the Pd capping.

# Static and Dynamic Magnetic Properties of Amorphous CoAlZr Films and Composition Modulated Heterostructures

*A. Tryggvason*, S. Ingvarsson, F. Magnus*
*Science Institute, University of Iceland, Reykjavik, Iceland*
*asgeir13@gmail.com

Thin film amorphous magnets can be synthesized to have a variety of static and dynamic magnetic properties. The advantage of working with amorphous materials is that the composition can be tuned continuously and with good precision over a large range without phase segregation or defect formation. This facilitates precise control over the magnetic properties. The amorphous magnets synthesized for this study are $Co_x(Al_{0.7}Zr_{0.3})_{1-x}$ alloys with $0.7 < x < 1.0$, where the ratio of Co to AlZr determines the observed magnetic properties. Post-annealing of the CoAlZr alloy is also examined as a way to control the magnetic properties and study the effect of crystallization [1].

This study evaluates the compositional and structural dependence of the coercivity, anisotropy, magnetization and ordering temperature by vibrating sample magnetometry and magneto-optical Kerr effect magnetometry. The magnetization varies linearly with composition, as seen in Fig. 1, and the coercivity is extremely low (<0.2 mT) for all amorphous composition. The magnetic damping, which is crucial for spintronics and magnonics, is measured with ferromagnetic resonance measurements. Post-annealing of CoAlZr is found to increase the damping. The damping is also shown to decreases substantially with increasing amount of Co, as seen in Fig. 1, from relatively high $\alpha$ Gilbert damping of 0.05 to the low value of 0.01, which is comparable to permalloy.

In addition to single layers we study $Co_x(Al_{0.7}Zr_{0.3})_{1-x}$ heterostructures, where the composition is continuously varied. Multilayers of two different compositions of $Co_x(Al_{0.7}Zr_{0.3})_{1-x}$ with abrupt interfaces have previously been shown to display giant magnetic proximity effects where ferromagnetism or superparamagnetism is induced in an intrinsically paramagnetic layer through proximity to a high-$T_c$ layer [2]. The heterostructures introduced here are without abrupt interfaces and the composition is continuously modulated throughout the depth of the sample. We study the static and dynamic magnetic properties of such structures and compare to those of multilayers with abrupt interfaces. These heterostructures offer a new route to tune the magnetic response and could potentially be used as ultrasoft magnets in high frequency applications.

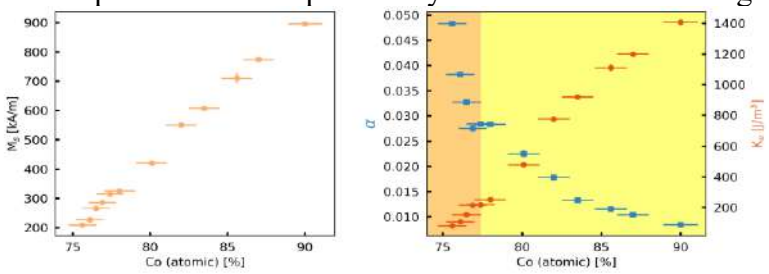



Figure 1: Magnetic properties of $Co_x(Al_{70}Zr_{30})_{100-x}$ for $x$ in the range 75-90 at%. The left figure displays the saturation magnetization and the right figure displays the Gilbert damping and the first order uniaxial anisotropy constant. The film thickness of compositions in the orange area is 60 nm and the compositions in the yellow area are 20 nm.

## Acknowledgements

This work was supported by funding from the Icelandic Research Fund Grants No. 228951, 218029 and 217843.

## References

1. A. Tryggvason, T. H. Frímannsdóttir, M. T. Sultan, K. A. Thórarinsdóttir, F. Magnus, S. Ingvarsson, "Structural and magnetic changes in CoAlZr thin films upon post annealing," 2022 International Semiconductor Conference (CAS), Poiana Brasov, Romania, 2022, pp. 181-184.
2. Thórarinsdóttir, K. A., Palonen, H., Palsson, G. K., Hjörvarsson, B., & Magnus, F. (2019). Giant magnetic proximity effect in amorphous layered magnets. Physical Review Materials, 3(5)

# Exchange Spring Evolution in $SmCo_5$/Fe and $NdCo_5$/Fe Magnetic Bilayers

*Thomas Saerbeck[1]*, Nina-Juliane Steinke[1], Marietta Seifert[2], Volker Neu[2]*

[1] *Institut Laue-Langevin, Grenoble, France*

[2] *Leibniz IFW Dresden, Dresden, Germany*

*saerbeck@ill.fr

High-anisotropy rare-earth alloys combined with soft magnetic transition metal materials, e.g. Fe, in thin film heterostructures form prime examples of exchange spring (ES) materials possessing high magnetizations and high magnetic anisotropy [1,2]. This promises a range of applications in logic circuits, sensor and magnetic storage technologies with high magnetic stability and durability.

Using polarized neutron reflectometry with polarization analysis (PNR), we present the chemical and magnetic structure as a function of depth in $SmCo_5$/Fe and $NdCo_5$/Fe bilayers grown on Cr-buffered MgO substrates. The films show epitaxial growth with magnetic easy axis of the rare-earth alloy oriented in the surface plane of the sample. Vibrating sample magnetometry is used to study the volume averaged response of the system to external magnetic fields. The proximity coupling between Fe and the rare-earth alloy is quantified through PNR by application of the magnetic field opposite to an initial saturation or away from the in-plane anisotropy easy axis, thus exerting a torque on the magnetizations of each layer. In the case of $SmCo_5$/Fe, we observe a significantly enhanced switching field of the whole Fe layer with a strongly depth dependent moment configuration as a function of in-plane magnetic field strength. Strong spin-flip signals of the neutron reflectometry as a function of field quantify the extension of moment canting away from the applied field towards the $SmCo_5$ interface.

The case of $NdCo_5$/Fe is particularly intriguing due to its giant magnetocaloric effect and a spin-reorientation transition observed at 255 K [3,4] during which the magnetic easy axis switches from the in-plane a-axis to the 90° rotated in-plane c-axis at low temperatures [4]. The transistion proceeds via an easy-cone formation in which both anisotropies are populated. PNR, recorded as a function of temperature and magnetic field orientation, shows a spiral moment configuration forming between the low and high temperature states. The field dependence indicates a larger extension of the moment canting into the Fe layer at low temperature, pointing towards a stronger proximity coupling.

## References

[1] Y. Liu *et al.*, Phys. Rev. B **83**, 174418 (2011);
[2] V.M. Uzdin *et al.*, Phys. Rev. B **85**, 024409 (2012).
[3] M. Seifert *et al.*, New J. Phys. **15**, 013019 (2013).
[4] B.M.S. Teixeira *et al.*, J. Phys. D: Appl. Phys. **49** 315002 (2016).

# Full Spin Polarized Ultrathin $La_{0.8}Sr_{0.2}MnO_3$ Films

*F. Stramaglia [1], G. Panchal [1], F. Nolting[1], C. A. F. Vaz [1]**

[1]*Paul Scherrer Institute, Forschungsstrasse 111, 5232 Villigen, Switzerland*

*carlos.vaz@psi.ch

We have studied the effect of using $LaMnO_3$ and $La_{0.45}Sr_{0.55}MnO_3$ buffer layers on the magnetic properties of epitaxial ultrathin $La_{0.8}Sr_{0.2}MnO_3$ films grown on $SrTiO_3$(001) substrate by molecular beam epitaxy. The surface quality is monitored during the deposition with reflection high energy electron diffraction (RHEED), shown in Fig. 1(a) for 5 unit cell (uc) thick $La_{0.8}Sr_{0.2}MnO_3$ on $LaMnO_3$ buffer layer. We characterize the samples using bulk magnetometry, electrical transport, and synchrotron x-ray spectroscopy as a function of $La_{0.8}Sr_{0.2}MnO_3$ thickness. We show that the introduction of the $LaMnO_3$ buffer layer leads to bulk-like magnetic moments for $La_{0.8}Sr_{0.2}MnO_3$ thickness down to 1 uc, while the $La_{0.45}Sr_{0.55}MnO_3$ buffer layer induces antiferromagnetic order on the first 3 uc $La_{0.8}Sr_{0.2}MnO_3$ and bulk-like moments at larger thicknesses. For both buffer layers (Figure 1(b)), the bulk-like moment is confirmed by a linear increase in the magnetization as a function of thickness, with a slope $m$=3.8 $\mu_B$/uc corresponding to the fully spin-polarized Mn moment. The results are confirmed by x-ray magnetic circular dichroism (XMCD), showing an increasing signal with $La_{0.8}Sr_{0.2}MnO_3$ thickness for both buffer layers. Moreover, our linear dichroism results show an orbital reorganization for the ferromagnetic samples. We attribute the full spin polarization of the $La_{0.8}Sr_{0.2}MnO_3$ thin films to a direct ferromagnetic/antiferromagnetic exchange coupling to the buffer layer. Our results highlight the role of spin exchange from the buffer layer in controlling the magnetic properties of ultrathin $La_{0.8}Sr_{0.2}MnO_3$.

**Acknowledgements**

This project is funded by the Swiss National Science Foundation (SNF) (Grant No. 200021_184684).

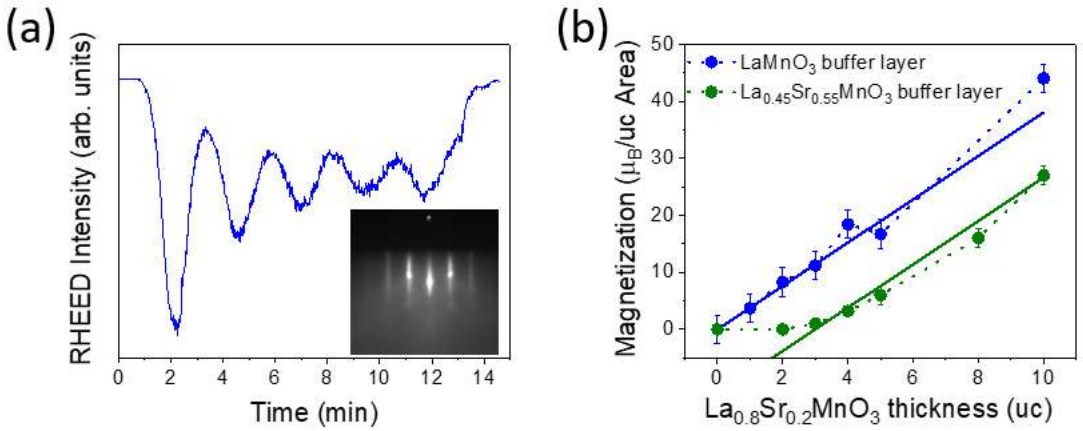



Figure1: (a) RHEED oscillations during the 5 uc $La_{0.8}Sr_{0.2}MnO_3$ deposition, indication of a layer-by-layer growth and used to monitor the film thickness in real time. Inset: final RHEED pattern showing that the film is epitaxial and the surface is smooth. (b) Magnetization of the $La_{0.8}Sr_{0.2}MnO_3$ layer as a function of its thickness extrapolated at zero field from the hysteresis loops for $LaMnO_3$ (blue) and $La_{0.45}Sr_{0.55}MnO_3$ (green) buffer layer. The straight lines have slope 3.8 $\mu_B$/uc, corresponding to the bulk-like fully spin polarized $La_{0.8}Sr_{0.2}MnO_3$ moment.

# Spin Reorientation Transition In Epitaxial Nd-Fe-B Thin Films With High Perpendicular Magnetic Anisotropy

*J. Soler-Morala[1]*, I. Arnay[1], G. Gzoukia[2], P. Pedraz[1], P. Perna[1], L. Alff[2], C. Navío[1], A. Bollero[1]*

[1] *IMDEA Nanoscience, C/ Faraday, 9, 28049 Madrid, Spain*

[2] *Institute of Materials Science, Technische Universität Darmstadt, Alarich-Weiss-Str.2, 64287 Darmstadt, Germany*

*Jimena.soler@imdea.org

Rare-earth intermetallic thin films posses high magnetic anisotropy and that makes them the perfect candidates for nanoscale applications such as high-density magnetic recording, spintronic devices, and microelectromechanical systems (MEMS) [1-4]. Among them, Nd-Fe-B is the most popular permanent magnet to date due to its outstanding instrinsic magnetic properties [8]. Nevertheless, few examples of Nd-Fe-B films with thicknesses below 50 nm are found in the literature.

In this study, Nd-Fe-B films grown by DC magnetron sputtering onto MgO substrates have been evaluated in the 20 - 40 nm thickness range. Structural, compositional, morphological and magnetic properties have been analysed (Fig. 1). Strong perpendicular magnetic anisotropy is achieved for a 40 nm film, reaching a coercivity of 8 kOe at room temperature (Fig. 1b). A detailed characterization of epitaxial NdFeB has been done by Grazing Incidence X-Ray Diffraction (GIXRD) (Fig. 1a). Lattice parameters are reported and the chemical states of Nd3d and Fe2p have been analysed by Hard X-Rays Photoelectron Spectroscopy (HAXPES). Furthermore, a low temperature study has been carried out, determining a Spin Reorientation Transition Temperature ($T_{SR}$) of 124.2 K (Fig. 1c), i.e., around 10 K below the theoretical value for $Nd_2Fe_{14}B$ (135 K) [6]. This change is attributed to the induced strain in the $Nd_2Fe_{14}B$ lattice, suggesting the possibility of tuning $T_{SR}$ with the appropriate design of parameters and architecture and, therefore, broadening the working temperature range of these film systems. Low temperature structural characterization has also been performed in order to provide some insight in the causes that promote the tilt of the magnetization.

## Acknowledgements

Authors acknowledge financial support from EU through the H2020 FET Open UWIPOM2 project (Ref. 857654) and from Comunidad de Madrid through NANOMAGCOST (P2028/NMT-4321). J. S-M. acknowledges financial support from Comunidad de Madrid (PEDJ-2029-PRE/IND-17045).

## References

[1] M. Nakano, S. Oshima, T. Yanai, H. Fukunaga, *J. Appl. Phys.* **115** (2014) 17A741
[2] A. Bollero, V. Neu, V. Baltz, D. Serantes *et al*, *Nanoscale* **12** (2020) 1155-1163
[3] T. S. Chin, *J. Magn. Magn. Mater*. **209** (2000) 75-79
[4] N. M. Dempsey, A. Walther, F. May, D. Givord et al. *Appl. Phys. Lett.* **90** (2007) 0925091-0925093
[5] A. C. Sun, Y. T. Lin, Y. S. Chen, *Thin Solid Films* **660** (2018) 879-884
[6] S. K. Pal, K. H. Müller, L. Schultz, *Phys. Lett. A* **385** (2021) 1269541-1269545

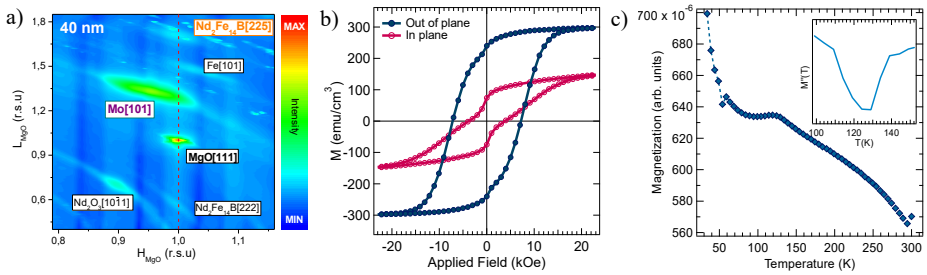



Figure 1: a) Reciprocal Space Map across MgO[111], Mo[101] and $Nd_2Fe_{14}B$[225] reflections measured by GIXRD using a beam energy of 15 keV (λ = 0.826 Å) for a 40 nm film, b) Out of plane and in plane hysteresis loops for *t* = 40 nm. c) Curve representing magnetization as a function of temperature for t = 40 nm. Inset shows the second derivative of magnetization with temperature used to obtain $T_{SR}$.

# Spin-Glass State in Strained $La_{2/3}Ca_{1/3}MnO_3$ Thin Films

*N. Marcano[1]*, I. Lucas[2,3], T. Prokscha[4], C. Magén[2,3,5], R. Corcuera[2,3], L. Morellón[2,3], J. M. De Teresa [2,3,5], M. R. Ibarra[2,3,5] and P. A. Algarabel[2,3]*

[1] *Centro Universitario de la Defensa, Academia General Militar, 50090 Zaragoza, Spain*
[2] *INMA, CSIC-Universidad de Zaragoza, 50009 Zaragoza, Spain.*
[3] *Dept. de Física de la Materia Condensada, Universidad de Zaragoza, 50090 Zaragoza, Spain.*
[4] *Laboratory for Muon Spin Spectroscopy, Paul Scherrer Institut, CH-5232 Villigen PSI, Switzerland.*
[5] *Laboratorio de Microscopías Avanzadas (LMA), Universidad de Zaragoza, 50018 Zaragoza, Spain.*
*marcanon@unizar.es

Manganites are one of the archetypes in the field of functional complex oxides, as they combine numerous exotic properties interplayed, such as colossal magnetoresistance, large spin polarization, multiferroicity, etc., which make them appealing canditates for fundamental studies as well as for spintronic applications. Their perovskite crystal structure allows the growth of epitaxial thin films and the fabrication of high quality heterostructured devices. Strain engineering has become one of the most popular routes to optimize the physical properties and introduce new functionalities into the deposited epitaxial thin film oxides. In the case of $La_{2/3}Ca_{1/3}MnO_3$ (LCMO), a previous work [1] demonstrates that, depending on the deposition conditions, a certain Non-FerroMagnetic (NFM) percentage of the thickness of the LCMO, can be obtained at the top of the LCMO film, the whole film being chemical and structurally homogeneous at room temperature. Theoretical calculations support that depending on the tetragonallity of LCMO, defined as $\tau = |c-a|/a$, a NFM layer can be obtained due to the competition between ferromagnetic (FM) and antiferromagnetic (AF) states. Such a NFM layer was previously characterized as AF in nature based on the presence of exchange bias (EB) in the magnetic hysteresis loops at low temperatures (10K).

Here, we present a comprehensive magnetic characterization of the strain-induced segregated NFM layer by means of field-cooling and zero-field-cooling magnetization versus temperature measurements, field-cooled hysteresis loops, relaxation of thermoremanent magnetization and low-energy muon spin relaxation experiments (LE-μSR). The relevance of understanding the magnetic nature of this NFM layer lies on the fact that this could be utilized as a native pinning layer of the same material in magnetic tunnel junction fabrication. The sensitivity and depth-profiling capacity of LE-μSR have evidenced the different magnetic behaviour of the two spatially segregated layers (NFM-FM). Our experimental results discard the AFM nature and support the spin-glass-like scenario for the NFM layer [2]. This scenario can be understood in the framework of the competing FM and AF interactions of similar energies, as suggested by the previous DFT+U calculations of the magnetic ordering of strained LCMO [1]. Additionally, the high spatial resolution of the Electron Holography (EH) technique has been used to locally image the magnetic changes present in this epitaxially strained LCMO thin films (Figure 1).

## Acknowledgements

This research was funded by Spanish MCIN/AEI/10.13039/5011000011033, grant number PID2020-112914RB-100, the Aragón Regional government grants number E13_17R, E13_20R and E28_20R.

## References

[1] L. Marín, L.A. Rodríguez, *et al.*, *Nano Lett.* **15** (2015) 492-497.
[2] I. Lucas, N. Marcano, *et al.*, *Nanomaterials* **12** (2022) 3646-3658.

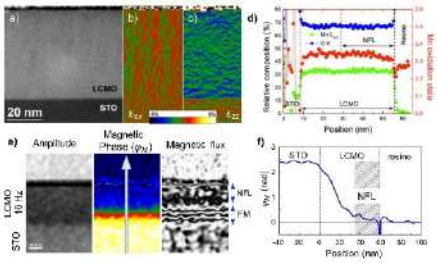



Figure 1: Structural, chemical, and magnetic characterization of the NFM of the LCMO thin film grown on STO (100).

# Polarised neutron reflectivity to resolve interfacial spin canting in a ferromagnetic metal-semiconductor bilayer

*A. J. Caruana[1]*, P. Muduli[2], H. Isshiki[3], J. Puebla[4], C. J. Kinane[1], Y. Otani[3, 4], N. Leo[5]*

[1] *ISIS Neutron and Muon Source, STFC, RAL, Oxfordshire, OX11 0QX, UK*
[2] *Department of Physics, Indian Institute of Technology Madras, Chennai, 600036, India*
[3] *Insitute for Solid State Physics, University of Tokyo, Kashiwa, 277-8581, Japan*
[4] *Center for Emergent Matter Science, RIKEN, Wako, Saitama 351-0198, Japan*
[5] *Instituto de Nanociencia y Materiales de Aragón (INMA), CSIC-Universidad de Zaragoza, Zaragoza, 50009, Spain*
*andrew.caruana@stfc.ac.uk

Ferromagnetic semiconductors and insulators have gained interest due to their potential to increase the efficiency of spintronic devices. This improvement arises from the fact that in insulators, such as europium sulphide EuS, spin currents and spin orbit torques do not involve dissipative charge currents, and thus avoid unnecessary losses due to Joule heating, as in ohmic resistive materials. Furthermore, crucial to developing more power-efficient devices for practical applications based on phenomena such as magnetoresistive response, dynamic behaviour, and spin-transfer torque switching – is the understanding the structural and magnetic coupling e.g., between ferromagnetic insulator/ferromagnetic metal bilayers, as key spintronic phenomena are typically localised at the thin-film interface between materials [1].

Inspired by interesting features found in the magnetoresistance of EuS|Py bi-layers indicating non-collinear interfacial magnetic coupling, here we present results from Polarised Neutron Reflectometry with Polarisation Analysis (PNR-PA), taken at the POLREF beamline to investigate the in-plane canting at the EuS|Py interface as a function of depth [2]. We found a significant neutron spin flip signal (Fig. 1a), indicating sizable spin canting in the bilayer system. From model fits to determine the depth-dependent magnetisation profile (Figs. 1b and c), we find that this spin canting is strongest at the EuS|Py interface and decays further away from the Py, aligning back to the guide field direction $H_g$ (i.e., the applied field direction). At low temperatures, this canting persists for applied magnetic fields up to 75 mT, and disappears above the EuS ferromagnetic transition temperature $T_C$=16 K. The strong spin canting localised at the interface between ferromagnetic metals and insulators could therefore be interesting for spintronic applications, such as spin-transfer torque switching.

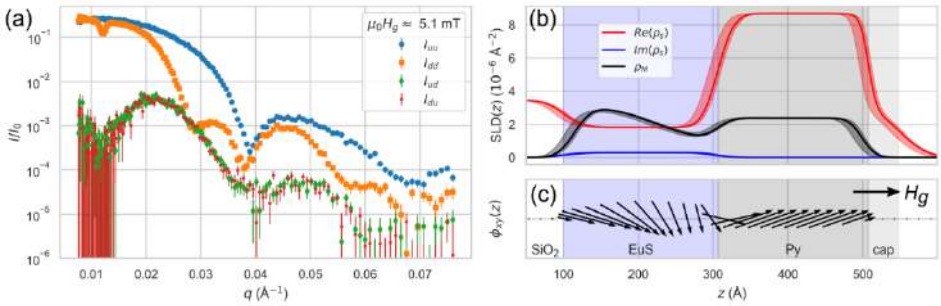



Figure: (a) Non-spin-flip and spin-flip neutron reflectometry data. (b,c) Fit results for the shown data, including depth-dependent structural and magnetic scattering length densities $\rho_s(z)$ and $\rho_M(z)$ as well as the depth-dependent in-plane spin canting $\phi_{xy}(z)$.

## Acknowledgements

NL acknowledges support by the European Community under the Horizon 2020 Program, Contract No. 101001290 (3DNANOMAG).

## References

[1] F. Hellman *et al.*, "*Interface-induced phenomena in magnetism*" Rev. Mod. Phys. **89** 25006, (2017).
[2] Naëmi Leo *et al.*; 2022: "*Understanding the magnetoresistance of a ferromagnetic metal-semiconductor bilayer*", STFC ISIS Neutron and Muon Source, https://doi.org/10.5286/ISIS.E.RB2220727-1 and https://doi.org/10.5286/ISIS.E.RB2220727-2.

# Magnetism of Magnetite/Hematite Epitaxial Bilayers

*Oscar Rodríguez de la Fuente[1]*, Mauricio Prieto[2], Lucas Caldas[2], Liviu C. Tanase[2], Eugenia Sebastiani[3], Jesús López-Sánchez[3], Thomas Schmidt[2], Juan Rubio-Zuazo[3]*

[1] *Departamento de Física de Materiales, Universidad Complutense de Madrid, Madrid, Spain*
[2] *Fritz-Haber-Institute of the Max-Planck-Society, Department of Interface Science, Berlin, Germany*
[3] *ICMM-CSIC, Madrid, Spain and SpLine-Spanish CRG BM25 beamline at the ESRF, Grenoble, France*
*oscar.rodriguez@fis.ucm.es

Oxides give rise to novel phenomena when different types are combined, and the production of magnetic heterostructures is an everlasting topic in the development of new materials. Thus, the exploration of new fabrication methods is mandatory. Low energy ion bombardment has been used on single-crystalline oxide thin films to produce epitaxial layers of the corresponding suboxides [1,2]. We have carried out an investigation of the magnetite/hematite epitaxial bilayer ($Fe_3O_4(111)/\alpha\text{-}Fe_2O_3(0001)$) on different substrates. This is an ideal oxide system to study magnetic exchange effects at the interface, since it combines two types of magnetism (antiferromagnetism in hematite, ferrimagnetism in magnetite) and two phase transitions (Morin in hematite, Verwey in magnetite). The characterization of this system has included the structural, chemical and magnetic properties. We have used synchrotron-based experimental techniques such as Surface X-ray Diffraction (SXRD), Hard X-ray Photoemission Spectroscopy (HAXPES), Low Energy Electron Microscopy (LEEM), Photoemission Electron Microscopy (PEEM) or X-ray Magnetic Dichroism (XMD). The aim of this work has been to obtain spectromicroscopic information on the as-grown material and on the evolution of the materials during the ion bombardment, as well as during high temperature annealing of the bilayers. Ion bombardment gradually reduces the initial hematite into a defective sub-oxide where magnetite nuclei start to grow with increasing ion doses and coalesce into a defective epitaxial magnetite upper layer. Rotational domains emerge, with small grains which are typically a few tens of nanometres large. They do not grow significantly with temperature annealings. This robustness may be explained by the incoherent growth of the grains which favour the formation of twin and antiphase boundaries. Finally, we study the distribution of magnetic domains (which do not apparently correlate with the structural domains) and the existence of exchange bias and other types of interfacial exchange effects.

## References

[1] B.M. Pabón et al., *Nat. Comm.* **6** (2015) 6147.
[2] S. Ruiz-Gomez et al., *Appl. Phys. Lett.* **110** (2017) 093103.

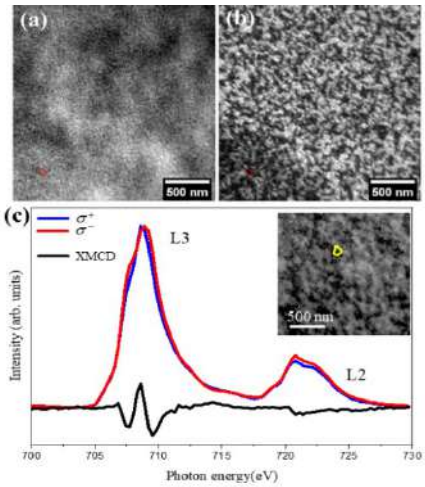



Figure: XMCD (a) and DF-LEEM (b) images of magnetite. A smooth dichroic contrast is obtained. c) XMCD spectra under different helicities ($\sigma^+$ and $\sigma^-$) integrated on a specific region (in yellow) of the magnetite surface. The insert is an XMCD image.

# Unravelling the Role of Sm in the Magnetic Interactions in $SmFeO_3$

*D. Amoroso*[1], *B. Dupé*[*1,2], *M. Verstraete*[1]

[1] *Nanomat/Qmat/CESAM, Université de Liège, 4000 Sart-Tilman, Belgium*

[2] *Fonds de la Recherche Scientifique (FNRS-FRS), 1000 Bruxelles, Belgium*

*bertrand.dupe@uliege.be

Magnetic rare-earth orthoferrites $RFeO_3$ exhibit a rich playground ranging from multiferroicity to spin-reorientation, strong magnetostriction and ultrafast light-driven manipulation of magnetism, which can be exploited in spintronics [1]. Such properties stem from the strong spin-orbit coupling (SOC) interaction combined with lattice vibrations (phonons) due to the presence of two magnetic ions ($R^{3+}$-$Fe^{3+}$) in different sublattices.

In $SmFeO_3$, recent experiments attempted to clarify the non-collinear magnetic ground-state, the origin of the spin-reorientation and low temperature magnetization compensation. Furthermore, a peculiar temperature evolution of vibrational mode has been measured which suggest a strong coupling between phonon and magnons [2]. Although a conclusive picture has not yet been reached, theoretical studies that explore the microscopic mechanisms at play are still rare.

Here, using density functional theory calculations to explore the magnetic ground state of $SmFeO_3$. We explore the interplay between magnetic states and phonons, and show the anomalous modes found experimentally are of two types, sensitive to either the f-electron magnetism and/or the volume. We obtain an effective magnetic Hamiltonian and highlight the fundamental role of the Sm f-orbital SOC contribution to the magnetocrystalline anisotropy. The combination of this anisotropy with a modest Fe-Sm magnetic exchange provides a possible mechanism for the spin-flop transition in the magnetic phase diagram.

**Acknowledgements**

This work was supported by the m-era.net project SWIPE funded by FNRS Belgium grant PINT-MULTI R.8013.20, and by computing time on the Belgian share of the LUMI supercomputer (EuroHPC JI hosted by CSC Finland), and on the Tier-1 supercomputer of the Fédération Wallonie-Bruxelles infrastructure (grant agreement n. 1117545), through the Consortium des équipements de Calcul Intensif.

**References**

[1] D. Afanasiev, J. R. Hortensius, B. A. Ivanov, A. Sasani, E. Bousquet, Y. M. Blanter, R. V. Mikhaylovskiy, A. V. Kimel, and A. D. Caviglia, *Nature materials* **20**, 607 (2021).

[2] M. C. Weber, M. Guennou, D. M. Evans, C. Toulouse, A. Simonov, Y. Kholina, X. Ma, W. Ren, S. Cao, M. A. Carpenter, B. Dkhil, M. Fiebig, and J. Kreisel, *Nature Communications* **13**, 443 (2022).

[3] D. Amoroso, B. Dupé and M. Verstraete *Phys. Rev B* in press

# Thermal Spin Trasnport Effects in Ferrimagnetic/Superconducting Hybrid Heterostructures

*Rubén Corcuera[1]*, Irene Lucas[1], Pilar Jiménez-Cavero[2], Pedro Alonso[1], Noa Varela[3], Pedro Antonio Algarabel[1], Francisco Rivadulla[3], Luis Morellón[1]*

[1] *Instituto de Nanociencia y Materiales de Aragón, Universidad de Zaragoza-CSIC, 50018 Zaragoza, Spain*

[2] *Centro Universitario de la Defensa, Academia General Militar, 50090 Zaragoza, Spain*

[3] *Centro Singular de Investigación en Química Biolóxica e Materiais Moleculares, Universidade de Santiago de Compostela, 15782 Santiago de Compostela, Spain*

*r.corcuera@unizar.es

Energetic efficiency is one of the bottlenecks in technological development of new devices; in that respect, spintronics proposes the use of the electron's spin degree of freedom in order to carry information in electrically insulating materials, thus avoiding power dissipation by Joule's effect.

Therefore, hybrid structures formed by ferrimagnetic (FM) and superconducting (SC) nanometric layers are an interesting combination taking into account that in a superconductor, spin-charge separation may give rise to different characteristic length scales [1,2]. Thermal injection of spin polarized currents into superconductors allows the spin current not to be supressed and being able to be propagated over long distances by thermally excited quasiparticles [3]. Therefore, the combination of SC and FM thin films forming bilayers of potential use in spintronic devices has recently attracted great interest from the scientific community.

Spin Seebeck Effect (SSE) induced by thermal gradients or spin pumping through ferromagnetic resonance (FMR) are two of the recently studied techniques for spin injection in SC materials. Both techniques are highly dependent on the interface quality between the FM and the SC layer, thus the understanding and control of this quality becomes crucial to optimize the combination of these materials for spintronic applications.

The detection of spin currents is typically achieved by the conversion of these currents into a measurable voltage; Inverse Spin Hall Effect (ISHE) is the response of a non-magnetic (NM) layer with a high spin-orbit coupling to a spin current.

This work is devoted to study the interactions between spin currents, electrical currents, and heat, in FM/SC hybrid structures. The selected materials are Yttrium Iron Garnet (YIG, $Y_3Fe_5O_{12}$) as FM, Yttrium-Barium Copper Oxide (YBCO, $YBa_2Cu_3O_{7-\delta}$) as SC, and Platinum (Pt) as NM.

The fabrication of the FM/SC bilayers has been performed via Pulsed Laser Deposition (PLD). The deposition of a thin layer of Platinum to detect the spin currents onto the bilayers was performed by DC Sputtering.

Both types of bilayers ($Gd_3Ga_5O_{12}$//YIG/YBCO and MgO//YBCO/YIG) were structurally, electrically and magnetically characterized. A complete structural characterization has been performed by means of X-Ray Scattering experiments (diffraction and reflection) in a high resolution diffractometer; also, detailed scattering experiments were performed using Synchrotron Radiation to ensure the crystalline quality of the bilayers.

SSE has been studied in both structures. For samples with the SC layer on top, ion milling has been used to remove part of the SC layer on the edges of the sample to get the NM layer in contact with the FM Layer. The effect of the SC layer has been examined by performing the SSE-ISHE experiments below and above the superconductor critical temperature.

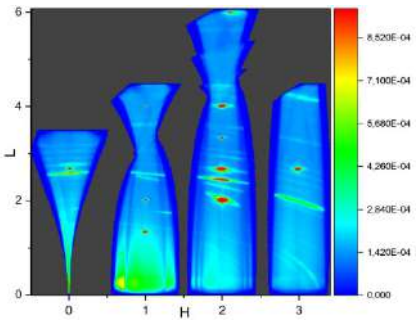



Figure: In-plane diffraction maps of the bilayers studied with synchrotron radiation.

## References

[1] S.A. Kivelson and d. S. Rokhsar, Phys. Rev. B, 41, 11693(R) (1990)

[2] F.S. Bergeret, M. silaev, P. Virtanen and T.T. Heikklä, Rev. Mod. Phys. 90, 041001 (2018)

[3] M. Silaev, P. Virtanen, F.S. Bergeret and T.T. Heikklä, Phys. Rev. Lett, 114, 167002 (2015)

# Tuning interfacial spin-orbit related effects at Co|Al interface

*S. Krishnia[1], N. Sebe[1], Y. Sassi[1], L. M. Vicente-Arche[1], S. Collin[1], T. Denneulin[2], A. Kovács[2], R. E. Dunin-Borkowski[2], A. Thiaville[3], A. Fert[1], J.-M. George[1], H. Jaffrès[1], N. Reyren[1] and V. Cros[1,]**

[1] *Unité Mixte de Physique, CNRS, Thales, Université Paris-Saclay, 91767, Palaiseau, France.*
[2]*Ernst Ruska-Centre for Microscopy and Spectroscopy with Electrons (ER-C 1) and Peter Grünberg Institut (PGI-5), Forschungszentrum Jülich GmbH, 52425 Jülich, Germany.*
[3]*Laboratoire de Physique des Solides, Université Paris-Saclay, CNRS, Orsay 91405, France*
*vincent.cros@cnrs-thales.fr

Spin-orbit coupling at interfaces in inversion-asymmetric magnetic multilayers is responsible for most of interesting phenomena occurring in modern nanomagnetism and spintronics, such as the interfacial anisotropies, the Dzyaloshinskii–Moriya interaction as well as for charge-to-spin interconversion mechanisms. A typical structure of the multilayers of interest is composed of an heavy material (e.g. Pt), a magnetic material (e.g. Co) and a lighter element (e.g. Ru, Ir, Cu etc..) in which chiral domain walls or skyrmions can be stabilized [1]. Indeed, the role of light element interface on all the properties mentioned earlier in such multilayers has been often overlooked and need some more attention.

In this study, we have investigated how the interfacial anisotropy and DMI evolve with the oxidation degree at the interface in the spintronics toy system: Ta|Pt|Co|Al(AlOx). Historically, it is considered that perpendicular magnetic anisotropy (PMA) in Pt/Co/AlOx system is promoted by the hybridization of d-orbits of Co with p-orbits of oxygen at the Co/AlOx interface and therefore an optimium oxidation condition is necessary. Manchon et al found that in the absence of considerable oxygen atoms at Co/AlOx interface, the Co magnetization goes in-plane [2]. Here, we have used magnetron sputtering to deposit Ta(5)|Pt(8)|Co(0.9)|Al($t_{Al}$) series of samples by varying Al thickness from 0.1 nm to 3 nm. Our XPS results revel that for $t_{Al}$ = 0.6 nm, the Co remains unoxidized, whereas the Al is fully oxidized, establishing perfect condition to achieve PMA. For this condition, the anisotropy of Co is found to be 1.2 T, which decreases with Al thickness and goes in-plane, as expected. However, the striking results we find is that the anisotropy of Co goes form in-plane to out-of-plane at $t_{Al}$ =1.6 nm, where we have flat metallic Al interface. The strength of PMA further increases with Al thickness and saturates at 1.8 T, a much higher value than the $H_k$ at Co/AlOx interface. Additionally, we also have quantified the DMI using Brillion light spectroscopy (BLS). Figure 1(b), shows the evoulation of $D_{eff}$ as a function of Al thickness. The DMI shows $1/t_{Al}$ dependace as a function of Al thickness in the measured range. As Al is a light element with negligible spin-orbital coupling, its strong impact of interfacial mechanisms suggests the occurance of novel interfacial phenomenon. Additionally, the charge-to-spin conversion mechanisms and resultant spin-orbit torques in this system will be discussed in details.

**Acknowledgements**

This work has been supported by the ANR-20-CE30-0022 “ORION”. N. Sebe benefits from a France 2030 government grant managed by the ANR (ANR-22-PEPR-Electronique-EMCOM).

**References**

[1] A. Fert *et al*, *Nat. Rev. Materials* **1** (2017) 17031.
[2] A. Manchon *et al*, *J. Appl. Phys.* **104**, 043914 (2008).

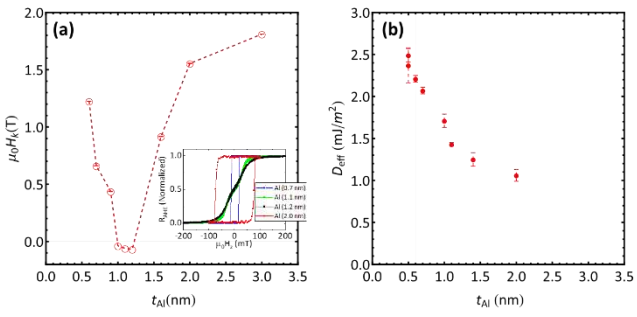



Figure: (a) Al layer thickness dependence of out-of-plane magnetic anisotropy in Ta(5)|Pt(8)|Co(0.9)|Al($t_{Al}$) series of samples. Inset shows the normalized anamolous Hall effect loops as a function of out-of-plane magnetic field for four different Al thicknesses. (b) The effective DMI constant as a function of Al thickness.

# Anomalies in the Modulation of Surface Acoustic Waves due to Magnetostrictive Effects

*J.D. Aguilera[1]*, L. Soria[1], R. Loriente[1], R. Rancha1[1,2], I. Gràcia[3], S. Vallejos[3], A. Hernando[1], P. Marín[1,2], P. de la Presa[1,2], D. Matatagui[1,2]*

[1] *Instituto de Magnetismo Aplicado (UCM-ADIF-CSIC), A6 km. 22.500; 28230 Las Rozas, Spain*
[2] *Dpto. de Física de Materiales, Facultad de Ciencias Físicas, Universidad Complutense de Madrid, Plaza de las Ciencias 1, 28040 Madrid, Spain*
[3] *Instituto de Microelectrónica de Barcelona (IMB, CSIC), Campus UAB, Bellaterra, 08193, Spain*
*juandiag@ucm.es

A wide range of high-performant sensors is based on the variations of the amplitude and phase of a surface acoustic wave (SAW) propagating through a piezoelectric substrate. On the other hand, magnetostrictive materials have found wide applicability as transducers in sensors and actuators due to their ability to transform mechanical energy into magnetic energy and viceversa.

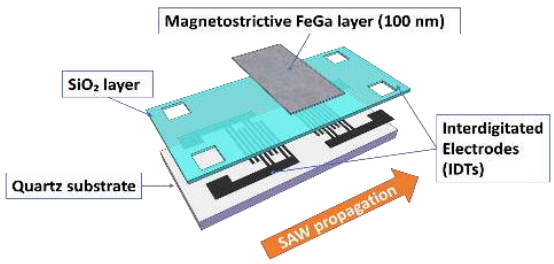
In this work, we study the behaviour of a device, which combines magnetostriction and SAWs by depositing a magnetostrictive film, which can induce strain when applying a magnetic field onto the SAW device. There are only a few precedents to this idea in literature [1-2]. A 3.6 μm film of amorphous $SiO_2$ deposited on crystalline quartz guides the shear horizontal SAW (Love wave), which is generated and detected by two interdigitated transducers (IDTs) (see Fig. 1). The graphs (Fig. 2) shown below were obtained from a sample consisting in a 100 nm polycrystalline magnetostrictive Galfenol ($Fe_{72}Ga_{28}$) layer, which is deposited by sputtering onto the piezoelectric substrate. The IDTs are connected to a feedback loop circuit which provides a resonant SAW frequency (around 160 MHz) that is tracked in real time as the external magnetic field varies.

Fig. 2 shows the frequency shift observed as the applied magnetic field sweeps from -40 to 40 mT (and back) forming an angle of 0º and 45º respectively with the direction of propagation of SAWs. For an angle of 45º, the coercivity of the Galfenol film can be deduced, corresponding to the position of the two central peaks, and the highest applied field nearly saturates the sample. This behaviour can be directly related to the hysteresis loop. In the second case, 40 mT is not enough to saturate the film and, most of all, four relative minima appear, while only two could be seen for 45º, revealing that the resonant frequency has a non-monotonic behaviour with the modulus of magnetisation. This anomalous feature, which has not been reported in precedent literature on the topic, arises from a more complex process of magnetization and its interaction with acoustic waves. We believe that this phenomenon shows great potential to be exploited for different sensing applications.

## Acknowledgements

This research has been funded by Ministerio de Ciencia, Innovación y Universidades (grant number PID2021-123112OB-C21 and PDC2022-133039I00).

(1)




(2)

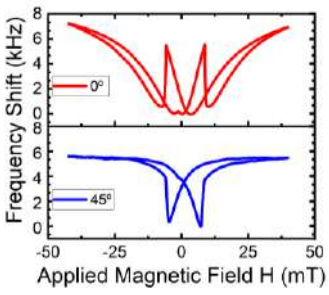



*Figure 1*: Schematics of the device. *Figure 2*: Frequency shift against applied field for 0º (a) and 45º (b) configuration

## References

[1] Fahim, et al., *A simple and novel SAW magnetic sensor with PVA bound magnetostrictive nanopowder film.* Sensors and Actuators A: Physical, 2021. **331**: p. 112926.

[2] Meyer, et al., Thin-film-based saw magnetic field sensors, Sensors, 2021. **24**, art. no. 8166, .

# Magnesium diboride thin films for superconducting spintronics

*C. Pfaff[1]*, L. Pasquier[1], S. Andrieu[1], S. Petit-Watelot[1], S. Migot[1], J. Ghanbaja[1], M. Emo[1], M. Hehn[1], S. Mangin[1], K. Dumesnil[1], T. Hauet[1]*

[1]*Institut Jean Lamour, Université de Lorraine, 2 allée André Guinier, 54011 Nancy, France*

*corentin.pfaff@univ-lorraine.fr

The interplay of magnetism and superconductivity has been heavily studied the past 30 years and recently led to the concept of superconducting spintronic [1]. Nevertheless proximity effect, inverse proximity effect or injection of quasiparticles at superconductor/ferromagnet (S/F) interfaces have been essentially investigated in niobium or aluminium-based heterostructures grown by sputtering [2] with critical temperatures (Tc) of the order of few Kelvin. S/F stacks with a higher Tc and carefully tuned interfaces would enable the thorough exploration of new superconducting spintronic features, as well as the possible implementation in operating quantum devices.

We report here the growth of epitaxial $MgB_2$ and $MgB_2$/F films by Molecular Beam Epitaxy. Depending on the UHV growth conditions, we were able to control either textured polycrystalline or single crystalline $MgB_2$ films, which were characterized by RHEED, XPS, XRD and TEM, as shown in Fig. 1(a). The critical temperature is systematically higher for single crystalline films and were measured for thicknesses as low as 5 nm (Fig. 1 (b)). Tc reaches 30K for films thicker than 15nm, in good agreement with Refs [3,4]. Using BCS model, electrical transport and magnetic measurements reveal a typical coherence length of 5nm at 0 K for the single crystalline $MgB_2$ films.

The growth of a Co or Permalloy layer on top of $MgB_2$ only reduces Tc by about 1K and allows us to investigate $MgB_2$/F stacks having Tc above or close to 30K. The temperature dependent magnetization damping of the F layer was extracted from FMR measurements, in order to probe spin transport from F into $MgB_2$. As expected from previous works on Nb/$Ni_{80}Fe_{20}$ [2] and NbN/$Ni_{80}Fe_{20}$ [5], the channel of momentum loss in the $MgB_2$ layer is suppressed by opening of the superconducting gap below Tc. This results in the drop of the damping parameter below Tc in $MgB_2$/$Ni_{80}Fe_{20}$ (Fig.1(c)) and $MgB_2$/Co bilayers.

Our results on epitaxial $MgB_2$-based thin films and heterostructures are promising for the investigation of superconducting spintronic physics over a large range of temperature and under temperatures larger than the $H_2$ liquid-gaz transition. They would also enable studying the properties related to this specific superconducting material in its single crystalline form.

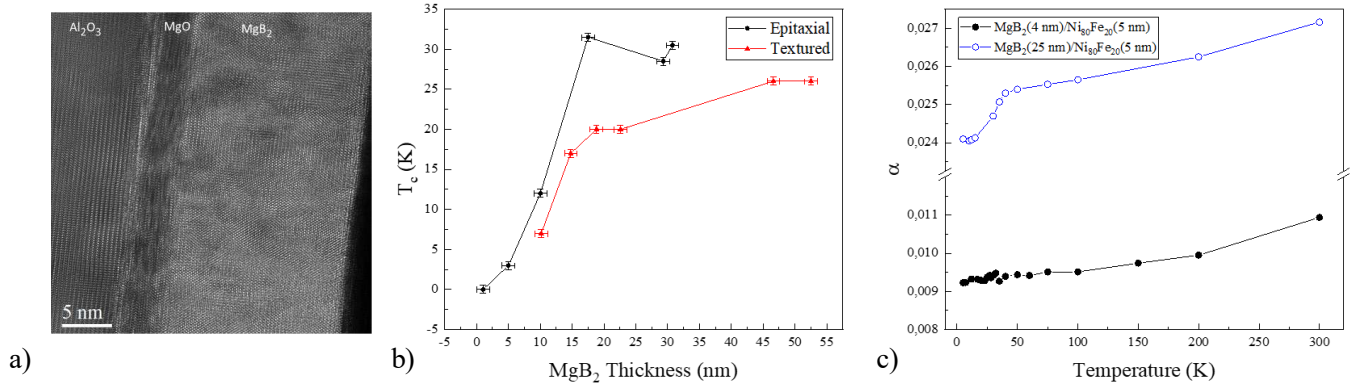



*Figure 1 : a) High resolution TEM micrograph of a $Al_2O_3$/MgO/$MgB_2$/Au structure. b) Critical temperature of single crystalline (black) and textured (red) $MgB_2$ thin films measured for various thicknesses. c) Variation of Gilbert damping for two $MgB_2$/$Ni_{80}Fe_{20}$ stacks, with (blue, empty symbols) and without (black, filled symbols) superconducting transition.*

## References

[1] J. Linder and J. W. A. Robinson, Nature Phys **11**, 307 (2015).

[2] C. Bell, S. Milikisyants, M. Huber, and J. Aarts, Phys. Rev. Lett. **100**, 047002 (2008).

[3] C. Zhang, Y. Wang, D. Wang, Y. Zhang, Z.-H. Liu, Q.-R. Feng, and Z.-Z. Gan, Journal of Applied Physics **114**, 023903 (2013).

[4] M. Naito and K. Ueda, Supercond. Sci. Technol. **17**, R1 (2004).

[5] M. Müller, L. Liensberger, L. Flacke, H. Huebl, A. Kamra, W. Belzig, R. Gross, M. Weiler, and M. Althammer, Phys. Rev. Lett. **126**, 087201 (2021).

# Tunable martensitic transformation towards cryogenic temperature range in all-d-metal Ni(Co)-Mn-Ti Heusler alloys

*Aun N. Khan[1]*, Álvaro Díaz-García [1], Luis M. Moreno-Ramírez[1], Jia Yan Law[1], Victorino Franco[1]*

[1] *Dpto. Física de la Materia Condensada. ICMS-CSIC. Universidad de Sevilla, P.O. Box 1065, Sevilla 41080, Spain*

*akhan@us.es

All-d-metal Ni(Co)-Mn-Ti Heusler alloys have recently gained popularity in the magnetocaloric community because of their intriguing functional and mechanical properties, especially that the type of elements used in their compositions can greatly improve performance: generally, Co substituting Ni allows for a significant magnetization change to occur during the magnetostructural transformation and stabilizes the austenitic phase while Fe substituting Mn decreases the entropy change of the transformation and improves the separation of martensitic and Curie transitions without lowering large magnetization difference between both phases [1]. Their properties are highly dependent on the synthesis route, microstructure, and composition [2], with the latter placing a lot of emphasis on ferromagnetic elements. According to the composition formula, Mn (anti-ferromagnetic nature) accounts for a considerable portion of the total composition. It is always used in excess due to its ease of evaporation loss during fabrication because of its high vapor pressure but it has received the least research attention regarding its integral role in the alloy properties. In this work, we present a series of $Ni_{35}Co_{15}Mn_{35+x}Ti_{15}$ alloys (x = 1.25, 2.5, 5, 10 wt.%), designated as $Mn_{1.25}$, $Mn_{2.5}$, $Mn_5$ and $Mn_{10}$, whose magnetostructural transformation temperatures are tuned to the cryogenic temperature range (see $Mn_{10}$ alloy in Figure 1 whose transition temperature is at 120 K while cooling) while Curie transitions of austenite remained unaltered. Furthermore, these alloys show large isothermal entropy changes for moderate magnetic fields of 1.5 T, which increase with Mn content, saturating at 13.1 J $kg^{-1}$ $K^{-1}$ (Figure 1 (b)). The observed saturation is associated to the increasing magnetic contribution of the martensitic phase (related to a lower transition temperature), which reduces the magnetization change during the transformation.

## Acknowledgements

Work funded by grant PID2019-105720RB-I00/AEI/10.13039/501100011033, Consejería de Economía, Conocimiento, Empresas y Universidad de la Junta de Andalucía (grant P18-RT-746), and Air Force Office of Scientific Research (FA8655-21-1-7044). ANK acknowledges a FPI scholarship from the Spanish MCIN/AEI. JYL acknowledges EMERGIA fellowship from Junta de Andalucía (ref. EMC21_00418).

## References

[1] A.N. Khan, L.M. Moreno-Ramírez, Á. Díaz-García, J.Y. Law, V. Franco, J Alloys Compd. **931** (2023) 167559.

[2] A. Taubel, B. Beckmann, L. Pfeuffer, N. Fortunato, F. Scheibel, S. Ener, T. Gottschall, K.P. Skokov, H. Zhang, O. Gutfleisch, Acta Mater. **201** (2020) 425–434.

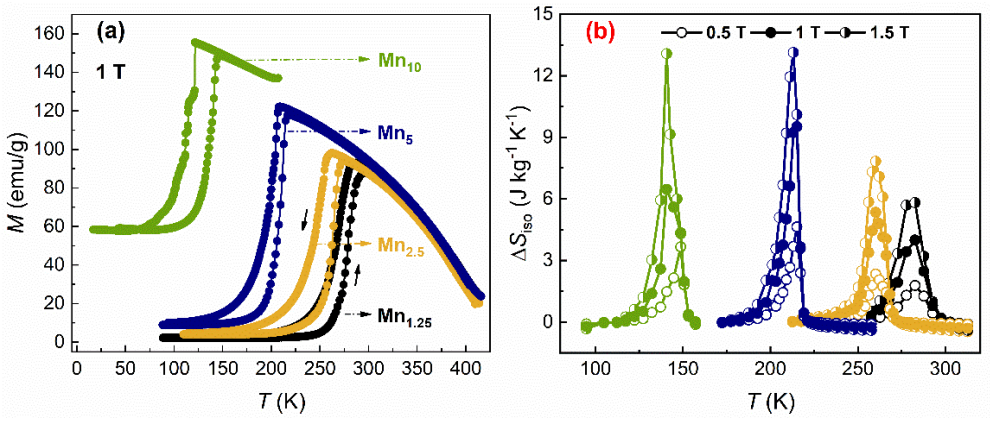



Figure 1: Temperature dependence of (a) magnetization at 1 T and (b) the isothermal entropy change at moderate magnetic fields for the studied alloy series.

# Spin-Gapless Semiconducting Heusler-Alloy Films

*Tiffany Sarfo [1], Wei Li [1,2] and Atsufumi Hirohata [1]*

[1] *School of Physics, Engineering and Technology, University of York, York YO10 5DD, U.K.*

[2] *State Key Laboratory of Metastable Materials Science and Technology, Yanshan University, 066004 Qinhuangdao, P.R. China*

* atsufumi.hirohata@york.ac.uk

Ternary Heusler alloys has demonstrated half-metallic ferromagnetic, antiferromagnetic and ferrimagnetic natures as well as topological insulating properties by achieving correct compositions out of over 2,500 combinations [1],[2]. They have been used in magnetoresistive (MR) junctions as well as the other spintronic devices, including spin caloritronics and spin Hall effects. Recently, a topological insulator demonstrated to generate a spin-polarised electrical current with almost 60% conversion ratio [3]. Typically, such a topological insulator has been grown by molecular-beam epitaxy in an ultrahigh vacuum, which is not compatible for possible device applications. In this study, we used conventional sputtering for the deposition of Heusler-alloy film, which has theoretically predicted to become a topological insulator [4].

20-nm-thick polycrystalline Heusler-alloy thin films of CoCrFeAl, CoFeMnSi and FeCrTiAl were deposited on a thermally-oxidised Si substrate using a sputtering system. The base pressure was about $3\times10^{-7}$ Pa and the Ar pressure during the deposition was $7\times10^{-3}$ Pa. The films were typically grown at the rate of 0.06 nm/s. The compositions of the films were measured using energy dispersive X-ray spectroscopy and adjusted by adding supplemental pegs on the stoichiometric targets. The Heusler-alloy films were post-annealed at elevating temperatures between 300 and 500ºC for up to two hours in a tube furnace for their crystallisation. The films were then structurally and magnetically characteristed for their optimisation by X-ray diffraction and vibrating sample magnetometer, respectively. The optimised film capped with a 2-nm-thick Pt layer was patterned into a Hall bar with the wire width of 1 mm for spin Hall measurements with a conventional direct-current four-terminal method with a sensing current of 50 μA.

For the case of CoCrFeAl, the films annealed at 400ºC for two hours show a very subtle principle peak at 44º in their XRD signals and exhibit clear ferromagnetic behaviour in their magnetisation curves. The saturation magnetisation is calculated to be 444 emu/cm$^3$, which is almost 57% of the bulk and the generalised Slater-Pauling curve as previously reported [5]. This may be due to the lower ordering of our films (at least in their *A*2 phase). We anticipate further improvement in the magnetic properties by optimising the film ordering. Such a polycrystalline film can be useful for device implementation.

**Acknowledgements**

This project was partially supported by EPSRC (EP/V007211/1) and JST CREST (No. JPMJCR17J5).

**References**

[1] C. Felser and A. Hirohata (Eds.), “Heusler Alloys,” Springer, Berlin, Germany, Nov. 2015.
[2] K. Elphick *et al.*, *Sci. Technol. Adv. Mater.* **22** (2020) 235.
[3] K. Kondou *et al.*, *Nat Phys.* **12** (2016) 1027.
[4] X. Wang *et al.*, *Phys. Rep.* **888** (2020) 1.
[5] R. Choudhary *et al.*, *AIP Adv.* **6** (2016) 056304.

# Fine-control of magnetic properties in NiMn-based Heusler compounds

*F. Cugini*[1,2], S. Chicco[1], F. Orlandi[3], L. Gallo[1,2], G. Allodi[1], P. Bonfà[1], V. Vezzoni[1], O. Miroshkina[4], M. E. Gruner[4], L. Righi[5], S. Fabbrici[2], R. De Renzi[1], F. Albertini[2], M. Solzi[1,2]*

[1] *SMFI Department, University of Parma, Parco Area delle Scienze 7/A I-43124 Parma, Italy*
[2] *IMEM-CNR Institute, Parco Area delle Scienze 37/A, I-43124 Parma, Italy*
[3] *ISIS Pulsed Neutron Facility, STFC, RAL, Chilton, Didcot, Oxfordshire OX11-0QX, UK*
[4] *Faculty of Physics and CENIDE, University of Duisburg-Essen, Lotharstr. 1, 47048 Duisburg, Germany*
[5] *SCVSA Department, University of Parma, Parco Area delle Scienze 11/A I-43124 Parma, Italy*
*francesco.cugini@unipr.it

The complex interplay between structural, electronic and magnetic degrees of freedom in Mn-based magnetic Heusler compounds gives rise to a great variety of functional properties suitable for technological applications (energy conversion devices, sensors, actuators, spintronic components…). The adaptive lattice allows, by changing composition, the development of different crystallographic and magnetic structures, thus offering the opportunity to tune properties and effects.

In this work, we show how to finely tune the magnetic properties of austenitic Ni-Mn-(In,Sn) Heusler compounds through changes in the composition, element substitution and control of atomic order [1-3]. In particular, we studied the $Ni_{48}Mn_{36}In_{16-x}Sn_x$ series as a test bench to understand the relation between atomic order, magnetic interactions and electronic features in off-stoichiometric NiMn-based Heusler compounds [1,2]. We combine bulk-sensitive magnetometry, neutron diffraction and $^{55}Mn$ hyperfine-field nuclear magnetic resonance (NMR) experiments with first-principles calculations and Monte Carlo simulations to decouple and clarify the different contributions that define the magnetic structure of these compounds. The replacement of In with Sn leads to a decrease of the saturation magnetization, whereas the Curie temperature displays a non-monotonic variation (Fig. 1.a). First-principles calculations demonstrated that the unusual behaviour of the Curie temperature is due to the variation of the electronic band structure induced by the electronic doping of the compound. Whereas, NMR and neutron diffraction experiments revealed that the decrease of saturation magnetization is related to an effective decoupling of the two ferromagnetic sublattices containing Mn. The replacement of In with Sn causes the reduction of the inter-sublattice exchange interaction, resulting in the appearance of two distinct order temperatures for the two Mn sublattices and the complete loss of long-range magnetic order in one sublattice for the Sn end-member composition (Fig. 1.b).

## References

[1] F. Cugini, S. Chicco, F. Orlandi, G. Allodi, P. Bonfá, V. Vezzoni, O. N. Miroshkina, M. E. Gruner, L. Righi, S. Fabbrici, F. Albertini, R. De Renzi, M. Solzi, *Phys. Rev. B* **105** (2022) 174434.
[2] G. Cavazzini, F. Cugini, M. E. Gruner, C. Bennati, L. Righi, S. Fabbrici, F. Albertini, M. Solzi, *Scripta Mater.* **170** (2019) 48.
[3] S. Fabbrici, F. Cugini, F. Orlandi, N. Sarzi Amadè, F. Casoli, D. Calestani, R. Cabassi, G. Cavazzini, L. Righi, M. Solzi, F. Albertini, *J. Alloys Compd.* **899** (2022) 163249.

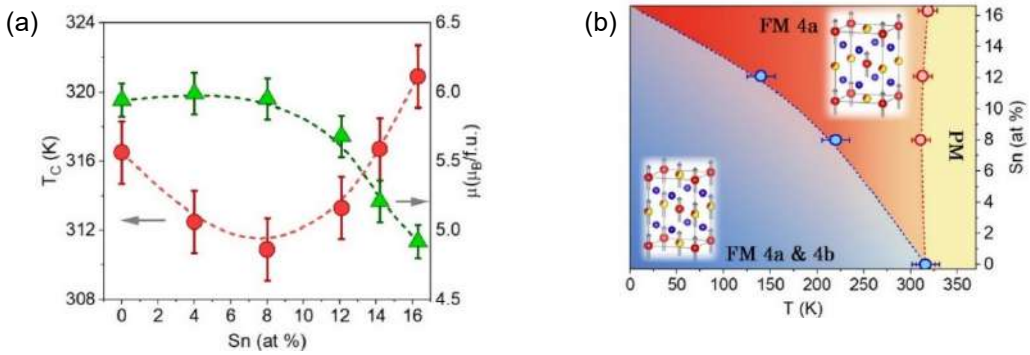



Figure 1: (a) Curie temperature and saturation magnetization of $Ni_{48}Mn_{36}In_{16-x}Sn_x$ compounds as a function of the Sn at.% content. (b) Magnetic phase diagram of $Ni_{48}Mn_{36}In_{16-x}Sn_x$ compounds.

# Ferromagnetic Phases in Sm–Fe–Ti and Sm–(Fe,Co)–Ti Alloy Systems

*A.M. Gabay[1]*, Chaoya Han[2], Chaoying Ni[2], G.C. Hadjipanayis[1,3]*

[1] *University of Delaware, 217 Sharp Lab, Newark, Delaware, USA*
[2] *University of Delaware, 201 DuPont Hall, Newark, Delaware, USA*
[3] *Northeastern University, 360 Huntington Avenue, Boston, Massachusetts, USA*
*gabay@udel.edu

The data on magnetism and equilibria of the Sm–Fe–Ti phases available since the 1990s are insufficient for the ongoing efforts to develop new rare-earth-lean permanent magnets based on the tetragonal $SmFe_{12-x-y}Co_xTi_y$ compound of the $ThMn_{12}$ type structure with $x \leq 2.8$, $0.5 \leq y \leq 1$ [1]. In this work, such data were systematically acquired for Fe-rich Sm–Fe–Ti alloys equilibrated at 1000 and 1100 °C, as well as for Sm–$Fe_{0.8}Co_{0.2}$–Ti alloys equilibrated at 1000, 1100 and 1200 °C. All ferromagnetic phases stable at these temperatures were characterized over an extended Ti range. An equilibrium between a liquid phase and the 1:12 phase, absent at 1000 °C, does exist at 1100 and 1200 °C – with immediate implications for liquid-phase sintering of the new magnets. At 1200 °C, the monoclinic 3:29 phase is replaced with a hexagonal 2:17 phase of the $Th_2Ni_{17}$ type [2], which is Sm-depleted compared to the rhombohedral phase of the same nominal stoichiometry. Replacing 20% of iron with cobalt does not significantly modify the phase equilibria. This replacement, however, dramatically affects the thermomagnetic properties of all ferromagnetic phases (see Figure), not only increasing the values of the Curie temperature, but also changing the sign of the Ti effect from positive (or zero for the 1:12 phase) to negative.

## Acknowledgement

This work was supported by the U.S. Department of Energy Office of Science, Award No. DE SC0022168.

## References

[1] Y.K. Takahashi, H. Sepehri-Amin, T. Ohkubo, *Sci. Technol. Adv. Mater.* 22 (2021) 449.
[2] G.V. Ivanova, G.M. Makarova, Ye.V. Shcherbakova, Ye.V. Belozerov, *J. Alloys Compd.* 224 (1995) 29.
[3] A. Paoluzi, L. Pareti, *J. Magn. Magn. Mater.* 189 (1998) 89.

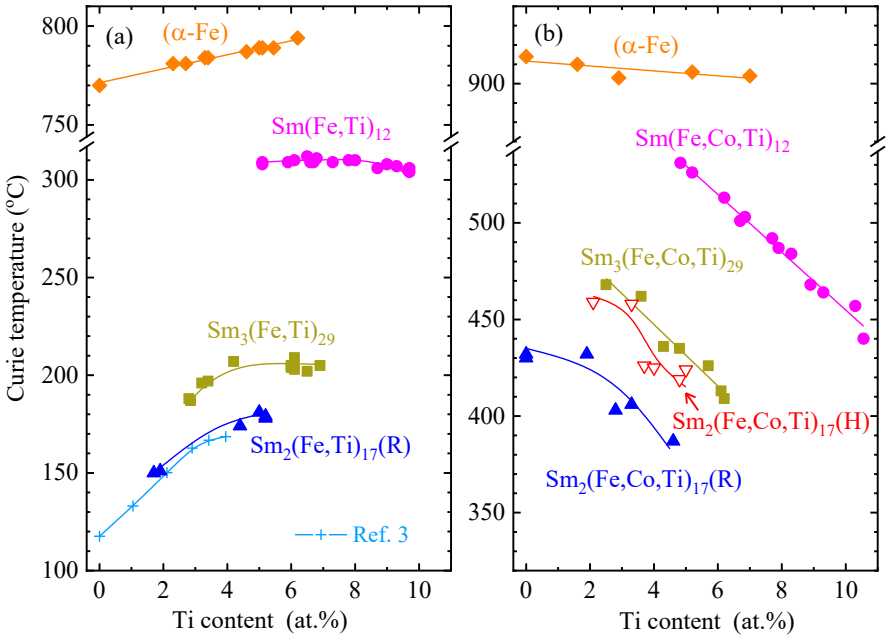



Figure: Effect of phase Ti content on the Curie temperatures of ferromagnetic phases found in (a) Sm–Fe–Ti alloys equilibrated at 1000–1100 °C and (b) Sm–$Fe_{0.8}Co_{0.2}$–Ti alloys equilibrated at 1000–1200 °C. Hexagonal $Sm_2(Fe,Co,Ti)_{17}$(H) phase was found only at 1200 °C, above the thermal decomposition of $Sm_3(Fe,Co,Ti)_{29}$ phase.

# Magnetic-Field-Biased Diffusion and its Connection to Ultra-Hard Ferromagnetic Precipitates in the Ni-Mn-Sn System

*Nicolas Josten[1]*, Steffen Franzka[2], Ziyuan Rao[3], Franziska Scheibel[4], Olga Miroshkina[1], Franziska Staab[5], Aslı Çakır[6], Markus Gruner[1], Mehmet Acet[1], and Michael Farle[1]*

[1] *Faculty of Physics and Center for Nanointegration (CENIDE), University Duisburg Essen, Duisburg, 47057, Germany*

[2] *Interdisciplinary Center for Analytics on the Nanoscale (ICAN), Carl-Benz-Straße 199, 47057 Duisburg, Germany*

[3] *Max-Planck-Institut für Eisenforschung, Max-Planck-Strasse 1, Düsseldorf 40237, Germany*

[4] *Functional Materials, Institute of Materials Science, Technical University of Darmstadt, Alarich-Weiss-Str. 16, 64287 Darmstadt, Germany*

[5] *Physical Metallurgy, Institute of Materials Science, Technical University of Darmstadt, Alarich-Weiss-Str. 2, 64287 Darmstadt, Germany*

[6] *Department of Metallurgical and Materials Engineering, Mugla University, 48000 Mugla, Turkey*

*Nicolas.Josten@uni-due.de

The $Ni_{50}Mn_{50-x}Sn_x$ ($0 < x < 25$) system is metastable and decomposes into ferromagnetic full-Heusler $Ni_2MnSn$ and antiferromagnetic tetragonal NiMn [1]. If this decomposition occurs in a magnetic field, it leads to ultra-hard ferromagnetic precipitates inside an antiferromagnetic matrix with coercive fields of 5T [2]. We performed structural and magnetic investigations on these precipitates using standard magnetometry, switching magnetization MFM (SM-MFM), and atom probe tomography (APT). We also investigated the matrix material NiMn and the closely related PdMn, in which an excess of about 2% of Ni or Pd leads to the emergence of strongly pinned magnetic moments after magnetic annealing.

**Acknowledgements**

We acknowledge funding by the Deutsche Forschungsgemeinschaft CRC/TRR 270 (Project-No. 405553726).

**References**

[1] W. Yuhasz, et al. Journal of alloys and compounds 492, 681 (2010)
[2] Scheibel, F., et al. Applied Physics Letters 110 (19), 192406 (2017)

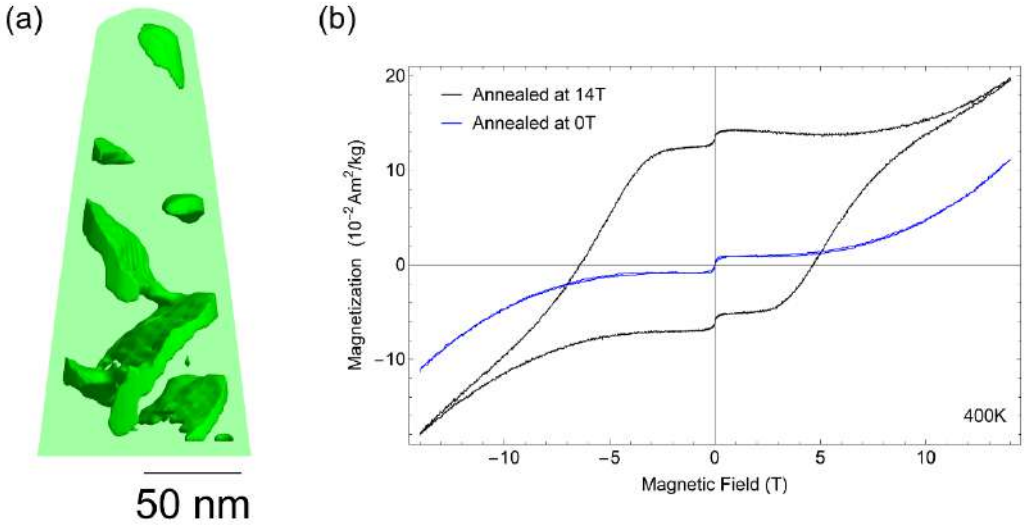



Figure: (a) Ferromagnetic precipitates with a laminar structure in annealed $Ni_{51}Mn_{44}Sn_{05}$ measured with APT. (b) Magnetic field dependent magnetization measurements of $Ni_{51}Mn_{44}Sn_{05}$ at 400K after annealing for 6h at 650K in 14T and 0T.

# Effect of Ga concentration on structural, thermal stability and magnetism of nanocrystalline CoCrFeNiGa$_x$ (x = 0.5 , 1) high entropy alloys

*N. F. Shkodich[1,2]*, M. Spasova[1], Z. Rao[2], M. Farle[1]*

[1] *Faculty of Physics and Center of Nanointegration (CENIDE), University of Duisburg-Essen, Duisburg, 47057 Germany*

[2] *Max-Planck-Institut für Eisenforschung, Max-Planck-Str. 1, 40237, Düsseldorf, Germany*

* natalia.shkodich@uni-due.de

The concept of high-entropy alloys (HEAs) introduces a fundamentally novel strategy for exploring unknown regions in multicomponent phase diagrams and opens ways to new materials [1, 2]. Most of the reported works on HEAs were mainly focused on microstructure, phase formation and mechanical behavior. The magnetic properties, however, have only begun to be investigated. The complexity in the magnetic response arises from the locally varying exchange coupling between the elements of HEAs. Such interactions are influenced by different sintering routes, by the morphology and microstructure in bulk or powder, and are related to their magnetic properties even when the stoichiometries are identical.

Here, we report the successful fabrication of nanocrystalline singe *fcc* phase CoCrFeNiGa$_x$ (x = 0.5, 1.0) particles with good structural and compositional homogeneity by short-term high energy ball milling (HEBM) in planetary ball mills at room temperature. We find an enhanced solid solubility and a homogenous alloy formation compared to other non-equilibrium processes. The XRD, SEM/EDX, and TEM results showed that a *fcc* phase with refined microstructure of nanosized grains (~10 nm) could be obtained after 190 min of HEBM. Based on DSC results, the HEA powders have a thermal stability of up to 1273 K despite the low melting point of Ga (302.9K).

Using these HEBM CoCrFeNiGa$_x$ (x = 0.5, 1.0) powders we produced homogeneous nanocrystalline bulk HEAs, which was not possible starting with elemental powders of Co, Cr, Fe, Ni and Ga ingots in a direct Spark plasma sintering (SPS) process. SPS at 1073 K of the CoCrFeNiGa$_{0.5}$ powder increased the crystallinity of the *fcc* phase, while for the equiatomic CoCrFeNiGa powder a partial transformation of *fcc* structure into a *bcc* one was observed. 3D local compositional mapping at atomic resolution by Atom probe tomography (APT) of CoCrFeNiGa$_{0.5}$ HEA showed a truly homogeneous distribution of all elements (Fig. 1).

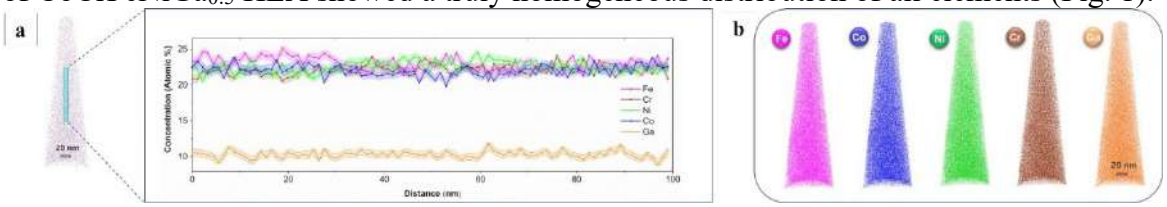



Figure 1: Atom probe tomography of CoCrFeNiGa$_{0.5}$ bulk HEA (a) a tip reconstruction with 1D concentration profiles across the cylinder (b) atom maps of Fe, Co, Cr, Ni and Ga.

The nanocrystalline HEA CoCrFeNiGa$_x$ (x = 0.5, 1.0) powders showed a paramagnetic behavior at room temperature with a Curie temperature ($T_c$) in the range of 130 K – 150 K. The SPS consolidation led to the enhancement of magnetic properties for the equiatomic CoCrFeNiGa HEA, and increased the $T_c$ up to 735-750 K, and saturation magnetization $M_s$ (FM) by 10 times compared to the HEA powder, while the magnetic nature of CoCrFeNiGa$_{0.5}$ bulk HEA alloy remains intact.

## Acknowledgements

This work has been supported by the Deutsche Forschungsgemeinschaft (DFG) within CRC/TRR 270, project S01 (project ID 405553726).

## References

[1] J.-W. Yeh, et al., *Adv. Eng. Mater.* **6** (2004) 299.
[2] B. Cantor, et al., *Mater. Sci. Eng. A* **375-377** (2004) 213.
[3] N.F. Shkodich, M. Spasova, M. Farle, et al., *J Alloys Comp*. **816** (2020) 152611.

# FCC-BCC phase transition and magnetism in $(MnFeCoNi)_{80}Cu_{20-x}Z_x$ (*Z*: Al, Ga) high-entropy alloys

*M. Acet[1]*, Y. Orbay[2], Z. Rao[3], A. Çakır[2], T. Tavşanoğlu[2], M. Farle[1]*

[1]*Faculty of Physics and Center for Nanointegration (CENIDE), Universität Duisburg-Essen, D-47048 Duisburg, Germany*

[2] *Department of Metallurgical and Materials Engineering, Muğla Sıtkı Koçman University, 48000 Muğla, Turkey*

[3]*Max-Planck-Institut für Eisenforschung GmbH, Düsseldorf, Germany*

*mehmet.acet@uni-due.de

We study the occurrence of FCC and BCC structural phases and the magnetic and mechanical properties of the six-component high-entropy alloy series $(MnFeCoNi)_{80}Cu_{20-x}Z_x$ (*Z*: Al, Ga). We find that the equiatomic five-component alloy MnFeCoNiCu is FCC and ferromagnetic with a Curie temperature lying close to room temperature. By partially replacing Cu with Al or Ga, the BCC structure emerges with 10 at% Al and 15 at% Ga. This brings along additional ferromagnetic interactions and stabilizes ferromagnetism in both phases leading to mixed ferromagnetic anisotropies and magnetic hardening. We estimate the magnetic moments in each phase by considering the phase fractions obtained from x-ray diffraction refinements and discuss the magnetic interactions in the multiphase alloys. The magnetic moments of these high-entropy alloys in the individual phases obey the Slater-Pauling rule in a region covering the valence electron concentration dependence of the magnetic moment of Heusler alloys.

**Acknowledgements**

Work supported by Deutsche Forschungsgemeinschaft (Project 405553726-CRC/TRR 270) and TUBITAK Science Fellowships and Grant Programs (Grant 1059B192000916).

# Controlling Magnetic Phase Coexistence in Spatially Confined FeRh

*Jon Ander Arregi[1,*], Lucie Motyčková[1], Friederike Ringe[1], Olena Gomonay[2], Vojtěch Uhlíř[1,3]*

[1] *CEITEC Brno University of Technology, Purkyňova 123, 612 00 Brno, Czechia*
[2] *Institute of Physics, Johannes Gutenberg-University, Staudingerweg 7, 55099 Mainz, Germany*
[3] *Institute of Physical Engineering, Brno University of Technology, Technická 2, Brno, 616 69, Czechia*
*ja.arregi@ceitec.vutbr.cz

The close-to-room-temperature phase transition from antiferromagnetic (AF) to ferromagnetic (FM) order in the FeRh alloy attracts a large interest for investigating the physics of coupled order parameters, as well as for its potential applications in magnetic recording, spintronics, and magnetocaloric cooling [1]. Experiments in epitaxial films confirmed the heterogeneous character of the transition, with typical AF/FM domain sizes of ~500 nm and below during phase coexistence [2].

Here, we explore the nature of phase coexistence in spatially confined FeRh systems, ranging from thin films to patterned and self-assembled nanostructures. We find that its presence and character are susceptible to parameters such as nucleations sites, lateral confinement, morphology, applied magnetic field, and strain.

We firstly visualize phase separation in relatively thick (200 nm) epitaxial FeRh films using the reflectivity contrast in wide-field optical microscopy, where we find considerably large, micron-sized phase domains. Their size and shape are primarily influenced by elastic stresses in the film, resulting in preferential domain growth along the principal crystal axes of FeRh. The application of a sufficiently high magnetic field enables selecting the growth direction of the emerging phase domain during both heating and cooling (see Fig. 1a), where magnetic dipolar interactions in the FM phase fraction partly override the influence of elastic stress [3].

Furthermore, we investigate the phase transition properties of self-assembled FeRh nanostructures fabricated via solid-state dewetting, reaching lateral sizes of ~200 nm (see Fig 1b). While AF/FM phase separation in sub-micron patterned FeRh wires is supressed during cooling [4], we find that phase coexistence in self-assembled nanostructures disappears for both heating and cooling cycles, recovering the abrupt character of the transition [5]. We additionally discuss aspects such as the effect of nanostructure morphology, and the observation of very pronounced supercooling, reaching 150 K.

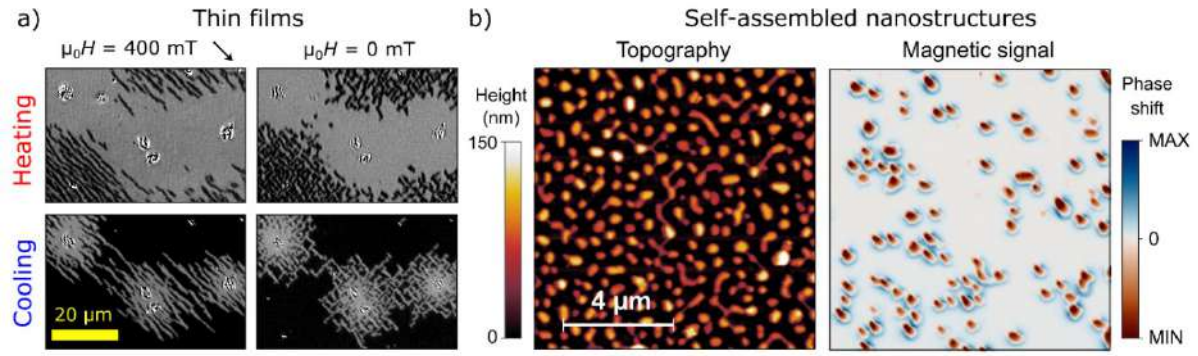



Figure 1: (a) Optical microscopy images of a 200-nm-thick FeRh film at the phase-separated state during heating and cooling for different values of the applied magnetic field. Dark/light regions indicate FM/AF domains. (b) Room-temperature topography and magnetic force micrographs of self-assembled FeRh nanoislands on a MgO substrate.

## Acknowledgements

We acknowledge CzechNanoLab Research Infrastructure supported by MEYS CR (LM2018110).

## References

[1] L. H. Lewis, C. H. Marrows, S. Langridge, *J. Phys. D: Appl. Phys.* **49** (2016) 323002.
[2] C. Baldasseroni, *et al.*, *Appl. Phys. Lett.* **100** (2012) 262401.
[3] J. A. Arregi, *et al.*, *J. Phys. Mater.,* submitted for publication.
[4] V. Uhlíř, J. A. Arregi, E. E. Fullerton, *Nat. Commun.* **7** (2016) 13113.
[5] L. Motyčková, J. A. Arregi, *et al.*, *ACS Appl. Mater. Interfaces* **15** (2023) 8653–8665.

# Tuning $L1_0$ Order and Magnetocrystalline Anisotropy in Rare-Earth-Free Transition Metal Magnets: an Integrated, First Principles Approach

*Julie B. Staunton*[1], *Christopher D. Woodgate*[1]*, *L. H. Lewis*[2,3]

[1] *Department of Physics, University of Warwick, Coventry, CV4 7AL, UK*
[2] *Department of Chemical Engineering, Northeastern University, Boston, MA 02115, USA*
[3] *Department of Mechanical and Industrial Engineering, Northeastern University, Boston, MA 02115, USA*
*C.Woodgate@warwick.ac.uk

We report results from an holistic approach for modelling both atomic ordering and the subsequent magnetocrystalline anisotropy energy (MAE) of magnetic materials with application to the design of novel, rare-earth-free permanent magnets. This computationally efficient technique allows for fast exploration of the materials design space and could open up a new route for materials discovery. In the present work, we study the class of magnetic materials which chemically order into the $L1_0$ structure. It is known that such materials often have large MAE values, with prime examples being FePt and CoPt. However, materials such as these use component elements which have a high criticality. Ab initio theory has previously confirmed that it is the tetragonal, $L1_0$ order that produces the high MAE values measured in these materials[1]. There is therefore a desire to discover new $L1_0$ materials which are made using more abundant elements, but which still retain desirable magnetic properties. One such candidate material is $L1_0$-FeNi, found in meteoritic, tetrataenite samples. This material is known to have a high uniaxial anisotropy, but a low chemical ordering temperature and sluggish kinetics make it challenging to manufacture in a laboratory setting.

Here, we consider introducing a third element into the Fe-Ni system at a low concentration to promote ordering tendencies and enhance its predicted MAE, studying systems with the general formula $Fe_{50-x}Ni_{50-y}X_{x+y}$ for a variety of additives, examples including X=Pd, Co, Pt, Al. Crucially, our modelling enables us to predict the nature of any chemical order[2] and then go on to predict the MAE for a given system[1], using the same ab initio formalism for both aspects of the modelling approach. The ordering behaviour predicted on adding these dopants is rich and a variety of chemical orderings are obtained. Interestingly, we find that it is often the addition of light elements such as Al which enhance the MAE the most. We are also able to study the impact of magnetic order on predicted atomic order and show that annealing samples in an applied magnetic field may enhance chemical ordering temperatures by altering the magnetic state of a material[3].

**Acknowledgements**

We gratefully acknowledge the support of the UK EPSRC, Grant No. EP/W021331/1. This work was also supported by the U.S. Department of Energy, Office of Basic Energy Sciences under Award Number DE SC0022168 and by the U.S. National Science Foundation under Award ID 2118164.

**References**

[1] J.B. Staunton et al., Phys. Rev. B 74, 144411 (2006).
[2] C. D. Woodgate and J. B. Staunton, Phys. Rev. B **105**, 115124 (2022).
[3] C. D. Woodgate D. Hedlund, L. H. Lewis, J. B. Staunton, arXiv:2303.00641 (2023).

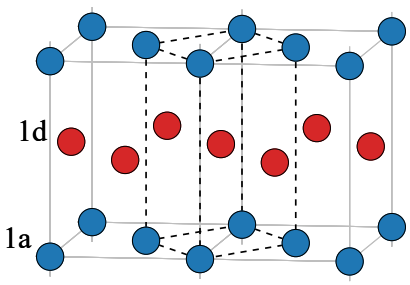



Figure: Visualisation of the $L1_0$ structure, with the body-centred-tetragonal unit cell highlighted by dashed lines.

# Magnetic properties of $(Cr_{1-x}Mn_x)_2GaC$ MAX phase epitaxial films: role of stoichiometry and chemical disorder

*Ivan Tarasov[1]*, Olga Miroshkina[1], Hanna Pazniak[1], Michael Farle[1], Ulf Wiedwald[1]*

[1] *Faculty of Physics and Center for Nanointegration (CENIDE), University of Duisburg-Essen, 47057 Duisburg, Germany*

*ivan.tarasov@uni-due.de

Due to their nanolaminated structure, tunable chemistry, and high oxidation resistance, MAX phases (where M is an early transition metal, A is a main group element, and X is carbon or nitrogen) are interesting materials for a wide variety of applications. The partial substitution of M atoms is one of the ways to tailor their properties to specific applications. In this study, we grow $(Cr_{1-x}Mn_x)_2GaC$ MAX phase films to fine-tune their magnetic response by stoichiometry variations for x = 0-1. High-quality epitaxial $(Cr_{1-x}Mn_x)_2GaC$ MAX phase films (thickness 50 – 70 nm) are synthesized by pulsed laser deposition on rigid MgO(111), Al2O3(0001) and flexible muscovite $KAl_3Si_3O_{10}(OH)_2(001)$ substrates using $(Mn_{0.5}Cr_{0.5})_{66}Ga_{34}$, $Mn_{66}Ga_{34}$, $Cr_{66}Ga_{34}$ and C targets. Structural and morphological characterization reveals a strong competition between the $(Cr_{1-x}Mn_x)_2GaC$ MAX phase and $(Cr_{1-x}Mn_x)_3GaC$, $(Cr_{1-x}Mn_x)_3Ga$ phases. We suppress the formation of side phases by variation of the growth temperature and the growth on seed layers.

The flexibility of muscovite substrates allows tuning the magnetic properties via different external strains applied. The $(Cr_{0.55}Mn_{0.45})_2GaC$ films under strain show a higher remanent and saturation magnetization. A delaminated $(Cr_{0.55}Mn_{0.45})_2GaC(0001)$ film transferred on a tape reveals a complex magnetic behaviour likely caused by a superposition of different local strain configurations of the flexible tape.

Vibrating sample magnetometry of the MAX phase reveals a general trend of increasing magnetization and ordering temperature with increasing Mn content. Nevertheless, for various $(Cr_{1-x}Mn_x)_2GaC$ MAX phase films for given x, the ordering temperature may vary within a temperature range of 150 K. The observed experimental behaviour is explained by preferential local chemical order at M sites for Cr and Mn atoms as derived from first-principles calculations coupled with Monte Carlo simulation within the Heisenberg model.

**Acknowledgements**

Funding by the Deutsche Forschungsgemeinschaft (DFG) within CRC/TRR 270, project B02 (Project-ID 405553726) is gratefully acknowledged. The authors thank Detlef Spoddig for assistance with SEM and EDX imaging. We thank Merlin Schmuck and Doru Lupascu for their assistance and for providing access to the XRD with the Eulerian cradle. We also thank Ulrich von Hörsten for his support in the optimization of the deposition chamber.

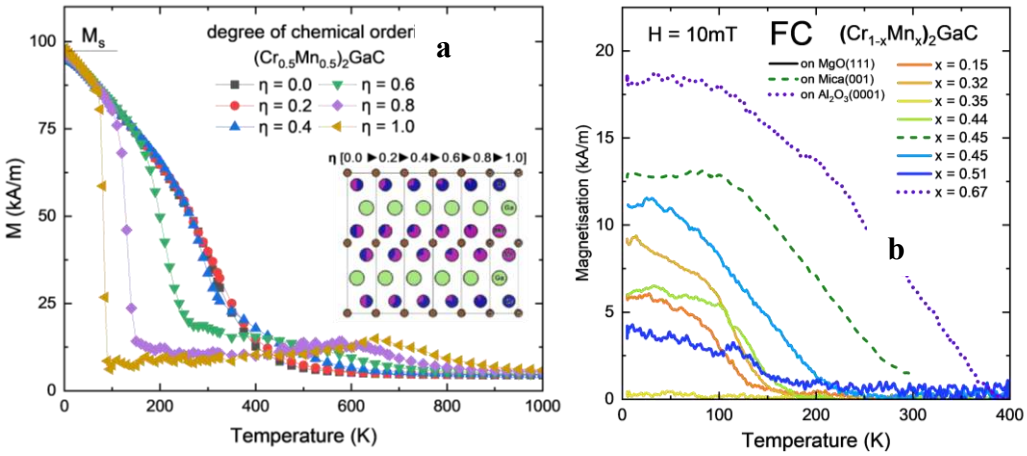



Figure: (a) Magnetization of a $(Cr_{0.5}Mn_{0.5})_2GaC$ MAX phase simulated for various degrees of chemical order at M-site as indicated in the inset. (b) Experimental magnetization curves for a series of thin film samples of $(Cr_{1-x}Mn_x)_2GaC$ MAX phases.

# Structure and magnetic properties of $Ca_3Mn_2O_7$ thin films prepared by pulsed laser deposition

*B. Silva[1]*, J. Oliveira[1], T. Rebelo[1], P. Rocha-Rodrigues[2], N. Lekshmi[2], A. Lopes[2], J. Araújo[2], B. Almeida[1]*

[1]*Center of Physics of the Universities of Minho and Porto (CF-UM-UP), Laboratory for Materials and Emergent Technologies (LAPMET), Dep. de Física, Universidade do Minho, Campus de Gualtar, 4710-057 Braga, Portugal*

[2] *Institute of Nanoscience and Nanotechnology and Photonics, LAPMET, Universidade do Porto, Rua do Campo Alegre, 687, 4169-007, Porto, Portugal*

*brunasilva@fisica.uminho.pt

Naturally Layered Perovskite structures with improper ferroelectricity such as the $Ca_3Mn_2O_7$ allow exploring nonpolar rotations of oxygen octahedra and displacement of cation sites, resulting in noncentrosymmetry. Moreover, their high sensitivity to lattice distortions allows for the manipulation of acentricity through preparation as thin films over crystalline substrates, which enables tuning of lattice, electric, and magnetic interactions[1,2]. As such, five thin films of calcium manganese oxide with variable thicknesses (40 to 300 nm) were prepared by pulsed laser deposition over $SrTiO_3$ substrates[2] and their structural, dielectric and magnetic properties were characterized. The $A2_1am$ ferroelectric phase was observed in the films, along with the orthorhombic Acaa phase. The X-ray diffraction results demonstrate that the thickness of the samples influences the preponderance of each phase. The magnetic properties show the presence of an antiferromagnetic (AFM) transition between 90 and 112 K (Figure). The AFM transition temperature was lower for thinner films and increases as the thickness of the films increases. For the thicker film, with around 300 nm, an AFM transition at 112 K was found. This temperature is characteristic of the $Ca_3Mn_2O_7$ Ruddlesden-Popper phase in bulk. The films show hysteresis loops typical of antiferromagnetic materials. The dielectric properties of the thicker film present a different behavior when changing to the antiferromagnetic region Additionally, for the thicker thin film, the Kohlrausch-Williams-Watts stretched exponential parameter showed an abrupt decrease below ~110 K, near the AFM transition temperature. This indicates the presence of magnetoelectric interactions and magnetically induced enhancement of dipolar-correlations in the AFM phase. In this work, a study of the magnetic properties of the films produced and its correlation with the structure and dielectric properties will be discussed and presented.

## References

[1] P. Ghosez and J.M. Triscone, Nat. Mater. 10, 269 (2011).

[2] B.M. Silva, J. Oliveira, T. Rebelo, V.B. Isfahani, P. Rocha-Rodrigues, N. Lekshmi, J.H. Belo, F.L. Deepak, A.M.L. Lopes, J.P. Araújo, and B.G. Almeida, Mater. Res. Bull. 158, 112066 (2023).

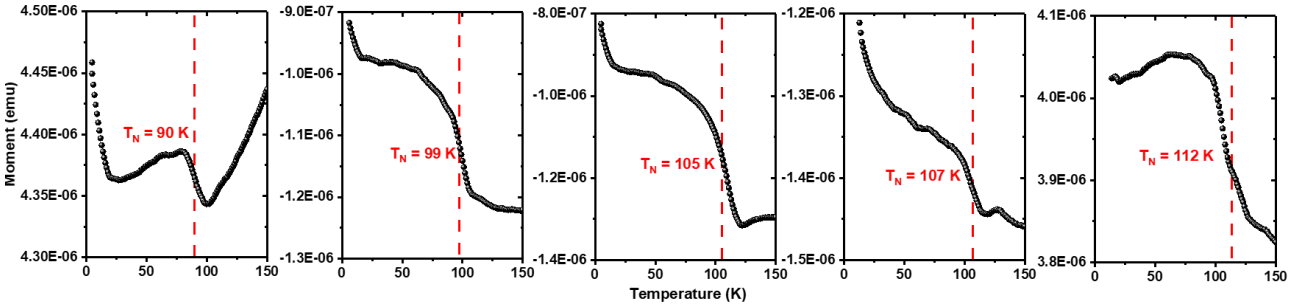



Figure: Temperature dependence of the magnetization measured in $Ca_3Mn_2O_7$ thin films with increasing thickness (from 40 to 300K - left to right in the graph) over $SrTiO_3$ substrates and their corresponding AFM transition temperatures.

# Highly Anisotropic Uniaxial Magnetic and Transport Properties in Thin Films Controlled by Their Nano-sheets Morphology

*C. Favieres[1,2]*, J. Vergara[1,2], C. Magén[3,4,5], M. R. Ibarra[3,4,5], V. Madurga[1]*

[1]*Laboratory of Magnetism. Department of Science. Physics. Public University of Navarre (UPNA). Campus de Arrosadía, E-31006, Pamplona, Spain .*

[2] *Institute for Advanced Materials and Mathematics (INAMAT²), (UPNA) E-31006, Pamplona, Spain*

[3]*Instituto de Nanociencia y de Materiales de Aragón (INMA) CSIC-Universidad de Zaragoza, 50009 Zaragoza, Spain.*

[4] *Departamento de Física de la Materia Condensada, Universidad de Zaragoza, 50009 Zaragoza, Spain.*

[5]*Laboratorio de Microscopías Avanzadas (LMA), Universidad de Zaragoza, 50018 Zaragoza, Spain*

*favieresc@unavarra.es

Ferromagnetic Co-rich thin films were grown by Pulsed Laser Deposition (PLD) with a special nano-morphology: the oblique-angle incidence of the plasma and the particular set-up during deposition allow the generation of films composed of tilted nano-sheets, ≈ 3.0-4.0 nm thick, ≈ 30-100 nm wide, and ≈ 200-300 nm long, with an inter-sheet distance of ≈ 0.9-1.2 nm. This nano-morphology conferred remarkable shape anisotropy to the samples in the direction perpendicular to the plane of incidence of the plasma during PLD, giving rise to large in-plane uniaxial magnetic and transport anisotropies [1-4]. The hysteresis loops showed an easy direction of magnetization in the longitudinal direction of the nano-sheets, with anisotropy fields ≈ 70 kA/m, whereas the hard direction loops were anhysteretic. The resistance of the films behaved also anisotropically: $(R_\perp - R_\parallel)/R_\parallel$ = 33% for the Co–V films and 18% for the Co–Zn samples, with $R_\perp$ being the resistance in the perpendicular direction of the nano-sheets and $R_\parallel$ the resistance in the parallel direction. The changes in nano-morphology caused by thermal treatments, and hence in their anisotropic properties, were studied and the origin of these anisotropic behaviors was established. The Co–V films increased their anisotropic properties after annealing at least up to 500 ºC, while they vanished for annealed Co–Zn films. The dependence of the anisotropic resistance on temperature, transmission electron microscopy, and chemical analysis at the nano-scale, allowed to establish the nature and the activation energy spectra of the atomic relaxation processes during annealing. These spectra were associated to the changes induced in the nano-morphology by thermal treatments.

## References

[1] C. Favieres, J. Vergara, C. Magén, M. R. Ibarra, V. Madurga. *J. Alloy. Compd*. **664** (2016) 695
[2] J. Vergara, C. Favieres, C. Magén, J.M. De Teresa, M. R. Ibarra, V. Madurga. *Materials* **10** (2017) 1390
[3] C. Favieres, J. Vergara, C. Magén, M. R. Ibarra, V. Madurga. *J. Alloy. Compd*. **911** (2022) 164950
[4] C. Favieres, J. Vergara, V. Madurga. *J. Appl. Phys*. (2023) in press

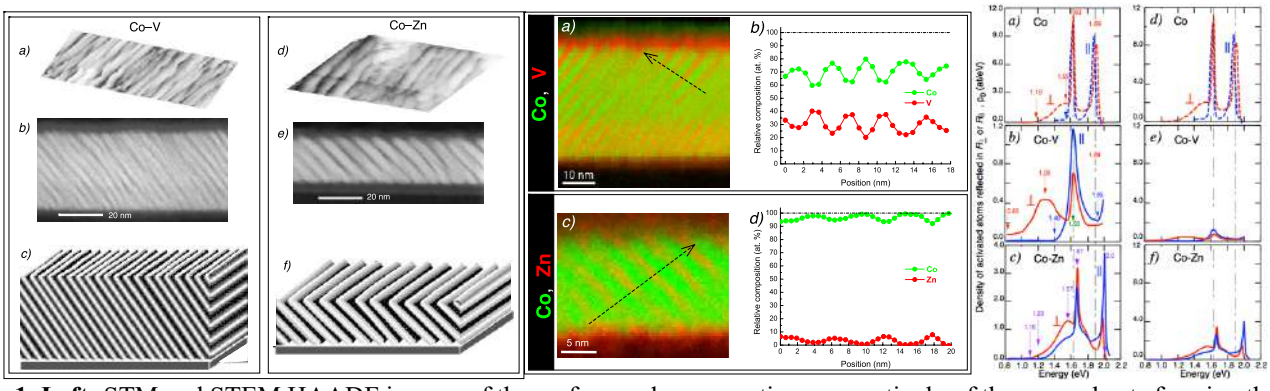



**Figure 1. Left:** STM and STEM HAADF images of the surface and cross sections respectively of the nano-sheets forming the films and schematic 3D diagrams of the nano-sheet morphologies. **Center:** Chemical maps of the cross-sections of the as-deposited films. **Right:** Activation energy spectra of the processes occurring during thermal treatments, which affected the anisotropic resistance [3].

# Observation of the Ideal Triple-Q State in Pd/Mn/Re(0001)

*Felix Nickel[1]*, Soumyajyoti Haldar[1], Roland Wiesendanger[2], Stefan Heinze[1] and Kirsten von Bergmann[2]*

[1] *Institute of Theoretical Physics and Astrophysics, University of Kiel, Leibnizstraße 15, Kiel, Germany*

[2] *Department of Physics, University of Hamburg, Jungiusstraße 11, Hamburg, Germany*

*nickel@physik.uni-kiel.de

The 3Q state – a three-dimensional spin structure on a two-dimensional lattice predicted about 20 years ago [1] – has been observed in a Mn monolayer on Re(0001) using spin-polarized scanning tunneling microscopy (SP-STM) [2]. The 3Q state is a superposition of three symmetry equivalent spin spirals with the same period and can be stabilized by higher-order exchange interactions (HOI) such as the biquadratic or four-spin interactions [1,2]. Recently, it has been suggested based on density functional theory (DFT) calculations that the 3Q state in Mn/Re(0001) is significantly distorted due to topological chiral magnetic interactions [3].

Here, we show using DFT that the competition of biquadratic, four-spin, and topological chiral interactions can be tuned in Mn/Re(0001) by single atomic adlayers of Pd or Rh. Thereby, the frustration of HOI present in Mn/Re(0001) is lifted which results in the occurrence of the ideal 3Q state with tetrahedron angles between adjacent magnetic moments. SP-STM measurements on Pd/Mn/Re(0001) are shown in the figure below. Two Pd/Mn islands on adjacent Re terraces can be seen for which the structure, and thus also the magnetic state, are rotated by 180° with respect to each other. On the left island a stripe pattern is observed in the SP-STM image, while the right island exhibits a hexagonal pattern. SP-STM images of the 3Q state were calculated based on DFT (see figure below) using the spin-polarized generalization of the Tersoff-Hamann model [4]. The magnetic moment directions of Mn (red) and Pd (green) atoms are given in the figure by arrows. With the indicated tip magnetization estimated from the experiment (yellow arrow), these simulations are in excellent agreement with the experimental SP-STM images. Thus the experiments confirm the theoretically predicted ideal 3Q state in Pd/Mn/Re(0001) and SP-STM measurements of Rh/Mn/Re(0001) show a similar contrast.

## References

[1] Ph. Kurz *et al.*, *Phys. Rev. Lett.* **86** (2001) 1106
[2] J. Spethmann *et al.*, *Phys. Rev. Lett.* **124** (2020) 227203
[3] S. Haldar *et al.*, Phys. *Rev. B* **104** (2021) L180404
[4] D. Wortmann *et al.*, *Phys. Rev. Lett.* **86** (2001) 4132

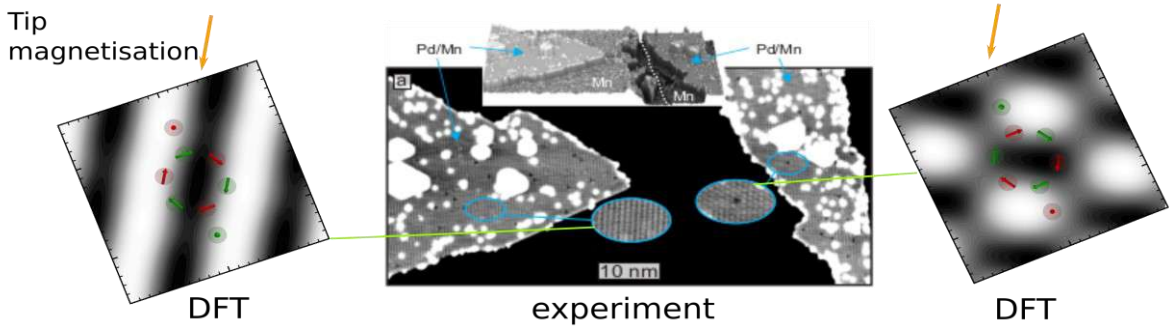



Figure: SP-STM image of Pd/Mn/Re(0001). Two islands can be seen in the experimental image (center). The blue ellipses show a zoom of regions on the islands. SP-STM images calculated for the 3Q state based on DFT are shown for comparison to the left and right. The yellow arrows indicate the tip magnetization used for these calculations.

# Skyrmions and Antiskyrmions in ferrimagnetic multilayers

*Sabri Koraltan[1]*, Michael Heigl[2], Florian Bruckner[1], Claas Abert[1], Manfred Albrecht[2] , and Dieter Suess [1]*

[1] *Faculty of Physics, University of Vienna, Vienna, Austria*

[2] *Institute of Physics, University of Augsburg, Augsburg, Germany*

*sabri.koraltan@univie.ac.at

Skyrmions and antiskyrmions are topologically protected spin structures with opposite vorticities [1]. While the former have been extensively researched over the past years in a variety of magnetic materials and systems, the latter were exclusive to materials with D2d-symmetry [2]. These magnetic spin textures are promising in realizing future spintronic devices and data storage technologies, Thus, their co-existence may show fascinating new physics and potential for novel spintronic devices.

In a recent publication [3], we have shown that by using ferrimagnetic multilayers we can tune the magnetic properties of the system, such that we can stabilize dipolar skyrmions, type-II bublles, as well as first and second order antiskyrmions. In this conference contribution we aim to explain with our micromagnetic modelling the nucleation, stability, and annihilation of the dipolar-stabilized spin textures. We reveal a phase of magnetic parameters, in which the dipolar antiskyrmions can be stabilized, which agrees well with our experimental findings, where the spin objects are images using Lorentz Transmission Electron Microscopy (LTEM). Furthermore, we develop a novel approach where we combine magnetic alyers of different magnetic parameters to enrich the antiskyrmion phase by tuning the stripe domained region.

Since the skyrmions and antiskyrmions in our numerical and experimental findings nucleate rather randomly, we show in a second study that skyrmions and antiskyrmions can be nucleated by making use of magnetic meta-materials situated on top of the skyrmionics layers. We show that magnetic vortices in nanodisks and antivortices in a square lattice, can lead to the nucleation of skyrmions and antiskyrmions, respectively. We investigate in great detail, under which circumstances this stabilization mechanism is reliable, and we demonstrate that the geometry of meta material can be tuned to tune the geometrical arrangement of skyrmions and antiskyrmions in the skyrmion layer [4].

**References**D. Metrio, E. Duardo, O. Legario, *Nat. Phys.* **15** (2019) 735. (Times New Roman 10 points, left-justified)

[1] Göbel, B. et al. Phys. Rev. 895, 1 (2021)
[2] Nayak et al. Nature volume 548, 561–566 (2017).
[3] M. Heigl, S.Koraltan et al. Nat. Comm. Vol. 12, 2611 (2021).
[4] S. Koraltan et al. https://arxiv.org/abs/2207.12260 (2022)

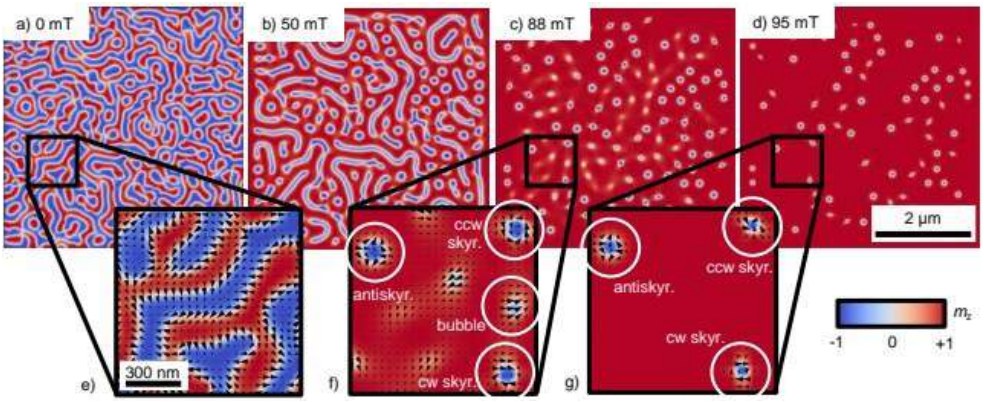



Figure: Magnetization states from the micromagnetic simulation of ferrimagnetic multilayers, where stripe domains are stable at vanishing fields in (a), the stripe start collapse at 50 mT out-of-plane field in (b), the skyrmions, antiskyrmions and type-II bubbles co-exist at 88mT in (c), and only skyrmions and antiskyrmions remain at 95 mT in (d).

# Helical magnetic structure in nanolaminated $Mn_2GaC$ film (MAX phase compound )

J. Dey[1], R. Kalvig[1], E. Jędryka[1], M. Wójcik[1], U. Wiedwald[2], M. Farle[2], and J. Rosen[3]
[1]Institute of Physics, Polish Academy of Sciences, Al. Lotników 32/46, 02-668 Warszawa, Poland
[2]Faculty of Physics and Center for Nanointegration (CENIDE), University of Duisburg-Essen, 47057, Duisburg, Germany
[3]Thin Film Physics, Department of Physics, Chemistry and Biology (IFM), Linköping University, SE-581 83, Linköping, Sweden

$Mn_2GaC$ heteroepitaxial films belong to the MAX phase family of compounds, described as $M_{n+1}AX_n$, where M is a transition metal, A stands for an A-group element (mostly group 13 and 14), X denotes carbon or nitrogen and n can take the numbers 1, 2 or 3. This rich group of materials has recenty attracted a lot of research interest following the reports on their outstanding ceramic and metallic properties [1]. As an important extenstion of their functionalities, some compositions displaying magnetic interactions have been successfully synthesized [2]. Among those, the ternary compound $Mn_2GaC$ crystallizing in the hexagonal structure is the only MAX phase to contain a single M element, facilitating the investigation of its complex, temperature-dependent magnetic structure.

The nanolaminated structure of $Mn_2GaC$ is formed by stacks of ferromagnetically coupled Mn-C-Mn trilayers interleaved with non-magnetic Ga layers repetitively along the hexagonal c-axis. The theoretical studies on $Mn_2GaC$ indicate that the strongly coupled Mn-C-Mn trilayer can be regarded as a single "supermoment", whereas the competing ferromagnetic (FM) and antiferromagnetic (AFM) interactions across the Ga layer result in a complex, temperature-depended magnetic structure, the nature of which has not been fully understood. To resolve some ambiguities resulting from the contradictory results reported in literature [3], we have undertaken a Nuclear Magnetic Resonance (NMR) study on a 100 nm film of $Mn_2GaC$ (0001) | MgO (111) .

The NMR experiments have carried out at 4.2 K in zero-field (ZF) as well as in the external magnetic field ($B_{ext}$) applied in the film plane. The $^{69,71}$Ga ZF NMR data yield large effective field at Ga ($\approx 15.75\ T$), which is due to the uncompensated magnetic moments from the nearest Mn neighbors. This result reveals a non-collinear magnetic structure in $Mn_2GaC$. The ZF $^{55}$Mn NMR indicates the presence of magnetically non-equivalent Mn-positions, where the average magnetic moment of Mn is found to be $\cong 2\,\mu_B/$ Mn-atom. Furthermore the in-field NMR experiments unveil the continuous distribution of orientations of magnetic moments from 0° to 180°. This distribution represents the helical magnetic structure extending along the c-axis (i.e. normal to the film plane) and consisiting of Mn supermoments lying in hexagonal base plane. The calculated pitch of the helix is incommensurate with the crystal lattice period. The helical structure deduced from the analysis of the NMR data can be explained by the presence of competing ferro-antiferro exchange interaction between supermoment layers.

## Acknowledgements

This work was supported by the National Science Center, Poland (grant number UMO-2019/35/B/ST3/03676).

## References

[1] A S Ingason, *J. Phys.: Condens. Matter* **28** (2016) 433003
[2] A.S. Ingason, *Materials Research Letters*, 2:2, (2014) 89-93
[3] M. Dahlqvist, *Phys. Rev. B 93*, (2016) 014410

# Properties of (Ga,Mn)N Insulating Ferromagnet Studied with Ferromagnetic Resonance

*Y.K. Edathumkandy[1], K. Gas[1], D. Sztenkiel[1], K. Das[1], D. Hommel[2,3], H. Przybylińska[1], M. Sawicki[1]*

[1] *Institute of Physics, Polish Academy of Sciences, Warsaw, Poland*
[2] *Lukasiewicz Research Network–PORT, Polish Center for Technology Development, Wrocław, Poland*
[3] *Institute of Low Temperature and Structure Research, PAS, Wrocław, Poland*

Dilute ferromagnetic semiconductors, in particular (Ga,Mn)N predicted to have an exceptionally high Curie temperature ($T_C$), have attained great research importance due to their unique ability to combine the properties of semiconductors and magnetic materials [1]. Moreover, GaN being a wide band gap semiconductor has been dominating the photonics [2] and high power electronics. So it is important to make an effort to understand the underling magnetic properties of (Ga,Mn)N.

We report ferromagnetic resonance (FMR) studies of a series of (Ga,Mn)N layers grown by molecular beam epitaxy [3,4]. All investigated samples showed ferromagnetic signatures, as evidenced by SQUID magnetometry, with $T_C$ ranging from 3 to 12 K. A broad angularly dependent FMR signal appears only at higher temperatures, closer to and above $T_C$, with intensities roughly scaling with magnetic susceptibility of the material, as shown in Fig, 1. However, apart from a very weak paramagnetic signal of $Mn^{2+}$, no ferromagnetic resonance is observed below 7 K, *i.e.*, where such ferromagnetic features as the hysteresis of magnetization curves and the remnant moment are the strongest. We relate this counterintuitive lack of low temperature FMR signal to inhomogeneous broadening caused by non-uniform distribution of magnetic ions and thus inhomogeneities in coupling strengths influencing the local magnetic anisotropies of $Mn^{3+}$ ions.

This study has been supported by the National Science Centre (Poland) through OPUS (UMO - 2018/31/B/ST3/03438) project.

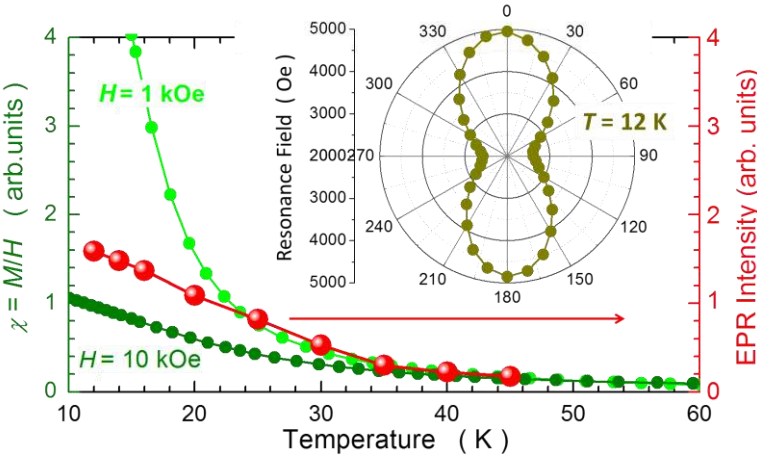



Fig.1 Comparison of magnetic susceptibilities determined by SQUID at fields of 1 and 10 kOe with that determined from FMR signal intensity at fields about 2 kOe. The inset shows the angular dependence of the resonance fields at 12 K

**References:**

[1] T. Dietl, H. Ohno, Rev. Mod. Phys. **86**, 1 (2000).
[2] S. Nakamura, T, Mukai, M. Senoh, App. Phy. Lett., **64**, 13 (1994).
[3] G. Kunert et al., Appl. Phys. Lett. **100**, 155321 (2012).
[4] K. Gas *et al.* J. Alloys Compd. **747**, 946 (2018).

# Magnetic, electronic and topological properties of HgTe and CdTe doped with V and Cr

*Giuseppe Cuono[1]*, Carmine Autieri[1], Tomasz Dietl[1,2]*

[1] *International Research Centre MagTop, Institute of Physics, Polish Academy of Sciences, Aleja Lotników 32/46, PL-02668 Warsaw, Poland*

[2] *WPI-Advanced Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan*

*gcuono@magtop.ifpan.edu.pl

Dilute magnetic semiconductors have played a central role in the demonstrating and describing a strong and intricate influence of the sp-d exchange interactions upon effective mass states in semiconductors, paving the way for the rise of dilute ferromagnetic semiconductors [1] and magnetic topological insulators [2,3,4]. In recent works, the exchange splittings of magneto-optical spectra in $Cd_{1-x}Mn_xTe$ and $Hg_{1-x}Mn_xTe$ have been analyzed [5], and it has been shown that superexchange dominates in magnetic topological insulators [6].
Here we analyze the interplay between magnetism and topology in the HgTe and CdTe doped with Cr and V by using density functional theory with functionals beyond the standard ones to correctly reproduce the topology and the band gap of these systems.
We describe the electronic features of these systems, the spin splitting, the topology and the exchange couplings.
We obtain that the coupling is ferromagnetic in the case of doping with V, unlike in the case of doping with Mn and Cr, where we find antiferromagnetic couplings. It is possible to get the quantum anomalous Hall phase in case of ferromagnetic topological insulators. Based on our results, the quantum anomalous Hall effect could be realized in HgTe doped with V.
It is essential to consider the distortions produced by Jahn-Teller effect, especially in the case of doping with Cr. We find that the distorted structure favors a ferromagnetic ground state.

## References

[1] T. Dietl and H. Ohno, *Rev. Mod. Phys.* **86**, 187 (2014).
[2] H. Ke, Y. Wang, and Q.-K. Xue, *Annu. Rev. Condens. Matter Phys.* **9**, 329 (2018).
[3] Y. Tokura, K. Yasuda, and A. Tsukazaki, *Nat. Rev. Phys.* **1**, 126 (2019).
[4] R. Islam, S. Mardanya, A. Lau, G. Cuono, T.-R. Chang, B. Singh, C. M. Canali, T. Dietl, and C. Autieri, *Phys. Rev. B* **107**, 125102 (2023).
[5] C. Autieri, C. Sliwa, R. Islam, G. Cuono and T. Dietl, *Phys. Rev. B* **103**, 115209 (2021).
[6] C. Sliwa, C. Autieri, J. A. Majewski, and T. Dietl, *Phys. Rev. B* **104**, L220404 (2021).

# Magnetic interactions and high-field magnetotransport properties of $Ge_{1-x-y}Sn_xMn_yTe$ multiferroic layers

*A. Khaliq[1]*, P. Dziawa[2], B. C. Camargo[2], R. Minikaev[1], S. Zakar[1], A. Reszka[1], M. Arciszewska[1], A. Avdonin[1], J. Szczytko[2] and L. Kilanski[1]*

[1] *Institute of Physics, Polish Academy of Sciences, Aleja Lotnikow 32/46, PL-02668 Warsaw, Poland*

[2] *Faculty of Physics, University of Warsaw,Ul. Pasteura 5, PL-02-093 Warsaw, Poland*

*abdul.khaliq@ifpan.edu.pl

Multiferroics offer exciting features in terms of coupling between ferroelectric and ferromagnetic orderings. In this work, we present the structural, magnetic and magnetotransport results of $Ge_{1-x-y}Sn_xMn_yTe$ epitaxial multiferroics with $x$ = 0.03 – 0.14 and $y$ = 0.11 – 0.35 over a broad range of temperature. The samples were grown via molecular beam epitaxy with thickness, $t$ ~ 150 nm. From structural point of view, the layers maintain the rhombohedral crystal symmetry inherited from its host lattice, GeTe which is ferroelectric below $T$ ~ 720 K. The ferroelectric polarization is well-maintained in doped layers even up to $x$ ~ 2 and $y$ ~ 0.15. Furthermore, for $x$ = 0.03 and $y$ = 0.11, the dc susceptibility, $\chi(T)$ manifests double-maxima in the zero-field-cooled curves which might represent paramagnetic to ferromagnetic ($T$ = 55 K) and ferromagnetic to an anticipated re-entrant spin-glass ($T$ = 25 K) phase transition. From high magnetic field results, a negative to positive crossover in magnetoresistance is observed at $T$ = 20 K with a weak (anti)-localization effect below about 50 K that vansihes at higher temperatures for the same sample. Additionally, we also present a modified scaling mechansim of anomalous Hall resistivity which displays a large magnitude, $\rho_{AH}$ = 2.6 – 3 μΩcm at $T$ = 4.3 K in the above mentioned doping range. The origin of scattering processes that induce such a large anomalous Hall resistivity and weak (anti)-localization effect is discussed.

**The research was financed by the National Science Centre, Poland under the project number 2018/30/E/ST3/00309.**

# Magnetization Reversal in FePt Thin Films: Experiments and Simulations

*Augusto Román[1], Agustín Lopez Pedroso[1], Karim Bouzehouane[2], Javier E. Gómez [3,4], Alejandro Butera [3,4], Myriam H. Aguirre[5,6,7], Marcio Soares Medeiros[8,9], Carlos Garcia [10] and Laura B. Steren[1]*

[1]Instituto de Nanociencia y Nanotecnología CNEA-CONICET, Nodo Constituyentes, Buenos Aires, Argentina
[2]Unité Mixte de Physique CNRS, Thales, Universite Paris-Saclay, Palaiseau, France
[3]Instituto de Nanociencia y Nanotecnología CNEA - CONICET, Nodo Bariloche, Río Negro, Argentina
[5] Insituto de Nanociencia y Materiales de Aragón, UNIZAR-CSIC, Zaragoza, Spain
[6]Departamento de Física de la Materia Condensada, Universidad de Zaragoza, Zaragoza, Spain
[7]Laboratorio de Microscopías Avanzadas, Universidad de Zaragoza, Zaragoza, Spain
[8]Laboratório Nacional de Luz Síncrotron, CNPEM, Campinas, São Paulo, Brazil
[9]Departamento de Física, Universidade Federal da Paraíba, 58051-900 João Pessoa, Brazil
[10] CCTVal - Departamento de Física, Universidad Técnica Federico Santa María, Valparaíso, Chile
*augusto.jre@gmail.com

In ferromagnetic thin films with moderate perpendicular magnetic anisotropy (PMA), the magnetic domain configuration transitions from planar to stripe-like domains above a critical thickness ($t_c$) due to the competition between shape and perpendicular magnetic anisotropies [1]. Thin films with striped domains present unusual magnetic behaviors such as rotatable anisotropy[1] and abnormal temperature dependence of the coercive field[2]. The analysis of the magnetization reversal mechanism is essential to the understanding of these unusual behaviors.

FePt films deposited at room temperature present a moderate PMA and, consequently, a transition in the domain configuration at $t_c$=30 nm [2]. In this work, we present a detailed research of the magnetization switching mechanisms in FePt thin films through micromagnetic simulations and experiments.

For the study, a series of thin films were grown by sputtering on silicon substrates with thickness varying between 10nm and 60nm in order to compare their magnetic properties below and over the critical value. The magnetic characterization of the samples was made through magnetometry measurements and magnetic force microscopy. Synchrotron x-ray diffraction and transmission electron microscopy experiments were used to analyze the effect of the strains induced by the substrate in the films structure. Finally, we complemented the analysis with micromagnetic simulations using Mumax3 [3].

We found that below $t_c$ the reversal mechanism is well described by a two-phase model that combines coherent rotation with domain wall movement. For films with $t > t_c$ we observed that the magnetization within each stripe reverses by coherent rotation. Moreover, running simulations of the temporal evolution of the magnetization for different values of KPMA (Figure 1), allowed us to explain the origin of the abnormal temperature variation of the coercive field observed in thick films. The effect was associated to a change in the magnetization reversal process dynamic due to the variation of the PMA by substrate-induced strains.

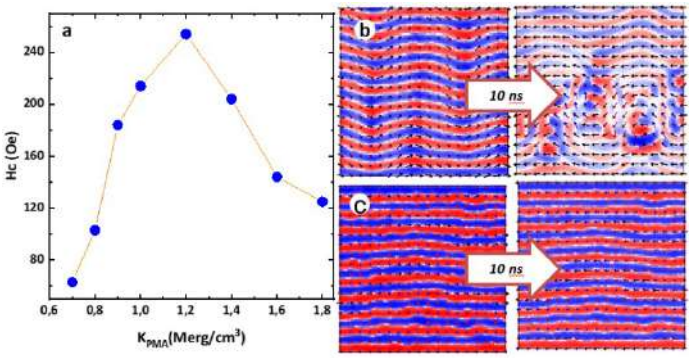



Figure 1: (a) Simulated coercive field vs magnetic anisotropy constant for a 60 nm FePt film. Simulated time evolution of the magnetic domain configuration after the application of $H>H_c$ for (b) $K_{PMA}$=0.9 Merg/cm$^3$ and (c) $K_{PMA}$=1.6 Merg/cm3

## Acknowledgements

Financial support of FONCYT PICT 0867-2016, PICT 02781-2019 and the European Commission through the Horizon H2020 funding by H2020-MSCA-RISE-2016 - Project N$^0$ 734187 –SPICOLOST. Beamtime was granted on the XRD2 beamline by the LNLS.

## References

[1] L.C. Garnier et al. *Journal of Physics: Materials*, **3** (2020) 024001.
[2] J. M. Guzmán et al. *Journal of magnetism and magnetic materials* **347** (2013) 61.
[3] A. Vansteenkiste et al. *AIP Advances* **4**, (2014) 107133.

# Tailoring Perpendicular Magnetic Anisotropy in [$Mn_3G$ / $Co_2YZ$] Heusler Super-Lattices

*C. Guillemard[1,†,*], V. Palin[2,3], C. De Melo[2,‡], P. Gargiani[1], M. Valvidares[1], F. Bertran[3] and S. Andrieu[2]*

[1]*ALBA Synchrotron Light Source, Cerdanyola del Vallès, Spain*
[2]*Institut Jean Lamour, CNRS - Université de Lorraine, Nancy, France*
[3]*SOLEIL synchrotron source, Saint-Aubin, France*
[†]*New affiliation: CINaM, CNRS - Aix Marseille Université, Marseille, France*
[‡]*New affiliation: ICMN, CNRS - Université d'Orléans, Orléans, France*
*charles.guillemard@cnrs.fr

The discovery and engineering of new materials that fully satisfy the requirements for low energy consumption devices is a continuous challenge in spin electronics. Besides the need of a high spin polarization at the Fermi energy and a low Gilbert damping coefficient, obtaining the magnetization perpendicular to the films' plane is one of the most important conditions to improve the efficiency of next generation devices. Perpendicular magnetic anisotropy increases the efficiency of the magnetization reversal by spin transfer torque [1], and opens the path to exploit new exciting and less energy consuming phenomena such as spin-orbit torque switching [2].

On the one hand, ferrimagnetic tetragonal $Mn_3G$ (G=Ga, Ge) Heusler compounds have attracted much attention in the last decade due to their strong magneto-crystalline anisotropy along their c-axis and small overall magnetic moments [3]. On the other hand, it has recently been demonstrated that epitaxial $Co_2MnZ$ (Z=Al, Si, Ga, Ge, Sn) Heusler compounds are half metal magnets with a spin gap in their electronic structure [4] and the lowest Gilbert damping values reported for conductive layers ($< 5 \times 10^{-4}$) [4]. For these reasons, Super-Lattices (SL) composed of those two types of materials represent excellent systems to combine small magnetic moments, full spin polarizations, reduced Gilbert damping values and perpendicular magnetic anisotropy, thus offering a new pathway for extremely efficient and stable spin torques devices.

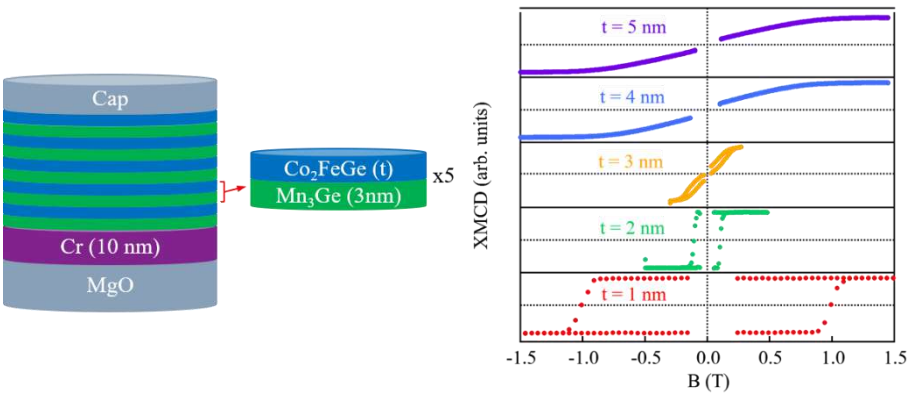



Figure: a) sketch of $[(Mn_3Ge)_{3nm}/(Co_2FeGe)_t]_5$ SL stack b) normal incidence hysteresis loops of the SLs with different $Co_2FeGe$ thicknesses measured by X-ray Magnetic Circular Dichroism (XMCD) at Fe $L_{2,3}$ edge.

This has motivated the investigation of $[(Mn_3G)/(Co_2YZ)]_n$ SL which we will present. Samples were grown by molecular beam epitaxy at the Institut Jean Lamour (France) (figure a), and present an excellent structural quality from X-ray diffraction and transmission electron microscopy characterization. The magnetic properties were investigated using magneto-optical Kerr effect and magnetometry in combination with element specific X-ray magnetic circular dichroism measurements (BOREAS beamline, ALBA synchrotron). Perpendicular magnetic anisotropy is achieved, as presented in the figure b. Moreover, we observed a strong parallel magnetic coupling between the sublayers and, remarkably, the possibility of tuning magnetic properties like the anisotropy, coercive fields or total magnetic moments by adjusting the sublayers thicknesses.

**References**

[1] S. Mangin *et al., Nat. Mater.* **5**, 210 (2006).
[2] I. M. Miron *et al., Nature* **476**, 189 (2011).
[3] B. Balke *et al., Appl. Phys. Lett.* **90**, 152504 (2007).
[4] C. Guillemard *et al., Phys. Rev. Appl.* **11**, 064009 (2019).

# Magneto-Ionic Control of Coercivity and Domain-Wall Velocity in Co/Pd-Multilayers

*Madeleine Bischoff*[1], *Olav Hellwig*[2,3], *Karin Leistner*[1], *Markus Gößler*[1*]

[1] *Institute of Chemistry, Chemnitz University of Technology, Chemnitz, Germany*

[2] *Institute of Physics, Chemnitz University of Technology, Chemnitz, Germany*

[3] *Institute of Ion Beam Physics and Materials Research, Helmholtz-Zentrum Dresden-Rossendorf, Dresden, Germany*

*markus.goessler@chemie.tu-chemnitz.de

Magnetic multilayers exhibiting strong perpendicular magnetic anisotropy (PMA) are an important material class for spintronic applications, where spin-polarized currents are used to change magnetic properties. An alternative, low-power approach to tuneable magnetic properties is offered by magneto-ionics, which is based on voltage-controllable electrochemical reactions, which affect magnetism in the electrode material [1].

Here we apply the concept of hydrogen-based magneto-ionics [2] to Co/Pd multilayer stacks with PMA [3]. In particular, we utilize the ability of Pd to host large quantities of hydrogen in its crystal lattice to control the magnetic properties of the multilayer stack via electrochemical hydrogen-loading. The magnetic multilayers consist of $[Co(0.4nm)/Pd(0.7nm)]_x$ building blocks providing the strong PMA, and supporting Pd layers on top and bottom, which facilitate hydrogen loading into the stack.

In order to probe magnetic properties upon electrochemical hydrogen-loading of the Co/Pd multilayers in an acidic electrolyte, an *in situ* electrochemical cell setup within a magneto-optical Kerr effect (MOKE) microscope is employed. This technique not only allows to acquire magnetization curves upon hydrogen-loading in real time, but also provides insight into the magnetization processes and dynamics by monitoring the magnetic field- and time-dependent behaviour of magnetic domains.

Our experiments demonstrate a controllable increase in coercivity in MOKE hystereis loops upon hydrogen-loading up to $\Delta H_c/H_{c,0}$ =20%, which is reversible over multiple loading- and unloading cycles (see Figure 1). Importantly, by adjusting the hydrogen concentration via loading voltage, the coercivity can be tuned continuously between the maximum coercivity for the hydrogen-loaded multilayer stack and the minimum coercivity upon complete unloading. The domain wall velocity in the regime of thermally activated domain creep magnetization reversal is found to decrease by a factor of up to 45 upon hydrogen-loading, which is an indirect result of our observed increase in coercivity. We discuss the observed coercivity increase on the basis of a changing anisotropy energy and orbital occupation caused by hydrogen-loading, as predicted by density functional theory calculations [4]. Lastly, an outlook of magneto-ionics in PMA multilayers is provided.

## References

[1] K. Leistner, *Curr. Opin. Electrochem.* **25** (2021) 100636

[2] M. Gößler, M. Albu, G. Klinser, E.-M. Steyskal, H. Krenn, R. Würschum, *Small* **15** (2019) 1904523

[3] S. Pan, O. Hellwig, A. Barman, *Phys. Rev. B* **98** (2018) 214436

[4] K. Klyukin, G. S. D. Beach, B. Yildiz, *Phys. Rev. Mater.* **4** (2020) 104416

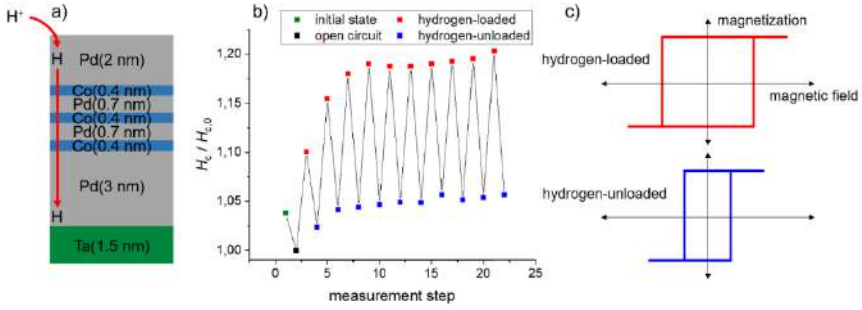



Figure: Magneto-ionic control of $[Co(0.4nm)/Pd(0.7nm)]_3$ multilayers via hydrogen loading. a) device structure, b) changes in coercivity upon several hydrogen-loading and -unloading cycles, c) schematic changes in the corresponding magnetic hysteresis loops in the hydrogen-loaded and -unloaded states.

# Magnetic switching of $Fe_3O_4$/Nb:STO heterostructures driven by electric-field

*Yifan Xu[1]*, Mai Hussein Hamed[1,2], Connie Berdnarski-Meinke[1], Steffen Tober[1], Patrick Schöffmann[3], Oleg Petracic[1], Thomas Brückel[1]*

[1] *Jülich Centre for Neutron Science (JCNS-2) and Peter Grünberg Institut (PGI-4), JARA-FIT, Forschungszentrum Jülich GmbH, 52425 Jülich, Germany*

[2] *Faculty of Science,Helwan University,11795 Cairo,Egypt*

[3] *Synchrotron SOLEIL, L'Orme des Merisiers, 91190 Saint-Aubin, France*

*corresponding author email address

The ability to tune magnetic oxide phases via redox reactions across their heterointerfaces could lead to useful spintronic and memristive device applications. By applying a small electric field, oxidation/ reduction occurs at the heterointerface which leads to a reversible phase transition [1-2]. We present the magnetic switching of epitaxial (001) $Fe_3O_4$ thin films grown on $TiO_2$-terminated (001) Nb:STO via pulsed laser deposition. Using magnetometry, we quantify the presence of the Verwey transition being an indicator of the oxygen content in the $Fe_3O_4$ films. We observe the disappearance of the Verwey transition with an applied positive electric field. This could be explained by oxygen diffusion through the interface which then leads to a reversible phase transition from $Fe_3O_4$(magnetite) to $\gamma$-$Fe_2O_3$(maghemite). In the next step, using in-situ XMCD, we probe the cation proportion of $Fe^{2+}$ and $Fe^{3+}$ while applying electric field. Additionally, magnetic depth profiling is investigated by PNR. This enables us to better understand the details of the phase switching at the interface. Combining the in-situ XMCD, PNR ad SQUID measurement, the magnetic switching phenomenon can be explained by the change of oxygen content across the thin film and the intralayer.

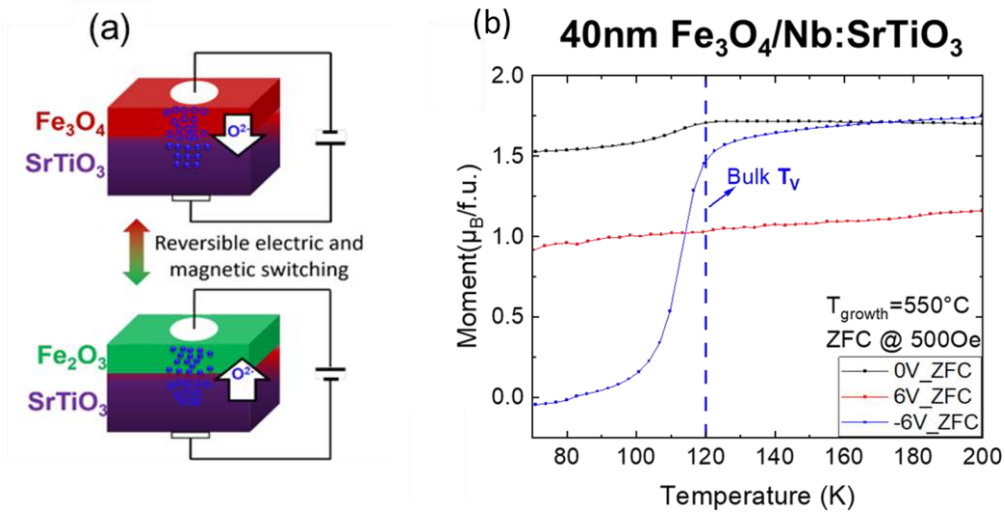



Figure: (a) Interfacial oxygen exchange through the active oxide interfaces motivates a reversible switching of complex oxide heterostructures via applied electric field; (b) In-situ zero filed cooling MT curves of 40 nm $Fe_3O_4$/Nb:$SrTiO_3$ film before and during applied ±6V.

## References

[1] M. H. Hamed, R. A. Hinz, P. Lömker, M. Wilhelm, A. Gloskovskii, P. Bencok, C. Schmitz-Antoniak, H. Elnaggar, C. M. Schneider and M. Müller, ACS Appl. Mater. Interfaces, 11 (7), 7576, 2019.

[2] M. H. Hamed, D. N. Müller and M. Müller, J. Mater. Chem. 8(4), 1335, 2020.

# Insights into Strain-induced Ferromagnetism in Epitaxial $SrMnO_3$

*Adriana I. Figueroa[1,2]*, Pedro A. Algarabel[3,4], José A. Pardo[3,5,6], Javier Blasco[3,4], Cinthia Piamonteze[7], Arantxa Fraile Rodríguez[1,2] Eric Langerberg[1,2],*

[1]*Depart. de Física de la Matèria Condensada, Universitat de Barcelona, Barcelona, Spain*
[2] *Institut de Nanociència i Nanotecnologia (IN2UB), Barcelona, Spain*
[3]*Instituto de Nanociencia y Materiales de Aragón (INMA), CSIC-Universidad de Zaragoza, Spain*
[4]*Depart. de Física de la Materia Condensada, Universidad de Zaragoza, Zaragoza, Spain*
[5]*Laboratorio de Microscopías Avanzadas, Universidad de Zaragoza, Zaragoza, Spain*
[6]*Depart. de Ciencia y Tec. de Materiales y Fluidos, Universidad de Zaragoza, Zaragoza, Spain*
[7]*Swiss Light Source, Paul Scherrer Institut, Villigen PSI, Switzerland.*
*aifigueroa@ub.edu

The development of multiferroic materials with strong magnetoelectric coupling would allow creating low energy consumption devices where the magnetic response could be controllable by an electric field. The coexistence between magnetism and polar order — driven by the off-centering of the magnetic $Mn^{4+}$ cation ($d^3$) — has been observed in $SrMnO_3$ perovskite compounds [1,2]. As the same cation is responsible for both the polar and magnetic orders, strong magnetoelectric coupling is expected. While in bulk $SrMnO_3$ the Mn—O—Mn superexchange magnetic interaction stabilizes the G type antiferromagnetic (AF) order, first-principles calculations suggest that different magnetic ground states may be accessible in $SrMnO_3$ epitaxial films by tuning the biaxial strain exerted by the substrate [3,4]. For tensile stress, a rich phase diagram is inferred, in which a progressive increase in the strain magnitude induces a gradual transition from different types of AF orders to eventually a ferromagnetic (FM) order [3,4].

Here, we study three fully strained 10 nm-thick $SrMnO_3$ epitaxial films grown on (La,Sr)(Al,Ta)$O_3$ (LSAT), $SrTiO_3$ (STO), and $DyScO_3$ (DSO) single-crystal substrates with mismatch values of +1.68%, +2.63%, and 3.78%, respectively, in order to determine the strain-oxygen vacancies-magnetic phase diagram. Synchrotron-based X-ray linear dichroism (XLD), X-ray magnetic circular dichroism (XMCD), and element-specific magnetic hysteresis loops, were performed in all samples around the $L_{2,3}$ -Mn edges at low temperature and high magnetic field. Preliminary results show sizeable XMCD signals and a slight opening of the hysteresis loops in the case of the DSO and LSAT, suggesting the emergence of FM order in these strained systems (see Figure 1). Besides, the comparison of the experimental data with calculated reference spectra is also in progress to determine the evolution of the $Mn^{3+}/Mn^{4+}$ ratio as a function of the strain and its effect on both the local magnetic moment and the resulting magnetic order.

## Acknowledgements

Part of this work was performed at the Swiss Light Source, PSI, Switzerland. Funding from the ART_2021_7 Ajut a la Recerca Transversal from IN2UB, and MINECO projects PGC2018-097789-B-I00, PID2020-112914RB-I00 and PID2021-127397NB-I00 is acknowledged. AIF and EL are Serra Húnter Fellows.

## References

[1] C. Becher, *et al*. *Nat. Nanotechnol.* **10** (2015) 661–666
[2] R. Guzmán, *et al*. *Nano Lett.* **16** (2016) 2221–2227
[3] J. H. Lee and K. M. Rabe, *Phys. Rev. Lett.* **104** (2010) 207204
[4] A. Edström and C. Ederer, *Phys. Rev. Mater.* **2** (2018) 104409

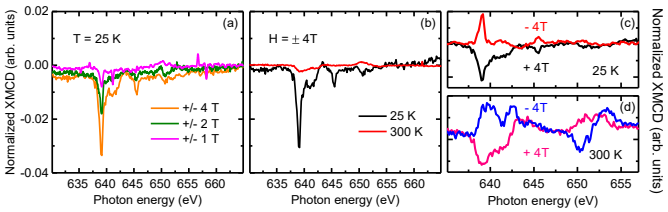



Figure 1: XMCD for the $SrMnO_3$ thin film grown on DSO. (a) Magnetic field dependence. (b) Temperature dependence. Panels (c) and (d) depict inversion of the signal with opposed magnetic fields.

# Strain-Engineering of Magnetic Anisotropy in Vertically Aligned Nanocomposites

*M. Hennes[1*], X. Weng[1], D. Demaille[1], C. Rodríguez Cortéz[1], Y. Zheng[1], F. Vidal[1]*

[1]*Sorbonne Université, CNRS, Institut des NanoSciences de Paris, INSP, UMR7588, F-75005 Paris, France*

*marcel.hennes@sorbonne-universite.fr

Self-assembled vertically aligned nanocomposites (VANs), i.e., dense arrays of nanopillars embedded in a host matrix, have emerged as a novel playground for strain engineering approaches in nanostructures [1]. In contrast to thin films obtained by classical planar heteroepitaxy, VANs consist of two (or more) intertwined phases exhibiting strong coupling along vertical interfaces (Figure 1a). This allows for an efficient steering of the properties of ultrasmall magnetic nano-objects via magnetoelastic effects.

In the present work, we show how sequential pulsed laser deposition can be used to obtain nanometer-sized wires and pillars consisting of Co-Ni alloys embedded in various oxide host matrices (Figure 1b). With a combined experimental-simulation approach, we shed light on the growth and self-assembly mechanisms of such metal-oxide VANs and illustrate the impact of the synthesis parameters on the final nanoarchitecture of these hybrid systems [2].

Special emphasis is put on the final strain state of the embedded metallic nanowires [3]. We show that under optimized conditions, huge out-of-plane dilations can be reached (>4%), which is evidenced with x-ray diffraction and transmission electron microscopy. In such samples, magnetoelastic effects become dominant and allow to enhance the uniaxial anisotropy of the nanowires leading to blocking temperatures exceeding 600 K in the case of pillars with a diameter of 5 nm and high Co content. These data are eventually complemented with angular dependent x-ray magnetic circular dichroism (XMCD) measurements at the Co and Ni $L_{2,3}$ edges. This allows to extract the value of the magnetic moment by applying sum rules and the anisotropy of the orbital moment, providing a microscopic interpretation of the observed effects [4].

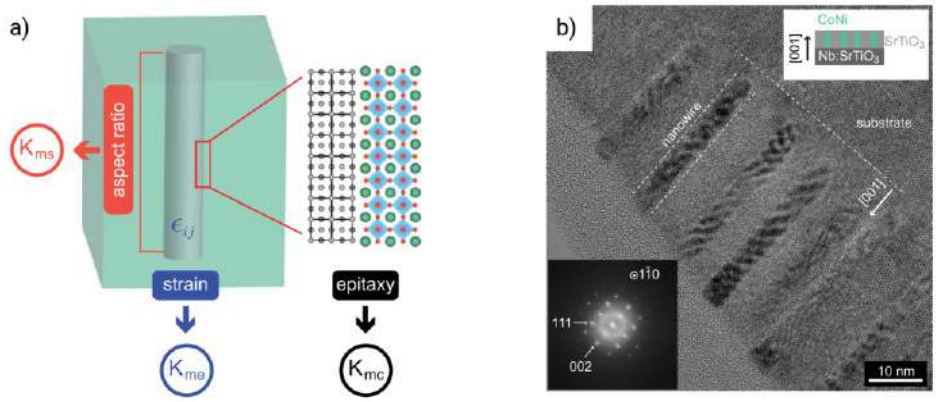



Figure 1: (a) Schematics of a VAN highlighting the distinct contributions to magnetic anisotropy. (b) Cross-sectional TEM image of a CoNi/$SrTiO_3$ composite thin film (adapted from [4])

## Acknowledgements

We acknowledge SOLEIL synchrotron for granting beam time and the DEIMOS beamline staff for support. We gratefully thank P. Sainctavit and A. Juhin for their help with regard to XMCD data interpretation and D. Hrabovsky for support with magnetometry measurements.

## References

[1] A. Chen et al., *Adv. Mater.* **31** (2019) 1803241
[2] M. Hennes et al., *Nanoscale* **10** (2018) 7666-7675
[3] X. Weng et al., *Phys. Rev. Materials* **2** (2018) 106003
[4] X. Weng et al., *Phys. Rev. Materials* **6** (2022) 046001

# Tailoring Exchange Bias Phenomena in V2O3/Co bilayers driven by Ferromagnetic Domains

*José Manuel Díez,[1,2]* Jose Luis F. Cuñado,[2] Pavel Lapa,[3] Inés Gacía-Manuz, [1,2] Guillermo de Arana, [2] Paolo Perna, [2] Alberto Bollero, [1] Rodolfo Miranda, [1,2] Ivan K. Schuller, [3] and Julio Camarero[1,2]*

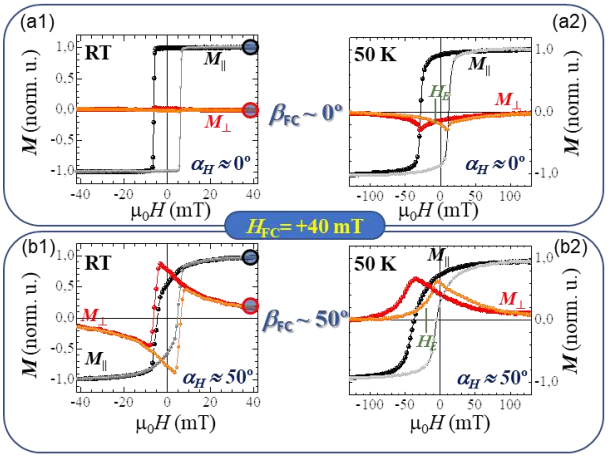
[1]*Dpto. Física de la Materia Condensada, IFIMAC, and Instituto "Nicolás Cabrera", Universidad Autónoma de Madrid, 28049Madrid, Spain*

[2]*IMDEA-Nanociencia, Campus de Cantoblanco, 28049 Madrid, Spain*
[3]*University of California San Diego, La Jolla, CA 92093, USA*

* josemanuel.diez@estudiante.uam.es

The research on hybrid systems combining dissimilar materials with artificial interfaces is among the major challenges of the nanotechnology during the last decades. Different functionalities may be engineered by combining dissimilar materials. To this regard, bilayers composed of a ferromagnetic (FM) layer interfaced with a transition-metal-oxide (TMO) layer can be considered as model hybrid systems $V_2O_3$ is an archetypical TMO system exhibiting a first-order metal insulator transition (MIT) when the temperature is decreased below $T_{MIT}$= 170 K [1], from a paramagnetic metallic state to an antiferromagnetic (AFM) insulating phase, accompanied simultaneously by a structural phase transition [2]. FM/TMO bilayers could provided with an additional degree of freedom in order to control the FM properties at the interface exploting control exchange bias (EB) phenomena. Increase in coercivity is general, but very different interfacial exchange-coupling effects have been observed even in similar systems, ranging from moderate to negligible exchange bias field ($H_E$) to dissimilar temperature effects. The discordant results are usually ascribed to uncontrolled parameters, namely dissimilar interfacial defects, structure, and thicknesses.

Here we present detailed angular and temperature dependent magnetic and transport (simultaneous) characterizations of a $Co/V_2O_3$ bilayer across the MIT, where the Co layer has a well-defined uniaxial magnetic anisotropy at room temperature (Fig. a1 and Fig. b1). Vectorial-resolved magnetic measurements performed in the very same sample during warming after different field cooling (FC) procedures exhibit a strong dependence on the FC angle ($\beta_{FC}$) and the domain structure of the FM layer, while equal transport features [3]. Just after the FC, the low temperature (50 K) loops display strong interfacial exchange coupling effects (Fig.1a2 and Fig.1b2), i.e., enhanced coercive field, clear negative exchange bias, and asymmetric magnetization reversal. Remarkably, up to a factor of two in $H_E$ value is found after different $\beta_{FC}$ whereas the magnetization reversal analysis during warming reveals 35 K of variation in blocking temperature ($T_B$). These observations can be explained within the random-field model for the interfacial exchange coupling with a fixed AFM domain structure in contact with a variable (angle-dependent) FM domain structure. The results highlight the importance of the domain structure and magnetization reversal of the FM layer (not previously considered) in the EB phenomena, with potential to tailor interfacial effects in future spintronic devices.




## References

[1] M. Imada, A. Fujimori, and Y. Tokura, Rev. Mod. Phys. **70**, 1039-1263 (1998).
[2] Y. Kalcheim, et al. Phys.Rev. Lett. **122**, 057601 (2019).
[3] J. M. Díez et al., Adv. Mater. Interfaces **9**, 2200331 (2022).

# Magneto-ionic control of magnetism via wireless bipolar electrochemistry in CoN thin films

*Zheng Ma[1*], Laura Fuentes-Rodriguez[2,3], Zhengwei Tan[1], Eva Pellicer[1], Llibertat Abad[3], Javier Herrero-Martín[4], Enric Menéndez[1], Nieves Casañ-Pastor[2], Jordi Sort[1,5]*

[1] *Departament de Física, Universitat Autònoma de Barcelona, 08193 Cerdanyola del Vallès, Spain*

[2] *Institut de Ciencia de Materials de Barcelona, CSIC, Campus UAB, 08193 Bellaterra, Spain*

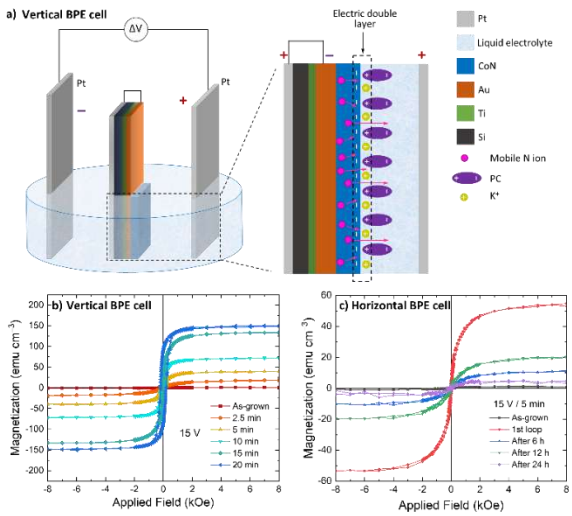
[3] *Institut de Microelectrónica de Barcelona-Centre Nacional de Microelectrónica, CSIC, Campus UAB, 08193 Bellaterra, Barcelona, Spain*

[4] *ALBA Synchrotron Light Source, 08290 Cerdanyola del Vallès, Spain*

[5] *Institució Catalana de Recerca i Estudis Avançats (ICREA), Pg. Lluís Companys 23, Barcelona 08010, Spain*

*mazheng2016@gmail.com

Modulation of magnetic properties through voltage-driven ion motion and redox processes, i.e., magneto-ionics, is a unique approach to control magnetism with electric field for low-power memory and spintronic applications. So far, magneto-ionics has been achieved through direct electrical connections to the actuated thin films. Here we evidence that an alternative way to reach such control is possible in a wireless manner. Induced polarization in the material immersed in the electrolyte, without direct wire contact, promotes wireless bipolar electrochemistry, an alternative pathway to achieve voltage-driven control of magnetism based on the same electrochemical processes involved in direct-contact magneto-ionics [see Figure a)]. A significant tunability of magnetization is accomplished for 50-nm CoN thin films, including transitions between paramagnetic and ferromagnetic states (e.g., a magnetization increment from zero to over 200 emu cm$^{-3}$). Such effects can be either volatile or non-volatile depending on electrochemical cell configurations [see Figure b) and c)]. These results represent a fundamental breakthrough that may inspire future device designs for applications in bioelectronics, catalysis, neuromorphic computing, or wireless communications.




## Acknowledgements

Financial support by the European Union's Horizon 2020 Research and Innovation Programme ('BeMAGIC' European Training Network, ETN/ITN Marie Skłodowska–Curie grant Nº 861145), the European Research Council (2021-ERC-Advanced 'REMINDS' Grant Nº 101054687), the Spanish Government AEI (PID2020-116844RB-C21 and PDC2021-121276-C31, RTI2018-097753-B-I00, PID2021-123276OB-I00, and Severo ochoa CEX2019-000917-S) and the Generalitat de Catalunya (2021-SGR-00651) is acknowledged. We thank ALBA synchrotron for providing beamtime for synchrotron X-ray spectroscopic experiments.

# SYMPOSIUM 10.
## MAGNETISM OF ALLOYS, INTERMETALLICS, THIN FILMS AND MULTILAYERS.
## S10. POSTERS

# Study on the magnetic properties of an additive manufactured Fe-Cr-Co alloy using microstructural EBSD investigations

*S. Arneitz 1*, R. Buzolin 1, S. Rivoirard 2, P. de Rango2 and C. Sommitsch 1*
*1 Institute of Materials Science, Joining and Forming, Graz University of Technology, Kopernikusgasse 24/I, 8010 Graz (Austria)*
*2 Université Grenoble Alpes, CNRS, Grenoble INP, Institute Néel, 38042 Grenoble (France)*
*corresponding author email address

## Incentive of the study

In recent years, additive manufacturing has become more and more relevant for producing magnetic materials due to higher demands for miniaturisation and complex-shaped magnet parts. Using laser powder bed fusion (LPBF), magnet parts of the Fe-Cr-Co system can be produced with notable shape accuracy. The chemical composition can be modified directly in the printing chamber with the in-situ alloying technique. With this novel method, complex alloys can be produced with a chemical composition accustomed to each specific application case. This study aimed to study the influence of the process conditions on the magnetic properties of selected 3D printed magnet parts.

## Description of the work

Fe-Cr-Co is an Fe-Co based magnetic alloy, which obtains its hard- magnetic properties due to a spinodal decomposition of a solid- solution alpha phase with bcc crystal structure into a ferromagnetic Fe-Co phase and a paramagnetic Cr-Fe matrix. The phase transformation occurs during heat treatment at a given temperature in the presence of a high magnetic field. The LPBF process can have a considerable influence on microstructure (e.g. homogeneity and texture) and the influence of printing parameter on the magnetic properties has been demonstrated before. [1] Based on these results, further investigations have been conducted measuring samples with additional printing parameter and/or post processing conditions and further differences in magnetic properties have been observed here. To correlate the occurring changes in the magnetic properties to the changes in the microstructure upon using different process and post- process conditions, EBSD measurements have been performed as a core part of the study. Measurements have been performed both on samples prepared before and after the thermomagnetic treatments in order to gain a detailed insight on the separate roles of process and post process conditions on the microstructure with the aim to find a correlation between microstructure and magnetic properties. A special focus has been set on the texture evolution during the thermomagnetic treatment.

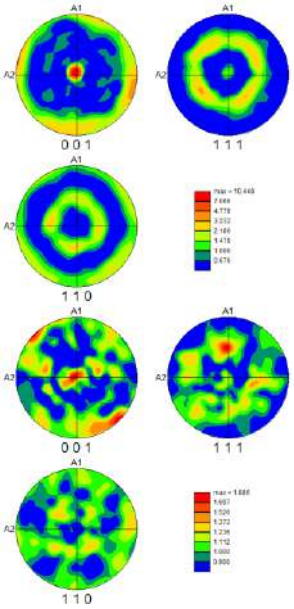



Figure 1: A strong focus in this work has been set on the texture analysis in the samples both after printing and post- processing

## Acknowledgements

The results presented were achieved in the course of the AddMag project which is funded by the 2021 m-era.net program.

## References

[1] S. Arneitz, R. Buzolin, S. Rivoirard, and C. Sommitsch, “Additive manufacturing of an Fe–Cr–Co permanent magnet alloy with a novel approach of in-situ alloying,” Eur. J. Mater., vol. 2, no. 1, pp. 475–497, Dec. 2022, doi: 10.1080/26889277.2022.2098832.

# Coupling of magnetism and structural phase transition in $V_2O_3$/Ni heterostructures

*K. Ignatova[1]*, E.O. Vlasov[2], T.P.A. Hase[3], U.B. Arnalds[1]*

[1] *Science Institute, University of Iceland, Dunhaga 3, 107 Reykjavik, Iceland*
[2] *Research Group EMAT, University of Antwerp, Groenenborgerlaan 171, Antwerp, Belgium*
[3] *Department of Physics, University of Warwick, Coventry CV4 7AL, United Kingdom*

*kri12@hi.is

We present a comprehensive study of the structural evolution of thin film $V_2O_3$ across its structural phase transition (SPT) [1] and the magnetic response of epitaxial $V_2O_3$/Ni bilayers [2]. Scanning transmission electron microscopy (STEM) reveals the abruptness of the $V_2O_3$ interfaces with both the sapphire substrate and Ni layer. The results show that the $V_2O_3$ layer exhibits a symmetry-breaking effect in the form of anti-phase boundaries (Fig. 1). Temperature-dependent x-ray diffraction and reflectivity measurements across the SPT reveal the transition to occur at a temperature of 135 K upon heating with an increase in layer thickness by 1.5% and a peak in roughness at the transition. Figure 1 shows the coercivity derived from hysteresis loops recorded as a function of temperature. A notable sharp strain-related decrease and a clear peak of enhanced coercivity are observed occurring at a temperature coinciding with the structural phase transition. This temperature range of enhanced coercivity corresponds well to the $V_2O_3$ phase coexistence.

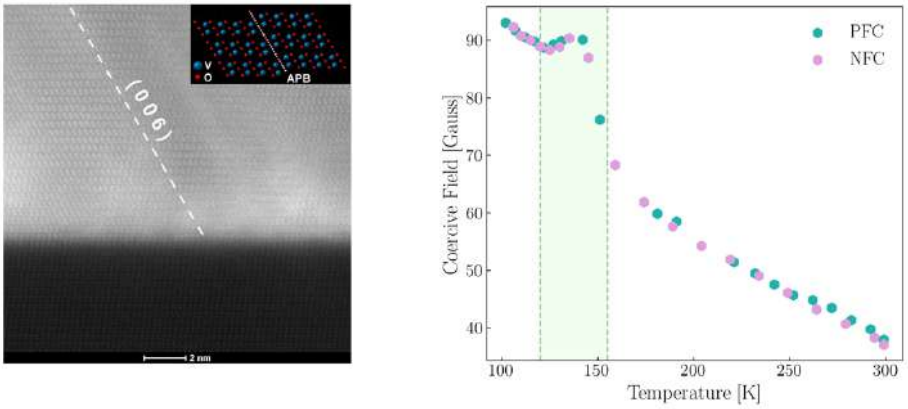



Figure 1: (Left) Experimental high-angle annular dark-field-STEM image of the substrate/$V_2O_3$ interface. The inset shows schematically the geometry of the antiphase domains, indicated by white dashed line in the STEM image. (Right) Coercivity of the $V_2O_3$/Ni film extracted from magnetization loops recorded using low-temperature magneto-optical Kerr effect measurements as a function of increasing temperature after positive and negative field cooling (PFC/NFC) under an applied field of +/- 400 G along the easy axis.

## Acknowledgements

This work was supported by the Icelandic Research Fund Grant No. 207111. This work is based on experiments performed at the BM28 (XMaS) beamline at the European Synchrotron Radiation Facility, Grenoble, France. STEM experiments were funded from the European Union's Horizon 2020 research and innovation programme under grant agreement No 823717 – ESTEEM3.

## References

[1] E. B. Thorsteinsson, S. Shayestehaminzadeh, A. S. Ingason, F. Magnus and U. B. Arnalds, "Controlling metal–insulator transitions in reactively sputtered vanadium sesquioxide thin films through structure and stoichiometry", *Sci. Rep.* **11** (2021) 123.

[2] K. Ignatova, et al., "Reversible exchange bias in epitaxial $V_2O_3$/Ni hybrid magnetic heterostructures", *J. Phys. Condens. Matter.* **34** (2022) 49.

# Interplay Between Magnetism and Short-Range Order in Ni-Based High-Entropy Alloys: CrCoNi, CrFeCoNi, and CrMnFeCoNi

*Christopher D. Woodgate[1]*, Daniel Hedlund[2], L. H. Lewis[2,3], Julie B. Staunton[1]*

[1] *Department of Physics, University of Warwick, Coventry, CV4 7AL, UK*
[2] *Department of Chemical Engineering, Northeastern University, Boston, MA 02115, USA*
[3] *Department of Mechanical and Industrial Engineering, Northeastern University, Boston, MA 02115, USA*
*C.Woodgate@warwick.ac.uk

We suggest a potential route for tuning atomic ordering in medium- and high-entropy alloys, by annealing samples in an applied magnetic field and thus altering the magnetic state. Controlling atomic order is critical for tuning materials properties, and our approach could open up new routes for discovery of novel materials. Specifically, the impact of magnetism on predicted atomic short-range order in Ni-based high-entropy alloys is studied using a first-principles, all-electron, Landau-type linear response theory, coupled with lattice-based atomistic modelling [1,2]. We perform two sets of linear-response calculations: one in which the paramagnetic state is modelled within the disordered local moment picture, and one in which systems are modelled in a magnetically ordered state. We show that the treatment of magnetism can have significant impact both on the predicted temperature of atomic ordering and also the nature of atomic order itself [3]. In CrCoNi, we find that the nature of atomic order changes from being $L1_2$-like when modelled in the paramagnetic state to $MoPt_2$-like when modelled assuming the system has magnetically ordered. In CrFeCoNi, atomic correlations between Fe and the other elements present are dramatically strengthened when we switch from treating the system as magnetically disordered to magnetically ordered. Our results show it is necessary to consider the magnetic state when modelling multicomponent alloys containing mid- to late-*3d* elements, and we suggest that, potentially, there could be a variety of multicomponent alloy compositions containing *3d* transition metals that will exhibit specific atomic ordering when thermally treated in an applied magnetic field.

## Acknowledgements

We gratefully acknowledge the support of the UK EPSRC, Grant No. EP/W021331/1. C.D.W. is supported by a studentship within the UK EPSRC-supported Centre for Doctoral Training in Modelling of Heterogeneous Systems, Grant No. EP/S022848/1. This work was also supported in part by the U.S. Department of Energy, Office of Basic Energy Sciences under Award Number DE SC0022168 and by the U.S. National Science Foundation under Award ID 2118164.

## References

[1] C. D. Woodgate and J. B. Staunton, Phys. Rev. B **105**, 115124 (2022).
[2] C. D. Woodgate and J. B. Staunton, Phys. Rev. Mater. **7** 013801 (2023).
[3] C. D. Woodgate D. Hedlund, L. H. Lewis, J. B. Staunton, arXiv:2303.00641 (2023)

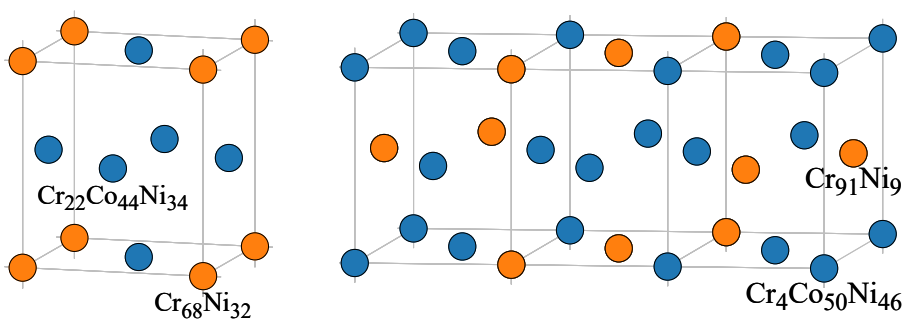



Figure: Competing chemical orderings in the medium-entropy alloy CrCoNi. In the paramagnetic state, it is $L1_2$-like order (left) which dominates, but once magnetic order has established, it is $MoPt_2$-like order (right) which is preferred.

# Growth and Characterization of $Ir_{1-x}Co_xO_2$ Thin Films

*M.A. Laguna–Marco[1, 2]*, J. Herrero-Albillos[1, 3], M. Rueda[3], M. H. Aguirre[1,2,4]*

[1]*Instituto de Nanociencia y Mat. de Aragón (INMA), CSIC – Univ. de Zaragoza, 50009 Zaragoza, Spain*

[2]*Departamento de Física de la Materia Condensada, Univ. de Zaragoza, 50009 Zaragoza, Spain*

[3]*Departamento de Ciencia y Tecnología de Materiales y Fluidos, Univ. de Zaragoza, 50018 Zaragoza, Spain*

[4]*Laboratorio de Microscopías Avanzadas, Univ. de Zaragoza, 50018 Zaragoza, Spain*

*anlaguna@unizar.es

$IrO_2$ is one of the most promising materials for spin–current detection applications due to the high spin–orbit coupling (SOC) of the Ir ions that makes possible an efficient conversion of pure spin-currents on charge-currents via the so-called inverse spin Hall effect (ISHE) [1, 2]. In order to improve the performance of $IrO_2$ as spin detector doping with different elements appears as a natural route. Indeed, in the current quest for optimized spintronic materials, doping is one of the most feasible and commonly used methods to modify the electronic structure of the host compounds [3,4]. The different ionic size of the dopant may give rise to contraction/elongation of the lattice and/or distortions that can significantly change the electronic structure. Similarly, the dopant ions can induce relevant changes in the oxidation state of the host. In addition, when a magnetic ion is inserted in the lattice it can give rise to the appearance of magnetic ordering and Slater- insulator behaviors. The doping of $IrO_2$ with 4+ ions, such as $Sn^{4+}$ and $Cr^{4+}$, has been reported to give rise to strong changes in its electronic and/or magnetic behavior [5,6]. On the other hand, the effect of increasing the oxidation state of Ir in $IrO_2$ films has remained unexplored to date, even when in other iridium oxide series a larger SOC has been reported for $Ir^{5+}$ relative to $Ir^{4+}$ [7]. Here, $Ir_{1-x}Co_xO_2$ thin films have been prepared by reactive magnetron co–sputtering deposition. Composition, structure, oxidation state and magnetic behavior have been analyzed. After annealing, an $Ir_{1-x}Co_xO_2$ substitutional solid solution phase was achieved for a wide Co-doping range ($0 \leq x \leq 0.6$). In all the cases a rutile crystal structure was found even when no binary Co oxide crystallizes in this structure. XAS spectra show that the oxidation state of cobalt is $Co^{3+}$ and that this triggers an increase in the oxidation state of Ir, up to ~ 5+ in the $Ir_{0.6}Co_{0.4}O_2$ sample. By application of sum rules, a 13% increase in the SOC is found despite the fact that lattice shrinkage causes a detrimental bandwidth broadening. In addition, our work shows that the magnetic response of the doped films is very similar to that of the paramagnetic parent $IrO_2$. No enhancement of the susceptibility is observed indicating a non-magnetic low spin Co3+ electronic configuration. This results indicate a route for further optimization of the performance of the $IrO_2$ as spin current detectors.

**Acknowledgements**

Spanish MICINN Project No. PID2020-115159GB-I00/AEI/10.13039/501100011033 and DGA Grant No. E12-20R (RASMIA). We acknowledge Diamond Light Source for beamtime under Proposal SP-32410 and the project CALIPSO plus under the Grant Agreement 730872 from the EU Framework Programme for Research and Innovation HORIZON 2020. We acknowledge the B18 staff for technical support at the beamline. Authors would like to acknowledge the use of Servicio General de Apoyo a la Investigación-SAI (Univ. Zaragoza) and the use ofthe National Facility ELECMI ICTS, node "Laboratorio de Microscopías Avanzadas" at the Univ. Zaragoza.

**References**

[1] K. Fujiwara et al., Nat. Commun. 4 (2013) 2893
[2] M. Morota et al., Phys. Rev. B 83 (2011) 174405
[3] N. Che Ani et al., Materials Science and Engineering: B 276 (2022) 115536
[4] V. K. Dwivedi and S. Mukhopadhyay, Journal of Applied Physics 126 (2019) 165112
[5] E. Arias-Egido, et al., Adv. Funct. Mater. 29 (2019) 1806754
[6] E. Arias-Egido et al., , Materials & Design, 196 (2020) 109083
[7] M. A. Laguna-Marco et al., Phys. Rev. B 91 (2015) 214433

# Multi phase driven operating temperature range for magnetocaloric effect in $Ho_2Co_2In$ alloy

*S. R. Athul[1], K. Arun [1], S. Swathi [1], U. D. Remya[1], A. Dzubinska[2], M. Reiffers[3]*, R. Nagalakshmi[1]*

[1] *Intermetallics and non-linear optics laboratory, Department of Physics, National Institute of Technology, Tiruchirappalli 620 015, India*

[2] *CPM-TIP, University Pavol Jozef Safarik, 04011 Košice, Slovakia*

[3] *Faculty of Humanities and Natural Sciences, Presov University, 081 16 Presov, Slovakia*

*marian.reiffers@unipo.sk

The magnetic and magnetocaloric characteristics of $Ho_2Co_2In$ alloy have been investigated. In sample two phases are present, which crystallize in cubic $HoCo_2$, and unidentified $HoCo_{1.5}In_{0.33}$ structures, resp.. At temperatures $T_1$ = 123 K and $T_2$ = 34 K, the titled alloy exhibits ferromagnetic ordering and at $T_3$ = 7 K it undergoes antiferromagnetic ordering. The latter transition is suppressed by applied magnetic field leading to an overall ferromagnetic behaviour. The ferromagnetic transitions are found to be second order in nature with negligible magnetic hysteresis. The magnetocaloric effect (MCE) is estimated from isothermal magnetization data using Maxwell's equations. The multiple magnetic transitions presented in the alloy, have lead to twin peaks in MCE with maximum relative cooling power (RCP) value of 595 J/kg, which is obtained for the applied magnetic field change of 0 - 9 T. Additionally, alloy exhibits a maximum in magnetoresistance with magnitude of -15 % at 123 K.

**Acknowledgements**

The author, Athul S R, thanks CSIR, Govt. India for awarding JRF through CSIR fellowship (No. 09/895(0013)/2019-EMR-I). The authors are grateful to Mr. Nilesh Kulkarni and Mrs. Bhagyashree Chalke, Department of Condensed Matter Physics, Tata Institute of Fundamental Research, Mumbai, India in structure measurements. This research work is a part of the Project implementation: University Science Park TECHNICOM for Innovation Applications Supported by Knowledge Technology, ITMS: 313011D232., supported by the Research & Development Operational Programme funded by the ERDF; and also by VEGA 1/0404/21 and VVGS-2022-2117.

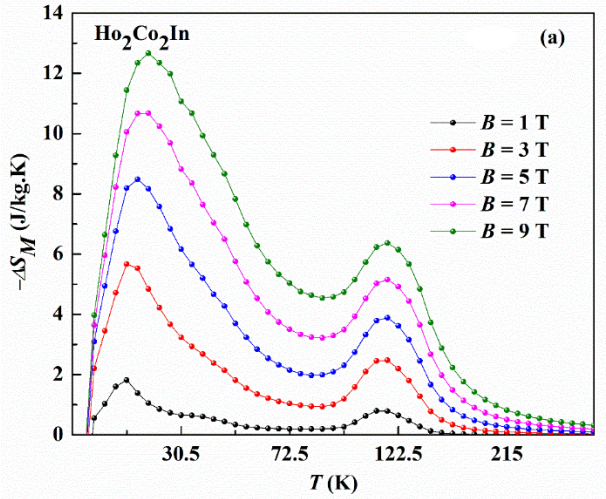



Figure: Temperature dependence of magnetic entropy change for magnetic field changes up to 9 T.

# Controlling Magnetic Properties of Ir/Co/Pt Layered System through Low-Energy $Ga^+$ Ion Bombardment

*M. Kowacz*, M. Schmidt, F. Stobiecki, and P. Kuświk*

*Institute of Molecular Physics, Polish Academy of Sciences, Mariana Smoluchowskiego 17, 60-179 Poznań, Poland*

*mateusz.kowacz@ifmpan.poznan.pl

Interfaces play a crucial role in inducing various important magnetic properties in magnetic thin-film systems [1-2], such as perpendicular magnetic anisotropy (PMA) and Dzyaloshinskii-Moriya interaction. The source of these properties can be associated with the interfaces, which can be influenced by the appropriate selection of the ferromagnetic layer and its surroundings, as well as the quality of the ferromagnet interfaces. It should be noted that the interface-induced properties arise independently from both interfaces. Therefore, it is crucial to precisely engineer the interfaces, and ion bombardment provides a means to achieve such controlled modification.

In this study, we investigated the modification of interfaces in a perpendicularly magnetized Ti(4nm)/Au(30nm)/Ir(30nm)/Co(0.8nm/)Pt(5nm) layered system induced by $Ga^+$ ion bombardment with energies ranging from 5 to 30 keV and doses ranging from $10^{12}$ to $10^{15}$ $Ga^+/cm^2$. Our results from polar magneto-optical Kerr effect (P-MOKE) measurements showed that as the ion energy ($E_{ion}$) was lowered, the spin reorientation transition (SRT) from PMA to easy plane anisotropy occurred for increasingly higher doses ($D$). This indicates there is progressively weaker ballistic mixing efficiency as the $E_{ion}$ is lowered. To explain these changes, Monte-Carlo simulations with the TRIDYN code were performed. The depth profile of Ir, Co, and Pt concentration distribution after bombardment for $E_{ion}$=5, 8, 30 keV, and $D$ corresponding to SRT ($D_{SRT}$) are presented in Fig. 1. The $D_{SRT}$ values were determined from P-MOKE measurements performed after ion bombardment with different values of $D$ and $E_{ion}$. For such selected $D_{SRT}$ values, it can be expected that changes in concentration profiles caused by ion bombardment will be similar for different $E$ values. However, as shown in Fig.1, these changes are similar only for the bottom (Ir/Co) interface. This result strongly suggests that the PMA of the studied layered films is mainly correlated with the structural modification of the lower interface.

Our results show that appropriately selected ion bombardment parameters, such as ion energy and ion dose, allow enabling stronger modification of the upper interface, which can be used to tailor magnetic properties induced by the upper interface.

**Acknowledgments**

This study was supported by National Science Centre Poland: OPUS (UMO-2019/33/B/ST5/02013)

**References**

[1] M.T. Johnson et al., *J. Magn. Magn. Mater.* **148** (1995) 118–124

[2] A. Soumyanarayanan et al., *Nature* **539** (2016) 509–517

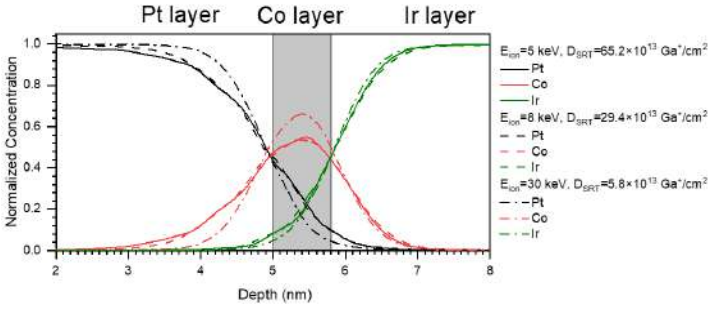



Figure 1: Concentration profiles of Ir/Co/Pt systems obtained by Monte-Carlo simulation of ion bombardment (TRIDYN code) for $Ga^+$ ions with energies: 5 keV (solid line), 8 keV (dashed line), 30 keV (dash-dot line) and for dose where is achieved SRT ($D_{SRT}$). The $D_{SRT}$ value was obtained from P-MOKE measurements. Black, red, and green lines indicate Pt, Co, and Ir concentration, respectively.

# Growth and Magnetic Properties of Vertically Aligned Photostrictive Nanocomposites

*C. Rodríguez[1]*, M. Hennes[1], D. Demaille[1], Y. Zheng[1], D. Hrabovsky[2], F. Vidal[1]*

[1] *Institut des NanoSciences de Paris, 4 place Jussieu, 75005 Paris, France*

[2] *Plateforme de Mesures Physiques à Basse Température, 4 place Jussieu, 75005 Paris, France*

*cortez@insp.jussieu.fr

Vertically aligned nanocomposites (VAN) consist of arrays of nanometric pillars perpendicularly embedded within a thin film (Fig. 1(a)). Because of their size and shape, the nanopillars in a VAN exhibit enhanced sensitivity to interfaces and strong lattice coupling to the surrounding matrix. This makes magnetostrictive VANs an ideal candidate for the study of magnetostriction [1] and allows the design of novel functional hybrid films where the magnetic properties of the pillars can be steered by playing with the strain state of the matrix.

In this work, we explore a photostrictive-magnetic model system consisting of spinel $CoFe_2O_4$ (CFO) pillars coupled to a light-responsive $SrRuO_3$ (SRO) photostrictive perovskite matrix [2,3]. While light-induced effects in these systems have been evidenced, a detailed understanding of the underlying processes, as well as a quantitative description of the timescales relevant to photostrictive expansion and magnetization control are currently lacking. However, such information is of crucial importance for the development of ultrafast hybrid devices, where femtosecond optical pulses can be used to manipulate the magnetization of nanometer-sized magnetic structures.

As a first step towards future time-resolved studies, we present a systematic investigation of deposition parameters on the final nanoarchitecture and magnetic properties of the thin films and their optimization for future femtomagnetism investigations. All samples have been synthesized by pulsed laser deposition under growth conditions optimized for epitaxy on $SrTiO_3$ substrates. Using X-ray diffraction and transmission electron microscopy, we have shown that the obtained nanopillars are approximately 5-10 nm in diameter with variable height (Fig. 1(b)) and compressively strained in the out-of-plane direction, inducing perpendicular magnetization, which makes them amenable to light-induced anisotropy control via optically induced matrix lattice dilation.

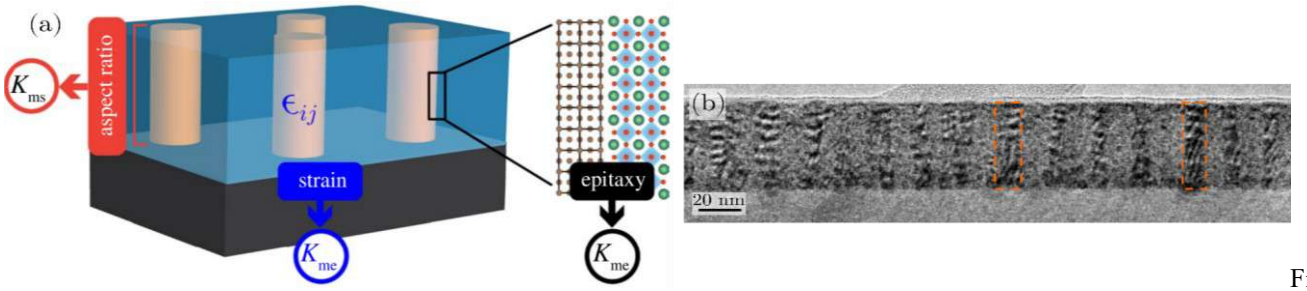



Figure 1: Schematics of a VAN (substrate: black, matrix: blue) highlighting contributions to magnetic anisotropy (a) (reproduced from [1]). Cross-sectional TEM image of one of our CFO-SRO VANs. Moiré patterns are observed at nanopillar locations, two of which are highlighted by dashed rectangles (b).

**Acknowledgements**

We acknowledge support from ANR, contract No. ANR-21-CE09-0042.

**References**

[1] M. Hennes, D. Demaille, G. Patriarche, T. Tran, Y. Zheng, F. Vidal, *MRS Bulletin* **46**, 136–141 (2021).
[2] H.-J. Liu, L.-Y. Chen, Q. He, C.-W. Liang, Y.-Z. Chen et al. *ACS Nano* **6**, 8 (2012), 6952–6959.
[3] T.-C. Wei, H.-P. Wang, H.-J. Liu, T.-S. Tsai, J.-J. Ke et al. *Nature Communications* **8**, (2017) 15018.

# Intrinsic Magnetic Properties of $ThMn_{12}$ - type Sm-Fe-Co-V Alloys

*D. Benea[1]*, V. Pop[1], R. Hirian[1] and J. Minar[2]*

[1] *Faculty of Physics, Babeş-Bolyai University,Cluj-Napoca, Romania*
[2]*New Technologies–Research Center, University of West Bohemia, Pilsen, Czech Republic*

*diana.benea@ubbcluj.ro

The present study shows the theoretical calculations on intrinsic properties of $SmFe_{12-x-y}Co_xV_y$ (x, y = 0,1,2) alloys, including some of the most promising permanent magnets between the 1:12 alloys. The theoretical investigations are based on the Korringa-Kohn-Rostoker (KKR) band structure method, using the Local Spin Density Approximation (LSDA) with Hubbard-U correction (LSDA+U)[1]. The calculated magnetic moments, magneto-crystalline anisotropies and Curie temperatures for the stable $SmFe_{12-x}V_x$ (x=1,2) and quasi-stable $SmFe_{10}Co_2$ magnetic alloys are compared with available experimental and theoretical data [2,3] in order to show the reliability of the present theoretical approach.

Afterwards, the intrinsic magnetic properties of the hypothetical $SmFe_{10}CoV$ alloy are evaluated. This 1:12 alloy is expected to be stable, due to V content exceeding the minimum required to stabilize the phase (y = 1), being also susceptible to maintain some of excellent intrinsic properties of the $SmFe_{10}Co_2$ alloy. Relatively high values of saturation magnetization (1.30 T) and magnetocrystalline anisotropy (9.35 $MJ/m^3$) have been obtained, whilst a Curie temperature of 945 K is predicted by mean-field approach (MFA) for this $SmFe_{10}CoV$ alloy. These theoretical results recommend the $SmFe_{10}CoV$ alloy as a valuable candidate for permanent magnets applications.

## Acknowledgements

The financial support from the Romanian Ministry of Education and Romanian Ministry of Research and Innovation, CCCDI-UEFISCDI grant PN-III-P2-2.1-PED-2019-3484 is acknowledged.

## References

[1] H. Ebert, D Ködderitzsch and J. Minar, Rep. Prog. Phys, 74 096501 (2011).
[2] Y. Hirayama, Y. K. Takahashi, S. Hirosawa and K. Hono, Scr. Mater.138 62 (2017).
[3] D. Odkhuu, T. Ochirkhuyag, S. C. Hong, Phys. Rev. Appl. 13, 054076 (2020).

# Ferroelectric Thermal Treatments: an Additional Lever in Magnetoelectric Heterostructures

*Deepak Dagur[1,2]*, Vincent Polewczyk[1], Aleksandr Yu. Petrov[1], Simone Dolabella[1], Alice Margherita Finardi[1,3], Claudia Fasolato[4], Andrea Giugni[1,3], Giorgio Rossi[1,3], Giancarlo Panaccione[1], Piero Torelli[1], and Giovanni Vinai[1]*

[1] *Instituto Officina dei Materiali (IOM)-CNR, Area Sci. Park, S.S.14, km 163.5, 34149, Trieste, Italy*
[2] *Department of Physics, University of Trieste, Via Alfonso Valerio 2, 34127, Trieste, Italy*
[3] *Department of Physics, University of Milan, Milano I, 20133, Milan, Italy*
[4] *Istituto dei Sistemi Complessi (ISC)-CNR, Piazzale Aldo Moro 5, 00185, Rome, Italy*
*dagur@iom.cnr.it

During the last years, multiferroic (MF) heterostructures have been extensively studied because of the possibility of playing with both ferromagnetic (FM) (break in time-reversal symmetry) and ferroelectric (FE) (break in space-inversion symmetry) degrees of freedom via direct and converse magnetoelectric (CME) coupling effects [1,2]. This offers a wealth of opportunities for technological implementations, specifically, in case of CME i.e., modifying the FM response with an external stimulus, for instance an applied bias [3] or via fully optical means [4]. Despite of comprehensive research efforts in this field, a wide number of parameters playing a role in both FM and FE components of the heterostructure, and how they induce modifications on their counterpart, leaves room for further implementations, exploiting characteristics up to now neglected.

In this framework, here we report the possibility of tailoring the magnetic properties of a FM/FE heterostructure by thermal annealing treatments, having deposited Fe thin films on (1-x)$Pb(Mg_{1/3}Nb_{2/3})O_3$-x$PbTiO_3$ (PMN-PT, x = 40) FE substrate, by exploiting the structural transitions of the latter. FE structural modifications were observed via X-ray diffraction (XRD), showing an increase of in-plane FE domains after annealing above PMN-PT first order phase transition (Fig. 1a). Magneto-Optic Kerr effect (MOKE) and X-ray Circular Dichroism (XMCD) measurements revealed how these FE structural modifications affect the magnetic anisotropy of Fe thin film. While as-grown sample displayed a FM isotropic response, thermally induced FE structural changes led to anisotropies after annealing (Fig. 1b,c), in combination with modifications of Fe dichroic signal. A detailed analysis of the structural modifications taking place in the complex PMN-PT morphotropic regime helped understanding the origin of such changes.

This work paves the way to further explore FE thermal treatments as an additional lever in tuning and maximizing CME coupling in MF heterostructures.

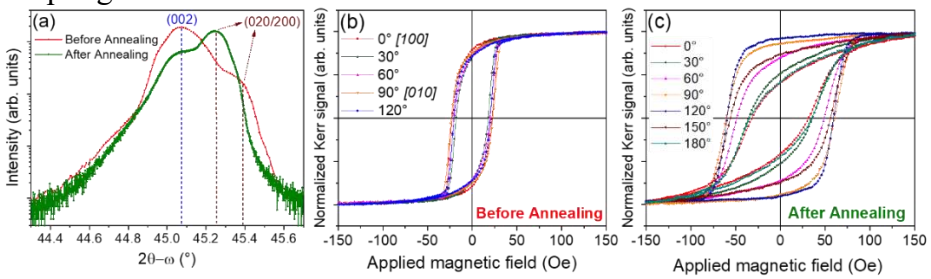



Figure1: a) XRD 2θ-ω scan, b,c) MOKE before (b) and after (c) annealing of PMN-PT/Fe heterostructure.

## Acknowledgements

This work was performed in the framework of the Nanoscience Foundry and Fine analysis (NFFA-MUR Italy Progetti Internazionali) project (www.trieste.NFFA.eu).

## References

[1] W. Erenstein, N.D. Mathur, J.F. Scott, *Nature* **442** (2006) 759.
[2] G. Vinai, F. Motti, V. Bonanni, A. Yu. Petrov, S. Benedetti, C. Rinaldi, M. Stella, D. Cassese, S. Prato, M. Cantoni, G. Rossi, G. Panaccione, and P. Torelli, *Adv. Electron. Mater.* **5** (2019) 1900150.
[3] D. Dagur, V. Polewczyk, A. Yu. Petrov, P. Carrara, M. Brioschi, S. Fiori, R. Cucini, G. Rossi, G. Panaccione, P. Torelli, and G. Vinai, *Adv. Mater. Interfaces* **9** (2022) 2201337.

# Atomic and Magnetic Ordering in Three Varieties of the Heusler Alloy $V_2FeAl$

*Ross Smith[1]*, Rui Zhang[1], Zsolt Gercsi[1], Karsten Rode[1], J.M.D. Coey[1]*

*[1] School of Physics & CRANN, Trinity College Dublin, Dublin 2, Ireland*

*rsmith1@tcd.ie

Magnetic Heusler alloys have been a topic of growing interest in recent years, specifically due to their potential applications in the field of spintronics. These materials, which have the form $X_2YZ$, where X and Y are transition metals and Z is a lighter element, consist of four interpenetrating face-centered cubic lattices. The Heusler alloy $V_2FeAl$ has not before been studied experimentally, but a number of different theoretical papers have been published, predicting several structural and magnetic configurations [1][2][3]. The two possible fully-ordered crystal structures of $V_2FeAl$, $L2_1$ and XA-type, are shown in the figure below. We have grown $V_2FeAl$ both in bulk and in thin-film form. Arc-melted ingots were found to crystallise in the partially-disordered B2 structure. Thin-film samples were prepared by magnetron-sputtering from high-purity V and FeAl targets onto single-crystal MgO (001) substrates. Analysis of X-ray diffraction patterns showed that there two distinct structures were being formed with differing integrated peak intensity ratios of the (002) to (004) reflections. Comparing these ratios to those predicted by structure factors, we associate these two structures with the $L2_1$ and XA-types. SQUID magnetometry shows that the bulk ingot is a Pauli paramagnet, whereas both types of thin-films were surprisingly found to be magnetically-ordered at room temperature. The $L2_1$ structured films show saturation magnetizations of approximately 0.9 $\mu_B$/f.u. and 1.1 $\mu_B$/f.u. at 300K and 5K respectively, whereas the XA-type films showed magnetizations of approximately 0.10 $\mu_B$/f.u. and 0.15 $\mu_B$/f.u. at 300K and 5K respectively. It is evident that disorder has an important effect on the magnetic properties of these systems, which we relate to the electrical transport properties of thin films.

## Acknowledgements

Research was funded through the ZEMS contract, Science Foundation Ireland, grant 16/IA/4534.

## References

[1] R. Watson, M. Weinert, *Phys. Rev. B.* **58** (1998) 5981

[2] M. Kumar, T. Nautiyal, S. Auluck, *J. Phys.: Condens. Matter* **21** (2009) 4460

[3] S. Skaftouros, K. Özdoan, E. Şaşıoğlu, I. Galanakis, *Phys. Rev. B* **87** (2013) 024420

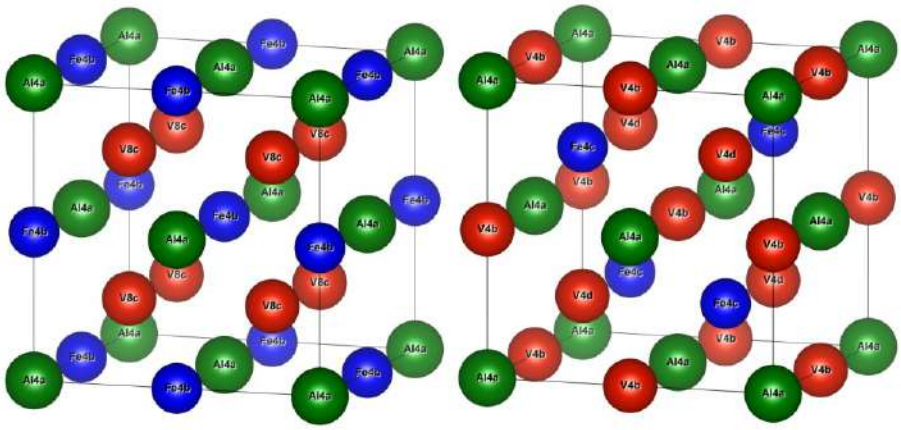


**Figure:** Possible fully-ordered crystal structure of V2FeAl; L21 (left) and XA-type (right).

# Defect-Mediated Interface Intermixing And Its Impact On Perpendicular Magnetic Anisotropy In Cobalt Based Alloys

*Anmol Mahendra[1,2,3]*, Peter P. Murmu[2], Susant Acharya[1,3], Atif Islam[1,3] Holger Fiedler[2] Prasanth Gupta[2,3] Simon Granville[1,3] John Kennedy[2,3]*

[1] *Robinson Research Institute, Victoria University of Wellington, Wellington, New Zealand.*
[2] *National Isotope Centre, GNS Science, PO Box 30368, Lower Hutt, New Zealand.*
[3] *The MacDiarmid Institute for Advanced Materials and Nanotechnology, Wellington, New Zealand.*
*Anmol.Mahendra@vuw.ac.nz

Mangetic multilayers consisting of Cobalt-based magnetic materials such as cobalt-iron-boron (CoFeB) and the full Heusler alloy of cobalt-manganese-gallium ($Co_2MnGa$) have excellent magnetic properties and high perpendicular magnetic anisotropy (PMA) at low film thicknesses. These characteristics make them ideal for integrating in magnetic tunnel junctions (MTJs) based devices for various technological applications such as magnetic sensors, storage devices, and read heads [1,2]. However, fabricating MTJs with specific characteristics can be challenging due to the high dependence of the magnetic properties on individual film thicknesses and interfaces. To address this challenge, we investigate the effect of Argon and Neon ion irradiation on the PMA of CoFeB and $Co_2MnGa$ Heusler alloy. Ion irradiation is a versatile post-growth modification tool that induces structural disorders in the material's lattice that enables altering the magnetic properties by varying fluence, energy, and ion current density of the beam [3,4]. The ion irradiation introduces several structural defects in the lattice which allows tunning of interface mixing and magnetic properties of materials that effect PMA in the stack [5].

In this study, we use a low-energy ion implanter to irradiate the materials with 30 keV $^{20}Ne^+$ and $^{40}Ar^+$ beams at normal incidence and ambient temperature [6]. The irradiation fluence ranges from $1\times10^{13}$ to $1\times10^{15}$ ions.cm$^{-2}$. We perform Monte Carlo based simulations using SRIM and SDTRIMSP to obtain depth profiles and estimate the displacements per atom (DPA) caused by the irradiation.Our results show that ion irradiation reduces the magnetic layer's anisotropy in the out-of-plane direction for both CoFeB and $Co_2MnGa$ stacks. We observe a 30% decrease in $Co_2MnGa$ thin films' anisotropy on Ar+ irradiation at a low fluence of $10^{13}$ ions.cm$^{-2}$, attributed to $Co_2MnGa$/MgO interface mixing. A similar trend is observed in ion-irradiated CoFeB films, and simulations confirm the effect of interfacial intermixing. We also demonstrate that the enhanced PMA of $Co_2MnGa$ can effectively be tuned using ion irradiation, making it a promising candidate for MTJ applications. Our results suggest that the DPA provides a very good estimate of interface intermixing. We will present detailed experimental results obtained from MOKE and SQUID magnetometer, supported by TRIM and SDTRIMSP simulations, at the conference.

## References

[1] Ikeda S, Miura K, Yamamoto H, Mizunuma K, Gan H D, Endo M, Kanai S, Hayakawa J, Matsukura F and Ohno H 2010 A perpendicular-anisotropy CoFeB–MgO magnetic tunnel junction *Nat Mater* **9** 721–4

[2] Ludbrook B M, Ruck B J and Granville S 2017 Perpendicular magnetic anisotropy in Co2MnGa and its anomalous Hall effect *Appl Phys Lett* **110** 062408

[3] Fassbender J, Ravelosona D and Samson Y 2004 Tailoring magnetism by light-ion irradiation *J Phys D Appl Phys* **37** R179–96

[4] Fassbender J and McCord J 2008 Magnetic patterning by means of ion irradiation and implantation *J Magn Magn Mater* **320** 579–96

[5] Mahendra A, Gupta P, Granville S and Kennedy J 2022 Tailoring of magnetic anisotropy by ion irradiation for magnetic tunnel junction sensors *J Alloys Compd* **910** 164902

[6] Kennedy J V., Trompetter W J, Murmu P P, Leveneur J, Gupta P, Fiedler H, Fang F, Futter J and Purcell C 2021 Evolution of Rutherford's ion beam science to applied research activities at GNS Science *J R Soc N Z* **51** 574–91

# Magnetic Domain Evolution in Ferrimagnetic Gd/Fe Alloys and Bilayers through the Curie Temperature

*A. González-García[1]*, J. Obando-Guevara[1,2], A. Locatelli[3], T. O. Mentes[3], M. Jugovac[3], A. Mascaraque[1], M. A. González Barrio[1]*

[1] *Depto. Física de Materiales, Universidad Complutense Madrid, 28034 Madrid, Spain*
[2] *Université Paris-Saclay, CNRS, Laboratoire de Physique des Solides, 91405, Orsay, France.*
[3] *ELETTRA Sincrotrone Trieste S.C.p.A., Basovizza, Trieste 34149, Italy*
*alvaroag@ucm.es

The design of more effective and power-efficient magnetic memories requires the development of methods for magnetization switching driven by currents rather than magnetic fields. In comparison to the more widely used ferromagnets, ferrimagnets have a number of benefits, such as a small net magnetization and ultra-fast magnetization dynamics at the compensation temperature, at which the net magnetization vanishes [1, 2, 3]. In order to employ ferrimagnets in future spintronic devices, a thorough knowledge of the domain structure and its temperature evolution close to the compensation temperature is crucial.

We have investigated the temperature evolution of the magnetic domains of different ferrimagnetic systems: thin Fe/Gd and Gd/Fe bilayers and FeGd alloy ultrathin films grown on W(110). For this purpose, XAS spectra and XMCD images at the Fe $L_{2,3}$-edge and Gd $M_{4,5}$-edge, as well as XPS measurements of the Fe 3*p*, W 4*f* and Gd 4*f* core levels were performed using synchrotron-based PEEM. Based on these results, the chemical composition and the temperature evolution of the magnetic domain structure were determined. We have monitored the magnetization of the Gd and Fe sublattices and the evolution of the magnetic domain structure as a function of temperature across the Curie temperature (see Figure).

## Acknowledgements

We thank the Ministerio de Ciencia e Innovacion PID2020-117024GB-C43 project and ELETTRA User Travel program for financial support.

## References

[1] T. Jungwirth*et al.Nat. Nanotech.***11** (2016) 231
[2] N. Thielemann-Kühn, *et al. Phys. Rev. Lett.***119** (2017) 197202
[3] V. Baltz. *Rev. of Modern Phys.* **90** (2018) 015005

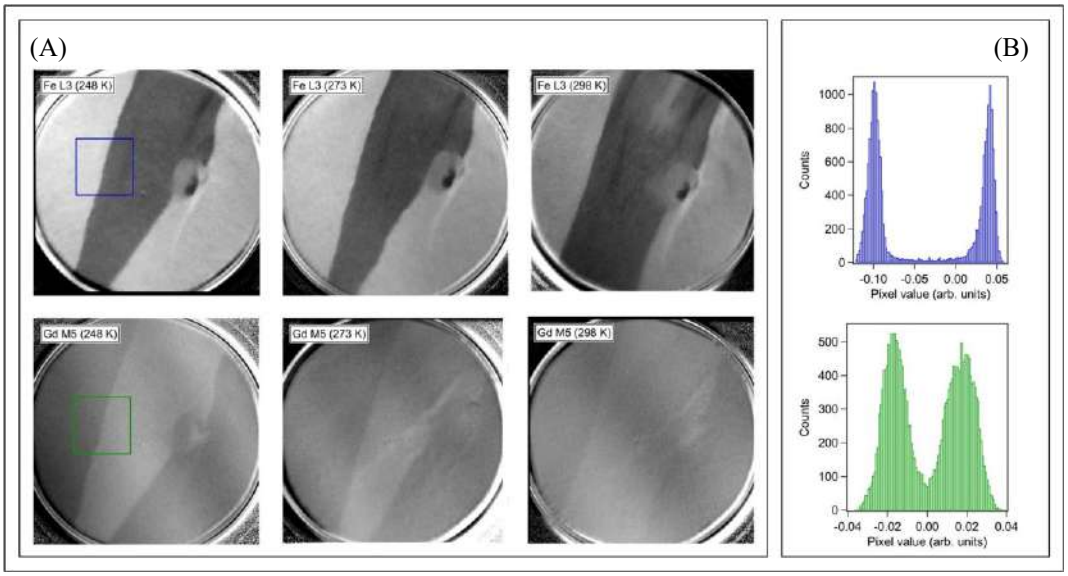



Figure: XMCD measurements of a Fe/Gd bilayer. (A) XMCD images (FOV = 10 μm) of the sample taken at different temperatures at the Fe $L_3$-edge (upper row) and at the Gd $M_5$-edge (lower row). (B) Histograms of the regions highlighted in the images of the previous panel.

# Cooling Stage Method for MBE Growth of *bct* $\alpha'$-$Fe_8N$ Thin Films

*Giovanni d'Andrea[1]*, Taewook Kim[1], Philipp Schreyer[1], Georgia Gkouzia[1], Márton Major[1] and Lambert Alff[1]*

[1] *Technische Universität Darmstadt, Institute for Materials Science, Alarich-Weiss-Str. 2, 64287 Darmstadt, Germany*

*giovanni.dandrea@tu-darmstadt.de

Non-equilibrium methods in the preparation of tetragonally distorted iron proved to be successful for the formation of $\alpha''$-$Fe_{16}N_2$ and $\alpha'$-$Fe_8N$ metastable phases in thin films [1]. For these structures and the analogous in the Fe-B and Fe-C systems [2, 3], the light atoms are found to be in interstitial positions, but it is no easy task to make them stable and to achieve a high enough interstitials solubility to allow the formation of a large volume ratio of these phases [4, 5]. With the prospect of overcoming these limitations for the formation of iron films with high magnetocrystalline anisotropy and increased average magnetic moment of the iron atoms, a cooling stage for a molecular beam epitaxy (MBE) deposition system is employed. This method of decreasing the kinetic energy of the evaporated atoms impinging on cold (down to -150 °C) substrates represents a novel non-equilibrium approach in the preparation of this material.

Preliminary work on an MBE deposition system provided us with reliable benchmark. The 30 nm thick Fe films grown on MgO (100) substrates have a strong preferential orientation, and their saturation magnetization ($\mu_0 M_S$) value is in agreement to the well-known 2.15 T of iron [6]. To validate and compare our results for the new approach, ion-implantation will be used as a secondary non-equilibrium method. For samples prepared under optimised conditions, different annealing procedures will be tested and compared to those employed for the films grown with the novel approach. This comparison will allow for a deeper understanding of the material synthesised on the cooling stage and to confirm the effectiveness of this method.

## Acknowledgements

We thank the Deutsche Forschungsgemeinschaft (DFG) for funding our research, as part of the CRC 1487 "Iron, upgraded!" project (nr. 443703006). Ion implantation experiments will be performed at the Ion Beam Center (IBC) at the Helmholtz Zentrum Dresden Rossendorf (HZDR).

## References

[1] P. Gupta, H. Fielder, S. Rubanov and J. Kennedy, *J. Magn. Magn. Mat.* **517** (2021) 167388.
[2] D. Gölden, H. Zhang, I. Radulov, I. Dirba, P. Komissinskiy, E. Hildebrandt and L. Alff, *Phys. Rev. B* **97** (2018) 014411.
[3] H. Ohtsuka, V.A. Dinh, T. Ohno, K. Tsuzaki, K. Tsuchiya, R. Sahara, H. Kitazawa and T. Nakamura, *ISIJ Int.* **55.11** (2015) 2483.
[4] T. Weber, L. de Wit, F.W. Saris, P. Schaaf, *Thin Solid Films* **249** (1996) 216.
[5] M. Widenmeyer, T.C. Hansen, and R. Niewa, *Z. Anorg. Allg. Chem* **639.15** (2013) 2851.
[6] J.M.D. Coey, Magnetism and magnetic materials, *Cambridge university press* (2010) p.66.

# SmCo-based permanent magnets processed by severe plastic deformatilon

*A. Paulischin[1]*, L. Weissitsch[1], S. Wurster[1], H. Krenn[2], R. Pippan[1], A. Bachmaier[1]*

[1] *Erich Schmid Institute of Materials Science of the Austrian Academy of Sciences, Leoben, Austria*

[2] *Institute of Physics, University of Graz, Graz, Austria*

* alexander.paulischin@oeaw.ac.at

The continually increasing global energy consumption and the growing awareness for sustainability and renerwable energy resources are accompanied with a rising demand for high performance rare earth based permanent magnets (REPMs). State of the art REPMs are based on combinations of rare-earth and transition metals. Especially for applications under severe working conditions, REPMs based on Sm and Co possess remarkable performance characteristics due to their high Curie temperature and thus their elevated maximum operating temperature as well as an enhanced corrosion resistance. Conventionally, these SmCo-based REPMs are manufactured using powder metallurgical techniques, which include manufacturing steps like powder refinement via milling, green compact formation with simultaneous magnetic field alignment as well as subsequent sintering and annealing processes. [1, 2]

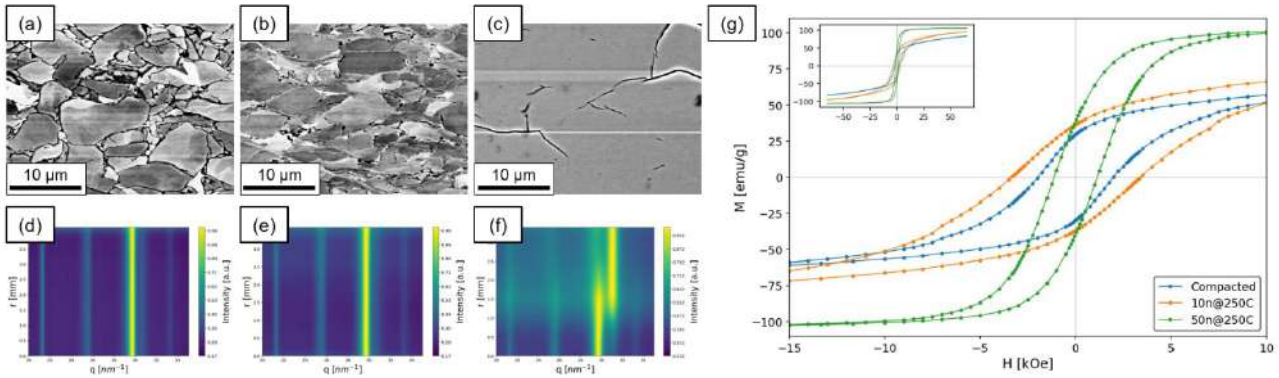



Figure 1: SEM images of the formed microstructure after (a) powder compaction (b) 10 rotations and (c) 50 rotations at 250°C as well as their corresponding X-ray patterns ((d) – (f)). (g) The formed microstructure has a massive influence on the achieved magnetic properties.

A novel approach to manufacture REPMs based on intermetallic SmCo-phases (e.g. $Sm_2Co_{17}$, $SmCo_5$) is presented in this study by using high pressure torsion (HPT), a method of severe plastic deformation. Processing by HPT possesses the opportunity of simultaneous powder compaction, grain refinement, formation of metastable phases as well as strain induced phase transformation just in a single processing step. Herein, we describe the evolution of the emerging microstructures due to HPT-deformation. For the microstructural characterization of the HPT-processed specimens, scanning electron microscopy (SEM) is used, while strain induced phase transformations are monitored by synchrotron high energy X-ray scattering techniques. The chosen deformation parameters (e.g. deformation temperature, applied strain) have a strong influence on the formed microstructure (Fig 1 (a) – (c)) and furthermore on the present phases after deformation (Fig. 1 (d) – (f)). Grain refinements down to the nanometer range and concurrent phase transformations have a severe impact on the achievable magnetic properties, which are determined by SQUID magnetometry (Fig. 1 (g)). Subsequent annealing treatments further influence microstructural features. Beside the reduction of defect density, a grain boundary tuning is apparent, fostering the magnetic properties of intermetallic SmCo-phases.

## Acknowledgements

This project has received funding from the European Research Council (ERC) under the European Union´s Horizon 2020 research and innovation programme (Grant No. 757333).

## References

[1] S. Liu, Chin. Phys. B Vol. 28, No. 1, 2019, 017501

[2] J. Cui, JOM 74, 2022, 1279 - 1295

# Strong interfacial pinning in Ni-Mn-based $L2_1$ Heusler films sandwiched between antiferromagnetic $L1_0$ NiMn

*A. Çakır[1]*, I. Sahin[2], F. Dreist[2], B. Gündogdu[2], M. Farle[2], M. Acet[2]*

[1]*Department of Metallurgical and Materials Engineering, Muğla Sıtkı Koçman University, 48000 Muğla, Turkey*

[2]*Faculty of Physics and Center for Nanointegration (CENIDE), Universität Duisburg-Essen, D-47048 Duisburg, Germany*

*cakir@mu.edu.tr

$Ni_{50}Mn_{45}X_5$ (*X*: Al, Ga, In Sn, Sb), when annealed around 650 - 750 K, decomposes and results in the formation of 2-5 nm full-Heusler cubic ferromagnetic $Ni_{50}Mn_{25}X_{25}$ precipitates embedded coherently in an antiferromagnetic tetragonal $Ni_{50}Mn_{50}$ matrix. When the annealing takes place in a magnetic-field, the spins at the interface become strongly pinned in the direction of the annealing field and give rise to the mono-polar magnetic behavior of shell-ferromagnetic effect. The precipitates acquire a coercivity exceeding 15 T and they comprise at most about 10 % of the material so that the overall magnetization remains low, at most about 8 $Am^2kg^{-1}$. Instead of dealing with isolated precipitates in a matrix, we can sandwich a 5-nm $Ni_{50}Mn_{25}Sn_{25}$ film between $Ni_{50}Mn_{50}$ layers so as to have spread the 'precipitate' over two dimensions as a film in order to increase the size of the magnetization. We prepare $Ni_{50}Mn_{50}/Ni_{50}Mn_{25}Sn_{25}/Ni_{50}Mn_{50}$ multilayered films by flash evaporation. Comparison of the magnetic properties of as-prepared and field-annealed multilayered films show that strong interfacial pinning can also be attained in the field-annealed case. The initially incoherent interface acquires coherence due to the stabilized structures of the Heusler and $Ni_{50}Mn_{50}$ layers, and the spins at the interface become strongly pinned in the direction of the applied magnetic field leading to a quasi-two-dimensional magnet.

**Acknowledgements**

Work supported by Deutsche Forschungsgemeinschaft (Project 405553726-CRC/TRR 270) and TUBITAK Science Fellowships and Grant Programs (Grant 1059B192000916).

# Superposed Shape Memory and Magnetocaloric Effects in Ferromagnetic Intermetallic Compound $Ni_{52}Fe_{20}Ga_{23}Co_3Al_2$

*F. Ţolea[1]*, M. Sofronie[1], B.Popescu[1], M. Ţolea[1], M. Enculescu[1]*

[1] *National Institute of Materials Physics, POB MG-7, 77125 Bucharest-Măgurele, România*

*felicia.tolea@infim.ro

The functionality of ferromagnetic shape memory alloys (FSMA) is related to the martensitic and magnetic order-disorder transformations; both, the martensitic ($T_M$) and Curie (Tc) temperatures, may be tailored by doping the alloys with other elements (to change the electronic concentration, e/a) or by suitable thermal treatments (to change the atomic order). So that, alloys with concomitant or sequential structural and magnetic phase transition may be obtained. These are accompanied by sharp change of magnetization as well as electronic structure, which provide the attractive multifunctional potential for both fundamental science and technological applications of these class of materials. The Magnetocaloric Effect around room temperature is an environmentally friendly alternative to conventional refrigeration and the most significant effect occurs at temperatures near magnetic transitions, and strongly depends on the type of transition being highest for first-order transitions [1, 2]. Recent studies have shown that the entropy change associated to the martensitic transformation in FSMA increase as the temperature difference between Tc and $T_M$ decreases [3, 4].

In the present work we investigate the martensitic transformation and magnetic properties of Heusler intermetallic compound $Ni_{52}Fe_{20}Ga_{23}Co_3Al_2$ prepared as melt spun ribbons subjected to different thermal treatments. X-ray diffraction, differential scanning calorimetry, magnetocaloric and magnetoresistive effects evaluations have been performed. The results highlight the role of concomitant or sequential structural and magnetic phase transitions.

**Acknowledgements**

This work was supported by project 35PFE/2021, funded by the Romanian Ministry of Research, Innovation and Digitization.

**References**

[1] V. K. Pecharsky, K. A. Gschneidner Jr., *Phys. Rev. Lett.* **78**, (1997) 4494.

[2] V. Franco, J.S. Blázquez, J.J. Ipus, J.Y. Law, L.M. Moreno-Ramírez, A. Conde, *Prog. Mater. Sci.* **93** (2018) 112–232.

[3] Recarte V, Pérez-Landazábal J I, Gómez-Polo C, Cesari E, Dutkiewicz J, *Appl. Phys. Lett.* **88** (2006) 132503.

[4] A. Planes, L.Mañosa, M. Acet, *J. Phys.: Condens. Matter* **21** (2009) 233201. (29pp).

# Studying strain effects on the magnetism of iron oxide thin films on $BaTiO_3$ single crystal substrates.

*Darla Mare*[1]*, *Nico Dix*[1], *Martí Gich*[1]

[1] *Institut de Ciència de Materials de Barcelona (ICMAB-CSIC), Bellaterra 08193, Barcelona, Spain.*

*dmare@icmab.es

Stabilizing complex ferroic oxides on piezoelectric substrates is appealing from a fundamental point of view to study the mutual control of electric and magnetic properties (magnetoelectric effect). It also opens the possibility of indirectly probing the influence of strain on magnetic films at structural phase transitions of the substrate. This has been mainly explored in the case of barium titanate ($BaTiO_3$) [1,2,3]. In this system, the single crystal substrates act as the active mechanical part during the transition and deform the magnetic films thanks to their tight chemical bonding, making it an effective tool for determining mechanical coupling.

Here we report our investigation on the strain-induced magnetic response in two iron oxides, cubic magnetite ($Fe_3O_4$, *Fd-3m*) [3] and metastable orthorhombic epsilon iron oxide (ε-$Fe_2O_3$, *Pna2₁*) [4]. The magnetic properties of these iron oxides are quite contrasting due to their respective crystal structures. While magnetite presents a cubic anisotropy with easy magnetization along the <111> crystal directions, the latter has a substantial uniaxial anisotropy along the [100] axis. Both phases were epitaxially stabilized by pulsed laser deposition on $BaTiO_3$ (111) substrates. The spinel phase grows with (*LLL*) texture, while epsilon iron oxide has out-of-plane (00*L*) texture, together with the formation of three distinct in-plane domains similar to the films grown on $SrTiO_3$ (111) substrates [5]. The magnetic properties of the thin films were studied with a SQUID magnetometer. We analyzed M(H) hysteresis loops to determine saturation magnetization, coercivity, and remanent magnetization for two perpendicular in-plane substrate directions ([1-10] and [11-2]) and the out-of-plane direction. The influence of the structural phase transitions of $BaTiO_3$ around 190 K (rhombohedral to orthorhombic) and 270 K (orthorhombic to tetragonal) is revised by a combined M(T) and M(H, T) approach. Epsilon iron oxide only shows a small sizable change of the magnetization at the orthorhombic to the rhombohedral phase transition of $BaTiO_3$ (Figure 1a). In contrast, for the magnetite, abrupt jumps (Figure 1b) are found for both transitions within the studied temperature range, with variations of the magnetization up to 40%. We discuss these differences considering the crystal structure, magnetic domain geometry, and measurement direction. The findings point towards a robust magnetization against deformation of the hard magnetic epsilon iron oxide, despite structural deformations suggesting a rather large magnetostructural response [4]. In contrast, the softer magnetite is more suitable for strain-tuned magneto-electric devices.

**References**

[1] M. Acosta *et al.*, Applied Physics Reviews 4, 041305 (2017).
[2] M . K. Lee *et al.*, Applied Physics Letters 77, 3547 (2000).
[3] C. A. F. Vaz *et al.*, Applied Physics Letters 94, 022504 (2009).
[4] M. Gich *et al.*, Journal of App. Physics 98, 044307 (2005).
[5] M. Gich *et al.*, Advanced Materials 26, 4645 (2014).

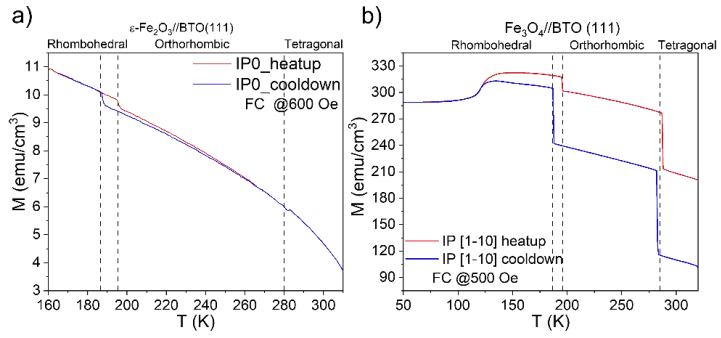



Figure 1: a) Magnetization response of the epsilon iron oxide with temperature dependence while applying an external field of 600 Oe. b) Magnetization response of the magnetite thin film with temperature dependence while applying an external field of 500 Oe.

# Element-Specific Study of Magnetic Anisotropy and Hardening in $SmCo_{5-x}Cu_x$ Thin Films

*Georgia Gkouzia[1]*, Damian Günzing[2], Ruiwen Xie[1], Teresa Weßels[2,3], András Kovács[3], Alpha T. N' Diaye[4], Márton Major[1], J. P. Palakkal[1,5], Rafal E. Dunin-Borkowski[3], Heiko Wende[2], Hongbin Zhang[1], Katharina Ollefs[2], and Lambert Alff[1]*

[1] *Institute of Materials Science, Technical University of Darmstadt, Germany*
[2] *Faculty of Physics and Center for Nanointegration (CENIDE), University of Duisburg-Essen, Germany*
[3] *Ernst Ruska-Centre for Microscopy and Spectroscopy with Electrons and Peter Grünberg Institute, Forschungszentrum Jülich, Germany*
[4] *Lawrence Berkeley National Laboratory, Berkeley, USA*
[5] *Institute of Materials Physics, Georg-August-University of Göttingen, Germany*
*georgia.gkouzia@tu-darmstadt.de

Permanent magnets play a vital role in numerous applications and are crucial for advancing sustainable energy[1]. The most well-known compounds $Nd_2Fe_{14}B$, $SmCo_5$, and $Sm_2Co_{17}$ combine a large energy density, high Curie temperature ($T_C$), and a large magnetocrystalline anisotropy (MCA). $SmCo_5$ magnets are well-known for their exceptional uniaxial magnetic anisotropy ($K_1$=17.2 MJ/m$^3$) and high $(BH)_{max}$ values[2]. In the same family, $Sm_2Co_{17}$-type of alloys are excellent candidates for high-temperature applications due to their strong combination of high $(BH)_{max}$ and $T_C$. As a result of their significance, researchers have turned their attention towards comprehensively understanding the hysteresis behavior of these materials[3].
Thin films of Sm-Co are excellent candidates for creating model structures that enable precise control over the nanostructure, allowing for the investigation of the specific impact of individual defects on magnetic properties. Recently, we have discovered a novel phase decomposition regime in molecular beam epitaxy (MBE) grown thin films, resulting in the coexistence of $SmCo_5$ and $Sm_2Co_{17}$ at the nanoscale, in a range of few nanometers[4].
In the present work, we have investigated how the substitution of copper affects the magnetic properties of $SmCo_5$ thin films synthesized by MBE. A series of thin films with varying concentrations of Cu, were grown on $Al_2O_3$ (0001) substrates and pre-characterized by X-ray diffraction (XRD) and superconducting quantum interference device (SQUID) magnetometer. We have observed that Cu at the $Co_{3g}$ sites not only stabilizes the formation of the $SmCo_5$ structure, but also enhances magnetic anisotropy and coercivity.
Density functional theory (DFT) calculations show that $Sm(Co_4Cu_{3g})_5$ has a higher single-ion anisotropy than pure $SmCo_5$. Furthermore, X-ray magnetic circular dichroism (XMCD) measurement indicates that Cu substitution causes an increasing decoupling of the Sm 4*f* and Co 3*d* moments. Scanning transmission electron microscopy (STEM) confirms that the primary phase formation is $SmCo_5$, and also identifies nanoscale inhomogeneities in the Cu and Co distribution. Through the utilization of advanced characterization and modeling techniques on thin film model systems, this study provides insight into the relationship between the intrinsic and extrinsic factors contributing to magnetic hysteresis in rare earth-based magnets.

## Acknowledgments

We acknowledge the financial support from the Deutsche Forschungsgemeinschaft (DFG) in the framework of the CRC/TRR270 (Project ID. 405553726), projects A02, A03, A05, B05, and Z01/02. JPP acknowledges DFG for the funding under project 429646908. The authors acknowledge Janghyun Jo for experimental help and Lea Risters for FIB sample preparation. This research used resources of the Advanced Light Source, which is a DOE Office of Science User Facility under contract no. DE-AC02-05CH11231.

## References

[1] O. Gutfleisch et al., *Adv. Mater.* **23** (2011) 821.
[2] K. J. Strnat & R. M. W. Strnat, *J. Magn. Magn. Mater.* **36** (1991) 100.
[3] H. Sepehri-Amin, *Acta Mat.* **126** (2017) 1-10.
[4] S. Sharma et al., *ACS Appl. Mater. Interfaces* **13** (2021) 32415.

# Electric field control of magnetization reversal in FeGa/PMN-PT

*G. Pradhan[a], F. Celegato[a], L. Mikuličková[b], A. Magni[a], G. Barrera[a], M. Coïsson[a], P. Tiberto[a]*
*a Istituto Nazionale di Ricerca Metrologica (INRIM), Strada delle Cacce 91, 10135, Torino, Italy*
*b Thin film Technological Service, s.r.o., Prague, Czech Republic*
*p.tiberto@imrim.it

The recent era of information technology devices largely focusses on energy and cost efficiency. Magnetoelectric (ME) materials possess huge potential to be used in these technologies due to their coupling between magnetization and electric-field induced strain which therefore consumes less power and significantly reduces heat losses [1,2]. The application of voltage generates strain at the interface and this strain transfer to the magnetic layer from the piezoelectric layer induces change in magnetic anisotropy which has been shown to be important in switching patterned structures from a multi-domain magnetized state to a flux closure vortex one [3].

In the framework of multiferroic heterostructures and their interfacial magnetoelectric interplay, here we propose to investigate the properties of FeGa/PMN-PT heterostructures using magnetometers and magnetic imaging techniques. FeGa thin films have been deposited by sputtering on PMN-PT substrates. The study of magnetic domains using Magneto-Optic Kerr effect (MOKE) revealed the presence of in-plane magnetic domains in the samples. Under the application of voltage, a change in magnetic hysteresis shape is observed as represented in Figure 1. The hysteresis curve at the tensile state (pink curve) has the highest magnetic remanence (*Mr*) and coercive field (*Hc*). The corresponding domain images with domain walls pointing along x, are shown in Figure 1(b). The hysteresis has the lowest *Mr* and *Hc* at the compressive state (orange curve) and new the magnetic domain walls starts appearing at an angle of 45° w.r.t x. Further, when the field is decreased towards negative polarization saturation (-10 kV/cm), the strain decreases again generating a tensile strain along x. The *Mr* and *Hc* values increases and the domain structures shown in Figure 1(e) become similar to ones recorded at 10 kV/cm. In order to better understand the effect of applying a voltage to PMT-PT substrate on the overall magnetisation process, the time evolution of remanent magnetization has been acquired by a vibrating sample magnetometer (VSM) after applying an electrical field of 10 kV/cm.

## Acknowledgements

This work has received funding from the European Union's Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement No 861145.

## References

[1] R. Lo Conte, et al. Nano letters 18.3, 1952-1961 (2018)
[2] Q. Li, et al. Applied Physics Letters 110.26, 262405 (2017)
[3] M.G. De Jesus, et al. Smart Materials and Structures 31.3, 035005 (2022)

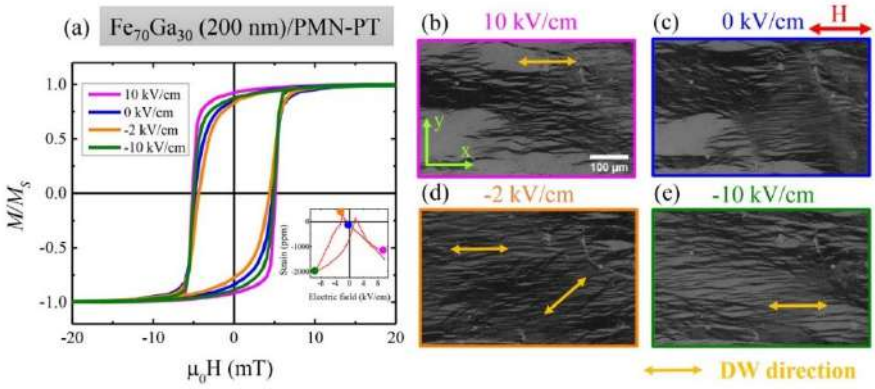



Figure 1: (a) represents the in-plane magnetic hysteresis loop for $Fe_{70}Ga_{30}$/PMN-PT (001) sample. The pink, blue, orange and green curves represent to the hysteresis recorded under applied field of 10 kV/cm, 0 kV/cm, -2 kV/cm and -10 kV/cm, respectively. The plot of strain as a function of electric field is shown in the inset. The magnetic domains near coercivity for the corresponding curves are shown by (b), (c), (d) and (e), respectively.

# Tuning Physical Properties of $La_{0.7}Sr_{0.3}MnO_{3-\delta}$ via Oxygen Off-Stoichiometry Using Thermal Annealing and Ionic Liquid Gating

*C. Yin [1]*, L. Cao[1], S. He[2], T. Duchon[3], Y. Zhou[4], E. Kentzinger[1], Th. Brückel[1], O. Petracic[1]*

[1] *Jülich Centre for Neutron Science (JCNS-2) and Peter Grünberg Institut (PGI-4), JARA-FIT, Forschungszentrum Jülich GmbH, Jülich, Germany*

[2] *Peter Grünberg Institut (PGI-7), JARA-FIT, Forschungszentrum Jülich GmbH, Jülich, Germany*

[3] *Peter Grünberg Institut (PGI-6), JARA-FIT, Forschungszentrum Jülich GmbH, Jülich, Germany*

[4] *Helmholtz-Zentrum Dresden-Rossendorf (HZDR), Dresden, Germany*

*c.yin@fz-juelich.de

In complex oxides, controlled oxygen release/uptake can induce changes of the crystal structure and of the magnetic and electrical properties [1-2]. Consequently, a systematic control of their oxygen stoichiometry might enable potential applications in information technology, solid oxide fuel cells and catalysts. Previously, the oxygen off-stoichiometry in $La_{0.7}Sr_{0.3}MnO_{3-\delta}$ (LSMO) thin films on $SrTiO_3$ (STO) substrates has been investigated employing Al-assisted vacuum annealing. The gradual deoxygenation during annealing induces a topotactic phase transition from the as-prepared Perovskite (PV, $ABO_3$) phase to a layered oxygen-vacancy-ordered Brownmillerite (BM, $ABO_{2.5}$) phase (Figure 1 (a)). Recently, we employed ionic liquid gating (ILG) and realized a ferromagnetic to antiferromagnetic transition (Figure 1 (e)). The "plateau"-like feature in XRD and the S-shape MH curve indicate the existence of subsystems after gating (Figure 1 (c) (d)). After gating at higher voltages, an additional hydrogen content inserted into the lattice with an atomic percent of ca. 30% is evidenced (Figure 1(f)). Our results demonstrate the tuning of film physical properties via a dual ionic transfer process by ILG, i.e. oxygen vacancy formation and hydrogen insertion.

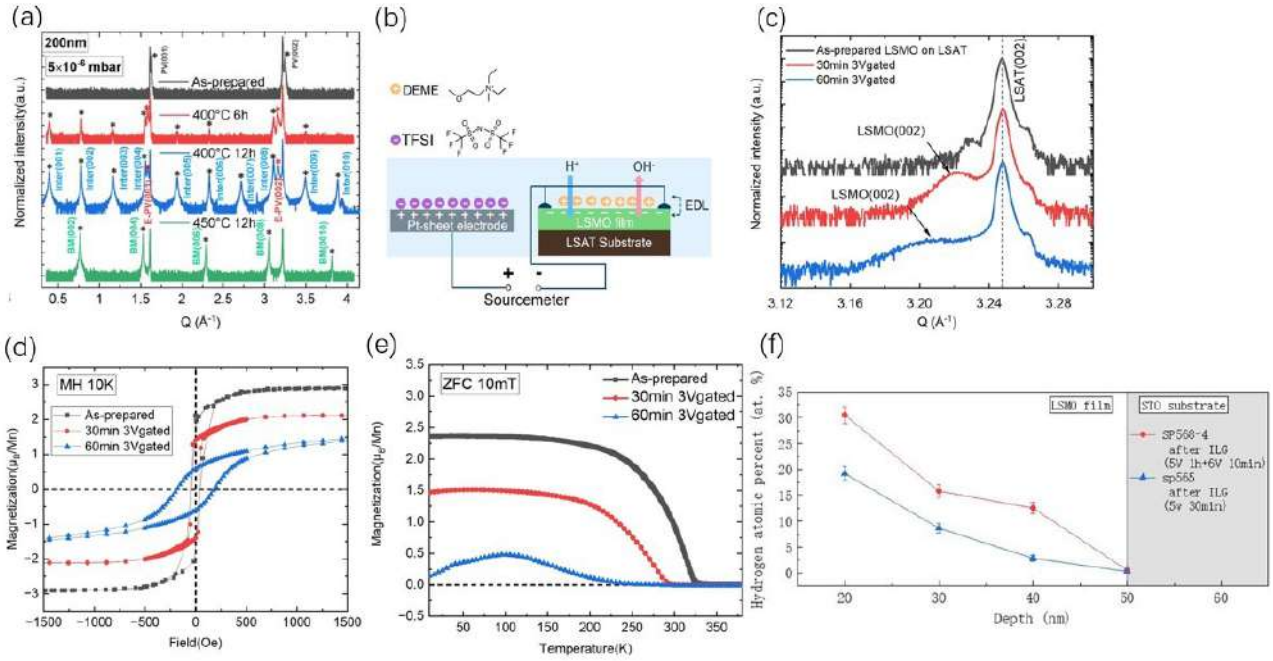



Figure 1: (a) XRD pattern of the 200nm as-prepared and annealed LSMO films. The PV phase under expansion is defined as "E-PV" phase. The newly emerged intermediate phase is labeled as "inter". (b) Schematics of the ILG process. (c) XRD pattern of the 40nm as-prepared and ILG-treated LSMO films. (d) Hysteresis loops of the as-prepared and ILG-treated LSMO films, measured at 10K. (e) Magnetization vs. temperature zero-field cooling (ZFC) curves of the as-prepared and ILG-treated LSMO films, measured at 10mT. (f) Hydrogen content depth profiles of ILG-treated LSMO films, measured by nuclear reaction analysis.

## References

[1] Cao L, Petracic O, et al., Advanced Materials, 2019.31(7): 1806183.

[2] Cao, L., Petracic, O., et al., Small, 2021.17(51): 2104356.

# Spin rectification by planar Hall effect in synthetic antiferromagnets

*D. E. Gonzalez-Chavez[1], M. A. Pervez[1], J. E. Gómez[2,3], A. Butera[2,3], L. Avilés-Félix[2,3,*], and R. L. Sommer[1]*

[1] *Centro Brasileiro de Pesquisas Físicas, 22290-180 Rio de Janeiro, RJ, Brazil*

[2] *Instituto de Nanociencia y Nanotecnología (CNEA-CONICET), Nodo Bariloche, Av. Bustillo 9500, (8400) Bariloche (RN), Argentina*

[3] *Laboratorio Resonancias Magnéticas, Centro Atómico Bariloche, Av. Bustillo 9500, (8400) Bariloche (RN), Argentina*

*Corresponding author: luis.aviles@ib.edu.ar

Electrically detected ferromagnetic resonance signals are originated by the radiofrequency electric field, the spin currents induced by the excitation fields, and their interaction with the ferromagnetic material or adjacent nonmagnetic layers. In this work, we study spin rectification effects in NiFe/Ru/NiFe/NM synthetic antiferromagnets at the saturated, non-collinear and antiparallel magnetic states. The change in the magnetization orientation at these states allows us to study the angle dependence of spin rectification phenomena by only changing the applied field [1]. This approach avoids the need of changing the sample orientation within the experiment for this kind of studies. Our results show large planar Hall effect contributions to the measured spin rectified voltage with interesting features in the non-collinear and antiparallel states, which bring new possibilities for device design and applications [2]. The results improve the understanding of electrically detected ferromagnetic resonance signals, not only for synthetic antiferromagnets, but also for other samples where the signals arising from the anisotropic magneto resistance effect and the inverse spin Hall effect are superimposed.

## Acknowledgements

This work was supported by the Brazilian agencies CNPq, FINEP, FAPERJ and CAPES. M. Asmat Pervez would like to acknowledge the Higher Education Commission (HEC) Pakistan, and Institute of Space Technology (IST) Pakistan for providing his fellowship. Technical support from Rubén E. Benavides, César Pérez, and Matías Guillén is greatly acknowledged. This work was partially supported by Conicet, Argentina under Grant PIP 201501-00213, ANPCyT, Argentina Grant PICT 2018-01138, and U.N. Cuyo Grant 06/C556, all from Argentina.

## References

[1] M. A. Pervez, D. Gonzalez-Chavez, R. Dutra, B. Silva, S. Raza, R. Sommer, *J. Magn. Magn. Mater.* **548** (2022) 168923 doi: https://doi.org/10.1016/j.jmmm.2021.168923

[2] D. E. González Chávez, M. Asmat Pervez, L. Avilés-Félix, J. E. Gómez, A. Butera and R. L. Sommer, *J. Magn. Magn. Mater.* **560** (2022) 169614 doi: https://doi.org/10.1016/j.jmmm.2022.169614

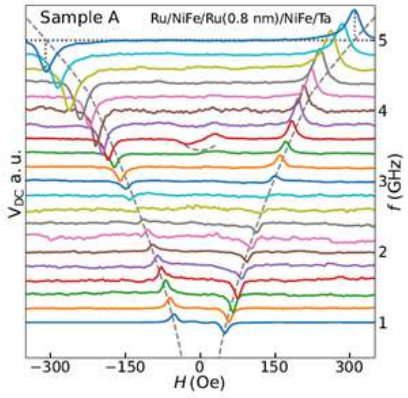



Figure 1: Measured DC voltages vs Magnetic field, along all the magnetization states, for a Ru/NiFe/Ru(0.8 nm)/NiFe/Ta. The dashed lines correspond to the dispersion relations of the acoustic FMR mode.

# Coherent Magnetization Reversal Promoted by Magnetoelastic Coupling

*N. Cotón[1], J. P. Andrés[2], A. Cabrera[1], M. Maicas[3] and R. Ranchal[1,4]*

[1] *Dpt. Física de Materiales, Fac. CC. Físicas. Universidad Complutense de Madrid. Plaza de las Ciencias 1, Madrid 28040, Spain*

[2] *Univ. Castilla La Mancha, Inst. Reg. Invest. Cient. Aplicada IRICA, Dept. Appl. Phys., Ciudad Real 13071, Spain*

[3]*Institute for Optoelectronics and Microtechnologies, Polythecnic University of Madrid, Madrid, Spain*

[4]*Instituto de Magnetismo Aplicado, UCM-ADIF-CSIC, Las Rozas 28232, Spain*

*rociran@ucm.es

One of the most active and successful investigation field is the development of innovative multilayers exploiting interfacial interactions [1,2]. Typically, for thicknesses above a critical value there appear uncoupled regions that independently switch their magnetization. Nevertheless, for applications such as exchange-biased systems or spring magnets it is needed a coherent magnetization reversal. Although the exact value of the critical thickness depends on the considered magnetic interaction, fundamental magnetic parameters as the exchange correlation length, and the magnetic anisotropy play an important role. In this work, we show how in a magnetostrictive bilayer structure comprised of two materials with magnetostriction constants of opposite sign, the magnetic switching is affected by the magnetoelastic coupling. Since magnetoelasticity is not an interfacial interaction, the mechanical strain promoted by the applied magnetic field affects the layers as a whole. The net effect is a coherent reversal of the magnetization of the two layers regardless of their thickness. This behavior has been studied in bilayers with different thicknesses ratio comprised of $Ni_{90}Fe_{10}$, negative magnetostriction, and $Fe_{70}Ga_{30}$ positive magnetostriction (Figure 1). These results demonstrate the possibility of using this physical mechanism to beat the critical limit thickness imposed in other interfacially coupled magnetic systems opening new ways to develop applications in the fields of energy or spintronics.

## Acknowledgements

This work has been financially supported through the projects PID2021-122980OA-C53 (FEDER) and CPP2021-008397 of the Spanish Ministry of Science and Innovation.

## References

[1] R. Bläsing, T. Ma, S.-H. Yang, C. Garg, F. K. Dejene, A. T. N'Diaye, G. Chen, K. Liu, S. S.P. Parkin, *Nat. Comm.* **9** (2018) 4984.

[2] Z. J. Guo, J.S. Jiang, J.E. Pearson, S.D. Bader, J.P. Liu, *Appl. Phys. Lett.* **81** (2002) 2029.

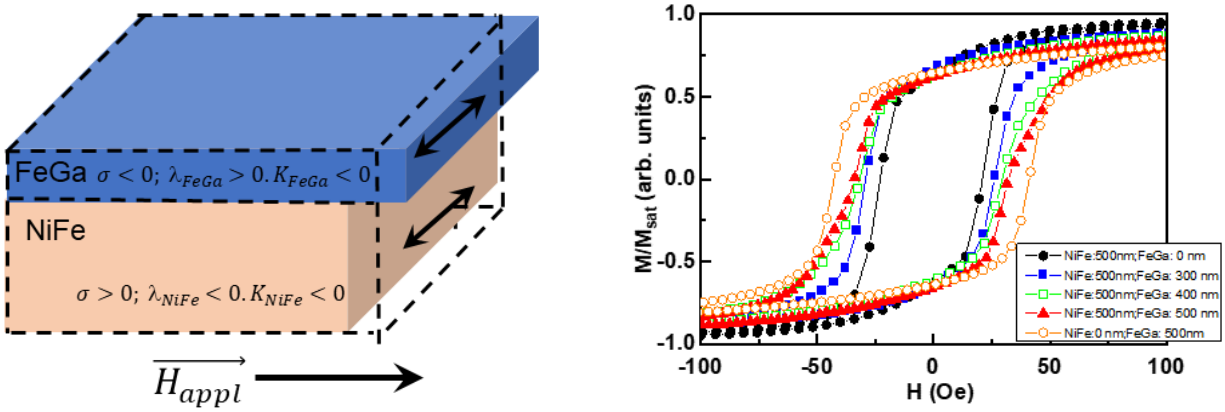



Figure 1. Left: Schematic view of a NiFe/Fe bilayer showing the expected mechanical deformation due to the magnetization process promoted by the applied magnetic field. Right: hysteresis loops for $Ni_{90}Fe_{10}$ and $Fe_{70}Ga_{30}$ single layers together with the hysteresis loops of different NiFe/FeGa bilayers in which the $Ni_{90}Fe_{10}$ is 500 nm and $Fe_{70}Ga_{30}$ varies from 300 nm to 500 nm.

# Influence of Electric Field on the Spin Wave Stiffness and Gilbert Damping in Metallic Layers

*Ondřej Šipr[1]*, Aki Pulkkinen[2], Ján Minár[2], Sergey Mankovsky[3], Hubert Ebert[3], Alberto Marmodoro[1]*

[1] *FZU-Institute of Physics of the Czech Academy of Sciences, Cukrovarnická 10, Prague, Czechia*

[2] *New Technologies Research Center, University of West Bohemia, Pilsen, Czechia*

[3] *Department Chemie, Ludwig-Maximilians-Universität, Butenandstr. 11, München, Germany*

*sipr@fzu.cz

Behaviour of magnetic systems in external fields has become an intensively studied topic. Controlling magnetism with electric fields could be a key for developing future energy-efficient devices. Recently, it was demonstrated that external electric field can significantly modify material properties such as magnetic moments, x-ray spectra or lifetimes of magnons in thin films and that this effect is strongly influenced by the substrate [1,2]. We push this research further by examining how external electric field influences dynamic magnetic properties, in particular, the spin wave stiffness and the Gilbert damping. We monitor how this effect depends on the choice of the magnetic material and on the choice of the substrate.

**Acknowledgements**

This work was supported by the project GA ČR 23-04746S.

**References**

[1] E. Simon, A. Marmodoro, S. Mankovsky, H. Ebert, *Phys. Rev. B* **103** (2021) 064406

[2] A. Marmodoro, S. Mankovsky, H. Ebert, J. Minár, O. Šipr, *Phys. Rev. B* **105** (2022) 174411

# Influence of Magnetic Relaxation on the performance of Magnetoelastic Resonance-Based Detection

*Beatriz Sisniega[1], Jon Gutiérrez[1, 2, *], José Manuel Barandiaran[1] and Alfredo García-Arribas[1, 2]*

*[1]Departamento de Electricidad y Electrónica, Universidad del País Vasco (UPV/EHU), Leioa, Spain*

*[2]BC Materials, Applications and Nanostructures, UPV/EHU Science Park, Leioa, Spain*

*jon.gutierrez@ehu.eus

Magnetoelastic resonance (MER) sensors are usually made of amorphous ribbon-shaped ferromagnetic alloys. The operation of these sensors is based on the strong coupling between the magnetic and mechanical properties of these materials, which allows their mechanical excitation by the application of an alternating magnetic field, and the corresponding detection of the induced magnetic changes. The phenomenon of magnetomechanical resonance occurs in these materials at specific frequencies of excitation, and is highly sensitive to different external parameters, which, together with its wireless operation, has driven the development of a large number of detection devices based on this effect [1]. Nevertheless, although these soft magnetic materials present excellent mechanical and magnetic properties, they suffer magnetic relaxation under the application of the bias magnetic field [2, 3], an issue which is not usually addressed in the literature concerning the application of such materials as sensors. We have observed that the time-dependence of the magnetization (and other magnetic related properties) associated with this relaxation, affects the resonance signal of the sensor and thus its resonance frequency, the parameter most commonly used for sensing (Fig. 1). To unveil such phenomenon, in the present work, the influence of this relaxation behavior was studied at room temperature, for an amorphous ferromagnetic ribbon of composition $Fe_{73}Cr_5Si_{10}B_{12}$ performing as the main element of a MER sensor [4]. The magnetic relaxation was observed by monitoring the evolution, under constant bias field, of the magnetoelastic resonance signal (Fig. 1a), and in particular, the value of the resonance frequency $f_r$ (Fig. 1b), which is the parameter commonly used in the magnetoelastic detection. The relaxation was studied under different bias field values ($H$ = 4, 7.8 and 10 Oe) and different amplitudes of the excitation field ($h$ = 20, 42, 100 and 180 mOe). The study has revealed that the relaxation has a considerable effect on the sensor signal, producing changes of the resonance frequency up to several hundreds of Hz in a time interval of 2000 seconds, but it is very sensitive to the conditions of the experiment. The amplitude of the excitation magnetic field ($h$) turned out to be a key factor on this phenomenon, since larger excitation amplitudes ($h \geq 100$ mOe) reduce considerably its effect, decreasing the relaxation time ($\tau$) and reducing the temporal variation of $f_r$ experience by the sensor. In addition, the effect of this relaxation on the sensor performance was evaluated for an experimental case of real-time monitoring of a precipitation reaction. An approach based on the relaxation modelization is proposed to rectify this effect and thus improve the sensitivity of the sensor. Besides, we have found that these measurements of magnetic relaxation through magnetoelastic resonance can provide a novel, simple and accurate method for the study of magnetic relaxation processes in these materials.

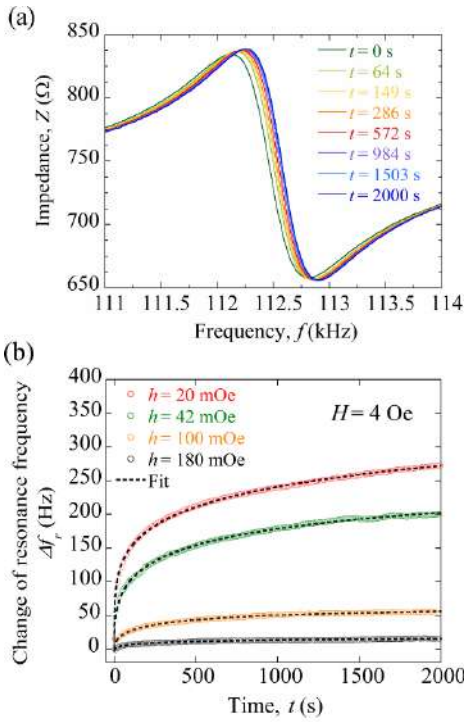



Figure 1: (a) Changes produced in the resonance signal of the sensor due to magnetic relaxation under a constant bias field. (b) Temporal evolution of the resonance frequency of the sensor under constant bias field of $\boldsymbol{H = 4}$ Oe and different excitation amplitudes $\boldsymbol{h}$.

## References

[1] C. A. Grimes *et al.*, *Sensors* **2** (2002) 294-313.
[2] J. Rivas *et al.*, *Journal of non-crystalline solids* **131** (1991) 1235-1239.
[3] P. Kwapuliński, G. Haneczok, *Acta Phys. Polonica A* **136** (2019) 701-704.
[4] B Sisniega *et al.*, *J. Phys. D: Appl. Phys.* (2023) **56** 105001.

# Disentangling the magentoresistance contributions of perovskites thin films

*I.García-Manuz[1,2]*, J.M. Diez, [1,2] R. Solís [1,3], G. De Arana [1] , J. L. F. Cuñado[1], A. Gudín[1], L.G. Enger [3] , B. Guillet [3], L. Méchin [3] , S. Flament [3] , P. Perna [1] and J.Camarero[1,2]*

*[1]IMDEA-Nanoscience, Campus de Cantoblanco, 28049 Madrid, Spain*

*[2]Dpto. Física de la Materia Condensada, IFIMAC, and Instituto "Nicolás Cabrera", Universidad Autónoma de Madrid, 28049Madrid, Spain*

*[3]CNRS GREYC (UMR 6072) ENSICAEN UNICAEN, 14050 Caen, France*

*E-mail: ines.garciam@uam.es*

Perovskite half-metallic manganites hold promise for spintronic applications, but are not yet available in today devices because of the lack of control of their magnetotransport properties. The reason for this relies on the complexity of the physical scenarios governing the interplay between a wide variety of coupled interactions. Different magnetoresistance (MR) contributions, such as colossal, Lorentz, spin-dependent scattering at grainboundaries, domain-walls and other magnetic inhomogeneities, typically hide the switchable anisotropic magnetoresistance (AMR) that is more amenable for technological applications. In addition, the mess can be even greater if each contribution presents different evolutions with temperature. Disentangling the origin of MR in manganites requires hence clear cut experiments to isolate the various contributions.

The most efficient way to investigate transport phenomena is to measure the field-driven magnetization and MR loops simultaneously in engineering epitaxial $La_{1-x}$ $Sr_xMnO_3$ (LSMO) films grown on top of vicinal (001)-oriented SrTiO3 (STO) single crystal substrates [1]. We present a detailed characterization of both magnetic and transport features at different applied field angles ($\alpha_H$), in the whole angular range, and in a broad temperature ($T$) range, from 400K to 100 K [2]. In general, the engineered epitaxial LSMO films display a wide variety of both magnetic and magnetoresistive behaviors which are related to each other. The data give direct views on the angular and temperature dependence of magnetoresistance and magnetization reversal pathways, from which characteristic axes, remanences, critical fields, domain wall types, effective magnetic symmetry, and MR contributions are obtained [3]. At a given temperature, MR display isotropic collosal magnetoresistance (CMR) and anisotropic magnetoresistance(AMR) contributions. The later originates from competing magnetic anisotropy terms: uniaxial $K_U$ (growth-induced) and biaxial $K_2$ (magnetocrystalline). The isotropic (linear) behavior of CMR is better identified at large fields while the AMR contribution is at low fields. Similar trends of the CMR linear factor and the resistance with temperature are found. AMR shows asymmetric (and clear hysteretic) behaviour at low $T$, that becomes more evident as $T$ decreases, which may be associated with the increasingly remarkable biaxial contribution. These findings have a strong impact on the real applications of manganite based devices for the high-resolution low field magnetic sensors and spintronics.

**References:**

[1] P. Perna *et al.* Adv. Funct. Mater. 27, 1700664 (2017).

[2] J.L.F. Cuñado *et al.,* Rev. Sci. Instrum. **86**, 046109 (2015).

[3] I.García-Manuz *et al.,* to be published.

# FERRO to ANTIFERROMAGNETIC TRANSITION in EPITAXIAL THIN FILMS of $Mn_5(Si_xGe_{1-x})_3$ GROWN on Ge(111) SUBSTRATE

*Sueyeong Kang [1], Roger Kalvig [2], T. Baujard [3], G. Bon [3], Vasile Heresanu [1], O. Cespedes [4], B. Hickey [4]*
*Ewa Jedryka [2], Marek Wojcik [2], Lisa Michez [1*], Matthieu Petit [1]*

[1] *Aix Marseille Univ, CNRS, CINaM, Marseille, France*
[2] *Institute of Physics, Polish Academy of Sciences, Aleja Lotników 32/46, PL–02668 Warsaw, Poland*
[3] *Aix Marseille Univ, Polytech, Materials Department, Marseille, France*
[4] *Condensed Matter group, University of Leeds, Leeds LS2 9JT, United Kingdom*
*lise.michez@univ-amu.fr

$Mn_5Ge_3$ and $Mn_5Si_3$ alloys crystallizes in the same hexagonal $D8_8$ structure ($P6_3/mcm$ space group) (see inset of Fig.1a). In both compounds, the manganese atoms are located in two different crystallographic sites, so-called $Mn_I$ and $Mn_{II}$, corresponding to the Wyckoff positions 4(d) and 6(g), respectively. Despite these similarities, their magnetic behaviour is very different. $Mn_5Ge_3$ is strongly ferromagnetic with a Curie temperature of 296 K, while $Mn_5Si_3$ is a complex antiferromagnet with a temperature- and field-dependent magnetic structure. The two compounds can be alloyed over the whole Ge-Si concentration range, which makes this system ideal to study the transition occurring from a ferromagnetic state ($Mn_5Ge_3$) to an antiferromagnetic state ($Mn_5Si_3$).

In this context, we present here the epitaxial growth of $Mn_5(Si_xGe_{1-x})_3$ thin films on Ge(111) substrate by Molecular Beam Epitaxy using a codeposition technique. Their structural properties have been thoroughly characterized using reflection high-energy electron diffraction, x-ray diffraction, atomic force microscopy and transmission electron microscopy while the magnetic properties have been investigated using vibration sample magnetometry (VSM) and $^{55}Mn$ Nuclear Magnetic Resonance (NMR) which acts as a very sensitive local probe of the Mn atoms environment.

Upon silicon substitution, two correlated effects are readily visible: i) the decrease of the lattice parameters (Fig. 1) and (ii) a diminution of the Mn magnetic moment on both Mn sites as evidenced by the shift towards lower frequencies in the NMR spectra. This is confirmed by the decrease of the average magnetic moment measured by VSM. The thorough analysis of the structural and magnetic data suggests a steady substitution of Ge by Si atoms as the alloy is enriched in Si until a concentration threshold of $x$~0.6. A higher Si concentration drives a drastic change in the thin films magnetic behaviour. NMR local probe indicate that the Si substitution mainly affects the $Mn_{II}$ atoms local environment.

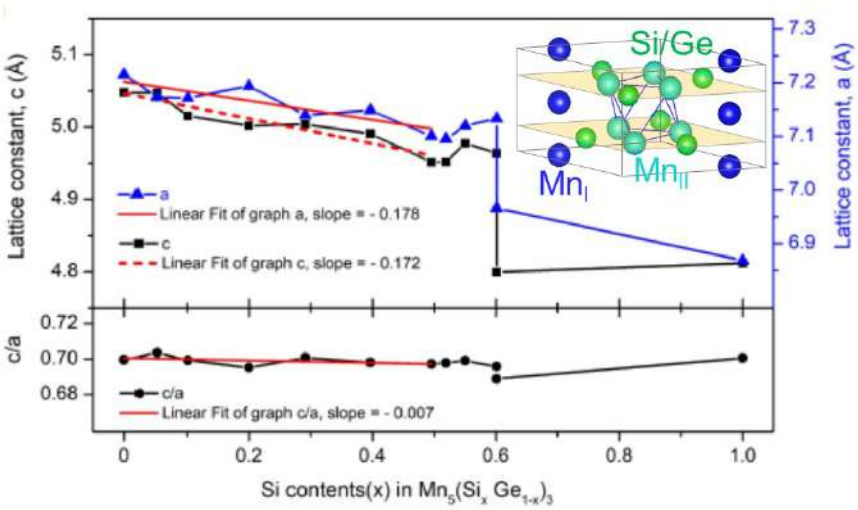



Figure 1: Dependence of the lattice parameters *a* and *c* and their ratio as a function of the silicon content *x* in $Mn_5(Si_xGe_{1-x})_3$ epitaxial thin films. Inset: unit cell of the $Mn_5Si_3$-type structure.

# Magnetoelastic stresses in epitaxial FeGa thin films

*Miguel Ciria[1]*, Adrián Begué[1,2], Maria Grazia Proietti[1], José I. Arnaudas[1]*

[1] *Instituto de Nanociencia y Materiales de Aragón(INMA), CSIC-Universidad de Zaragoza, Zaragoza 50009, España.*

[2] *Universidad Complutense de Madrid (UCM), dpto. de Física de Materiales, Madrid 28071, España*

*miguel.ciria@csic.es

$Fe_{100-x}Ga_x$ is a magnetic alloy whose properties, such as magnetostriction or anomalous Nernst effect, are strongly linked to crystal structure and doping, and coexistence of multiple crystalline phases. These factors can significantly affect the performance of the alloy. Magnetostriction is directly tied to magnetoelastic (ME) stress, but the stiffness elastic constants for this family of alloys also play an important role. The ME stresses $B_1$ and $B_2$ are crucial in determining the anisotropies induced by external strains in thin films. In this study, we present measurements of both coefficients for $Fe_{100-x}Ga_x$ epitaxial films with thicknesses below 50 nm and compositions of x = 21, 28, and 33 at room temperature using the cantilever method [1]. Our measurements indicate that both irreducible coefficients exhibit bulk-like values. Specifically, $B_1$ takes the largest value in modulus for films with x = 21 while $B_2$ is positive for x = 21 and negative for the other compositions. Interestingly, we observe no significant variation in $B_1$ and $B_2$, which extends previous observations in thicker films [2]. To investigate possible local structural changes associated with Ga concentration and film thickness, we conducted X-ray absorption fine structure experiments (XAFS). The results show that there are no appreciable structural changes with changing thickness. However, we do observe a significant increase in the static disorder of the Ga environment next-nearest-neighbors distribution as x increases from 21 to 28 and 33.

**Acknowledgements**

This work was supported by Spanish MICINN (Grants No. TED2021-131064B-I00 and PID2021-124734OB-C21). A.B. would like to acknowledge the funding received from the Ministry of Science and Innovation for the Ph.D. contract BES-2016-076482, as well as the funding received from the Ministry of Universities and the European Union-Next Generation for the Margarita Salas fellowship.

**References**

[1] P. Bartolomé, A. Begué, A. Muñoz-Noval, M. Ciria, and R. Ranchal. *J. Phys. Chem. C* **124** (2020) 474

[2] M. Barturen, D. Sander, J. Milano, J. Premper, C. Helman, M. Eddrief, J. Kirschner, and M. Marangolo, *Phys Rev B.* **99** (2019) 134432.

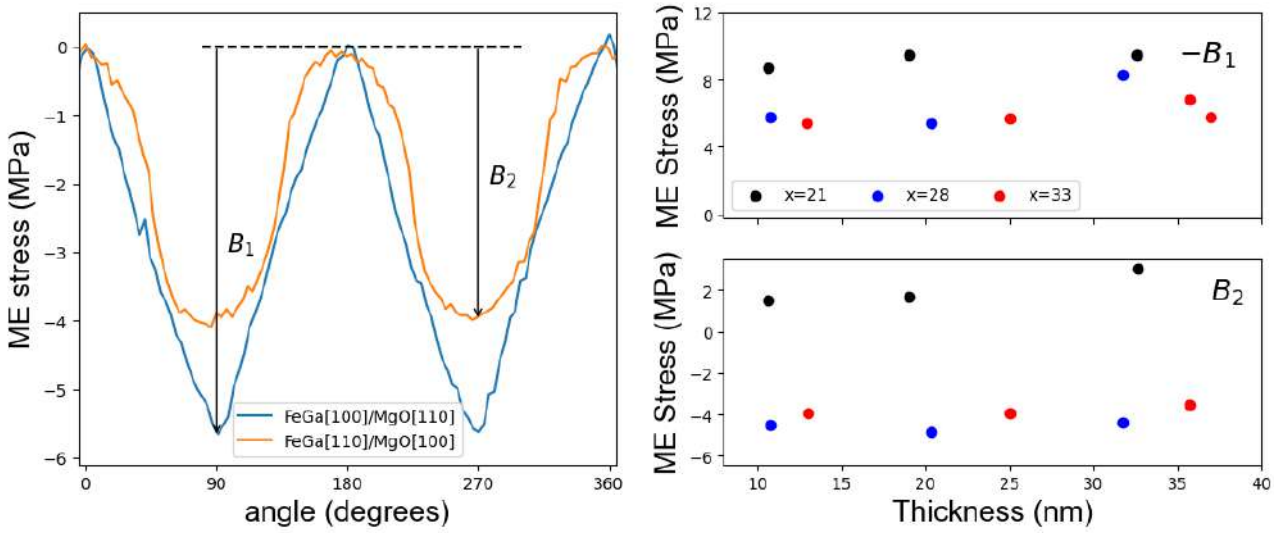



Figure. Left: ME stresses as a function of the angle between the applied field and the cantilver beam. Right: ME stress coefficients as function of the FeGa film thickness and Ga atomic content *x*.

# Anomalous Nernst Effect on Magnetic Multilayers with Flexible Substrate

*Cantia Belloso-Casuso[a*], G. López-Polín[b], A. Asenjo[a]*
*[a]Instituto de Ciencia de Materiales de Madrid (CSIC), C/Sor Juana Inés de la Cruz, 3. 28033, Madrid, Spain*
*[b]Departamento de Física de Materiales, Universidad Autónoma de Madrid, Madrid, E-28049, Spain*
** cantia.belloso@icmm.csic.es*

One of the thermoelectric effects that is currently attracting a considerable interest because of its potential for energy harvesting is the anomalous Nernst effect (ANE). ANE is a thermomagnetic phenomenon with potential applications in thermal energy harvesting. Our aim is to study and increase the ANE coefficient of materials and record the power density generated by the ANE by using flexible substrates in order to increase the number of applications it may be used.

Therefore, we make use of a micrometer-sized Hall bar device consisting of [Co0.5nm/Pt0.5nm]10 sputtered multilayers, which present a high ANE coefficient and thermopower (~1 μV/K), low electrical resistivity, and perpendicular magnetic anisotropy (PMA) when grown on a SiNx/Si substrate. Furthermore, flexible substrates (like polymide, acetate…) allow to analyze the effect strain has in the properties of the samples. This way, the devices shown in this work, are composed by magnetic multilayers with high PMA exposed to in-plane thermal gradient. VSM and VF-MFM are other experimental techniques employed to accomplish the magnetic characterization. Additionally, a MATLAB software will be developed to measure Seebeck and Hall effects too in order to fully characterize the manufactured devices.

We believe that this design may find uses in harvesting wasted energy, e.g., in electronic devices.

**References**

[1] S. Hashimoto et al. *J. Appl. Phys.* **66** (1989) 4909.
[2] M. Mizuguchi et al. *Sci. Technol. Adv. Mater.,* **20** (2019) 262−275.
[3] F. Zhang et al. *J. Appl. Phys*., **110** (2011) 033921.
[4] G. Lopez-Polin et al. *ACS Appl. Energy Mater.,* **5** (9) (2022) 11835-11843.
[5] G. Dai et al. *Appl. Phys. Lett*., **100** (2012) 122407.

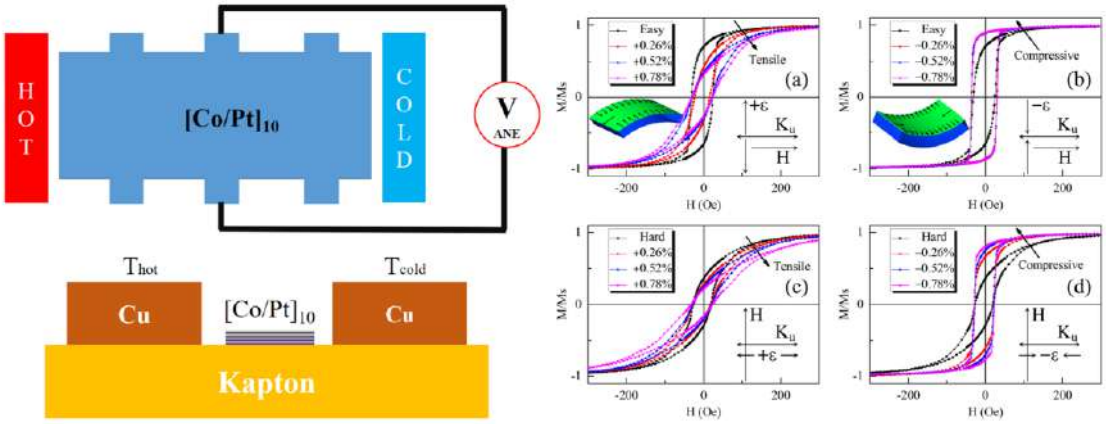



Figure: Description of the image Scheme of the device and ANE measurement used technique, and hysteresis loops for Fe81Ga19/ PET obtained under various external strains using different measuring configurations [5].

# Temperature-dependence of ferromagnetic resonance in YIG garnets thin films

*Lara M. Solis[1,2,3]*, Santiago Carreira[4], Javier Briático[4], Abdelmadjid Anane[4], Myriam H. Aguirre[3,6,7], and Laura Steren[1,5]*

[1] *Instituto de Nanociencia y Nanotecnología CNEA-CONICET, Nodo Constituyentes, Buenos Aires, Argentina*

[2] *Instituto Sábato, Universidad Nacional de San Martín, San Martín, Argentina*

[3] *Instituto de Nanociencia y Materiales de Aragón, UNIZAR-CSIC, Zaragoza, Spain*

[4] *Unité Mixte de Physique, CNRS, Thales, Université Paris-Saclay, Palaiseau, France*

[5] *Consejo Nacional de Investigaciones Científicas y Técnicas, Argentina*

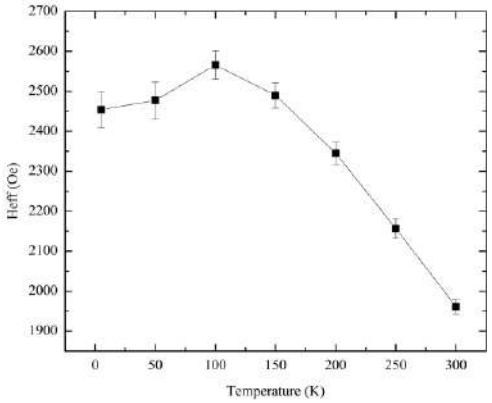
[6] *Departamento de Física de la Materia Condensada, UNIZAR, Zaragoza, Spain*

[7] *Laboratorio de Microscopías Avanzadas, UNIZAR-CSIC, Zaragoza, Spain*

*laramsolis@gmail.com

The ferrimagnetic insulator Y3Fe5O12 (YIG) is a model material that exhibits low magnetic damping, making it promising for applications in magnonic devices with ultrafast magnetization dynamics. Furthermore, its optical attributes, particularly a large Faraday rotation, can be used to fabricate optical valves or devices. Due to these interesting properties YIG thin films have been extensively investigated and have also been integrated into countless heterostructures, combined with topological insulators and heavy metals, among other materials. For example, YIG/Pt systems allow efficient spin-charge conversion and pure detection of spin-current effects owing to their unique properties [1].

In order to obtain a thorough characterization of the magnetic anisotropy of pure epitaxial YIG, thin films grown by PLD have been prepared on sGGG substrate. The magnetic characterization was performed by FMR measurements in the range from 4 to 40 GHz and temperatures between 5K and 300K. In Fig. 1, the temperature dependence of the effective field for a 60nm-YIG/sGGG thin film is shown. The effective field, $H_{eff}$, derived from the resonance field of the spectra is related to the magnetic anisotropy of the films by the foolwing expression Heff = 4πMs – 2K/Ms where K is the magnetic anisotropy constant and Ms the saturation magnetization. The effective field, $H_{eff}$, decreases up to 25% at temperatures above 100K. We analyze the temperature dependence of the magnetic anisotropy and the magnetization [2] of YIG thin films in terms of magnetoelastic effects.




## Acknowledgements

This work is funded by ANPCYT Grant PICT 2019-02781. We thank the funding received from the MSCA Project ULTIMATE-I under the European Union's Horizon 2020 research and Innovation program under grant agreement No 101007825.

## References

[1] C. Du, H. Wang, P. C. Hammel, and F. Yang, Journal of Applied Physics 117, 172603 (2015).

[2] Gouéré, D., Merbouche, H., El Kanj, A., Kohl, F., Carrétéro, C., Boventer, I, & Anane, A. Physical Review Materials, 6(11), 114402 (2022).

Figure 1: Temperature dependence of the effective field, Heff for 60nm-YIG on sGGG substrate.

# Modification of the magnetocrystalline anisotropy at molecule-cobalt interfaces

*Anita Halder*, Sumanta Bhandary, David O'Regan, Stefano Sanvito, and Andrea Droghetti*
*School of Physics and CRANN, Trinity College, Dublin 2, Ireland*
*haldera@tcd.ie

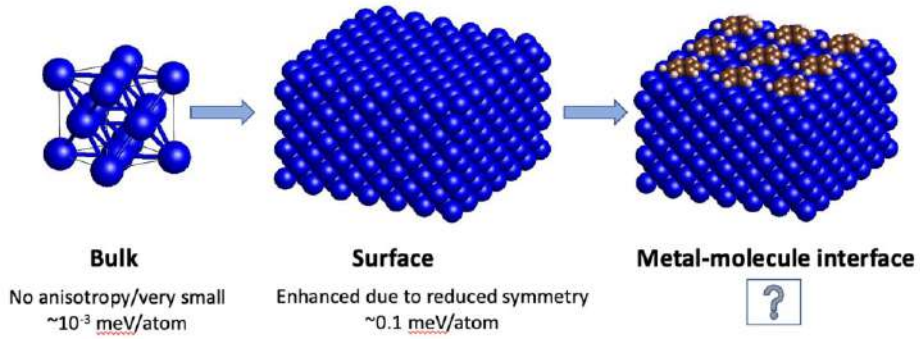
Magnetocrystalline anisotropy(MCA) plays an important role in high-density data storage because it can be used to control the orientation of magnetic domains within the storage medium. By controlling the orientation of the magnetic domains, it is possible to create stable, long-lived magnetic patterns that can be used to represent digital information. In this regard, the adsorption of nonmagnetic organic molecules on ferromagnetic materials offers an opportunity to tune their magnetic properties for promising applications in data storage devices. In the present work, we report the manipulation of the MCA of Co slabs through the adsorption of small molecules, such as benzene, cot, etc. We have used Density Functional Theory and the magnetic force theorem to calculate magnetic anisotropy. The results indicate that molecular adsorption tends to favor perpendicular MCA at surfaces by reducing the in-plane MCA of the slab. Further, we analyze our results considering a simple model based on 2nd-order perturbation theory to explain the modification of MCA due to molecular adsorption in a qualitative way. A detailed analysis of various atom-resolved quantities from DFT calculations demonstrates that the underlying physical mechanism is the metal-molecule interfacial hybridization, and, in particular, it is related to the chemical bond between the molecular $p_z$ and the surface $d_{z^2}$ orbitals. Generalizing the same argument, we also show that the complex molecules $C_{60}$ and $Alq_3$ deposited on fcc-Co induce a similar modification of the in-plane MCA, and we related the results to recent experimental observations.

**Acknowledgements**

Marie Curie postdoctoral fellowship (VOLTEMAG - 101065605), Science Foundation Ireland (SFI) and the Royal Society through the University Research Fellowship URF-R1-191769. The computational resources from Trinity College Dublin Research IT and the Irish Center for High End Computing.

**References**

https://doi.org/10.48550/arXiv.2302.07330



# Soft magnetism in single phase Fe3Si thin films deposited on SrTiO3(001) by pulsed laser deposition

*Icíar Arnay1,2,3*, Aída Serrano1,2,4, Verónica Braza5, Rosalía Cid1,2,6, Ana M. Sánchez7, Jesús López-Sánchez1,2, Germán R. Castro1,2, Juan Rubio-Zuazo1,2*

*1. Spanish CRG BM25-SpLine at The European Synchrotron. 71 Av. des Martyrs 38000 Grenoble, France*

*2. Instituto de Ciencia de Materiales de Madrid, CSIC, C/Sor Juana Inés de la Cruz 3, 28049 Madrid, Spain*

*3. IMDEA Nanociencia C/Faraday 9, 28049 Madrid, Spain*

*4. Instituto de Cerámica y Vidrio, CSIC, C/Kelsen 5, 28049 Madrid, Spain*

*5. Instituto de Microscopía Electrónica y Materiales (IMEYMAT) Universidad de Cádiz, Facultad de Ciencias, 11510 Puerto Real, Cádiz, Spain*

*6. CIC EnergiGUNE, Parque Tecnológico de Álava, Albert Einstein, 48, 01510 Vitoria-Gasteiz, Spain*

*7. Department of Physics, University of Warwick, Coventry CV4 7AL, United Kingdom *iciar.arnay@imdea.org*

The development of devices relying on spin phenomena requires of an ideal spin polarized electron source. This can be achieved by taking advantage of half-metallic full Heusler alloy thin films. $Fe_3Si$ presents ferromagnetic behavior up to 840 K and it is characterized by a soft magnetism, with coercive fields in the order of few mT, and a low damping constant, which make it a great alternative for common ferromagnetic electrodes as permalloy for the study of spin based phenomena [1,2]. However, its implementation requires a controlled growth of stoichiometric films with large activation volumes. Moreover, its half metallic behaviour is surrogated to structural properties thus the preparation of high quality films as well as a thoughtful characterization of their structural properties becomes mandatory. [3]. In this work, we report on the growth of epitaxial $Fe_3Si$ ultra-thin films by pulsed laser deposition on $SrTiO_3(001)$ substrates, analyzing the effect of deposition temperature in the structural, morphological and magnetic properties of the deposited films. We conclude that optimal compromise between phase purity and interface quality is obtained at 200 ºC, obtaining the best magnetic response under this condition [4].

**Acknowledgements**

This project received financial support from the Spanish MCIN and CSIC [Grant No. 2010-6-OE-013 and 2021 60 E 030]. IA acknowledges support by the Community of Madrid (CM) through Project P2018/NMT4321 (NANOMAGCOST), by MINECO through Project RTI2018-097895-B-C42 (FUN-SOC), PCI2019-111867-2 (FLAG-ERA JTC 2019, SOgraphMEM), and by the “Severo Ochoa” Programme for Centres of Excellence in RD, MINECO grant SEV-2016-0686.. A.S. acknowledges the financial support from the CM for contract (No. 2017-t2/IND5395).

**References**

[1] W. Rotjanapittayakul, W.Pijitrojana, T. Archer, S. Sanvito, J. Prasongkit, Sci Rep 8 (2018) 4779

[2] H. Y. Hung, G. Y. Luo, Y. C. Chiu, P. Chang, W. C. Lee, J. G. Lin, S. F. Lee, M. Hong, J. Kwo, J.Appl. Phys. 113 (2013) 17C507

[3] L. L. Tao, S. H. Liang, D. P. Liu, H. X. Wei, Jian Wang, and X. F. Han, Appl. Phys. Lett. 104 (2014) 172406

[4] I. Arnay, A. Serrano, V. Braza-Blanco, R. Cid, A. M. Sánchez, J. López-Sánchez, G. R. Castro, J. Rubio-Zuazo, J. Alloys. Compd. 947 (2023) 169540

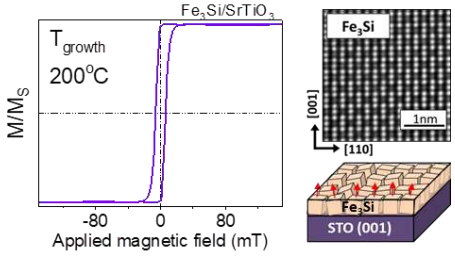



Figure: Longitudinal magnetic optical Kerr effect (L-MOKE) characterization of $Fe_3Si$(20 nm)/SrTiO3(001) stacks deposited by PLD and high magnification STEM-HAADF image of the layer of interest ($Fe_3Si$) proving epitaxial growth.

# Origin of local magnetic moments in Cr-V alloys

*A. J. A. de Oliveira[1,2]*, L. M. de Oliveira[3], P. E. N. de Souza[4], Fabiano Yokaichia[5], P. C. de Camargo[1]*

[1]*Departamento de Física – Universidade Federal de São Carlos, 13565-905, São Carlos, SP, Brazil*

[2]*Instituto de Estudos Avançados e Estratégico - Universidade Federal de São Carlos, 13565-905, São Carlos, SP, Brazil*

[3]*Colegiado de Ciências da Natureza, Campus Senhor do Bonfim - Universidade Federal do Vale do São Francisco, 48970- 000, Senhor do Bonfim, BA, Brazil*

[4]*Núcleo de Física Aplicada, Instituto de Física - Universidade de Brasília, DF 70910-900, Brasília, DF, Brazil*

[5]*Departamento de Física, Universidade Federal do Paraná, 82590-300 Curitiba, PR, Brazil*

*corresponding author adilson@df.ufscar.br

Chromium-rich Cr-V alloys exhibit three phases: Paramagnetic phase (P) above Néel temperature ($T_N$), a transverse polarization spin-density-wave (SDW) phase ($AF_1$) below T and above the spin-flip temperature ($T_{SF}$), and a longitudinal polarization SDW phase ($AF_2$) below $T_{SF}$. In the paramagnetic phase, Cr exhibit a Pauli susceptibility with slight temperature dependence [1]. Otherwise, the introduction of small amounts of V in Cr not only changes T, but also induced a Curie-Weiss behavior (CW) that we have associated to local magnetic moments. This behavior is limited up to 0.67%V and magnetic fields of 15 kOe [2,3] . This behavior was also observed for different Cr alloys. In magnetic susceptibility as a function of temperature using zero field-cooling (ZFC) and field-cooling (FC) protocol is observed that in ZFC measurements the local moments only appears in paramagnetic phase. Otherwise, in FC measurments this behvaior is is not only observed in the paramagnetic phase, but also in the AF1 phase, exhibiting a strong magnetic irreversibility. The origin of local magnetic moments has been associated with the establishment of local spin-density waves (LSDW) around V impurities. In this work, we presented an investigation of the effects of local magnetic moments in antiferromagnetic phases in Cr-x at.% V alloys (x=0, 0.1, 0.2, 0.4 and 0.67). Results show that presence of LSDW induces a strong magnetic anisotropy in long-range order of SDW in $AF_1$ phase.

## Acknowledgements

The authors acknowledge the financial support from CNPq and FAPESP (2013/07296-2)

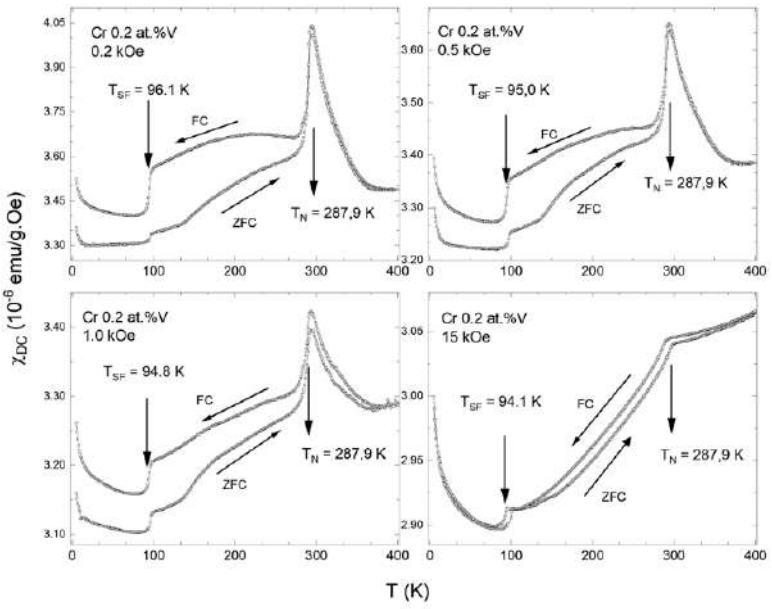



Figure: Magnetic susceptibility of function of temperature for Cr-0.2 at.%V with different magnetic fields using ZFC and FC protocol (a) H= 0.2 kOe, (b) 0.5 kOe, (c) 1.0 kOe, and (d) 15 kOe

## References

[1] E. Fawcett, H. L. Alberts, V. Yu. Galkin, D. R. Noakes, and J. V. Yakhmi - Rev. Mod. Phys. **66**, (1994) 25

[2] L. M. de Oliveira, W. A. Ortiz, and A. J. A. de Oliveira - J. of Applied Physics **93**, (2003) 7154.

[3] A. J. A, de Oliveira, et al. Yokaichiya - AIP Advances, **13**, (2023) 015310.

# Microfabrication of Complex Soft Magnetic FeCo(V) Components by Combining Lithography and Electrodeposition

*Ester M. Palmero*, Manuel R. Osorio, Andrés Valera, María Acebrón,*
*Alberto Bollero, and Daniel Granados*
*IMDEA Nanoscience, 28049 Madrid, Spain*
*ester.palmero@imdea.org

The microfabrication of functional parts is opening the path to new and innovative applications (e.g., micro-electro-mechanical systems, MEMS) in key high-tech sectors such as electronics, aerospace and medical [1]. Combining lithography and electrochemistry allows for manufacturing unique functional components with complex shapes and tunable properties accompanied by high dimensional and replication accuracies [2,3]. Additional advantages of the combination of these technologies are the possibility of fabricating parts with a high aspect ratio, smooth surface finish, and with parallel side walls with flank angles really close to 90º [3,4].

In this work FeCo(V) micrometer size parts with complex shapes were manufactured by an optimized combination of lithography and electrodeposition. This process allowed to engineer the shape of the parts and their magnetic performance by tuning the growth parameters, e.g., electrolyte stoichiometry and electrodeposition voltage and duty cycle (Fig. 1). FeCo-based alloys have been used to obtain functional soft ferromagnetic parts showing a low coercivity (50 Oe) accompanied by a high saturation magnetization (1300 emu/cm$^3$) (Fig. 1b). Doping the $Fe_{65}Co_{35}$ alloy with vanadium (1.5 at.%) has enhanced the finishing quality of the fabricated parts (Fig. 1c). Optimization of the process has allowed for obtaining parts with a high aspect ratio (>1:20), complex contours, and micrometer accuracy. The developed approach enables the fabrication of hundreds of units per run (see inset in Fig. 1b), thus opening the path to an industrially scalable process with a drastic reduction of the unit cost of each component.

## Acknowledgements

Authors acknowledge financial support from EU by the FET-Open project *UWIPOM2* (Ref. 857654). IMDEA Nanoscience thanks support from the ‘‘Severo Ochoa’’ Programme for Centers of Excellence in R&D (MICINN, Grant CEX2020-001039-S) and Comunidad de Madrid by TEC2SPACE-CM-P2018-NMT-4291. E.M.P. acknowledges support from AEI through the Juan de la Cierva – Incorporación program (IJC2020-043011-I/MCIN/AEI/10.13039/501100011033) and EU by NextGenerationEU/PRTR.

## References

[1] N. Jackson et al., *J. Microelectromech. Syst.* **25** (2016) 716.
[2] Y. Wang et al., *J. Microelectromech. Syst.* **28** (2019) 311.
[3] H. Zhang et al., *J. Micromech. Microeng.* **30** (2020) 103002.
[4] F. Cros et al., *J. Microelectromech. Syst.* **15** (2006) 427.

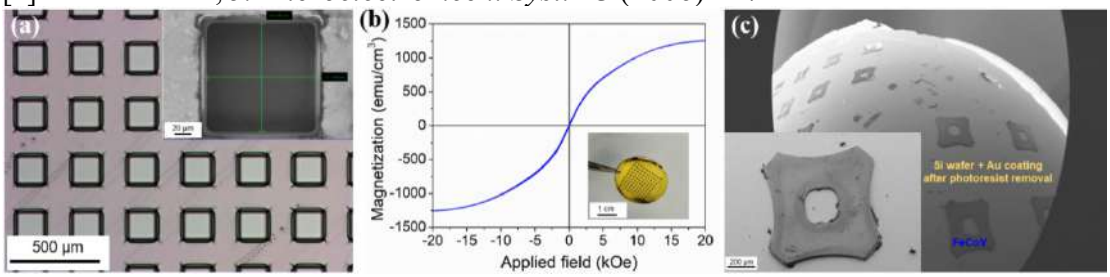



Figure 1: (a) Top view Scanning Electron Microscopy (SEM) image of an array of micrometer size cubes made of photoresist by lithography. Inset shows a FeCoV cube grown by electrodeposition on the lithographed molds in (a). (b) Room temperature hysteresis loop measured by VSM for the electrodeposited FeCoV alloy showing its soft ferromagnetic behaviour. Inset in (b) shows a representative array of FeCoV parts grown by electrodeposition into the photoresist molds used for the magnetic characterization. (c) SEM image of an array of FeCoV parts with complex shape after the selective chemical removal of the photoresist. Inset in (c) shows a close view of a single FeCoV part after mold removal.

# Left-Handed Properties of the Nanocrystaline Ferromagnetic Microwires for GHz Shielding

*G. Ababei*[*], *I. Murgulescu, S. Corodeanu, V. Dobrea, G. Filipciuc, N. Lupu*
*National Institute for Research and Development for Technical Physics,*
*47 Mangeron Boulevard, Iaşi, Romania*
*gababei@phys-iasi.ro

In the last years, the specific properties of ferromagnetic microwires have been intensively investigated in order to develop metastructures known as electromagnetic absorber with negative values of the dielectric permittivity, $\varepsilon$, and magnetic permeability, $\mu$, for high frequency shielding applications [1-3]

The aim of this paper is to report our lastest experimental and theoretical results on the left-handed properties of FeCuNbSiB nanocrystaline magnetic microwires, arranged in parallel configurations, as free-standing systems, in order to develop new metastructures for GHz shielding applications.

$Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ (at.%) microwires with diameters of 60 μm were obtained by cold-drawing process, by successive reductions of the diameter of an amorphous microwire with the initial diameter of 130 μm, prepared by in-rotating-water spinning method.

The nanocrystalline structure of the FeCuNbSiB amorphous microwires was induced by annealing at 500ºC for 1 h. The nanocrytalline microwires with length of L = 0.8 cm, were fixed in parallel arrangement at different inter-microwire distance, (d = 1 mm, and, respectively, d = 3 mm), on a dielectric holder. The left-handed characteristics of the free standing FeCuNbSiB - based metastructure have been studied using a X-band microwave guide connected to the emission-reception ports of a Vector Network Analyzer (VNA) in the frequency range 8.2 ÷ 12.4 GHz The left-handed characteristics of the metastructure were computed using the measured reflection and the transmission coefficients of the VNA, as imput terms for theoretical calculations the magnetic permeability, and of the dielectric permittivity, respectively, of the metastructures through the Nicolson-Ross-Weir (NRW) analytical method [4]. External d.c. magnetic fields, $H$, in the range of 0 to 32 kA/m were applied parallel to the long axis of the microwires.

Experimental results shown that in the absence of the external magnetic field, the metastructures with the inter-microwire distance of d = 1 mm present a microwave absorption window of about 2 GHz, while the metastructures with the inter-microwire distance of d = 3 mm present a microwave absorption window of about 3 GHz. Both absorption windows have the central frequency at about 9.7 GHz. In the presence of the maximun magnetic field, the width of the absorption window is slightly increased with about 1 GHz and the central frequencies are shifted to higher frequencies with about 200 MHz for both type of metastructures.

The negative values of the real term of the magnetic permeability, and of the dielectric permittivity, respectively, of the metastructures obtained by theoretical calculations through the NRW analytical method confirms the existence of left-handed behavior of the developed metastructures in the frequency range 8.2 ÷ 12.4 GHz.

The variation of the interwire distance proves to be a useful tool to obtain metastructures with suitable left-handed characteristics for GHz applications. The properties of such metastructures are found dependent on their geometrical parameters and also on the intrinsic properties of the used microwires, offering flexibility for achieving different engineering requirements.

**Acknowledgements**
Financial support from NUCLEU Programme - Contract No. 18N/01.01.2023, project PN 23 11 01 01 is highly acknowledged.

**References**

[1] H. Li, L. Hua Yuan, B. Zhou, X. P. Shen, Q. Cheng and T. Jun Cui, *J. Appl. Phys*. **110** (2011) 014909.
[2] G. Ababei, C. S. Olariu, N. Lupu, and H. Chiriac H, *AIP Advances* **6** (2016) 055901.
[3] Q.Jiang, Y. Qiao, A. Uddin, F. Qin, L. Chen, L. Wu, *Composites Part B*, **247** (2022) 110298.
[4] J.B. Pendry, A.J. Holden, W.J. Stewart, and I. Youngs, *Phys. Rev. Lett.* **76** (1996) 4773.

# Laser-Induced Manipulation of Magnetization in Nanopatterned GdFe Thin Films

*Victor Deinhart[1, 2,] *, Lisa-Marie Kern[1], Christopher Klose[1], Felix Steinbach[1], Clemens von Korff Schmising[1], Dieter Engel[1], Katja Höflich[3], Stefan Eisebitt[1, 4], Bastian Pfau[1]*

[1] *Max-Born-Institut, 12489 Berlin, Germany*
[2] *Helmholtz-Zentrum für Materialien und Energie GmbH, 14109 Berlin, Germany*
[3] *Ferdinand-Braun-Institut, Leibniz-Institut für Höchstfrequenztechnik, 12489 Berlin, Germany*
[4] *Technische Universität Berlin, 10623 Berlin, Germany*
*victor.deinhart@helmholtz-berlin.de

Since the discovery of all-optical switching (AOS) in rare-earth transition-metal (RE-TM) systems, these materials have been proposed to be used in future computational applications, e.g., as novel data storage devices [1]. Yet, the deterministic manipulation of the magnetization on the nanometer scale via ultra-short laser pulses remains a challenge [2, 3]. In this work, we explore the ability to modify the AOS property of GdFe thin films using He-ion irradiation with the aim to reduce the switching area down to the nanoscale. Using focused 30-kV He-ion irradiation allows us to modify the magnetic properties on spatial scales of ~10 nm without changing the film's topography [4, 5].

In agreement with literature [5], we find a reduction of the perpendicular magnetic anisotropy (PMA) after ion irradiation until the magnetizations turns fully in-plane for sufficiently high doses. In addition, we observe that the laser fluence range suitable for triggering AOS reduces in the ion-irradiated GdFe films and, finally, the AOS ability disappears above a composition-dependent dose threshold. Still, this threshold dose is insufficient to turn the magnetization in-plane.

In our GdFe films, we create islands with 80 nm width by irradiating grid lines of square lattices with an He-ion dose which entirely quenches the PMA. The untreated magnetic islands are imaged using scanning X-ray microscopy (STXM) at the MAXYMUS beamline located at the BESSY II synchrotron-radiation source. In this microscope, photoexcitation is provided by a 1030-nm infrared laser with 10–15 ps pulse duration. While AOS is not supported by this pulse length, we demonstrate that heat assisted magnetization switching of individual islands can be achieved using ps laser pulses (Fig. (a)). Statistical analysis of magnetization patterns after several applied laser pulses indicates a spatial dependence of the islands' switching behavior (cf. Fig. (b)) for which we account lateral material inhomogeneities as already reported by Liu et al. [3] on lengths scales of 80 nm and above. Further, we observe a clustering of islands with same magnetization direction supposedly to be a result of a transient coupling of the islands during laser-induced heating. In conclusion, He-ion patterning allows to create magnetic islands that can be addressed with laser excitation, providing a promising platform to achieve deterministic AOS on the nanoscale.

[1] Stanciu et al., Phys. Rev. Lett. **99** (2007) 047601.
[2] Finazzi et al., Phys. Rev. Lett. **110** (2013) 177205.
[3] Liu et al., Nano Lett. **15** (2015) 6862.
[4] Fassbender et al., J. Phys. D: Appl. Phys. **37** (2004) R179.
[5] Krupinksi et al., Phys. Rev. Mater. **5** (2021) 024405.

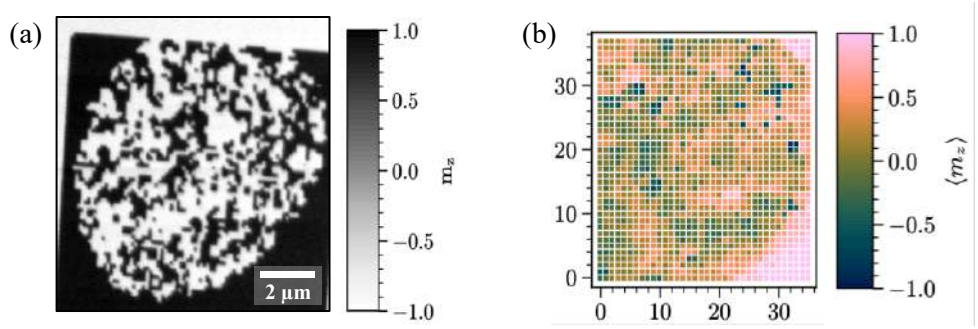



Figure: (a) Normalized out-of-plane magnetization pattern after applying a single ~15 ps laser pulse on a He-irradiated grid. (b) Averaged island out-of-plane magnetization for 20 applied laser pulses. Each square represents a single 80 nm island. All magnetization patterns are extracted from STXM data.

# Multipole magnetic expansion in conical-helical magnetic textures stabilized by Dzyaloshinskii-Moriya interaction

*Felipe Brevis[1]*, Rodolfo Gallardo[1], Paula Mellado[2], Pedro Landeros[1]*

[1]*Departamento de Física, Universidad Técnica Federico Santa María, Avenida España 1680, Valparaíso, Chile.*

[2]*Facultad de Ingeniería y Ciencias, Universidad Adolfo Ibáñez, Diagonal las Torres 2640, Peñalolén, Santiago, Chile.*

*felipe.brevis@usm.cl

The relationship between the multipolar magnetic expansion of conical-helical (CH) magnetic textures stabilized by Dzyaloshinskii-Moriya interaction (DMI) and the magnetoelectric effect is theoretically investigated. These magnetic textures correspond to non-collinear magnetization orderings in the form of a spiral that can be useful to give rise to a magnetoelectric effect. Such CH textures are formed due to the competition between magnetic interactions such as dipolar, Zeeman, anisotropy, symmetric and antisymmetric exchange (or DMI) [1]. The textures are characterized by a pitch vector ($q$) which defines the direction and period of the texture and by a cone angle ($\theta$) that sets the inclination of the magnetization with respect to $q$. The physical system corresponds to a square ferromagnetic film of side $L$ and thickness $d$ where all the interaction previously mentioned are considered. On the other hand, the magnetoelectric effect corresponds to the coupling between the magnetic (ferroelectric) order with electric (magnetic) fields and has been reported in spiral-type magnetic structures [2-3]. This effect is modeled through the magnetoelectric tensor related to the magnetic multipolar expansion, which gives rise to monopolar, toroidal and quadrupole terms [3-4], where the toroidal moment is closely linked to the magnetoelectric response. Analytical results of these terms are presented as a function of the different types of DMI: bulk and interfacial. For instance, by reducing the bias field from saturation, one can go from zero toroidal moments for the uniform magnetization state to a nonzero toroidal moment when the conical-helix state is reached by reducing the field. The toroidal moment for the bulk case was found to have an oscillatory behavior concerning to the size of the system and is always zero when $L = n\lambda$, where $L$ is the film side, $n$ is an integer, and λ is the period of the texture (Figure 1). These results pave the way to explore other types of textures and coupling with electric fields and their effect on magnetic order.

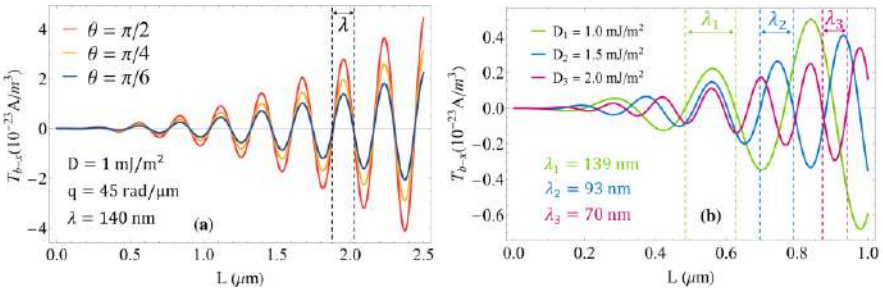



Figure 1: Toroidal moments (x-component) for bulk DMI as a function of the system size ($L$) for: (a) Three magnetization cone angles for a fixed DMI constant (D), pitch vector ($q$) and texture period ($\lambda$). For $\theta = 0$ (saturated state) the toroidal moment is zero. (b) Three values of the DMI constant, which changes the modulus of the pitch vector and the texture periods which phase shift between curves. The other components (y, z) are zero for the bulk case.

## Acknowledgements

The authors acknowledge financial support from ANID through PhD Fellowship No. 2021-21211469, Fondecyt through Grants No. 1201153 and No. 1210607, and the Basal Program for Centers of Excellence through Grant No. AFB180001 CEDENNA, CONICYT.

## References

[1] C. Ríos-Venegas, F. Brrevis, R.A. Gallardo, P. Landeros, *Phys. Rev. B* **105** (2022) 224403.
[2] T. Kimura, *Annual Review of Material Research* **37** (2007) 387-413.
[3] N.A Spaldin, M. Fiebig, M. Mostovoy, *J. Phys.: Condens. Matter* **20** (2008) 434203.
[4] N.A. Spaldin, M. Fechner, E. Bousquet, A. Balatsky, L. Nordström, *Phys. Rev. B* **88** (2013) 094429.

# Interfacial Exchange Coupling in TbFe/[Co/Pd]$_N$ Films

*A.J. Dalmeida*, U. Herr*

*Institute for Functional Nanosystems,Ulm University, Albert-Einstein-Allee 47, Ulm, Germany*

*ashley.dalmeida@uni-ulm.de

The exchange-bias (EB) effect is most commonly observed in ferromagnetic/antiferromagnetic (FM/AFM) bilayers, after cooling down below the blocking temperature of the antiferromagnet in the presence of an external magnetic field and the exchange coupling to the ferromagnet. The effect was discovered by Meiklejohn and Bean in 1956 in CoO/Co nanoparticles and has become an essential ingredient of spin valve giant magnetoresistance (GMR) and tunnelling magnetoresistance (TMR) devices [1-3]. The effect is based on the presence of uncompensated spins of the AFM at the FM/AFM interface which couple to the FM spins. More advanced models consider the formation of interfacial domain walls (iDW) in the FM or AFM layer [4-5]. Exchange bias effects have also been observed in ferrimagnetic/ferromagnetic (FI/FM) and FI/FI bilayers [6-9]. For example, TbFe/[Co/Pt]$_N$ heterostructures have been shown to exhibit exchange bias of up to 8 kOe [9]. Amorphous sputtered TbFe thin films have been intensively studied due to their extraordinary magnetic properties such as high magnetostriction and the existence of a compensation temperature for certain compositions [10]. Sputtered thin films of TbFe show perpendicular magnetic anisotropy (PMA) at compositions around 25 at% of Tb and 75 at% of Fe [11]. More recently studies of TbFe have focussed on its applications in spintronic devices [12-13]. [Co/Pt]$_N$ multilayers show PMA for Co layers thinner than 14Å [14]. Therefore, TbFe /[Co/Pt]$_N$ multilayers can potentially replace the traditional FM/AFM pinned layer of a GMR or TMR device. The PMA of the heterostructure would be beneficial for realization of perpendicular GMR and TMR devices which are potentially packed more densely than traditional GMR and TMR devices with in-plane magnetic anisotropy [3,15].

A related but less studied system are TbFe/[Co/Pd]$_N$ heterostructures. [Co/Pd]$_N$ layers show PMA for Co layers thinner than 8Å [14]. We present investigations of the interfacial exchange coupling in the TbFe/[Co/Pd]$_N$ system for different TbFe compositions and varying thickness of FI and FM layers. The heterostructures were prepared by DC magnetron sputtering from elemental targets onto Si(001) wafers in an UHV chamber. The TbFe alloy was prepared by co-sputtering from elemental targets. The chemical composition of the films was measured using EDX in a Zeiss Leo 1550 SEM. The microstructure was characterized by X-ray diffraction. The interfacial exchange interaction and the magnetization reversal processes are studied by characterizing the heterostructures using vibrating sample magnetometry (VSM), polar-MOKE, and vector-MOKE methods.

## References

[1] W. H. Meiklejohn and C. P. Bean, Physical Review 102, 1413 (1956).
[2] B. Dieny et al., Journal of Applied Physics 69, 4774 (1991).
[3] S. Z. Peng et al., Wiley Encyclopedia of Electrical and Electronics Engineering 1 (2014).
[4] A. P. Malozemoff, Journal of Applied Physics 63, 3874 (1988).
[5] D. Mauri et al., Journal of Applied Physics 62, 3047 (1987).
[6] S. Mangin, G. Marchal, and B. Barbara, Physical Review Letters 82, 4336 (1999).
[7] F. Canet et al., Europhysics Letters (EPL) 52, 594 (2000).
[8] S. Mangin, F. Montaigne, and A. Schuhl, Physical Review B 68, 140404 (2003).
[9] C. Schubert et al., Physical Review B 87, 054415 (2013).
[10] Y. Mimura et al., Journal of Applied Physics 49, 1208 (1978).
[11] C. Prados, E. Marinero, and A. Hernando, Journal of Magnetism and Magnetic Materials 165, 414 (1997).
[12] J. Finley and L. Liu, Applied Physics Letters 116, 110501 (2020).
[13] S. K. Kim et al., Nature Materials 21, 24 (2021).
[14] P. F. Carcia, Journal of Applied Physics 63, 5066 (1988).
[15] S.-E. Lee, T.-H. Shim, and J.-G. Park, NPG Asia Materials 8, e324 (2016).

# Relaxation of Strain in Terbium Iron Garnet Layers Detected by Magneto-optical Kerr Effect Spectroscopy

*Michal Vančík[1]*, Takian Fakhrul[2], Allison Kaczmarek[2], Kensuke Hayashi[2], Jakub Zázvorka[1], Caroline A. Ross[2], Martin Veis[1]*

[1] *Faculty of Mathematics and Physics, Charles University, Prague, Czech Republic*
[2] *Department of Materials Science and Engineering, Massachusetts Institute of Technology, Cambridge, MA, United States*
*vancikm@o365.cuni.cz

Ferrimagnetic iron garnets are promising materials for various applications, such as magneto-optical (MO) isolators, spintronic and spin wave devices. Terbium iron garnet grows on (111) gadolinium gallium garnet substrate under compressive strain, that induces perpendicular magnetic anisotropy (PMA), which is a desired property for various applications. The performance of particular device based on this material usually depends on the thickness of the layer. However, the desired PMA may not be retained in the whole layer above certain thicknesses [1].

To determine the state of the strain in the 264 nm thick TbIG layer, we measured spectral dependence of polar MO Kerr effect (MOKE) hysteresis loops in the range from 1.4 to 4.5 eV. The measurement showed, that the hysteresis loops are composed of two contributions (Fig. 1) in the whole spectral range, which represent the PMA bottom layer, and top layer with relaxed strain. Using the Yeh formalism [2] [3], we obtained the spectral dependence of the off-diagonal elements of permitivity tensor from the saturated state at a magentic field of 1 T. Subsequently, using Yeh formalism again, we separately calculated the magneto-optical response of the strained and relaxed layers (Fig. 2). By matching the position of the spectral peaks, we estimated the thicknesses of the strained and relaxed layers to be 160 nm and 104 nm, respectively. The strain relaxation was further confirmed by the X-ray reciprocal space mapping (RSM) measurement (Fig. 3). The results show that the strain relaxation in thicker TbIG films may cause a magnetic anisotropy separation and field dependent MOKE spectroscopy is a robust and sensitive technique for probing the quality of the films.

## References

[1] Ortiz, V.H. et al. APL Materials 6, 121113 (2018)
[2] Yeh, P. Surface Science 96, 41–53 (1980)
[3] Višňovský, Š. Czech J Phys 36, 625–650 (1986)

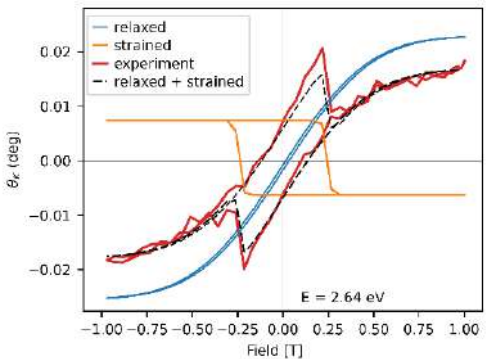



Figure 1: MOKE hysteris loop modelledof 264 nm TbIG modeled as a sum of two hysteresis loops of a strained and relaxed layers.

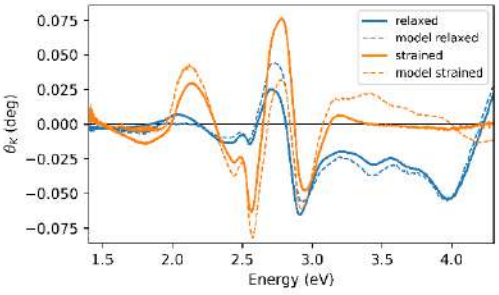



Figure 2: MOKE spectra of the strained and relaxed layers and modeled spectra using Yeh formalism.

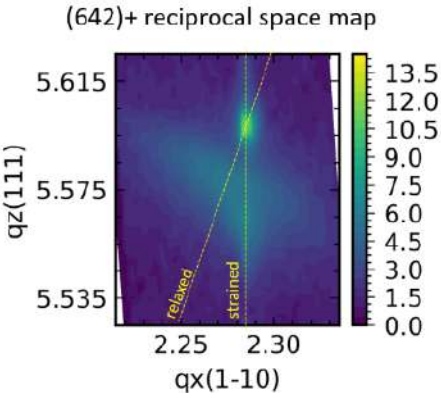



Figure 3: RSM measurement showing a film strain relaxation.

# Magnetic and Dielectric Properties of $CoFe_2O_4$\$BaTiO_3$ Bilayers

*J. Oliveira[1,2]*, B. Silva[1], T. Rebelo[1], P. N. Lekshmi[3], L. Francis[2], J.A. Mendes[1], B.G. Almeida[1]*

[1] *Center of Physics of the Universities of Minho and Porto (CF-UM-UP), Laboratory for Materials and Emergent Technologies (LAPMET), Departamento de Física, Universidade do Minho, Campus de Gualtar, 4710-057 Braga, Portugal*

[2] *Nanostructured Materials Group, International Iberian Nanotechnology Laboratory, Av. Mestre José Veiga s/n, 4715-330 Braga, Portugal*

[3] *Institute of Nanoscience and Nanotechnology and Photonics, LAPMET, Universidade do Porto, Rua do Campo Alegre, 687, 4169-007, Porto, Portugal*

*b8171@fisica.uminho.pt

Nanostructured multiferroic thin films constructed by combining magnetostrictive and piezoelectric materials have attracted much scientific and technological interest [1, 2]. In addition to possessing ferroelectricity and ferromagnetism in each individual phase, they are shown to exhibit stress-mediated coupling between their magnetic and electric properties, called the magnetoelectric effect. Nevertheless, the dynamical magnetic-electric properties have been poorly explored in nanoscopic bilayer magnetostrictive-piezoelectric composite systems. Also, their integration into current silicon technology would further increase their applicability. Barium titanate ($BaTiO_3$ - BTO) is a ferroelectric material with good piezoelectric, pyroelectric, and electro-optical properties. Cobalt ferrite ($CoFe_2O_4$ - CFO) presents a high magnetocrystalline anisotropy and magnetostriction, making it suitable for application in magnetoelectric composite thin films.

Here, bilayer composite thin films, composed of a $BaTiO_3$ layer deposited over a $CoFe_2O_4$ film, have been prepared by laser ablation on highly doped conductive Si (001) substrates. Their structural, dielectric, and magnetic properties were characterized. The X-ray diffraction and Raman spectroscopy measurements performed at room temperature show the presence of the tetragonal ferroelectric structure of $BaTiO_3$ and the cubic spinel structure of $CoFe_2O_4$. The SEM micrographs show dense films, with cobalt ferrite and barium titanate layer thicknesses in the range of 80 nm and 150 – 470 nm for $CoFe_2O_4$ and $BaTiO_3$, respectively. Their dielectric properties were characterized by impedance spectroscopy. The magnetic properties were measured using a SQUID magnetometer. A thickness and interfacial strain-dependent magnetization behaviour was observed. To obtain the relaxation times and activation energies, the electrical permittivity was fitted, using appropriate models for the behaviour of the polarization and including a conductivity contribution. As such, here, the magnetic and dielectric behaviour of the films will be discussed and presented.

## References

[1] N. A. Spaldin, R. Ramesh, Advances in magnetoelectric multiferroics, *Nat. Mater.* **18** (2019) 203–212, https://doi.org/10.1038/s41563-018-0275-2.

[2] A. Aubert, V. Loyau, Y. Pascal, F. Mazaleyrat, M. LoBue, Dynamic Magnetostriction of $CoFe_2O_4$ and Its Role in Magnetoelectric Composites. *Phys. Rev. Applied* **9** (2018) 044035, https://doi.org/10.1103/PhysRevApplied.9.044035.

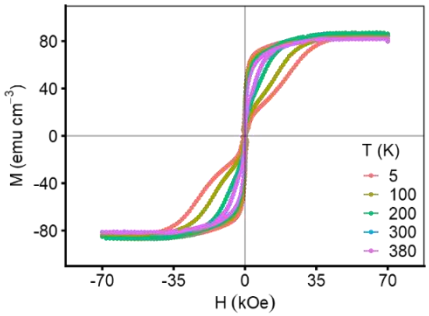



Figure: Temperature-dependent hysteresis loops measured in the $CoFe_2O_4$\$BaTiO_3$ bilayer thin film with layer thicknesses of 80 and 150 nm, respectively.

# Controlling DMI in Magnetic Heterostructures by Electric Fields

*Donya Mazhjoo[1,2]*, Gustav Bihlmayer[1], Stefan Blügel[1,2]*

[1] *Peter Grünberg Institut and Institute for Advanced Simulation, Forschungszentrum Jülich and JARA, 52425 Jülich, Germany*

[2] *Department of Physics, RWTH Aachen, 52056 Aachen, Germany*

*d.mazhjoo@fz-juelich.de

Using external electric fields allows controlling the interfacial Dzyaloshinskii-Moriya interaction (DMI), which opens the way to the realization of spintronic devices with a tuneable DMI. In addition, the magnetocrystalline anisotropy energy (MAE) changes as function of electric field intensity. Graphene (Gr) on ferromagnetic/heavy metals (FM/HM) structures has been proposed for the exploration of novel spin-orbitronic devices as they possess a perpendicular magnetic anisotropy (MA) as well as a sizeable DMI. Experiments reveal that DMI at the Gr/Co interface compensates the spin-orbit coupling induced DMI at the Co/HM interface [1]. Hence, these structures are susceptible to electric fields.

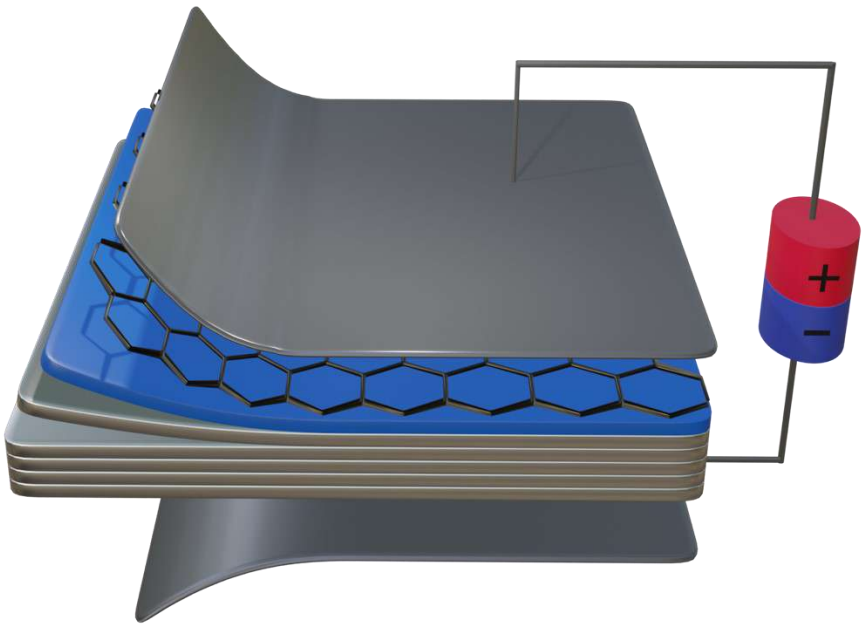
Here, we study the influence of electric fields on Gr/Co/HM structures by using density functional theory (DFT) as implemented in the FLEUR-code [2]. Sandwiching Gr/Co/HM(111) films with different FM and HM thickness between two electrodes of opposite polarity, self-consistent spin-spiral calculations were performed with these boundary conditions shown schematically in the figure below. The spin-orbit effects were considered in first order perturbation theory for the DMI and self-consistently for the MAE. We analyse the influence of the polar oxide BeO on Gr/Co/Pt(111) film within the electric field. We find that the external field gives us control of of spin-orbit induced quantities, in particular variations of the DMI up to 30% can be achieved.

**Acknowledgements**

Support from the FLAG-ERA JTC 2019 grant SOgraphMEM is gratefully acknowledged.

**References**

[1] F. Ajejas et al., *Nano Lett.* **18** (2018), 5364-5372.
[2] https://www.flapw.de



# First Order Reversal Curves of joule and laser annealed Vitroperm and Metglass ribbons

*Elisa Garcia de Blas[1], Victor de Manuel[2], Carolina Martín-Rubio[1], Boris Martín[1], Santiago Martín[1], Rafael P. del Real[3], Ruy Sanz[1]**

[1] *National Institute for Aerospace Technology (INTA), Torrejón de Ardoz, Spain.*
[2] *Institute of Experimental and Applied Physics, Kiel University, Germany.*
[3] *Material Science Institute of Madrid (ICMM, CSIC), Madrid, Spain.*
*sanzgr@inta.es

Nanocrystalline and amorphous magnetic materials [1] due to their soft magnetic properties, are critical elements in key technologies e.g. sensor and transformer cores, magnetic shielding and EMI suppression. Thermal treatments are standard protocols applied in these materials for improving their magnetic properties. However, these thermal treatments can also degrade their magnetic response due to the modification of their structure, i.e. crystallization, generation and precipitation of different phases. The degradation of their magnetic properties is evidenced by the substantial modification of their hysteresis loops. These changes may include a reduction of the magnetization and increasing of the coercive field. Nevertheless, the hysteresis loops provide limited qualitative information about the nature of the modifications. First Order Reversal Curves (FORC) technique that provides valuable information about the type and intensity of the magnetic interactions and, thus complementary data about the nature of the magnetic phases of the material [2],[3].

In this work we have measured and analysed first magnetization curves, hysteresis loops and FORC of joule annealed (Ar atmosphere) nanocrystalline Vitroperm ($Fe_{73.5}Si_{15.5}B_7Nb_3Cu_1$) [4] and laser annealed Metglass 2826MB ($Fe_{40}Ni_{38}Mo_4B_{18}$) [5] ribbons. Some of the obtained FORC diagrams are shown in the Figure 1. Depending on the applied annealing conditions Vitroperm FORC diagram show interactions compatible with the precipitation of FeSi and complex intermetallic phases [4]. For laser annealed Metglass sample, its FORC diagram exhibit a pseudo-single domain distribution pointing-out the crystallization of the material.

**Acknowledgements**

This work was supported by PID2020-115325GB-C31 and PhD fellowship PRE2019-088640, funded by MCIN/AEI/ 10.13039/501100011033.

**References**

[1] G. Herzer, *Acta Materialia*, 61, (2013) 718-734.
[2] L. Zha et al. *AIP Advances* (2021) 11, 015209.
[3] M. Rivas, J. C. Martínez-García, I. Škorvánek, J. Marcin, P. Švec, P. Gorria, *Appl. Phys. Lett.* 107 (2015) 132403
[4] V. de Manuel, V, R. P. del Real, *J. Phys. D: Appl. Phys.* 41 (2008). 085001.
[5] J. Timmerwilke, S.-H. Liou, S. F. Cheng, A. S. Edelstein. *J. Phys. D: Appl. Phys.* 49 (2016) 165005

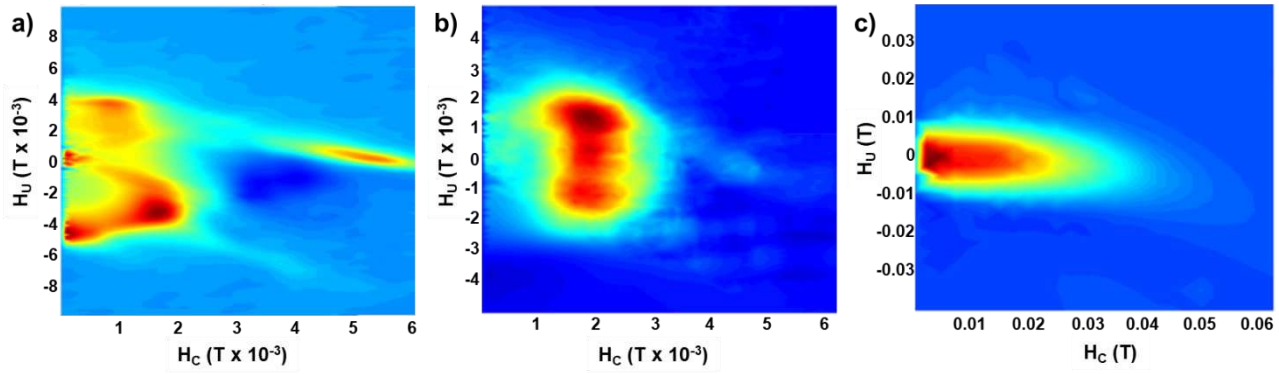



Figure: FORC distributions of: a) 7 A, 60 s in 10 mbar Ar atmosphere Vitroperm b) 7 A, 60 s in 20 mbar Ar atmosphere Vitroperm c) laser annealed Metglass

# Exchange Bias in Bulk Nanocomposites Processed by Severe Plastic Deformation

*M. Zawodzki[1], H. Krenn[2], R. Pippan[1], L. Weissitsch[1],S. Wurster[1], A. Bachmaier[1]**

[1] *Erich Schmid Institute of Materials Science of the Austrian Academy of Sciences, 8700 Leoben, Austria*

[2] *Institute of Physics, University of Graz, 8010 Graz, Austria*

*andrea.bachmaier@oeaw.ac.at

Thermomechanical processing by high pressure torsion (HPT), a severe plastic deformation method, allows the successful synthesis of novel bulk nanocomposites consisting of FeNi-NiO or Ni-NiO. Such phase combinations, where FeNi or Ni is used as ferromagnetic phase and NiO as antiferromagnetic phase, have been predominantly studied in thin films to investigate the magnetic phenomenon of exchange bias (Heb). The origin of Heb is related to a coupling of spins over the phase interface between the ferromagnetic and antiferromagnetic phases, whereas the achieved Heb depends strongly on the FM phase dimensions with a Heb ~1/x behaviour. As a consequence, the phenomenon of Heb is limited to the nanometre regime and vanishes for too large ferromagnetic phase dimensions [1,2].

HPT deformation at elevated temperatures (thermomechanical processing) proved to be the key to obtaining homogeneous bulk nanocomposites. Additionally, the influence of prior ball milling (BM) was studied. X-ray diffraction investigations detected nanocrystalline grain size of the ferromagnetic and antiferromagnetic phases. The obtained bulk nanocomposites extend the magnetic characterisation of Heb from 2D to 3D structures and allow an insight in this novel permanent magnetic material. A tailoring of the Heb was demonstrated for the $Fe_{10}Ni_{40}NiO_{50}$ composition through the application of strain. The steady increase of Heb with applied strain was correlated to the evolving microstructure The influence of NiO content on the deformation process was analysed for the Ni-NiO combination. A strong dependence of the microstructure and magnetic properties on the NiO content could be confirmed, reaching a maximum for the $N_{i50}NiO_{50}$ composition (Fig. 1).

**Acknowledgements**

This project has received funding from the European Research Council (ERC) under the European Union's Horizon 2020 research and innovation programme (grant agreement No:757333).

**References**

[1] W.H Meiklejohn,. C.P. Bean, *Phys. Rev.* **102 (**1956) 1413

[2] T. Blachowicz, A. Ehrmann, *Coatings* **11 (**2021) 122

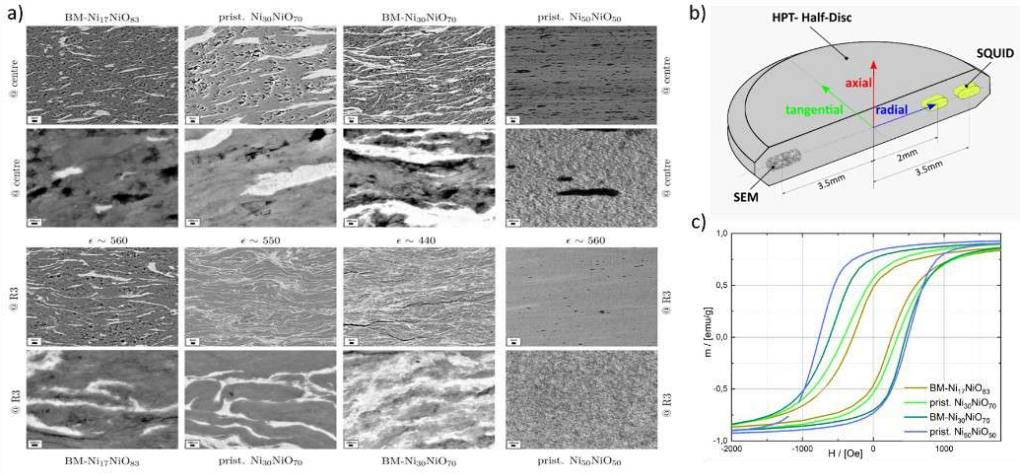



Figure 1: a) Microstructure for various Ni-NiO nanocomposites at similar applied strain. b) Schematic of a HPT half disc. Regions of measurements are highlighted. c) Magnetic hysteresis loops of the nanocomposites depicted in a).

# Anomalous magnetic anisotropy behaviour in Co-rich and Fe-rich glass-coated microwires under applied stress

*Alfonso García-Gómez*[*1,2,3], *Juan M. Blanco*[1,2,3], *Paula Corte-León*[1,2,3], *Mihail Ipatov*[1,2,3], *Álvaro González*[1,2,3] *Julián González*[1,2,3], *Arkady Zhukov*[1,2,3,4] *and Valentina Zhukova*[1,2,3]

[1]*Dept. Polymers and Advanced Mater., Univ. of Basque Country, UPV/EHU, San Sebastian 20018, Spain*

[2]*Dept. Appl. Phys., EIG, Univ. of Basque Country ,UPV/EHU, San Sebastian 20018, Spain*

[3]*EHU Quantum Center, University of the Basque Country, UPV/EHU, Bilbao 48009, Spain*

[4]*IKERBASQUE, Basque Foundation for Science, Bilbao 48011, Spain*

*alfonso.garciag@ehu.eus

The studies of glass-coated magnetic microwires fabricated by the Taylor-Ulitovsky [1] technique has attracted much attention due to their cheap fabrication costs and excellent soft magnetic properties. These properties include the appearance of the giant magnetoimpedance effect GMI, high magnetic field sensibility, magnetic bistability and fast domain wall propagation potentially suitable for the development of the robust magnetic memories and a strong dependence of magnetic properties on applied stress. The magnetic properties of this microwires family is strongly related to the internal stresses distribution due to the fabrication technique involving rapid melt quenching, producing internal stresses within the metallic core, the glass-coating and the interface between them. This interactions along with applied stresses and relaxations generated by annealing has been proved to affect significantly the domain structure and magnetic anisotropy and the magnetostriction coefficient $\lambda_s$ of glass-coated microwires. Accordingly, the tunability of magnetic properties makes glass-coated magnetic microwires a perfect candidate for their integration in health monitorization devices for structures under high applied stresses.

In this work we studied the variation in the value of $\lambda_s$ in as-prepared and annealed at different temperatures Co-rich ($\lambda_s \sim 0$) and Fe-rich ($\lambda_s > 0$) [2] microwires with applied stress. This is done by measuring the coercitive field Hc while applying longitudinal stress and the $\lambda_s$–value by Small-angle magnetization rotation method in several samples previously annealed at different temperatures.

## References

[1] Baranov, S.A.; Larin, V.S.; Torcunov, A.V. Technology,. *Crystals* 7 (2017) 136.

[2] Zhukov, M. Churyukanova, S. Kaloshkin, V. Semenkova, S. Gudoshnikov, M. Ipatov, A. Talaat, J. Blanco, and V. Zhukova, *J. Alloys. Compund. X***7** (2017) 031037.

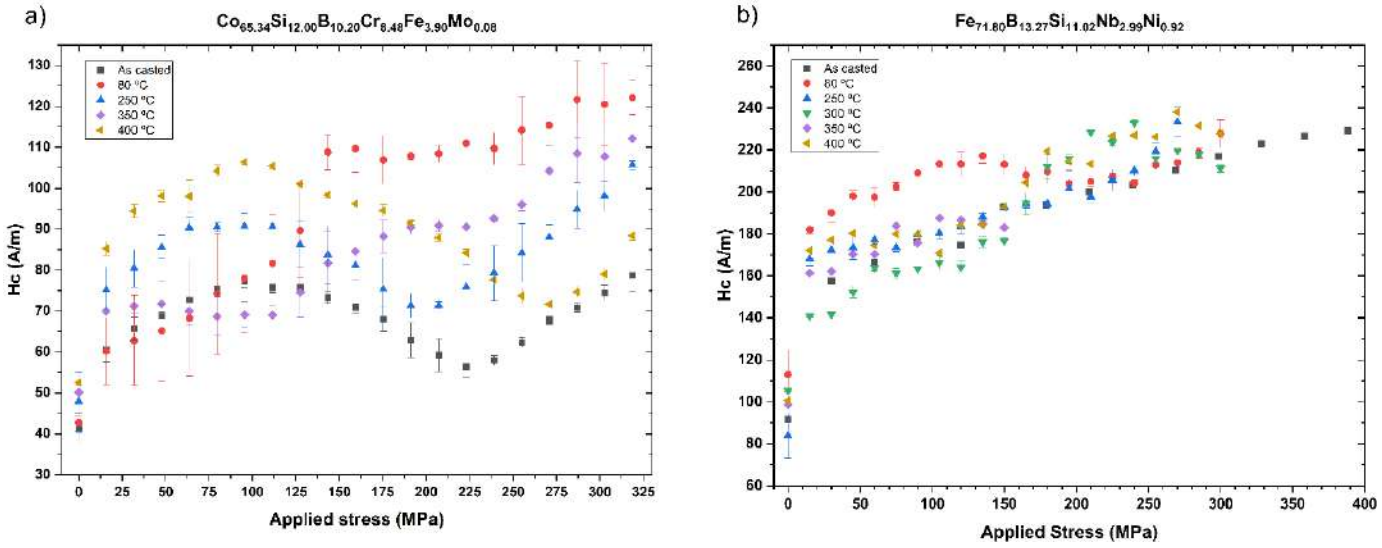



Figure: Coercitive field evolution with applied stress of ***a)*** a Co-rich microwire ($d_{core} = 17.5\ \mu m, D_{wire} = 22.2\ \mu m$) ***b)*** and Fe-rich microwire ($d_{core} = 15.9\ \mu m, D_{wire} = 24.5\ \mu m$).

# Interplay between Microstructural and Magnetic Properties in Nanostructured $R_2Fe_{17}$ (R = Pr, Nd) Ribbons

*J.L. Garrido-Álvarez[1]*, P. Álvarez-Alonso[2], C.F. Sánchez-Valdés[3], P. Gorria[2], J.L. Sánchez Llamazares[1,4]*

[1]*Departamento de Física, Universidad de Oviedo, Calvo Sotelo s/n, 33007 Oviedo, Spain*
[2]*IUTA, Universidad de Oviedo, 33203 Gijón, Spain*
[3]*Depto de Física y Matemáticas, División Multidisciplinaria, Ciudad Universitaria, Instituto de Ingeniería y Tecnología, Universidad Autónoma de Ciudad Juárez (UACJ), Ciudad Juárez, Chihuahua 32310, México*
[4]*Instituto Potosino de Investigación Científica y Tecnológica A.C., Camino a la Presa San José 2055, Col. Lomas 4ª, San Luis Potosí, S.L.P. 78216, México*
*garridojose@uniovi.es

We have studied the interplay between the microstructural and magnetic properties of rapidly solidified $Pr_2Fe_{17}$ and $Nd_2Fe_{17}$ melt-spun ribbons. XRD analyses confirm that the main phase in samples exhibits the rhombohedral $Th_2Zn_{17}$-type crystalline structure [1], which coexists with a small amount of impurity α-Fe phase (~ 1-4 % wt.). SEM and HRTEM analyses reveal that the $Pr_2Fe_{17}$ and $Nd_2Fe_{17}$ ribbons are composed by nanograins, with mean sizes of 15 and 13 nm, agglomerated to form averaged in size nanoparticles of 70 and 40 nm, respectively. Nanocrystallites are surrounded by a poorly crystalline intergranular phase which causes the presence of two successive magnetic transitions in the low-field thermomagnetic $M(T)$ curves at 290, 323 K and 326, 350 K for $Pr_2Fe_{17}$ and $Nd_2Fe_{17}$ ribbons, respectively. The temperature of the low temperature transition corresponds with the reported for bulk 2:17 crystalline phases, according to previous studies [2], while the higher temperature transitions are related to the poorly crystalline phases. This phase separation at the nanoscale, which is indicative of a significant atomic disorder, is also responsible for the broadening in the temperature region of the second-order ferro-to-paramagnetic transition. The latter, causes a remarkable increase of the full-width at the half-maximum of the magnetic entropy change curve $\Delta S_M(T)$, in comparison with the reported for bulk alloys [3], which becomes table-like in shape (see Fig.1).

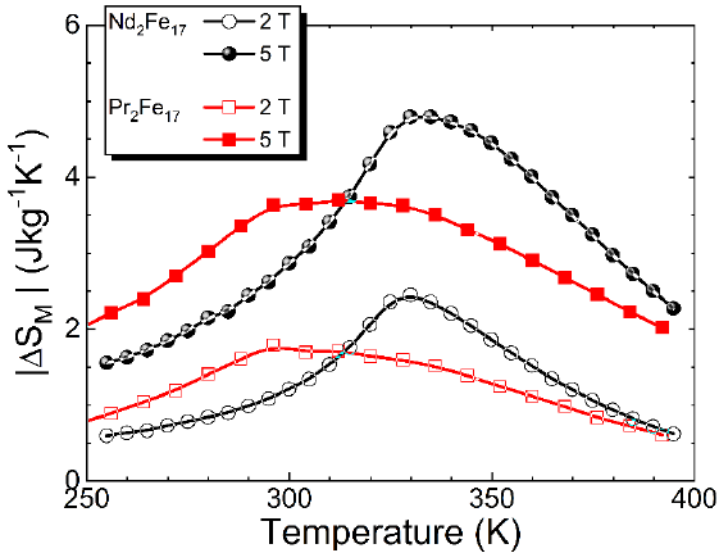



Figure 1. $\Delta S_M(T)$ *curves* for magnetic field changes of 2 and 5 T for $Pr_2Fe_{17}$ and $Nd_2Fe_{17}$ melt-spun ribbons.

## Acknowledgments

Work supported by MCIU, AEI, European FEDER (MCIU-19-RTI2018- 094683-B-C52), Principado de Asturias (SV-PA-21-AYUD/2021/51822), SEP-CONACyT, Mexico (project A1-S-37066), and LINAN (IPICyT). J.L.S.LL. acknowledges the support received from the EU-NGEU, MIU, and Plan de Recuperación, Transformación y Resiliencia (Maria Zambrano program; Ref: MU-21-UP2021-030 71741542).

## References

[1] Q. Johnson, D.H. Wood, G.S. Simth, A.E. Ray, *Acta Cryst.* B**24** (1968) 274-276.
[2] X.C. Kou *et al.*, *J. Magn. Mater.* **177** (1998) 1002-1007.
[3] P. Álvarez-Alonso *et al.*, *Acta Phys. Pol. A* **118** (2010) 867-869.

# Magnetic anisotropy of Au/$Fe_{0.75}Co_{0.25}$/Au thin films: Combined experimental and first-principles study

*Hubert Głowiński[1], Piotr Kuświk[1], Justyna Rychły-Gruszecka[1], Justyn Snarski-Adamski[1], and Mirosław Werwiński[1]**

[1] *Institute of Molecular Physics, Polish Academy of Sciences, Smoluchowskiego 17, 60-179 Poznań, Poland*

*werwinski@ifmpan.poznan.pl

Magnetic thin films of FeCo alloy with a thickness of about 1 nm are currently being used in spintronic devices, such as magnetic tunnel junctions and spin valves [1,2]. In this work, we present the experimental and computational results for $Fe_{0.75}Co_{0.25}$ films in the thickness range from zero to two nanometers, surrounded by gold layers. We found a strong contribution of surface magnetic anisotropy to effective magnetic anisotropy in the Au/$Fe_{0.75}Co_{0.25}$/Au system. Although the saturation magnetization of the $Fe_{0.75}Co_{0.25}$ alloy is one of the largest known, the surface contribution to the magnetic anisotropy is strong enough to overcome the shape magnetic anisotropy, as we observed a rectangular hysteresis loop in the out-of-plane configuration. The theoretical model developed within the framework of density functional theory allows for a 1:1 atomic scale representation of the considered FeCo layer thickness range and an unambiguous representation of its chemical composition [3]. Based on the calculations, we determined the details of the geometry of the system, magnetic moments, magnetocrystalline anisotropy energies, the thickness ranges of the occurrence of perpendicular magnetic anisotropy, as well as the effect of film thickness and the presence of a substrate and cap layers on the direction of the magnetization easy axis. Calculations reveal an irregular oscillatory dependence of magnetic anisotropy with a change in FeCo layer thickness every single atomic monolayer starting from one monolayer, nevertheless averaging over several thickness values leads to a close to linear dependence of magnetic anisotropy on thickness similar to the experimental results, see Figure 1. Although extrapolation of the calculation result also suggests a transition to perpendicular magnetic anisotropy for the thinnest layers, the theoretical relationship is shifted downward, most likely due to the infinitely periodic character of the model (no shape anisotropy).

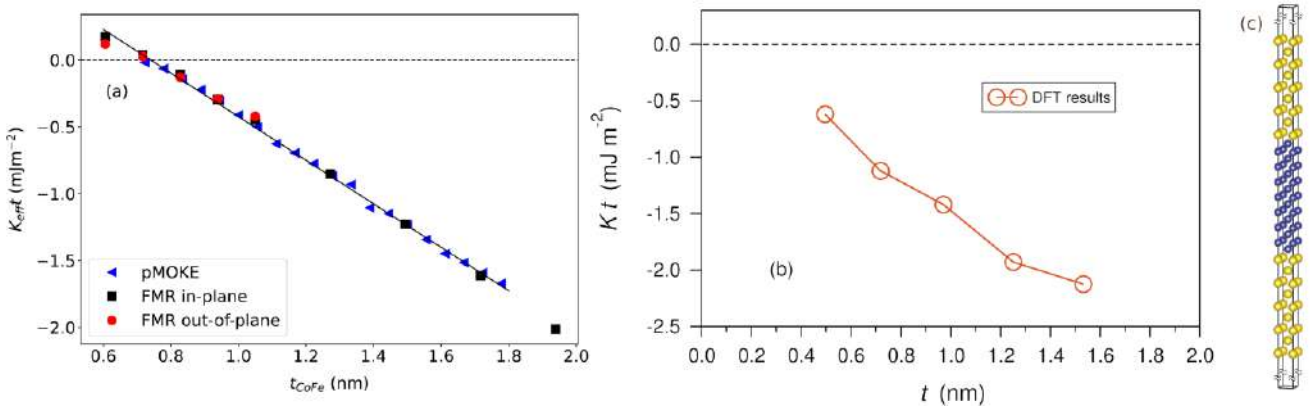



Figure 1: (a, b) The experimental and calculated dependence of the product of effective magnetic anisotropy constant and film thickness ($K_{eff}\,t$) on film thickness ($t$) for the $Fe_{0.75}Co_{0.25}$ thin films. (c) The exemplary computational model of Au/$Fe_{0.75}Co_{0.25}$/Au heterostructure with about 2 nm thickness of FeCo central layer.

## Acknowledgements

We acknowledge the financial support of the National Science Center Poland under the decisions DEC-2018/30/E/ST3/00267 (SONATA-BIS 8) and DEC-2021/41/B/ST5/02894 (OPUS 21).

## References

[1] H. Głowiński, *et al.*, Influence of adjacent layers on the damping of magnetization precession in $Co_xFe_{100-x}$ films, J. Alloys Comp. 785 (2019) 891–896.

[2] P. Kuświk, *et al.*, Colloidal domain lithography for regularly arranged artificial magnetic out-of-plane monodomains in Au/Co/Au layers, Nanotechnology. 22 (2011) 095302.

[3] M. Werwiński, *et al.*, Magnetocrystalline anisotropy of $Fe_5PB_2$ and its alloys with Co and 5d elements: A combined first-principles and experimental study, Phys. Rev. B. 98 (2018) 214431.

# Optimization of $CoFe_2O_4$ / Ba:Pb($Zr_xTi_{1-x}$)$O_3$ bilayers for magnetoelectric applications

*Federica Luciano[1,2]*, Stefan De Gendt[1,2], Florin Ciubotaru[2], Christoph Adelmann[2]*

[1] *KU Leuven, Celestijnenlaan 200F, Leuven, Belgium*

[2] *IMEC, Kapeldreef 75, Leuven, Belgium*

*Federica.Luciano@imec.be

Magnetoelectric (ME) materials have received much attention in recent years for their fundamental properties and application potential. The ME effect describes the coupling between ferro- or antiferromagnetic order with dielectric or ferroelectric polarization in solids. The research interest has focused overwhelmingly on the control of the magnetization by electric fields [1]. By comparison, much less work has been devoted to the inverse ME effect, especially at the nanoscale. In our work, we combine piezoelectric and magnetostrictive or piezomagnetic materials (layers), in which the ME coupling occurs indirectly via stress and voltages can be generated as a response to magnetization changes.

Among magnetostrictive phases, $CoFe_2O_4$ (CFO) stands out because of its high magnetostrictionas well as a finite piezomagnetic coefficient [2]. For ME applications, we have combined CFO with Ba-substituted PZT [$(Ba_{0.1}Pb_{0.9})(Zr_{0.52}Ti_{0.48})O_3$] (BPZT) as the piezoelectric material in bilayers with nm thickness. The bilayer stacks have been deposited by pulsed laser deposition (PLD) on (1 1 1)-oriented Pt (70nm) bottom electrode (BE) and Si/SiO2 (400nm) substrates. Chemical mechanical polishing (CMP) was performed to reduce the BPZT roughness by an order of magnitude to < 1nm [F. Luciano et al. (2023) - unpublished]. Subsequently, 50 nm thick CFO was deposited on both as deposited (AD) BPZT and after CMP. The properties of CFO on BPZT were then compared to CFO deposited directly on a Pt BE. The deposition parameters have been controlled to obtain optimized crystalline structure, magnetic properties, and stoichiometry.

Vibrating sample magnetometry analysis has been used to access the hysteresis loop and to measure the saturation magnetization (Ms) [Fig. 1a] for all CFO layers (AD BPZT, on CMPed BPZT, on Pt). Values of $Ms_{AD}$ = 80KA/m, $Ms_{CMP}$ = 65KA/m, and $Ms_{Pt}$ =125KA/m, were obtained. The lower saturation magnetization for CFO on BPZT can be linked to the (poly)crystalline structure of CFO as measured by x-ray diffraction analysis [Fig. 1b]. The results show that CFO deposited on Pt follows the same (1 1 1) orientation of the BE, which is not the case on BPZT. Further optimization is ongoing to improve the CFO quality on BPZT, *e.g.* by using (oxide) interlayers to improve crystallinity. Additionally, an compositional depth profiling by time-of-flight secondary ion mass spectrometry of CFO/BPZT bilayer and x-ray photoelectron spectroscopy found negligible intermixing of CFO and BPZT and indicated that the layers had the expected stoichiometry. The analysis of ferroelectric and piezoelectric properties of the BPZT layer after CFO deposition showed no deterioration, rendering this stack a good candidate to assess the (inverse) ME coupling.

F. L. acknowledges funding by *The Research Foundation – Flanders (FWO)* – grant number 1183722N.

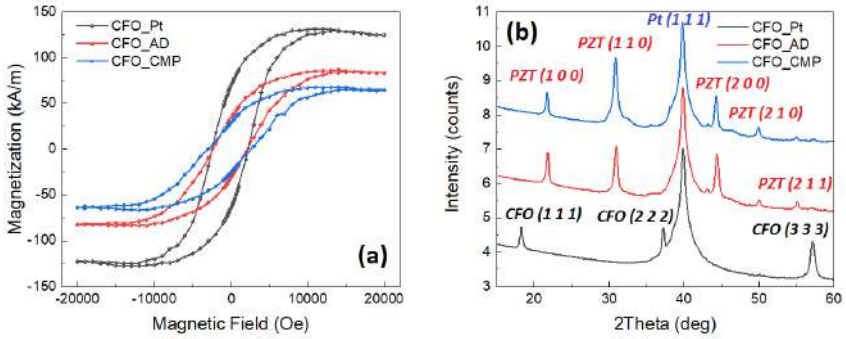



Figure 1: Magnetic hysteresis loop (M-H) (a) and XRD spectrum (b) of $CoFe_2O_4$ deposited on BPZT and Pt

## References

[1] Nan, C.-W., et al. *Journal of Applied Physics* **103.3** (2008): 1.

[2] Aubert, Alex, et al. *IEEE Transactions on Magnetics* **53.11** (2017): 1-5.

# Composition and Thickness Dependent Magnetic Properties of Co/Tb-Co Bilayers

*D. Kiphart*, F. Stobiecki, Ł. Frąckowiak, M. Matczak, , M. Schmidt, G.D. Chaves O'Flynn, P. Kuświk*
*Institute of Molecular Physics, Polish Academy of Sciences, Poznań, Poland*
*dkiphart@ifmpan.poznan.pl

The presence of two magnetic sublattices in ferrimagnetic RE-TM films (RE-rare earth, e.g., Tb, TM - transition metal, e.g., Co) allows the magnetic properties to be tuned by changes to their composition (e.g. magnetization, coercivity, as well as compensation and Curie temperatures). Here, we demonstrate that the addition of a Co underlayer constitutes a convenient method to manipulate magnetic properties of Tb-Co alloy films.

This talk describes studies of several Co/Tb-Co bilayer systems deposited by magnetron sputtering. The Tb-Co alloy layers were prepared with mutually perpendicular gradients of Tb concentration ($16 \leq c_{Tb} \leq 59$ at. %) and thickness ($3 \leq t_{alloy} \leq 10$ nm) as described in Ref. [1]. The Co thicknesses ($t_{Co}$) were kept uniform ($t_{Co}$ = 0.5, 1.0, 2.0, 4.0 nm). The magnetic characterization of samples was performed using PMOKE magnetometry.

The measurements were used to determine the range of parameters ($t_{Co}$, $t_{alloy}$, $c_{Tb}$) for which this system has Perpendicular Magnetic Anisotropy and exhibit simultaneous magnetization reversal of the Co and the alloy layers. We demonstrate that the switching fields of the bilayers can be tailored by appropriate choice of $t_{Co}$. The addition of the Co underlayer shifts the concentration for which the Tb and Co sublattices in the alloy are compensated at room temperature towards higher *cTb*. Surprisingly, at higher $c_{Tb}$, a second transition point occurs where the Co sublattice dominates. This transition is found for $c_{Tb} \gtrsim 40\%$ , for which we expected that Tb-Co layers are paramagnetic at room temperature [2]. To understand this type of transition we performed atomistic simulations on Vampire [3]. These simulations show that, in Co/Tb-Co bilayers, a small part of the Tb-Co layer exhibits spontaneous magnetization close to the Co/Tb-Co interface. This indicates that the newly observed transition results from the spin polarization of the paramagnetic Tb-Co layer because of a proximity effect which causes the system to behaves as a ferrimagnet with Co domination.

**Acknowledgements**
The work was supported by National Science Centre Poland under OPUS funding Grant No. 2020/39/B/ST5/01915

**References**

[1] Ł. Frąckowiak, et al., *J. Magn. Magn. Mater.* **544** (2022) 168682.
[2] P. Hansen, et al., *J. Appl. Phys.* **66** (1989) 756.
[3] R F L Evans, et al., *Phys.: Condens. Matter* **26** (2014) 103202.

# Temperature Dependent Exchange Interaction in $Fe_{1-x}V_x$

*J. Solano[1]*, J. Thoraval[1], J. Robert[1], B. Gobaut[1], Y. Henry[1], D. Halley[1], M. Bailleul[1]*

[1] *Institut de physique et de chimie des Matériaux de Strasbourg, CNRS-Université de Strasbourg, Strasbourg, France*

*jose.solano@icms.unistra.fr

In metallic ferromagnets, the exchange interaction shapes both their magnetic and electron-transport properties. Late developments in computational physics [1, 2] have attempted to expand on the earlier modelling [3, 4] of such phenomena, but incomplete understanding and characterization of the complex interplay between the electron and magnetic systems makes progress difficult. In our study we take an experimental approach to these problems by combining two different temperature dependent measurements in single-crystalline $MgO/Fe_{1-x}V_x/MgO$ thin films: spectroscopy of exchange spin wave modes through broadband ferromagnetic resonance (FMR) and magnetoresistance measurements at high magnetic fields.

For the first experiment [Figure 1 (a)], we build upon the characterization of MgO/Fe/MgO films [5] to probe inhomogeneous magnetization dynamics and extract the spin-wave exchange stiffness *D* as a function of temperature (5K-400K). We observe that *D* follows a polynomial law ([Figure 1(c)]) that agrees well with a model of magnon mass renormalization produced by both electron and magnon interactions, as expected in itinerant ferromagnets.

The second experiment [Figure 1 (b)] measures the longitudinal magnetoresistance as function of applied magnetic field and temperature (300-400K). We observe a decrease of the resistivity due to the reduction of the magnon magnetoresistance component by the application of large magnetic fields [6]. We fit this magnetoresistance with a model that takes into account the exchange mediated electron-magnon scattering [3]. From this analysis, we also estimate *D* as a function of temperature, and we observe that it has a different temperature dependence compared ([Figure 1(c)]) to our result from magnetization dynamics.

Finally, we argue that our former result is a more accurate description of the spin-wave exchange stiffness of the system. We ascribe the disagreement between the two results to the crudeness of our transport model. We suggest that this could be improved using temperature dependent exchange coupling constants $J_{ij}(T)$ between the i, j=s, d electron bands.

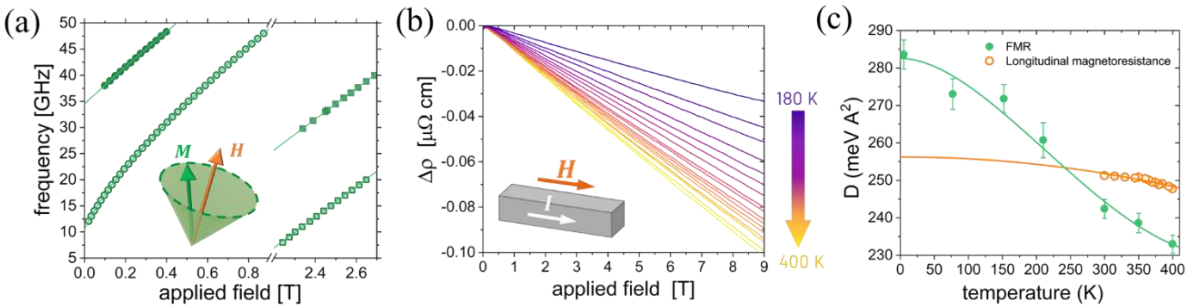



Figure 1: a) Broadband FMR spectra for a 20nm thick $Fe_{94}V_6$ film at 300 K. b) Longitudinal magnetoresistance as a function of applied magnetic field and temperature. c) Extracted spin-wave exchange stiffness as function of temperature.

## Acknowledgements

We acknowledge technical support from the STnano and XRD platforms, in particular: Hicham Majjad, Romain Bernard and Marc Lenertz.

## References

[1] D. Alfè, Phys. Rev. B. **85**, (2012) 024102.
[2] R. Moreno, Phys. Rev. B. **94**, (2016) 104433.
[3] D. Goodings, *Phys. Rev.* **132** (1963) 2.
[4] A. Fert, J. Phys. C. **2** (1969) 2.
[5] J. Solano, et al, *Phys. Rev. Mat.* **6** (2022) 124409.
[6] B. Raquet, et al., Phys. Rev. B. **66** (2002) 024433.

# Laser-Assisted Antiferromagnetic-Ferromagnetic Transition in FeRh

*C. Gourdon[1]*, K. Alhada-Lahbabi[1], G. Olivetti[1], V. Uhlíř [2], J.A. Arregi [2], M. LoBue[3], M. Almanza [3], A. Pecheux [3], D. Fournier [1], L. Thevenard [1]*

[1] *Institut des Nanosciences de Paris, Sorbonne Université, CNRS, 4, Place Jussieu, 75005 Paris, France*
[2] *CEITEC BUT, Brno University of Technology, Purkyňova 123, 612 00 Brno, Czech Republic*
[3] *Université Paris-Saclay, ENS Paris-Saclay, CNRS, SATIE, 91190 Gif-sur-Yvette, France*
*gourdon@insp.jussieu.fr

Among magnetic materials near equiatomic FeRh alloys, with their temperature driven antiferromagnetic (AF) to ferromagnetic (FM) hysteretic transition near room temperature were studied long ago in bulk form. In the recent years they have aroused a renewed interest as thin films with particular focus on their AF nature at room temperature, the electrical detection of their magnetic state, and their giant magneto-caloric effect (MCE) [1]. FeRh is also a good candidate to investigate laser-assisted transient or irreversible phase transition. Most experiments have used ultrashort excitation/detection schemes either all optical [2], or with optical pump and X-rays or photoemission probe [3]. However, at short time scale electron, spin and phonon dynamics are entangled whereas at much longer timescale (µs range) only thermal effects are at play.

We investigate the AF-FM transition using a modulated reflectance experiment [4], taking advantage of the 4% reflectivity difference between the AF and FM phases. In the optical microscopy setup the pump beam (532 nm laser with square modulation at 100 kHz) and the probe beam (488 nm laser) are focused on the same spot. Two components (at $f_0$=0 (CW) and $f_1$=100 kHz) of the reflected probe are detected while the temperature is scanned across the transition. The $f_0$ reflectance shows the modification of the hysteresis cycle with increasing pump power. The average transition temperature shifts by 2.2°C per mW of pump power (Fig.(a)), consistently with the thermal conductivities of the two phases, which are determined in this work, while the hysteresis loop shape is strongly affected. Under pump heating the nucleation of the FM phase occurs at lower temperature whereas the FM-to-AF transition is hardly affected (Fig.(a)). At about 4 mW of pump power hysteresis reduces, and eventually disappears, which is not the result of irreversible damage since removing the pump restores the initial cycle. This on-demand suppression of the hysteresis while maintaining the AF and FM reflectance (hence the magnetization in the FM phase) might be of great use for magneto-calorics. The $f_1$ signal varies linearly with pump power in the in pure FM and AF phases where it can be ascribed to thermoreflectance only (dashed curves in (c,e)). In the mixed AF-FM phase it shows a strongly non-linear behavior with pump power. We show that it is consistent with breathing FM domains at the modulation frequency. The AF-FM domain wall velocity, smaller than 1 m/s, can then be estimated, which opens new insight for the yet little investigated AF-FM interfaces.

## Acknowledgements

This work was funded by the French Agence Nationale de la Recherche (project ANR-20-CE30-0027) and the LabEx LaSIPS (ANR-10-LABX-0032-LaSIPS) under the program (ANR-11-IDEX-0003).

## References

[1] X. Marti *et al.*, Nat. Mater. 13, 367 (2014), S. Cervera et al., Phys. Rev. Mater. 1, 065402 (2017)
[2] G. Ju *et al.*, Phys. Rev. Lett. 93, 197403 (2004),
[3] S. O. Mariager *et al.*, Phys. Rev. Lett. 108, 087201 (2012), F. Pressacco *et al.*, Nat. Com.., 12, 5088 (2021)
[4] D. Fournier, M. Marangolo, and C. Fretigny, J. Appl. Phys., 128, 241101 (2020)

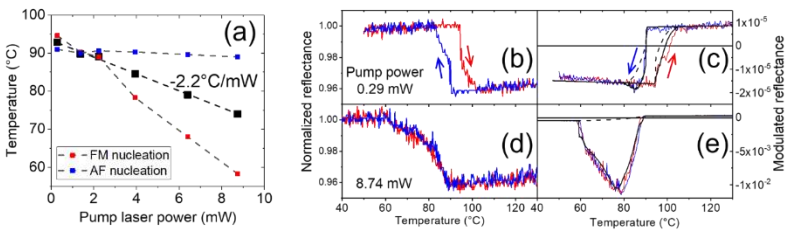



Figure: (a) Transition and nucleation temperatures, $f_0$ (b,d) and $f_1$ (c,e) reflectance and fits for different pump powers

# Growth Of Graded Composition FeRh Over GaAs for Piezoelectric Acoustic Wave Excitation

*D. Nguyen[1], G. Olivetti[1], A.K. Vythelingum[1], C. Gourdon[1], A. Castellano[2], M. J. Verstraete[2], V. Laude[3], L. Thevenard[1]**

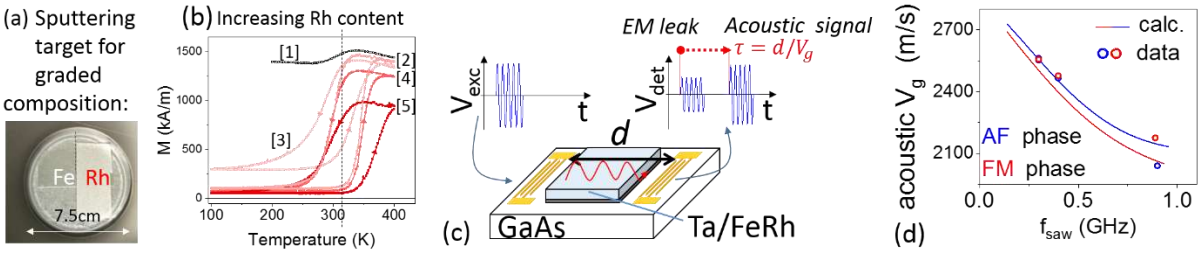
[1] *Institut des Nanosciences de Paris, Sorbonne Université, CNRS, 4 place Jussieu, Paris, France*
[2] *NanoMat/Q-Mat/CESAM Université de Liège, and European Th. Spec. Liège,5 Belgium*
[3] *FEMTO-ST, TEMIS,15B avenue des Montboucons, 25030 BESANCON cedex,France*
*thevenard@insp.jussieu.fr

FeRh is a room-temperature antiferromagnet (AF) which undergoes a spectacular first order transition to a ferromagnetic (FM) state, with a large 0.7 % volume increase. Discovered almost 90 years ago, it is generating renewed interest for spintronics and magnetocaloric energy-harvesting applications, but also for fundamental studies of laser-triggered phase changes [1]. Films are generally grown over lattice-matched non-piezoelectric substrates (MgO and $Al_2O_3$), and more rarely on piezoelectric but brittle materials (PZT or PMN-PT), an interesting approach to control the magnetic phase via static strain [2]. Here we show that we can grow FeRh over a piezoelectric substrate, GaAs, with which we excite Rayleigh surface acoustic waves (SAWs). Combined with the good static and dynamic magnetic properties of the polycrystalline FeRh layer deposited, this new system is appealing to excite ferromagnetic resonance remotely using magnetoacoustics [3].

Using magnetron sputtering, we grow FeRh(270nm)/Ta(100nm)/GaAs(001) films using a graded composition approach (Fig. a). This yields in a single growth a series of samples of varying Rh concentration (#1->5 in Fig. b), among which a few will always present the right Fe/Rh stoichiometry for room-temperature AF behavior, independently of the history of the target. Using optical microscopy [4], we show that AF and FM domains coexist on a very small scale, below our ~µm microscopy resolution. Static hysteresis cycles show a coercivity close to that encountered in epitaxial MgO, with a similar increase upon cooling down from the FM to the mixed AF/FM phase. Ferromagnetic resonance evidences a very weak magnetic anisotropy, fairly large linewidths, and a hysteretic low-field microwave absorption often found in thick metallic compounds. Finally, we deposit transducers on either side of a FeRh/GaAs mesa in order to excite SAWs (Fig. c). Their group velocity is measured by the transit time τ of the acoustic signal transmitted through the FeRh. The average dispersion $V_g$ versus $f_{SAW}$ is reproduced decently by calculations based on elastic constants determined by anharmonic phonon DFT calculations (Fig. d), but not relative values of velocities in the AF and FM phases.

## References

[1] Marti *et al*, *Nat. Mat.* **13** 367 (2014), Thiele *et al*, *Appl. Phys. Lett*. **85** 2857 (2004), Cervera *et al*, *Phys. Rev. Mat.* **1** 065402 (2017)
[2] J. A. Arregi, *et al*, J. Physics D: Applied Physics **51**, 105001 (2018), J. D. Clarkson *et al.*, Sci. Rep. **7**, 15460 (2017).
[3] A. Vythelingum *et al* JEMS2023 abstract, Thevenard *et al*, *Phys. Rev. B.* **90** 094401 (2014)
[4] Saidl *et al New J. Phys*., **18** 083017 (2016)




Figures: (a-b) A piece of Rh on a Fe target faces a non-rotating sample holder, inducing a gradient of Rh concentration (samples #1->5) from which an evolution from FM to AF room-temperature behaviour is obtained (dashed line in VSM magnetometry curves in b). (c) Interdigitated transducers excited by a pulsed rf signal generate SAWs. The velocity is estimated in the AF and FM phases by measuring the delay of the acoustic signal with respect to the electromagnetic (EM) leak signal, and compared to calculations using first-principles-determined elastic constants of FeRh.

# Optimizing the magnetic properties of MM-FeCo-B Ribbons by Addition of Si, Ti, Ta Elements

*M. Grigoras, M. Lostun, M. Porcescu, G. Stoian, N. Lupu*

*National Institute of Research & Development for Technical Physics, 700050 Iasi, Romania*

*mgrigoras@phys-iasi.ro

Currently, the permanent magnet market is dominated by Nd-Fe-B based magnets, which are critically dependent on the rare earth elements Nd / Pr / Dy / Tb. At the same time there is a significant imbalance in the rare earth element market due to the fact that while Nd and Pr represented 75% of the REE market value, they represented only 20% of the volume. On the other hand, La and Ce represented around 70% of the volume but only 8% of the value [1]. Therefore, due to the high price and low supply of Nd, many researchers have tried to replace it using low-priced rare earth metals such as Ce or La [2, 3]. On the other hand, the extraction and separation of rare earths are processes that lead to environmental pollution [4]. Replacing of Nd, by Misch-metal (MM), can lead to a balanced use of rare earth elements, reduce the price of the magnet, and reduce environmental pollution. However, the total replacement of Nd with MM leads to the degradation of Curie temperature (Tc), saturation magnetization (Ms), coercivity (Hc) and as a result the energy product (BH)max of MM-Fe-B alloys, which still remains superior to hard ferrites, making of these alloys a potential candidate for filling the energy gap between ferrites and NdFeB type magnets. But of course there are continuous concerns about improving their magnetic properties.

In this work, we report the effect of Fe substitution by medium-high melting point elements such as Si, Ti, and Ta on the hard magnetic properties of $MM_{16}Fe_{76-x}Co_2M_xB_6$ ribbons, where M= Si, Ti, Ta, and $0<x<2$. The structure, magnetic properties, exchange interactions and phase transition temperatures of annealed $MM_{16}Fe_{76-x}Co_2M_xB_6$ ribbons were investigated methodically. It is found that the addition of a critical amount of: *(i)* Si, has the effect of refining the microstructure, decreasing the lattice constants of the 2:14:1 phase increasing the anisotropy field and exchange interaction between Fe atoms, resulting in improved coercivity, remanence, and the Curie temperature, *(ii)* Ti, increases the coercivity by suppressing the formation of the α-Fe phase, inhibiting grain growth and forming pinning centers, however, at the same time the remanence decreases slightly, *(iii)* Ta, refines the grains and reduces the volume fraction of non-magnetic phases, as a result the coercivity, remanence and $(BH)_{max}$) of the alloys are improved, however the Curie temperature remains unchanged. The saturation magnetization decreased for all types of additions used in this study.

The optimized magnetic properties of: *(i)* coercivity Hc=8.7 kOe, remanence magnetization Mr=81 emu/g, $(BH)_{max}$= 12.33 MGOe and Curie temperature Tc= 254 ºC were obtained for the $MM_{16}Fe_{75}Co_2Si_1B_6$ ribbons, *(ii)* coercivity Hc=9.8 kOe, remanence magnetization Mr=72 emu/g and $(BH)_{max}$= 10.23 MGOe were obtained for the $MM_{16}Fe_{74.5}Co_2Ti_{1.5}B_6$ ribbons, and *(iii)* coercivity Hc=9.1 kOe, remanence magnetization Mr=83 emu/g and $(BH)_{max}$= 12.84 MGOe were obtained for the $MM_{16}Fe_{75.4}Co_2Ta_{0.6}B_6$ ribbons. This work shows that the magnetic properties of MM-FeCo-M-B alloys can be improved by optimizing the addition element (M) content.

## Acknowledgements

This work was supported by a grant of the Ministry of Research, Innovation and Digitization, CCCDI - UEFISCDI, project number PN-III-P2-2.1-PED-2021-1386, within PNCDI III.

## References

[1] G. Blengini, et al. *European Commission, Critical raw materials factsheets* (2020) doi:10.2873/92480.
[2] M. Grigoras et al, *J. Magn. Magn Mater.* **432** (2017) 119.
[3] Z. Y. Zhang et al, *J. Alloys Compd.* **859** (2021) 157780.
[4] Le Zhou and Jianping Ge, *The Extractive Industries and Society*, **8** (2021) 340-354.

# Direct Imaging of Magnetoelectric Coupling in $La_{0.9}Ba_{0.1}MnO_3/BaTiO_3$ Multiferroic Heterostructures

*F. Stramaglia [1], G. Panchal [1], L. Tovaglieri[2], C. Lichtensteiger[2], F. Nolting[1], C. A. F. Vaz [1]**

[1]*Paul Scherrer Institute, Forschungsstrasse 111, 5232 Villigen, Switzerland*

[2] *DPMC, University of Geneva, 24 Quai E.-Ansermet 1211, Geneva 4, Switzerland*

*carlos.vaz@psi.ch

We report here the direct imaging of the magnetic response of a 4.8 nm $La_{0.9}Ba_{0.1}MnO_3$ film to the voltage applied across a 5 nm $BaTiO_3$ film in a $BaTiO_3/La_{0.9}Ba_{0.1}MnO_3$ multiferroic heterostructure using x-ray photoemission electron microscopy (XPEEM). The samples were grown by molecular beam epitaxy and are found to be atomically smooth, as determined by reflection high energy electron diffraction (RHEED) and atomic force microscopy. Specifically, we have written square ferroelectric patterns on the $BaTiO_3$ layer with an atomic force microscope in contact mode by applying a voltage to the tip ranging between -8 V and +8V and measured the change in magnetic contrast through the X-ray circular dichroic effect imaged at the Mn L-edge with high spatial lateral resolution using XPEEM. We find an increase in the magnetic contrast when going from negative to positive writing voltages, as shown in Fig. 1 for measurements taken at 80 K, consistent with the presence of a magnetoelectric effect through changes in the hole carrier density. Temperature dependent measurements show a decrease in the Curie temperature in the areas where positive voltage above +7 V was applied corresponding to hole depletion state, in agreement with our expectations. The magnetic contrast of the written areas are also compared with SQUID magnetometry results, showing the typical behaviour of accumulation and depletion state of manganite for the two polarization of the $BaTiO_3$. Our results are the first direct imaging of magnetoelectric coupling in such multiferroic heterostructure.

**Acknowledgements**

This project is funded by the Swiss National Science Foundation (SNF) (Grant No. 200021_184684).

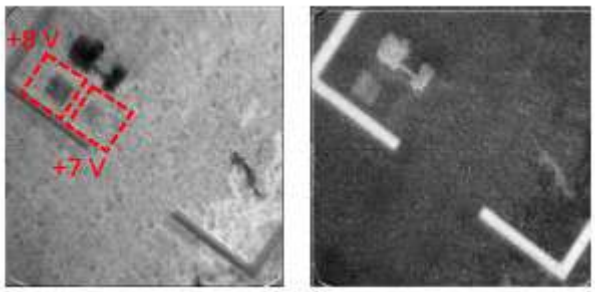



Figure1: XMCD images collected at 80 K at Mn $L_3$ (left) and $L_2$ edge (right), showing inversed magnetic contrast as expected because of the opposite sign of the XMCD for the two edges. The black squares to the top left image are damaged areas (applied voltage of -7, -8 V), while the grey ones marked by a red dotted square correspond to applied voltages of +8 and +7 V.

# Effect of nonmagnetic Hf addition on magnetic properties melt-spun Misch Metal-Fe-B ribbons

***M. Lostun****, *M. Grigoraș, G. Ababei , L.C. Răcilă, T. Roman, N. Lupu*
*National Institute of Research and Development for Technical Physics, 700050, Iasi, Romania*
*mlostun@phys-iasi.ro

The increasingly wide range of applications for rare earth permanent magnets (REPMs) has a direct influence on the growing global market of REPMs [1]. At present, strong research activity is thus focused on finding viable alternatives to rare earth (RE) elements -based permanent magnets, [2] which can allow their replacement at least in those applications where high performances are not strictly required.

To balance the utilization of RE resources and develop Misch Metal (MM) -Fe -B permanent magnets with a high performance/cost ratio, the role of the effect of Hf addition on the microstructure, magnetic properties, and thermal stability of $MM_{14}Fe_{80-x}Hf_xB_6$ (x=0–3.0) ribbons melt-spun and annealed was investigated. It has been confirmed that the hard magnetic properties of MM-Fe-B magnets can be effectively enhanced by the addition of Hf. Magnetic characterization using a vibrating sample magnetometer (VSM) showed that the addition of nonmagnetic Hf had a significant role in improving the magnetic properties at room temperature. The coercivity Hc of the optimally processed ribbons increased monotonically with increasing Hf content, from 3.85 kOe for x=0 to 5.7 kOe for x=3.0. Unlike the coercivity, the remanence Mr increased first with Hf addition, from 25 emu/g up to 37 emu/g at x=1.5, and then decreased with further Hf addition. The maximum energy product (BH)max behaved similarly, increasing from 6.5 MOe to a maximum of 7.45 MOe at x=2. Structural and morphological studies using X-ray powder diffraction (XRD) and scanning electron microscopy (SEM) had shown a significant microstructure refinement with Hf addition. The grain refinement and microstructure uniformity are essential for improving the magnetic properties of $MM_{14}Fe_{80-x}Hf_xB_6$ alloy. This paper may shed light on the further development of MM-based magnets and offer a feasible way for using rare earth resources efficiently.

**Acknowledgements**

This work was supported by a grant of the Ministry of Research, Innovation and Digitization CNCS–UEFISCDI, project number PN-III-P1-1.1-TE-2021-0976, within PNCDI III

**References:**

[1] Deng, S.; Kpodzro, E.; Maani, T.; Li, Z.; Huang, A.; Yih, Y.; Zhao, F.; Sutherland, J.W. Planning a circular economy system for electric vehicles using network simulation. J. Manuf. Syst. 2022, 63, 95–106.
[2] https://ec.europa.eu/growth/sectors/raw-materials/areas-specific-interest/critical-raw-materials_en

# Nanocrystalline Fe-Co based Soft Magnetic Powdered Alloys

*J. Daza[1]*, Z. Curbelo-Cano[2], C.M. Montero[2], J. Saurina1[1], L. Escoda[1], E.M. Palmero[2], A. Bollero[2], J.J. Suñol[1]*

[1] *Department of Physics, University of Girona, Campus Montilivi s/n, 17003 Girona, Spain*

[2] *Group of Permanent Magnets and Applications, IMDEA Nanoscience, C/Faraday 9, 28049 Madrid, Spain*

*joanjosep.sunyol@udg.edu

In this work, monocrystalline Fe-Co based ferromagnetic alloys have been obtained as powders using the mechanical alloying technique. Fe-Co based alloys can be used in actuators, transformer cores and sensors amongst other possible applications. These alloys are also candidates to be used in soft-hard composites to develop spring magnets. However, general applications usually require the formation of nanocrystalline structures within the alloy as this will improve their soft magnetic behavior [1].

The morphology of the Fe-Co based powders obtained by mechanical alloying was analyzed by scanning electron microscopy, and the composition was determined by using the energy dispersive X-ray spectroscopy detector. The studied samples were prepared using milling times from 10 to 50 h, both without and with the addition of cyclohexane as a process control agent. It was observed a reduction in the particle size by increasing the milling time, being the process more efficient when using cyclohexane (i.e., both the mean particle size and the particle size distribution were further reduced for the same milling time). For a milling time of 50 h the mean particle size was 1-2 µm (using cyclohexane) and in the range between 5-15 µm (with no cyclohexane). For all the samples a composition of $Fe_{65}Co_{35}$ was confirmed.

The magnetic analysis of the Fe-Co based alloys has been performed by vibrating sample magnetometry. Its magnetically soft behavior at room temperature has been verified in hysteresis cycle measurements. The saturation magnetization ($M_s$) values are in the 208-230 $A \cdot m^2 \cdot kg^{-1}$ interval and the coercivity ($H_c$) in the 50-$80 \cdot 10^{-4}$ T range.

Concerning the microstructure, the X-ray diffraction patterns analysis confirms the formation of a nanocrystalline Fe-rich *bcc* solid solution. The thermal analysis (differential scanning calorimetry) show broad exothermal processes beginning at about 200 ºC linked to the structural relaxation of the microstrain induced during milling. At high tamperatures (~575ºC) an exothermic peak appears, corresponding to the crystalline growth of the nanocrystalline phase. These results are comparable to those obtained in previous studies for Fe-Co-Nb based [2] and Finemet-like [3] alloys.

## Acknowledgements

Authors acknowledge financial support from MICINN through *NEXUS* project (Ref. PID2020-115215RB-C22 and PID2020-115215RB-C21). E.M.P. acknowledges support from AEI through the Juan de la Cierva – Incorporación program (IJC2020-043011-I/MCIN/AEI/10.13039/501100011033) and EU by NextGenerationEU/PRTR. IMDEA Nanoscience thanks support from the ‘‘Severo Ochoa’’ Programme for Centers of Excellence in R&D (MICINN, Grant CEX2020-001039-S) and from the Regional Government of Madrid through *NANOMAGCOST* project (Ref. S2018/NMT-4321).

## References

[1] M. Raja, K. Chattopadhyay, B. Majumdar, A. Narayanasamy, *J. Alloys. Compd.* **297** (2000) 199.

[2] A. Carrillo, J. saurina, L. Escoda, J.J. Suñol, *Metals* **11(3)** (2021) 379.

[3] J. Daza, W. Ben Mbarek, L. Escoda, J.J. Suñol, *Metals* **11(12)** (2021) 1896.

# Mn-Al Alloys Produced by Mechanical Alloying and Melt-spinning

*J. Daza[1]*, A.J. Campos-Hernández[2], J. Saurina1[1], L. Escoda[1], E.M. Palmero[2], A. Bollero[2], J.J. Suñol[1]*
[1]*Department of Physics, University of Girona, Campus Montilivi s/n, 17003 Girona, Spain*
[2]*Group of Permanent Magnets and Applications, IMDEA Nanoscience, C/Faraday 9, 28049, Madrid, Spain*
*joanjosep.sunyol@udg.edu

Mn-Al alloys with Mn content between 50 and 60 at.% and the minor addition of a third element (e.g., C, Ni or Tb) are promising candidates for technological applications due to their interesting combination of magnetic properties, mechanical strength and resistance to corrosion [1,2]. The only ferromagnetic phase of the Mn-Al system is the τ-phase, which together with an optimized content of other phases (i.e., β-phase and $Mn_3AlC$ carbides), tuned grain size and induced strain allow for obtaining tailored magnetic properties [2-4].

In this work, Mn-Al alloys have been produced by mechanical alloying (as powders) and by melt-spinning (as ribbons). Mechanical alloying favors the formation of a nanostructured crystalline phase with a high density of crystallographic defects as dislocations. Meanwhile, melt-spinning allows for the production of amorphous alloys. In order to obtain Mn-Al-based alloys with an optimized crystallographic structure, it is required a subsequent thermal treatment, being one option the heating up to 1000 – 1100 ºC, followed by quenching and, finally an annealing at 500 – 550 ºC. The parameters of the thermal treatments carried out in the Mn-Al system play a key role for obtaining the desired crystallographic phases in the resulting material. The cooling rate is crucial as quenching with a fast cooling rate originates the formation of the ε-phase, intermediate quenching rate the formation of the τ-MnAl phase, and a slow cooling rate promotes the decomposition of the metastable τ-phase into the more stable phases such as the β- and $\gamma_2$-phases, originating multiphase samples.

Calorimetric mesaurements (by differential scanning calorimetry) confirms that Mn-Al alloys with Co, V or C addition show on heating the formation of the desired τ-phase, at temperatures close to 500 ºC. It has been also found that an exothermic process appears at lower temperatures, indicating the previous formation of the intermediate and metastable ε'-phase. The optimization of the processing conditions (for both mechanical alloying and melt-spinning) and the compositions is under development. The approach consisting in using the proposed scalable fabrication tecniques opens a new path for the preparation of novel Mn-Al-based alloys with tailored magnetic properties.

## Acknowledgements

Authors acknowledge financial support from MICINN through *NEXUS* project (Ref. PID2020-115215RB-C22 and PID2020-115215RB-C21). A.J.C.-H. and E.M.P. acknowledge support from "La Caixa" Foundation (ID 100010434) through Doctoral INPhINIT Incoming program (LCF/BQ/DI20/1178002) and AEI (JdC-I, IJC2020-043011-I/MCIN/AEI/10.13039/501100011033) and EU NextGenerationEU/PRTR, respectively. IMDEA Nanoscience thanks support from the ''Severo Ochoa'' Programme for Centers of Excellence in R&D (MICINN, Grant CEX2020-001039-S) and from the Regional Government of Madrid through *NANOMAGCOST* project (Ref. S2018/NMT-4321).

## References

[1] F. Jiménez-Villacorta, J.L. Marion, J.T. Oldham, M. Daniil, M.A. Willard, L.H. Lewis, *Metals* **4** (2014) 8-19.
[2] J. Rial, P. Švec, E.M. Palmero, J. Camarero, P. Švec Sr, A. Bollero, *Acta Materialia* **157** (2018) 42-52.
[3] J. Rial, E.M. Palmero, A. Bollero, *Engineering* **6** (2020) 173-177.
[4] C. Muñoz-Rodríguez, L. Feng, E.M. Palmero, T. Mix, J. Rial, F. Olsson, B. Skårman, H. Vidarsson, P-O. Larsson, T.G. Woodcock, A. Bollero, *J. Alloys. Compd.* **847** (2020) 156361.

# Control of Magnetic Compensation Temperature in Co/Tb Multilayers Via Buffer Layer Modification

*Jakub Zázvorka*[1]*, *Carlos H. Verbeno*[1], *Lukáš Nowak*[1], *Martin Veis*[1]

[1] *Institute of Physics, Faculty of Mathematics and Physics, Charles University, 121 16, Prague, Czech Republic*

*zazvorka@karlov.mff.cuni.cz

Materials with antiferromagnetic exchange have been extensively studied due to their potential applications in spintronics and related industry. As utilization of compensated antiferromagnets poses a challenge due to their low interaction with external magnetic fields, ferrimagnetic materials utilize the advantages of both ferromagnets and antiferromagnets and stand a bridging point for both material classes. It has been shown that rare earth-transition metal ferrimagnets in forms of thin films and multilayers can be of importance in the field of magneto-optical recording, as all-optical switching was observed in such systems (e.g. GdFeCo and CoTb) [1, 2]. Studying and optimizing the material parameters of ferrimagnetic multilayer systems is therefore an interesting topic with application potential. One of the advantages of materials with antiferromagnetic coupling is the higher frequency of magnetic resonance, which can lead to faster magnetic response in devices [3]. Ferrimagnets consisting of several elements in the form of compound or individually coupled layers exhibit the so-called magnetic compensation temperature. As the magnetic moments in the individual sublattices have different thermo-magnetic dependence, the net magnetisation of the system is strongly temperature dependent and exhibits a point of zero net magnetic moment. This temperature is called the magnetization compensation temperature $T_{MC}$. The effect of magnetic imprinting was shown in the vicinity of $T_{MC}$ and therefore studying the physical effects (e.g. domain wall velocity) at that temperature is of interest [4]. It has been shown that the compensation temperature is dependent on the material composition and the level of intermixing of elements in a compound material [5]. Our previous investigation (unpublished data at the date of submission) has revealed that the magnetic parameters in a multilayer system of Pt/Co can be affected by introducing a buffer layer which changed the interface roughness. The coercivity of the multilayer was modified proportionally to the roughness without changing the saturation magnetization. Therefore, we concluded that the buffer layer can be used to prepare a material with desired coercivity.

Here, we present results on introducing the same buffer layer to a Co/Tb multilayer system. While the different thicknesses of the Au buffer layer result in a different interface roughness, the magnetization compensation temperature of Au(y = 0-20 nm)/Pt(5)/[Co(0.7)/Tb(x = 0.6-1.0 nm)]$_{\times 6}$/Au(5) model system is shifted. Higher interface roughness results in a higher compensation temperature with good correlation. Therefore, the conclude that interface engineering can be used to prepare ferrimagnetic multilayer materials with desired magnetic parameters for further development of devices.

## References

[1] C. D. Stanciu, F. Hansteen, A. V. Kimel, et al., *Phys. Rev. Lett.* **99**, (2007).
[2] M. S. El Hadri, P. Pirro, C.-H. Lambert, et al., *Phys. Rev. B* **94**, (2016).
[3] H. Bai, X. Zhou, Y. Zhou, et al., *J. Appl. Phys.* **128**, (2020) 210901.
[4] L. Caretta, M. Mann, F. Buttner, et al., *Nat Nanotechnol* **13**, (2018) 1154-1160.
[5] L. Avilés-Félix, L. Álvaro-Gómez, G. Li, et al., *AIP Advances* **9**, (2019).

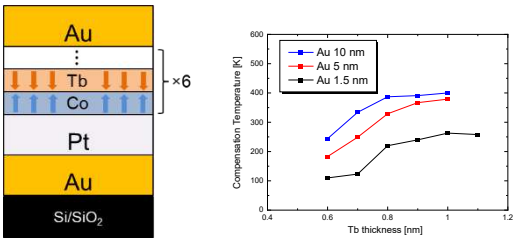



Figure 1. Left: Schematic of the investigated multilayer system. Right: Magnetization compensation temperature $T_{MC}$ in dependence on Tb layer thickness and the thickness of the Au buffer layer.

# Thickness Dependent Magneto-optical Properties of $La_{2/3}Sr_{1/3}MnO_3$ Thin Films on Silicon with a Nanosheet Seed Layer

*Tomáš Maleček[1*], Aleš Melzer[1], Guillaume Agnus[2], Thomas Maroutian[2], Lukáš Horák[1], Petr Machovec[1], Valérie Demange[3], Jan Prokleška[1], Philippe Lecoeur[2], Martin Veis[1]*

[1] *Charles University, Faculty of Mathematics and Physics, Ke Karlovu 2027/3, 121 16, Prague, Czech Republic*

[2] *Centre for Nanoscience and Nanotechnology (C2N), CNRS UMR 9001, Univ Paris-Sud, Université Paris-Saclay, 91120 Palaiseau, France*

[3] *Univ Rennes, CNRS, ISCR – UMR 6226, F-35000 Rennes, France*

*tomas.malecek@matfyz.cuni.cz

Ultrathin films of the hole-doped perovskite manganite $La_{2/3}Sr_{1/3}MnO_3$ (LSMO) attract interest due to their unique combination of physical properties. LSMO is ferromagnetic at room temperature ($T_C$ = 370 K [1]), it exhibits colossal magnetoresistance [2], and is almost fully spin polarized [3], which makes it an auspicious candidate for novel applications in the field of spintronics.

In this poster presentation we demonstrate the possibility of growing textured LSMO thin films on Si substrates using pulsed laser deposition. Their magneto-optical and optical properties are comparable to high quality epitaxial layers on bulk $SrTiO_3$ (STO). Successful growth of LSMO on (100) Si substrate is achieved via two-dimensional nanosheet (NS) seed layer of $Ca_2Nb_3O_{10}$ (CNO), which induces epitaxial stabilization of the layers. Investigated samples on NS exhibit larger Curie temperature and lower magnetic moment than the samples on STO with same thickness. This indicates almost full strain relaxation, since the strain induces suppression of $T_C$, and was confirmed by XRD measurements.

Optical and magneto-optical properties of the samples are presented along with the spectral dependencies of the full permittivity tensor deduced from the measurements. Optical response of the LSMO/NS layers is comparable to epitaxial layers, indicating similar electronic structure and therefore confirms fully developed perovskite structure. The thickness dependent spectra of magneto-optical polar Kerr rotation at room temperature in the field of 1T can be seen in the figure 1. Since magneto-optical phenomena are very sensitive to magnetic moment, electronic structure and interfaces, the thickness-dependent measurements allowed us to investigate subtle changes in the permittivity as a result of the onset and evolution of magnetic order in ultrathin layers of this material.

## Acknowledgements

The study was supported by the Charles University, project GA UK No. 363321.

## References:

[1] G.H. Jonker, J.H. Van Santen, Physica, 16 (1950) 337-349.
[2] A.P. Ramirez, Journal of Physics: Condensed Matter, 9 (1997) 8171-8199.
[3] M. Bowen, et al., Applied Physics Letters, 82 (2003) 233-235.

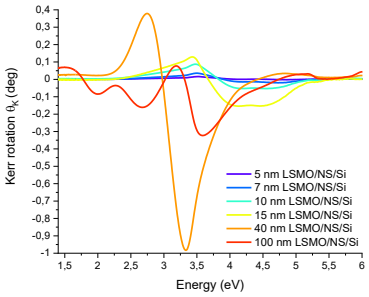



Figure 1: Polar magneto-optical Kerr rotation spectra of textured LSMO thin films with varying thicknesses on silicon with a CNO seed layer.

# Discerning the Magnetization Reversal Mechanism and Magnetic Interactions in Arrays of Hexagonally Ordered FeNi Nanowires

*Alonso J. Campos-Hernandez*, Ester M. Palmero, Carlos Iglesias Fernández-Cuevas, and Alberto Bollero*

*Group of Permanent Magnets and Applications, IMDEA Nanoscience, 28049 Madrid, Spain*

*alonsojose.campos@imdea.org

Electrodeposited nanowires (NWs) are quickly emerging as a useful platform for the development of multiple applications, going from next-generation information technologies or sensors [1] to nanocomposite magnetic materials profiting from the complementary properties of the coexisting phases. In all of these use cases, a proper understanding of the magnetization reversal processes and the magnetic interactions between the NWs is needed (e.g., to optimize memory writing in NWs-based magnetic memories or to enhance the magnetic performance of multifunctional materials under operation conditions).

In this work, FeNi NWs with a well-controlled mean diameter of 40 nm and a different Fe:Ni ratio were synthesized by electrodeposition into nanoporous anodized aluminum oxide membranes (see inset in Fig. 1b). The goal of this study was to assess the magnetization reversal mode and the magnetic interactions between adjacent NWs in an array configuration, in order to gain an in-deep understanding of the magnetic phenomena controlling the behavior of these systems. For this purpose, a combined experimental and modelling approach (Fig. 1a) has been developed based in previous results [2] . It was found that all the arrays of FeNi NWs under study follow a magnetization reversal process dominated by transverse domain wall reversal. Additionally, evidence was found for the enhancement of magnetostatic interactions between the NWs as the Fe content increased. To advance further in the understanding of these magnetic interactions, First-Order Reversal Curve (FORC) analysis was carried out. Enlarged magnetic interactions between the NWs with increasing the Fe content was confirmed by FORC, thus allowing to establish analogies with well-known magnetic systems. In particular, the FORC diagram obtained for the $Fe_{0.24}Ni_{0.76}$ NWs (Fig. 1b) shows limited interactions and similar characteristics to that of comercial magnetic memories [3], while the diagram for the $Fe_{0.80}Ni_{0.20}$ NWs (Fig. 1c) shows the same configuration as those corresponding to highly-interacting magnetic systems [4].

## Acknowledgements

Authors acknowledge financial support from Robert Bosch GmbH through an IMDEA Nanoscience-Bosch Research cooperation project. A.J.C.-H. and E.M.P. acknowledge support from "La Caixa" Foundation (ID 100010434) through Doctoral INPhINIT Incoming program (LCF/BQ/DI20/1178002) and AEI (JdC-I, IJC2020-043011-I/MCIN/AEI/10.13039/501100011033) and EU NextGenerationEU/PRTR, respectively.

## References

[1] P. Fischer, D. Sanz-Hernández, R. Streubel, A. Fernández-Pacheco, *APL Mater.*, **8** (2020) 010701.
[2] L.G. Vivas, M. Vazquez, J. Escrig, S. Allende, D. Altbir, D. C. Leitao, J.P. Araujo, *Phys. Rev. B*, **85** (2012) 035439.
[3] C.R. Pike, C.A. Ross, R.T. Scalettar, G. Zimanyi, *Phys. Rev. B*, **71** (2005) 134407.
[4] E.M. Palmero, F. Béron, C. Bran, R.P. del Real, and M. Vázquez, *Nanotechnology*, **27** (2016) 435705.

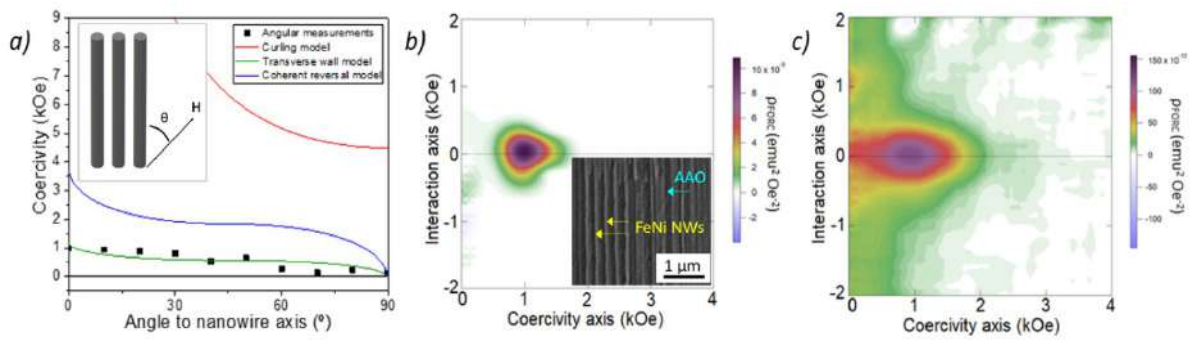



Figure 1: a) Angular coercivity measurements and model fits for the array of $Fe_{0.24}Ni_{0.76}$ NWs; FORC diagrams for b) the array of $Fe_{0.24}Ni_{0.76}$ NWs and c) the the array of $Fe_{0.80}Ni_{0.20}$ NWs. Inset in a) shows a scheme of the applied magnetic field (H) with an angle (θ) to the NWs longitudinal axis. Inset in b) shows a cross-section SEM image of the FeNi NWs embedded in the alumina membrane.

# Effect of thermal treatments on the microstructure of Ni-Mn-Ga-(Co, Fe) ribbons for mechanical actuation

M. Namvari [1], J. J. Rosero-Romo[2], A. García[2], B. Rodriguez[2], P. G. Saiz[2], K. Ullakko[1], D. Salazar[2]

[1] *Material Physics Laboratory, Lappeenranta-Lahti University of Technology LUT, 53850 Lappeenranta, Finland*

[2] *BCMaterials, Basque Center for Materials,Applications & Nanostructures, 48940 Leioa, Spain*

**Abstract:** Ferromagnetic shape memory alloys (FSMAs) are mostly off-stoichiometric Heusler type metals that have attracted attention due to their potential in applications as giant magnetic field induced strain effect (MFIS), which is linked to the reorientation of the martensitic microstructure. Rapid solidification of these alloys via melt-spinning allows to obtain homogeneous, chemically ordered and single phase polycrystalline ribbons of FSMAs [1]. In this work, $Ni_{49.8}Mn_{28.5-x}Ga_{21.7}(Fe, Co)_x$ melt-spun ribbons were prepared by melt-spinning technique. The effect of the substitution of Mn by additions of Fe (1 at.%) and Co (1.5 and 2 at.%) on the crystal structure, microstructure and magnetic properties of heat treated ribbons was studied. Melt-spun ribbons were heat-treated at 1313 K and cooled at different rates. The crystalline phases and the microstructure of the rapidly and slow-cooled ribbons were analyzed by X-ray diffraction patterns and SEM-EDX, respectively. Thermomagnetic measurements at different applied magnetic fields were recorded using a vibrating sample magnetometer. These results evidenced an ideal microstructure for MFIS applications.

***Keywords***:*NiMnGa Heusler alloys, Ferromagnetic shape memory alloy, melt spun ribbons, magnetic field induced strain*

[1] C.O. Aguilar-Ortiz, D. Soto-Parra, P. Álvarez-Alonso, P. Lázpita, D. Salazar, P.O. Castillo-Villa, H. Flores-Zúñiga, V.A. Chernenko, Influence of Fe doping and magnetic field on martensitic transition in Ni–Mn–Sn melt-spun ribbons, Acta Materialia **107** (2016) 9-16.

# Novel Magnetic Properties of RF-Sputtered $Mn_4N$ Thin Films

*Lucy Prendeville[1]*, Gwenaël Atcheson[1], Karsten Rode[1], Yangkun He[2], Simon Lenne[1] and J. M. D. Coey[1]*

[1] *School of Physics and CRANN, Trinity College, Dublin, Ireland*

[2] *Beihang University, Beijing, China*

*prendevl@tcd.ie

$Mn_4N$ has an fcc Mn structure with N in the body center interstitial position. It is a ferrimagnet with a non-collinear spin structure. The two Mn sublattices (1*a* corner sites and 3*c* face center sites) couple anti-ferromagnetically but unequal and opposite net sublattice magnetisations yield a resultant along the [111] direction in bulk form. The 3*c* moments have components at 120° to each other in the (111) plane [1]. When grown as a thin film on a (100) MgO substrate, $Mn_4N$ adopts a (001) orientation with a small tetragonal distortion (c/a=0.99) and exhibits perpendicular magnetic anisotropy [2]. Y. He *et al.* [3] proposed a possible magnetic structure based on the known spin arrangements of bulk $Mn_3ZN$ compounds, considering appropriate exchange interactions between different Mn sites. The material has potential for ferrimagnetic spintronics.

The properties of $Mn_4N$ films depend sensitively on the deposition parameters, including substrate temperature, sputtering gas and chamber pressure. Our $Mn_4N$ films were deposited on (100) MgO substrates by RF reactive magnetron sputtering at 440°C in a 1:20 $N_2$:Ar atmosphere. The structure of the films is particularly sensitive to the growth pressure. Figure 1 (a) shows a section of the XRD patterns of films grown at 2 mTorr and 10 mTorr with the (002) $Mn_4N$ peaks indicated. The out-of-plane *c*-parameters are 3.852 and 3.863 Å respectively. In the 2 mTorr films we observed an impurity phase with 2θ close to 40.4° which is absent in the 10 mTorr films and associated with the (002) peak of MnO. The top right insert shows how this peak disappears as chamber pressure increases. The insert in the bottom right shows how the variation in *c* of the $Mn_4N$ films is not linear with pressure. Crystallite sizes in the 2 mTorr and 10 mTorr films were determined as 23.7 and 14.4 nm from the Scherrer formula.

The purer 10 mTorr films show smaller saturation magnetisation and higher coercivity compared to the 2 mTorr films. Figure 1 (b) shows that they have lower remanence and the hysteresis loop measured by SQUID is wasp-waisted but that measured by anomalous Hall effect (AHE), intriguingly, is not, Fig. 1 (c). Furthermore the loop becomes increasingly wasp-waisted when the substrate is tilted with respect to the applied field. An explanation of these interesting results in terms of the domain state magnetisation will be presented.

## Acknowledgements

This work was supported by the FFP grant FISTMAP (21/FFP-P/10175). Lucy Prendeville is funded by the IRC under grant GOIPG/2019/4430.

## References

[1] D. Fruchart, D. Givord, P. Convert, P. L'Héritier, and J. P. Sénateur, *J. Phys. F: Metal Phys*. **9** (1979) 12.

[2] Y. Yasutomi, K. Ito, T. Sanai, K. Toko, and T. Suemasu, *J. Appl. Phys*., **115** (2014) 10.

[3] Y. He, S. Lenne, Z. Gercsi, G. Atcheson, J. O. Brien, D. Fruchart, K. Rode, and J. M. D. Coey, *Phys. Rev. B* **106** (2022) L060409.

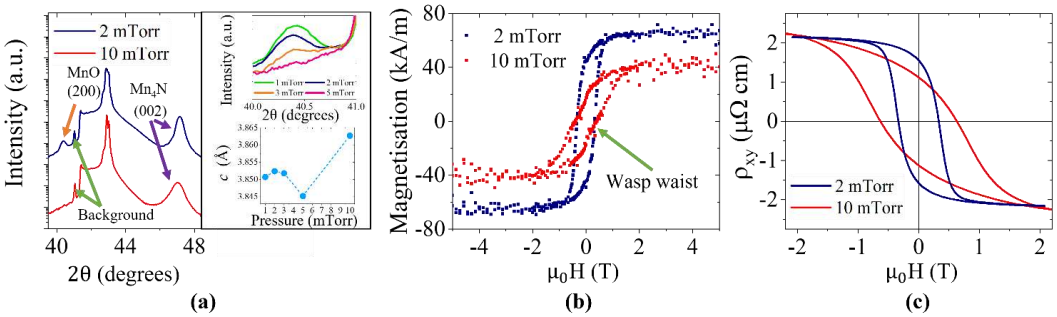



Figure 1: (a) XRD patterns, (b) SQUID loops and (c) AHE loops for $Mn_4N$ films grown at different chamber pressures. The wasp waist of the red SQUID loop is indicated in (b), which is absent in the AHE data in (c).

# SYMPOSIUM 11.
## SPIN-TRANSFER BASED PHENOMENA AND DEVICES.
## S11. INVITED ORAL PRESENTATIONS

# Advances of MRAM Devices for Memory and Computing Application

*S. Couet*, S. Rao, R. Carpenter, K. Cai, W. Kim, F. Yassin, G. Talmelli, S. Van beek, N. Franchina-Vergel, N. Jossart, A. Trovato, D. Crotti, K. Wostyn*

Imec, Kapeldreef 75, B3001 Leuven, Belgium

*sebastien.couet@imec.be

Magnetic tunnel junction (MTJ) based memories have seen a tremendous effort the last 10 years to bring them into production. Since 2019, spin transfer torque (STT) magnetic random access memory (MRAM) is in volume production at major foundries [1]. While this is in many aspects an important milestone for magnetic memories, this first applications is somewhat limited to embedded flash replacement at Nxx node. The R&D focus is currently shifting towards enabling MRAM as a (partial) last level cache (LLC) replacement specification. This is expected to make full use of MRAM's advantage: high endurance, CMOS voltage compatibility at advanced nodes and high read/write speeed.

Key to achieving LLC functionality are a high read/write speed well below 5ns, an 'unlimited' endurance while keeping switching current low and a high bit-density. Here as well STT devices are a potential candidate as being the most mature, but more advanced concepts such as spin orbit torque (SOT) MRAM or even voltage controlled (VC) MRAM display significant advantage in terms of write speed and/or power compared to STT. Over the last 5 years, we have made several advances in demonstrating SOT-MRAM functionality on 300mm integrated platform. Despite the progress, there remains several challenges along the way.

In this presentation, we will review the state-of-the-art of MRAM technologies and position them from an application standpoint. We will also review the key challenges that remains ahead, drawing a particular emphasis at where fundamental progress and understanding remains to be achieved.

**Acknowledgements**

This work was supported by IMEC's industrial affiliation program on MRAM device.We also acknowledge the support from the ECSEL Joint Undertaking Program (grantNo.876925- project ANDANTE).

**References**

[1] Lee, Y.K.; Song, Y.; Kim, J.; Oh, S.; Bae, B.-J.; Lee, S.; Lee, J.; Pi, U.; Seo, B.; Jung, H.; et al., Embedded STT-MRAM in 28-Nm FDSOI Logic Process for Industrial MCU/IoT Application. In Proceedings of the 2018 IEEE Symposium on VLSI Technology, Honolulu, HI, USA, 18–22 June 2018; pp. 181–182

# Stochastic magnetic tunnel junction for probabilistic computing

*Shunsuke Fukami*[1,2,3,4,5]*

[1] *Research Institute of Electrical Communication, Tohoku Univ., Sendai, Japan*
[2] *Center for Science and Innovation in Spintronics, Tohoku Univ., Sendai, Japan*
[3] *Center for Innovative Integrated Electronic Systems, Tohoku Univ., Sendai, Japan*
[4] *Center for Innovative Integrated Electronic Systems, Tohoku Univ., Sendai, Japan*
[5] *Inamori Research Institute for Science, Kyoto, Japan*
*s-fukami@tohoku.ac.jp

Conventional electronics relies on deterministic operation of computing hardware and stochastic behavior is attempted to be reduced as low as possible. Contrary to this perception, in 1981, R. P. Feynmann gave a suggestion of unconventional computing paradigm in which probabilistic behavior of physical system is effectively used in computing hardware, so-called the probabilistic computing [1]. Probabilistic bit (p-bit) is a fundamental unit of the probabilistic computer and stochastic magnetic tunnel junction is proposed to be used as a promising ingredient for the p-bit [2].

In this talk, I will show some proof-of-concepts of the spintronic probabilistic computers and describe how the computer can be constructed from the stochastic magnetic tunnel junction and how it solves computationally hard problems [3-5]. I will also discuss the physics governing the probabilistic behavior of the stochastic magnetic tunnel junction and strategy to develop the devices for high-performance probabilistic computers [6-10].

**Acknowledgements**

This study is carried out in collaboration with H. Ohno, S. Kanai, W. A. Borders, K. Hayakawa, K. Kobayashi, K. Y. Camsari, A. Z. Pervaiz, J. Kaiser, S. Datta, and G. Finocchio and was partly supported by JST-CREST JPMJCR19K3 MEXT X-NICS JPJ011438 and RIEC Cooperative Research Projects.

**References**

[1] R. P. Feynman, Int. J. Theor. Phys. **21**, 467 (1982).
[2] K. Camsari et al., Phys. Rev. X **7**, 031014 (2017).
[3] W. A. Borders et al., Nature **573**, 390 (2019).
[4] J. Kaiser et al., Phys. Rev. Appl. **17**, 014016 (2022).
[5] A. Grimardi et al., IEEE IEDM 2022, 22.4 (2022).
[6] S. Kanai et al., Phys. Rev. B **103**, 094423 (2021).
[7] K. Hayakawa et al., Phys. Rev. Lett. **126**, 117202 (2021).
[8] K. Kobayashi et al., Appl. Phys. Lett. **119**, 132406 (2021).
[9] T. Funatsu et al., Nat. Comm. **13**, 4079 (2022).
[10] K. Kobayashi et al., Phys. Rev. Appl. **18**, 054085 (2022).

# Spin-orbit torque switching in ferrimagnets and ferromagnets with picosecond electrical pulses

*E. Díaz[1], A. Anadón[1], M. Morassi[2], A. Lemaître[2], M. Hehn[1], J. Gorchon[1,*]*

[1] *Université de Lorraine, CNRS, Institut Jean Lamour (IJL), Nancy, France*

*2 Centre de Nanosciences et de Nanotechnologies (C2N), CNRS, Université Paris Sud, Université Paris-Saclay, Palaiseau, France*

*jon.gorchon@univ-lorraine.fr

Spintronic phenomena could potentially lead to devices with lower energy consumption than current electronics, but in order to make this technology more competitive, a faster reversal of the magnetic cell should be achieved. Current spintronics devices are based upon the utilization of spin-transfer torque [1,2], but the switching speed of this mechanism is limited by an incubation time and the maximum current density applicable before the breakdown of the device. On the other hand, spin-orbit torque (SOT) offers a substantial improvement in terms of speed and also a low energy consumption [3]. SOT has been studied thoroughly down to the ns scale and somewhat at the sub-ns scale [3], but the ps scale remains to this day largely unexplored [4,5], and the mechanisms at play during reversal are still unclear. Here we show the SOT switching of various ferromagnetic and ferrimagnetic materials induced by picosecond pulses (see MOKE images of GdCo switching due to single ps pulses in Figure 1), along with the first critical current and energy characterization done at this time scale.

## References

[1] D J. C. Slonczewski, Journal of Magnetism and Magnetic Materials 159, L1–L7 (1996).
[2] L. Berger, Phys. Rev. B 54, 9353–9358 (1996).
[3] Grimaldi et al., Nature Nanotechnology 15, 111-117 (2020)
[4] K. Jhuria et al., Nature Electronics 3, 680–686 (2020)
[5] Debanjan et al., under submission

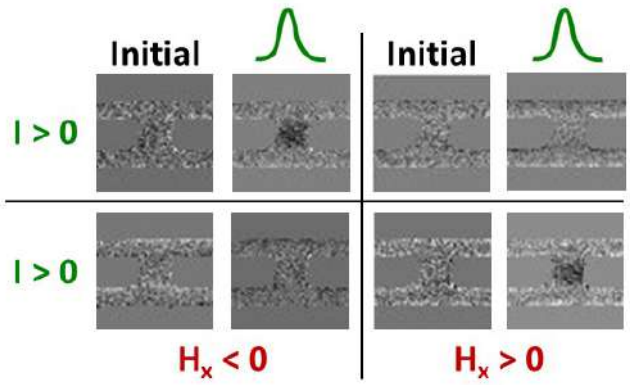



Figure 1: Switching of a CoGd ferrimagnet by single 5-8 ps-wide SOT pulses for different configurations of current (I) and external in-plane magnetic field (Hx).

# Field-free all optical switching and electrical read-out of Tb/Co based magnetic tunnel junctions

*D. Salomoni[1], Y. Peng[2], L. Farcis[1], M. Hehn[2], G. Malinowski[2], S. Mangin[2], B. Dieny[1], L. D. Buda-Prejbeanu[1], R.C. Sousa[1] and I. L. Prejbeanu[1],**

[1] *Univ. Grenoble Alpes, CEA, CNRS, Grenoble INP, SPINTEC, 38000 Grenoble, France*

[2] *Institut Jean Lamour, UMR CNRS 7198, Université de Lorraine, Nancy 54011, France*

*lucian.prejbeanu@cea.fr

All optical switching (AOS) of a magnetic tunnel junction provides an energy efficient way to enable writing at speeds 3 orders of magnitude faster than electrical alternative methods based on spin torque methods, providing a possible path for THz frequency memory operation. Using [Tb/Co] multilayers is particularly interesting, because their large perpendicular magnetic anisotropy would allow data retention at sub-20nm sizes, while integration into magnetic tunnel junction has already been proven. This work reports successful magnetization reversal driven by femtosecond light pulses on a [Tb/Co] based storage layer, on a perpendicular magnetic tunnel with a bottom reference electrode junction after patterning to sub-100nm lateral dimensions. It is possible to observe toggle switching between low and high resistance states, corresponding to parallel and anti-parallel alignment of the two electrodes, after each 50fs long laser pulse applied on the device. Nano-fabricated MTJ devices have diameters down to 80nm with TMR values up to 74%. A systematic study of the switching probability is compared to AOS experiments on continuous films. It was possible to demonstrate that full amplitude and reliable toggle switching can be achieved for specific Tb and Co multilayer compositions, with a Tb thickness ranging from 0.6 to 0.9nm, and 1.2 to 1.5nm for Co. No external field is required for the reversal, however an in-plane field of ~20Oe or an out-of-plane field of ~50Oe is sufficient to shift switching threshold fluences. Surprisingly, at film level for laser fluences higher than 11mJ/cm², concentric rings start appearing with opposite magnetic directions, indicating that a precessional reversal mechanism is taking place. Furthermore the fluences required to reverse and stabilize a given number of rings does not depend on the duration of the laser pulses investigated. Both these aspects are of great interest for the application as a large fluence operation window with high resilience to pulse length variability are expected. These results pave the way towards the development of a nanoscale ultra-fast and energy-efficient memory that exploits all-optically-switching.

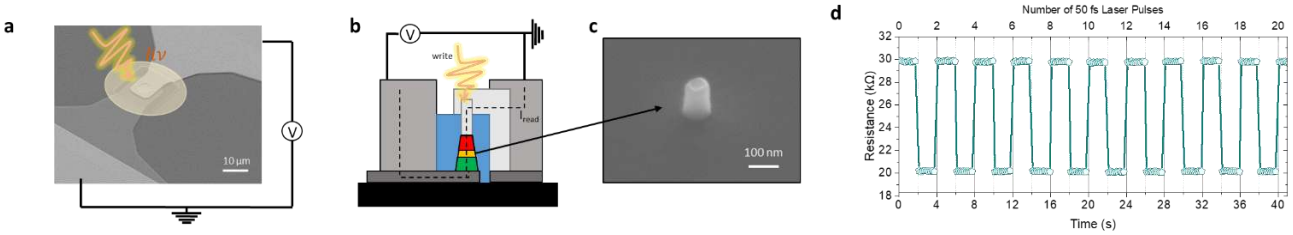



**Figure:** a) Top SEM view of the magnetic tunnel junction b) schematic illustration of All-optical "writing" and electrical reading of AOS-MTJ c) SEM image of an individual magnetic tunnel junction cell d) AOS field-free toggle switching between P and AP states.

**Acknowledgements**

This work has received funding from the European Union's Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement No 861300 and fom the ANR UFO

# SYMPOSIUM 11.
## SPIN-TRANSFER BASED PHENOMENA AND DEVICES.
## S11. ORAL PRESENTATIONS

# Electrodeposition viability for ultra-scaled MTJ

*Nuno Caçoilo[1]*, Laurent Cagnon[2], Ricardo Sousa[1], Bernard Dieny[1], Florian Disdier[1], Olivier Fruchart[1] and Ioan-Lucian Prejbeanu[1]*

[1] *Univ. Grenoble Alpes, CEA, CNRS, Grenoble INP, SPINTEC, 38000 Grenoble, France*
[2] *Univ. Grenoble Alpes, CNRS, Grenoble INP, Institut Neel, 38000 Grenoble, France*
*nuno.cacoilo@cea.fr

The spin-transfer-torque magnetic random-access memory is one of the most promising non-volatile memory technologies. In addition to non-volatility, it offers a quasi-infinite write endurance, high speed and moderate write power consumption. While holding a promise for high density, challenges in its fabrication towards very small nodes are acute, related to the need to define a hard mask with a large vertical aspect ratio [1, 2]. Starting from the definition of the hard-mask (HM), with a pillar etched using Reactive Ion Etching (RIE), to the Ion Beam Etching (IBE) to reach very small dimensions, the shape of the pillar has an impact on the magnetic and electrical properties of the MTJ. To reduce the diameter to values equal or below the ones from the EBL it is necessary to use IBE at a grazing angle to reduce the pillar diameter. As the stack is etched faster than the HM, eventually the base diameter can be smaller than the HM diameter, leading to faller or tilted pillars, limiting the device yield. In this work we present an alternative route to fabricate very small node magnetic tunnel junctions (MTJ), using a Pt hard mask electroplated inside nano-vias created with EBL. For this study, we use a typical MTJ stack SiO2 | Ta (22) | FeCoB(0.8) | Pt(8) | SyAF | W(0.2) | FeCoB(1) | MgO(1.25) | FeCoB(1.5) | W(2) | Ta(2) | Pt(5) (nominal thicknesses in nm), where the SyAF is composed of Co|Pt multilayers spaced by a thin 0.9 nm Ru layer. The RxA of the device is estimated using CIPT (current in-plane tunneling) to be around 8 $\Omega \cdot \mu m^2$. On top of the stack, 2% PMMA is coated with a thickness of around 220 nm, in which nano-vias with different nominal sizes are defined with EBL. Then, the Pt is electroplated within the vias above the Pt-capped MTJ stack which is used as electrode. Filling the vias with 100nm thick Pt pillars takes about 7 s (inset of Figure 1). After PMMA stripping, no significant increase in the vias diameter is observed (Figure 2). IBE then allows to achieve sub-20 nm diameter with large TMR values (Figure 2). We achieve 100% STT switching without applied magnetic field in these devices, confirming the viability of the electrodeposited Pt as a HM wih no aggressive trimming for the lateral downsizing of MTJ technology.

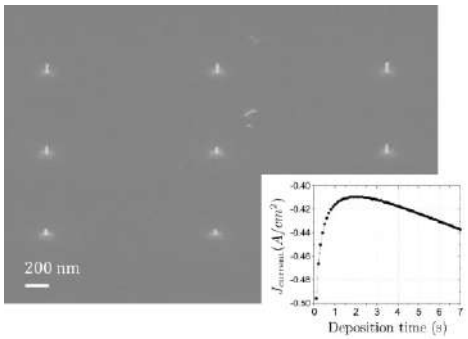



**Figure 1:** SEM measurements after etching and light trimming of the MTJ. The inset shows the current density as a function of the deposition time.

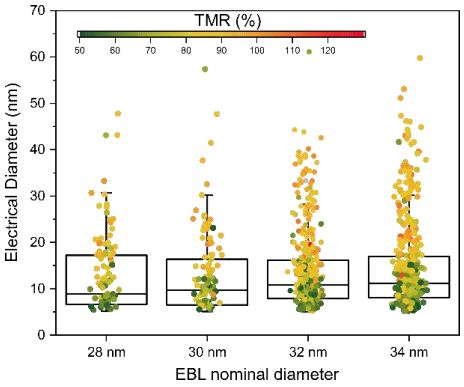



**Figure 2:** Dispersion of the electric diameter for each initial nominal diameter defined by EBL in the nano-via. Colormap shows the TMR of the fabricated device (from 50% to 130%).

**Acknowledgements**

This work was partly supported by the French RENATECH network.

**References**

[1] L. Wan, IEEE Transactions on Magnetics 58 (5), 1-6
[2] Y. Kim et al, VLSI Symposium, pp. 210-211 (2011)

# Spin Current Generation Using Amorphous Materials

*J. Karel*[*1,2], *C.-H. Hsu*[3], *N. Roschewsky*[4], *D.S. Bouma*[4], *S. Sayed*[3], *C. Fuchs*[4,5], *S. Cheem*[6], *S. Bennett*[1], *P. Corbae*[6], *S.B. Song*[7], *B.H. Zhang*[7], *R.Q. Wu*[8] *F. Hellman*[4,9], *S. Salahuddin*[3,9]

[1] *Department of Materials Science and Engineering, Monash University, Clayton, Victoria, Australia*
[2] *ARC Centre of Excellence in Future Low-Energy Electronics Technologies, Monash University, Clayton, Victoria, Australia*
[3]*Department of Electrical Engineering and Computer Science, University of California, Berkeley, Berkeley, California, USA*
[4]*Department of Physics, University of California, Berkeley, Berkeley, California, USA*
[5]*Department of Physics and Astronomy, Julius-Maximilians-Universität, Würzburg, Germany*
[6]*Department of Materials Science and Engineering, University of California Berkeley, Berkeley, CA USA*
[7] *State Key Laboratory of Surface Physics, Key Laboratory of Computational Physical Sciences, and Department of Physics, Fudan University, Shanghai, China*
[8]*Department of Physics and Astronomy, University of California, Irvine, CA, USA*
[9]*Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, Berkeley, California, USA*
*julie.karel@monash.edu

The efficient generation of spin currents is critical to numerous low-energy electronic devices, including the attojoule logic gate and spin orbit torque magnetoresistive random access memory (SOT-MRAM). Efforts to identify materials that act as good spin current sources have focused primarily on crystalline systems. In this talk, it will be shown that amorphous materials are potential candidates to generate spin currents. First, the talk will examine the anomalous Hall angle (AHA) in a series of ferromagnetic amorphous transition metal thin films $M_xY_{1-x}$ (M=Fe, Co; Y=Si, Ge; x=0.40-0.71). It will be shown that the AHA ($=\sigma_{xy}/\sigma_{xx}$) is as large as 5%, which is substantial even for crystalline systems. In the amorphous materials it was found that the AHA increases with increasing Hall conductivity ($\sigma_{xy}$). This trend, which is opposite to that which occurs in crystalline systems, is attributed to low $\sigma_{xx}$, while $\sigma_{xy}$ and M remain high. [1] The talk will then report observation of a large spin-orbit torque in an amorphous non-magnetic $Fe_xSi_{1-x}$/cobalt bilayer via spin-torque ferromagnetic resonance and harmonic Hall measurements. The origins of this SOT will be discussed. [2]

## References

[1] J. Karel *et al.*, *Phys Rev Materials* **4** (2020) 114405
[2] C.-H. Hsu, J. Karel *et al.*, https://arxiv.org/abs/2006.07786

# Single Domain Spin Orbit Torque Enabled Magnetic Field Sensor With Offset Compensation

Johannes Guettinger[1], Sebastian Zeilinger[1,2], Armin Satz[1], Sabri Koraltan[2] and Dieter Suess[2]
[1] *Infineon Technologies Austria, Siemensstraße 2, Villach, Austria*
[2] *University of Vienna, Kolingasse 14-16, Vienna, Austria*
*johannes.guettinger@infineon.com

Reducing the zero-field offset is crucial for precise field measurements as a small offset allows for a higher amplification and less parasitic effects from temperature and mechanical stress. In this work we present a device that can be used to measure in-plane magnetic fields with an out of plane sensitive sensor and compensate the respective offsets. The device employs a modulated spin-orbit-torque (SOT) to reduce the offset. The sensor is sensitive to in-plane fields parallel to the bias current direction ($I_{xx}$ in Fig. 1b). Depending on whether a positive or negative current is applied in the heavy metal layer (HM), the damping like spin-orbit torque results in a positive or negative Mz component of ferromagnetic layer (FM), respectively. This out of plane Mz component is proportional to the external B field ($B_{ext}$) and it is measured via the anomalous Hall effect ($V_{xy}$). The signal is obtained by subtracting the measured Mz components of the two opposing current directions (see Fig. 2). We are able to reproduce the observed characteristic with a single spin model in contrast to previous work, where out of plane polarization of the FM and domain wall motion was observed [1]. It is shown that bias fields applied orthogonal to the sensitive direction do not affect the offset or the sensitivity as long as they are smaller than the linear range of the sensor.

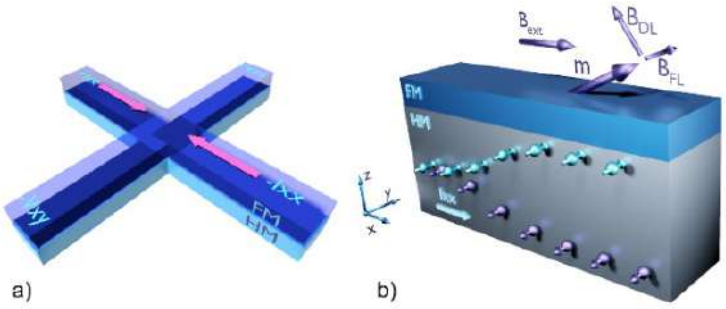



Figure 1: Current induced SOT. a) The Hall cross consists of a ferro magnetic layer on top of a heavy metal layer. b) The SOT current (Ixx) induces a torque on the magnetization of the ferromagnetic layer which can be described by an dampinglike (BDL) and the fieldlike (BFL) effective field.

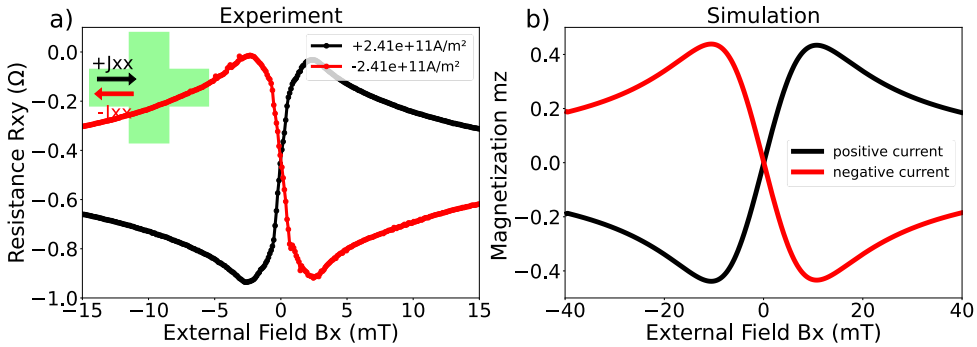



Figure 2: a) Measurement of an external Bx field. a) Flipping the current direction changes the sign of the signal, but the offset stays the same. b) Simulation of the normalized z-magnetization (mz) with a single spin model.

**References**

[1] Li, R., Zhang, S., Luo, S. et al. A spin–orbit torque device for sensing three-dimensional magnetic fields. Nat Electron 4, 179–184 (2021). https://doi.org/10.1038/s41928-021-00542-8

# Orbital Hanle Magnetoresistance in Light Transition Metals

*Giacomo Sala*[1]*, *Hanchen Wang*[1], *William Legrand*[1], *and Pietro Gambardella*[1]

[1] *Department of Materials, ETH Zürich, Hönggerbergring 6, 8093 Zürich, Switzerland*

*giacomo.sala@mat.ethz.ch

Recent theories have shown that orbital moments play a pivotal role in current-induced effects in transitional-metal thin films [1-2]. In particular, electric currents in 3*d* elements can generate a substantial non-equilibrium orbital accumulation that is comparable to or even larger than the spin accumulation caused by the spin Hall effect and the Rashba-Edelstein effect in 4*d* and 5*d* elements. The orbital accumulation can exert spin-orbit torques on ferromagnets and lead to magnetoresistance effects in multi-layered samples [3-5].

Here, we present room-temperature measurements of the orbital Hanle magnetoresistance in single Mn layers with variable thickness. We show that magnetic fields orthogonal to the orbital accumulation cause a modulation of the longitudinal and transverse resistances of the order of $6.5 \cdot 10^{-5}$ and $2 \cdot 10^{-5}$, respectively, which is comparable in magnitude to the spin Hall and Hanle magnetoresistance measured in Pt-based heterostructures [6]. The dependence of the Hanle magnetoresistance on the thickness of Mn and the magnetic field allows us to estimate an orbital relaxation time of 1.5 ps and a diffusion length of the order of 2 nm. In addition, the combination of a large Hanle magnetoresistance with a vanishingly-small spin Hall magnetoresistance in BiYIG/Mn confirms the orbital origin of the observed effects because orbital moments do not interact with the magnetization.

Our findings reveal that current-induced orbital moments can be responsible for magnetoresistance effects as large as those determined by spin moments, and provide a tool to investigate the orbital physics.

## References

[1] D. Go, *Phys. Rev. Lett.* **121** (2018) 86602.
[2] L. Salemi, *Nat. Comm.* **10** (2019) 1-10.
[3] D. Lee, *Nat. Comm*. **12** (2021) 6710.
[4] S. Ding, *Phys. Rev. Lett*. **128** (2022) 067201.
[5] G. Sala, *Phys. Rev. Res.* **4** (2022) 033037.
[6] S. Velez, *Phys. Rev. Lett*. **116** (2016) 016603.

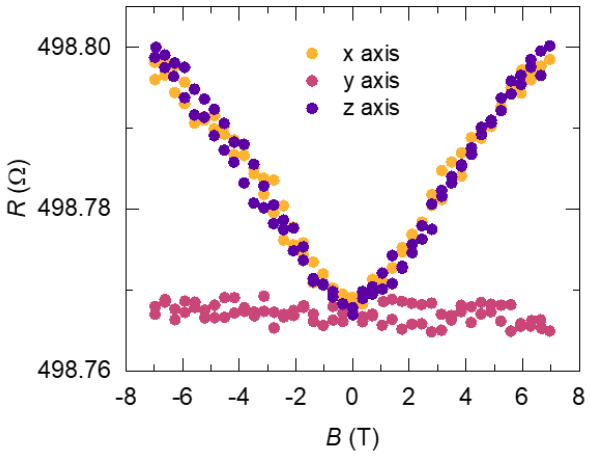



Figure: Hanle magnetoresistance in single layers of Mn. Dependence of the longitudinal resistance of a Mn(9 nm) Hall bar device on a magnetic field oriented along three orthogonal directions; the electric current flows along *x*, and *z* is the direction normal to the sample plane.

# Evidence of orbital current and orbital torques in transition metals using oxidized Cu light element

*B. Bony[1], S. Krishnia[1], S. Collin[1], J. M. George[1], N. Reyren[1], V. Cros[1], H. Jaffrès[1]*

[1] *Unité Mixte de Physique CNRS, Thales, Université Paris-Saclay, 91767 Palaiseau, France*

*benjamin.bony@cnrs-thales.fr

In the field of spin-orbitronics, recent works [1] demonstrated experimentally a significant magnetic torque in light elements which may be only explained by considering the occurrence of an orbital angular momentum current and then its conversion into spin current through spin-orbit coupling. Large orbital currents have been indeed predicted to be generated at interface between Cu and CuOx by orbital Rashba-Edelstein effect [2]. In this work, we quantify the current-induced torques by harmonic Hall voltage measurements on Co(2)/Pt(t)/Cu*(3) (Cu* corresponding to a naturally oxidized copper layer) and for comparison Co(2)/Pt(t), Co(2)/Pt(t)/AlOx(1). We extract the amplitude of damping-like torque exerted on the magnetization (see Fig. 1a) in systems Co(2)/Pt(t)/Cu*(3) compared to the reference systems Co(2)/Pt(t) and Co(2)/Pt(t)/AlOx(1). In the system including a Cu/CuOx interface (namely Cu*(3)), we observe a noteworthy enhancement of the torque up to two fold for 4 nm Pt (Fig. 1a). Assuming an orbital current generation in the top Cu/CuOx interface, such an increase can be interpreted through the existence of a large orbital-to-spin conversion as well as a diffusion of the orbital current through the Pt layer. Then we further characterize the spin Hall magnetoresistance measurements (Fig. 1b) on Co(2)/Pt(t)/Cu*(3) as well as on Co(2)/Pt(t), with also an increase about a factor 2.

Recently, similar behaviour showing an increase of the torque have been found in the case of an insulating garnet film covered by Pt/Cu* either using harmonic Hall measurement [3] or ferromagnetic resonance [4]. The Pt thickness for which the maximum of the torque stands is shifted compared to our Co/Pt/Cu*. To understand this difference, we have measured the Co thickness dependence of the effective torque in Co(t)/Pt(3-4)/Cu*(3) in order to differentiate the orbital and spin currents thanks to their different critical length scales. Finally, we also characterize some pure orbital torque contribution in Co(t)/Cu*(3) as well as in spin-pumping measurements.

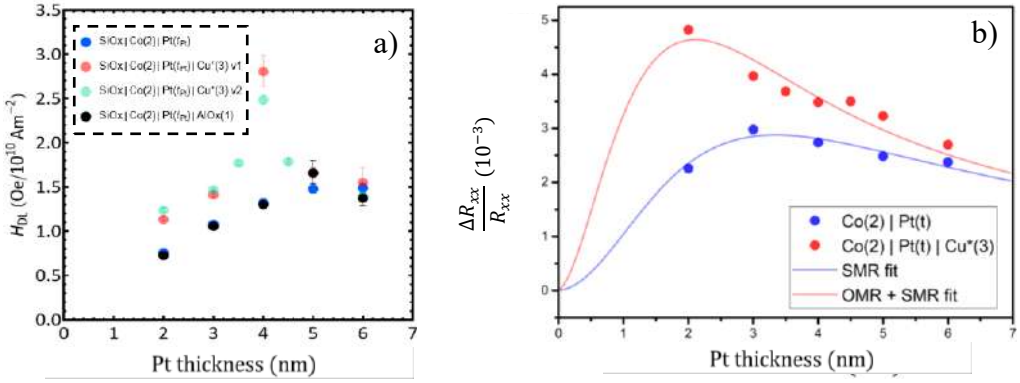



Fig. 1: Pt thickness dependence of a) the normalized damping-like torque evaluated by harmonic Hall voltage measurements b) the spin Hall magnetoresistance.

**Acknowledgements**

Financial support from the Agence Nationale de la Recherche, France, No. ANR-20-CE30-0022 (ORION)

**References**

[1] S. Lee et al., Commun. Phys. 4, **234** (2021)
[2] D. Go, D. Jo, *Phys. Rev. B* **103**, L121113 (2021)
[3] S. Ding et al., *Phys. Rev. Lett.* **125**, 177201 (2020)
[4] E. Santos, arXiv:2204.01825v2

# Ultrathin NiO Interlayer for Enhanced SOT Efficiency in Pt/Co Systems for SOT MRAM

*M. Wijshoff[1,2]*, R. Carpenter[1], G. Talmelli[1], S. Couet[1], C. Fleischmann[1,2], K. Temst[1,2]*

[1] *imec, Kapeldreef 75, 3001 Leuven, Belgium*

[2] *Quantum Solid-state Physics, KU Leuven, Celestijnenlaan 200D, 3001 Leuven, Belgium*

*michelle.wijshoff@imec.be

The demand for faster, smaller and more power-efficient devices is increasing while conventional memories such as static random-access memory (SRAM) and dynamic RAM (DRAM) are reaching their scaling limits. Magnetic RAM (MRAM) is a non-volatile alternative which will be a next generation memory for consumer applications of spintronics. Spin orbit torque (SOT) MRAM is promising for high-speed (<1 ns) applications such as Last Level Cache memories. In SOT MRAM, a spin-polarised current is generated via the spin Hall effect in a heavy metal (HM). Diffusion of this spin current into a neighbouring magnetic free layer (FL) exerts a torque, reversing the layer's magnetic orientation. The bottleneck for SOT MRAM is the write efficiency; spin polarisation is lost at the HM/FL interface. To increase this efficiency, a spin transparent metal oxide, such as NiO, CoO, $Fe_2O_3$ [1-3], can be added at the interface of the HM/FL. W. Lin et al. [3] have shown a 10 times increase in the spin current transport after introducing a 1 nm NiO layer. Furthermore, several studies have reported a decrease in the spin backflow and memory loss after an insertion of 10-20 Å of NiO at the HM/FL interface [4,5].

In this work, a thin (5-20 Å) NiO layer is added at a Pt(40 Å)/Co(10 Å) interface in order to study the effect on the damping-like (DL) SOT efficiency; the percentage of write current contributing to magnetic switching [4]. Additionally, a sample with a 200 Å NiO interlayer is included to investigate if antiferromagnetic ordering is required [3,5]. The magnetic tunnel junctions (MTJ) consists of a seed layer, Pt/NiO/Co and a capping layer and are deposited by magnetron sputtering. These are shown to be crystalline, as confirmed by transmission electron microscopy (TEM) (figure 1A).

The magnetic properties were measured before and after annealing. After deposition, perpendicular magnetic anisotropy (PMA) is observerd only in the samples with 5 Å and 20 Å of NiO. After annealing for 5 minutes at 350 °C, all samples have a PMA. This is seen to increase depending on the temperature, in addition to an increase in the $M_s$ and $H_c$ with respect to the reference, as shown in figure 1B. Figure 1C shows the DL SOT efficiency of the NiO samples as measured by the second harmonic method [6]. The efficiency depends on the NiO thickness, increasing from 6.5 % (no NiO) to 26 % (15 Å NiO, 350 °C anneal). This shows that NiO is a promising material to improve the write energy. In the full work, the reason for this improvement will be explored.

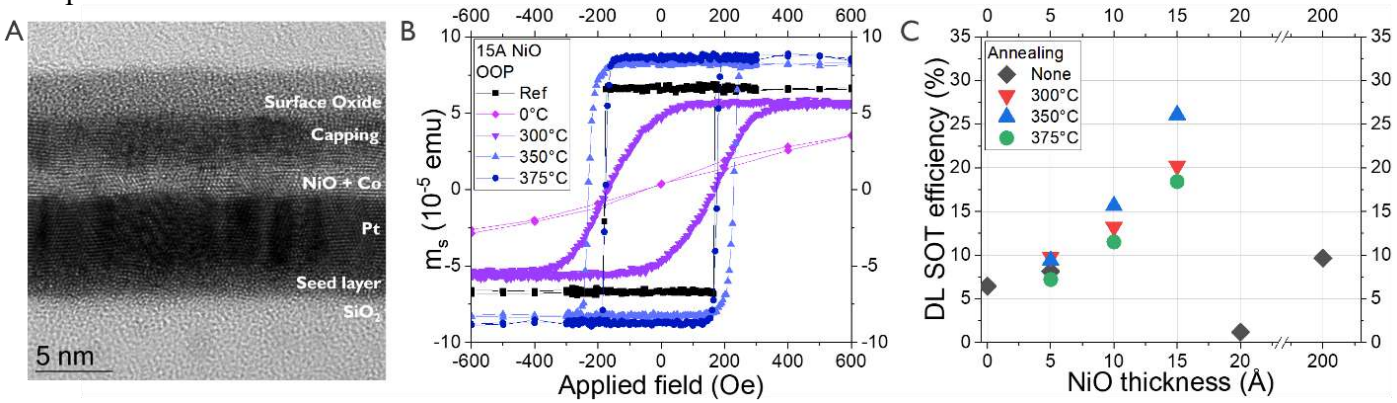



Figure 1: A) TEM analysis of the MTJs with 15 Å NiO shows crystalline layers. B) The OOP hysteresis loops of the 15 Å NiO samples with different annealing temperatures and 'Ref' stands for the reference Pt/Co sample. C) The DL SOT efficiencies depending on the annealing temperatures and NiO thickness measured by the second harmonic method.

**References**

[1] C. Hahn et al., Europhys Lett. 108, 57005 (2014).
[2] R. Lebrun et al., Nature 561, 222–225 (2018).
[3] W. Lin et al., Phys Rev Lett. 116, 186601 (2016)
[4] L. Zhu et al., Phys Rev Lett. 126, 107204 (2021).
[5] H. Wang et al., Phys Rev Appl. 11, 044070 (2019)
[6] C. O. Avci et al., Phys Rev B. 90, 224427 (2014).

# Switching of Magnetization in Synthetic Antiferromagnet Probed by Time-Resolved Magneto-Optical Kerr Effect

*Meiyang Ma [1], Zhuoyi Li [1], Xuezhong Ruan [1], Jing Wu [2]*, Ruifeng Wang [1], Tianyu Liu [3], Jun Du [3], Xianyang Lu [1]*, Yongbing Xu [1,2]**

[1] *Jiangsu Provincial Key Laboratory of Advanced Photonic and Electronic Materials, School of Electronic Science and Engineering, Nanjing University, Nanjing 210093, P. R. China*

[2] *York-Nanjing Joint Center (YNJC) for Spintronics, Department of Electronic and Physics, The University of York, York YO10 5DD, UK*

[3] *Department of Physics, Nanjing University, Nanjing 210093, P. R. China*

* jing.wu@york.ac.uk, xylu@nju.edu.cn, ybxu@nju.edu.cn

Since the discovery in 1996 by Beaurepaire et al. that a femtosecond laser pulse can quench the magnetization in nickel thin films within less than a picosecond [1], the field of femtomagnetism has attracted thriving interest. Ultrafast demagnetization, as one of the most key issues, has been the subject of intense research for more than twenty years. In the last decade, a new model based on superdiffusive spin transport has been mentioned, which couples the fields of ultrafast spin dynamics and stationary spin transport [2]. It has been demonstrated that laser excited spin-polarized hot electrons can transport angular momentum to neighboring FM layers, thus changing the orientation and magnitude of magnetization [3].

The discovery of synthetic antiferromagnetic (SAF) materials is well suited to explore the possible magnetization switching and their mechanisms by the laser excited spin-polarized hot electrons in an antiferromagnetic coupled system [4]. The SAF structure can switch the magnetization between parallel and antiparallel under the regulation of the external magnetic field. This unique structure serves as a promising approach for the designing of spin current-controlled spintronic devices.

To uncover the effect of spin-polarized hot-electrons generated by laser excitation on magnetization switching in SAF, we have carried out a systematic study of a $[Pt/Co]_4/Ru/[Co/Pt]_4$ based SAF structure by using the time-resolved magneto-optical Kerr effect (TRMOKE). The optically induced changes in the magnetization of the sample were recorded by measuring the transient changes in the polar Kerr rotation. We have experimentally observed that the evidence of spin-polarized hot electrons will assist the magnetization switching of the lower ferromagnetic layer at low pump fluences of less than 1.60 mJ $cm^{-2}$. This phenomenon highly depends on the relative orientation of magnetization between the layers. Additionally, the strong thermal stability of the Ruderman-Kittel-Kasuya-Yosida (RKKY) exchange interaction effectively prevents further degradation of the switching field at pump fluence above 1.60 mJ $cm^{-2}$.

Our discovery provides fundamental insight into the effect of spin-polarized hot electrons on magnetization switching and is relevant for identifying the role of the interface in ultrafast spin dynamics. We demonstrated that it is possible to regulate the magnetization of the individual ferromagnetic layer in an antiferromagnetically coupled system in a controlled way by injecting photoexcited spin-polarized hot electrons.

## References

[1] E. Beaurepaire, J. C. Merle, A. Daunois, J. Y. Bigot, *Phys. Rev. Lett.* **1996**, 76, 4250.
[2] M. Battiato, K. Carva, P. M. Oppeneer, *Phys. Rev. Lett.* **2010**, 105,027203.
[3] G. Malinowski, F. D. Longa, J. H. H. Rietjens, P. V. Paluskar, R. Huijink, H. J. M. Swagten, B. Koopmans, *Nat. Phys.* **2008**, 4, 855.
[4] R. A. Duine, K. J. Lee, S. S. P. Parkin, M. D. Stiles, *Nat. Phys.* **2018**, 14, 217.

# Bulk and Interface Spin-Orbit Torques in Pt/Co/MgO Structures

*M.S. Gabor[1]*, T. Petrisor jr.[1], M. Belmeguenai[2], I.M. Miron[3]*

[1] *Centre for Superconductivity, Spintronics and Surface Science, Physics and Chemistry Department, Technical University of Cluj-Napoca, Str. Memorandumului, 400114 Cluj-Napoca, Romania*

[2] *LSPM–CNRS, UPR 3407, Université Sorbonne Paris Nord, F-93430 Villetaneuse, France*

[3] *SPINTEC, UMR-8191, CNRS/CEA/Université Grenoble Alpes, Grenoble, 38054 France*

*mihai.gabor@phys.utcluj.ro

Spin-orbit torques (SOTs) are usually evidenced in heavy-metal (HM)/ferromagnetic (FM) bilayers, where an in-plane charge produces a spin accumulation at the HM/FM interface via Spin-Orbit Coupling (SOC). The spin accumulation gives rise to the Damping-Like (DL) and the Field-Like (FL) torques acting on the magnetization of the FM. They can switch the magnetization, induce magnetization precession or can lead to magnetic domain walls (DWs) or skyrmions motion in the FM layer [1]. Several works proposed different methods to engineer either the bulk of the HM or the FM/HM interface to increase the SOTs efficiency: from using resistive HMs to alloying or using insertion layers and spin sinks. One particularly attractive method to enhance the SOTs is to oxidize the HM/FM. The main advantage is that the Oxide/FM interfacial SOTs could be stronger than the HM induced one, while also consuming less power, due to the insulating oxide. Most of the published works to date report an increase of the SOT efficiency by oxidation [2], however there is an ongoing debate as some studies do not observe a clear improvement [3]. One of the main difficulties when quantifying the SOTs in such systems is that during the oxidation of the HM layer, the resistivity varies strongly and affects the current distribution within the stack. Moreover, the oxidation could affect the saturation magnetization of the FM layer leading to an overestimation of the SOTs strength. In order to avoid these limitations, in this work, we grow Pt (4nm)/Co (2m)/MgO (0-3.6 nm)/Pt (4 nm) stacks using ultrahigh vacuum e-beam evaporation, where the MgO layer forms a continuous wedge. The samples are patterned as Hall crosses using standard UV lithography technique. We evaluate the SOTs using the harmonic Hall voltage method. For a MgO film thickness above 0.4 nm the layer is continuous and the electrical resistance of the stack remains rather independent of the MgO layer thickness. Moreover, the saturation magnetization of the Co layer is only slightly affected by increasing the MgO layer thickness and, thus, the availability of oxygen at the Co interface. However, we observe a strong variation of the surface magnetic anisotropy, as expected, since it is well known that the surface anisotropy is highly sensitive to the Co layer interfacial oxidation. The surface magnetic anisotropy corelates well with the FL torque, which also shows a strong variation with MgO layer thickness as the oxygen present at the interface affects the Rashba field at this interface. The DL torque is rather insensitive to MgO layer thickness, suggesting that the Spin Hall Effect in the HM layer is its main origin. The effective Dzyaloshinskii-Moriya interaction (DMI) shows a similar trend, indicating that the Pt/Co interface is the most important source of DMI (Fig.1).

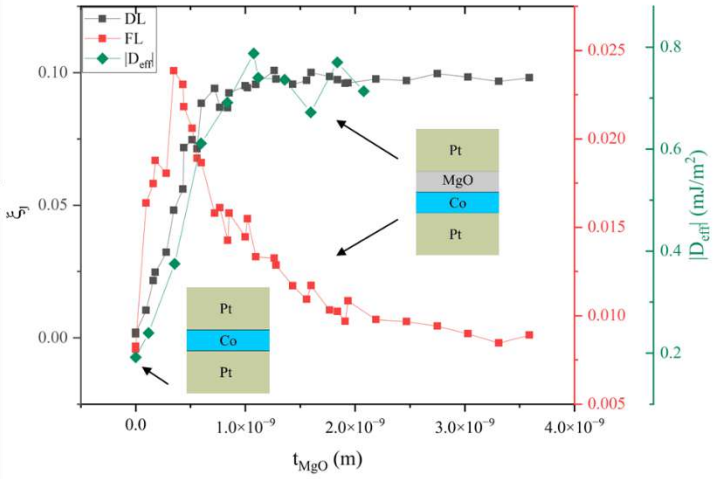



Fig.1: SOTs efficiency and effective DMI constant as a function of the MgO layer thickness.

## References

[1] A. Manchon *et al., Rev. Mod. Phys.*, **91**, 035004 (2019).
[2] K.-U. Demasius *et al.*, Nat. Commun. 2016, 7, 10644.
[3] J. Feng *et al.*, Phys. Rev. Appl. 2020, 13, 044029.

# Temperature-dependent quantification of spin-orbit-torques for Ta/CoFeB/MgO interfaces at low and high current regime

*J. Salazar-Mejía[1]*, S. Koraltan[1], C. Abert[1], P. Flauger[1], M Agrawal[2], S. Zeilinger[1], A. Satz[3], C. Schmitt[4], G. Jakob[4], R. Gupta[4], M. Kläui[4], H. Brückl[5], S. Güttinger[3] and D. Suess[1]*

[1)] *University of Vienna, Faculty of Physics, Physics of Functional Materials, Vienna, Austria*
[2)] *Infineon Technologies AG, Am Campeon 1-15, 85579 Neubiberg, Germany*
[3)] *Infineon Technologies Austria, Siemensstrasse 2, Villach, Austria*
[4)] *Institute of Physics, Johannes Gutenberg-University Mainz, 55128 Mainz, Germany*
[5)] *Universität für Weiterbilding Krems, Department für Integrierte Sensorsysteme, 2070 Wiener Neustadt, Austria*
*joshua.salazar@univie.ac.at

Spin-current manipulation in spintronic devices has demonstrated tremendous potential for technological improvement of nowadays traditional electronic devices. Devices based on ferromagnet/heavy metal (FM/HM) interfaces and Spin-Orbit Torque (SOT) effects are deeply studied for sensing applications [1]. Due to the high spin-orbit coupling of the heavy metal in such interfaces, current-induced SOTs can be employed to produce changes in the magnetization direction of the FM layer. The changes in the magnetization are used to produce a sensor readout depending on the external field applied [2].

To adequately describe the magnetization dynamics of the FM/HM interfaces under study, the current-induced torques need to be properly quantified. Within different ranges of current densities applied on the studied devices, the current-induced Joule heating causes the temperature to change. At different temperatures, the material parameters that describe the system, such as saturation magnetization and/or effective anisotropy, change and thus SOT effective fields are not linear w.r.t current density anymore.

To quantify the SOT coefficients as adimensional constants, adequate material parameters are needed to be characterized for different temperatures, to afterwards perform the SOT coefficients extraction correctly. Different methods are employed to measure the SOT coefficients, longitudinal and transversal (w.r.t to the current direction) field sweeps for out-of-plane configurations [3], and rotational field sweeps along the film plane for in-plane configurations [4]. In this work, we show the results from comparing those two extraction methods for a range of current densities where the saturation magnetization and anisotropy field show significant change. Hence, the extracted SOT coefficients are extracted using temperature-dependent material parameters to adequately describe constant behavior with respect to the current density applied.

## References

1 D. Suess, A. Bachleitner-Hofmann, et al. Nat. Elec. 1, 362–370 (2018)
2 R. Li, S. Zhang, et al. Nat. Elect. 4, 179–184 (2021)
3 M. Hayashi, J. Kim, et al. Phys. Rev. B 9, 144425 (2014)
4 Dutta, S., Bose, Phys. et al. Rev. B, 103(18), 184416 (2021)

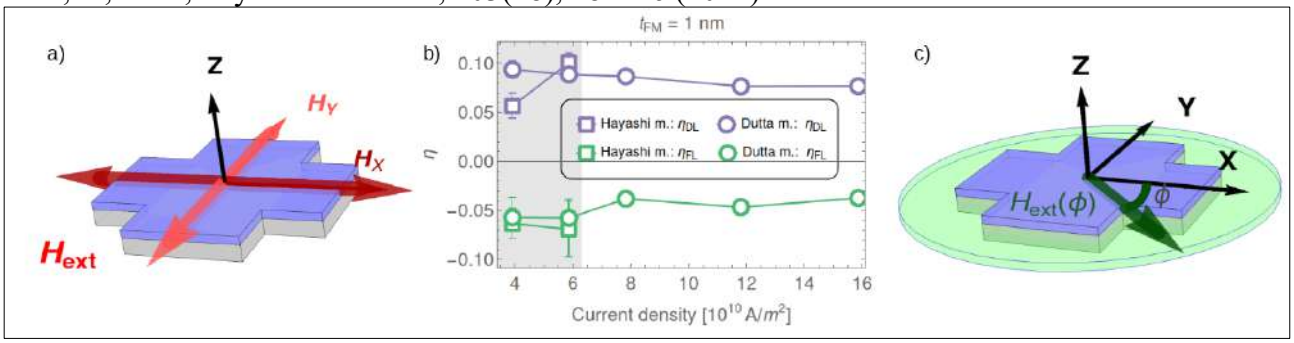



Figure 1: Extracted SOT coefficients under different ranges of current densities. The subfigure a) show the external field sweep direction, red arrows, required for the Hayashi method. Subfigure b) depicts the extracted coefficients from voltage harmonics measurements from a FM/HM interface with CoFeB thickness of 1 nm, where the gray background represents the current density regimes where the device is in OOP configuration. Subfigure c) is a schematic of the external field sweep direction, green disk for the Dutta extraction method.

# Large interfacial Rashba torques in atomically thin ferromagnet

*N Sebe[1], S. Krishnia[1], Y. Sassi[1], F. Ajejas[1,*], N. Reyren[1], S. Collin[1], T. Denneulin[2], A. Kovács[2], R. E. Dunin-Borkowski[2], A. Fert[1], J.-M. George[1], V. Cros[1] and H. Jaffrès[1]*

*[1] Unité Mixte de Physique, CNRS, Thales, Université Paris-Saclay, 91767, Palaiseau, France, vincent.cros@cnrs-thales.fr*

*[2]Ernst Ruska-Centre for Microscopy and Spectroscopy with Electrons (ER-C 1) and Peter Grünberg Institut (PGI-5), Forschungszentrum Jülich GmbH, 52425 Jülich, Germany*

**Present address: Department of Physics and Center for Advanced Nanoscience, University of California, San Diego, La Jolla, CA, 92093, USA*

Spin-orbit interaction (SOI) at metallic interfaces with broken inversion symmetry [1] has been recently the center of spintronic research because it enables fast and efficient magnetization dynamics for spin-based memory (e.g. SOT-MRAM), skyrmion and neuromorphic devices. Indeed, the dynamics of magnetization in spintronics devices is mainly controlled by the efficient spin-orbit torques (SOT) exerted by the generated out-of-equilibrium transverse angular momentum like played by the spin currents. Two main mechanisms *i.e.* (a) spin Hall effect (SHE) in heavy metals such as Pt, Ta and (b) spin or orbital Rashba effect (REE) at interfaces may thus contribute leading to a damping-like (DL) and a field-like (FL) torque [2]. More recently, the occurrence of orbital Hall and Rashba effects in devices integrating light elements were also shown to generate torque or to enhance the already existing SOT in well engineered stack and heterostructures [3-4].

In order to tackle these fundamental issues and provide some routes for improvement of SOT-based devices, we demonstrate here how the insertion of a light metal element interface profoundly affects with a strong benefit both the nature of spin-orbit torque and its efficiency in terms of damping-like and field-like effective fields acting on a very adjacent thin Co layer. We will more focus on the case of Pt/Co/Al/Pt systems with variable thicknesses integrating a top Co/Al interface [5]. We show how the insertion of a Co/Al interface leads to a huge enhancement, by about one order of magnitude, of the FL torque upon the increase of the Al thickness up to 3 nm. By varying the Al and Pt thicknesses, we undoubtedly demonstrate the occurrence of a large Rashba interaction at metallic Co/Al interface that we will discuss in terms of spin vs. orbital Rashba interactions. On the other hand, from the variation of the torque vs. the Co thickness at the very low thickness limit ($t_{Co}$ varying from 0.55 to 1.4 nm) [Fig.1], we extract the main parameters governing the transverse spin-dissipation related to the spin-precession and spin-decoherence and discuss the ensemble of those phenomena that we correlate to the respective spin-Hall magnetoresistance (SMR) and anomalous Hall effect (AHE) response.

## References

[1] A. Soumyanarayanan, N. Reyren, A. Fert, and C. Panagopoulos, *Nature* 539, 509–517, 2016.
[2] A. Manchon *et al.*, *Rev. Mod. Phys.*, 91, 3, 2019
[3] J. Kim, D. Go, H. Tsai, D. Jo, K. Kondou, H.-W. Lee, and Y. Otani, Phys. Rev. B 103, L020407, 2021
[4] S. Ding et al., Phys. Rev. Lett. 125, 177201
[5] S. Krishnia *et al.*, "Large interfacial Rashba interaction and giant spin-orbit torques in atomically thin metallic heterostructures," http://arxiv.org/abs/2205.08486

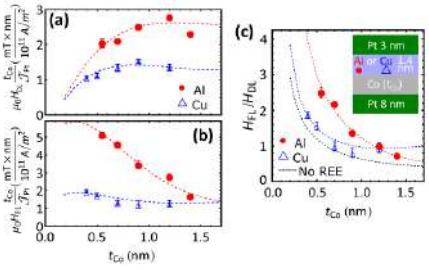



Fig. 1. Co layer thickness dependence of **(a)** DL-SOT field and **(b)** FL-like SOT fields multiplied by Co thickness for $10^{11}$ A/m$^2$ current density in Pt in Pt8|Co ($t_{Co}$)|Al1.4|Pt3 (red circles) and Pt8|Co($t_{Co}$)|Cu1.4|Pt3 (blue triangles) samples (c) $\zeta = H_{FL}/H_{DL}$ as a function of $t_{Co}$. The dashed lines are fits with the theoretical model.

# Electrically-Driven Spin Current Modulation in Silicon

*F. Scali[1]*, C. Zucchetti[1], P. Grassi[1], M. Bollani[1], L. Anzi[1], G. Isella[1], M. Finazzi[1], F. Ciccacci[1] and F. Bottegoni[1]*

[1] *Dipartimento di Fisica, Politecnico di Milano, Piazza Leonardo da Vinci 32, 20133 Milano, Italy*

*francesco.scali@polimi.it

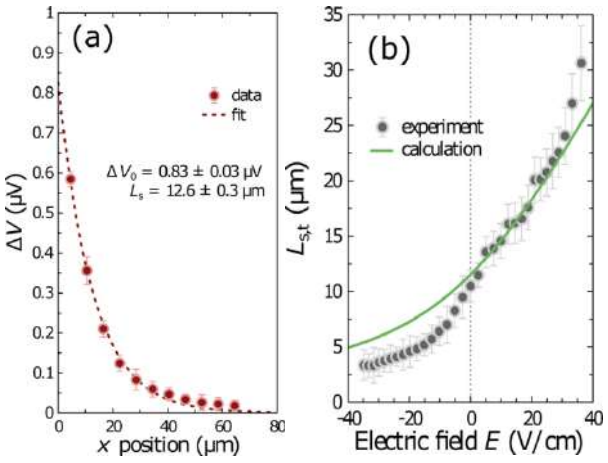
A primarily aim for spintronics is to develop spin-based devices in bulk Si substrates to enhance the functionalities of CMOS-compatible circuits without altering the electronics supply chain. However, due to the weak spin-orbit coupling, the manipulation of the spin degree of freedom of carriers directly inside Si is a difficult task if one excludes the application of energy-consuming magnetic fields [1]. Here, we present an alternative method that takes advantage of the Si spin-transport features to accomplish electrical control of spin-dependent output signals by modulating the spin current rather than the spin itself.

We created a non-local architecture for spin current generation, modulation and detection in lightly *n*-doped ($N_d \approx 1 \times 10^{15}$ cm$^{-3}$) bulk Si at room temperature. Spin-polarized electrons are injected by means of optical orientation technique exploiting phonon-mediated transitions allowed by dipole selection rules at the Si indirect gap [2]. The generation of in-plane spin polarized electrons is obtained by illuminating the edges of Pt stripes deposited onto the Si substrate [3]. Spin currents are detected by means of the Inverse Spin Hall Effect that mediates spin-to-charge conversion inside a Pt stripe that is electrically contacted. The output signal consists in the voltage drop $\Delta V$ across this Pt detector measured in an open circuit condition. We demonstrate that spin-polarized electrons can be driven by an in-plane electric field that fosters or hinders spin diffusion. Electric fields up to 35 V/cm are investigated (applied voltage $V$ up to 7.5 V). As a result, we verify that the ISHE voltage drop can be electrically manipulated to have high or low output values.

The spin diffusion length $L_s$ and the *spin transport length* $L_{s,t}$ (typical distance travelled by spin-polarized electrons before depolarizing under the application of external electric fields) are estimated by fitting the $\Delta V$ profile peaks within a 1D spin transport model (see Fig. 1(a)) [4]. We observe an increase (decrease) of about a factor 3 of the spin transport length with respect to the diffusion length when the electric field is antiparallel (parallel) to the electron diffusion velocity and fosters (hampers) spin diffusion (see Fig. 1(b)). The result is in good agreement with the 1D spin drift-diffusion model proposed in Ref. [5]. Finally, we observe that the output ISHE voltage drop can be electrically-driven between two well-defined logic states when the spins are injected at a distance larger than $L_s$ (Fig. 1(c)), revealing that our architecture essentially acts as a spintronic logic gate.

## References

[1] R. Jansen, *Nat Mater* **11** (2012), 400.
[2] F. Bottegoni *et al.*, *Appl Phys Lett* **110** (2017), 042403.
[3] F. Bottegoni *et al.*, *Nat Mater* **13** (2014), 790.
[4] C. Zucchetti *et al.*, *Phys Rev B* **96** (2017), 014403.
[5] Z. G. Yu and M. E. Flatté, *Phys Rev B* **66** (2002), 2012021.



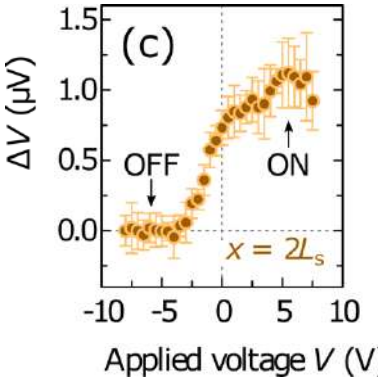




Figure 1. (a) ISHE signal $\Delta V$ (dots) measured as a function of the distance $x$ between the spin injection point and the ISHE detector. The dashed line represents an exponential fit that provides an estimate of the spin diffusion length $L_s$. (b) Spin transport length $L_{s,t}$ (see text) as a function of the electric field $E$. (c) ISHE signal $\Delta V$ as a function of the applied bias $V$ for spin generation at a distance $x = 2L_s$ from the Pt detector. The light power on the sample is 7 μW. The spot is diffraction limited and is obtained with a 0.7 numerical aperture microscope objective.

# Non-Volatile Electric-Field Control of Spin-Orbit Torques in Perpendicular Ferromagnet - $SrTiO_3$ System

*Paolo Sgarro[1*], Aurélie Kandazoglou[1], Cécile Grezes[1], Maxen Cosset-Cheneau[1], Luis M. Vicente Arche[2], Salvatore Teresi[1], Théo Frottier[1], Maxime Culot[1], Filip Osana[1], Paul Noël[1], Stephane Auffret[1], Kevin Garello[1], Manuel Bibes[2], Laurent Vila[1], and Jean-Philippe Attané[1]*

[*] paolo.sgarro@cea.fr

[1] *Université Grenoble Alpes / CEA / IRIG/ SPINTEC, Grenoble, France*

[2] *Unité Mixte de Physique, CNRS, Thales, Université Paris-Saclay, Palaiseau, France*

The realization of magnetization switching induced by in-plane current injection in heavy metal/ferromagnetic heterostructures has drawn increasing attention to spin-orbitronics, leading to the advent of spin-orbit torques magnetoresistive random access memories (SOT-MRAM). Oxide 2D electron gases (2DEG) have emerged as alternative spin-orbitronics systems, which benefit from an efficient spin-charge interconversion through the Rashba-Edelstein effects. Recently, we have demonstrated an enhancement of the spin-to-charge conversion efficiency by two orders of magnitude in $SrTiO_3$-based 2DEG compared to conventional heavy metals [1], along with a non-volatile electric-control of the spin-to-charge conversion [2]. While the sign and efficiency of the SOTs are fixed by the stack in conventional SOT devices, achieving an electric-control of the charge-to-spin conversion would be of great interest for developing reconfigurable SOT-MRAMs.

Here we report electric-control of SOTs with electrical remanence in a perpendicular ferromagnet-$SrTiO_3$ system. Non-volatile electric-control of the sheet resistance is achieved with 1150% contrast, and two switchable remanent resistivity states of the 2DEG. SOT effective fields are further measured using second harmonic Hall methods. A remanent electric-control of the SOT efficiency is demonstrated, with sign inversion of the anti-damping-like effective field. These results are consistent with a combination of both intrinsic modulation of the SOT efficiency together with extrinsic modulation due to the non-volatile electric-control of the current injection in the 2DEG. The non-volatile control of the SOT effective field is evidenced by reproducible inversion of the SOTs after initializing with voltage pulses of ±130 V (applied across the 500 µm $SrTiO_3$ substrate), opening the way to reconfigurable SOT memories and logic-gate architectures.

## References

[1] D. C. Vaz et al., Nature Materials, 18, 1187–1193(2019).
[2] P. Noel et al., Nature 580, 483–86 (2020).

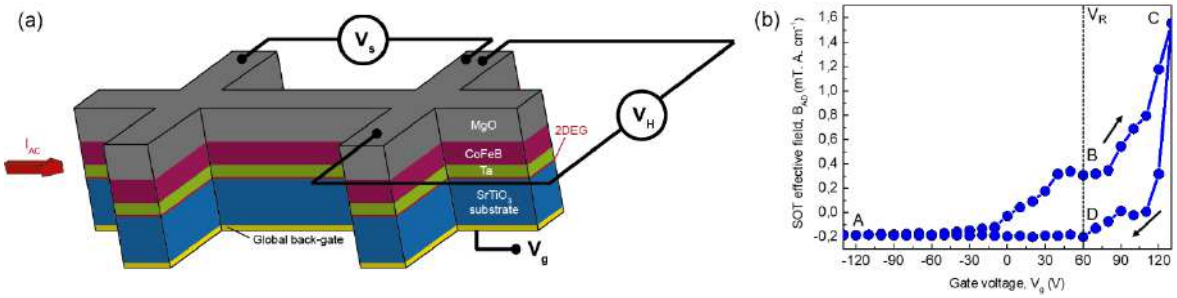



**Figure 1. (a)** Structure of the sample and measurement geometry. **(b)** Electric-field control of spin orbit torques with electrical remanence.

# Engineering of the ferroelectric Rashba semiconductor $Ge_xSn_{(1-x)}Te$ towards ultralow power spintronics

*F. Fagiani[1*], L. Nessi[1], G. Gandini[1], S. Cecchi[2], F. Delodovici[3], G. Vinai[4], J. Fujii[4], I. Vobornik[4], R. Calarco[5], K. Rohit[6,7], A. Rubano[6], M. Fiebig[7], S. Picozzi[3], R. Bertacco[1], M. Cantoni[1], C. Rinaldi[1]*

[1] *Dipartimento di Fisica, Politecnico di Milano, via G. Colombo 81, 20133, Milan, Italy*
[2] *Paul-Drude-Institut für Festkörperelektronik, Hausvogteiplatz 5-7, 10117, Berlin, Germany*
[3] *Consiglio Nazionale delle Ricerche, CNR-SPIN c/o Università G. D'Annunzio, 66100, Chieti,*
[4] *Istituto Officina dei Materiali (CNR-IOM), Laboratorio TASC, Q2 Building, 34149, Trieste, Italy*
[5] *Istituto Microelettronica e Microsistemi (CNR-IMM), via del Fosso 100, 00133, Rome, Italy*
[6] *Dipartimento di Fisica, Università Federico II, via Cinthia, 80126, Naples, Italy*
[7] *Department of Materials, ETH Zurich, 8093, Zurich, Switzerland*
*federico.fagiani@polimi.it

The power consumption of information and communication technology is constantly increasing and on the verge of becoming unbearable. A thrilling solution towards atto-joule beyond-CMOS devices was outlined by Intel in an avantgarde scientific work [1], which proposed to use the combination of spin polarization and collective phenomena (e.g. ferroelectricity and ferromagnetism) for the development of non-volatile logic capable to eventually reach the atto-joule regime. Nonetheless, as recently demonstrated [2], the performance of available materials are far from being satisfying.

In the quest for ultralow power devices, ferroelectric Rashba semiconductors (FERSC) stand out, as they can provide non-volatility, voltage-based control and spin-based processing capability within the very same material. Indeed, we demonstrated an intrinsic coupling between the ferroelectric polarization and the spin-to-charge current conversion mediated by giant Rashba effect in epitaxial GeTe(111) films [3]. This may enable information storing and reading in one single semiconductor endowed with silicon compatibility.

To meet the requirements of applications (e.g. switching voltages below 1 V, low power consumption, …), it is important to provide a way to tune some of the properties of the material to a certain extent. Here we show that by alloying GeTe [4] and SnTe [5], it is possible to tune both the ferroelectric and the Rashba properties of the ternary compound $Ge_xSn_{(1-x)}Te$. The addition of Sn weakens the ferroelectricity with respect to pure GeTe, in turn, lowering the voltage (and therefore the energy) required to switch the ferroelectric polarization (see Fig. 1a). Accordingly, the Curie temperature decreases with the percentage of Sn, while we identify the range of compositions compatible with room temperature ferroelectricity (see Fig. 1b). Endorsed by *ab-initio* calculations, spin and angular resolved photoemission spectroscopy experiments prove the persistence of giant Rashba effect in the ternary compound, possibly allowing for spin-to-charge current conversion processes.

This research sheds light concerning the possibility to engineer the ferroelectric and spin properties by composition towards the development of non-volatile logic-in-memory devices based on ferroelectric Rashba semiconductors.

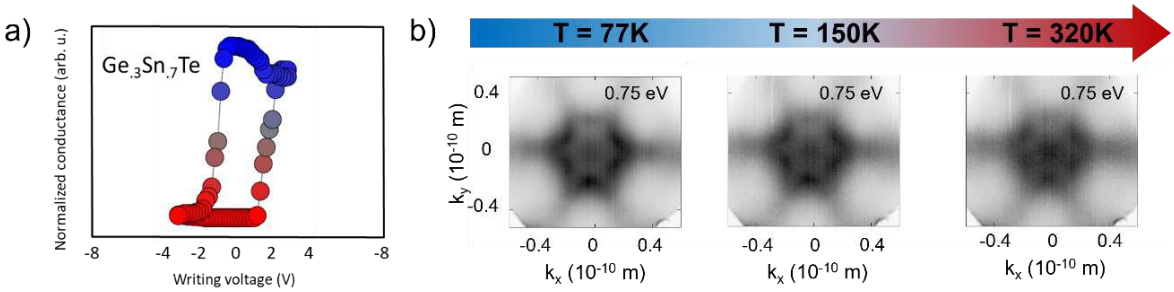



Figure 1: a) Ferroelectric hysteresis loop of $Ge_{30}Sn_{70}Te$ obtained by electro-resistive measurements. b) The Fermi contour of Rashba bands persists up to room temperature in the $Ge_{70}Sn_{30}Te$, indicating the presence of giant Rashba spin splitting.

## References

[1] S. Manipatruni *et.al.*, *Nature* **565** (2019) 35
[2] D.C. Vaz, F. Casanova *et.al.*, arXiv:2302.12162 (2023)
[3] S. Varotto, L. Nessi, F. Fagiani, C. Rinaldi *et.al.*, *Nat. Electron.*, **4** (2021) 740
[4] D. Di Sante *et.al.*, *Adv. Mater.*, **25** (2013) 509
[5] E. Plekhanov, S. Picozzi, *Phys. Rev. B*, **90** (2014) 161108R

# Spin-to-Charge Conversion in Ferroelectric Germanium Telluride (GeTe) For a New Generation of Logic Devices

*Salvatore Teresi[1], Aurélie Kandazoglou[1*], Maxen Cosset-Cheneau[1], Théo Frottier[1], Paolo Sgarro[1], Michael Wissmann[1], Maxime Culot[1], Pierre Noé[2], Jessy Paterson[2], Françoise Hippert[3], Louis Hutin[2], Fabien Cheynis[4], Frederic Leroy[4], Boris Croes[4], Manuel Bibes[5], Guillaume Prenat[1], Alain Marty[1], Jean-Philippe Attané[1], Laurent Vila[1]*

*[1]Université Grenoble Alpes / CEA / IRIG / SPINTEC, Grenoble, France*
*[2]Université Grenoble Alpes / CEA / LETI, Grenoble, France*
*[3]Université Grenoble Alpes / CNRS / Grenoble INP / LMGP, Grenoble, France*
*[4]Aix Marseille Université / CNRS / CINAM / AMUTECH, Marseille, France*
*[5]Unité Mixte de Physique CNRS/Thalès, Université Paris-Saclay, Palaiseau, France*
*aurelie.kandazoglou@cea.fr

The recent announcement of Intel for a new kind of logic device called MESO (Magneto-Electric Spin-Orbit) [1] opens the door to ultra-low energy computing schemes with enhanced logic density. One of the key element is the reading block of the device that relies on the interconversion between spin and charge current using the spin orbit coupling. Finding a material with high spin to charge conversion is indeed crucial for the MESO technology to reach the 100 mV signal required as an output.

In the meantime research on spin to charge conversion led to the discovery of materials possessing both spin-charge interconversion and ferroelectricity [2,3]. These materials exhibit a change of the sign of the spin to charge conversion by reversing the ferroelectric polarization of the material. This recent discovery led to the design of a new type of logic device called the FESO (FerroElectric Spin-Orbit). This device exhibits the same important features than the MESO device like the non-volatile control of the information for in-memory computing. The output signal is controlled electrically but there are no magnetization switching thus excluding the need of an efficient magnetoelectric material like in the writing block of MESO. Indeed the output signal is controlled directly by the ferroelectric state and does not need to reverse the magnetic state of a spin injection layer. Finding the best spin-charge conversion signal controlled by ferroelectricity is thus of great importance. One possibility is the use of two-dimensional electron gases (2DEG) like STO [2]. Another possibility is the use of ferroelectric Rashba semiconductors (FERSC). Among the available FERSC one good candidate is Germanium Telluride (GeTe). Recent studies [3] show the experimental demonstration at room temperature of the spin-charge conversion in GeTe and the control of the sign of this conversion by the ferroelectric state of the material. In order for GeTe to be relevant for the FESO technology it must exhibit the same properties in nanopatterned stacks than in the pristine one.

In this work, we first introduce the FESO principle as a solution for in-memory computing, and present how it simplifies the MESO principle. We then briefly present the FERSC and the state of the art on GeTe as one of them. Finally, we show our all-electrical measurements on the spin to charge conversion in FESO devices nanopatterned on large-scale grown GeTe. We also show that our concept scales favorably with the geometrical downscaling.

## References

[1] Manipatruni, Sasikanth, et al., Nature 565.7737 (2019): 35-42.
[2] Noël, Paul, et al., Nature 580.7804 (2020): 483-486.
[3] Varotto, Sara, et al., Nature Electronics 4.10 (2021): 740-747.

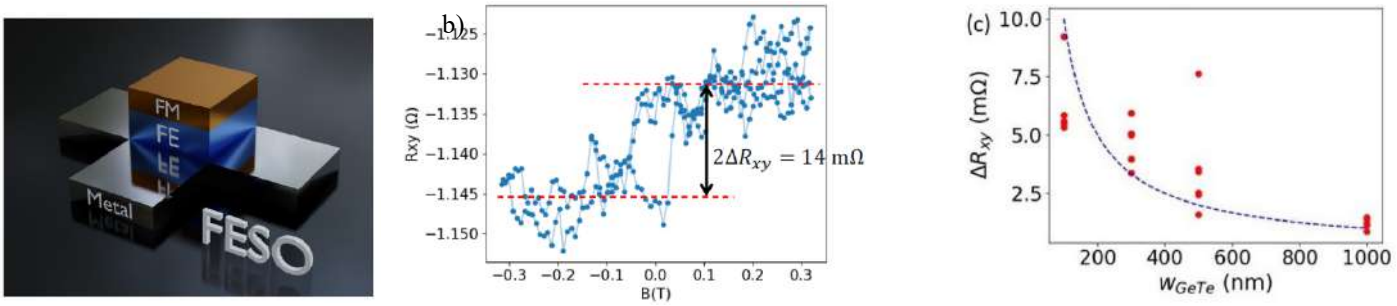



**Figure 1. (a)** Concept of a FESO device. **(b)** Spin-charge Interconversion on nanopetterned 200 nm GeTe. **(c)** Signal amplitude versus the width of the channel.

# Mutual Synchronization of 50 Serially Connected Spin Hall Nano-Oscillators

*Akash Kumar[1,2], Roman. Khymyn[1], Artem Litvinenko[1], Mona Rajabali[2], Nilamani Behera[1], Xiaotian Zhao[1], Ahmad A.Awad[1,2] and Johan Åkerman[1,2]*

[1]*Department of Physics, University of Gothenburg, 412 96 Gothenburg, Sweden*
[2] *Center for Science and Innovation in Spintronics, Tohoku University, Sendai 980-8577 Japan*
[3]*NanOsc AB, Electrum 229, 164 40 Kista, Sweden*
*akash.kumar@physics.gu.se

Mutual synchronization of two or more spin-torque nano-oscillators (STNOs) has been of intense interest since their introduction [1], as it not only improves both microwave signal power and signal quality factor (Q-factor), appealing to communication technology, but can also be used directly for neuromorphic computing due to the tunable magnetic nature of the interaction between STNOs [2]. Thanks to the spin Hall effect, a new class of spintronic oscillators, known as spin Hall nano-oscillators (SHNOs) [3], has emerged. In comparison to STNOs, they rely on current flowing in-plane, which simplifies their fabrication and allows a large number of SHNOs to be synchronized in both 1D chains and 2D arrays [4-6].

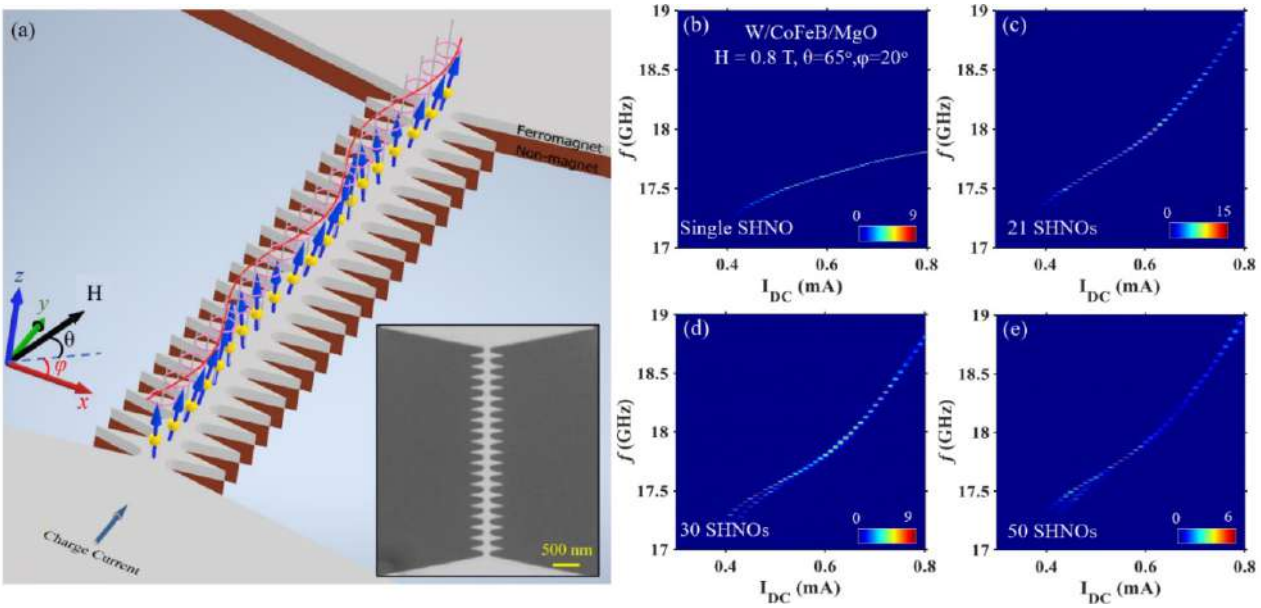



Figure 1 (a) Schematic of 21 serially connected SHNOs (inset shows the SEM image for the fabricted sample). (b-e) Power spectral density for single, 21, 30 and 50 serially connected nano-constriction SHNOs.

Here, we study mutual synchronization in SHNO chains of up to 50 serially connected nano-constrictions fabricated from W(5nm)/CoFeB(1.4nm)/MgO(2nm) and W(5nm)/NiFe(3nm) stacks. Figure 1(a) shows the schematic and SEM micrograph of the fabricated samples. We find that robust and complete mutual synchronization can persist in chains of up to 21 oscillators (power spectral density shown in Fig. 1b-c), resulting in a significantly lower linewidth (< 134 KHz) and higher output power (>4000 $(nV)^2/Hz$) compared to single SHNOs. We also observe mutual synchronization in the longer chains (Fig. 1d-e) but with deteriorated performance, which we ascribe to the longer chains separating into shorter mutually synchronized regions. The low current and low field operation of these oscillators, along with large frequency tunability, make them ideal for various spintronic applications such as neuromorphic computing.

## References

[1] S. I. Kiselev, *et al.,* Nature, vol. 425, no. 6956, pp. 380–383, 2003.
[2] S. Tsunegi, *et al.,* Sci. Rep., vol. 8, no. 1, pp. 1–7, 2018.
[3] V. E. Demidov, *et al.*, Appl. Phys. Lett., vol. 105, no. 17, p. 172410, 2014.
[4] A. A. Awad, *et al.*, Nat. Phys., vol. 13, no. 3, pp. 292–299, nov 2016.
[5] A. Kumar, *et al.*, arXiv, vol. 2301.03859, 2023.
[6] M. Zahedinejad, *et al.*, Nat. Nano., vol. 15, no. 1, pp. 47–52, 2020.

# Ultra-low-current Spin Hall Nano-oscillators

*Nilamani Behera[1]*, Avinash Kumar Chaurasiya[1], Victor H. González[1], Lakhan Bainsla[1], Akash Kumar[1], Ahmad A. Awad[1], Himanshu Fulara[2], and Johan Åkerman[1]*

[1]*Physics Department, University of Gothenburg, 41296 Gothenburg, Sweden,*
[2]*Department of Physics, Indian Institute of Technology Roorkee, Roorkee 247667, India*
*nilamani.behera@physics.gu.se*

Recently, nano-constriction based spin Hall nano-oscillators (SHNOs) have emerged as a new versatile class of devices because of their easy fabrication [1], [2], their direct control using voltage gating [3], [4] or laser heating [5], and their tendency towards mutual synchronization in one [6] and two [7] dimensions for potential use in Ising Machines [8]. In all these applications, power consumption, primarily governed by the SHNO auto-oscillation threshold current, is an important figure of merit that needs to be minimized. While the first SHNOs required several mA of current, increased SOT efficiencies [9], reduced lateral dimensions [10] and the addition of perpendicular magnetic anisotropy (PMA) [11], have been different approaches to reduce the threshold current by about an order of magnitude. Here, we combine all three approaches in an attempt to further reduce the threshold current. We report on the magnetodynamic properties and the magnetization auto-oscillations of ultra-low 20 nm wide nanoconstriction SHNOs fabricated from W-Ta/CoFeB/MgO stacks grown on different substrates and seed layers (see Fig.1). Combining an optimized $W_{88}Ta_{12}$ alloy, low damping CoFeB, and a moderate perpendicular magnetic anisotropy, we push the threshold current down to 35 μA in the best devices (see Fig.1). The best overall magnetodynamic properties and lowest threshold currents are obtained when using a 3 nm thick AlOx seed layer in between the high-resistance Si substrate and the $W_{88}Ta_{12}$ layer. The state-of-the-art ultra-low operational current leads to less power consumption in truly nano-scopic dimension SHNO devices, and paves an energy-efficient way to scale oscillator-based neuromorphic computing in networks of linear chains and two-dimensional arrays.

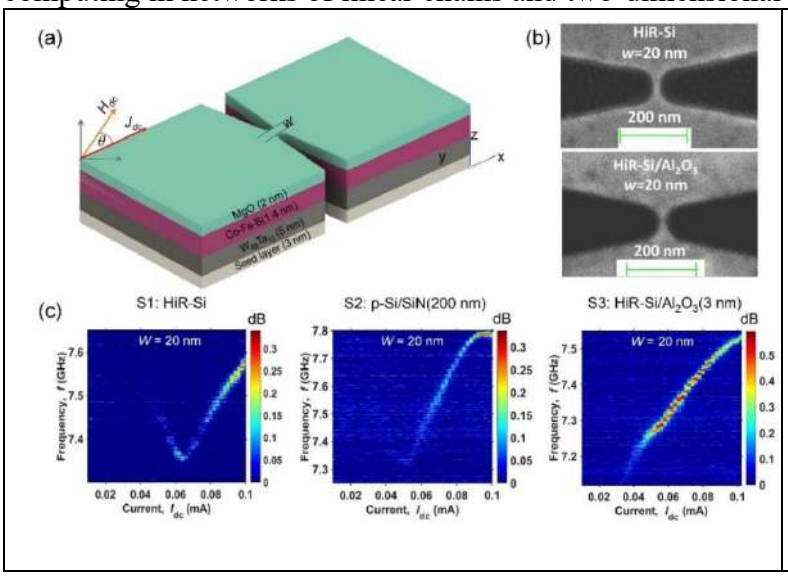



Figure: Fig. 1. (a) Schematic of a nano-constriction-based SHNO device. (b) Scanning electron micrograph (SEM) and of nano-constriction SHNOs with two different seed layers. (c) PSD plots of magnetization auto-oscillation vs. current for 20 nm wide nano-constriction SHNOs. The measurements are performed at an in-plane angle of 22◦, an OOP angle of 60◦, and a constant magnetic field magnitude of 0.3 T.

## References

[1] M. Haidar *et al.*, *Nat. Commun.* **10** (2019), 2362.
[2] V. E. Demidov *et al.*, *Appl. Phys. Lett.* **105** (2014) 172410.
[3] H. Fulara *et al.*, *Nat. Commun.* **11** (2020), 4006.
[4] M. Zahedinejad *et al., Nat. Mat.* **21** (2022) 81–87.
[5] S. Muralidhar *et al., Appl. Phys. Lett.* **120** (2022) 262401.
[6] A. A. Awad *et al.*,, *Nat. Phys.* **13** (2017) 292–299.
[7] M. Zahedinejad *et al., Nat. Nanotechnol.* **15** (2020) 47–52.
[8] A. Houshang *et al., Phys. Rev. Appl*. **17** (2022) 014003.
[9] K. U. Demasius *et al.*, *Nat. Commun.* **7** (2016) 10644.
[10] P. Dűrrenfeld, *Nanoscale* **9** (2017)1285-1291.
[11] H. Fulara *et al.*, *Sci. Adv.* **5 (2019)**, eaax8467.

# Inverse Spin Hall Detection of Surface Magnetostatic Waves in sub-µm W/CoFeB Waveguide

*Daniele Narducci*[1,2]*, *Enrico Tonon*[2], *Johan De Boeck*[1,2], *Inge Asselberghs*[2], *Christoph Adelmann*[2], *Florin Ciubotaru*[2]

[1] *Department of Electrical engineering (ESAT), Kasteelpark Arenberg 10, 3001, Leuven, Belgium*
[2] *IMEC, Leuven, Kapeldreef 75, 30001, Leuven, Belgium*
*Daniele.Narducci@imec.be

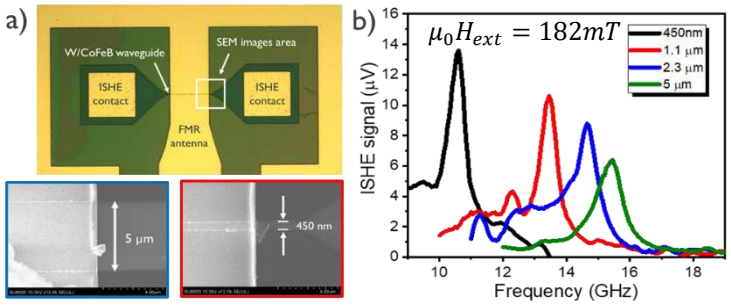



Figure 1: a) Optical image and SEM images of two devices based on 5 µm (blue) and 450 nm (red) W/CoFeB waveguides. b) ISHE voltage as a function of the excitation frequency for waveguide widths as indicated.

In recent years, spin wave devices have gained increasing interest as promising candidates for information carriers in beyond CMOS applications. To remain competitive with other technologies, it is essential for spin wave devices to be scaled down to sub-µm dimensions while maintaining high detection efficiency. While the inverse spin Hall effect (ISHE) has been shown to be independent of the lateral dimension of the waveguide in the case of surface magnetostatic waves [1], previous studies were limited to µm to mm wide devices, where quantization phenomena do not take place or the magnetization non-uniformity is negligible. In this study we investigate the ISHE detection of surface magnetostatic waves in waveguides with lateral dimensions ranging from 5 µm to 450 nm, in which both the quantization and the nonuniformities are highly pronounced [2]. The devices consist of W(10nm)/ CoFeB(40nm)/ Ta(3nm) waveguides covered by a 200nm thick SiN layer for electrical insulation, whereas a 200nm thick Au layer is patterned into antenna and coplanar waveguides. Figure 1(a) displays optical and SEM images of some devices, with a zoom on the 5 µm (blue) and 450 nm (red) waveguides. Two types of excitation antennas were fabricated: a 50 µm wide used to only excite ferromagnetic resonance (FMR), and 600 nm wide antenna for a broadband excitation. The devices were subjected to a magnetic field of 182 mT. The inverse spin Hall voltage was measured as a function of the excitation frequency for devices with different magnetic waveguide widths. The ISHE voltage is shifted to lower frequency when scaling down the width of magnetic waveguide. This is the due to the increase of the shape anisotropy field that decrease the effective magnetic field inside the magnetic waveguides. Additionally, the susceptibility of the scaled devices changes, resulting in an enhanced signal. Figure 2(b) shows the measurement of the resonance peak frequency increasing the external magnetic field for both 600nm and 50µm wide antenna with 450 nm wide waveguides. A frequency shift of up to 1.5 GHz for the same applied magnetic field can be seen once the waveguide reaches saturation above 100 mT [2]. This frequency shift can be understood using the dispersion relation of surface magnetostatic waves. In large waveguides the ISHE peak position is found near the FMR frequency, whereas in scaled waveguides it is shifted to higher wavevectors and consequently, higher frequencies due to quantization effects.

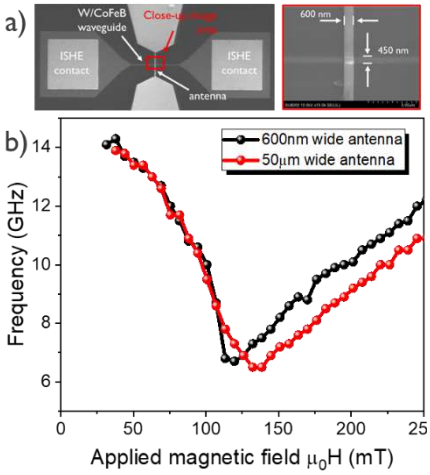



Figure 2: a) SEM image of a 450 nm wide magnetic waveguide and 600 nm wide antenna device b) Frequency peak position of the ISHE voltage for different external applied field of the FMR devices and antenna devices with a 450 nm wide waveguide.

## Acknowledgements

D.N. acknowledges financial support from the Research Foundation – Flanders (FWO) through Grant Nos. 1SB9121N.

## References

[1] H. Nakayama, K. Ando, K. Harii, Y. Fujikawa, Y. Kajiwara, T. Yoshino and E. Saitoh, *J. Phys.: Conf. Ser.* **266** (2011) 012100

[2] G. Talmelli, D. Narducci, F. Vanderveken, M. Heyns, F. Irrera, I. Asselberghs, I. P. Radu, C. Adelmann and F. Ciubotaru, *Appl. Phys. Lett.* **118** (2021) 152410

# Acoustic Spin Hall Effect Induced by Piezoelectric Acoustic Waves

*Takuya Kawada[1*], Hiroki Matsumoto[1], Masashi Kawaguchi[1], Hiroshi Kohno[2], and Masamitsu Hayashi[1]*

[1] *Department of Physics, The University of Tokyo, 113 0033, Tokyo, Japan*
[2] *Department of Physics, Nagoya University, 464 8602, Nagoya, Japan*
*takuya.kawada@phys.s.u-tokyo.ac.jp

Spintronics is a field to study spin-dependent electronic properties in solids. Spin current [1], a flow of spin angular momentum, plays a central role in recent spintronics as a carrier of spin information. It is of significant importance to explore means to induce spin current not only to develop next-generation spintronic devices but also to unveil the coupling mechanisms between spin and other degrees of freedom. Studies have shown that spin current can be generated by applying electric current [2], thermal gradient [3], and circularly polarized light [4]. Recently, mechanical generation of spin current was reported [5][6] using surface acoustic wave (SAW), a vibrational mode localized at solid surfaces.

In this work, we demonstrate that SAW induces AC spin current in a heavy metal (HM) via spin-orbit interaction (acoustic spin Hall effect [7]). A nonmagnetic metal (NM)/ferromagnet (FM) heterostructure is deposited on a ferroelectric substrate. SAW is coherently excited by applying an rf signal to an interdigital transducer (IDT) fabricated on the substrate. DC voltage across the film is measured under the excitation of the SAW as a fuction of an in-plane magnetic field. We find a sizable field-dependent DC voltage (acoustic voltage) develops only when the NM layer is composed of HM such as W and Pt. The amplitude of the acoustic voltage increases with increasing the HM layer thickness and it takes the maximum at a certain HM layer thickness, suggesting that the applied SAW induces the spin current in the HM layer. Experimental results for the HM/FM heterostructures with different layers indicate that the magnetoelastic coupling contributes to the acoustic voltage. Acoustic voltages are also measured with applying SAWs in various directions with respect to the electric polarization of a ferroelectric substrate. We find the magnetic field angle dependence of the acoustic voltage is "phase-shifted" between devices with different SAW propagation directions, implying that the electric field associated with the SAW may play an essential role in the spin current generation. Taking all the experimental results into account, we conclude that the acoustic voltage (DC) is generated due to rectification of the SAW-induced AC spin current by an in-phase oscillation of the FM layer magnetization via magnetoelastic coupling. The origin of the SAW-induced spin current will be discussed in the presentation.

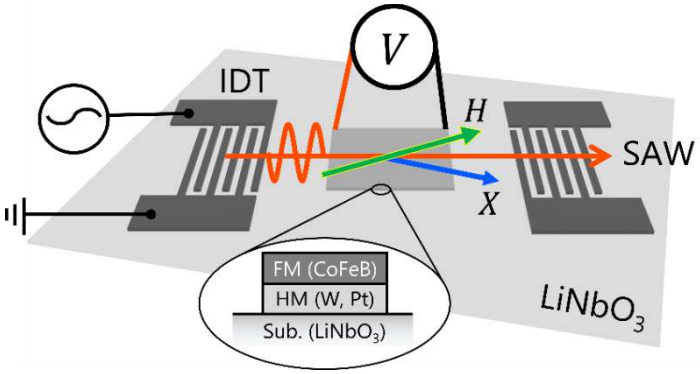



Figure: Schematic illustration of the setup. HM/FM heterostructures and IDTs are deposited on a ferroelectric $LiNbO_3$ substrate. Acoustic voltage is measured as a function of the in-plane magnetic field ($H$). The measurements are performed for SAW devices with different SAW propagation direction with respect to a crystal axis ($X$) of

**Acknowledgements**

This work was partly supported by JSPS KAKENHI (Grant No. 20J21915) from JSPS.

**References**

[1] M. Dyakonov et al, *Phys. Lett. A* 35, 459 (1971).
[2] J. E. Hirsch, *Phys. Rev. Lett.* 83, 1834 (1999).
[3] K. Uchida et al, *Nature* 455, 778 (2008).
[4] H. Hirose et al, *Phys. Rev. B* 103, 174437 (2021).
[5] L. Dreher et al, *Phys. Rev. B* 86 (2012).
[6] D. Kobayashi et al, *Phys. Rev. Lett.* 119 (2017).
[7] T. Kawada et al, *Sci. Adv.* 7, eabd9697 (2021).

# Microwave µW emission power of vortex spin-torque nano-oscillators with a $Co_{20}Fe_{60}B_{20}$/Ta/$Ni_{80}Fe_{20}$ free layer

*Mohammad Qaid*[1], Karsten Rott[2], Gregor Buettel[3], Jürgen Langer[4], Berthold Ocker[4], Uwe Hartmann[3], Günter Reiss[2], Georg Schmidt[1,5]*

[1] *Institut für Physik, Martin-Luther-Universität Halle-Wittenberg, 06120 Halle, Germany*
[2] *Center for Spinelectronic Materials and Devices, Physics Department, Bielefeld University, 33615 Bielefeld, Germany*
[3] *Institute of Experimental Physics, University of Saarbrücken, 66041 Saarbrücken, Germany*
[4] *Singulus Technologies AG, 63796 Kahl am Main, Germany*
[5] *Interdisziplinäres Zentrum für Materialwissenschaften, Martin-Luther-Universität Halle-Wittenberg, Nanotechnikum Weinberg, 06120 Halle, Germany*
*mohammad.qaid@physik.uni-halle.de

Spin-torque nano-oscillators (STNO) based on magnetic tunnel junctions (MTJ) for operation in the MHz-regime were nanofabricated. The multilayer stack, deposited on a *SINGULUS TIMARIS* device, processed in this report was previously reported to allow fabrication and operation of zero-bias spin torque diodes with ultra-high sensitivity in the MHz range [1]. Here, a nominal similar stack was postprocessed by ion beam etching controlled by mass spectroscopy slightly above its $Co_{20}Fe_{60}B_{20}$ free layer and insitu re-sputtering of (8.5)$Ni_{80}Fe_{20}$/(3)Ta/(5)Ru (thickness in nm in brackets) on top to force a vortex domain formation in the oscillating combined free layer upon the MgO barrier. This stochiometry of a $Co_{20}Fe_{60}B_{20}$/Ta/$Ni_{80}Fe_{20}$ combined free layer was so far not reported. CoFeB and NiFe thicknesses were varied and investigated by structural and electrical characterizations to elucidate the behaviour of this material system concerning the vortex dynamics. The output power is enhanced by an order of magnitude in comparison to previously reported STNO based on the gyration of magnetic vortices in circular Permalloy discs and shows a high tunabililty of the power and frequency by varying bias field and current. (Fig. 1). Additionally, the vortex yield is very high for this reported free layer system. Such factors are important for applications like energy harvesting, neuromorphic computing or sensing purposes based on vortex spin-torque devices. The option to fabricate on-chip from the same stack zero-bias macrospin spin-torque diodes in the same frequency range could pave a way to highly integrate and ease new electronic device concepts based on spin-torque oscillators, diodes and circuitries implementing such.

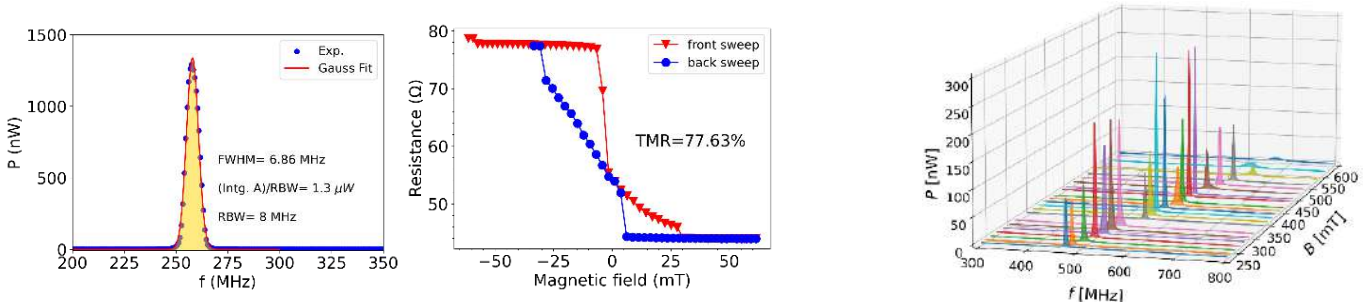



Fig. 1. a) Rf power spectrum of STNO device with a size of 500nm. b) TMR and vortex curve of the same device. c) The power tunability of STNO across a broad range of f and B values, while also maintaining high power.

## Acknowledgements

This work is supported by the Federal Ministry of Education and Research (BMBF) (no. E52FMK024 “spinGMI”)

## References

[1] Fang, B., Carpentieri, M., Hao, X., Jiang, H., Katine, J. A., Krivorotov, I. N., Ocker, B., Langer, J., Wang, K. L., Zhang, B., Azzerboni, B., Amiri, P. K., Finocchio, G., & Zeng, Z., *Nat. Comm.* 7 (2016) 1–7

# Effect of Beam Shift on Helicity Dependent Photoresistance Measurement

*H. Yang[1,3], E. Schmoranzerová[1,2], P. Jang[1],*
*J. Nath[1], T. Guillet[1], I. Joumard[1], S. Auffret[1], P. Němec[2], G. Gaudin[1], and I. M. Miron[1]*
*[1] Univ. Grenoble Alpes, CNRS, CEA, SPINTEC, F-38000 Grenoble, France*
*[2] Faculty of Mathematics and Physics, Charles University, Prague, Czech Republic*
*[3]CIC nanoGUNE BRTA, 20018 Donostia-San Sebastian, Basque Country, Spain*
*eva.schm@karlov.mff.cuni.cz

Efficient generation and detection of spin currents is one of the key ingredients for the modern spintronic applications. Optical methods of spin generation are orders of magnitude faster than the conventional electronics, while simultaneously providing a high spatial resolution. Helicity-dependent generation of spin currents combined with simultaneous detection of photocurrent/photovoltage represents a prominent experiment in this field. The coupling between light polarization and spin current/accumulation has been observed in many systems and is often associated with topologically non-trivial spin structures [1]. Experimentally, a periodical modulation of the light helicity is typically achieved by polarization components such as photoelastic modulator or mechanically-rotated λ/4 waveplate, all of which are based on a change of (complex) index of refraction. These components are extremely sensitive to any imperfection, which limits the laser pointing stability and leads to a periodical beam shift during the measurement [Fig. 1 (c)].
In this contribution, we show how the effective motion of beam can generate artificial signals in the electro-optical experiment via periodically-generated heat gradients. The "beam-shift" signals are indistinguishable from the real spin-related photoresistance in the conventional Hall-bar devices [see Fig. 1 (a), (d)]. We propose a ring-shape device structure that allows for separation of the parasitic signals [see Fig. 1 (b), (e)]. Here the angle difference Δθ between the zero photoresistance position and the diameter across the other side of the zero photoresistance position indicates the mixture of both spin accumulation effect (Δθ = 180°) and beam shift effect (Δθ = 0°) . We show that the "beam-shift" signals dominate the experimental data in various systems under study [1]. This leads us to conclusion that this artefact should be estimated and corrected in all experiments where the circularly polarized laser beam is used to locally influence the electrical properties.

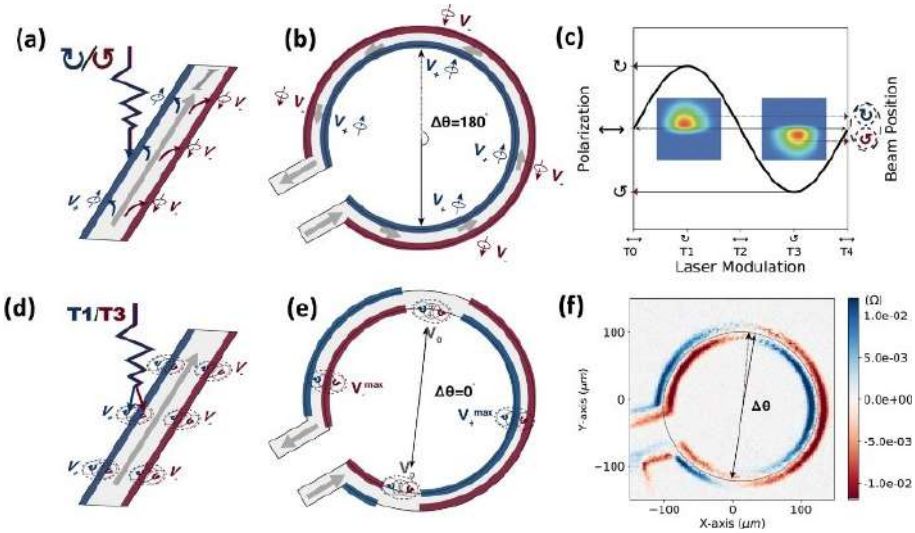



Figure 1: Illustration of spin accumulation and the corresponding helicity dependent photoresistance for (a) a straight current path and (b) a ring-shaped current path. (c) Illustration of the laser spot barycenter displacement associated to the polarization change. The thermal gradients produced by the oscillating beam induce opposite longitudinal resistance variations at the two edges (blue and red) for (d) straight current path and (e) ring-shape device (f) Measured photoresistance mapping for 3 nm thick Pt ring shaped device.

## References

[1] A. McIver et al., Nature Physics **16** (2020) 38.
[2] H. Yang et al., *Nature Communications* **13** ( 2022) 6790.

# Anomalous Hall Enhancement and Field-free Perpendicular Magnetization Switching in Synthetic Antiferromagnets

*Shishen Yan[1,2]*, Tie Zhou[2], Xuejie Xie[2], Yufeng Tian[2], Lihui Bai[2], Yanxue Chen[2] and Qikun Huang[1]*

[1] *Spintronics Institute, University of Jinan, Jinan 250022, China*

[2] *School of Physics, State Key Laboratory of Crystal Materials, Shandong University, Jinan 250100, China*

*corresponding author email: shishenyan@sdu.edu.cn

There are two problems to be overcome for the strong-coupling synthetic antiferromagnetic structure with perpendicular magnetic anisotropy to serve as spin-orbit-torque-based memory and logic devices: first, the Hall signals of the two ferromagnetic layers in the synthetic antiferromagnetic structure are usually weakened by each other, which is not beneficial to the use of Hall voltage to read [1]; secondly, it is usually necessary to apply a large in-plane auxiliary magnetic field to obtain the deterministic magnetization switching caused by the spin orbit torque [2]. In order to solve these problems, we prepared Pt/CoPt/Ru/CoTb synthetic antiferromagnetic heterojunctions. By controlling the long-ranged interlayer exchange coupling and the neighboring ferrimagnetic coupling, we integrate the advantages of ferromagnetic CoPt, ferrimagnetic CoTb, and interlayer antiferromagnetic CoPt/Ru/CoTb spin configurations into a heterojunction. In the CoPt/Ru/$Co_{65}Tb_{35}$ heterojunction with macroscopic interlayer antiferromagnetic coupling, the compensation of magnetization at room temperature and the enhancement of residual anomalous Hall resistance are simultaneously realized [1], as shown in Figure 1. Moreover, it is proved theoretically and experimentally that the field-free magnetization switching can be induced by the spin orbit torque when there is a significant difference in the Dzyaloshinsky Moriya interaction between the lower magnetic layer and the upper magnetic layer of the synthetic antiferromagnet [2]. These findings provide key insights for understanding the role of spin orbit torque and Dzyaloshinsky Moriya interaction in perpendicular magnetization switching of magnetically coupled chiral system, and pave the way to practical applications of synthetic antiferromagnets.

## Acknowledgements

This paper was supported by the Major Basic Research Project of Shandong Province ZR2020ZD28.

## References

[1] X. J. Xie, X. J. Wang, W. Wang, X. N. Zhao, L. H. Bai, Y. X. Chen, Y. F. Tian,* and S.S. Yan*, Adv. Mater. 35 (2023) 2208275.

[2] T. Zhou, X. J. Xie, X. N. Zhao, Y.N. Dong, J. Wang, W. B. Chen, Q. K. Huang, Q. W. Leng, L. H. Bai, Y. X. Chen, S. S. Yan , and Y. F. Tian, Phys. Rev. B 105 (2022) 144434.

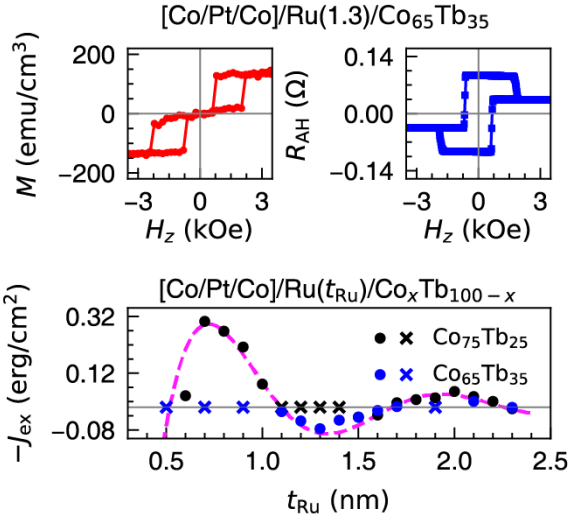



Figure 1. (a) The compensated magnetization, (b) enhanced anomalous Hall resistance in the remanence state, and (c) oscillatory interlayer coupling in synthetic antiferromagnetic heterostructure .

# Broadband terahertz spin-conductance spectroscopy of ultrafast spin transport

*J. Jechumtál[1*], R. Rouzegar[2,3], L. Nádvorník[1], O. Gueckstock[2,3], P. Kubaščík[1] P. W. Brouwer[2], G. Woltersdorf[4], T. Kampfrath[2,3]*

1. *Faculty of Mathematics and Physics, Charles University, 121 16 Prague, Czech Republic*
2. *Department of Physics, Freie Universität Berlin, 14195 Berlin, Germany*
3. *Department of Physical Chemistry, Fritz Haber Institute of the Max Planck Society, 14195 Berlin, Germany*
4. *Institut für Physik, Martin-Luther-Universität Halle, 06120 Halle, Germany*

**email address: jiri.jechumtal@matfyz.cuni.cz*

The understanding the spin transport has been a critical prerequisite for development of current spintronic applications, such as giant-magnetoresistance sensors or spin-torque-based magnetic random-access memories [1]. Nowadays, novel concepts of spintronic devices operating beyond the gigahertz clock rate, based on optical spin-orbit torque switching [2] or antiferromagnetic recording media, are promising sub-picosecond base operation times [3] and require new insights into the ultrafast spin transport. Althought this topic has already attracted considerable attention in past years [4,5,6], the dynamics, propagation and theoretical models of the femtosecond spin transport are still not completely understood.

In this contribution, we report on a new robust method for investigation of out-of-plane ultrafast spin transport based on the principle of the spintronic emission of pulses of terahertz (THz) radiation [7], complemented with a new theoretical description. Using time-domain THz spectroscopy, we reveal spectral dependence of spin current relaxation and speed of propagation in copper, and compare it to the diffusive, super-diffusive and ballistic models. Moreover, our new experimental technique is universal and can be used for investigation of THz spin conductance of any metallic thin layer, which can help future development of new devices based on ultrafast spin transport.

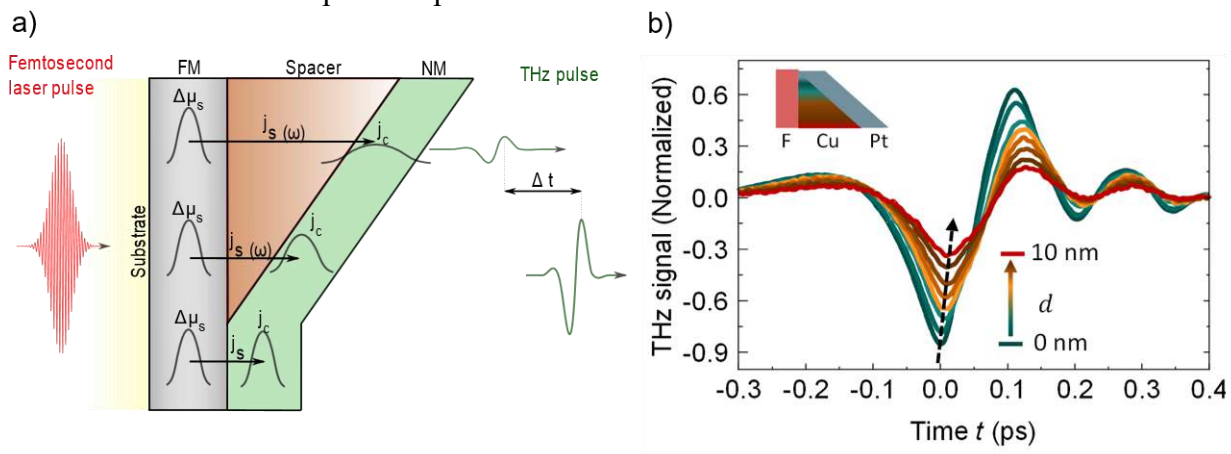



Figure 1. a) Principle of experiment: a ferromagnet (FM) photoexcited by ultrashort laser pusle creates spin voltage $\Delta\mu_s$ at its interface and generates spin current $j_s$ through variable thick copper spacer into nonmagnetical (NM) platinum. Here, $j_s$ is converted via spin-orbit interaction into in-plane charge current $j_c$, acting as a Hertz dipole and emiting a THz pulse. b) Emitted THz waveforms whose shapes and temporal shifts allow us to renocstruct frequency-dependent spin transport properties in the spacer, such as speed and relaxation length.

## Acknowledgements

We would like to acknowledge to Czech Science Foundation (EXPRO Grant No. 19-28375X), GAUK?

## References

[1] M. Cubukcu et al., *IEEE Transactions on Magnetics* **54** (2018) 9300204.
[2] Gyung-Min Choi et al., *Nat. Com.* **11** (2020) 1482.
[3] T. Jungwirth et al., *Nat. Nanotech.* **11** (2016) 231.
[4] M. Battiato et al., *Phys. Rev. Lett* **105** (2010) 027203.
[5] Y. H. Zhu et al., *Phys. Rev. B* **78** (2008) 054429.
[6] Alexey Melnikov et al., *Phys. Rew. B* **106** (2022) 104417.
[7] T. Seifert et al., *Nat Photonics* **10** (2016) 483.

# On-Chip Multilayer Spintronic THz Emitters

*Wolfgang Hoppe[1]*, Amine Wahada[2], Stuart Parkin[2], Georg Woltersdorf[1]*

[1] *Institute of Physics, Martin Luther University Halle-Wittenberg, Von-Danckelmann-Platz 3, 06120 Halle Germany*

[2] *Max Planck Institute for Microstructured Physics, Weinberg 2, 06120 Halle, Germany*

*wolfgang.hoppe@physik.uni-halle.de

Nanometer thin ferromagnet/heavy metal bilayers illuminated by intense short laser pulses have proven to be a realiable source for THz emission [1]. When integrated into a gold waveguide structure, the bilayer can be used as an on-chip source for ultrafast current pulses from the GHz to the THz regime [2]. Stacking severeral bilayers, each separated by a thin MgO interlayer enhances the charge current amplitude, as the MgO suppresses spin-currents in between the individual bilayers [3]. In this way we construct multilayers where all charge currents add up constructively, enhancing the signal up to a factor of three. As one possible application these ultrafast currents could be used to switch the magnetization of an adjacent ferromagnet, similar to previous experiments[4]. Electroooptic sampling is employed to characterize the charge current with sub-ps time resolution.

**Acknowledgements**

Financial support from the German research foundation (DFG) through the collaborative research center CRC/TRR 227 (project ID 328545488, project B02) and Priority Programm SPP 2137 (Skyrmionics) and the European Union through project ASPIN (Grant 766566) is gratefully acknowledged.

**References**

[1] Seifert et al., *Nat. Photonics* **10** (2016) 438-488
[2] W. Hoppe et al., *ACS. Appl. Nano Mater.* **4**,7 (2021) 7454-7460
[3] A. Wahada et al., *ACS Nano Lett.* **22**, 9 (2022) 3539-3544
[4] Y. Yang et al., *Sci. Adv.* **3**, 11 (2017) e1603117

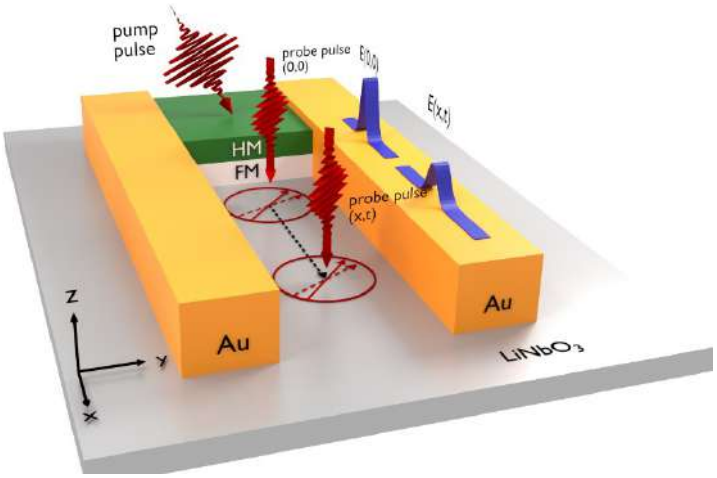



Figure: The electrooptic sampling scheme consisting of a pump-probe experiment to characterize the on-chip THz field **E(x,t)** induced in the ferromagnet/heavy metal bilayer.

# SYMPOSIUM 11.
## SPIN-TRANSFER BASED PHENOMENA AND DEVICES.
## S11. POSTERS

# Potentiation in a Crystallised Heusler-Alloy Giant Magnetoresistive Junction for Neuromorphic Computation

*Zhenyu Zhou [1], William Frost [1], David C. Lloyd [1], Takeshi Seki [2], Takahide Kubota [2], Rafael Ramos [3], Eiji Saitoh [3,4], Koki Takanashi [2,5] and Atsufumi Hirohata [1]*

[1] *School of Physics, Engineering and Technology, University of York, York YO10 5DD, U.K.*
[2] *Institute for Materials Research, Tohoku University, Sendai, 980-8577, Japan*
[3] *WPI Advanced Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan*
[4] *Department of Applied Physics, University of Tokyo, Tokyo 113-8656, Japan*
[5] *Advanced Science Research Center, Japan Atomic Energy Agency, Tokai 319-1195, Japan*
* atsufumi.hirohata@york.ac.uk

As a post-Moore processing unit, a series of new computation architectures has been proposed and demonstrated. Among them, neuromorphic computation to mimic a brain, where a neuron only sends a signal when a spike input to the neuron exceeds a threshold, is advantageous over the others due to the energy efficiency for example **¡Error! No se encuentra el origen de la referencia.**. Recently, current-induced crystallisation of a ferromagnetic Heusler-alloy film used in a giant magnetoresistive (GMR) junction has been demonstrated **¡Error! No se encuentra el origen de la referencia.**, which can be used as data-logging and potentiation functionality into the neuromorphic computation. Here, almost 85% reduction in the crystallisation energy is employed by depositing the films on a (110) plane to promote the layer-by-layer crystallisation process. However, the corresponding GMR ratio is found to be very small (0.04%), which cannot be used for device applications. In this report, we extend our potentiation functionality to the conventional GMR junctions grown on a (001) plane.

A multilayer of Cr (20)/Ag (40)/$Co_2Fe_{0.4}Mn_{0.6}Si$ (5)/$Ag_{0.78}Mg_{0.22}$ (5)/$Co_2Fe_{0.4}Mn_{0.6}Si$ (5)/Ag (2)/Au (5) (thickness in nm) was grown on a MgO(001) substrate at room temperature using an ultrahigh vacuum sputtering system [3]. The seed and Heusler alloy layers were annealed at 650 and 500ºC, respectively, for their crystallisation. The multilayer was patterned into a series of elliptical GMR pillars with long axes between 100 and 800 nm using a combination of photo lithography and Ar-ion milling. The fabricated GMR nanopillars were measured using a non-magnetic probe station with a Keithley 2400 sourcemeter and a Keithley 2182A nanovoltmeter in a similar manner under an application of a magnetic field of ±1 kOe and a sensing current of 50 μA flowing perpendicular to the pillar as reported previously **¡Error! No se encuentra el origen de la referencia.**. The nanopillar was then exposed to current pulses between 100 and 500 μA for 100~300 μs. Note that the sensing current was set to be ~1/10 of the annealing current. Between every pulse introduction, the nanopillar was left for 1 ms to avoid any influence of Joule heating.

In these crystallised GMR pillars, we found the monotonic decrease in their resistance and the corresponding increase in their GMR ratios after the applications of 100 μA for 200 μs for 1,000 times, 200 μA for 100 μs for 1,000 times and 500 μA for 200 μs for 1,000 times. These results confirm the potentiation can be achieved even in a crystallised GMR junction. It should be noted that by applying a current above 600 μA, the resistance increases as the number of sweeps increases due to the Joule heating. Our finding confirms that such potentiation functionality can be implemented in a conventional GMR junction, allowing more realistic neuromorphic computation.

## Acknowledgements

This project was partially supported by EPSRC (EP/V007211/1) and JST CREST (No. JPMJCR17J5).

## References

[1] B. V. Benjamin *et al.*, *Proc. IEEE* **102** (2014) 699.
[2] W. Frost *et al.*, *Sci. Rep.* **11** (2021) 17382.
[3] T. Kubota *et al.*, *Phys. Rev. Mater.* **1** (2017) 044402.
[4] Z. Zhang *et al.*, *J. Magn. Magn. Mater.* **571** (2023) 170575.

# Spin-Charge Current Interconversion in High-Quality Epitaxial Co/Pt Systems

*A. Gudín[1,*], A. Anadón[2], I. Arnay[1], R. Guerrero[1], J. Camarero[1,3], R. Miranda[1,3],S. Petit-Watelot[2], P. Perna[1], J.C. Rojas-Sánchez[2]*

[1]*IMDEA Nanociencia, 28049 Madrid, Spain.*

[2]*Univ. Lorraine, CNRS, Institut Jean Lamour, F-5400 Nancy, France.*

[3]*DFMC Instituto "Nicolás Cabrera" & IFIMAC, Universidad Autónoma de Madrid, 28049 Madrid, Spain.*

*adrian.gudin@imdea.org

The conversion of spin current into charge current and vice versa is one of the main operation for the development of novel low energy consumption spintronic devices. In the last years, an active investigation in materials, interfaces and spin injection schemes aims at increasing the efficiency of the spin-to-charge conversion, which is mainly due to the spin-orbit coupling (SOC).

Pt is a material that exhibits one of the largest intrinsic SOC and hence one of the most studied and employed in magnetic multilayers[1]. We have recently exploited the strong SOC interaction in FM/Pt interfaces for the induction of spin orbit torques in Co [2] as well as the stabilization of chiral spin textures [3]. Nevertheless, there is still an open debate on the efficiency of spin-charge interconversion measured by using different techniques in the same material. The discrepancy found in the experimental results points to a possible anisotropy of such efficiency, so that a renewed interest in the study of the spin charge interconversion in epitaxial Pt along different crystallographic directions and interfaced with different FM layers such as Fe [4,5], CoFeB [6], Co[7], etc [8] has speeded up.

In this context, we have evaluated the in-plane anisotropy of the spin-charge interconversion efficiency in epitaxial [111]Co/Pt deposited on (0001)-oriented $Al_2O_3$ single crystals. Spin pumping – ferromagnetic resonance (SP-FMR) method was used to estimate the spin Hall angle, $\theta_{SH}$, as a function of the thickness of Pt. We find out a slight dependence with Pt-thickness but an appreciable difference (~20%) between the two in-plane crystallographic directions, i.e. $\beta = 0° \parallel Al_2O_3[\bar{1}100]$ and $\beta = 90° \parallel Al_2O_3[11\bar{2}0]$. Whereas, in complementary techniques such as Spin Torque – ferromagnetic resonance (ST-FMR) or Longitudinal Spin Seebeck effect (LSSE) measurement experiments under the same samples, do not yield similar outcomes, being difficult to discern the anisotropic behaviour of our epitaxial [111]Co/Pt samples.

## Acknowledgements

This work has been possible thanks to the funds granted by the IEEE Magnetics Society Support for special projects: HISTRA (High quality spin injectors and transparent interfaces).

## References

[1] J.C. Rojas-Sánchez et al, *Phys. Rev. Letter.* **116** (2016) 096602.
[2] A. Anadón, et al, *ACS Applied Nano Materials*, **4** (2020) 487-492.
[3] F. Ajejas, et al, *Nano letters*, **18**(2018) 5364-5372.
[4] C. Guillemard, et al, *Applied Physics Letters*, **113** (2018) 262404.
[5] S. Keller et al, *New Journal of Physics*, **20** (2018) 053002.
[6] R. Thompson et al, *Physical Review Applied*, **15** (2021) 014055.
[7] Q. Bai et al, *Applied Physics Letters*, **118** (2021) 132403.
[8] P. Li et al, *Physical Review Materials*, **5** (2021) 064404.

# X-ray detection of spin accumulation by XMCD-PEEM: Spin Hall effect in CuBi and beyond

*S. Ruiz-Gómez[1], R. Guerrero[2], M. W. Khaliq[3,4], C. Fernández-González[2,5], J. Prat[3], S. Finizio[6], P. Perna[2], J. Camarero[2,7], L. Pérez[2,5], M.A. Niño[3,*], L. Aballe[3], M. Foerster[3]*

[1]*Max Planck Institute for Chemical Physics of Solids. 01069 Dresden, Germany.*
[2]*Instituto Madrileño de Estudios Avanzados, IMDEA Nanociencia, 28049 Madrid, Spain*
[3] *ALBA Synchrotron Light Facility, CELLS, E-08290 Cerdañola del Vallés, Barcelona, Spain*
[4]*Departamento de Física de la Materia Condensada, Univ. de Barcelona, 08028 Barcelona, Spain*
[5]*Departamento de Física de Materiales, Universidad Complutense de Madrid, 28040 Madrid, Spain*
[6]*Swiss Light Source, Paul Scherrer Institut, CH-5232 Villigen PSI, Switzerland*
[7]*Universidad Autónoma de Madrid (UAM), Campus de Cantoblanco. Madrid 28049, Spain*
*mnino@cells.es

X-ray magnetic circular dichroism (XMCD) is a synchrotron based use reference tool for magnetic measurements of small or diluted magnetic signals with the advantage to be element sensitive. Here we show the combination of XMCD with Photoemission Electron Microscopy (XMCD-PEEM) for the direct observation in different materials of the spin Hall effect (SHE), a spin-charge conversion mechanisms widely investigated for spintronics devices. Its measurement has been mostly investigated with electrical detection schemes involving interfaces with another magnetic material and thus, a combination of the properties of both materials as well as the interface are measured. Optical detection techniques have been used for semiconductors but are challenging for metallic systems due to their considerably shorter spin diffusion lengths, and only recently optical measurements for Pt and W have been reported [1].
We have performed laterally resolved interface free x-ray spectro-microscopy measurements at the Cu $L_{3,2}$ absorption edges, while applying electrical current to the sample, of a single layer of Bi-doped Cu ($Cu_{95}Bi_5$), a material in which giant SHE has been already reported [2,3]. The sign of spin accumulation depends on the direction of the current (Figure 1) and the amplitude of the X-ray magnetic circular dichroism (XMCD) signal scales with the current density and it has opposite sign when measuring at the $L_2$ or $L_3$ absorption edges, as expected for SHE. These results constitute a proof of concept for the direct, interface free and element-selective measurement of the SHE in a metallic material by means of X-ray spectro-microscopy [4]. We will also discuss recent results for further materials applying the same technique.

## References

[1] C. Stamm et al. *Phys. Rev. Lett.* **119**, 087203 (2017).
[2] Y. Niimi et al. *Phys. Rev. Lett*. **109**, 156602 (2012).
[3] S. Ruiz-Gómez et al. APL Materials 6, 101107 (2018).
[4] S. Ruiz Gómez et al. *Phys. Rev. X* **12**, 031032 (2022).

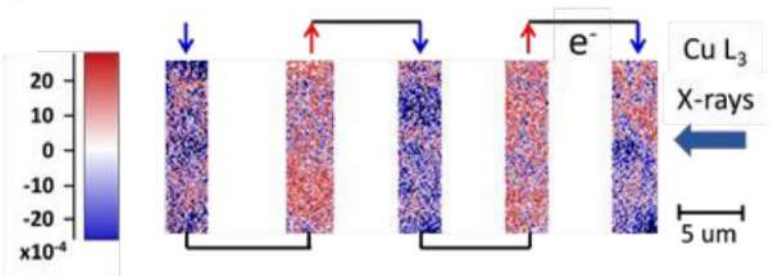



Figure 1. Visualization of the spin accumulation in a $Cu_{95}Bi_5$ electrode showing the inverted XMCD signal when inverting the current direction.

# Spin-orbit Ferroelectric RAM, concept and study of spin-dependent transport via Finite Element Method simulations

*Paolo Sgarro[1]*, Theo Frottier[1], Aurélie Kandazoglou[1], Salvatore Teresi[1], Maxime Culot[1], Maxen Cosset-Chéneau[1], Lorena Anghel[1], Alain Marty[1], Laurent Vila[1], Guillaume Prenat[1] and Jean-Philippe Attané[1]*

[1] *Spintec, IRIG, DEFI, Univ. Grenoble Alpes, CEA, CNRS, F-38000 Grenoble, France*

*paolo.sgarro@cea.fr

Spin-related phenomena are attracting increasing interest from information technology, due to the possibility to bring together the advantages of non-volatile devices and low energy state switching. Physical effects like spin-orbit torque and spin-transfer torque have found application for fast non-volatile memories, and recently, the MagnetoElectric Spin-Orbit (MESO) device, proposed by INTEL, is expected to bring a dramatic improvement with respect to CMOS in terms of power consumption and logic density [1].

In a previous work [2], a novel device featuring the interplay of ferromagnetism and ferroelectricity has been proposed; in which the data is stored in a ferroelectric layer, and can be read by injecting a spin-polarized current and exploiting the ferroelectric control of spin-to-charge interconversion. This phenomenon can be observed in several materials, ranging from oxide-based two dimensional electron gases [2], to Ferroelectric Rashba semiconductors [3]. We expect this device to be interesting for applications in logic, memories and non-conventional computing.

In this work, we want to propose a novel technology called Spin-Orbit Ferroelectric RAM (sofRAM), in which the pheonomenon described above is exploited for a non volatile memory with a lower writing energy with respect to magnetic memories, and with a non destructive reading of the ferroelectric state.

We show the development of the compact model of the device, which is later tested by means of Finite Element simulations for spin-dependent transport. Later, these simulations are also used to optimize the performances of the device, with a particular focus on the output signal, and materials choice is also discussed.

In the last part, we propose a few architectures, with reading and writing strategies, in which the sofRAM can be implemented for a large-scale memory array.

## Acknowledgements

This project has received funding from the European Union's Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement Nº 955671.

## References

[1] Manipatruni, Sasikanth, et al. *Nature* 565.7737 (2019) 35-42.
[2] Noël, Paul, et al. *Nature* 580.7804 (2020): 483-486.
[3] Varotto, Sara, et al *Nature Electronics* 4.10 (2021): 740-747.

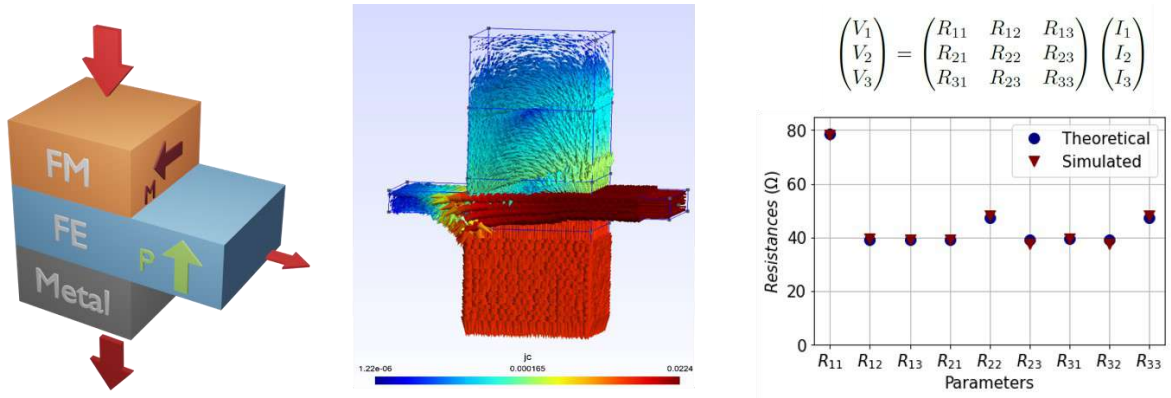



Figure : (left) Sketch of the sofRAM, (middle) a representation of the current lines computed via FEM, and (right) the comparison of the compact model parameters computed analytically with the results of simulations.

# Spin transport and magnetic dynamics in ultra low damping epitaxial $Co_{100-x}Fe_x$/Ta bilayers

*D. Velázquez Rodriguez[1,2], J. E. Gómez[1,2], T. E. Torres[1], M. H. Aguirre[3,4], P. Costanzo[5], J. Milano[1], L. Avilés-Félix[1,2,*], A. Butera[1,2]*

[1] *Instituto de Nanociencia y Nanotecnología (CNEA-CONICET), Nodo Bariloche, Av. Bustillo 9500, (8400) Bariloche (RN), Argentina*

[2] *Laboratorio Resonancias Magnéticas, Centro Atómico Bariloche, Av. Bustillo 9500, (8400) Bariloche (RN), Argentina*

[3]*Laboratorio de Microscopías Avanzadas, Universidad de Zaragoza, E-50018, Zaragoza, Spain*

[4]*Instituto de Nanociencia y Materiales de Aragon, INMA-CSIC-Univ de Zaragoza, Zaragoza, Spain*

[5]*Instituto Balseiro, Universidad Nacional de Cuyo, Comisión Nacional de Energía Atómica, Consejo Nacional de Investigaciones Científicas y Técnicas (CONICET), Av. Bustillo 9500, Bariloche, Rio Negro 8400, Argentina.*

*luis.aviles@ib.edu.ar

The manipulation of the electron charge and the spin for the development of more efficient spintronic devices has been a topic of increasing interest in recent years. In particular, systems based on materials with very low damping, a parameter that determines the speed and energy consumption during the operation of modern electronic devices, are excellent candidates for spin current injection. Recently, it has been reported that the $Co_{25}Fe_{75}$ alloy exhibits low magnetic damping, due to the features of the band structure in $Co_{100-x}Fe_x$ alloys. This work discusses the magnetic characterization and spin transport in $Co_{100-x}Fe_x$ (20 nm)/Ta (10 nm) bilayer systems, with x = 65, 70, 75, 80, 85, grown on MgO (100) single crystal substrates. Characterization by Kerr magnetometry, transmission electron microscopy and ferromagnetic resonance allowed to determine that the $Co_{100-x}Fe_x$ films grow epitaxially with a cubic structure rotated 45° with respect to the (100) direction of the MgO substrate plane, determining the easy and hard magnetization axes. On the other hand, spin transport characterization using spin pumping and inverse spin Hall effect allowed the detection of induced voltages associated to the conversion of spin current to charge current, of the order of 100 μV ($Fe_{80}Co_{20}$), demonstrating efficient conversion of spin current to charge current in Ta layers.

## References

[1] D. Velázquez Rodriguez et al. – Manuscript in preparation (2023)
[2] D. Velázquez Rodriguez et al. J. Phys. D: Appl. Phys 54, 325002 (2021)
[3] D. Velázquez Rodriguez et al. J. Magn. Magn. Mat. 504, 166692 (2020)

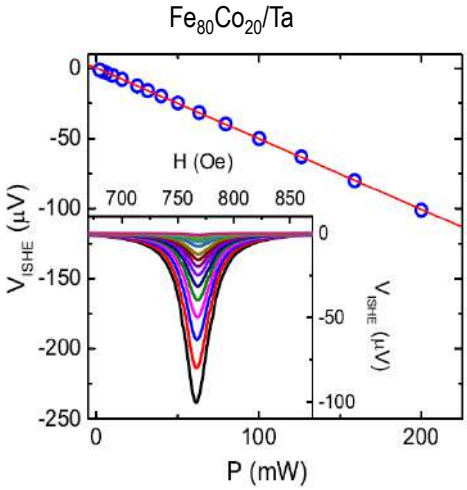



Figure: Linear dependence of ISHE signals vs. the microwave power in spin pumping-inverse spin Hall effect measurements for $Co_{20}Fe_{80}$/Ta bilayers.

# Interplay between Oxygen Vacancies and Tunneling Spin-Transfer Torque

L. M. Kandpal[1,2*], B. Taudul[1], A. Kumar[3], F. Schleicher[4], E. Monteblanco[4], D. Lacour[4], P. Gupta[3], S. Boukari[1], W. Weber[1], M. Alouani[1], M. Hehn[4], F. Montaigne[4], J. D. Costa[5], T. Böhnert[5], R. Ferreira[5], P. Freitas[5], L. Thomas[6] , P. K. Muduli[3], M. Bowen[1].

[1]Institut de Physique et Chimie des Matériaux de Strasbourg (IPCMS), UMR 7504 CNRS, Université de Strasbourg, 23 Rue du Lœss, BP 43, 67034 Strasbourg, France.

[2]*Université Grenoble Alpes, CNRS, CEA, Grenoble INP, SPINTEC, 38000 Grenoble, France.*

[3]*Department of Physics, Indian Institute of Technology, Delhi, New Delhi 110016, India.*

[4]Institut Jean Lamour, UMR 7198 CNRS, Université de Lorraine, BP 70239, 54506 Vandœuvre les Nancy, France.

[5]*International Iberian Nanotechnology Laboratory (INL), Av. Mestre José Veiga s/n 4715-330, Braga, Portugal.*

[6]TDK-Headway Technologies Inc., Milpitas, California, USA.

* lalit-mohan.kandpal@cea.fr

Flowing an electrical current that is both of high areal density and large spin polarization across a magnetic tunnel junction (MTJ) can, through spin transfer torque (STT) [1], alter the relative magnetic orientation of the MTJ's ferromagnetic electrodes. This effect has enabled key next-generation MTJ applications, from memories [2] to artificial synapses [3]. Using measurements across MTJ nanopillars and ab-initio theory, we demonstrate that, contrary to conventional wisdom established over nearly 20 years, oxygen vacancies in the MgO barrier play a key enabling spintronic role for these technologies [4,5]. We observe that the orientation of electrode magnetizations determines whether charge transport across the MTJ is metallic or semiconducting, depending on whether localized states at and near the MTJ Fermi level are involved in transport. Ab-initio theory and ST-FMR measurements suggests that these states within the nanotransport path might be due to several oxygen vacancies forming a chain (F-chain) across the MgO barrier [5]. Taken together, these results indicate that the localized states promoted by oxygen vacancies constitute the linchpin for combined high TMR and spin transfer torque in MgO-based spintronics. This has implications on how small an MTJ can be laterally[6,7]. Mastering oxygen vacancies in industrial MTJs could enable their dual-use not only as memory devices, but also as energy harvesters at ambient heat[8,9].

**Acknowledgement:**

L. M. Kandpal sincerely thanks; 'Indo-French Center for the Promotion of Advanced Research (IFCPAR)' for the doctoral scholarship; Dr. Olivier Fruchart for the financial support to present this research work in JEMS 2023; Dr. Kevin Garello, Dr. Ricardo Sousa, and Dr. Bernard Dieny, for the scientific discussion on the topic during InMRAM school 2022; and Dr. K. Katcko for the technical assistance in the transport measurements.

**References:**

[1] J. C. Slonczewski, *Current-Driven Excitation of Magnetic Multilayers*, Journal of Magnetism and Magnetic Materials **159**, L1 (1996).

[2] B. Dieny et al., *Opportunities and Challenges for Spintronics in the Microelectronics Industry*, Nature Electronics **3**, 8 (2020).

[3] J. Torrejon et al., *Neuromorphic Computing with Nanoscale Spintronic Oscillators*, Nature **547**, 428 (2017).

[4] U. Halisdemir et al., *Oxygen-Vacancy Driven Tunnelling Spintronics across MgO*, in *Spintronics IX*, Vol. 9931 (International Society for Optics and Photonics, 2016), p. 99310H.

[5] L. M. Kandpal, Localized States in the Spacer Layer of Spintronic Nanodevices, These de doctorat, Strasbourg, 2021.

[6] R. C. Sousa et al., *Efficient Spin Transfert Torque Switching of Perpendicular Magnetic Tunnel Junctions and Scaling Outlook (Conference Presentation)*, in *Spintronics XV*, Vol. PC12205 (SPIE, 2022), p. PC122050D.

[7] T. P. Almeida, S. Lequeux, A. Palomino, R. C. Sousa, O. Fruchart, I.-L. Prejbeanu, B. Dieny, A. Masseboeuf, and D. Cooper, *Quantitative Visualization of Thermally Enhanced Perpendicular Shape Anisotropy STT-MRAM Nanopillars*, Nano Lett. **22**, 4000 (2022).

[8] K. Katcko et al., *Spin-Driven Electrical Power Generation at Room Temperature*, Communications Physics **2**, 116 (2019).

[9] B. Chowrira et al., *Quantum Advantage in A Molecular Spintronic Engine That Harvests Thermal Fluctuation Energy*, Advanced Materials **34**, 2206688 (2022).

# Atomic Resolution Studies of Low-dimensional Bi-doped Cu Nanowires for Spintronic Applications

*Alejandra Guedeja-Marrón[1,2]*, Matilde Saura-Múzquiz[1], Sandra Ruiz-Gómez[3], Claudia Fernández-González[3], Inés García-Manuz[4], Henrik Lyder Andersen[1], Lucas Pérez[1,4], and María Varela[1,2]*

[1] *Dept. Fisica de Materiales, Universidad Complutense de Madrid, Spain*
[2] *Instituto Pluridisciplinar, Universidad Complutense de Madrid, Spain*
[3] *Max Planck Institute for Chemical Physics of Solids, Dresden, Germany*
[4] *Instituto Madrileño de Estudios Avanzados – IMDEA Nanociencia, Madrid, Spain*
*aguedeja@ucm.es

The spin Hall effect (SHE) is a spin-charge conversion phenomenon that plays a key role in spintronics,[1] so the search for materials that exhibit giant SHE has grown significantly within the last decades.[2] CuBi alloys with Bi contents around 0.5% exhibit a large spin Hall angle (SHA).[3] Recently, SHE in $Cu_{95}Bi_5$ films has been directly detected by X-ray spectroscopy ,[4] confirming this material as a good candidate for integration in spintronic devices. Therefore, achieving CuBi structures with tunable composition and structure [5] can provide an ideal model-system for exploring the origin of extrinsic SHE and optimizing the spin-charge conversion. Furthermore, it facilitates the study of the thermal stability of the material, which determines the threshold parameters for transport and resistivity measurements.

In this work, we present a detailed structural characterization of Bi-doped Cu nanowires (NWs) with different doping levels and degrees of crystallinity, grown using template-assisted electrochemical deposition. Performing *in-situ* heating experiments with atomic resolution scanning transmission electron microscopy (STEM) combined with electron energy-loss spectroscopy (EELS) and high-resolution synchrotron powder X-ray diffraction (PXRD), we are able to describe in detail the atomic structure of the NWs and its evolution with temperature. These results pave the way towards the realization of spin-dependent transport measurements in CuBi NWs.

## Acknowledgements

Electron microscopy observations were carried out at the Centro Nacional de Microscopía Electrónica at Universidad Complutense de Madrid. PXRD experiments were performed at the MSPD beamline, ALBA, Spain, and ID22 beamline, ESRF, France.

## References

[1] J. Sinova et al., *Rev. Mod. Phys*. **87** (2015) 1213.
[2] T. Jungwirth et al., *Nature*. **11** (2012) 5.
[3] Y. Niimi et al., *Phys. Rev. Lett*. **109** (2012) 15.
[4] S. Ruiz-Gomez et al., *Phys. Rev. X*. **12** (2022) 3.
[5] S. Ruiz-Gomez et al., *J. Magn. Magn. Mater*. **545** (2022) 168645.

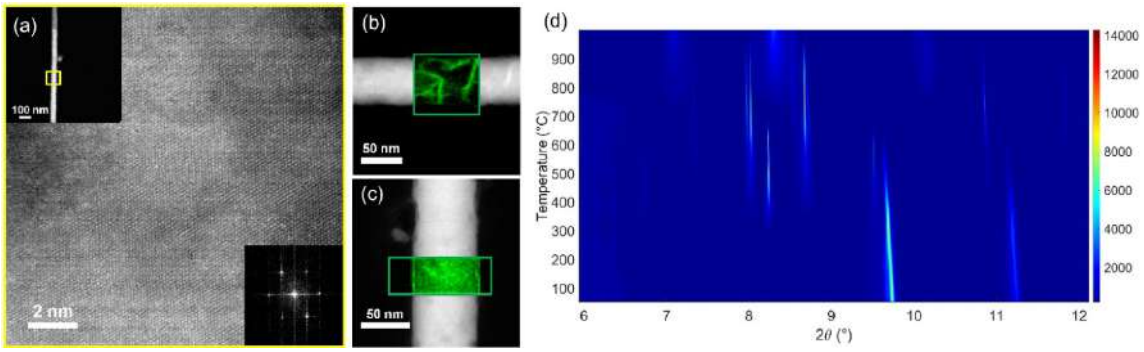



Figure 1: (a) High-angle annular dark field STEM atomic resolution images corresponding to a single-crystal CuBi nanowire. Bottom right (a) image displaying the fast Fourier transform [111] zone axis. EELS maps depicting the Bi $M_{4,5}$ edge integrated signal in a (b) polycrystal and (c) single-crystal NW. (d) PXRD heating 2D map showing the material conversion at around 300ºC.

# Kinetic Rashba-Edelstein effect and interface spin-Hall effect at the metal-insulator border

*A.V. Shumilin*[1]*, *V.V. Kabanov*[1]

[1] *Jozef Stefan Institute, Jamova cesta 39, SI-1000 Ljubljana, Slovenia*

* andrei.shumilin@ijs.si

Most often the spin torque induced by a heavy metal layer to the layer of ferromagnetic metal in spintronic devices is described either by spin-Hall effect related to the bulk of heavy metal or by Rashba-Edelstein effect that leads to the current-induced spin polarization in several monolayers near the interface. However, recent experiments [1] show that modification of the second surface of the heavy metal can significantly change the spin torque.

We theoretically predict that when the surface of heavy metal and insulator contains impurities, two novel mechanisms of spin torque emerge. The quantum interference of different impurity scattering paths results in spin-dependent skew scattering (fig a). The skew scattering itself leads to the kinetic Rashba-Edelstein effect: the spin polarization that spans over a length scale comparable to the mean free path into the bulk of the heavy metal (fig. b). When the spin relaxation at the boundary is taken into account, it becomes asymmetric due to the skew scattering. This asymmetry leads to the interface spin-Hall effect, i.e., to the charge-spin current conversion at the heavy metal–insulator boundary (fig c). In contrast to the ordinary spin-Hall effect, it exists in the Born approximation.

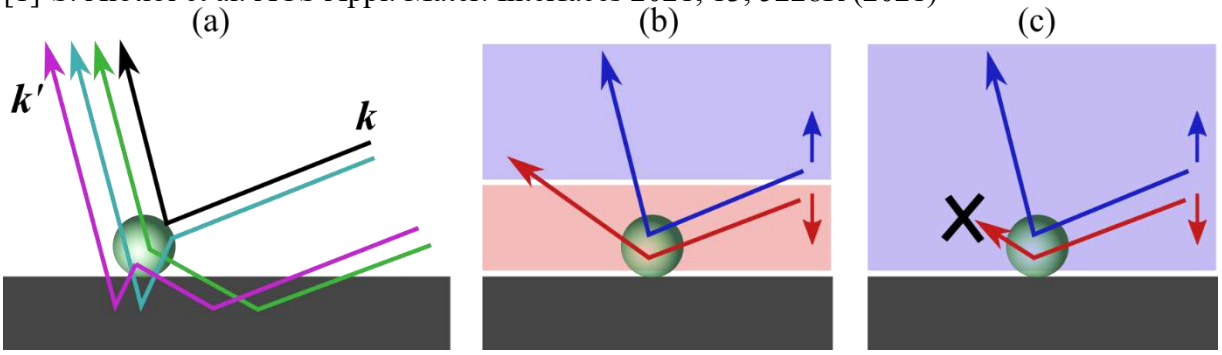
Both phenomena are sensitive to the properties of the interface itself and introduced impurities allowing the control of spin torque with surface engineering.

**Acknowledgements**

We acknowledge the support of the EC projects INTERFAST (H2020-FET-OPEN-965046).

**References**

[1] S. Alotibi et al. ACS Appl. Mater. Interfaces 2021, 13, 5228R (2021)




(a) The interference of the four reflection paths that leads to skew scattering. (b) Kinetic Rashba-Edelstein effect. (c) Interface spin-Hall effect.

# Ultra-Sensitive Low-Power Skyrmion-Based Spintronic Diode

*Mario Carpentieri[1]*, Davi R. Rodrigues[1], Riccardo Tomasello[1], Giulio Siracusano[2], Giovanni Finocchio[3]*

[1] *Department of Electrical and Information Engineering, Politecnico of Bari, I-70125 Bari, Italy*

[2] *Department of Electric, Electronic and Computer Engineering, University of Catania, I-95125 Catania, Italy*

[3] *Department of Mathematical and Computer Sciences, Physical Sciences and Earth Sciences, University of Messina, I-98166, Messina, Italy*

*mario.carpentieri@poliba.it

Spintronic diodes (SDs) are high performance detectors with applications in Internet-of-Things, energy harvesting, and artificial intelligence that exploit resonant magnetic excitations. We propose a passive spintronic diode based on the excitation of single skyrmion by voltage-controlled anisotropy (VCMA) and Dzyaloshinskii-Moriya interaction (VDMI), see Fig. 1. The use of VCMA or VDMI allows to increase the sensitivity while decreasing the required applied electric current and associated Joule losses. We demonstrate that with realistic physical parameters and geometry the sensitivity (rectified voltage over input microwave power) can be larger than 10 kV/W, an order order of magnitude better than diodes employing a uniform ferromagnetic state. The skyrmion is can be easily excited beyond the linear regime, showing a frequency dependence of the amplitude. Moreover, the device shows remarkable scalability, since skyrmions with a smaller radius produced higher sensitivities. These results combined to current efforts to improve anisotropy and DMI control paves the way for ultra-sensitive and low power skyrmionics devices.

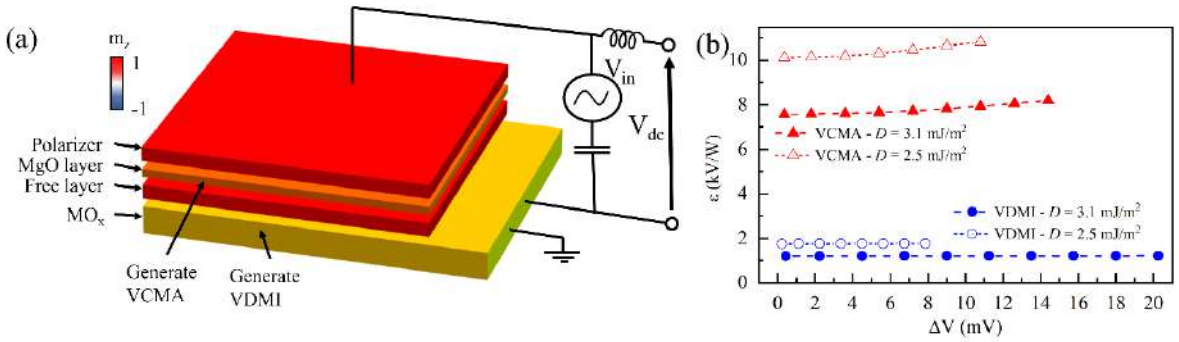



Figure 1: Properties of the proposed device. (a) Multi-stack structure of the device with the indication of the different layers of the MTJ, a polarizer, a MgO layer, a free layer, and a metal oxide ($MO_x$) layer. The MgO and $MO_x$ layers can independently generate the VCMA and VDMI respectively. A schematics of the electrical circuit to excite the skyrmion dynamics and detect the output dc voltage is also included. (b) Sensitivity (ε) of the device as a function of the applied voltage for different values of DMI and considering excitation by VCMA or VDMI. Notice that smaller VDMI, corresponding to a smaller skyrmion, yields a higher sensitivity.

## Acknowledgements

This work was supported under the project number 101070287 — SWAN-on-chip — HORIZON-CL4-2021-DIGITAL-EMERGING-01, the project PRIN 2020LWPKH7 “The Italian factory of micromagnetic modelling and spintronics”, and D.M. 10/08/2021 n. 1062 (PON Ricerca e Innovazione) funded by the Italian Ministry of University and Research, and by PETASPIN association (www.petaspin.com).

# Phonon Spin Valves and Phonon-mediated Interlayer Exchange Coupling

*Łukasz Karwacki[1,2]*, Rembert A. Duine[1,3]*

[1] *Institute for Theoretical Physics, Utrecht University, 3584CC Utrecht, The Netherlands*
[2] *Institute of Molecular Physics, Polish Academy of Sciences, ul. M. Smoluchowskiego 17, 60-179 Poznań, Poland*
[3] *Department of Applied Physics, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, The Netherlands*
*l.karwacki@uu.nl

In order to achieve components for future information technologies with low-energy consumption it is necessary to abandon electric currents and utilize spin currents and, e.g., magnons as information carriers. Recent rediscovery of the concept of a phonon spin [1-4] and, more generally, phonon angular momenta [5], might prove to be either another alternative or be used in fully insulating junctions and devices for information processing working in tandem with magnons, since it has been shown that, due to magnetoelastic coupling, the phonons can mediate information between magnetic layers [6].

By using Landau-Lifshitz-Gilbert equation for magnetization dynamics and Euler-Lagrange equations for phonon dynamics we show the possibility to fine-tune phonon spin and phonon spin current flowing between the ferromagnetic layers with generalized directions of magnetization. Moreover we show the emergence of an effective interlayer exchange coupling between the ferromagnets mediated by phonons in the central non-magnetic insulator.

## Acknowledgements

This work is part of the Fluid Spintronics research programme with project number 182.069, which is financed by the Dutch Research Council (NWO).

## References

[1] S. V. Vonsovskii, M. S. Svirskii, *Sov. Phys. – Solid State* **3**, (1962), 1568.
[2] L. Zhang, Q. Niu, *Phys. Rev. Lett.* **112**, (2014) 085503.
[3] A. Rückriegel, S. Streib, G. E. W. Bauer , R. A. Duine, *Phys. Rev. B* **101**, (2020) 104402.
[4] A. Rückriegel, R. A. Duine, *Phys. Rev. Lett.* **124**, (2020) 117201.
[5] D. M. Juraschek, N. A. Spaldin, *Phys. Rev. Mat.* **3**, (2019) 064405.
[6] K. An et al., *Phys. Rev. B* **101**, (2020) 060407(R).

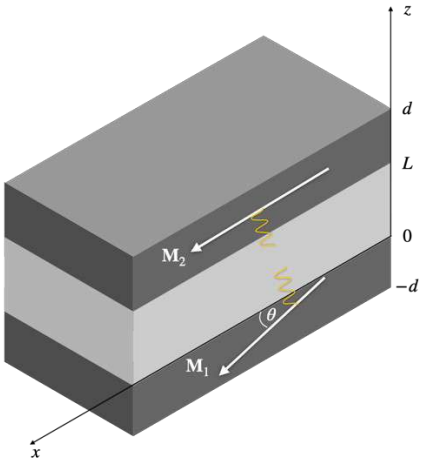



Figure: Schematic representation of the spin valve system consisting of a central non-magnetic insulator of thickness $L$ and two ferromagnets of thickness $d$ and magnetizations $\mathbf{M}_1$ and $\mathbf{M}_2$, respectively.

# Comparison of antidamping spin-orbit torques estimated by harmonic Hall and Kerr effect measurements

*Emir Karadža[1]*, Paul Noël[1], Richard Schlitz[1], Pol Welter[2], Charles-Henri Lambert[1], Luca Nessi[1,3], Federico Binda[1], Christian L. Degen[2], and Pietro Gambardella[1]*

[1] *Department of Materials, ETH Zürich, CH-8093 Zürich, Switzerland*
[2] *Department of Physics, ETH Zürich, CH-8093 Zürich, Switzerland*
[3] *Dipartimento di Fisica, Politecnico di Milano, Via G. Colombo 81, 20133 Milano, Italy*
*emir.karadza@mat.ethz.ch

Spin-orbit torques (SOTs) are current-induced magnetic torques that permit the electrical manipulation of the magnetization in thin ferromagnet/heavy metal (FM/HM) heterostructures and have application in low-power magnetic memory and logic devices [1,2]. Since the first SOT studies, the accurate estimation of these torques appear to be a cornerstone to understand them and further improve their efficiency. Multiple techniques have been developed to evaluate the SOTs, including magnetotransport, magnetooptical and resonance measurements. Nevertheless, there is no clear consensus on the exact torque efficiencies with strong discrepancies from group to group and when using different measurement techniques even for the most studied systems such as Pt [3].

In this presentation we will compare the antidamping like torque in several FM/HM bilayers obtained from the widely used harmonic harmonic Hall (HH) analysis technique [4] and the magneto-optical Kerr effect (MOKE) [5] in the same devices. We find that the antidamping like torque estimated by MOKE and HH measurements can differ by as much as 100%, and this discrepancy depends on both the ferromagnet and the heavy metal [6]. We identify the source of these discrepancies and provide a self-consistency check of the MOKE technique by calibrating the magneto-optic measurement of the Oersted field with nitrogen-vacancy scanning magnetometry. Our results show that the HH and MOKE results coincide for a broad variety of material systems if all the current-induced Hall voltages are properly accounted for.

[1] Manchon, A. *et al.* Rev. Mod. Phys. 91, 035004 (2019).
[2] Krizakova, V. et al., J. Mag. Magn. Mater. 562, 169692 (2022).
[3] Zhu, L., Ralph, D. C. & Buhrman, R. A. Applied Physics Reviews 8, 031308 (2021).
[4] Avci, C. O. *et al.* Phys. Rev. B 90, 224427 (2014).
[5] Fan, X. *et al.* Nat Commun 5, 3042 (2014).
[6] Noël, P., Karadža, E. *et al.* (in preparation)

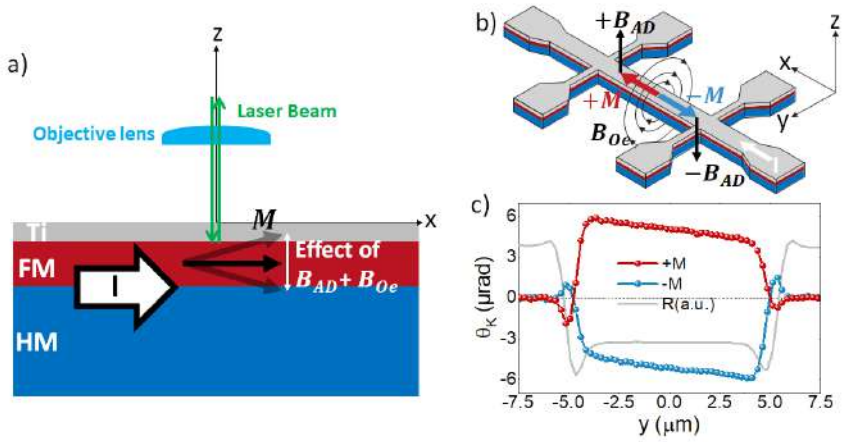



Figure: a) Schematic drawing of the scanning MOKE microscopy setup, the change of the direction of the magnetization due to torque affects the MOKE signal. b) The effect of the AD torque depends on the magnetization direction while the Oersted field is constant. c) MOKE detection of SOTs in Pt (5 nm)/CFB (2.5 nm) at 3.6 mA for positive and negative external magnetic field. The gray line shows the optical reflectance of the sample.

# Evaluating Spintronic Ising Machines for solving Max-Cut

*Luciano Mazza[1], Andrea Grimaldi[2], Eleonora Raimondo[2], Pietro Tullo[1], Davi Rodrigues[1], Kerem Camsari[3], Vincenza Crupi[2], Mario Carpentieri[1], Vito Puliafito[1], Giovanni Finocchio[2]*

*[1]Department of Electrical and Information Engineering, Politecnico di Bari, 70126, Bari, Italy*

*[2]Department of Mathematical and Computer Sciences, Physical Sciences and Earth Sciences, University of Messina, 98166, Messina, Italy*

*[3]Department of Electrical and Computer Engineering, University of California Santa Barbara, 93106, Santa Barbara, CA, USA*

*luciano.mazza@poliba.it

Heuristic paradigms like Ising machines are recently gaining interest in the academic and industrial world for the realization of highly scalable solutions for combinatorial optimization problems (COPs).
Ising machines map a COP with the Hamiltonian energy function of physical system in such way that the natural evolution to a ground low-energy state of the system leads to a configuration corresponding to a solution of the starting problem. We have analyzed how magnetic tunnel junctions (MTJs) can be used for realizing a coherent and probabilistic Ising machine for the solution of the Max-Cut problem.
The coherent Ising machine (CIM) is realized exploiting the coupling mechanisms of oscillators governed by the Hamiltonian energy and the binarization, necessary for the readability of the solution, is realized with a second harmonic signal, as shown in [1].
We implemented the CIM with the state-of-the-art Kuramoto model, and with a mathematical model that well represents the behavior of spintronic oscillators, observing comparable performances for randomly generated cubic problems; the latter model is characterized by a higher computational complexity which limited the size of the explored problems, but underlines the benefits of the nonlinearities of spintronic devices in such implementation.
The probabilistic implementation [2] relies on the use of random binary numbers efficiently implementable aligning the magnetization of the free layer of an MTJ with an hard axis and letting the system relax into one of the two possible states of the easy axis, characterized by different resistances.
Both systems provide excellent results for the solution of Max-Cut problems, and our results underline the effectiveness of a future implementation for hybrid CMOS-MTJ chips.

## References

[1] T. Wang, L. Wu, P. Nobel, and J. Roychowdhury, *Nat. Comput.* **20** (2021) 287.
[2] K. Y. Camsari, R. Faria, B. M. Sutton, and S. Datta, *Phys. Rev. X* **7** (2017) 1.

# SYMPOSIUM 12.
## ANTIFERROMAGNETIC SPINTRONICS.
## S12. INVITED ORAL PRESENTATIONS

# Optical read-out and manipulation of Néel vector in a metallic antiferromagnet

*V. Grigorev[1,2] M. Filianina[1,2,3], Y. Lytvynenko[1,4], S. Sobolev[1], A.R. Pokharel[1], A.P. Lanz[1], A. Sapozhnik[5], A. Kleibert[6], S. Bodnar[7], P. Grigorev[8], Y. Skourski[9], M. Kläui[1,2], H-J. Elmers[1,2], M. Jourdan[1], and J. Demsar[1,2,*]*

[1] *Institute of Physics, Johannes Gutenberg University, 55128 Mainz, Germany*

[2] *Graduate School of Excellence Materials Science in Mainz, 55128 Mainz, Germany*

[3] *Department of Physics, AlbaNova University Center, Stockholm University, 10691 Stockholm, Sweden*

[4] *Institute of Magnetism of the NAS and MES of Ukraine, 03142 Kyiv, Ukraine*

[5] *Institute of Physics, École Polytechnique Fédérale de Lausanne, 1015 Lausanne, Switzerland*

[6] *Paul Scherrer Institute, Swiss Light Source, 5232 Villigen, Switzerland*

[7] *Walter Schottky Institute and Physics Dept., Technical University Munich, 85748 Garching, Germany*

[8] *Aix-Marseille Université, CNRS, CINaM UMR 7325, 13288 Marseille, France*

[9] *Dresden High Magnetic Field Laboratory (HLD-EMFL), Helmholtz-Zentrum Dresden-Rossendorf, 01328 Dresden, Germany*

*demsar@uni-mainz.de

The absence of stray fields, their robustness against external magnetic fields, and the inherently fast dynamics make antiferromagnets promising candidates for active elements in spintronic devices [1]. Especially the metallic collinear antiferromagnets CuMnAs and $Mn_2Au$, which enable current-driven bulk (Néel) spin-orbit torques [2], have been in the research focus recently. The lack of sensitivity to external magnetic fields, however, poses challenges in terms of manipulation and read-out of the staggered magnetization (Néel vector). Thus, while ultrafast optical control of magnetization in several ferro- and ferri-magnets has been demonstrated in recent years [3-5], such approaches in antiferromagnets are scarse. Here, we address the optical read-out in a metallic collinear antiferromagnet $Mn_2Au$ [6] and present an approach of its optical manipulation by combining tensile strain and excitation with femtosecond optical pulses [7]. The dependence of switching on the laser fluence and strain suggests the Néel vector alignment is a result of optically-triggered depinning of 90° domain walls and their sliding in the direction governed by the magneto-elastic coupling. The switched, metastable, state is stable at room temperature and insensitive to magnetic fields. Thus, this approach may provide ways to realize robust high-density memory device with switching timescales in the picosecond range.

**Acknowledgements**

This work was funded by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) Grant No. TRR 173-268565370 (project A05, with contribution from A01) and TRR 288−422213477 (project B08) and received support from Horizon 2020 Framework Program of the European Commission under grant agreement No. 863155 (S-NEBULA). We acknowledge the Paul Scherrer Institute, Villigen, Switzerland for the beamtime allocation under proposal 20200977 at the SIM beamline of the SLS.

**References**

[1] P. Nemec, M. Fiebig, T. Kampfrath, A.V. Kimel, Nat. Phys. 14, 229–241 (2018).
[2] J. Železny, et al., Phys. Rev. Lett. 113, 157201 (2014).
[3] A. Kimel, et al., Nature 435, 655–657 (2005).
[4] A. R. Khorsand, et al., Phys. Rev. Lett. 108, 127205 (2012).
[5] C.-H. Lambert, et al., Science 345, 1337–1340 (2014).
[6] V. Grigorev, et al., Phys. Rev. Appl. 16, 014037 (2021).
[7] V. Grigorev, et al., ACS Nano 16, 20589–20597 (2022).

# Magnonics In Collinear and Canted Antiferromagnets: From Spin-Pumping To Magnon-Photon Coupling

I.Boventer[1*], H.T. Simensen[2], B.Brekke[2], M.Weides[3], A. Anane[1], M.Kläui[2,4,5], A. Braatas[2], R. Lebrun[1]

[1] Unité Mixte de Physique CNRS/Thales, Université Paris Saclay, Palaiseau 91767 France
[2] Center for Quantum Spintronics, Department of Physics, Norwegian University of Science and Technology, Trondheim Norway
[3] James Watt School of Engineering, Electronics & Nanoscale Engineering Division, University of Glasgow, Glasgow G12 8QQ, United Kingdom
[4] Institut für Physik, Johannes-Gutenberg Universität Mainz, D-55099, Mainz, Germany
[5] Graduate School of Excellence Materials Science in Mainz (Mainz), Staudinger Weg 9, D-55128 Mainz, Germany
*isabella.boventer@cnrs-thales.fr

Recent years have been the frame of a renewal of activity of magnonic and spintronic research on antiferromagnetic (AFM) materials due to some of their intrinsic and potentially advantageous properties [1]. Their vanishing stray fields, higher resonance frequencies, and the possibility to control them by spin currents renders them scalable, fast and tunable for future ICT devices. However, the implementation of magnonic AFM devices requires a profound understanding of the spin dynamics of AFMs and of their ability to couple to other systems to be integrable with other platforms. Despite huge progress, the capacity to generate coherent and sizeable electrical signal from their magnetization dynamics, and to couple efficiently with photonic excitation in cavities remain mainly elusive.

In this contribution, we will first report on our latest results on the investigation of inverse spin Hall effects generated by AFM resonances using hematite $Fe_2O_3$ and chromium oxide $Cr_2O_3$ as model systems [2].

In parallel, we discuss on the dynamics of antiferromagnets and how they can strongly couple with cavity photons and form AFM cavity magnon polaritons. We evidence that the presence of DMI in the canted phase of Hematite leads to an enhanced spin pumping signal as well as an increase of the magnon-cavity photon coupling strength compared to the collinear phase, achieving cooperativities C>70 for the canted phase [3]. These results pave the way to integrate canted AFMs in future AFM magnonic devices and for information processing with cavity magnonics

**Acknowledgements**

I.B. and R.L acknowledge support from the French Agence Nationale de la Recherche (ANR) under grant ANR-22-CE24-0008-03 (ICARUS). R.L., I. B., A.A. and M.K. acknowledge financial support from the Horizon 2020 Framework Programme of the European Commission under FET-Open grant agreement no. 863155 (s-Nebula). I. B. and R. L. acknowledges financial support from the Horizon 2020 Framework Programme of the European Commission under FET-Open grant agreement No. 964931 (TSAR). M.K. acknowledges support from the Graduate School of Excellence Materials Science in Mainz (MAINZ) DFG 266, the DAAD (Spintronics network, Project No. 57334897). M.K. acknowledges support from the DFG project number 423441604 and 268565370 (Project A01). The Research Council of Norway supported H.T.S., B.B, M.K., and A.B. through its Centers of Excellence funding scheme, project number 262633 “QuSpin”.

**References**

[1] V. Baltz et al., *Rev. Mod. Phys.* **90** (2018) 015005
[2] I. Boventer et al., *Phys. Rev. Lett.* **126** (2021) 187201
[3] I. Boventer et al. *Phys. Rev. Appl.* **19** (2023) 014071

# Observation of Nonreciprocal Magnon Hanle Effect

*J. Gückelhorn[1,2], S. Peña[3], M. Scheufele[1,2], M. Grammer[1,2], M. Opel[1], S. Geprägs[1], J.C. Cuevas[3], R. Gross[1,2,4], H. Huebl[1,2,4], A. Kamra[3], M. Althammer[1,2,]**

[1] *Walther-Meißner-Institut, Bayerische Akademie der Wissenschaften, Garching, Germany*
[2] *Technische Universität München, TUM School of Natural Sciences, Physik-Department, Garching, Germany*
[3] *Condensed Matter Physics Center (IFIMAC) and Departamento de Física Teórica de la Materia Condensada, Universidad Autónoma de Madrid, Madrid, Spain*
[4] *Munich Center for Quantum Science and Technology (MCQST), München, Germany*
*matthias.althammer@wmi.badw.de

In an ordered antiferromagnet, the quantized spin excitations with opposite chirality represent pairs of spin-up and -down magnons and thus can be characterized by a magnonic pseudospin. The precession of magnon pseudospin about the equilibrium pseudofield, the latter capturing the nature of magnonic eigen-excitations in an antiferromagnet, gives rise to the magnon Hanle effect [1]. Its realization via electrically injected and detected spin transport in an antiferromagnetic insulator demonstrates its high potential for devices and as a convenient probe for magnon eigenmodes and the underlying spin interactions in the antiferromagnet [2,3]. Here, we observe a nonreciprocity in the Hanle signal measured in hematite using two spatially separated platinum electrodes as spin injector/ detector [4]. Interchanging their roles was found to alter the detected magnon spin signal. The recorded difference depends on the applied magnetic field and reverses sign when the signal passes its nominal maximum at the so-called compensation field. We explain these observations in terms of a spin transport direction-dependent pseudofield. The latter leads to a nonreciprocity, which is found to be controllable via the applied magnetic field. The observed nonreciprocal response in the readily available hematite films opens interesting opportunities for realizing exotic physics predicted so far only for antiferromagnets with special crystal structures.

**Acknowledgements**

We gratefully acknowledge financial support from the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) under Germany's Excellence Strategy – EXC-2111 – 390814868 and project AL2110/2-1, and the Spanish Ministry for Science and Innovation – AEI Grant CEX2018-000805-M (through the "Maria de Maeztu" Programme for Units of Excellence in R&D).

**References**

[1] A. Kamra et al., Phys. Rev. B **102**, 174445 (2020).
[2] T. Wimmer et al., Phys. Rev. Lett. **125**, 247204 (2020).
[3] J. Gückelhorn et al., Phys. Rev. B **105**, 094440 (2022).
[4] J. Gückelhorn et al., arXiv:2209.09040 (2022).

# Spin Space Groups and Magnon Band Topology

*Paul McClarty*[1]*, *Roderich Moessner*[1], *Alberto Corticelli*,

[1] *Max Planck Institute for the Physics of Complex Systems, Nöthnitzer Strasse 38, 01187, Dresden*

*pmcclarty@pks.mpg.des

Magnetic crystalline symmetries as encoded in the magnetic space groups are central to condensed matter physics with implications ranging from structure determination to constraints on matrix elements and response functions. These groups combine anti-unitary symmetries such as time reversal with crystal translation and point group symmetries with the assumption that spin and space transformations are locked together. In real materials, though, spin and space symmetry operations may be wholly or partially decoupled leading to a much larger set of groups called the spin space groups. In this talk, I explain how these enhanced symmetries can be relevant to materials focussing on magnon band structure and topology. One surprising fact is that nontrivial space space symmetries can be present in magnets with significant spin-orbit coupled anisotropy with Kitaev magnets as a prominent example. We reveal that that systems with such anisotropies harbour a tower of different spin space groups and we show how the representation theory and compatibility relations can characterize key features of magnon bands revealing a proliferation of nodal points, lines, planes, and volumes.

## References

1. A. Corticelli, R. Moessner, P. McClarty, *Phys. Rev. B* **105** (2022) 064430.

# Interplay of Spin and Orbital Transport in Electrically and Optically Excited Antiferromagnets

*Yuriy Mokrousov*[1,2]*

[1] *Peter Grünberg Institut, Forschungszentrum Jülich, Jülich, Germany*
[2] *Institute of Physics, Johannes Gutenberg University Mainz, Mainz, Germany*
*y.mokrousov@fz-juelich.de

The progress in the field of antiferromagnetic spintronics has been firmly associated with the advantages that antiferromagnets bare for generation of spin currents. On the other hand, orbital currents have recently emerged as a novel paradigm and foundation for the field of orbitronics, which is able to naturally accommodate abundant lighter materials for the purpose of generating currents of angular momentum. In my talk, I will report on our understanding of specific microscopic mechanisms which drive non-trivial geometry of electronic bands in antiferromagents and altermagnets thus giving rise to complex intertwined spin and orbital physics out of equilibrium. Moreover, I will demonstrate that optical pulses can be used to drive large non-linear photocurrents of spin and orbital angular momentum in antiferromagnets by photospin and photoorbital Hall effects. While latter effects are extremely sensitive to the combined symmetry of the system in spin and real space, I will demonstrate how the orbital properties of antiferromagnets can be significantly promoted by lowering crystal symmetry in the system, thus suggesting a way to exploit an interplay between spin and orbital degrees of freedom for tracking the complex dynamics of staggered magnetization during electrical or optical switching.

# SYMPOSIUM 12.
## ANTIFERROMAGNETIC SPINTRONICS.
## S12. ORAL PRESENTATIONS

# Fast Magnetic Octupole Domain-wall Motion Driven by Spin-transfer Torque in Noncollinear Antiferromagnets

*Mingxing Wu[1, 2], Taishi Chen[1, 3, 4], Takuya Nomoto[5], Hironari Isshiki[1, 6], Yoshinobu Nakatani[7], Tomoya Higo[1, 4, 6], Takahiro Tomita[1,4,6], Kouta Kondou[2, 6], Ryotaro Arita[2, 5, 6], Satoru Nakatsuji[1, 4, 6], Yoshichika Otani[1, 2, 6*]*

[1] *The Institute for Solid State Physics, The University of Tokyo; Kashiwa, Chiba 277-8581, Japan.*

[2] *Center for Emergent Matter Science, RIKEN; 2-1 Hirosawa, Wako 351-0198, Japan.*

[3] *School of Physics, Southeast University; Nanjing 211189, China.*

[4] *Department of Physics, University of Tokyo; Hongo, Bunkyo-ku, Tokyo 113-0033, Japan.*

[5] *Research Center for Advanced Science and Technology, University of Tokyo; 4-6-1 Meguro-ku, Tokyo, 153-8904, Japan.*

[6] *CREST, Japan Science and Technology Agency (JST); 4-1-8 Honcho Kawaguchi, Saitama 332-0012, Japan.*

[7] *Department of Computer Science, University of Electro-Communications; 1-5-1 Chofugaoka, Chofu-Shi, Tokyo 182-8585, Japan.*

*corresponding author email address: yotani@issp.u-tokyo.ac.jp

Recent progress has shown the antiferromagnetic states are electrically manipulable in collinear antiferromagnets (AFMs) such as CuMnAs[1], $Mn_2Au$[2] owing to inversion asymmetry, or noncollinear chiral AFMs $Mn_3X$ (X = Sn, Ge)[3] with time-reversal asymmetry. Compared to collinear AFMs, the noncollinear $Mn_3X$ exhibits giant magnetic responses, beneficial for more efficient detection of their antiferromagnetic states. Recent experiments indicate that the electrical current may induce intrinsic spin torques to switch the magnetic octupole domain[4] and displace the magnetic octupole domain wall (MODW)[5] in $Mn_3X$. However, the direct evidence of current-induced spin-transfer torque (STT) for MODW motion has not been reported yet. Until now, the STT contribution to the MODW motion has not been elucidated in noncollinear AFMs.

Here, we demonstrate the current-induced STT can efficiently drive the fast MODW motion in $Mn_3X$. Our magneto-optical Kerr observation shows the current-induced STT can accelerate the Néel-like wall of $Mn_3Ge$ up to 750 m $s^{-1}$ with a small current density only of $7.56\times10^{10}$ A $m^{-2}$ under no magnetic field. The MODW exhibits surprisingly high mobility with a significantly small critical current density. In addition, we apply the general s-d exchange model[6] to formulate the current-induced STT for MODW motion. Our results open a new route to developing a mechanism responsible for domain-wall-based antiferromagnetic spintronic devices.

## Acknowledgements

This work was partially supported by JST-CREST(No. JPMJCR18T3) and JST-Mirai(JPMJMI20A1). M. Wu. would like to acknowledge support from JSPS “Research Program for Young Scientists (no. 21J21461).

## References

[1] Wadley, P. *et al*. *Science.* **351**, 587–590 (2016).
[2] Bodnar, S. Y. *et al. Nat. Commun.* **9**, 348 (2018).
[3] Tsai, H. *et al. Nature* **580**, 608–613 (2020).
[4] Xie, H., Chen, X., Zhang, Q., Yan, B. & Wu, Y. *Nat. Commun.* **13**, 5744 (2022).
[5] Sugimoto, S. *et al.Commun. Phys.* **3**, 1–9 (2020).
[6] Tatara, G., Kohno, H. & Shibata, J. *Phys. Rep.* **468**, 213–301 (2008).

# Spin Torque-Generated Antiferromagnetic Droplet Soliton

*Roman Ovcharov[1]*, Roman Khymyn[1], Boris Ivanov[2,3], Johan Åkerman[1]*

[1] *Department of Physics, University of Gothenburg, Gothenburg, Sweden*

[2] *Institute of Magnetism of NASU and MESU, Kyiv, Ukraine*

[3] *Radboud University, Institute for Molecules and Materials, Nijmegen, Netherlands*

*roman.ovcharov@physics.gu.se

Self-localized spin textures in antiferromagnets (AFMs) have unique properties advantageous for future spintronic applications, such as intrinsic high-frequency dynamics with low magnetic sensitivity and weak stray fields, spatially confined spin dynamics with robust alternate torque output [1, 2]. We demonstrate theoretically and by micro-magnetic simulations that the application of a spin current, inflowing from the nano-contact (NC), adjunct to an extended AFM film, can excite a dynamic self-localized soliton, equivalent to the ferromagnetic droplet [3, 4].

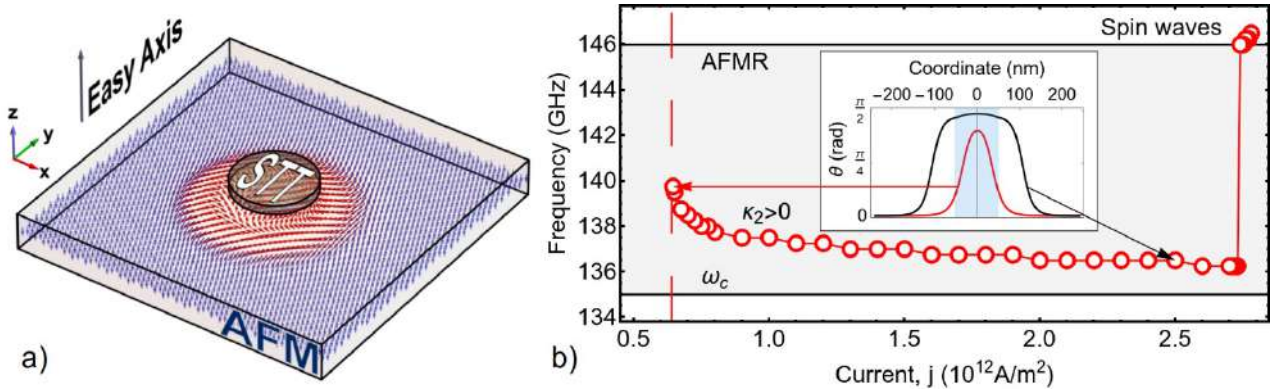



Figure 1: (a) The sketch that illustrates an AFM NC oscillator; (b) results of micro-magnetic simulations: frequency and profile of the AFM droplet.

We consider a system, schematically shown in Fig. 1 (a), which consists of a thin extended AFM film and NC, which provides STT. The anisotropy of the AFM is of the easy-axis type with the second-order term, and the STT polarization is parallel to the easy axis. At a particular sign of the secondary anisotropy, we observe a stable soliton at the currents above the threshold, see Fig. 1 (b). A noticeable gap between the droplet frequency and AFM resonance appears at the threshold, which is also a remarkable feature of the droplet in the ferromagnetic oscillators [3]. With increasing current, the soliton frequency decreases with the droplet profile's expansion. At a high current, the edge of the droplet can be expanded far beyond the NC area. With an even further increase in the current, the droplet loses its stability, and the area under the NC instead emits propagating spin waves [5], see Fig. 1 (b). We also show that the presence of an additional small anisotropy, perpendicular to the easy axis, can provide robust AC readout of the droplet spin dynamics at the double frequency of the soliton precession.

## References

[1] O. Gomonay, V. Baltz, A. Brataas, and Y. Tserkovnyak, *Nature Phys.* **14**, 213–216 (2018)

[2] R. Ovcharov, E. Galkina, B. Ivanov, and R. Khymyn, *Phys. Rev. Appl.* **18**, 024047 (2022).

[3] S. M. Mohseni, S. Sani, J. Persson, T. A. Nguyen, S. Chung, Y. Pogoryelov, P. Muduli, E. Iacocca, A. Eklund, R. Dumas, et al., *Science* **339**, 1295–1298 (2013).

[4] M. Ahlberg, S. Chung, S. Jiang, A. Frisk, M. Khademi, R. Khymyn, A. A. Awad, Q. T. Le, H. Mazraati, M. Mohseni, et al., *Nature communications* **13**, 1–7 (2022).

[5] M. Hamdi and D. Grundler, *arXiv preprint* arXiv:2206.07844 (2022)

# Large Spin Hall Magnetoresistance and Unidirectional Exchange Anisotropy at Antiferromagnet/$C_{60}$ Thin Film Interfaces

*Hari Babu Vasili[1]*, Satam Alotibi[1], Manuel Valvidares[2], Pierluigi Gargiani[2], Matthew Rogers[1], Mannan Ali[1], Timothy Moorsom[1], Gavin Burnell[1], Bryan J. Hickey[1], and Oscar Cespedes[1]**

[1] *School of Physics and Astronomy, University of Leeds, LS2 9JT, U.K.,*

[2] *ALBA Synchrotron Light Source, 08290 Cerdanyola del Vallès, Barcelona, Catalonia, Spain*

* h.vasili@leeds.ac.uk, o.cespedes@leeds.ac.uk

Recently, metallic antiferromagnets (IrMn, PtMn, etc) have attracted attention in spintronics due to high frequency (~THz) applications, strong inverse spin Hall effect (SHE) and the absence of stray magnetic fields. [1,2] Of particular interest in spintronics is transporting spin (only) angular momentum (pure spin currents) achieved in ferrimagnetic insulator [e.g. $Y_3Fe_5O_{12}$(YIG)] / normal metal (e.g. Pt) bilayers with a large spin-orbit coupling [3]. A parallel approach is the use of molecular layers (such as $C_{60}$) to tune the electronic properties of metallic and ferromagnetic interfaces in spintronic devices [4,5]. However, the effects of molecules on metallic antiferromagnet interfaces had so far not been studied. In this work, we report on the magnetic and spin transport of YIG/PtMn(t)/$C_{60}$(15 nm) (t = 1-10 nm), aiming at tunable SHE devices, and in search of spin order, enhanced spin transport, anisotropy anomalies and other emergent properties.

Figures 1(a,b) show that the spin Hall magnetoresistance (SHMR) of YIG/PtMn at 290 K is significantly larger when interfaced with $C_{60}$, resulting in an enhanced spin Hall angle ($\sim J_s/J_c$) from ~0.02 in PtMn to ~0.04 in PtMn/$C_{60}$. Soft X-ray Absorption Spectroscopy (XAS) and X-ray Magnetic Circular Dichroism (XMCD) at the Mn-$L_{2,3}$ absorption edges reveal that the Mn valence is reduced to +2 with $C_{60}$ *via* a charge transfer from Mn→C. This transfer causes an additional electron conducting layer and spin state (see Figure 1(d)). The total magnetic moment ($m_{tot}$) of PtMn at the Mn-$L_{2,3}$ edge is < 0.1 $\mu_B$ at 300 K, arising from uncompensated spins. At 2 K, the $m_{tot}$ of the pristine sample remains almost the same, but for $C_{60}$ interfaced PtMn is nearly double -inset Figure (d). This confirms that the $C_{60}$ interface is inducing uncompensated spins. PtMn -being antiferromagnetic (AF)- introduces an exchange bias with the ferromagnets at low T (Figure 1(e)). This is correlated with a SHMR sign change for YIG/PtMn below a blocking temperature, $T_B$, of ~ 50 K -data shown in Figure 1(c) – due to a change in the Neel vector direction during the spin flop [6]. The $C_{60}$ interface alters the magnetic order and Néel vector, leading to a positive SHMR until 10 K for t < 2 nm. At 2 K the AF exchange dominates and all samples show a negative SHMR. We note that the SHMR magnitude is asymmetric while cooling down -see Figure 1 (c)- attributed to the presence of unidirectional exchange anisotropy (UEA) at the YIG/PtMn interface in the out-of-plane direction. We disentangle the spin transfer torque for the UEA (~cosθ) which is different from the conventional SHMR (~$\cos^2\theta$). These results show for the first time the effects of molecules in the spin transport and magnetic order of AF metals.

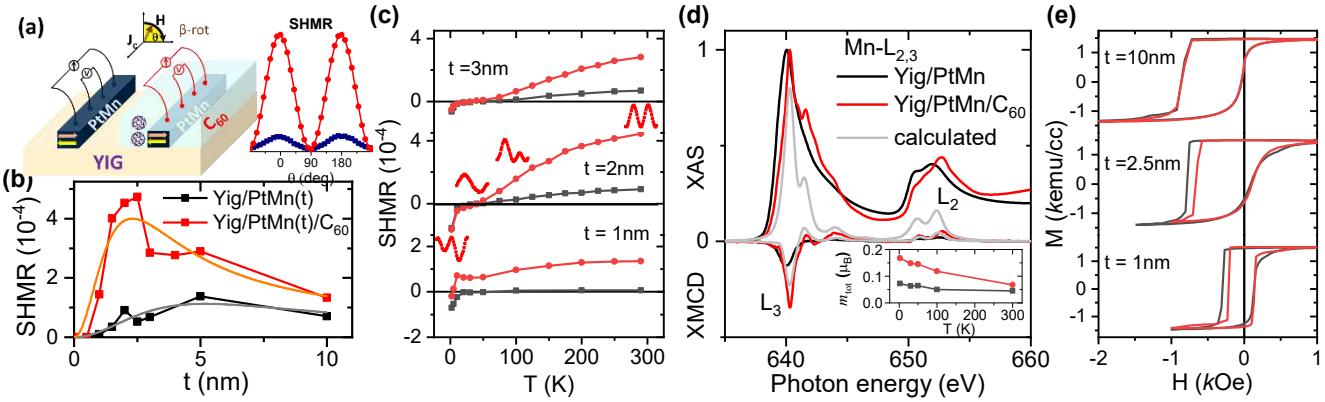



**Figure 1: (a)** Sketch of SHMR experiment on Yig/PtMn(t) with and without $C_{60}$. **(b)** The SHMR data at 290 K. **(c)** Temperature dependent SHMR for different PtMn thicknesses. **(d)** Mn-$L_{2,3}$ XAS and XMCD show $Mn^{2+}$ for the $C_{60}$ counterpart. Inset show the temperature variation of $m_{tot}$. **(e)** Magnetization of Co/PtMn(t) at 2K.

## Acknowledgements

We acknowledge support from the EU Horizon 2020 FET Open research grant 'INTERFAST' (No. 965046).

**References**

[1] Jungwirth et al., *Nat. Nanotechnol.* **11**, 231 (2016)
[2] Al Ma'Mari et al., *Nature* **524**, 69 (2015)
[3] Ou et al., *Phys. Rev. B* **93**, 220405(R) (2016)
[4] Alotibi et al., *ACS Appl. Mater. Interfaces* **13**, 5228 (2021)
[5] Fontcuberta et al. *Adv. Mat. Interfaces.* **6**, 1900475 (2019).
[6] Hou et al., *Phys. Rev. Lett.* **118**, 147202 (2017)

# Strain Control of Magnetoelectric Order in Multiferroic $SrMnO_3$ Thin Films

*J.J.L. van Rijn[1]*, D. Wang[2], I. Bhaduri[1], A. Das[1], V.E. Phanindra[1], B. Sanyal[2] and T. Banerjee[1]**

[1] *University of Groningen, Zernike Institute for Advanced Materials, Groningen, The Netherlands*
[2] *Department of Physics and Astronomy, Ångströmlaboratoriet, Uppsala University, Sweden*
*j.j.l.van.rijn@rug.nl & *t.banerjee@rug.nl

Multiferroic materials characterized by magnetoelectric coupling, allows for electric control on non-volatile memory in addition to magnetic control of electric polarization. These properties render them highly desirable both for fundamental exploration and for technological applications such as in novel computing architectures. In this work, we will discuss $SrMnO_3$, a rare example of a perovskite multiferroic, where the manganese atom hosts both the orders, and is theoretically predicted to exhibit a strong magnetoelectric coupling upon straining. This also predicts a giant magnetoelectrocaloric effect for an epitaxial strain at which the critical temperature of both order parameters coincide. The magnetic order is typically G-type antiferromagnetic in bulk but can be tuned by strain. We study this by depositing ultrathin $SrMnO_3$ films using pulsed laser deposition on five different substrates with the epitaxial strain varying from -0.4% to 3.9%. Detailed strain and crystal structure analysis of these films are followed by fabricating devices for studying the magnetic order and eventual coupling with the ferroic order. For this, we employ Spin Hall Magnetoresistance (SMR) studies using Pt nanostructures and study the ferroic order using Piezo-electric Force Microscopy and electrical Ferroelectric property measurements. Interestingly, we find that strain modifies the magnetic anisotropy, magnetic order and domain structures in these films. DFT studies predict a multiferroic phase above 1% epitaxial strain with a strong magnetoelectric coupling which corroborates with our findings.

## References

[1] J.J.L. van Rijn, D. Wang, B. Sanyal, T. Banerjee, *Phys. Rev. B* **106(21)** (2022) 214415.
[2] A. Das, V. E. Phanindra, A.J. Watson, T Banerjee, *Appl. Phys. Lett.* **118** (2021) 052407.

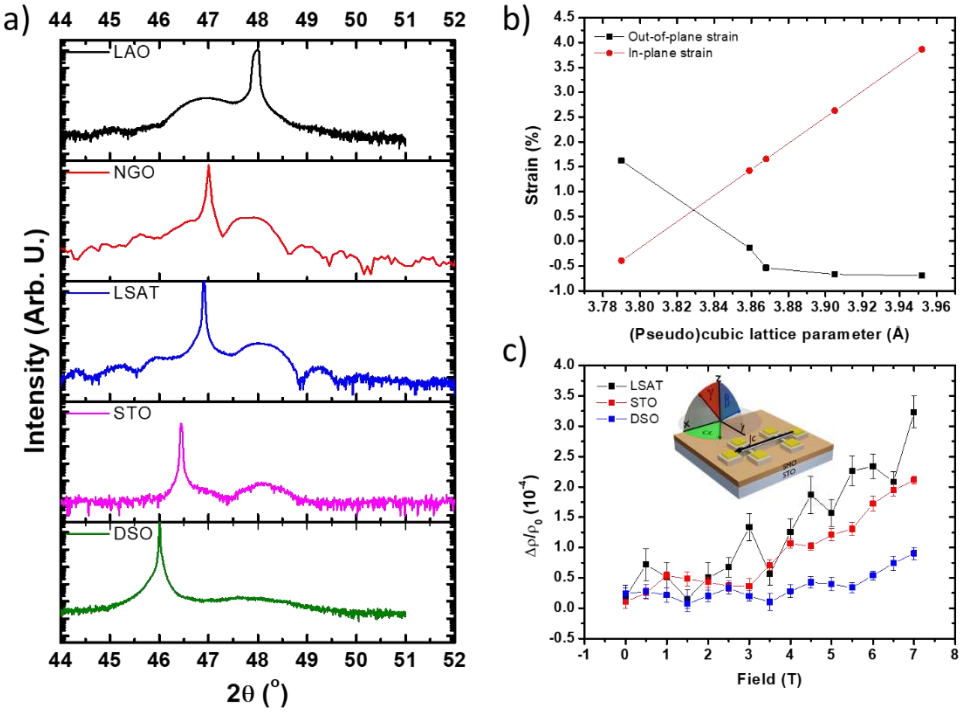



Figure: In a) the 2θ scan results for 12 nm $SrMnO_3$ films grown on five different substrates: $LaAlO_3$ (LAO), $NdGaO_3$ (NGO), $(LaAlO_3)0.3(Sr_2TaAlO_6)_{0.7}$ (LSAT), $SrTiO_3$ (STO) and $DyScO_3$ (DSO), are shown. From this, the out-of-plane strain is extracted and displayed in b) in addition to the in-plane strain obtained from Reciprocal space maps. In c) the field dependent magnitude of Spin Hall Magnetoresistance for three $SrMnO_3$ films is shown upon applying a current in a Pt structure as displayed in the inset, while rotating a magnetic field in the α-direction at 5 K.

# Impact of Strain on the Magneto-Optical Response of Ferrimagnetic and Antiferromagnetic $Mn_3NiN$ Thin Films

*F. Johnson[1], J. Zázvorka[2], L. Beran[2], D. Boldrin[1,3], L. F. Cohen[1], J. Zemen[4], M. Veis[2]*
[1] *Department of Physics, Imperial College London, London SW7 2AZ, UK*
[2] *Charles University, Faculty of Mathematics and Physics, Prague, 121 16, Czech Republic*
[3] *SUPA, School of Physics and Astronomy, University of Glasgow, Glasgow G12 8QQ, UK*
[4] *Czech Technical University in Prague, Faculty of Electrical Engineering, Czech Republic*
*veis@karlov.mff.cuni.cz

Antiferromagnets (AFMs) with frustrated exchange interactions between Mn ions offer unique opportunities for spintronic applications because of their chiral magnetic structures. In case of $Mn_3AN$ (A = Ga, Sn, Ni) systems that support the $\Gamma^{4g}$ spin arrangement in the (111) plane, the magnetic symmetry along with Weyl points in the band structure close to the Fermi energy allow for non-zero Berry phase curvature underpinning functional properties below the Néel temperature ($T_N$), such as anomalous Hall effect [1], anomalous Nernst effect [2], and magneto-optical Kerr effect (MOKE) [3]. A long range ordered ferrimagnetic (FIM) phase was also predicted in a strained state of these materials [4].

Here, we present a systematic theoretical and experimental study of magneto-optical response of strained $Mn_3NiN$ films on BTO, STO and LSAT substrates in AFM and FIM phases. The MOKE spectra showed notable evolution across $T_N$ as depicted in Fig. 1, confirming existence of a high temperature FIM phase. We employed ab-inito calculations using spin density functional theory and linear response approximation, and symmetry analysis to simulate the MOKE spectra. From the good agreement between the form of the measured and predicted MOKE spectra, we propose the AFM and FIM phases share the magnetic space group C2'/m' and that the symmetry driven magneto-optic and magneto-transport properties are maximized at room temperature in the FIM phase due to the enhanced intrinsic Berry phase contribution. More detailed comparison between the calculations and the experiment across the different strains induce by different substrates will be discussed. A room temperature FIM phase with large optical and transport signatures, as well as sensitivity to lattice strain and magnetic field has useful prospects for high-speed spintronic applications.

## Literature

[1] Chen, H., Niu, Q. & MacDonald, A. H. Anomalous Hall Effect Arising from Noncollinear Antiferromagnetism. *Physical Review Letters* **112**, 017205 (2014)
[2] Ikhlas, M., Tomita, T., Koretsune, T., Suzuki, M.-T., Nishio-Hamane, D., Arita, R., Otani, Y. & Nakatsuji, S. Large anomalous Nernst effect at room temperature in a chiral antiferromagnet. *Nature Physics* **13**, 1085-1090 (2017).
[3] Johnson, F., Zázvorka, J., Beran, L., Boldrin, D., Cohen, L. F., Zemen, J., Veis, M. Room temperature weak collinear ferrimagnet with symmetry driven, large intrinsic magneto-optic signatures. *Physical Review B* **107**, 014404 (2023).
[4] Zemen, J., Mendive-Tapia, E., Gercsi, Z., Banerjee, R., Staunton, J. B. & Sandeman, K. G. Frustrated magnetism and caloric effects in Mn-based antiperovskite nitrides: Ab initio theory. *Physical Review B* **95**, 184438 (2017).

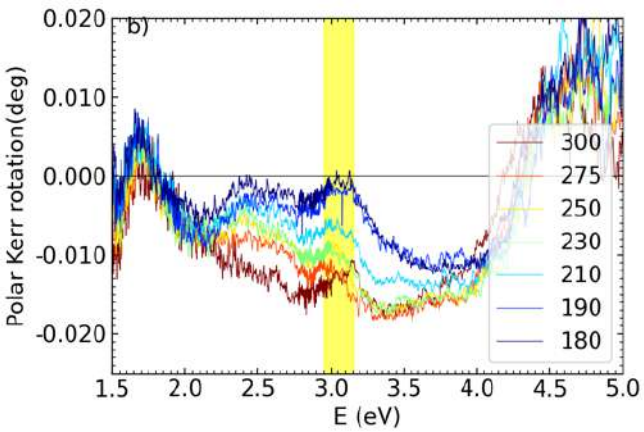



Fig 1. Temperature evolution of experimental spectra of polar MOKE rotation of Mn3NiN on BTO.

# Spin-Phonon Coupling in the 2D magnet CrSBr

*Alberto M. Ruiz*[1]*, *Gonzalo Rivero-Carracedo*[1], *Andrey Rybakov*[1], *José J. Baldoví*[1]

[1] *Instituto de Ciencia Molecular (ICMol), Universitat de València, Catedrático José Beltrán 2, Paterna, Spain*

*alberto.montoya@uv.es

2D layered antiferromagnetic materials have opened unprecedented avenues for spintronics at the limit of minituarization. CrSBr is a 2D magnetic van der Waals material that crystallizes in the orthorhombic P*mmm* space group, formed by ferromagnetic layers ($T_C$ = 146), which are antiferromagnetically coupled between them. [1, 2] In this talk, we will explore the spin-phonon coupling (SPC) interaction in CrSBr monolayer by means of first-principles calculations in order to determine the influence of the magnetic state in the amplitude and direction of phonon modes. By applying a finite displacement approach, combined with DFT calculations we obtained second derivatives of total energies as a function of displacement for different magnetic configurations following the corresponding phonon eigenvector direction ($u_j$). [3] This allows us to obtain the phonon frequency for each magnetic configuration and thus the SPC between each of them. Our results show that for some particular modes there is a strong SPC, indicating that there is an intimate dependency between the phonon vibrations and the magnetic ordering. Furthermore, for the modes presenting the strongest SPC, we determined the evolution of the magnetic exchange interactions $J_1$, $J_2$ and $J_3$ and magnon dispersion as a function of displacement following the corresponding $u_j$, revealing that there is a strong magnon-phonon coupling in CrSBr.

## Acknowledgements

The authors acknowledge the financial support from the European Union (ERC-2021-StG-101042680 2D-SMARTiES), the Spanish MIU (Grant No. FPU21/04195) and the Excellence Unit María de Maeztu Grant No. CEX2019-000919-M from the Spanish MICINN.

## References

[1] C. Boix-Constant, S. Mañas-Valero, A. M. Ruiz, A. Rybakov, K. A. Konieczny, S. Pillet, J. J. Baldoví, E. Coronado, *Adv. Mater*. **34** (2022) 2204940.
[2] D. L. Esteras, A. Rybakov, A. M. Ruiz, J. J. Baldoví, *Nano Lett.* **22** (2022) 8771-8778.
[3] J. Wu, Y. Yao, L. Miao-Ling, M. Rosner, Z. Du, K. Watanabe, T. Taniguchi, T. Ping-Heng, S- Haas, H. Wang, *Adv. Mater*. **34** (2022) 2108506.

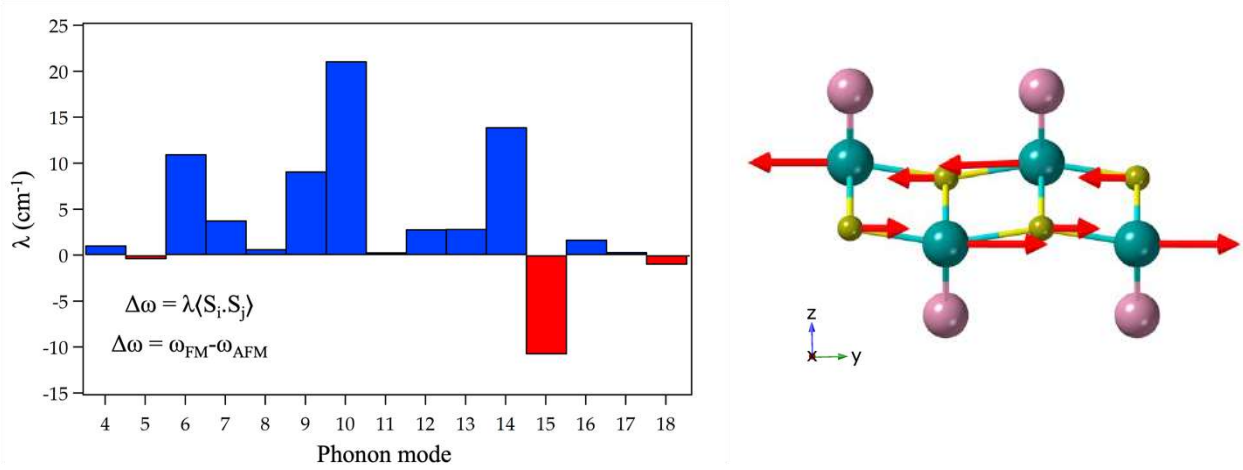



Figure: SPC of CrSBr for the 15 optical phonon modes (left) and graphical representation of movement of atoms for mode 10 (right). Cyan, yellow and pink balls represent chromium, sulfur and bromine atoms, respectively.

# ALTERMAGNETIC SPIN STRUCTURE STABILIZED by EPITAXY in $Mn_5Si_3$ THIN FILMS GROWN on Si(111)

*I. Kounta [1], H. Reichlova, [2, 3], D. Kriegner [2, 3], R. Lopes Seeger [4], A. Bad'ura [5], M.Leiviskä[4], J. Rial [4], S. Bertaina [6], M. Petit [1], P. Doležal [5], E. Schmoranzerova [5], L. Smejkal [7,3], J. Sinova [7,3], V. Baltz [4], S. T. B. Goennenwein [8,2], L. Michez [1,*]*

[1] *Aix Marseille Univ, CNRS, CINaM, Marseille, France*
[2] *IFM, Technische Universität Dresden, Dresden, Germany*
[3] *Institute of Physics, Czech Academy of Sciences, Czech Republic*
[4] *Univ. Grenoble Alpes, CNRS, CEA, Grenoble INP, Spintec, Grenoble, France*
[5] *Faculty of Mathematics and Physics, Charles University, Prague, Czech Republic*
[6] *Aix Marseille Univ, CNRS, IM2NP, Marseille, France*
[7] *Institut fur Physik, Johannes Gutenberg Universität Mainz, Germany*
[8] *Universitat Konstanz, Fachbereich Physik, Konstanz*
*lise.michez@univ-amu.fr

The antiferromagnetic $Mn_5Si_3$ compound has been known to give rise to a topological Hall effect (THE) in its non-collinear arrangement, i.e below 70 K. This effect has been observed in single crystal and polycrystalline thin films [1]. Here, we report on a route to grow epitaxial $Mn_5Si_3$ thin films on Si(111), with drastically different magnetotransport properties with respect to the bulk material. To this end, we use Mn and Si codeposition in a molecular beam epitaxy system, and carefully tune the deposition rates, the growth temperature, and the annealing temperature. The silicide phase-formation and morphology were assessed using reflection high-energy electron diffraction, X-ray diffraction, high resolution transmission electron microscopy (HRTEM) and atomic force microscopy. We show that it is possible to grow high quality crystalline $Mn_5Si_3$ thin films using interface engineering by means of a thin interfacial MnSi layer [2]. The latter acts as a seed layer for stabilizing the $Mn_5Si_3$ phase onto Si(111) substrate (Fig.1a). The magnetic behaviour of our films differs from the bulk and polycrystalline materials showing an enhanced transition temperature (~240 K) between the magnetically compensated phase and the paramagnetic state. This is attributed to its substrate-stabilized singular crystal structure that stays hexagonal in the whole temperature range as evidenced by the monotonous temperature variation of the in-plane lattice parameter '*a*' shown in Fig.1b, unlike in bulk $Mn_5Si_3$ in which the magnetic arrangement transitions are followed by structural changes. Interestingly, the specific crystal and spin symmetry of the $Mn_5Si_3$ thin films leads to a large spontaneous Hall effect (Fig. 1c) up to ~240 K [3, 4], in addition of the THE observed up to ~70 K. Epitaxy can therefore be used as a versatile tool to stabilize new spin structure and broaden the pool of materials in which spontaneous Hall is expected.

## References

[1] C. Sürgers *et al.*, *Nature Communications* **5** (2014) 3400.
[2] I. Kounta *et al.*, *Phys. Rev. Materials* **7** (2023) 024416.
[3] H. Reichlova *et al.*, arXiv:2012.15651 (2020).
[4] L. Smejkal, J. Sinova and T. Jungwirth, *Phys. Rev. X* 12 (2022) 040501.

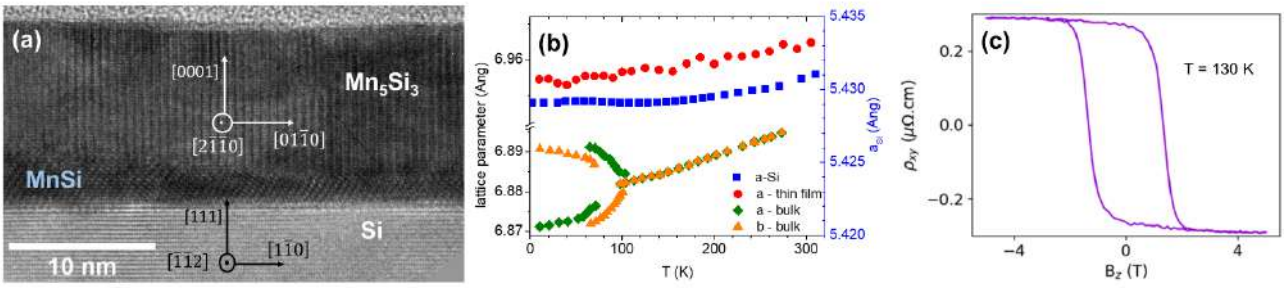



Figure 1: (a) HRTEM cross-section of a 13-nm thick $Mn_5Si_3$ thin film grown on Si(111). (b)Temperature dependence of in-plane lattice parameters '*a*' of $Mn_5Si_3$ thin films (red dots) following the same monotoneous trend of the Si substrate (blus square). For the sake of comparison, the temperature variation of the '*a*' and '*b*' parameters in bulk material is shown by green losanges and orange triangle, respectively. (c) Anomalous Hall resistivity in the antiferromagnetic phase measured at 130K, corresponding to a spontaneous Hall conductivitu of ~5 S/cm..

# Magneto-Transport Measurements in Altermagnetic Semiconductor MnTe

*Ruben D. Gonzalez Betancourt[1,2,3*], Jan Zubáč[3,4], Philipp Ritzinger[3], Rafael J. Gonzalez Hernandez[5], Kevin Geishendorf[3], Zbynek Šobáň[3], Gunther Springholz[6], Kamil Olejník[3], Libor Šmejkal[3], Tomas Jungwirth[3,7], Sebastian T. B. Goennenwein[1,8], Andy Thomas[1,2], Helena Reichlová [3], Jakub Železný[3], and Dominik Kriegner[1,3]*

[1]*IFMP, Technical University Dresden, Dresden, Germany*
[2]*IFW, Dresden, Germany*
[3]*Institute of Physics, Academy of Science of the Czech Republic, Prague, Czech Republic*
[4]*Charles University, Faculty of Mathematics and Physics,Prague, Czech Republic*
[5]*Departamento de Fisica y Geociencias, Universidad del Norte, Barranquilla, Colombia*
[6]*Semiconductor Physics, Johannes Kepler University Linz, Linz, Austria*
[7]*School of Physics and Astronomy, University of Nottingham, United Kingdom*
[8]*Department of Physics, University of Konstanz, Konstanz, Germany*
*ruben_dario.gonzalez_betancourt@tu-dreden.de

In altermagnets [1], the spin polarization in both real space and electronic structure alternates. Consequently, altermagnetism enables effects that were believed to be limited to ferromagnets. However, many of the predicted altermagnetic phenomena [2] await their experimental confirmation. Here, we present a magneto-transport characterization of the semiconducting, compensated collinear altermagnet MnTe (Fig.1-a) [3].

We experimentally confirm the symmetry components of the longitudinal (Fig.1-b) and transversal anisotropic magnetoresistance in thin films of MnTe. Furthermore, we discuss which contributions to the measured transversal and longitudinal signals can be signatures of the unconventional altermagnetic phase.

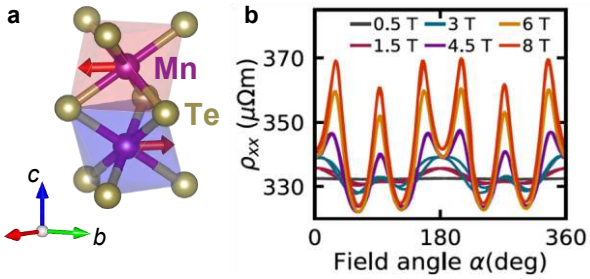



Figure 1: (a) Atomic and magnetic configuration of MnTe with hexagonal iAs structure. (b) Longitudinal magnetoresistance recorded in *c* plane oriented thin films and an in-plane rotating applied magnetic field.

**References:**

[1] L. Šmejkal et al., *Phys. Rev. X.*, 12.031042 (2022)
[2] L. Šmejkal et al., *Phys. Rev. X.*, 12.011028 (2022)
[3] R. D. Gonzalez Betancourt et al., *Phys. Rev. Lett.* **130**, 036702 (2023)

# Direct evidence for relativistic effects in hexagonal MnTe

*K. Gas[1]*, K. P. Kluczyk[2], P. Skupiński[1], M. J. Grzybowski[2], T. Fąs[2], J. Suffczyński[2], M. Baj[2], J. Z. Domagala[1], W. Wołkanowicz[1], E. Łusakowska[1], K. Grasza[1], A. Mycielski[1], M. A. Borysiewicz[3], K. Vyborny[4], U. Zülicke[5], R. Winkler[6,7], M. Sawicki[1] and M. Gryglas-Borysiewicz[2]*

[1] *Institute of Physics, Polish Academy of Sciences, Warsaw, Poland*
[2] *Faculty of Physics, University of Warsaw, Warsaw, Poland,*
[3] *Lukasiewicz Research Network - Institute of Microelectronics and Photonics, Warsaw, Poland,*
[4] *Institute of Physics, Academy of Sciences of the Czech Republic, Praha, Czech Republic,*
[5] *MacDiarmid Institute, School of Chemical and Physical Sciences, Victoria University of Wellington, Wellington, New Zealand*
[6] *Department of Physics, Northern Illinois University, DeKalb, USA*
[7] *Materials Science Division, Argonne National Laboratory, Lemont, Illinois, USA*
*kgas@ifpan.edu.pl

Recent theoretical and experimental studies of antiferromagnetic (AFM) materials have shown a breadth of new phenomena apparently similar to those of ferromagnetic (FM) counterparts but having different fundamental origin [1]. This is the case for the recently discovered subclass of collinear antiferromagnets, the so-called altermagnets [2]. Here, the nonrelativistic effects like the time reversal symmetry (TRS) breaking, mandated by the compensated antiparallel order, is accompanied by a locally anisotropic crystal environment giving rise to properties typical for either FM or AFM materials.

Hexagonal MnTe (α-MnTe) is a semiconducting compound with AFM order present up to just above room temperature. Numerous studies have elaborated that Mn magnetic moments exhibit a parallel alignment within the *c* planes, which are in turn oriented antiparallel one with respect to another. Yet, the Te atoms that form octahedra around the Mn atoms occupy noncentrosymmetric positions. This precludes crystallographic translation or inversion, yielding a strong nonrelativistic TRS breaking, which forms the basis for the altermagnetic phase and gives rise to such outstanding features as hysteretic anomalous Hall effect (AHE) existing in a material system without net magnetization [3]. Between the two directions of the maximal symmetry in the *c* plane, that is along the *a* axis or 30 deg away from it, the AHE is expected to vanish in the former and to achieve the highest values in the latter case [3]. Importantly, in this case, an axial vector parallel to *c* becomes allowed leading to the spontaneous formation of *c*-axis magnetization. Since it is of a higher order (relativistic) origin, its magnitude is expected to be weak, and in fact, its existence has never been reported to date.

Here we present structural, transport and magnetization studies of high quality bulk MnTe crystals. Raman spectra, temperature dependencies of magnetization and of the resistivity define the Néel temperature sharply at $T_N$ = 308 K. Our studies document the presence of a sizable and magnetic field dependent magnetic anisotropy with a magnitude expected for hexagonal antiferromagnets. Similarly as in [3] hysteresis in the Hall resistivity $\rho_{xy}(B)$ vanishing at $T_N$, is evidenced, corresponding to the predicted anomalous Hall effect (AHE) [2]. However, in contrast to [3], the signs of ordinary and anomalous Hall effects are opposite. Most importantly, for the first time we document the existence of the *c*-axis magnetization, which disappears precisely at $T_N$. Its weak magnitude – as low as $3 \times 10^{-5}$ $\mu_B$/Mn at 100 K exhibits a Brillouin-like dependence on temperature, serving as a measure of the AFM order parameter in this material. This *c*-axis magnetization is very robust, more than 2 T needs to be applied in the opposite direction to reverse its sign. It can be explained by a macrospin model of an easy-plane antiferromagnet with relativistic Dzyaloshinskii-Moriya interaction, which also reproduces the hysteretic behavior.

**Acknowledgements:** This work was partially supported by the Polish National Centre for Research and Development through grant TECHMATSTRATEG1/346720/8/NCBR/2017 and by National Science Centre, Poland through grant number 2021/40/C/ST3/00168.

**References**

[1] V. Baltz *et al*., Rev. Mod. Phys. **90**, 015005 (2018)
[2] L. Šmejkal, J. Sinova, and T. Jungwirth, Phys. Rev. X. **12**, 040501 (2022)
[3] R.D. Gonzalez Betancourt *et al*., *Phys. Rev. Lett*. **130**, 036702 (2023)

# Anisotropic Magnetoresistance in Systems with Non-collinear Magnetic Order

*Philipp Ritzinger*[1] *, *Karel Vyborny*[1]

[1] *Institute of Physics of the Czech Academy of Sciences, Cukrovarnicka 112/10, 162 00 Praha 6, Czech Republic*

*ritzinger@fzu.cz

The anisotropic magnetoresistance (AMR) has been the subject of extensive research since its discovery in 1857 by William Thomson. While theoretical studies have explored the microscopic mechanisms behind this effect, ranging from simple s-d models to complex ab initio calculations, most of the attention has been focused on ferromagnets. Recently, however, there has been an increasing interest in antiferromagnets (AFMs), whose magnetic order without net magnetic moment renders them excellent candidates for future spintronic devices [1].

AMR can arise from either anisotropic scattering (extrinsic) or an anisotropic Fermi surface (intrinsic) [2]. In this work, we focus on the latter, much less investigated intrinsic mechanism [3,4] in the context of non-collinear magnetic order inspired by real materials such as CrSe, δ-FeMn, $Mn_3Ge$, or $RbFe(MoO_4)_2$. We explore various types of lattices on a toy model level, including trigonal, tetrahedral, parallelogram, and kagome lattices. Magnetic moments can be arranged in many different ways on such lattices, and small changes can alter the Fermi surface symmetry, spin texture, and transport properties. We systematically investigate the influence of magnetic ordering on these properties, which allows us to predict general features of spin texture and transport by only considering the symmetry of the underlying system.

Preliminary ab initio calculations in real materials are used to guide the choice of magnetic configurations for our toy models. We employ Stoner-Wohlfahrt calculations to confirm the positions of magnetic moments in the toy models. Our results demonstrate that AFM systems without spin-orbit coupling on kagome and parallelogram lattices can develop anisotropy in the electric conductivity under an applied in-plane magnetic field (while orbital effects are neglected). This phenomenon relies on non-collinear magnetic order and does not occur in ferromagnets without spin-orbit coupling.

We will also propose a way to experimentally confirm our findings, which may have important implications for the development of future spintronic devices. Our findings shed light on the complex interplay between magnetic order, crystal symmetry, and electronic transport properties in magnetic materials.

**Acknowledgements**

We would like to express our gratitude to Jakub Železny for productive discussions and valuable coding contributions. We would like to express our gratitude to Jan Střeleček for help with the ab initio calculations.

Support from the Czech Science Foundation (GACR) via 22-21974S is appreciated.

**References**

[1] P. Ritzinger, K. Vyborny, https://arxiv.org/abs/2212.03700

[2] K. Výborný, J. Kučera, J. Sinova, A. W. Rushforth, B. L. Gallagher, T. Jungwirth, *Phys. Rev. B* **80** (2009) 165204.

[3] F. L. Zeng, Z.Y. Ren, Y. Li, J.Y. Zeng, M.W. Jia, J. Miao, A. Hoffmann, W. Zhang, Y.Z. Wu, Z. Yuan, *Phys. Rev. B* **125** (2020) 097201.

[4] L. Nadvornik, M. Borchert, L. Brandt, R. Schlitz, K. A. de Mare, K. Výborný, I. Mertig, G. Jakob, M. Kläui, S. T.B. Goennenwein, M. Wolf, G. Woltersdorf, T. Kampfrath, *Phys. Rev. X* **11** (2021) 021030.

# SYMPOSIUM 12.
## ANTIFERROMAGNETIC SPINTRONICS.
## S12. POSTERS

# Field-free spin-orbit torque-driven switching of Néel vector of antiferromagnetic insulator

*Fernando Ajejas[1, *], Felipe Torres[2, 3], Ali C. Basaran[1], Pavel Salev[1, †], and Ivan K. Schuller[1]*

[1] *Department of Physics and Center for Advanced Nanoscience, University of California, San Diego. La Jolla, CA, 92093, USA*

[2] *Departamento de Física, Facultad de Ciencias, Universidad de Chile, Casilla 653, Santiago, 7800024 Chile*

[3] *Centro de Nanociencia y Nanotecnología (CEDENNA), Avda. Ecuador 3493, Santiago, 9170124 Chile.*

*fajejas@physics.ucsd.edu

Enabling electrical control of antiferromagnetic (AFM) properties is one of the key requirements for developing novel spintronic technologies. We present a device in which the spin configuration of an AFM insulator ($FeF_2$) is modified by electrical current, taking advantage of the spin-orbit coupling existing in an adjacent heavy metal (HM) layer, such as W or Pt. We designed a trilayer, HM|AFM|FM, where the top ferromagnetic (FM) layer is exchange coupled to the AFM layer and thus can be used to monitor the changes in the AFM spin configuration. We performed magneto-optical Kerr effect measurements to probe the FM hysteresis loops as a function of temperature (T) and applied current ($I$). We found that the exchange bias field ($H_{EB}$) and coercivity ($H_c$) behavior due to the bulk AFM spins and the top AFM|FM interface can be strongly modified by the current applied at the bottom HM layer [Fig. 1]. We attribute this effect to an active spin-orbit torque generated at the HM|AFM interface that reaches the AFM|FM top interface, modifying amplitude and sign of $H_{EB}$. We found a critical current beyond which the effects on the $H_{EB}$ and $H_c$ are irreversible. AFM blocking temperature is shifted and the Néel vector is switched once a critical current threshold is overcoming. Applying a blocking temperature distribution approach, we present a model revealing two independent mechanisms of unpinned AFM moments. Temperature-dependent control experiments using normal metals (NM) such as Au in NM|AFM|FM and without AFM in HM|FM confirmed that the effect is produced by the HM-induced SOT and is not caused by thermal heating, Oersted field, or other potentially spurious effects.

This research was supported by the Department of Energy's Office of Basic Energy Science, under grant # DE-FG02-87ER45332.

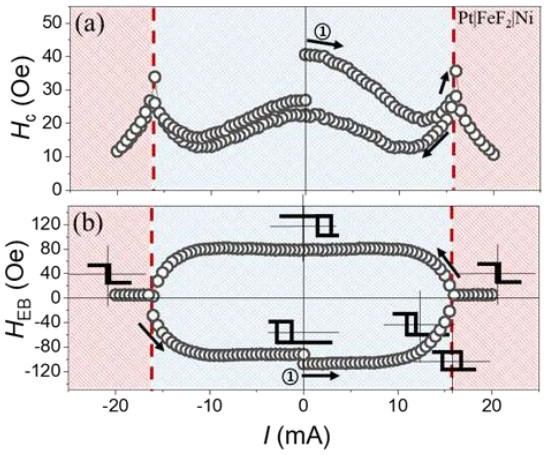



Figure 1 Pt|$FeF_2$|Ni (a) $H_c$ and (b) $H_{EB}$ as a function of the applied current $I$ at 20 K. The arrows represent the direction of the $I$ cycle. The blue area represents the region where the current effects are volatile. The dashed lines represent the critical current $I_c$, where the magnetization is switched. The red area represents the region where the current effects are permanent. The sketches represent the exchange bias sign and the coercive field amplitude.

# THz spectroscopy of optically induced quench switching of CuMnAs

*P. Kubaščík[1]*, D.Ch. Joshi[1], J.Jechumtál[1], K.Geischendorf[1], R. Matouška[1], M. Bušina[1],Z. Kašpar[1], M. Surýnek[1], A. Farkaš[1,2], J. Zubáč[1,2], E. Schmorranzerová[1], R. P. Campion[3],V. Novák[2], K. Olejník[2], T. Jungwirth[2,3], P.Němec[1], L.Nádvorník[1]*

[1]*Faculty of Mathematics and Physics, Charles University, Ke Karlovu 3, Prague 2 12116, Czech Republic*

[2]*Institute of Physics of the Czech Academy of Sciences, Cukrovarnická 10, Prague 6 16200, Czech Republic*

[3]*School of Physics and Astronomy, University of Nottingham, NG7 2RD, Nottingham, United Kingdom*

*kubascik@karlov.mff.cuni.cz

Nowadays, antiferromagnets (AF) attract significant attention because of the possibly higher integration density of devices and expected THz dynamics of magnetic moments. However, this advantageous robustness against external magnetic fields simultaneously complicates manipulation with magnetic moments and further development of antiferromagnetic spintronic devices. In recent years, AF with local symmetry breaking, such as CuMnAs, have attracted considerable attention because of manipulation with magnetic ordering via Néel spin-orbit torque [1]. Besides this, an additional writing mechanism based on a nano-fragmentation of AF domains by electrical [2], laser [2] and THz [3,4] pulses was demonstrated in CuMnAs. In all these experiments, stored information has been detected by electrical or optical measurements.

Here, we show that also picosecond pulses of THz radiation can be used to detect the laser induced nano-fragmentation of CuMnAs, as demonstrated in Fig. 1 where the resistence recovery dynamics is depicted (Fig. 1). We revealed that the THz detection provides results fully consistent with standard electrical measurements. Moreover, this THz detection shares advantages with optical detection, such as the noninvasive and contact-free nature or, possibly, the sub-picosecond temporal resolution. Thanks to this, we were able to measure the anisotropy of the quench switching in CuMnAs due to defects and the connected spectral changes in THz conductivity. These results are the first steps towards stroboscopic pump-probe experiments, which would allow us to study the nano-fragmentation processes at the ultimate sub-ps time scales.

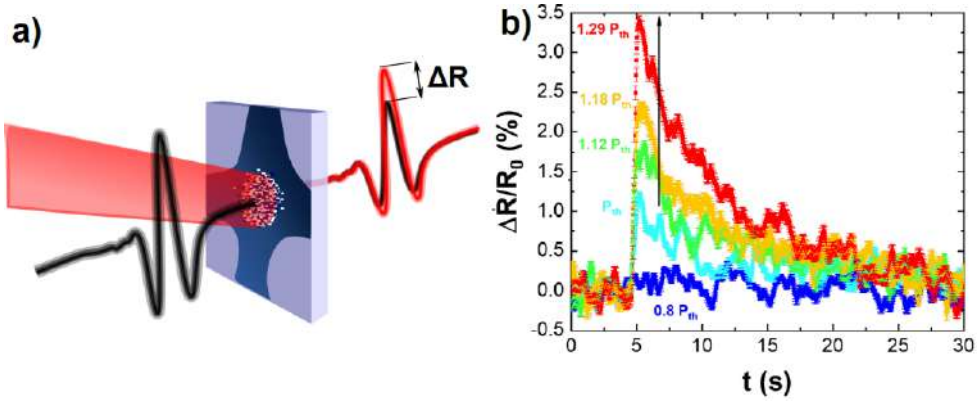



Figure: a) Schematics of the experiment. Femtosecond laser pulses induce nano-fragmentation of domains which are consequently detected by a change of the transmission of THz pulses. b) Temporal dependence of relative variation of THz resistance in laboratory time for laser pulse fluences bellow (blue points), at (azure) and above (the rest) the switching threshold fluence $P_{th}$.

## Acknowledgements

We would like to acknowledge to Czech Science Foundation (GA CR, Grant No. 21-28876J).

## References

[1] Wadley, P. et al. *Science 351*. 587-590(2016).
[2] Kašpar, Z. et al. Nature Electronics 4, 30-37 (2021).
[3] Olejník, K. et al. *Science advances 4*, eaar3566(2018).
[4] Heitz, J.J.F. et al. *Physical Review Applied 16*, 064047(2021).

# Finite size effect on the Hall response of altermagnetic $Mn_5Si_3$

*J. Rial* [1,2]*, *M. Leiviskä* [1], *H. Reichlova* [3,4], *I. Kounta* [5], *A. Badura* [4,6], *S. Beckert* [3], *R. Lopes Seeger* [1], *I. Joumard* [1], *F. Disdier* [1], *G. Gaudin* [1], *D. Kriegner* [3,4], *E. Schmoranzerova* [6], *L. Smejkal* [4,7], *J. Sinova* [4,7], *T. Jungwirth* [4,8], *S. T. B. Goennenwein* [3,9], *L. Michez* [5], *V. Baltz* [1]

[1] *Univ. Grenoble Alpes, CNRS, CEA, Grenoble INP, Spintec, Grenoble, France*
[2] *Univ. Autonoma de Madrid, Madrid, Spain*
[3] *IFMP, Technische Universität Dresden, Dresden, Germany*
[4] *Institute of Physics, Czech Academy of Sciences, Czech Republic*
[5] *Aix Marseille Univ, CNRS, CINaM, Marseille, France*
[6] *Faculty of Mathematics and Physics, Charles University, Prague, Czech Republic*
[7] *Institut für Physik, Johannes Gutenberg Universität Mainz, Germany*
[8] *School of Physics and Astronomy, University of Nottingham, Nottingham, United Kingdom*
[9] *Universität Konstanz, Fachbereich Physik, Konstanz, Germany*
*javier.rialrodriguez@cea.fr

Epitaxial thin film of $Mn_5Si_3$ [1,2] is a candidate to the recently predicted class of magnetic materials called 'alter'magnet [3], and more specifically to the anisotropic d-wave magnetic phases that features an unconventional combination of strong time reversal symmetry breaking responses and a vanishing net magnetization. The $Mn_5Si_3$ compound displays a large spontaneous anomalous Hall (AHE) response of a few $S.cm^{-1}$ below its Néel temperature of about 200 K, to which adds a topological-like (THE-like) signal below about 90 K, wherein some twisting of the magnetic moments is expected to occur, as opposed to a collinear arrangement above [1,2]. What is the anisotropic response of the AHE signal and what is the exact origin of the THE-like signal are some questions which are still pending and that we will present here. To address the first point, measurements were taken for a current and/or an external magnetic field applied along several crystallographic directions. For the second point a systematic study of the finite size effects was performed so as to understand whether the THE-like signal is the result of an actual topological effect related to the electrons picking up a Berry phase on interaction with the non-collinear spin structure of $Mn_5Si_3$ below 90 K or whether it is the bare summation of several AHE signals of opposite sign, in an inhomogeneous medium [4].

## Acknowledgements

This work was supported by the French national research agency (ANR) and the Deutsche Forschungsgemeinschaft (DFG) (Project MATHEEIAS- Grant No. ANR-20-CE92-0049-01 / DFG-445976410). JR acknowledges Spanish Ministry of Universities for the Margaritas Salas (MS) postdoctoral fellowship (CA1/RSUE/2021-00421).

## References

[1] I. Kounta et al., Phys. Rev. Mater. 7 (2023) 024416.
[2] H. Reichlova et al., arXiv:2012.15651 (2021).
[3] L. Smejkal et al., Phys. Rev. X 12 (2022) 040501.
[4] A. Gerber, Phys. Rev. B 98 (2018) 214440.

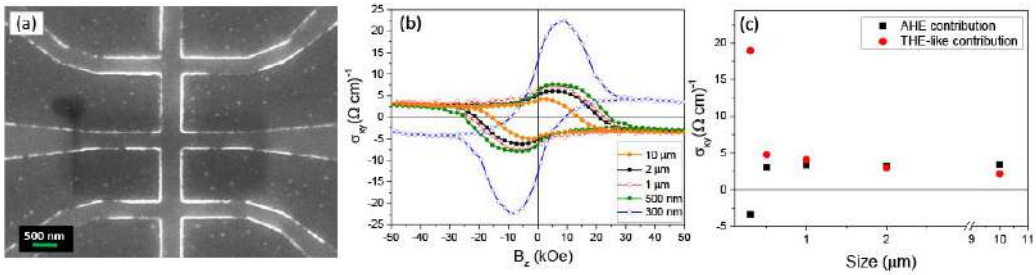



Figure 1: (a) SEM image of a ~300 nm $Mn_5Si_3$ Hall bar. (b) Hall conductivity of $Mn_5Si_3$ Hall bars of several lateral sizes, measured at 90 K. (c) Lateral size dependence of the spontaneous anomalous (AHE) and topological-like (THE-like) contributions to the Hall conductivity.

# Effects of magnetic field on spin-reorientation in antiferromagnetic $Eu^{3+}$-modified $TmFeO_3$ single crystal

*Poorva Sharma [1, 2, *] Ashwini Kumar [1], Arvind Yogi[3], Rubin Li[2], Baojuan Kang[2], Wei Ren[2], and Shixun Cao[2,4]*

*[1]Luzhou Key Laboratory of Intelligent Control & Electronic Device Application Technology, School of Electrical and Electronic Engineering, Luzhou Vocational and Technical College, Luzhou, 646000, China,*

*[2]Department of Physics, Materials Genome Institute and International Center for Quantum and Molecular Structures, Shanghai University, Shanghai, 200444, China*

*[3]UGC-DAE Consortium for Scientific Research, Khandwa Road, Indore 452001, India*

*[4]Shanghai Key Laboratory of High Temperature Superconductors, Shanghai University, Shanghai, 200444, China**corresponding author email address: poorva@lzy.edu.cn

Strongly correlated electron systems are an excellent platform for functional materials. $RFeO_3$ have attracted great attentions for various future applications in spintronics such as temperature and magnetic field control of spin-reorientation, inertia driven spin switching, laser-induced spin-reorientation, and large rotating-field entropy change [1 − 3]. $TmFeO_3$ crystallizes in an orthorhombic perovskite structure with *Pbnm* space group ($D\frac{16}{2h}$ symmetry) as schematically shown in Fig. 1. The magnetic anisotropy of $TmFeO_3$ undergoes dramatic changes as a function of temperature and due to this large magnetic anisotropy, magnetic phases such as $\Gamma_4(G_x, F_z)$, $\Gamma_{24}(G_{x,z}, F_{x,z})$, and $\Gamma_2(F_x, G_z)$ can appear with respect to temperature [4]. Here, we present a strong magnetic effect in the $Eu^{3+}$-doped $TmFeO_3$ single crystal, where half of the $Tm^{3+}$ sites are replaced by $Eu^{3+}$ dopants in the orthorhombic lattice of $TmFeO_3$ ($Tm_{0.5}Eu_{0.5}FeO_3$). We demonstrated by combining the *dc*-magnetic susceptibility $\chi(T)$ as well as magnetization isotherm $M(H)$, the temperature dependent magnetization results under small changes in the magnetic field ($H$) shows the evidence for the formation of such significant effect of $TmFeO_3$ as replaced by $Eu^{3+}$ ions. Interestingly, $Eu^{3+}$ doping protects the antiferromagnetic (AFM) spin-reorientation transition along crystallographic *c*-axis as observed by a sharp transition at $T_{SR}$ = 90 K, similar as parent $TmFeO_3$. Further, an external applied field ($H$) of 500 Oe is enough to fully suppress the spin-reorientation transition in $Eu^{3+}$ doped $TmFeO_3$. We emphasize that the transition from almost free-state of $Tm^{3+}$ spin to AFM state changes by 90 K at such low magnetic field. Experimentally observe significant changes in terms of shift in the magnetic transition (SRT) under an applied very low magnetic field due to $Eu^{3+}$ doping in $TmFeO_3$ lattice is truly robust effect.

**Acknowledgements**

This work is supported by the National Natural Science Foundation of China (NSFC, Nos. 11774217, 51672171, 51861145315 and 51911530124), and the National Key Research and Development Program of China (No. 2018YFB0704400). We also acknowledge the financial support from Luzhou Municipal Science and Technology Plan Project (grant numbers: 2021-JYJ-98; 2021-JYJ-99).

**References**

[1] A. V. Kimel *et al. Nat. Phys*. **5** (2009) 727–731.
[2] H. Wu, S. X. Cao, M. Liu, Y. Cao, B. Kang, J. Zhang, and W. Ren, *Phys. Rev.* B **90**, (2014) 144415.
[3] X. Li *et al.*, *Science* **361** (2018) 794.
[4] A. V. Kimel, A. Kirilyuk, A. Tsvetkov, R. V. Pisarev, Th. Rasing, *Nat*. **429** (2004) 850.

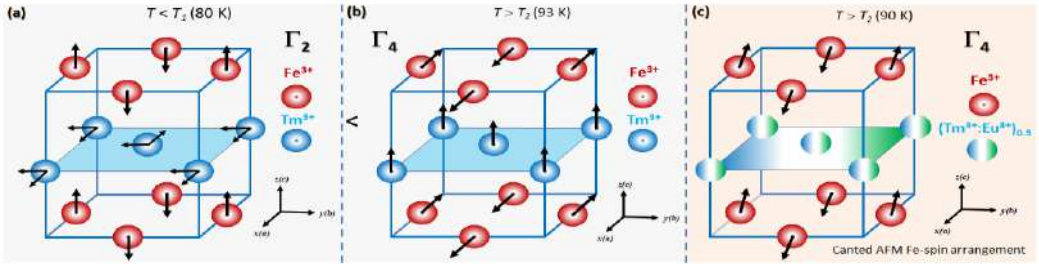



Figure 1: Schematic represents the magnetic structure of $TmFeO_3$ and $Eu^{3+}$-doped $TmFeO_3$ and spin structures for $\Gamma_2$, and $\Gamma_4$ configurations.

# Effect of samarium (Sm) doping on structural, magnetic and ferroelectric properties of $BiFeO_3$ for spintronics

*Ashwini Kumar[1,]*, Poorva Sharma[1], Qiu Fujun[1], Jingyou Tang[1], Guolong Tan[1,2]*

*[1]Luzhou Key Laboratory of Intelligent Control & Electronic Device Application Technology, School of Electrical and Electronic Engineering, Luzhou Vocational and Technical College, Luzhou, 646000, China,*
*[2] Institute of New Materials, Wuhan University of Technology, Wuhan, 430070, China*
*corresponding author email address: kumar1984@lzy.edu.cn

In recent years, multiferroics have gained popularity due to its possibility to combine two or more ferroic orders (ferroelectric, ferromagnetic and ferroelasticity) in the same phase, which can lead to coupling between them [1, 2]. The coupling between ferroelectric and magnetic ordering leads to the magnetoelectric effect (ME effect), where the polarization can be tuned by the applied magnetic field and vice versa, which is very desirable feature of multifunctional devices such as spintronic devices, magnetic data storage devices, sensors, etc. [3, 4]. In this way, bismuth ferrite ($BiFeO_3$, BFO) remains the promising candidate for lead-free type-1 multiferroic materials, which also makes it environment friendly and harmless. Here, we report the effect of samarium ($Sm^{3+}$) doping on structural, thermal, magnetic, and ferroelectric properties of $BiFeO_3$. Polycrystalline Sm-doped $BiFeO_3$ samples were synthesized using a conventional solid-state reaction route. $Bi_{1-x}Sm_xFeO_3$ ($x$ = 0.02-0.08) samples crystallize in rhombohedral structure ($R3c$). With the increase of $Sm^{3+}$ content, the decrease in crystal size is attributed to the change in lattice parameters, since the ionic radius of $Sm^{3+}$ (0.95 Å) is smaller than that of $Bi^{+3}$ (1.03 Å). A clear magnetic transition temperature ($T_N$) peak was obtained by DSC measurements. With the increase of $Sm^{3+}$ content, the peak temperature gradually decreased. Sm-doped $BiFeO_3$ samples exhibit ferromagnetic and antiferromagnetic behaviors. Magnetism is strongly influenced by the substitution of $Sm^{3+}$ ions in $BiFeO_3$, and thus the magnetism of the synthesized samples is greatly changed compared to that of pure $BiFeO_3$. $Bi_{1-x}Sm_xFeO_3$ ($x$ = 0.02-0.06) samples have weak pointed hysteresis loops. While, the BSFO-0.08 sample exhibit unsaturated rounded hysteresis loops possibly due to the large electrical leakage. Overall, we observed enhanced magnetic and ferroelectric properties by varying the $Sm^{3+}$ content in $BiFeO_3$. Therefore, the $Sm^{3+}$ doping in $BiFeO_3$ makes it a very prospective material for applications in spintronic devices, data storage devices and sensors.

**Acknowledgements**

We acknowledge the financial support from High-end Foreign Experts Introduction Program of Ministry of Science and Technology of China Project (grant number: G2022036014L), Luzhou Municipal Science and Technology Plan Project (grant numbers: 2021-JYJ-98, 2021-JYJ-99), Department of Science and Technology of Sichuan province Key R&D research Program (grant number 22ZDYF3765, 2022YFG0265) and Luzhou Vocational and Technical College Project (grant numbers: XJ2021005801, XJ2021005901)..

**References**

[1] J.T. Heron, M. Trassin, K. Ashraf, M. Gajek, Q. He, S.Y. Yang, D.E. Nikonov, Y. Chu, S. Salahuddin, R. Ramesh, *Phys. Rev. Lett*. **107** (2011) 217202.
[2] S.W. Cheong, M. Mostovoy, *Nat. Mater*. **6** (2007) 13–20.
[3] R. Ramesh, N.A. Spaldin, *Nat. Mater*. **6** (2007) 21.
[4] F. Kubel and H. Schmid, *Acta Crystallogr*. B **46** (1990) 698.

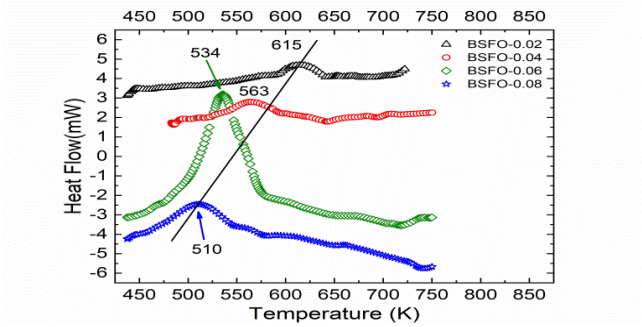



Figure 1: DSC curve of $Bi_{1-x}Sm_xFeO_3$ ($x$ = 0.02, 0.04, 0.06, and 0.08) samples. Arrow marks indicate its corresponding shift in magnetic transition temperature peak.

# Negative Energy Modes in Antiferromagnets for Amplification and Analogue Gravity

*V.Errani*[1] ,*R.A.Duine*[2]

[1] *Institute for Theoretical Physics, Utrecht University, 3584CC Utrecht, The Netherlands*
[2] *Institute for Theoretical Physics, Utrecht University, 3584CC Utrecht, The Netherlands*
[2] *Department of Applied Physics, Eindhoven University of Technology, P.O. Box 513, 5600 MB*
*v.errani@uu.nl

Magnonic black holes[1]– analogue event horizons for the spin-wave collective excitations of ordered magnets - can be used for fundamental research, for example for investigating Hawking radiation, but also for technological applications of spin waves, such as sensors[2], amplifiers[3], performing logical operations.

Here we will show how to engineer magnonic black holes in antiferromagnets[4], which have the attractive feature of fast magnetization dynamics. Our approach is both classical and quantum. We consider the set-up in Figure 1. The set-up consists of two antiferromagnets that are weakly coupled by exchange, and have different uniaxial magnetocrystalline anisotropies, magnetic fields, pumping mechanisms of angular momentum, i.e. Spin Orbit Torque (SOT) or Spin transfer Torque(STT).

We compute the values of parameters to have amplification of spin waves and moreover we individuate which configuration is more suitable for analogue gravity research purposes. Particular attention is paid to the differences between the classical and quantum regimes and on the challenges in observing the quantum effects.

**References**

1. Molina, A. S. Nunez, and R. Duine, “Magnonic black holes”, Physical review letters 118, 061301(2017)2
2. R. Duine, V. Errani, and J. Harms, “Non-linear dynamics near exceptional points of synthetic antiferromagnetic spin-torque oscillators”,10.48550/arXiv.2302.07607 (2023).
3. J. S. Harms, H. Y. Yuan, and R. A. Duine, Enhanced magnon spin current using the bosonic klein paradox,2021.
4. T. Jungwirth, X. Marti, P. Wadley, and J. Wunderlich, “Antiferromagnetic spintronics”, Nature nanotechnology 11, 231–241 (2016).

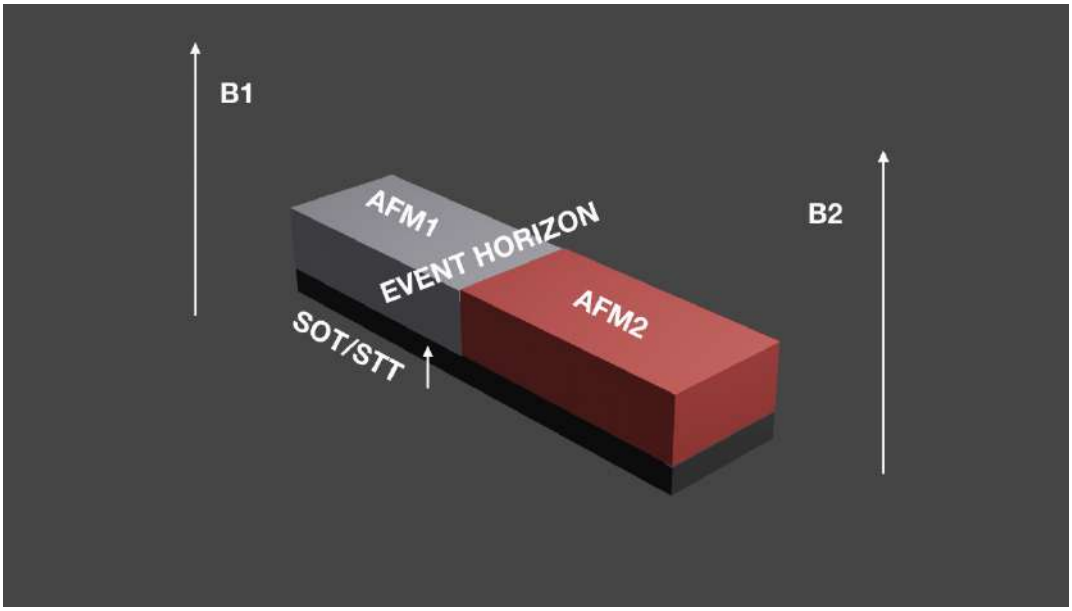



Figure: The set-up we study, in which uniaxial magnetocrystalline anisotropies, magnetic fields, angular momentum pumping mechanisms are varied between the two antiferromagnets

# Symmetry and Manipulation of the Magnetotransport Response in Altermagnetic $Mn_5Si_3$

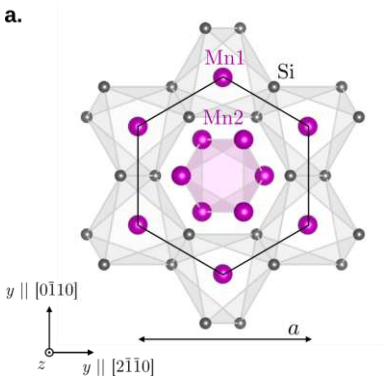
*Antonin Badura[1]*, Zeynab Sadeghi[2], Sebastian Beckert[3], Kamil Olejník[1], Ismaïla Kounta[4], Lisa Michez[4], Karel Výborný[1], Miina Leiviskä[5], Rafael Lopes Seeger[5], Javier Rial[5], Vincent Baltz[5], Daniel Scheffler[3], Libor Šmejkal[6], Jairo Sinova[6], Sebastian T. B. Goennenwein[7], Jakub Železný[1], Eva Schmoranzerová[2], Dominik Kriegner[1], and Helena Reichlová[1]*

[1] *Institute of Physics, Czech Academy of Sciences, Na Slovance 1999/2, 182 21 Prague 8, Czechia*
[2] *Faculty of Mathematics and Physics, Charles University, Ke Karlovu 3, 121 16 Prague, Czechia*
[3] *Institut für Festkörper- und Materialphysik, TU Dresden, 01062 Dresden, Germany*
[4] *Centre Interdisciplinaire de Nanoscience de Marseille, Aix-Marseille University, 163 Avenue de Luminy, 13288 Marseille Cedex 9, France*
[5] *Univ. Grenoble Alpes, CNRS, CEA, Grenoble INP, Spintec, F-38000 Grenoble, France*
[6] *Institut für Physik, Johannes Gutenberg Universität Mainz, 55128 Mainz, Germany*
[7] *Universität Konstanz, Fachbereich Physik, 78457 Konstanz, Germany*
*badura@fzu.cz

The recently introduced concept of altermagnetism [1] relies on the macroscopic time-reversal symmetry breaking in collinear ordered, compensated magnets. In contrast to antiferromagnets with time-reversal and space-inversion symmetries in their point groups, altermagnets can exhibit effects depending on the polarity of the ordering vector, e.g. the anomalous Hall effect. $Mn_5Si_3$ is a compensated magnet which, as a thin epitaxial layer, shows altermagnetic ordering. This was confirmed by our measurement of the spontaneous anomalous Hall effect which shows hysteretic behaviour with sizeable coercive field despite the negligible net magnetization of the $Mn_5Si_3$ thin films [2].

In this contribution, we focus on 10- to 20-nm epitaxial $Mn_5Si_3$ layers grown using molecular-beam epitaxy on a Si(111) substrate [3]. We introduce the origin of altermagnetic properties in our samples and we further discuss the symmetry of the measured magneto-transport response. The lowering of the symmetry connected with the onset of magnetic order in $Mn_5Si_3$ also allows for the presence of even contributions to the transverse magnetoresistance which we detect in precise experiments performed on lithographically defined Hall bar structures.

In the last part, we discuss the means of manipulation of the magnetotransport response in $Mn_5Si_3$ epitaxial layers. In particular, we present the key role of crystal quality. We also discuss the possibility to influence altermagnetism by external strain induced by piezoelectric stressors [4].



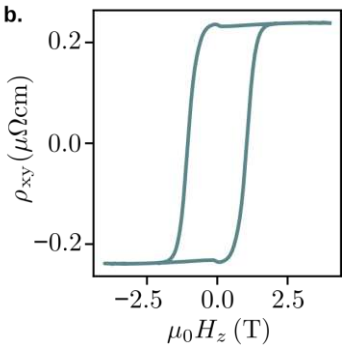




Figure: **a.** Top view of hexagonal crystal structure of the $Mn_5Si_3$ epilayers [2]. **b.** Transverse resistivity measured on lithographically patterned devices in a $Mn_5Si_3$ epitaxial film at 80 K shows clear hysteresis loop due to the anomalous Hall effect.

## References

[1] L. Šmejkal et al. *Phys. Rev.* X **12** (2022), 011028.
[2] H. Reichlová, et al. arXiv preprint (2020), 2012.15651.
[3] I. Kounta et al. *Phys. Rev. Mat.* **7** (2023), 024416.
[4] D. Butkovičová et al. *Rev. Sci. Instrum.* **84** (2013), 103902.

# Magnetic Switching and Exchange Bias Studies on Modified $SmCrO_3$

Manu Mohan*, Sanjib Nayak and P Murugavel
*Perovskite material laboratory, Functional Oxides Research group, Department of physics, Indian institute of technology Madras, Chennai, India*
*meetmanumohan@gmail.com

The study of compounds containing highly localized rare earth *f* - electrons, and their interaction with 3*d*-block transition-metal moments, which usually order at relatively high temperatures have always been a source of interesting physics. Among the rare-earth and transition-metal-based magnetic distorted perovskites, the rare-earth orthochromites ($RCrO_3$) shows fascinating behavior due to their coexisting functionalities comprising multiferroicity, spin-reorientation phase transition (SRPT), and sign reversal of magnetization. These features make $RCrO_3$ a potential candidate for possible novel applications in fast magnetic switching devices [1-3]. The $SmCrO_3$ is one of the interesting compounds in the $RCrO_3$ family which exhibits antiferromagnetic ordering ($T_N$) at ~ 191 K and spin reorientation ($T_{SR}$) at ~ 37 K [4]. In this work we prepared $SmCr_{0.8}Fe_{0.2}O_3$ by standard solid state reaction method. XRD studies revealed that the system belong to orthorhombic structure with Pbnm space group.The change in lattice parameter and strain due to the incoperation of $Fe^{3+}$ ions give raise to blue shift in the Raman spectrum. The analysis of the magnetic data obtained from the SQUID-VSM shows that the system exhibits antiferromagnetic ordering with $T_N$ ~ 185 K followed by a magnetic compensation temperatures ($T_{COMP}$) at ~ 137 K and 50 K and $T_{SR}$ at 64 K. The system shows an increase in the magnetic moment due to ordering of the $Sm^{3+}$ at low temperature region. Additionally, we noticed an interesting feature of magnetic field and temperature-dependent magnetization switching behavior in this sample. Notably, this sample is observed to exhibit high value of exchange bias at 10 K due to the existence of uncompensated spins. Exchange bias effect is confirmed by field cooled M-H measrements and training effect studies. The temperature-dependence specific heat curve confirms the short range ordering below $T_N$. Analysis of these results along with the complex interaction between the cations will be reported in this work.

## References

1. Y.Cao,S. Cao, W. Ren, Z. Feng, S. Yuan, B. Kang, B. Lu, and J. Zhang, Appl. Phys. Lett., **104** (2014)232405.
2. A. Kimel, B. Ivanov, R. Pisarev, P. Usachev, A. Kirilyuk, and T. Rasing, Nat. Phys., **5** (2009)727.
3. A. Ghosh, K. Dey, M. Chakraborty, S. Majumdar, and S. Giri, Europhys. Lett., **107** (2014)47012.
4. M.Tripathi, R. J. Choudhary, D. M. Phase, Phys. Rev. B., **96** (2017)174421.

# Epitaxy-driven altermagnetic phase in $Mn_5Si_3$ thin films

*I. Kounta[1*], H. Reichlova[2, 3], D. Kriegner[2, 3], R. Lopes Seeger[4], A. Badura[5], M. Leiviskä[4], J. Rial [4], S. Bertaina[6], M. Petit[1], P. Doležal[5], E. Schmoranzerova[5], L. Smejkal[7,3], J. Sinova[7,3], V. Baltz[4], S. T. B. Goennenwein[8,2], L. Michez[1]*

*[1] Aix Marseille Univ, CNRS, CINaM, Marseille, France*

*[2] IFM, Technische Universität Dresden, Dresden, Germany*

*[3] Institute of Physics, Czech Academy of Sciences, Czech Republic*

*[4] Univ. Grenoble Alpes, CNRS, CEA, Grenoble INP, Spintec, Grenoble, France*

*[5] Faculty of Mathematics and Physics, Charles University, Prague, Czech Republic*

*6 Aix Marseille Univ, CNRS, IM2NP, Marseille, France*

*[7] Institut fur Physik, Johannes Gutenberg Universität Mainz, Germany*

*[8] Universitat Konstanz, Fachbereich Physik, Konstanz*

*ismaila.kounta@univ-amu.fr

Bulk $Mn_5Si_3$ is of particular interest, due to its temperature- and field-dependent magnetic structure [1]. Recently, a new magnetic order has been evidenced in crystalline $Mn_5Si_3$ thin films through the observation of a spontaneous Hall signal up to ~240K [2]. This is in sharp contrast with bulk $Mn_5Si_3$ for which only a topological Hall effect was measured up to 70 K, i.e. in its non-collinear antiferromagnetic phase. The outstanding behaviour of the $Mn_5Si_3$ films is explained in terms of altermagnetism [3] arising from crystal and spin symmetry which macroscopically breaks the time reversal symmetry despite the absence of net magnetization. In this context, we present here the epitaxial growth of $Mn_5Si_3$ thin films on Si(111) and sapphire substrates by molecular beam epitaxy using a codepositon technique. Their structural properties have been characterized using reflection high-energy electron diffraction, x-ray diffraction, atomic force microscopy, and transmission electron microscopy. We have systematically studied the effect of the growth parameters (stoichiometry of the codeposit, growth and annealing temperature) on the structural and magnetic properties as well as on the magnetotransport. Concerning the growth on Si(111), the $Mn_5Si_3$/Si(111) heteroepitaxial growth followed the $Mn_5Si_3(0001)$-$[0\bar{1}10]$//Si(111)-$[1\bar{1}0]$ orientation through a complex interface composed either of an amorphous phase or a MnSi layer [4]. We show that interface engineering using a Sn-induced $(\sqrt{3}\times\sqrt{3})$R30°-Si(111) surface reconstruction can promote the direct $Mn_5Si_3$ growth on Si(111), yielding however a textured structure with a roughening of the top surface as shown in Fig.1(a). Our work also demonstrates a strong correlation between the $Mn_5Si_3$ crystal quality and the strength of the Hall effect (Fig. 1b), the signal being absent in polycrystalline films. This emphasizes the role played by epitaxy in the stabilization of a new crystal symmetry that can host an altermagnetic phase.

## References

[1] N. Bisniskos, *Phys. Rev. B* **105** (2023) 104404.
[2] H. Reichlová, Helena et al., arXiv preprint (2020), 2012.15651.
[3] L. Smejkal et al., *Phys. Rev.* X **12** (2022), 011028.
[4] I. Kounta et al., *Phys. Rev. Materials* **7** (2023) 024416.

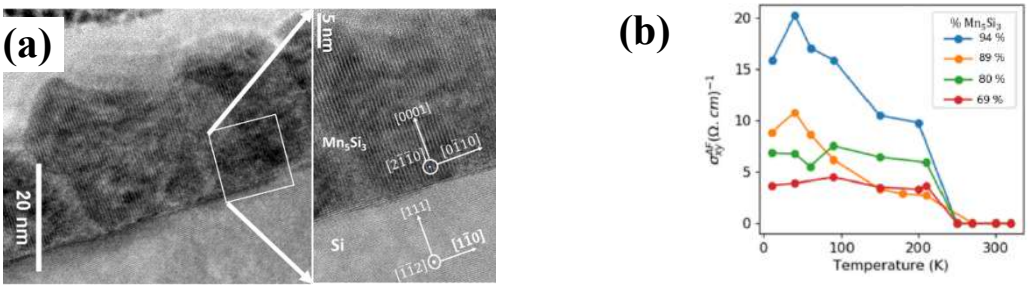



Figure: (a) HRTEM cross-section of a $Mn_5Si_3$ thin film grown on a Si(111)-$(\sqrt{3}\times\sqrt{3})$R30°:Sn surface. (b) Temperature dependence of the Hall conductivity for various samples with different crystal quality corresponding to distinct concentrations of Mn5Si3 within the Mn silide film. Lower Mn5Si3 proportion comes with higher mosaicity.

# Hybrid Magnetic Spinterfaces Based on Antiferromagnetic Oxides as Potential Transducers Between Light and Spin Waves

*A. Brambilla*[1], *A. Picone*[1], *M. Capra*[1], *A. Giampietri*[1], *F. Ciccacci*[1], *S. Fiori*[2], *G. Vinai*[2], *G. Panaccione*[2], *E. Molteni*[3], *S. Achilli*[3], *G. Fratesi*[3], *J. Nitschke*[4], *G. Zamborlini*[4] *and M. Cinchetti*[4]

[1] *Dipartimento di Fisica, Politecnico di Milano, Milano, Italy*

[2] *Istituto Officina dei Materiali (IOM)-CNR, Laboratorio TASC, Trieste, Italy*

[3] *Dipartimento di Fisica "Aldo Pontremoli", Università degli Studi di Milano, Milano, Italy*

[4] *Experimentelle Physik VI, Technische Universität Dortmund, 44221 Dortmund, Germany*

*alberto.brambilla@polimi.it

*Spinterfaces*, i.e. interfaces between an organic semiconductor (OS) and a ferromagnetic (FM) substrate, have been raising an ever increasing interest in the last two decades, first through the realization of organic spintronics prototypical devices, then by showing new intriguing phenomena related to the formation of hybridized interface states (HIS) [1]. As a promising development of the spinterface approach within the rapidly developing fields of *Antiferromagnetic (AF) Spintronics* and *AF Magnonics* [2], we have been extending those concepts to OS/AF interfaces, with the aim of creating a novel form of transducer between electromagnetic radiation (visible and near-IR) and spin waves (SW), based on the magnetic character of the HIS at the AF spinterface. It is well-known that AF materials are particularly suited as propagating media for SW at THz frequencies, while the light-induced perturbation of the magnetic moment associated to the HIS would allow for an electrode-free excitation of the nearby AF moments, otherwise hardly accessible by external stimuli. These ideas are under development in the ongoing EU-FET project SINFONIA [A1], during which we have been investigating different combinations of AF oxides, in particular $Cr_2O_3$, NiO and CoO (grown on different substrates), with suitable molecular materials. The latter include several different organic molecules, ranging from well-known ones like $C_{60}$ and Pentacene, to more complex compounds, such as Metal-Tetra Phenyl Porphyrins (MTPP; e.g., CoTPP) and Metal Phthalocyanines (MPc; e.g., FePc) [3], both of which are ideal candidates for building spinterfaces, since their ion core can have its own magnetic moment, due to the presence of unpaired spins [4,5].

Here, we are going to present in details the concept of our project and a series of results related to the growth and characterization (including crystalline, morphologic, and electronic properties) of the mentioned AF oxides and of related spinterfaces, along with some specific results related to the magnetism at the interfaces. Recently, in particular, we were able to show, by X-ray Magnetic Circular Dichroism (still unpublished), that FePc magnetically couples to $Cr_2O_3$, while similar measurements are foreseen in the near future, also based on the suggestions of various computational results based on first-principle theoretical approaches, which have in good part already been completed for selected AF spinterfaces. Finally, the expected development of the current research and its potential applications will be discussed.

## Acknowledgements

[A1] This research has received funding from the EU H2020 programme under Project SINFONIA, Grant 964396 - www.sinfonia-fet.eu

## References

[1] Mirko Cinchetti, V. Alek Dediu and Luis E. Hueso, *Nat. Materials* **16** (2017) 507.
[2] V. Baltz, A. Manchon, M. Tsoi, et al., *Rev. Mod. Phys.* **90** (2018) 015005.
[3] J. M. Gottfried, *Surf. Sci. Rep.* **70** (2015) 259.
[4] M.S. Jagadeesh, A. Calloni, A. Brambilla, et al., *Appl. Phys. Lett.*, **115** (2019) 082404.
[5] Annese, E.; Fujii, J.; Vobornik, I.; Panaccione, G. and Rossi, G. *Phys. Rev. B* **84** (2011) 174443.

# Parameters affecting the optically induced quench-switching of the antiferromagnetic CuMnAs

*Andrej Farkaš[1,2] *, Miloslav Surýnek[2], Prabhat Kumar[1], Filip Křížek[1], Vít Novák[1], Petr Němec[2], Tomáš Jungwirth[1], Kamil Olejník[1]*

[1] *FZU - Institute of Physics of the Czech Academy of Sciences, Prague, Czechia*
[2] *Charles University, Faculty of Mathematics and Physics, Prague, Czechia*
*farkasan@fzu.cz

Antiferromagnetic materials represent a new area of research with potential applicability in spintronic devices[1]. Compared to their ferromagnetic counterparts, antiferromagnetic materials exhibit faster dynamics, resiliency to the external magnetic field, and the absence of stray fields allowing for high integration density. Some functionalities are derived directly from their ferromagnetic counterparts, such as readout using anisotropic magnetoresistance or magnetic axis reorientation using current-induced spin-orbit torque [2]. Other are unique to antiferromagnets, such as recently discovered quenching into high resistivity states [3]. The quench-switching effect is based on changes in the magnetic domain structure of the epitaxially grown thin antiferromagnetic films of CuMnAs. These changes can be induced by current or optical pulses, which bring the system to the vicinity of its Neél temperature. The subsequent fast cooling results in a quenched magnetic state characterized by increased resistivity. The change in the resistivity can reach tens of percent at room temperature and even up to a hundred percent at low temperatures, which exceeds the spin-orbit torque-based switching by two orders of magnitude [2]. The quench-switching signals exhibit a temperature-dependent relaxation with characteristic times in the range of seconds at room temperature (Fig. b) [3], making it interesting for memory applications.

In this contribution, we explore differences in the response caused by changes in material parameters, such as CuMnAs layer thickness (Fig. c), device size, MBE growth conditions, and protective capping. The device performance is evaluated based on the observed amplitude of the quench-switching response. We also evaluate the durability of devices by monitoring the surface damage induced by the laser pulses.

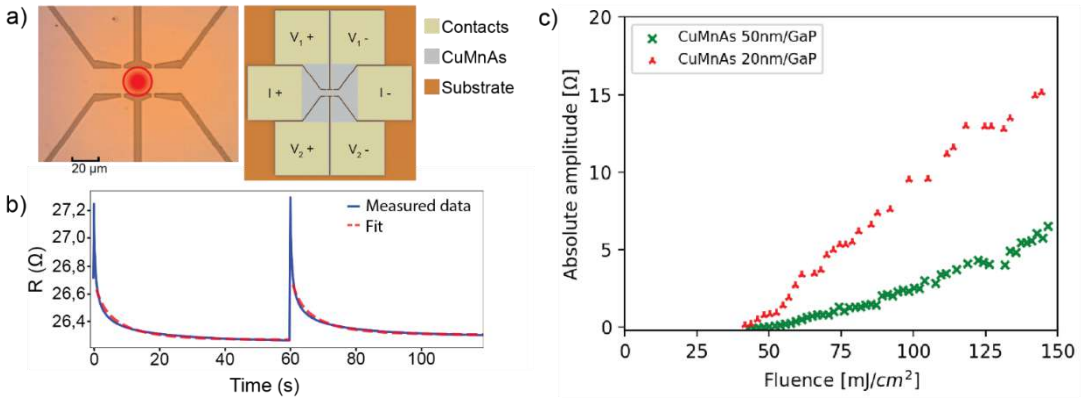



Figure: a) Example of the Hall bar device used for characterization. The position of the laser spot is depicted in the active area. b) Example of the measured quench-switching signal with a fitted stretch exponential function. c) Quench-switching amplitude as a function of the laser pulse fluence for 50 nm and 20 nm thick CuMnAs films.

## Acknowledgments

This work was supported by the Czech Science Foundation grant no. 21-28876J.

## References

[1] Jungwirth, T., et al. Antiferromagnetic spintronics. Nature Nanotech 11, 231– 241 (2016). https://doi.org/10.1038/nnano.2016.18

[2] Wadley, P. et al. Electrical switching of an antiferromagnet. Science 351, 587–590 (2016). https://doi.org/10.1126/science.aab1031

[3] Kašpar, Z., et al. Quenching of an antiferromagnet into high resistivity states using electrical or ultrashort optical pulses. Nat Electron 4, 30–37 (2021). https://doi.org/10.1038/s41928-020-00506-4

# Experimental Methods of Quantifying Magnetism of Thin Antiferromagnetic Layers Deposited on MgO Substrates

*K. Gas[1]*, S. Wakabayashi[2,3], T. Uchimura[2,3], J.-Y. Yoon[2,3], Y. Takeuchi[4], S. Kanai[2-4], S. Fukami[2-5], H. Ohno[2-5],and M. Sawicki[1]*

[1] *Institute of Physics, Polish Academy of Sciences, Aleja Lotnikow 32/46, PL 02-668 Warsaw, Poland*
[2] *Laboratory for Nanoelectronics and Spintronics, Research Institute of Electrical Communication, Tohoku University, Katahira 2-1-1, Aoba-ku, Sendai 980-8577, Japan*
[3] *Graduate School of Engineering, Tohoku University, Katahira 2-1-1, Aoba-ku, Sendai 980-0845, Japan*
[4] *WPI-Advanced Institute for Materials Research, Tohoku University, Katahira 2-1-1, Aoba-ku, Sendai 980-8577, Japan*
[5] *Center for Innovative Integrated Electronic Systems, Tohoku University, Aramaki-Aza-Aoba 468-1, Aoba-ku, Sendai 980-0845, Japan*
*kgas@ifpan.edu.pl

Non-collinear antiferromagnets e.g., $D0_{19}$-$Mn_3Sn$ and $Mn_3Ge$, possessing non-collinear spin configurations due to geometrical frustrations, are attracting a great deal of attention because of a large anomalous Hall effect due to a non-vanishing Berry curvature arising from the topologically non-trivial spin texture [1]. Technologically viable thin layers are deposited on bulky substrates, possessing a considerable challenge for precise volume magnetic characterization. Things get worse when the substrate material does not exhibit ideal diamagnetic properties. MgO, frequently used as a substrate material, has been already recognized to exhibit strong paramagnetic (PM) (dominating at low temperatures) and ferromagnetic-like (FM-like) responses. Particularly, in the case of antiferromagnetic materials, the signal of interest is only a small perturbation to the sizable response of MgO. In our study we aim to establish a scheme to characterize the magnetic properties of very thin $Mn_3Sn$ films.

The layers are epitaxially deposited by magnetron sputtering on MgO single-crystal substrates. Our dedicated magnetic characterization of these substrates confirms the existence of both the strong PM and FM-like responses. The former originates from the bulk of the substrates and can be accurately described by spins obeying the Brillouin function for $L = 0$, $S = 5/2$ with a concentration of $4\times10^{18}$ cm$^{-3}$. The strong FM-like response is found to be of a superparamagnetic origin and survives up to the highest attainable temperature of 400 K. Its $m(H)$ exhibits a Langevin-like sigmoidal shape saturating around 10 kOe with a magnitude depending on the batch and changing between $2.5\times10^{-5}$ and $5\times10^{-5}$ emu/cm$^2$. Typical coercive field is around 250 Oe. We show that this FM-like contamination is located exclusively on the epi-ready side of the substrate.

In order to eliminate both these PM and FM-like signals as well as the diamagnetism of MgO in our volume magnetometry studies, we adopt the concept of *in situ* compensation elaborated for mitigating the contribution brought about by bulky substrates [3]. In our approach the $Mn_3Sn$/MgO specimens are abutted on either side with seven $10\times5$ mm$^2$ pieces of MgO substrates, to form a quite uniform 15 cm strip of MgO which vastly restores the translational symmetry of the sample holder. Importantly the compensating MgO bits come from the same batch as the investigated samples. By doing so we achieve a compensation level of about 95%. Additional important helping hints will be presented and discussed. Our approach allows us to minimize error bar about 20 times, leading to precisely establishing the absolute values of magnetization in a series of $Mn_3Sn$ layers with thickness ranging from 10 to 50 nm, as it will be discussed elsewhere. Our findings and the experimental technique offer a solid foundation to study the unconventional physical properties of antiferromagnetic thin films.

**Acknowledgements:** This study has been partly supported by JSPS Kakenhi 19H05622.

## References

[1] Y. Takeuchi *et al*., *Nat. Mater.* **20** (2021) 1364-1370.
[2] M. Khalid *et al*., *Phys. Rev. B* **81** (2010) 214414.
[3] K. Gas and M. Sawicki, *Meas. Sci. Technol.* **30** (2019) 085003.

# Optical detection of collective spin-wave excitation in semiconducting NaMnAs crystal

*Jan Dzian[1]*, Martin Veis[1], Jiří Volný[1], Klára Uhlířová[1], Karel Výborný[2], Milan Orlita[3]*

[1] *Charles University, Faculty of Mathematics and Physics, Ke Karlovu 2027/3, 121 16, Prague, Czech Republic*

[2] *Institute of Physics, Academy of Science of the Czech Republic, Cukrovarnická 10, Praha 6, CZ-16253, Czech Republic*

[3] *Laboratoire National des Champs Magnétiques Intenses, CNRS, 25, avenue des Martyrs, 38042 Grenoble, France*

*dzian.jan@karlov.mff.cuni.cz

Materials with antiferromagnetic ordering are one of the most active research topics in current condensed matter physics [1-2]. NaMnAs is a semiconducting antiferromagnet from group of I-II-V semiconductors. The Neél temperature is above room temperature, and the material exhibits uniaxial anisotropy [3]. This makes NaMnAs an interesting material on the field of novel magnetic materials.

Our poster presentation focuses on magneto-optical properties of bulk single-crystal samples of antiferromagnetic semiconductor NaMnAs. We present low temperature infrared magneto-transmission spectroscopy at low energies, where we observe magnon-like mode at $\hbar\omega$ = 7 meV. With application of magnetic field along material's tetragonal axis, we observe splitting of the mode into 2 branches. The dispersion is linear with magnetic field and the extrapolated g-factor is approximately 2, which is in agreement with the expected value. The absence of interacting phonon modes in the vicinity of the magnon-like mode shows, that NaMnAs provides textbook example of antiferromagnetic resonance in easy-axis material. This statement is supported by magneto-transmission spectroscopy in additional geometry configurations.

The experimental results are accompanied with linear spin-wave theory simulation of dispersion of magnon in k-space. The simulation is based on the experiment discussed above and on ab initio theoretical predictions of exchange interactions. Theoretical predictions are developed from experimental measurements of optical properties of the material. To calculate the spin-wave simulation, we apply software package SpinW written for MATLAB. By correlating simulation results with experiment, we are able to determinate single-ion anisotropy energy.

**Acknowledgements**

None to be declared.

## References

[1] Jungwirth T. et al., Antiferromagnetic spintronics. Nature Nanotechnol. 11, 231 (2016).
[2] Baltz V. et al., Antiferromagnetic spintronics. Rev. Mod. Phys. 90, 015005 (2018).
[3] Volny, J. et al., Single-crystal studies and electronic structure investigation of the room-temperature semiconductor NaMnAs. Physical Review B (Vol. 105, Issue 12). American Physical Society (APS).

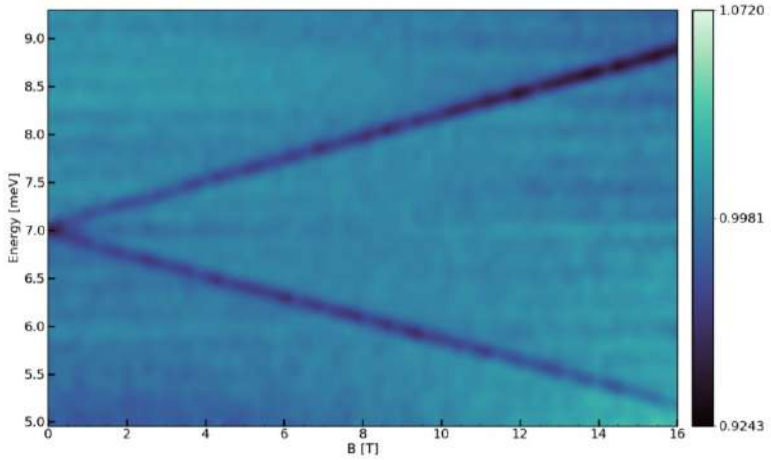



Figure: Magnetic field dispersion of magnon-like mode observed in low energy IR transmission spectra - false map $I(B)/I(B_{avg})$

# Antiferromagnetic domain distribution for different stoichiometries in mixed Ni-Co oxide films

A. Mandziak [1], C. Granados-Miralles[2], A. Berja[2] A. Quesada[2], J.E. Prieto[3*], M. Foerster[4], L. Aballe[4], M.A. Nino[4], J. de la Figuera[3] and P. Nita[5]

[1]*National Synchrotron Radiation Centre SOLARIS, Jagiellonian University, 30392 Cracow, Poland*
[2]*Instituto de Cerámica y Vidrio (CSIC), 28049 Madrid, Spain*
[3]*Instituto de Química Física "Rocasolano", 28006 Madrid, Spain*
[4]*Alba Synchrotron Light Facility, CELLS, 08290 Barcelona, Spain*
[5]*Faculty of Physics, Astronomy and Applied Computer Science, Jagiellonian University, 30348 Cracow, Poland*

Email: jprieto@iqfr.csic.es

Antiferromagnets (AFM) are promising materials for future spintronic applications owing to their advantageous properties: They are magnetically ordered but neighboring magnetic moments point in opposite directions, which results in zero net magnetization. This means antiferromagnets produce no stray fields and are insensitive to external magnetic field perturbations. Furthermore, they show intrinsic high frequency dynamics (in the THz regime) and exhibit considerable spin–orbit and magneto-transport effects. Over the past decade, it has been realized that antiferromagnets have more to offer than just being utilized as passive components in exchange bias applications. Among the wide variety of AFM materials, transition metal oxides (e.g. NiO or CoO) play an important role, especially in systems of reduced dimensionality, as pointed out by a number of studies [1, 2]. However, in thin film form, as required for devices, their quality is often disappointing, dictated by the defect density.
Here we demonstrate a route for preparing high quality ultrathin antiferromagnetic oxide films on a metallic substrate. Mixed nickel-cobalt oxides have been grown on Ru(0001) by high temperature oxygen-assisted molecular beam epitaxy. A comprehensive characterization has been performed combining LEEM and LEED for structural characterization and PEEM (PhotoEmission Electron Microscopy) with synchrotron radiation for chemical and magnetic analysis via X-ray Absorption Spectroscopy and X-ray Magnetic Linear Dichroism. For different chosen stoichiometries, the growth leads to the formation of high quality 2D islands. The high crystalline and morphological quality of the prepared films result in optimized properties with respect to films grown by other methods. In particular, their magnetic domains are larger by several orders of magnitude. By means of vectorial magnetometry, the spin axis orientation has been determined with nanometer spatial resolution and found to depend on the Ni:Co ratio.

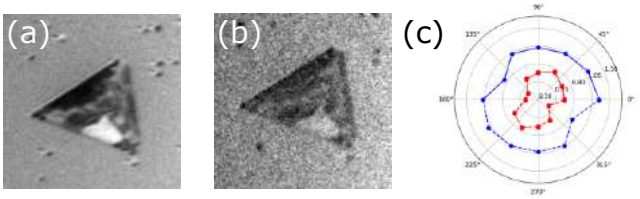



**Figure 1**. XMLD PEEM images obtained at (a) Co and (b) Ni absorption edges. (c) Polar plots of the experimental $L_2$ ratio extracted from a single-domain area for Co (red) and Ni (blue).

**Acknowledgements:** This research was funded by the SciMat Priority Research Area budget under the program "Excellence Initative - Research University" at the Jagiellonian University in Kraków - Grant No. 75.1920.2021, by Grant PID2021-124585NB-C31 funded by MCIN/AEI/10.13039/501100011033, and by "ERDF A way of making Europe" and by Grant TED2021-130957B-C54 funded by MCIN/AEI/10.13039/501100011033, and by the "European Union NextGenerationEU/PRTR".

[1] D. Alders et al. *Temperature and thickness dependence of magnetic moments in NiO epitaxial films*. Phys. Rev. B 57, 11623 (1998). DOI:10.1103/PhysRevB.57.11623.
[2] S. D. Peacor and T. Hibma. *Reflection high-energy electron diffraction study of the growth of NiO and CoO thin films by molecular beam epitaxy*. Surf. Sci. 301, 11 (1994). DOI:10.1016/0039-6028(94)91283-1.

# Nonrelativistic Spin Currents in Altermagnets

*Rodrigo Jaeschke-Ubiergo[1]*, V.K. Bharadwaj[1], Tomaš Jungwirth[2], Jairo Sinova[1], Libor Šmejkal[1]*

[1] *Institut für Physik, Johannes Gutenberg Universität Mainz, 55128, Mainz, Germany.*

[2] *Institute of Physics, Czech Academy of Sciences, Cukrovarnická 10, 162 00, Praha 6, Czech Republic.*

*rjaeschk@Uni-mainz.de

Altermagnetism has emerged recently as a third basic collinear magnetic phase [1], in addition to ferromagnets and antiferromagnets. Conventional antiferromagnets exhibit two sublattices with opposite magnetic moments related by translation or inversion. In altermagnets, the magnetic sublattices are connected by a rotation or a mirror operation. The particular symmetry causes altermagnets to display time-reversal (T) symmetry breaking and spin-split band structure even in the absence of spin-orbit coupling [2].

In this work, we study the spin conductivity tensor in altermagnets by using spin group theory formalism [1]. We also use Kubo's linear response to calculate the spin conductivity tensor in all the altermagnetic spin point group models. Additionally, we identify and sort 200 altermagnetic candidates into spin conductivity tensor classes. We will discuss some spin point groups that allow for a transverse spin current in detail. This is the case of spin splitter current in RuO2 [3,4], a nonrelativistic effect that conserves spin unlike in general magnetic spin Hall effect in noncollinear magnets. Moreover, the spin conductivity tensor is symmetric and T-odd, which makes it different from the conventional spin Hall effect.

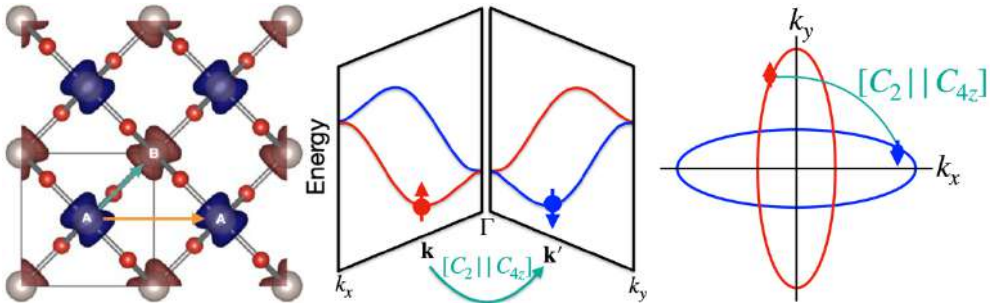



Figure: (left) Magnetization density $RuO_2$, from [4]. (center) Spin split bands in a d-wave altermagnet. (right) Fermi surfaces in a d-wave altermagnet.

**Acknowledgements**

This work is supported by SPIN + X (Grant No. DFG SFB TRR 173) and Elasto-Q-Mat (Grant No. DFG SFB TRR 288). We acknowledge the computing time granted on the supercomputer Mogon at Johannes Gutenberg University Mainz.

**References**

[1] Šmejkal et al., arXiv:2105.05820 Physical Review X, 12, 031042 (2022).
[2] Šmejkal, et al. Sci. Adv. 6, eaaz8809 (2020).
[3] Gonzalez-Hernandez, et al. Physical Review Letters, 126, 127701 (2021).
[4] Šmejkal, et al. PRX 12, 011028 (2022).

# SYMPOSIUM 13.
## DOMAIN WALLS, SKYRMIONS AND SPIN-ORBIT RELATED PHENOMENA.
## S13 INVITED ORAL PRESENTATIONS

# Gate-Controlled Skyrmion and Domain Wall Chirality

*Charles-Élie Fillion[1], Johanna Fischer[1], Raj Kumar[1], Capucine Gueneau[1], Aymen Fassatoui[2], Stefania Pizzini[2], Laurent Ranno[2], Djoudi Ourdani,[3] Mohamed Belmeguenai[3], Yves Roussigné[3], Salim-Mourad Chérif[3], Stéphane Auffret[1], Isabelle Joumard[1], Olivier Boulle[1], Gilles Gaudin[1], Liliana D. Buda-Prejbeanu[1], Claire Baraduc[1], Hélène Béa[1,4]*

[1] *Univ. Grenoble Alpes, CNRS, CEA, SPINTEC, Grenoble, France*

[2] *Univ. Grenoble Alpes, CNRS, Institut NEEL, Grenoble, France*

[3] *Laboratoire des Sciences des Procédés et des Matériaux (LSPM), Villetaneuse, France*

[4] *Institut Universitaire de France (IUF), Paris, France*

*helene.bea@cea.fr

In ultrathin magnetic films with perpendicular magnetic anisotropy, an antisymmetric exchange interaction, called Dzyaloshinskii-Moriya Interaction (DMI) [1] may exist due to interfaces and spin-orbit coupling. DMI may stabilize Néel domain walls (DWs) with a given sense of rotation, called chirality. The magnetic bubbles in these samples are thus non-trivial and called magnetic skyrmions [2] (Fig. 1a,b). Skyrmions move under current due to spin-orbit torques [3], which make them interesting for spintronics. This motion depends on the DW texture and chirality, defined by DMI sign and amplitude.

We showed that a gate voltage can invert the chirality of magnetic skyrmions and DWs in Ta/FeCoB/$TaO_x$ trilayers by tuning the sign of the DMI coefficient D [4]. We used Brillouin Light Scattering to measure DMI coefficient and sign as well as polar magneto-optical Kerr effect microscope to track, through a transparent electrode, the current-induced motion of skyrmions and DWs and determine their chirality. We attribute this control of chirality to a tuning with the gate voltage of the oxidation state at the top FeCoB/$TaO_x$ interface, which has been shown to control DMI sign [5].

Micromagnetic simulations show that this chirality control may be downscaled to nanometric skyrmions. This local and dynamical reversal of the chirality paves the way to an efficient and individual control of skyrmions, enabling new functionalities for spintronic logic devices and memories.

**Acknowledgements**

French ANR (ELECSPIN ANR-16-CE24-0018, ADMIS ANR-19-CE24-0019), DARPA TEE program No. MIPR HR0011831554, Nanosciences Foundation, REA grant agreement no. PCOFUND-GA-2013-609102

**References**

[1] I. E. Dzyaloshinskii, *J. Exptl. Theoret. Phys*., **46** (1964) 960; T. Moriya, *Phys. Rev*. **120** (1960) 91
[2] A. Fert, et al., *Nature Review Materials*, **46** (2017) 17031
[3] A. Thiaville et al., *Europhys. Lett.,* **100** (2012) 57002 ; S. Emori et al., *Nat. Mater*.**12** (2013) 611
[4] C. E. Fillion, et al., *Nat. Commun.,* **13** (2022) 1
[5] R. Kumar et al., *Phys. Rev. Appl.,* **19** (2023) 024064; M. Arora, et al., *Physical Review B*, **101** (2020) 054421

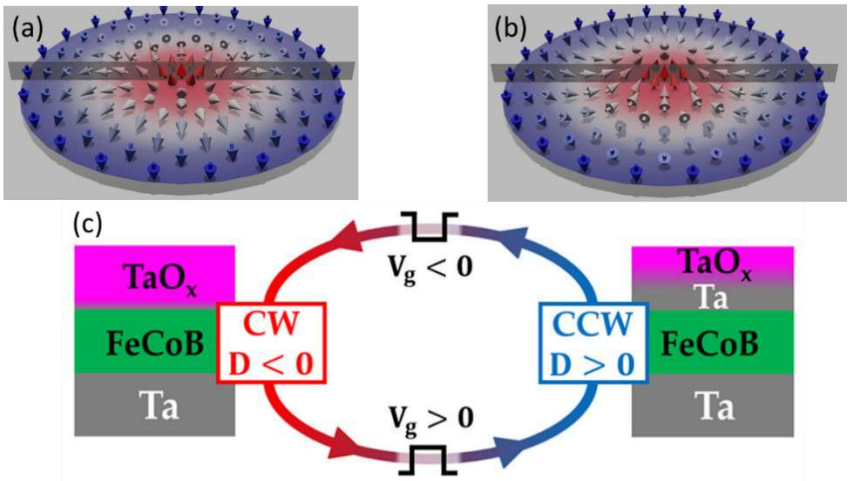



Figure: Representation of (a) clockwise (CW) and (b) counterclockwise (CCW) Néel skyrmions. (c) Effect of a gate voltage on skyrmion and domain wall chirality, induced by oxygen ion migration effects in Ta/FeCoB/$TaO_x$ stack.

# Crucial Role of Long-Range Orbital Angular Momentum Response and Transport in Spintronic Phenomena

*Dongwook Go*[1,2]*

[1] *Peter Grünberg Institut and Institute for Advanced Simulation, Forschungszentrum Jülich and JARA, 52425 Jülich, Germany*

[1] *Institute of Physics, Johannes Gutenberg University Mainz, 55099 Mainz, Germany*

*d.go@fz-juelich.de

Within the paradaigm of spintronics so far, most spintronic phenomena such as current-induced magnetic torques are interpreted in terms of spin-transfer physics and transport of angular momentum in the form of spin currents. At a fundamental level, however, the angular momentum of electrons is not only carried by the spin degree of freedom but also by the orbital degrees of freedom, but the possibility of transport and transfer of angular momentum via the orbital channel has been overlooked so far. In recent years, we have shown that even if the orbitals in the ground state is entirely quenched, OAM can be induced and transported via non-equilibrium states driven by external perturbations such as electric and magnetic fields [1,2]. The induced OAM can also be transferred to local moments to exert a torque, which offers a promising route for enhancing the efficiency of spin torque devices [3]. Despite many promising features of OAM for spintronics, one of the biggest challenges is to unambigiously quantify it, and in particular, disentangle the signals coming from the spin and orbital, whose phenomenological behaviors are nearly identical.

In this talk, I will show that nonequilibrium OAM exhibits long-range correlation in its response and transport, which are different from the behavior of the spin both quantitatively and qualitatively. I will show that injection of OAM in a ferromagnet exerts a torque on the local moment over unexpectedly long distance [4]. The injected OAM decays monotonically without any oscillation, which is distinct from the dephasing of spins in the mechanism of the spin-transfer torque. I will explain that this behavior originates from the momentum-space hotspots where Bloch states with different orbital characters are degenerate, which is imposed by the symmetry of the crystal field. This results in the nontrivial behavior of the current induced torque: the torque efficiency increases as the thickness of the ferromagnet grows. This prediction was verified by a recent experiment by Hayashi *et al.* [5], which found long (~ 20 nm ) orbital dephasing length in Ni by magnetic torque measurements. The experiemnt also revealed an strikingly large orbital relaxation length ~ 70 nm in W. The long-range transport of the OAM in W was also experimentally verified by Seifert *et al.* [6], where the OAM transport triggered by a THz pulse was directly observed in the time domain. I will discuss the physical origin of the robustness of the OAM transport and its exceptionally long relaxation length. I will also discuss outlook for utilizing long-range OAM for spintronic and orbitronic device applications.

**Acknowledgements**

We acknowledge the fundings by the Deutsche Forschungsgemein- schaft (DFG, German Research Foundation) − TRR 173/2 − 268565370 (project A11), TRR 288 − 422213477 (project B06).

**References**

[1] D. Go, D. Jo, C. Kim, and H.-W. Lee, Phys. Rev. Lett. **121**, 086602 (2018).
[2] D. Go, D. Jo, H.-W. Lee, M. Kläui, and Y. Mokrousov, Europhys. Lett. **135**, 37001 (2021).
[3] D. Go and H.-W. Lee, Phys. Rev. Research **2**, 013177 (2020).
[4] D. Go, D. Jo, K.-W. Kim, S. Lee, M.-G. Kang, B.-G. Park, S. Blügel, H.-W. Lee, and Y. Mokrousov, arXiv: 2106.07928.
[5] H. Hayashi, D. Jo, D. Go, T. Gao, S. Haku, Y. Mokrousov, H.-W. Lee, and K. Ando, Comm. Phys. **6**, 32 (2023).
[6] T. S. Seifert, D. Go, H. Hayashi, R. Rouzegar, F. Freimuth, K. Ando, Y. Mokrousov, and T. Kampfrath, arXiv:2301.00747.

# Topological Magnetic Structures and their Dynamics

*Karin Everschor-Sitte*

[1] *Faculty of Physics and Center for Nanointegration Duisburg-Essen (CENIDE), University of Duisburg-Essen, 47057 Duisburg, Germany*

*karin.everschor-sitte@uni-due.de

The current-induced dynamics of magnetic structures is quite complex. For example, magnetic skyrmions allow for 'banana kicks' in magnetism, i.e., not only a motion of the skyrmions along but also transverse to the current direction. This effect, which has become known as the skyrmion Hall effect [1,2,3], is often disruptive for device applications. In this talk, we will present possibilities of how to eliminate the skyrmion Hall effect [4,5]. As a particular example, we will discuss helical phases which provide confined one-dimensional channels for high-speed skyrmion motion. We discuss how skyrmions can be generated in such helical backgrounds and analyze their stability [6].

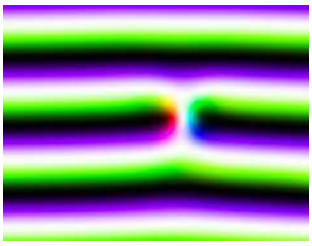


**Fig. 1:** *Skyrmion in a helical background [6].*

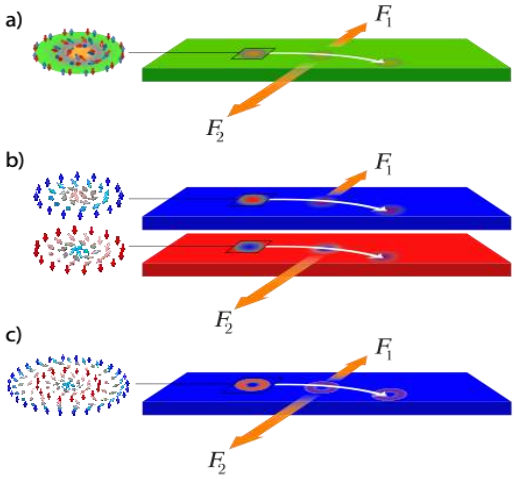



**Fig. 2:** *Net Skyrmion Hall effect in topologically neutral skyrmionics structures [7].*

Moreover, we will address the role played by topology in the physics of the skyrmion Hall effect. For example, it is widely believed that the skyrmion Hall effect, vanishes for overall topologically neutral structures such as (synthetic) antiferromagnetic skyrmions and skyrmioniums due to a compensation of Magnus forces. While this is true for spin-transfer torque-driven skyrmions, we show that this simple picture is generally false for spin-orbit torque-driven objects [7]. We find that the skyrmion Hall angle for spin-orbit torque-driven skyrmions is directly related to their helicity, which imposes an unexpected roadblock for developing faster and lower input racetrack memories based on spin-orbit torques.

**Acknowledgements**

The financial funding from DFG (Projects 320163632, 403233384, 268565370 (B12)) and the Emergent AI Center funded by the Carl-Zeiss-Stiftung is gratefully acknowledged.

**References:**

[1] T. Schulz, et al., *Nat. Phys.* **8**, 301 (2012)
[2] K. Litzius, et al., *Nat. Phys.* **13**, 170 (2017)
[3] W. Jiang, et al., *Nat. Phys.* **13**, 162 (2017)
[4] R. Zarzuela, et al., *Phys. Rev. B* **101**, 054405 (2020)
[5] K.-W. Kim, et al., *Phys. Rev. B* **97**, 224427 (2018)
[6] R. Knapman, et al., *J. Phys. D: Appl Phys.* **54**, 404003 (2021)
[7] R. Msiska, et al., *Phys. Rev. Appl.* **17**, 064015 (2022)

# Dynamic Investigation of Magnetic Skyrmions in the 2D van der Waals Magnet $Fe_3GeTe_2$

*K. Litzius[1,*], L. Powalla[2,3], M. T. Birch[4], S. Wintz[2,5], M. Burghard[2], M. Weigand[5], K. Kern[2,3], G. Schütz[5]*
[1]*Institute of Physics, University of Augsburg, 86149 Augsburg, Germany.*
[2]*Max-Planck-Institute for Solid State Research, 70569 Stuttgart, Germany.*
[3]*École Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland.*
[4]*Max-Planck-Institute for Intelligent Systems, 70569 Stuttgart, Germany.*
[5]*Helmholtz-Zentrum Berlin für Materialien und Energie, 12489 Berlin, Germany.*
*corresponding author email address

The vast versatility of 2-dimensional (2D) magnetic van der Waals materials has attracted immense interest within recent years. The biggest advantage of this material class is its ability to form versatile heterostructures that can be tailored towards a broad spectrum of properties, ranging from spintronic to optomagnetic and electrical applications. It is also capable of hosting chiral topological spin textures, such as skyrmions, interesting, e.g., for spintronic data storage applications. Unique to this class of materials is their mechanical flexibility and the option to exfoliate perfectly crystalline sheets on pre-pattered substrates. This offers the prospect of highly efficient low-dimensional devices and extreme ease to fabricate versatile heterostructures by stacking separate individual layers.

In this work, we utilize real-space imaging of the magnetic texture in thin flakes of the 2D van der Waals magnet $Fe_3GeTe_2$ (FGT) to characterize the dynamics of skyrmions in trilayer devices consisting of an FGT sheet capped by hexagonal boron nitride (hBN) and a graphite electrode to inject currents through a vertical nanocontact (see figure). With this device we realize the local injection of skyrmions and investigate dynamical effects of the generation process. Ultimately, the choice of composition and nucleation mechanism result in a selective stabilization of individual skyrmions that can then be manipulated by targeted current pulses. Our findings thus open novel perspectives and strategies for designing van der Waals heterostructure-based devices.

**References:**

[1] Powalla, L. et al. Nano Lett. (published at 10.1021/acs.nanolett.2c01944, 2022).
[2] Birch, M. T. et al. Nat. Commun. 13, 3035 (2022).

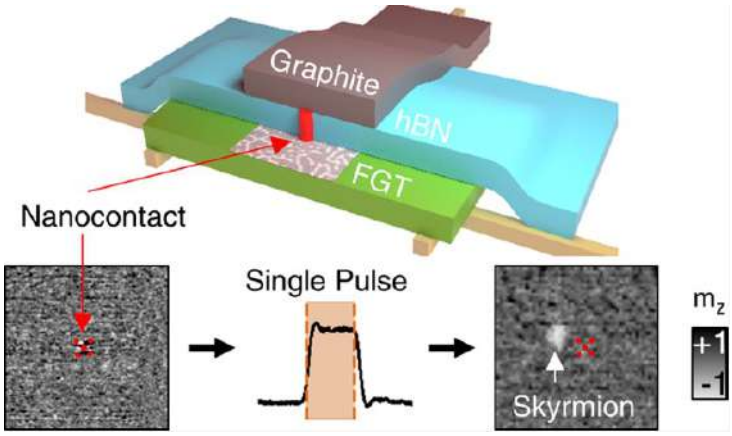



Figure: FGT flake heterostructure device with vertical interlayer nanocontact and lateral connections for current injection. The device consists of an FGT/hBN/graphite trilayer with the nanocontact marked in red. [1]

# SYMPOSIUM 13.
## DOMAIN WALLS, SKYRMIONS AND SPIN-ORBIT RELATED PHENOMENA.
## S13 ORAL PRESENTATIONS

# A Review on the Measuring Methods of the Interfacial Dzyaloshinskii-Moriya Interaction in Ultra-Thin Magnetic Films

*M. Kuepferling[1*], A. Casiraghi[1], G. Soares[1], G. Durin[1],*
*F. Garcia-Sanchez[2], L. Chen[3], C. H. Back[3], C. H. Marrows[4], S. Tacchi[5], and G. Carlotti[6]*

[1] *Istituto Nazionale di Ricerca Metrologica, Strada delle Cacce 91, 10135, Torino, Italy*
[2] *Departamendo de Fisica Aplicada, Universidad de Salamanca, Plaza de la Merced s/n 37008, Salamanca, Spain*
[3] *Technical University Munich, James-Frank-Str. 1, 85748 Garching, Germany*
[4] *School of Physics and Astronomy, University of Leeds, Leeds LS2 9JT, United Kingdom*
[5] *CNR, Istituto Officina dei Materiali - Perugia, c/o Dipartimento di Fisica e Geologia, Università Perugia, Via A. Pascoli, 06123 Perugia, Italy*
[6] *Dipartimento di Fisica e Geologia, Università di Perugia, Via Pascoli, 06123 Perugia, Italy*
*m.kuepferling@inrim.it

We aim to present our recent review [1] of the state-of-the-art of measuring the coefficient of the Dzyaloshinskii-Moriya interaction (DMI), the DMI constant *D*, focusing on systems where the interaction arises from the interface between two materials (i.e. interfacial DMI). The DMI, being one of the origins of chiral magnetism, is currently attracting considerable attention in the research community focusing on applied magnetism and spintronics. For future applications, we believe that an accurate measurement of its strength is indispensable. The review presents the experimental techniques as well as their theoretical background and models for the quantification of the DMI constant. The measurement techniques are divided into three categories: a) domain wall-based measurements, b) spin wave-based measurements and c) spin-orbit torque-based measurements. The advantages and disadvantages of each method and comparison of the *D* values at different interfaces are presented. In particular, we focus on the two most popular stacks reported in the literature, i.e. Pt/Co/X (with X a metal or an oxide) and X/CoFeB/MgO (with X a metal), as shown in the figures. The review aims to obtain a better understanding of the applicability of the different techniques to various stacks and of the origin of apparent disagreements among literature values.

## References

[1] M. Kuepferling, A. Casiraghi, G. Soares, G. Durin, F. Garcia-Sanchez, L. Chen, C. H. Back, C. H. Marrows, S. Tacchi, and G. Carlotti, *Rev. Mod. Phys* **95** (2023) accepted. (preprint: https://arxiv.org/abs/2102.00173)

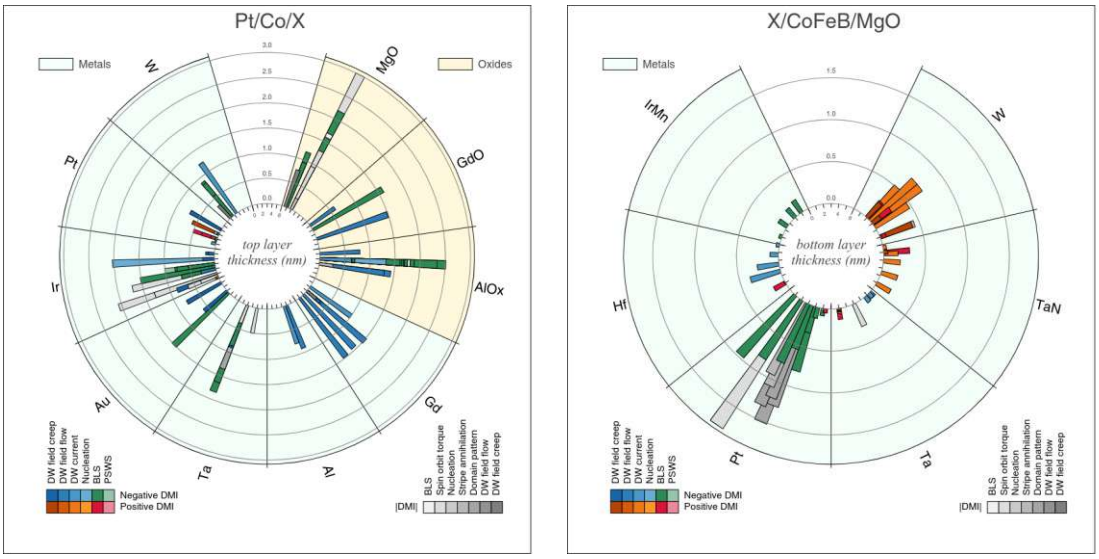



Figure: Measurements of the DMI constant $D_s = Dt$ (with t the thickness of the ferromagnetic layer) in Pt/Co/X (with X a metal or an oxide, left) and X/CoFeB/MgO (with X a metal, right) reported in the literature.

# Enhancing domain wall motion in W-CoFeB-MgO materials using $He^+$ ion irradiation

J.W. Van Der Jagt[1], L.Herrera Diez[2], A.Thiaville[3], V.Jeudy[3], L. Lopez Diaz[4], M.Fattouhi,[4] A.Lamperti[5], M.Belmeguenai[6], E.Monteblanco[1], B.Borie[1], R.Juge[1] and D.Ravelosona[1,2]

[1]*Spin-Ion Technologies, 91120 Palaiseau, France*

[2]*Centre de Nanosciences et de Nanotechnologies, CNRS, Université Paris-Saclay, 91120 Palaiseau, France*

[3]*Laboratoire de Physique des Solides, Université Paris-Saclay, CNRS, Orsay 91405, France*

[4]*Departamento de Física Aplicada, Universidad de Salamanca, Salamanca 37008, Spain*

[5]*Consiglio Nazionale delle Ricerche (CNR), Agrate Brianza, 20864, Italy*

[6]*Laboratoire des Sciences des Procédés et des Matériaux, CNRS, Université Sorbonne Paris Nord, Villetaneuse 93430, France*

Spintronic devices based on domain wall (DW) motion offer new exciting opportunities for non-volatile data storage, neuromorphic, and logic applications. Pinning of DWs due to structural disorder limits the efficiency of DW motion based devices. Such disorder in materials and devices usually takes the form of spatial variation of magnetic properties due to interface roughness, intermixing, crystalline texture or grain boundaries as well as edge defects induced by nanofabrication processes. $He^+$ irradiation is a powerful tool to engineer magnetic materials at the atomic scale enabling the control of magnetic properties [1]. The utilization of light ions provides the precise control of inter-atomic displacements through low energy transfer. In this work, we use He ion irradiation to reduce domain wall pinning in W-CoFeB-MgO systems with perpendicular anisotropy by (i) enhancing the crystallization process [2] and (ii) locally tuning the edges of micro sized wires. We have studied domain wall motion in W/CoFeB/MgO thin films with perpendicular magnetic anisotropy crystallized by annealing at 400◦C and a process based on He+ irradiation combined with moderated temperatures. We show that despite similar magnetic properties (effective anisotropy, magnetization, Gilbert damping, Dzyaloshinskii-Moriya interaction) for the whole series of samples, domain wall mobility is critically improved in the irradiated samples (see Figure 1). This is due to a smoother pinning potential with a narrower distribution of energy barriers for samples crystallized through ion irradiation. Finally, we have locally irradiated the edges of W-CoFeB-MgO wires, with widths between 5 µm and 40 µm. We demonstrate that the edge pinning can be quenched resulting in a significant increase in the DW velocity in the narrower wires.

In summary, the possibility to reduce structural defects through He ion irradiation paves the way toward power-efficient memory and neuromorphic devices.

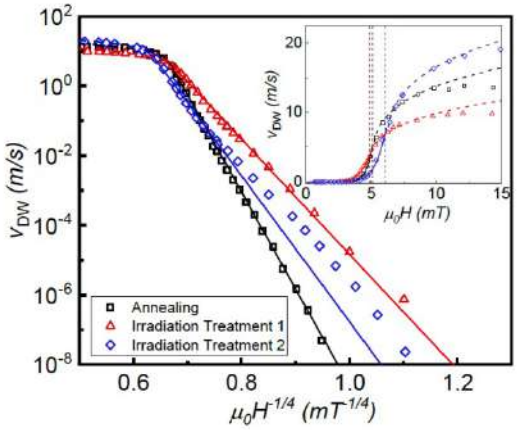



***Figure 1.*** *DW velocity as a function of the perpendicular magnetic field, rescaled to show creep law behavior. The solid lines are fits in the creep regime, and the dashed lines are fits in the depinning regime. The inset is a representation with linear axes around the depinning transition. The dotted vertical lines in the inset denote the depinning fields Hdep.*

**References**

[1] R.Juge, et al, Helium Ions Put Magnetic Skyrmions on the Track, Nano Letters 21, 2989, (2021)

[2] G. Van Der Jagt et al, Revealing Nanoscale Disorder in W/Co-Fe-B/MgO Ultrathin Films Using Domain-Wall Motion, Phys. Rev. Applied 18, 054072 (2022)

# Domain wall statics and dynamics in magnetic nanowires with arbitrary Dzyaloshinskii-Moriya tensors

*Adriano Di Pietro* [1,2] *, *Felipe García-Sánchez* [3], *Gianfranco Durin* [1]

[1] Istituto Nazionale di Ricerca Metrologica, Strada delle Cacce 91, 10135, Torino, Italy

[2] *Politecnico di Torino, Corso Duca degli Abruzzi 24, 10129, Torino, Italy*

[3] *Departamento de Fisica Aplicada, Universidad de Salamanca, Salamanca 37008, Spain*

*a.dipietro@inrim.it

The influence of different Dzyaloshinskii-Moriya interaction (DMI) tensor components [1] on the static and dynamic properties of domain walls (DWs) in magnetic nanowires is investigated using one dimensional collective coordinates models and micromagnetic simulations [2]. It is shown how the different contributions of the DMI can be compactly treated by separating the symmetric traceless, antisymmetric and diagonal components of the DMI tensor. First, we investigate the effect of all different DMI components on the static DW tilting in the presence and absence of in plane (IP) fields [3]. We discuss the possibilities and limitations of this measurement approach for arbitrary DMI tensors. Secondly, the interplay of different DMI tensor components and their effect on the field driven dynamics of the DWs is studied and reveals a non-trivial effect of the Walker breakdown field [4] of the material. It is shown how DMI tensors combining diagonal and off-diagonal elements can lead to a non-linear enhancement of the Walker field, in contrast with the linear enhancement obtainable in the usual cases (interface DMI or bulk DMI).

## Acknowledgements

This project has received funding from the European Union's Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement No. 860060 "Magnetism and the effect of Electric Field" (MagnEFi).

## References

[1] A. Di Pietro, P. Ansalone, V. Basso, A. Magni, and G. Durin. 2022. EPL 140, **4** (2022), 46003.
[2] A. Vansteenkiste, J. Leliaert, M. Dvornik, M. Helsen, F. Garcia-Sanchez, B.V. Waeyenberge. AIP Advances. 4**, 10** (2014) 107133.
[3] O. Boulle, S. Rohart, L.D. Buda-Prejbeanu, E. Jué, I.M. Miron, S. Pizzini, J. Vogel, G. Gaudin, A. Thiaville. PRL, 111, **21** (2013) 217203.
[4] S. Glathe, R. Mattheis, and D. V. Berkov. Applied Physics Letters 93, **7** (2008) 072508.

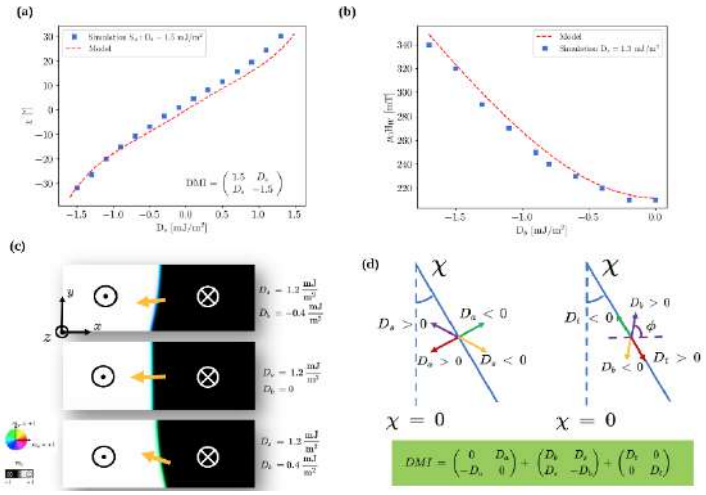



Figure 1: (a) DW tilting angle as a function of $D_s$ (b) Walker field $H_W$ as a function of $D_b$ (c) DW tiling in the absence of IP fields in three representative cases (d) Scheme of stabilized DW angles in the presence of the different DMI contributions shown in the green box below.

# Solid-State Lithium-Ion Supercapacitor for Voltage Control of Skyrmions

*Maria Ameziane, Joonatan Huhtasalo, Lukáš Flajšman, Rhodri Mansell, Sebastiaan van Dijken**

*Department of Applied Physics, Aalto University School of Science, Espoo, Finland*

*sebastiaan.van.dijken@aalto.fi

Ionic control of magnetism gives rise to high magneto-electric coupling efficiencies at low voltages [1-3], which is essential for low-power magnetism-based non-conventional computing technologies. However, for on-chip applications, magneto-ionic devices typically suffer from slow kinetics, poor cyclability, impractical liquid architectures or strong ambient effects. As a route to overcoming these problems, we demonstrate voltage control of a magnetic skyrmion state in an LiPON-based solid-state ionic supercapacitor with a 2 nm Ta/4 nm Pt/0.9 nm CoFeB/0.2 nm Pt/100 nm LiPON/1 nm SiN/4 nm Pt structure. Under the application of a positive voltage to the top electrode, skyrmions nucleate in the CoFeB film as a result of Li ion accumulation at the magnetic interface (Figure 1). Reversal of the voltage polarity results in skyrmion annihilation. Cyclic voltammograms (CVs) of the supercapacitor structure show a largely rectangular shape with no peaks indicative of redox processes. Instead, the CVs reveal the formation of an electric double layer at small voltage, followed by Li ion intercalation above +0.8 V. Because of the limited amount of Li ions migrating in the LiPON layer compared to the battery structures we studied previously [4], the system is highly cyclable and fast. Single 60-µs voltage pulses already trigger skyrmion nucleation while the supercapacitors can be cycled 750,000 times without loss of electrical performance. Our results demonstrate a simple and robust approach to ionic control of magnetism in spin-based devices.

**Acknowledgements**

This work was supported by the Academy of Finland (Grant No. 316857).

**References**

[1] M. Nichterwitz, S. Honnali, M. Kutuzau et al., *APL Mater.* **9** (2021) 030903.
[2] Y. Gu, C. Song, Q. Wang et al., *APL Mater.* **9** (2021) 040904.
[3] J. de Rojas, A. Quintana, G. Rius et al., *Appl. Phys. Lett.* **120** (2022) 070501.
[4] M. Ameziane, R. Mansell, V. Havu, P. Rinke, S. van Dijken, *Adv. Funct. Mater.* **32** (2022) 2113118.

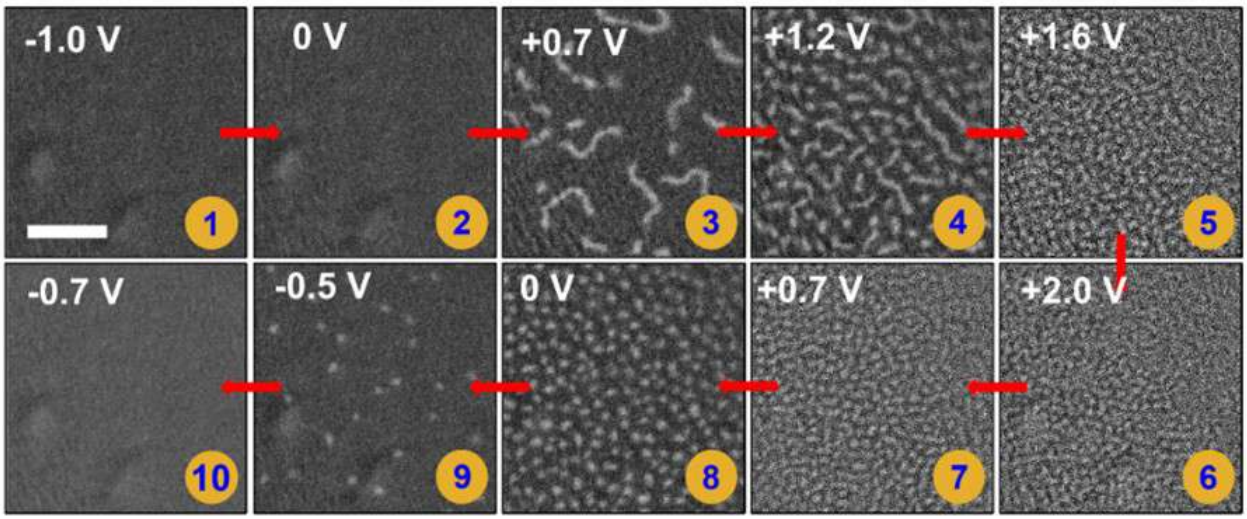



Figure 1: Voltage control of the skyrmion state in a LiPON-based solid-state ionic supercapacitor. Polar MOKE microscopy images recorded while sweeping the applied voltage from –1.0 V to +2.0 V and back. The perpendicular magnetic field is +0.7 mT. The scalebar corresponds to 10 µm.

# Near Landauer Reversible Skyrmion Logic with Voltage-Based Propagation

*B. W. Walker[1], F. Garcia-Sanchez[2]*, A. J. Edwards[1], X. Hu[1], M. P. Frank[3], J. S. Friedman[1]*

[1]*Department of Electrical & Computer Engineering, The University of Texas at Dallas, Richardson, Texas 75080, USA*

[2]*Departamento de Física Aplicada, Universidad de Salamanca, 37008 Salamanca, Spain*

[3]*Cognitive and Emerging Computing Department, Center for Computing Research, Sandia National Laboratories, Albuquerque, NM 87123 USA.*

*fgs@usal.es

Due to their particle like character and their particle-particle repulsion and their detectability, magnetic skyrmions have been proposed as a potential candidate for conservative logic [1-2]. The ideal realization for such logic implementation is the billiard ball logic proposed by E. Fredkin and T. Toffoli [3]. In the initial proposals for the skyrmion conservative logic, the skyrmion was propagated to produce logic operations using current induced torques [1-2]. In this work, we propose a scheme to produce skyrmion motion driven only by voltage using the voltage controlled magnetic anisotropy (VCMA) effect [4]. This modification will reduce drastically the energy consumption due to the reduced Joule dissipation. The magnetic device and the stack that produce such motion is shown in the figure. It consists of a time varying VCMA, which modifies the perpendicular anisotropy, with three phases in three consecutive tracks as depicted in Figure (a) and (c). The motion was confirmed with micromagnetic simulations as shown in the Figure (d). The implementation of the different basic logic gates was analysed using simulations.

To check the performance of such devices, we quantified the total dissipation of a device, including the magnetic and electrical dissipation, and found that the minimum dissipation of this VCMA-driven magnetic skyrmion logic at 0 K is found to be ~6× the room-temperature Landauer limit [5], indicating the potential for sub-Landauer dissipation through further engineering.

**Acknowledgements**

This work is supported in part by the Advanced Simulation and Computing (ASC) program at the U.S. Department of Energy's National Nuclear Security Administration (NNSA), by the National Science Foundation of USA under CCF award 1910800 and by project PID2020117024GB-C41 funded by Ministerio de Ciencia e Innovacion from the Spanish government.

**References**

[1] M. Chauwin et al., *Phys. Rev. Applied* **12** (2019) 064053.
[2] B. W. Walker et al., *Appl. Phys. Lett.* **118** (2021) 192404.
[3] E. Fredkin and T. Toffoli, *Int. J. Theor. Phys.* **21** (1982) 219.
[4] B. W. Walker et al., https://arxiv.org/abs/2301.10700.
[5] R. Landauer, *IBM J. Res. Dev.* **5** (1961) 183.

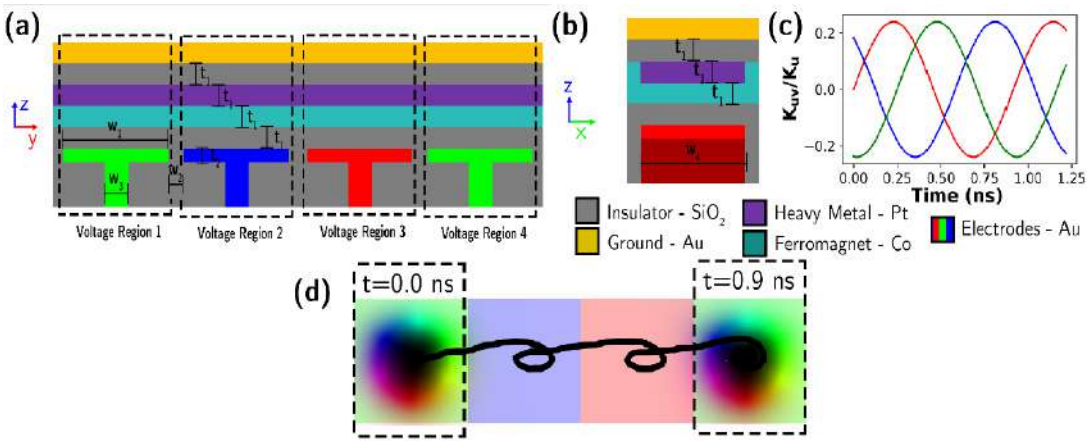



Figure: Scheme of the proposed device and implementation of skyrmion motion using pulses of VCMA (a) YZ and (b) XZ Device Cross Section (c) VCMA anisotropy waveform applied to electrodes of corresponding color. (d) Micromagnetic simulation of skyrmion propagation. The black path indicates skyrmion trajectory.

# Enhancement of Spin-Transfer-Torque and Spin-Orbit-Torque effects in magnetic core-shell nanotubes

*J. Hurst[1], C. Thirion[2], O. Fruchart[1], J.-C. Toussaint[2], Daria Gusakova[1]**

[1] *Univ. Grenoble Alpes, CNRS, CEA, Grenoble INP, Spintec, Grenoble, France*

[2] *Univ. Grenoble Alpes, CNRS, NEEL Institute, Grenoble, France*

*daria.gusakova@cea.fr

Three-dimensional spintronics exploits the interaction of magnetic moments with electron spins in non-planar objects, such as cylindres. These 3D building blocks of nanoscale devices are fascinating objects for fundamental research as well as for data storage advanced technologies. In particular, the information stored in a magnetic wire or tube, should be encoded by magnetic domains separated by magnetic domain walls which motion may be achieved by applying spin-polarized current. In addition to conventional single material wires, recent progress in nanofabrication gives rise to a new variety of multi-layered core-shell geometries giving access to Spin-Transfer-Torque (STT) and Spin-Orbit-Torque-induced (SOT) driving forces (Figure 1).

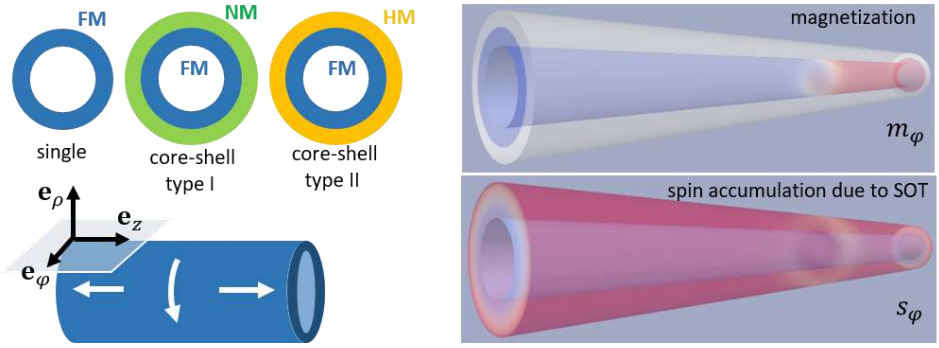



Figure 1: Left column: studied tubular geometries including STT and SOT effects. Right column: magnetization distribution and corresponding spin accumulation induced in core/shell ferromagnet/heavy metal structure.

To simulate non-trivial 3D micromagnetic textures and the impact of current on its dynamics we have developed the multi-physics finite element C++ software *feeLLGood* which is suitable for irregular or curved geometries [1,2,3]. Previous version of the code included self-consistent calculation of the magnetization and spin dynamics resulting in non-trivial Spin-Transfer-Torque (STT) effects within single ferromagnetic material. Our recent interest focuses on the SOT-induced effects and corresponding driving force acting on the domain wall within ferromagnet/heavy metal bilayers.

In the presentation we discuss the details of the domain wall current-induced motion under spin polarized current including STT and SOT effects. We characterise and compare STT and SOT efficiency as a function of geometry type and spin transport characteristic lengths, and highlight the differences with the usual planar geometry. In addition we also quantify the interplay between STT and SOT effects within the same structure.

## Acknowledgements

This study was partially supported by French National Research Agency (JCJC MATEMAC-3D, DIWINA).

## References

[1] http://feellgood.neel.cnrs.fr/

[2] M. Sturma et al, Phys. Rev. B **94**, 104405 (2016).

[3] J. Hurst et al. Phys. Rev. B **103**, 024434 (2021).

# Spin Orbit Torque for Field-Free Switching in $C_{3v}$ Crystals

*Diego García Ovalle[1]*, Armando Pezo[1], Aurélien Manchon[1]*

[1] *Aix-Marseille Université, CNRS, CINaM, Marseille, France*

*diego-fernando.garcia-ovalle@univ-amu.fr

In this work [1] we characterized the spin-orbit torques permitted by symmetry in trigonal crystals, revealing the fundamental role of the cubic order correction to the (Rashba) spin-orbit coupling and the trigonal warping of the Fermi surface to observe such responses. In particular, we demonstrate the microscopic origin of the so-called "3m" torque, which was previously reported in CuPt/CoPt [2] heterostructures and $Fe_3GeTe_2$ [3], and that has the ability to promote field-free switching of the magnetization in ferromagnetic interfaces. To do so, we firstly derived the effective fields in crystals with $C_{3v}$ point group symmetry by means of the invariant theory [4], looking up to first order in electric field and third order in magnetization components. Then, we corroborate our predictions with tight-binding calculations in a hexagonal lattice, where we consider linear and cubic spin-orbit coupling terms. Finally, we verify our findings with realistic calculations in CuPt/Co, which exhibits strong trigonal warping as it is depicted in Fig.1, and showing that the "3m" contribution can be as large as the regular damping-like torque in these systems. Our article motivates further exploration of low-symmetry crystals for magnetic memory devices.

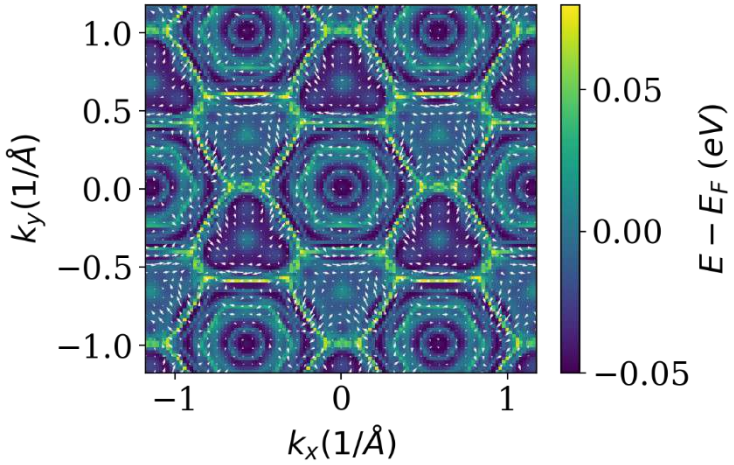



Fig. 1. (Color online) Spin texture of a selected band close to the Fermi level of CuPt(111)/Co slab geometry.

## Acknowledgements

This work was supported by the ANR ORION project, grant ANR-20-CE30-0022-01 of the French Agence Nationale de la Recherche. D. G.O. and A. M. acknowledge support from the Excellence Initiative of Aix-Marseille Universite - A*Midex, a French "Investissements d'Avenir" program.

## References

[1] D. G. Ovalle, A. Pezo, and A. Manchon, "Spin-orbit torque for eld-free switching in $C_{3v}$ crystals," 2023. [Online]. Available: https://arxiv.org/abs/2301.01133. *Submitted for publication.*

[2] L. Liu, C. Zhou, X. Shu, C. Li, T. Zhao, W. Lin, J. Deng, Q. Xie, S. Chen, J. Zhou, R. Guo, H. Wang, J. Yu, S. Shi, P. Yang, S. Pennycook, A. Manchon, and J. Chen, "Symmetry- dependent field-free switching of perpendicular magnetization," *Nature Nanotechnology*, vol. 16, no. 3, pp. 277–282, Mar 2021. [Online]. Available: https://doi.org/10.1038/s41565-020-00826-8

[3] O. Johansen, V. Risinggård, A. Sudbø, J. Linder, and A. Brataas, "Current control of magnetism in two-dimensional $Fe_3GeTe_2$," *Phys. Rev. Lett.*, vol. 122, p. 217203, May 2019. [Online]. Available: https://link.aps.org/doi/10.1103/PhysRevLett.122.217203

[4] M. J. Lax, *Symmetry Principles in Solid State and Molecular Physics*. Dover, UK: Dover Publications, 2012.

# Internal Twist of Domain Walls Driven by Spin-Orbit Torque Under Uniform Strain

*Mouad Fattouhi[1,]*, André Thiaville[2], Felipe García Sánchez[1], Eduardo Martínez[1] and Luis López-Diaz[1]*

[1]*Departamento de Física Aplicada, Universidad de Salamanca, 37008, Salamanca, Spain*
[2]*Laboratoire de Physique des Solides, Université P-Saclay, CNRS, Orsay 91405, France*
*mfa@usal.es

The internal dynamics of magnetic domain walls (DWs) driven by magnetic fields and electric currents exhibit characteristic oscillatory instabilities, such as what happens above the Walker breakdown field [1]. Depending on the dimensions, theses instabilities can be uniform or space-dependent [2]. In this work, we show a different class of DW instabilities that are stationary in time and induced by Slonzcewski-like spin-orbit torque in the presence of uniform in-plane mechanical strain [3]. Using micromagnetic (μM) simulations, we observe two regimes depending on the current injected. For small current densities, after a short transient the DW reaches a steady state regime and moves at a constant velocity. Above a certain current threshold ($J_c$) the DW stops moving completely and its internal magnetization modulates in space developing a characteristic ripple-like pattern like the one shown in Fig. 1, which cancels out the net torque over the DW. Using a 2D model for the DW internal angle and position, which can both vary along the width of the nanowire, together with μM simulations, we show that both the number of ripples and their curvature are given by the SOT current ($J_{SOT}$) and the nanowire width ($W$). These results are explained with analytical formulas that display their dependence on the material and geometrical parameters. In our view, our findings show that SOT-driven DWs in presence of in-plane strain could be a new platform for information storage in static memory elements as well as efficient energy manipulation.

## Acknowledgements

We gratefully acknowledge financial support from the European Union H2020 Program (MSCA MagnEFi ITN Grant No. 860060).

## References

[1] A. Malozemoff, and J. C. Slonczewski. Magnetic domain walls in bubble materials (Academic press, 1979).
[2] Y. Yoko, et al. *Nature Physics* **12**.2 (2016). 157-161.
[3] M. Fattouhi, et al. *Phys. Rev. Appl*. **18** (2022). 044023.

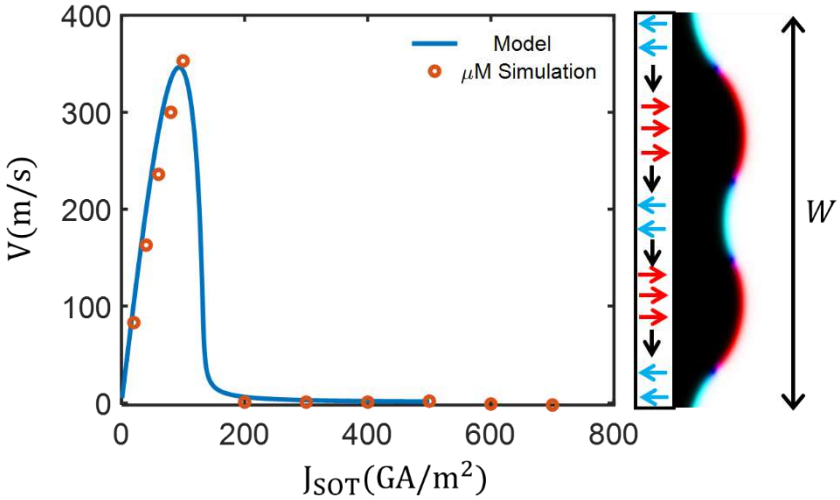



Figure: DW velocity versus current from both simulation (dots) and model (solid line). After a threshold current the DW completely stops, and a spatially modulated structure is stabilized as shown in the right part of the figure.

# Local $He^+$ Ion Irradiation on Spin-Orbit Torque Devices

*Aleena Joseph[1*], Jinu Kurian[1], Gregor Hlawacek[2], Salia Cherfi Hertel[1], Ciaran Fowley[2], Peter Dunne[1], Gwenaël Atcheson[3], M Venkatesan[3], J. M. D. Coey[3], Bernard Doudin[1]*

[1]*Université de Strasbourg, CNRS, IPCMS UMR 7504, 23 rue du Loess, F-67034 Strasbourg, France*

[2]*Institute of Ion Beam Physics and Materials Research, Helmholtz-Zentrum Dresden - Rossendorf, Bautzner Landstraße 400, 01328 Dresden, Germany*

[3]*AMBER and School of Physics, Trinity College, Dublin 2, Ireland*

* aleena.joseph@ipcms.unistra.fr

We illustrate here a promising approach for improved data storage and high-performance computation in Spin-Orbit Torque (SOT) -based spintronic devices by means of nm resolution ion beam microscopy that pattern a magnetic energy landscape triggering preferential current-driven magnetisation switching. SOT-induced magnetization switching devices have potential applications in the field of magnetic memory, nanomagnetic logic circuits and neuromorphic computing[1]. The critical current required to switch the magnetic state depends on saturation magnetization and magnetic anisotropy, which are highly interface dependent and can be tuned by $He^+$ irradiation [2]. We show that the critical current could be reduced by >80% by modifying the interface properties of Pt/Co/W-based thin films using a focused helium ion beam (fig.1a)[3]. Our key advancement is the i*n-situ* Hall measurement under local ion irradiation which allows us to monitor the evolution of PMA with ion dose (fig. 1a). Furthermore, we illustrate how multilevel switching can be achieved using an $He^+$ beam by partially irradiating the junction with appropriate doses (fig. 1b,c), chosen based on the *in-situ* irradiation curve[4]. Domain nucleation and domain wall motion [5] extend the portfolio of irradiation engineering of SOT devices towards an enabling patterning technology for new SOT device applications. Novel device fabrications and their spatial and time-resolved properties will be presented.

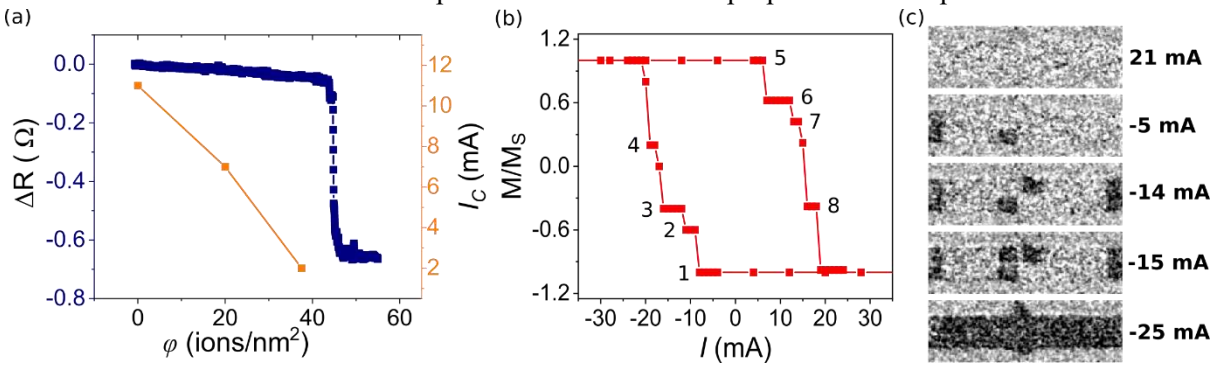



*Figure 1 (a) Evolution of in-situ anomalous Hall resistance and switching current with irradiation. (b) MOKE acquired SOT-induced magnetization switching curve of an 8-level switching device under a bias field of 125 mT. (c) MOKE images of the SOT induced magnetization switching of the junction when sweeping from 21 to –25 mA.*

## References

[1] A. Manchon *et al.*, *Rev. Mod. Phys.*, vol. 91, no. 3, p. 035004, Sep. 2019.
[2] C. Chappert *et al.*, *Science*, vol. 280, no. 5371, pp. 1919–1922, Jun. 1998.
[3] P. Dunne *et al.*, *Nano Lett.*, vol. 20, no. 10, pp. 7036–7042, Oct. 2020.
[4] J. Kurian *et al.*, *Appl. Phys. Lett.*, vol. 122, no. 3, p. 032402, Jan. 2023.
[5] A. Beguivin, D. C. M. C. Petit, R. Mansell, and R. P. Cowburn, *AIP Adv.*, vol. 7, no. 1, p. 015203, Jan. 2017.

# Electrical Detection and Nucleation of Magnetic Skyrmions in a Magnetic Tunnel Junction

*J. Urrestarazu Larrañaga[1], Naveen Sisodia[1], Florian Disdier[1], Van Tuong Pham[1], Liliana Buda-Prejbeanu[1], Ricardo Sousa[1], Gilles Gaudin[1], Sebastian Wintz[2], Markus Weigand[3] and Olivier Boulle[1]**

*[1]Univ. Grenoble Alpes, CEA, CNRS, Grenoble INP, SPINTEC, 38000 Grenoble, France*

*[2]Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, D-14109 Berlin, Germany*

*[3]Max Planck Institute for Intelligent Systems, Heisenbergstraße 3, 70569 Stuttgart, Germany*

*olivier.boulle@cea.fr

Magnetic skyrmions have recently attracted a large interest owing to their rich physics at the frontier of topology and magnetism and promising applications for non-volatile technological memory and logic devices. Skyrmions are local chiral whirlings of the magnetization texture with particle like properties, owing to their small lateral dimensions (down to the nanometer scale) and topological stability. They were recently demonstrated at room temperature in ultrathin heavy metal/ferromagnetic films as well as their fast manipulation by electrical current [1]. Those were first important steps toward technological applications where skyrmions in tracks are the information carriers. However, important challenges still need to be faced regarding the electrical detection and the low power nucleation of the skyrmions, which are required for the read and write operation in devices.

Here we demonstrate the nucleation by gate voltage and the electrical detection of a magnetic skyrmion in a magnetic tunnel junction (MTJ) using tunnelling magnetoresistance (TMR). To demonstrate this result, we combined scanning tunnelling magnetic microscopy (STXM) and *operando* magneto-transport measurements of a MTJ fabricated on top of an ultrathin SiN membrane. This allowed us to perform simultaneously high spatial resolution magnetic imaging of the spin texture within the MTJ and transport measurements to enable an unambiguous electrical detection of the magnetic skyrmion. Figure 1a shows the hysteresis loop of a 500nm diameter MTJ showing a TMR of 53% with sharp reversal and a resistance area product around 1.5kΩ.µm². Starting from the parallel magnetization resistance state with a uniform magnetization, the application of a voltage pulse (10ns) leads to a jump of the resistance (470Ω) to a stable intermediate resistance state. XMCD-STXM imaging shows that this new resistance state is associated with the nucleation of a skyrmion in the free layer of the MTJ (see figure 1b). Micromagnetic simulations using experimental parameters show that the nucleation can be explained by the transient nucleation of a vortex state, via voltage-induced decrease of the magnetic anisotropy, which eventually relaxes towards a skyrmionic state. These results demonstrate the read and write operations of a skyrmion based device and are an important milestone for low power applications based on the manipulation of magnetic skyrmions.

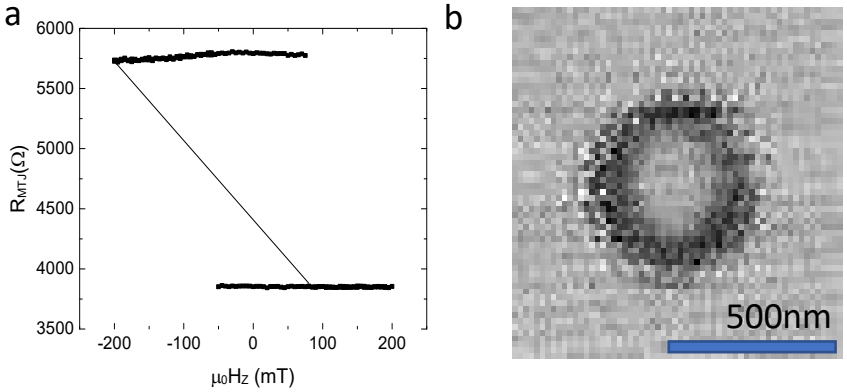



Figure 1: (a) Resistance vs out of plane magnetic field hysteresis loop of the device. (b) XMCD-STXM image of a magnetic skyrmion in a MTJ pillar of 500nm.

## References

[1] A. Fert, N. Reyren, and V. Cros, Nature Reviews Materials 2, 17031 (2017).

# Antiskyrmion-skyrmion hybrid strings

*Jan Masell*[1,2]*, *Fehmi S. Yasin*[2], *Xiuzhen Yu*[2]

[1]*Institute for Theoretical Solid State Physics, Karlsruhe Institute of Technology (KIT), Karlsruhe, 76049, Germany*

[2]*Center of Emergent Matter Science (CEMS), RIKEN, Wako, 351-0198, Japan*

* *jan.masell@kit.edu*

Magnetic skyrmions are nowadays found in a variety of different material classes, ranging from single crystals to sputtered films. They often emerge on length scales much larger than the atomic distance where they appear as vortex-like, rotationally symmetric whirls. Their anti-vortex-like partners, consequently dubbed *anti*-skyrmions, naturally break this rotational symmetry which leads to a plethora of new effects. However, only a limited number of antiskyrmion-hosting materials are known and, previously, they all belonged to the $D_{2d}$ symmetry class, even though crystals with $S_4$ symmetry were predicted to also host antiskyrmions.[1] Recently, we found thermodynamically stable antiskyrmions in $S_4$-symmetric doped schreibersite compounds, where they coexist as square-shaped solitons alongside elliptical skyrmions.[2,3]

In this talk, I will present our recent finding that antiskyrmions in 3D systems may not be pure antiskyrmion tubes but, in addition, feature skyrmion caps where they connect to the surfaces of the material. Extensive 3d micromagnetic simulations suggest that this topological transition in the soliton string is a quite ubiquitous phenomenon in soft magnets, where antiskyrmions gain Dzyaloshinskii-Moriya exchange energy in the bulk whereas Néel skyrmions on the surfaces are optimal in terms of magnetostatic energy. Tomographic reconstructions of field-free off-axis electron holography images of antiskyrmions in thin films of Pd-doped $(Fe,Ni)_3P$ indeed show these hybrid textures and confirm that there is a zoo of new 3D textures awaiting discovery.

## References

[1] A. N. Bogdanov, D. A. Yablonskii, *Sov. Phys. JETP* **68** (1989) 101-103.
[2] K. Karube, L. C. Peng, J. Masell, *et al. Nat. Mater.* **20** (2021) 335-340.
[3] K. Karube, L. C. Peng, J. Masell, *et al.* Adv. Mater. **34** (2022) 2108770.

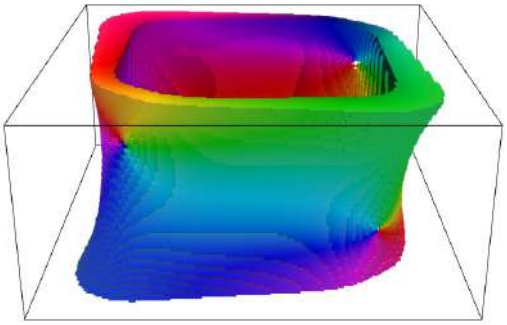


Figure: Hybrid antiskyrmion-skyrmion tube which is antiskyrmion in the bulk but exhibits Néel-skyrmion caps on the surfaces. Result of a micromagnetic simulation.

# Stabilization and switching of room temperature magnetic merons in epitaxial perpendicularly magnetized nanostructures

*Jose Peña-Garcia[1], Lorenzo Camosi[1,2], Stefania Pizzini[1], Aymen Fassatoui[1], Olivier Fruchart[3], Tevfik O. Mentes[4], Andrea Locatelli[4], André Thiaville[5], Stanislas Rohart[5], Jan Vogel[1]**

[1] *Univ. Grenoble Alpes, CNRS, Institut Néel, 25 Rue des Martyrs, Grenoble, France*

[2] *Catalan Institute of Nanoscience and Nanotechnology (ICN2), CSIC, BIST, Campus UAB, Barcelona, Spain*

[3] *Univ. Grenoble Alpes, CEA, CNRS, Spintec, Grenoble, France*

[4] *Elettra Sincrotrone, S.C.p.A, S.S 14 - km 163.5 in AREA Science Park, Basovizza, Trieste, Italy*

[5] *Univ. Paris-Saclay, CNRS, Laboratoire de Physique des Solides, 91405 Orsay Cedex, France*

*jan.vogel@neel.cnrs.fr

Magnetic thin films sandwiched between two different non-magnetic layers can show remarkable properties. The absence of structural inversion symmetry can give rise to an anti-symmetric exchange interaction, the Dzyaloshinsii-Moriya interaction (DMI), which favors an orthogonal alignment of neighboring magnetic moments. The competition with the Heisenberg exchange interaction, which favors a parallel alignment of the magnetic moments, can give rise to non-trivial magnetic textures : chiral domain walls, skyrmions or merons. Magnetic merons are topological objects which can be described as half-skyrmions. We have observed such magnetic merons in magnetic microstructures in epitaxial $Au_{0.58}Pt_{0.42}$/Co/W(110) trilayers using photoemission electron microscopy combined with x-ray magnetic circular dichroism (XMCD-PEEM) [1]. Magnetic merons are observed in nanostructures with diameters comparable to the spin-cycloid length of the system, but can also be found in a meta-stable state in much larger microstructures. We also show that the meron polarity can be switched by the application of a perpendicular magnetic field. We discuss the stability conditions of the magnetic merons and the possible effects of the microstructure edges on their formation.

**Acknowledgements**

JPG acknowledges a PhD Grant from GRECQUE and the Laboratoire d'excellence LANEF (Grant No. ANR-10-LABX-51-01). LC, JPG, SP and JV acknowledge financial support of the Agence Nationale de la Recherche, project ANR-17-CE24-0025 (TOPSKY).We also acknowledge financial support from the DARPA TEE program through Grant No. MIPR HR001183155 (SP and JV). We thank V. Guisset and P. David for their help with sample preparation.

**References:**

[1] Lorenzo Camosi1, Jose Peña-Garcia, Olivier Fruchart, Stefania Pizzini, Andrea Locatelli, Tevfik Onur Mentes, Francesca Genuzio, Justin M. Shaw, Hans T. Nembach and Jan Vogel, New J. Phys. 23 (2021) 013020

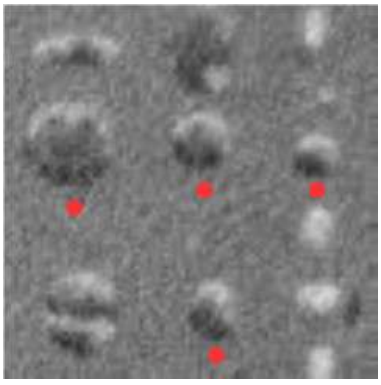


Figure 1: Magnetic image of micron-sized circular and elliptical dots of $Au_{60}Pt_{40}$/Co(0.78nm)/W(110), taken by XMCD-PEEM imaging. The structures marked with a red dot contain merons, with magnetization in the plane of Co layer at the borders (black and white contrast), and magnetization out of the Co plane in the center (dark grey).

# Field-Driven Chaos of Magnetic Skyrmions and its Potential Application to Dynamics-Based Computation

*Gyuyoung Park, and Sang-Koog Kim**

*Seoul National University, 1 Gwanak-ro, Gwanak-gu, Seoul, Republic of Korea*

*sangkoog@snu.ac.kr

Recent research on the nonlinear magnetization dynamics of various topological spin solitons has revealed the presence of chaos in nanoscale spin systems [1,2]. Despite the established inherent nonlinearity of magnetic skyrmions [3,4], their chaotic dynamics have received little attention compared to other topological spin solitons. In this study, we present micromagnetic simulation results of skyrmion chaos driven by external magnetic fields. We map the overall nonlinear dynamics of field-driven skyrmions over time through the calculation of the local Lyapunov exponents. In the chaotic regime, the skyrmion dynamics are highly sensitive to the initial conditions, while their stability or lifetime follows a power law. In the transition zone between the regular and chaotic regimes, also known as the edge-of-chaos, the skyrmion dynamics remain stable, similar to the regular regime, while the entropy is at its highest.

We explore the potential of the magnetic skyrmions performing simple and complex computations based on chaotic dynamics [5] by analyzing their chaotic maps for various initial conditions. This skyrmion chaos system executes all types of logic gates and morphs between them quickly. Furthermore, we show that the skyrmion dynamics at the edge-of-chaos are highly suitable for dynamics-based unconventional computing.

**Acknowledgements**

This research was supported by the Basic Science Research Program through the National Research Foundation of Korea (NRF) funded by the Ministry of Science, ICT and Future Planning (Grant No. 2021R1A2C2013543). The Institute of Engineering Research at Seoul National University provided additional research facilities for this work.

**References**

[1] T. Devolder et al., *Phys. Rev. Lett.* **123** (2019) 147701.
[2] L. Shen et al., *Phys. Rev. Lett.* **124** (2020) 037202.
[3] Y. Chen et al., *J. Magn. Magn. Mater.* **458** (2018) 123.
[4] D. R. Rodrigues et al., *Phys. Rev. Appl.* **16** (2021) 014020.
[5] S. Sinha, W. L. Ditto, *Phys. Rev. Lett.* **81** (1998) 10.

# Dynamics of Bistable Néel Domain Walls under Spin-Orbit Torque

*Eloi Haltz[1,2]*, Kévin J. A. Franke[1], Christopher H. Marrows[1]*

[1] *School of Physics and Astronomy, University of Leeds, Leeds LS2 9JT, United Kingdom*
[2] *LSPM-CNRS, UPR3407, Sorbonne Paris Nord University, Villetaneuse, France*
*eloi.haltz@univ-paris13.fr

The electrical manipulation of magnetic domain walls (DW) and their potential applications for data storage and processing devices have motivated an intense research in magnetism in the last decades. In particular, the archetypal magnetic layer / heavy metal (FM/HM) stacks are of great interest since the spin-orbit torque (SOT) generated in the HM layer acts on chiral textures in the FM layer such as Néel DW; this one being satabilized, with a given chirality, due to the asymmetry of the magnetic layer interfaces [1]. Recently, it has been proposed that an in-plane magnetic anisotropy can also stabilize Néel DW even in the absence of interface asymmetry [2-3]. The main difference with the classical case is the new bistable nature of the Néel DW where the two chiralities are energetically degenerate.

In this theoretical study [4], we explore SOT driven dynamics of such bistable Néel DW. An asymmetric trie-layer HM/FM/HM shaped with an appropriate wire-width is considered, allowing to stabilise bistable Néel DWs and drive there propagation under SOT. We find that, for a given up/down or down/up domain wall, two propagation directions along a nanowire are possible, depending on its initial state (shown in the Figure 1(a) for an up/down DW). These dynamics also exhibit complex dependence on the SOT magnitude, with a critical current (Jc) above which the DW is locked in its Bloch state and a maximum of velocity for an intermediate structure of the DW (shown in the Figure.1(b)). We show that the non-uniformity of the relation between the DW structure and its velocity leads to important transient regimes and non-Newtonian dynamics. Finally, we find that the chirality of the DW, and therefore its propagation direction, can be reversed under certain conditions: strong current pulses leading to the Brownian motion of the DW (shown in the Figure.1(c)), out-of-plan field inducing a quasi-random reversal of the DW propagation or, electrical-controlled step of magnetic anisotropy with a deterministic revesal or locking of the DW propagation.

A robust analytical model which handles all the obtained behaviors of such domain walls is developed and validated by comparing with numerical simulations. All these new dynamics of bistable Néel DWs open the way for new uses of DWs in information storage and processing devices.

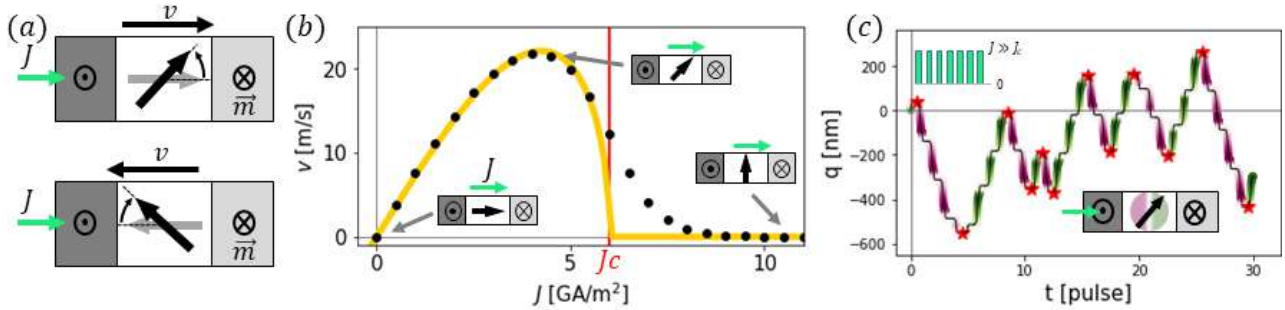



Figure 1: Dynamics of a bistable up-down Néel DW driven by SOT. (a) DW velocity and internal tilting driven by SOT, depending on the initial state (shown as a grey arrow). (b) Velocity curve of a bistable Néel DW driven by SOT obtained with micromagnetic simulations (black points) and our analytic model (yellow solid line). The insets show the evolution of the DW steady state when the current increases. (c) Brownian motion of a DW under a series of 20 GA/m² (>>Jc) 20 ns pulses with 20 ns delay. The color highlight indicate the DW structure and the red stars indicate when the DW reversal occurs.

## References

[1] A. Thiaville, S. Rohart, E. Jué, *et al.*, *EPL* **100** (2012) 57002.
[2] K. Franke , C. Ophus, A. Schmid *et al.*, *Phys. Rev. L* **127** (2021) 127203.
[3] B. Boehm, A. Bisig, A. Bischof, *et al. Phys. Rev. B* **95** (2017) 180406(R).
[4] E. Haltz, K. Franke, C. Marrows. *arXiv* (2023) 2303.06142

# SAW assisted Domain wall propagation

*A.Rivelles*, R. Izquierdo-López, M. Abuín, M. Maicas, M.M. Sanz and J.L. Prieto*

*ISOM, Universidad Politécnica de Madrid, , Av. Complutense 30, 28040, Madrid, Spain*

*alejandro.rivelles.garcia@upm.es

The field of spintronics is in a constant search for more efficient ways to manipulate the local magnetization in a ferromagnetic material without the need for an external magnetic field. Spin Transfer Torque (STT) offers the possibility of manipulating local magnetization with an electric current but often, at the expense of using a large current density, thus leading to detrimental thermal effects. Surface acoustic waves (SAW) are one of the candidates to solve these challenges. The stress carried by the SAW can affect domain wall (DW) movements and therefore alter local magnetization in ferromagnetic materials. In the literature, there are proposals to use SAW to control Domain Wall (DW) movement [1] and experimental results manifesting the effect of a SAW on the DWs [2] [3]. In this work we study how a SAW influences DW propagation on the basic unit of many spintronic devices, a magnetic nanostrip.

We have fabricated hybrid devices [Figure Left] where we can excite the nanostrip with SAW. The devices consist of a magnetostrictive nanowire placed between two InterDigitated Transducers (IDT) facing each other, allowing us to study the movement of a DW with respect to the SAW propagation direction. The entire device is grown on the surface of 128º Y-cut LiNbO3, and the SAW frequency lies in the range of GHz. We have performed electrical transport measurements on several devices where we extract the probability of the stochastic pinning of the DW with respect to the injection field. When adding the SAW we observe that it influences the DW propagation fields. Furthermore, we found that this influence depends on the SAW propagation direction by means of a mechanism still not fully understood. In [Figure Right] we show an example of one of this probabilistic maps obtained for one of our devices.

## References

[1] J. Dean et al. *Appl Phys Lett* **107** (2015) 14.
[2] D. Castilla *et al.*, *Sci Rep* **10** (2020) 1
[3] W. Edrington et al. *Appl Phys Lett* **112** (2018) 5

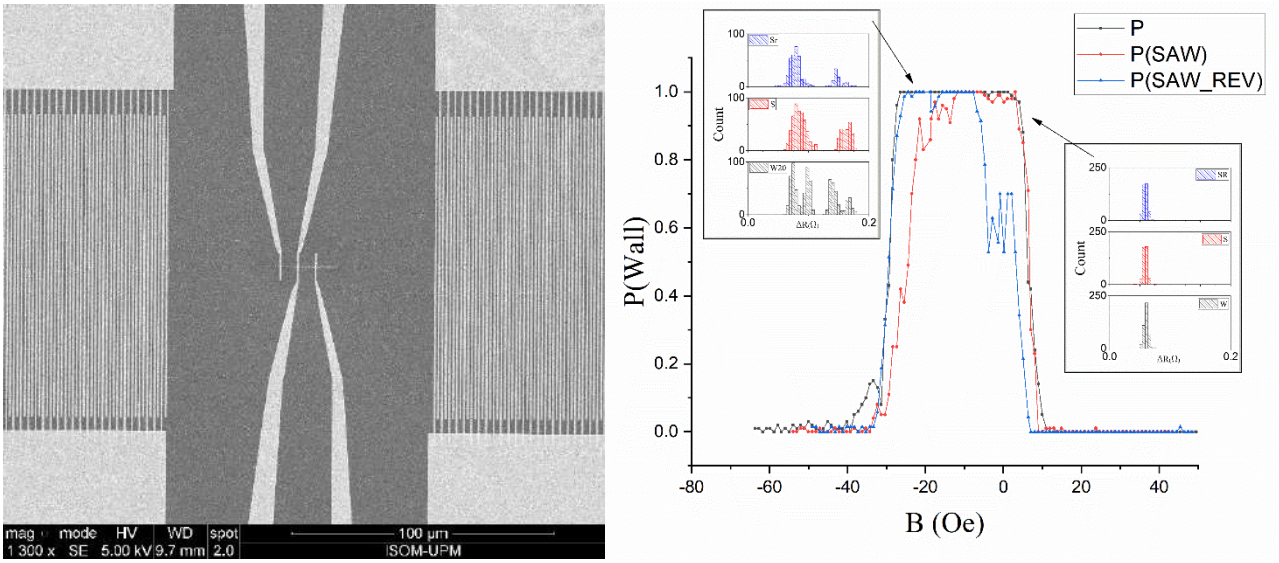



Figure: On the left: SEM image of the whole device. On the right: Probability of stochastic pinning of the DW with respect to the injection field, without SAW (Black), with SAW in the direction of the DW propagation (Red) and with SAW in the opposite direction of the DW propagation.

# Chiral Non-Collinear Spin Textures in Nominally Centrosymmetric van der Waals Magnet

*Rana Saha[1]*, Holger L. Meyerheim[1], Stuart S. P. Parkin[1]*
[1] *Max Planck Institute of Microstructure Physics, Weinberg 2, Halle (Saale), Germany*
*rana.saha@mpi-halle.mpg.de

A major topic in spintronics today is the study of magnetic materials in which neighboring magnetic moments are neither parallel nor anti-parallel but are non-collinear and thereby may form complex, topological, spin textures. Among them skyrmions represent an intensively studied group. Skyrmions are tiny magnetic nano-objects with a chiral magnetic boundary and a typically circular in shape. A prerequisite for the formation of such chiral nano-objects is that the parent material has a non-centrosymmetric crystal structure which allows the emergence of the chiral Dzyaloshinskii-Moriya exchange interactions. Recently skyrmions have been observed in several materials that were reported to be centrosymmetric [1]. Here we show evidence for skyrmions in a member of the family of van der Waals (vdW) materials, $CrTe_2$, that is nominally centrosymmetric [2]. As a result of Cr self intercalation into the vdW gaps between the Te-Cr-Te trilayers together with the formation of a three-dimensional long-range ordered (2×2×2) superstructure, the space group (SGR) symmetry is reduced from centrosymmetric $P\bar{3}2/m$ (#164) to the acentric SGR #156 (P3m1). This is a consequence that the two vdW gaps per unit cell are differenty occupied by Cr atoms and neighboring Te-Cr-Te tri-layers experience an asymmetric environment (SGR P3m1) that is compatible with the appearance of Néel-type skyrmions, which were directly imaged using in-situ Lorentz transmission electron microscopy over a wide temperature range and fields. Our studies show that self-intercalation in vdW materials is a novel route to the formation of synthetic non-collinear spin textures.

**Acknowledgements**

We acknowledge the funding by the German Research Foundation-SPP 2137 (project number-403505322) and thank P. Gargiani, M. Valvidares for assistance with the XMCD experiments.

**References**

[1] A. Chakraborty, A. K. Srivastava, A. K. Sharma, A. K. Gopi, K. Mohseni, A. Ernst, H. Deniz, B. K. Hazra, S. Das, P. Sessi, I. Kostanovskiy, T. Ma, H. L. Meyerheim, S. S. P. Parkin, *Adv. Mater.* 34 (2022) 2108637.
[2] R. Saha, H. L. Meyerheim, B. Göbel, B. K. Hazra, H. Deniz, K. Mohseni, V. Antonov, A. Ernst, D. Knyazev, A.B - Pinto, I. Mertig, S. S. P. Parkin, *Nat. Commun.* 13 (2022) 3965.

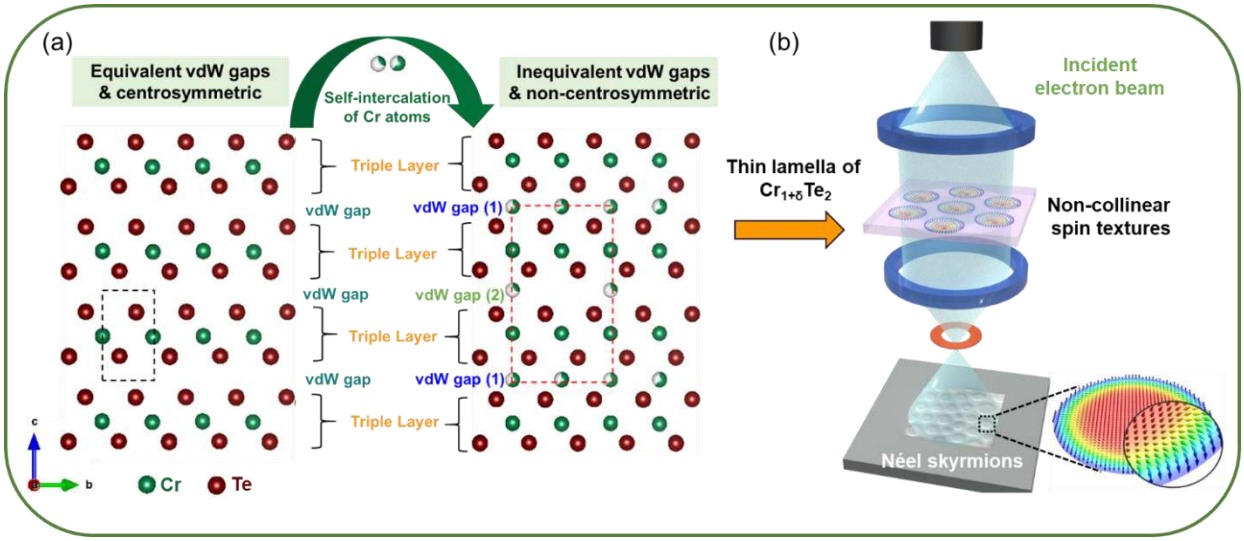



Figure: (a) Un-intercalated $CrTe_2$ with equivalent van der Waals (vdW) gaps [left] and self-intercalated $Cr_{1+\delta}Te_2$ with two inequivalent van der Waals gaps shown as vdW(1) and vdW(2) [right]. The (2×2×2) superstructure in the self-intercalated material is shown by the red dotted line. (b) Schematic of Lorentz TEM showing imaging of Néel skyrmions from the thin lamella prepared from a single crystal of self-intercalated $Cr_{1+\delta}Te_2$. Magnetic contrast of Néel skyrmions appear as circular features that are characterized by dark and bright contrast at their opposite edges. Schematic of Néel skyrmion is shown in the bottom corner.

# Magnetoelectric Effects in $CrI_3$

*Alexander Edström[1*], Massimiliano Stengel[2,3], Paolo Barone[4], Silvia Picozzi[5]*

[1*]*Department of Applied Physics, School of Engineering Sciences, KTH Royal Institute of Technology, AlbaNova University Center, 10691 Stockholm, Sweden*
[2]*Institut de Ciència de Materials de Barcelona (ICMAB-CSIC), Campus UAB, 08193 Bellaterra, Spain*
[3]*ICREA—Instituciò Catalana de Recerca i Estudis Avançats, 08010 Barcelona, Spain*
[4]*Consiglio Nazionale delle Ricerche CNR-SPIN, Area della Ricerca di Tor Vergata, Via del Fosso del Cavaliere 100, I-00133 Rome, Italy*
[5]*Consiglio Nazionale delle Ricerche CNR-SPIN, c/o Università degli Studi 'G. D'Annunzio', 66100 Chieti, Italy*
*aleeds@kth.se

We recently showed that curvature can stabilize non-collinear magnetic states, including cycloids or domain walls (DWs) in 2D magnets, such as $CrI_3$. [1] These non-collinear spin textures lead to electric polarization via magnetoelectric coupling. [2,3] Meanwhile, the flexoelectric effect generally leads to an electric polarization in curved materials. [4] This gives rise to a complex interplay between curvature, magnetism and electric polarization, motivating further investigation of magnetoelectric effects in 2D magnets. Moreover, phenomenological models predict that magnetic DWs, which are generally present in magnetic materials, are accompanied by an electric polarization. However, an understanding of this effect, in real materials, has been hindered as realistic, first principles modelling is typically not applicable at the length scales of magnetic DWs. This is an important challenge to overcome, as electric manipulation of magnetic DWs has important technological implications.

Here, we use first principles calculations combined with the generalized spin current (gKNB) model [5] to investigate the magnetoelectric coupling in a flat monolayer of $CrI_3$. The gKNB model allows us to predict the electric polarization of arbitrary, non-collinear spin states, with features well beyond what is expected from the earlier (KNB) spin current model [3]. We use the gKNB model to show that spin helices and cycloids, which can be stabilized by curvature, induce a homogenous electric polarization of magnitude 0.1 $\mu C/cm^2$. At magnetic DWs, we predict a local electric polarization of similar size, with direction dependent on the orientation and type of DW. The latter effect enables detection and control of magnetic DWs by electric means, and will be present also in flat structures.

## Acknowledgements

We acknowledge support from the Swedish Research Council (2018-06807 and 2022-04720), ÅForsk and the Göran Gustafsson foundation. Calculations were done on resources provided by the Swedish National Infrastructure for Computing (SNIC).

## References

[1] A. Edström, D. Amoroso, S. Picozzi, P. Barone, M. Stengel, Curved Magnetism in $CrI_3$. Physical Review Letters **128** (2022) 177202.
[2] M. Mostovoy, Ferroelectricity in Spiral Magnets, Physical Review Letters **96** (2006) 067601.
[3] H. Katsura, N. Nagaosa, and A. V. Balatsky, Spin Current and Magnetoelectric Effect in Noncollinear Magnets, Physical Review Letters **95** (2005) 057205.
[4] P. Zubko, G. Catalan, A. K. Tagantsev, Flexoelectric Effect in Solids, Annual Review of Materials Research **43** (2013) 387.
[5] H. J. Xiang, E. J. Kan, Y. Zhang, M.-H. Whangbo, and X. G. Gong, General Theory for the Ferroelectric Polarization Induced by Spin-Spiral Order, Physical Review Letters **107** (2011) 157202.

# Exploring New Limits to Control Domain Wall Chirality: 2Ni/Co Stacks Combined with Bi

*S. Gallego[1]*, M.A. González Barrio[2], G. Chen[2], S. Ruiz Gómez[2], A. González-García[2], A.K. Schmid[4], E.G. Michel[3], A. Mascaraque[2]*

[1] *Instituto de Ciencia de Materia1es de Madrid-CSIC, 28049 Madrid. Spain*
[2] *Dpto. Física Materiales, Universidad Complutense de Madrid, 28040 Madrid. Spain*
[3] *Dpto.Física Materia Condensada, Universidad Autónoma de Madrid, 28049 Madrid. Spain*
[4] *National Centre for Electron Microscopy, LBNL, Berkeley, CA 94720 USA.*

*sgallego@icmm.csic.es

Ultrathin magnetic metal films in contact to heavy metal (HM) layers are well known to develop robust spin-orbit (SO) derived properties ultimately leading to stabilization of chiral objects. At the basis of these phenomena are local interface Dzyaloshinskii-Moriya interactions (DMIs) that can be manipulated through different designs. The magnitude and sign of the DMI can be tuned by interface engineering, which allows to stabilize chiral or non-chiral Néel and Bloch domain walls in a given magnetic system, opening technological opportunities to handle information in electronic devices without the need of magnetic fields. The main drawback of the approach is the lack of descriptors easing prediction of the best conditions to enhance the DMI and gain control on the chirality sign. This confers additional interest to explore different systems and conditions that make possible the identification of trends.

Here we investigate a special system that adds new ingredients to previously explored metal/HM heterostructures: epitaxial [2Ni/Co] stacks on Cu(111) modified by Bi. We perform Spin Polarized Low Energy Electron Microscopy (SPLEEM) measurements demonstrating that Bi influences the spin texture of the system, with magnetic domain walls evolving from non-chiral Bloch to homo-chiral Néel configurations when Bi is added. Eventhough the surfactant properties of Bi render a disordered distribution, the system preserves constant homochirality both at low and room temperatures.

In this presentation we will focus on understanding the origin of this behavior based on ab initio calculations within the density functional theory, exploring the balance of magnetic energy terms (exchange and SO-derived) under different geometries. Bi contributes with singular properties: it has a large size, that introduces incomplete interface layer coverage; and it is a *p* valence band metal, weakly polarizable. Also the 2Ni/Co stacks confer an inherent asymmetry that conditions interface additive effects, further supported by the different chiral response of Ni and Co. Our results evidence 2Co/Ni heterostructures combined with Bi as unique systems to tune and control stable homochiral structures where the DMI can even be made the dominant energy scale.

## Acknowledgements

This work has been performed under grants PID2021-123295NB-I00 and PID2021-122980OB-C55, financed by MCIN/AEI/10.13039/501100011033/ and FEDER *A way to make Europe*. Further support is acknowledged from projects MAT2017-87072-C4-2-P and S2108 –NMT4321.

## References

[1] G. Chen, et al., Ap. Phys. Lett. 106, 062402 (2015).

# Eigenmodes of Magnetic Skyrmion Lattices

*Louise Desplat[1]*, and Bertrand Dupé[1,2]*

[1]*Nanomat/Q-mat/CESAM, Université de Liège, Belgium*

[2]*Fonds de la Recherche Scientifique (FNRS), Bruxelles, Belgium*

*ldesplat@uliege.be

We explore the interplay between topology and eigenmodes by changing the stabilization mechanism of skyrmion lattices (skX). We focus on two prototypical ultrathin films hosting an hexagonal (Pd/Fe/Ir(111) [1]) and a square (Fe/Ir(111) [2]) skyrmion lattice, which can both be described by an extended Heisenberg Hamiltonian.

We first examine whether the Dzyaloshinkskii-Moriya, or the exchange interaction when taken as the leading energy term affects the modes of the hexagonal skX of Pd/Fe/Ir(111). In both cases, we find that the lowest frequency modes correspond to internal degrees of freedom of individual skyrmions, and suggest a classification based on azimuthal and radial numbers *(l,p)*, with up to $l=6$, and $p=2$. We also show that the gyration behavior induced by an in-plane field [3] corresponds to the excitation of $l=1$ deformation modes with varying radial numbers.

Second, we examine the square lattice of skyrmions of Fe/Ir(111). Its stabilization mechanism is dominated by the 4-spin interaction as obtained in Ref. [2]. After relaxation, the unit cell does not carry a topological charge, and the eigenmodes do not correspond to internal skyrmion deformations. By reducing the 4-spin interaction, an integer topological charge is recovered, but the charge carriers do not possess internal degrees of freedom, nor are they separated by energy barriers. We conclude that a 4-spin dominated Hamiltonian does not yield skyrmion lattice solutions, and that therefore*, a nontrivial topology does not imply the existence of skyrmions* [4].

## Acknowledgements

This research was supported by the University of Liège under Special Funds for Research, IPD-STEMA Programme.

## References

[1] B. Dupé, M. Hoffmann, C. Paillard, S. Heinze, *Nat. Commun*. **5** (2014) 4030.

[2] S. Heinze, *et al. Nat. Phys.* **7** (2011) 713.

[3] M. Mochizuki, *Phys. Rev. Lett.* **108** (2012) 017601.

[4] L. Desplat, B. Dupé, *under review at Phys. Rev. B* (2023).

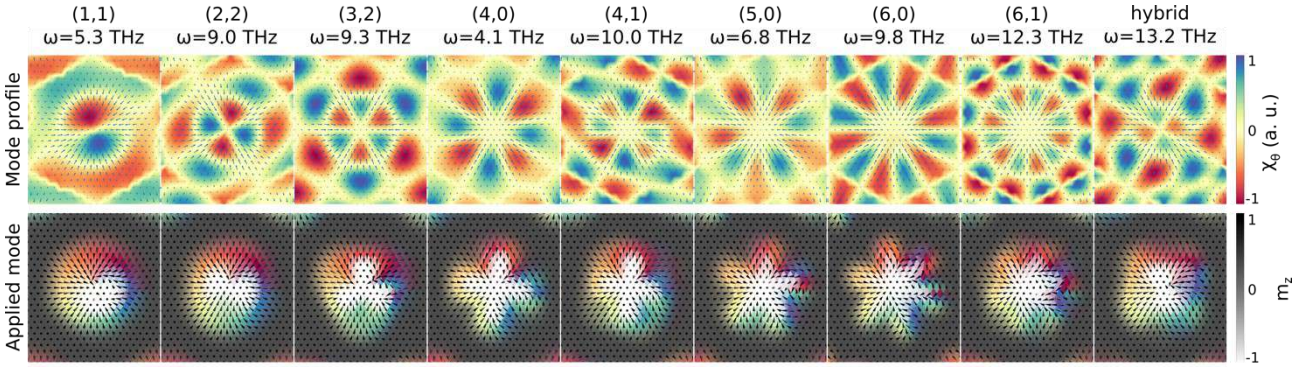



Figure: Example of modes of the skX of Pd/Fe/Ir(111), classified by azimuthal and polar numbers *(l,p)*. Top: mode out-of-plane profile, bottom: mode applied to the skX state.

# Evaluation of the Repulsive Forces Acting on Skyrmions

*Y. Sassi, D. Sanz-Hernández, T. da Câmara Santa Clara Gomes, S. Krishnia, N. Reyren, V. Cros**

[1] *Unité Mixte de Physique, CNRS, Thales, Université Paris-Saclay, 1 av. Augustin-Fresnel, Palaiseau, France*

* Vincent.cros@cnrs-thales.fr

Often the study of skyrmions is motivated by technological applications where the skyrmion acts as a bit of information in a circuit made of magnetic materials [1]. The skyrmions are interesting in such applications, because, having a soliton behaviour, they can move around corners or even edge defects, keeping their shape and properties [2]. Moreover, skyrmions will not easily merge with each others, so their number is conserved. All these desirable properties result from repulsive forces from any type of "walls", notably track edges, magnetic domain walls, and other skyrmions. Despite the prime importance of these repulsion forces, reports about their experimental measurements are scarse [3].

Here, we present experiments in metallic multilayers allowing these repulsion forces to be estimated. We used (Pt|Co|Al) based multilayers, with typically ten repetitions, leading to stable skyrmions under moderate external field (~20 mT). The skyrmions are driven by SOT provided by the Pt and Al layers. The corresponding force, which is proportional to the current density, can be calculated. We observe the deviation, annihilation or stop of the skyrmions in different situations as a function of the (local) current density, and we deduce the effective repulsion force. A few examples are illustrated in the Figure.

**Acknowledgements**

This work has been supported by DARPA TEE program grant (MIPRHR - 0011831554), ANR grant TOPSKY (ANR-17-CE24-0025), the FLAG - ERA Sograph MEM (ANR-15-GRFL-0005) and the Horizon2020 Framework Program of the European Commision, under FETProactive Grant agreement No. 824123 (SKYTOP) (H2020 FET proactive 824123).

**References**

[1] A. Fert, N. Reyren, V. Cros, *Nat. Rev. Materials* **1** (2017) 17031.
[2] J. Sampaio *et al*, *Nat. Nanotechnol.* **8** (2013) 839.
[3] S. Yang *et al*, *Phys. Rev. Appl.* **18** (2022) 024030.

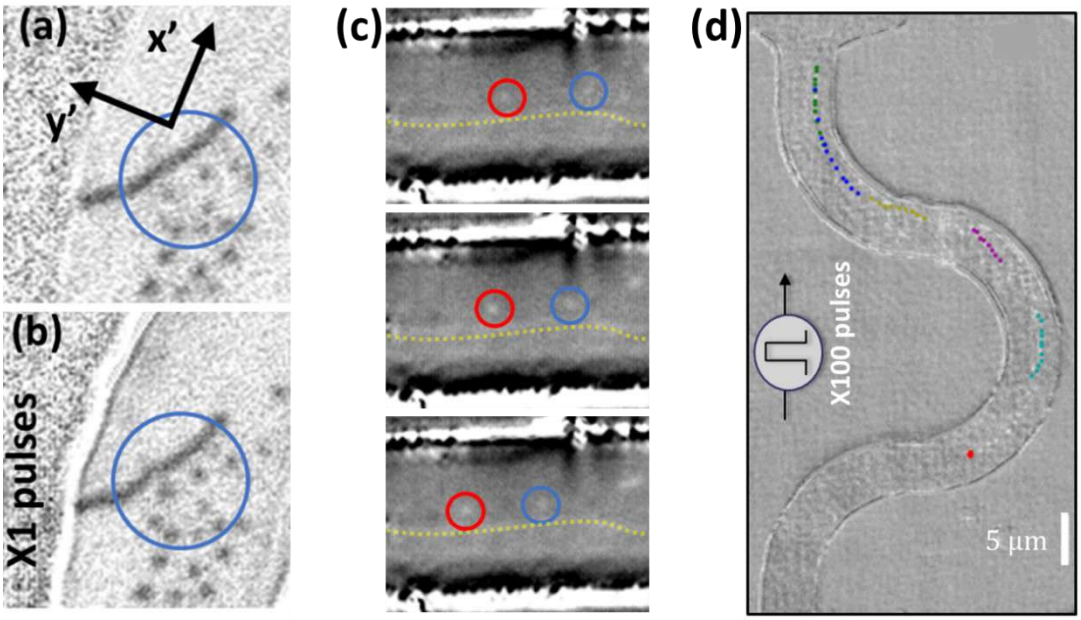



Figure: Examples of skyrmion interaction. (a-b) Skyrmion pushing a worm domain, (c) skyrmions moving along a worm domain (materialized by the dashed yellow line) and (d) skyrmions moving along curved edges.

# Pinning disorder and domain walls interaction controlled by He+ ion irradiation in Pt/Co/AlOx ultrathin films

*C. Balan[1], J. W. van der Jagt[2, 3], J. Peña Garcia[1], J. Vogel[1], L. Ranno[1], M. Bonfim[4], D. Ravelosona[2,3], S. Pizzini[1],1 and V. Jeudy[5]**

[1]Univ. Grenoble Alpes, CNRS, Institut Néel, 38042 Grenoble, France

[2]Spin-Ion Technologies, 10 Boulevard Thomas Gobert, 91120 Palaiseau, France

[3]Centre de Nanosciences et de Nanotechnologies, CNRS, Université Paris-Saclay, 10 boulevard Thomas Gobert, 91120 Palaiseau, France

[4]Dep. de Engenharia Elétrica, Universidade Federal do Parana, Curitiba, Brasil

[5]Laboratoire de Physique des Solides, Université Paris-Saclay, CNRS, 91405 Orsay, France

*vincent.jeudy@universite-paris-saclay.fr*

We have studied the effect of $He^+$ irradiation on the dynamics of chiral domain-walls (DWs) in ultra-thin Pt/Co/AlOx trilayers. Through a self-consistent description of the creep and depinning dynamics [1] completed by a scaling model of DW depinning [2] (relating microscopic DW pinning properties to DW dynamics and micromagnetic parameters), we reveal an excellent scaling between the variations of the DW pinning length scale ξ and the DW width parameter. This scaling strongly suggests that the modification of the DW pinning by irradiation is essentially dominated by the variations of DW magnetic texture (via the variation of the magnetic anisotropy), while short range atomic displacement produced by irradiation [3] have a weak impact on the pinning disorder [4].

The DW velocity versus applied perpendicular magnetic field is reported in the Figure (a) for increasing $He^+$ fluence. As it can be observed, the irradiation leads to a strong decrease of the depinning field and an enhancement of DW velocity. The velocity curves present a good agreement with the self-consistent description of the creep (solid lines) and depinning (dashed lines) dynamics, which allows extracting the values of the pinning parameters [1] (depinning field, temperature and velocity (not shown)). Complementary characterizations of the trilayers (determination of the saturation magnetization, anisotropy, and Dzyaloshinskii-Moriya interaction (DMI)) reveal that irradiation essentially reduces the anisotropy, which increases the DW width, while the other micromagnetic parameters remain only weakly affected.

Using the scaling model of DW depinning [2], we deduce the pinning length ξ (see Fig. (b)) and the pinning strength $f_{pin}$ (see Fig. (c)). Their variations with $He^+$ fluence are found to present a perfect scaling with those of the DW width Δ and DW energy per unit length $\sigma t$, respectively.

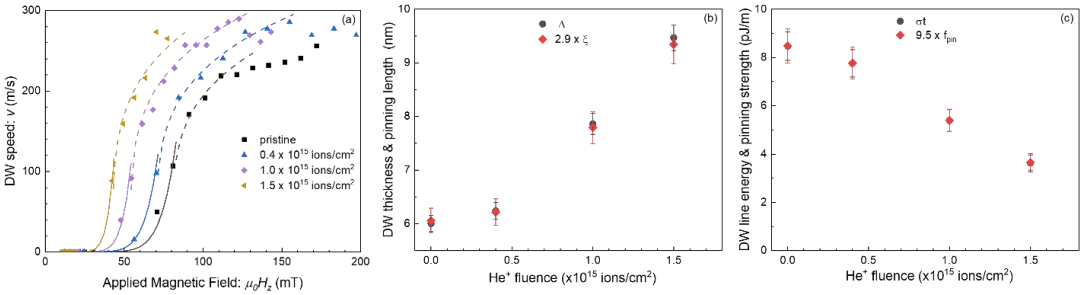



Figure: Effect of $He^+$ ion irradiation on DW dynamics and DW-disorder interaction: (a) domain wall velocity versus out-of plane field $\mu_0 H_z$ for increasing irradiation fluence; comparison between (b) DW-disorder characteristic length scale ξ and DW parameter Δ, (c) pinning strength $f_{pin}$ and DW energy per unit length $\sigma t$.

## References

[1] V. Jeudy, R. Díaz Pardo, W. Savero Torres, S. Bustingorry, & A. Kolton, Phys. Rev. B **98**, 054406 (2018).
[2] P. Géhanne, S. Rohart, A. Thiaville, and V. Jeudy, Phys. Rev. Research **2**, 043134 (2020).
[3] T. Devolder, Phys. Rev. B **62**, 5794–5802 (2000).
[4] C. Balan *et al.*, arXiv:2303.04577, submitted to Appl. Phys. Lett..

# Manipulating Skyrmion Nucleation in Magnetic Multilayers with Ion Irradiation and by Exploiting Néel Caps

*M.C.H. de Jong[1]*, B.H.M. Smit[1], M.J. Meijer[1], J. Lucassen[1], H.J.M. Swagten[1], B. Koopmans[1], R. Lavrijsen[1]*

[1] *Department of Applied Physics, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, the Netherlands*

*m.c.h.d.jong@tue.nl

Magnetic skyrmions are promising candidates for data carriers in future spintronic memory and data processing applications. They can be readily stabilized and studied in magnetic multilayers. These are systems consisting of repetitions of magnetic layers sandwitched between heavy metal layers [1]. In such a system, the skyrmion properties and behavior are determined by a balance between the domain wall energy density and the dipolar interaction [2]. In this contribution we investigate what effect tuning each of these contributions to this energy balance has on current-driven skyrmion nucleation.

We have previously shown that the domain wall energy density in an Ir|Co|Pt based magnetic multilayer can be controlled using $Ga^+$ ion irradiation [3]. The irradiation results in increased intermixing at the Ir|Co and Co|Pt interfaces which modifies the interfacial magnetic properties: *i.e.* perpendicular anisotropy and the Dzyaloshinskii-Moriya Interaction (DMI). Here, we quantitatively study the effect of this tuning on Joule heating-driven skyrmion nucleation by measuring the number of skyrmions as a function of the applied current density [Fig. 1(a)] and find that the energy required for skyrmion nucleation, $E_{sks}$, is significantly affected and scales approximately linearly with the domain wall energy density. This enables local control of the skyrmion nucleation and energy.

The second part of the energy balance, the dipolar energy, can also be used to tune the skyrmion energy [4]. Traditionally, every repeat of a magnetic multilayer is identical resulting in a mismatch between the dipolar field and the DMI effective field in half of the stack. By reversing the stacking order halfway up the stack—and thereby the sign of the DMI—the dipolar field and DMI effective field are aligned throughout [Fig. 1(b, c)]. This is verified using micromagnetic simulations [Fig. 1(d)] and measurements of the stripe domain width [4]. In this contribution we will report on the effect that this increase in effective DMI has on the current-driven nucleation of skyrmions.

## References

[1] C. Moreau-Luchaire, C. Moutafis, N. Reyren, *et al.*, Nature Nanotechnology **11**, 444-448 (2016)
[2] F. Büttner, I. Lemesh, G.S.D. Beach, Scientific Reports **8**, 4464 (2018)
[3] M.C.H. de Jong, M.J. Meijer, J. Lucassen, *et al.*, Physical Review B **105**, 064429 (2022)
[4] J. Lucassen, M.J. Meijer, M.C.H. de Jong, *et al.*, Physical Review B **102**, 014451 (2020)

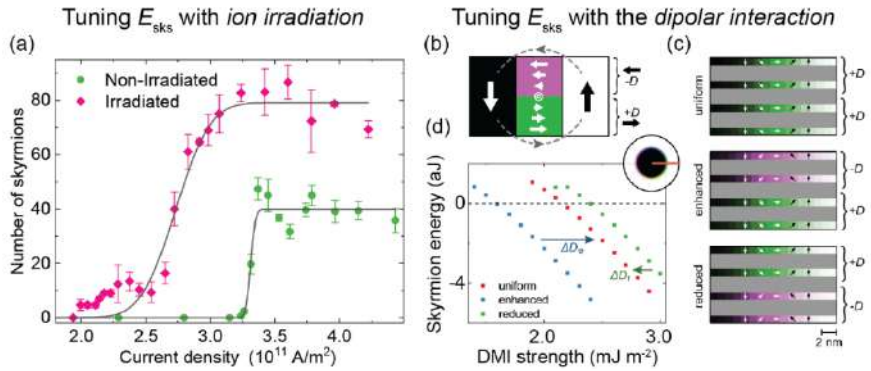



Fig. 1: (a) Experimentally observed number of skyrmions after Joule heating-driven skyrmion nucleation in a device with and without ion irradiation. Ion irradiation reduces the threshold current and increases the number of nucleated skyrmions. (b) Schematic representation of a multilayer. The dipolar field (grey) due to the domains (black and white) creates two Néel caps in the domain wall (pink and green). Since the sign of the DMI is opposite in both halves, the dipolar field and DMI effective field are always aligned. (c) Different possible stacks with different DMI sign in each half. (d) Skyrmion energy as a function of the DMI strength calculated using MuMax[3] for a skyrmion in a 256 nm dot (shown in the inset). The three colours correspond to the stacks shown in (c). Adapted from Ref. [4].

# Ultrafast time-evolution of chiral Néel magnetic domain walls in multilayers probed by XRMS.

*Cyril Leveillé[1], Yanis Sassi[2], Emanuele Pedersoli[3], Flavio Capotondi[3], Vincent Cros[2], Jean-Yves Chauleau[4], Michel Viret[4], Nicolas Reyren[2], and Nicolas Jaouen[1*]*

[1]*Synchrotron SOLEIL, Saint-Aubin, Boite Postale 48, 91192 Gif-sur-Yvette Cedex, France*

[2]*Unité Mixte de Physique, CNRS, Thales, Université Paris-Saclay, 91767 Palaiseau, France*

[3]*Elettra-Sincrotrone Trieste, 34149 Basovizza, Trieste, Italy*

[4]*SPEC, CEA, CNRS, Université Paris-Saclay, 91191 Gif-sur-Yvette, France*

*nicolas.jaouen@synchrotron-soleil.fr

Non-collinear spin textures in ferromagnetic ultrathin films are attracting a renewed interest fueled by possible fine engineering of several magnetic interactions, notably the interfacial Dzyaloshinskii-Moriya interaction in magnetic multilayers. This allows the stabilization of complex chiral spin textures such as chiral magnetic domain walls (DWs), spin spirals, and magnetic skyrmions among others. We have recently shown that circular dichroism in x-ray resonant magnetic scattering (XRMS) is a powerful tool to determine the domain wall characteristics, i.e., their type (Néel or Bloch) and sense of rotation (chirality) in ferromagnetic or antiferomagnetic multilayers [1,2,3].

In the first part of this presentation, we will report on the ultra-fast (in the first few picoseconds) evolution of the magnetic chirality in multilayers as well as their recovery on longer timescale (few hundreds of picoseconds) using XRMS [4]. Beyond the simple evolution of the magnetic domains' period in magnetic multilayers with large perpendicular anisotropy we have investigated how the chirality of the non-collinear spin texture, and their long-range ordering, evolves in the few picoseconds after a strong optical pulse. The change in the ratio between the chiral and magnetic signals have been related to an ultra-fast distortion of the homochiral Néel shape to transient Bloch-Néel-Bloch domain wall. After a few ps, the DWs return to a homochiral Néel configuration preserving the original sense of rotation (i.e., chirality). At longer timescale, the whole system relaxes towards its original homochiral Néel type equilibrium but the chiral magnetic order in the domain walls recovers faster than the collinear magnetic order in the domains.

In the second part, we will present new result obtained from spin-spiral in synthetic antiferromagnetic multilayers. This recent work has been done thanks to a new mode of operation of FERMI light source allowing to perform time resolved XRMS at Fe L edges (700eV) following the approach we developed at synchrotron [3].

## References

[1] J.-Y. Chauleau, W. Legrand, N. Reyren, D. Maccariello, S. Collin, H. Popescu, K. Bouzehouane, V. Cros, N. Jaouen, and A. Fert,"Chirality in magnetic multilayers probed by the symmetry and the amplitude of dichroism in x-ray resonant magnetic scattering", Phys. Rev. Lett 120, 037202 (2018)

[2] W. Legrand, J.-Y. Chauleau, D. Maccariello, N. Reyren, S. Collin, K. Bouzehouane, N. Jaouen, V. Cros, and A. Fert, "Hybrid chiral domain walls and skyrmions in magnetic multilayers", Science Adv. 4, eaat0415 (2018)

[3] C. Léveille, S. Flewett, E. Burgos-Parra, Y. Sassi, W. Legrand, F. Ajejas, V. Cros, N. Reyren, N. Jaouen, "Chiral spin spiral in synthetic antiferromagnets probed by circular dichroism in x-ray resonant magnetic scattering" Physical Review B., 104(6): art.n° L060402. (2021).

[4] C. Léveille, E. Burgos-Parra, Y. Sassi, F. Ajejas, V. Chardonnet, E. Pedersoli, F. Capotondi, G. De Ninno, F. Maccherozzi, S. Dhesi, D.M. Burn, G. van der Laan, O.S. Latcham, A.V. Shytov, V.V. Kruglyak, E. Jal, V. Cros, J.Y. Chauleau, N. Reyren, M. Viret, N. Jaouen, "Ultrafast time-evolution of chiral Néel magnetic domain walls probed by circular dichroism in x-ray resonant magnetic scattering" Nature Communications., 13: art.n° 1412. (2022).

# The Instability In Domain Wall Dynamics In Almost Compensated Ferrimagnets

*Roman Ovcharov[1]*, Roman Khymyn[1], Boris Ivanov[2,3], Johan Åkerman[1]*

[1] *Department of Physics, University of Gothenburg, Gothenburg, Sweden*

[2] *Institute of Magnetism of NASU and MESU, Kyiv, Ukraine*

[3] *Radboud University, Institute for Molecules and Materials, Nijmegen, Netherlands*

*roman.ovcharov@physics.gu.se

Topological spin structures, such as domain walls (DW) and skyrmions, have intriguing features and can play a key role in spintronics. In both ferromagnets (FM) and antiferromagnets (AFM), a DW, driven by applied torque, behaves like a solid particle, which can be described by collective variables: DW position and the angle of the magnetic order vector, albeit the dynamic properties are dramatically different. For an AFM, the DW energy as a function of its momentum is the same as that of a standard relativistic particle, see Fig. 1 (a), because of the Lorentz invariance of the AFM dynamics [1]. For FMs, the gyrotropic dynamic term couples two degrees of freedom; it slows down an FM DW and results in the periodic dependence of the DW energy on momentum.

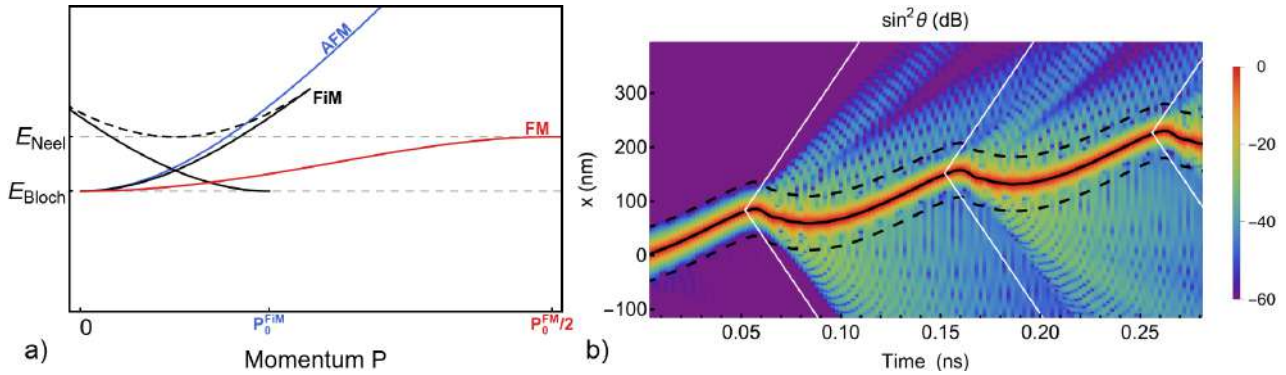



Figure 1: (a) Dependence of the energy on momentum for a DW in FM, AFM, and FiM near the spin compensation point; (b) The spatiotemporal evolution of a FiM DW. The white lines show the propagation with the maximum speed of magnons drawn from the points at which the DW reaches the critical point. The solid black line follows the trajectory of the domain wall center, while the dashed black lines are 50 nm away from the center.

We demonstrate that the DW dynamics in ferrimagnetic (FiM) materials near the spin compensation point is entirely different from the FM and AFM cases [2]. The dispersion law for FiM DWs exhibits two crossed branches with opposite group velocities, see Fig. 1 (a), where each branch contains an endpoint [3]. Driven by a field-like torque, a FiM DW accelerates to a higher speed and momentum than its FM counterpart and can reach the endpoint. Wherein it falls into a region of instability, where the internal dynamics of the DW is excited, and the collective variables approach fails. We show by micromagnetic simulations that the dynamical state of a DW, in this case, covers a wide frequency range, far beyond the magnon bandgap of a FiM. In this way, the FiM DW driven by external field-like torque acts as an efficient source of the spin waves (SW), propagating out of the DW region, see Fig. 1 (b).

## References

[1] Bar'yakhtar, V. G., Chetkin, M. V., Ivanov, B. A., et al. Dynamics of Topological Magnetic Solitons: Experiment and Theory, *Springer*, **129** (2006).

[2] Ivanov, B. A., Galkina, E. G., Kireev, V. E., et al. *Low Temperature Physics*, **46** (2022) 841-850.

[3] Galkina, E. G., Zaspel, C. E., Ivanov, B. A., et al. *JETP Letters*, **110** (2019) 481-486.

# Room-temperature skyrmions in symmetric RKKY multilayers: implications for spintronic without fields

*Lucas Palhares*[1,2], *Thiago Batistel*[1,2], *Fanny Béron*[2], ***J. Brandão****[1]

[1] Laboratório Nacional de Luz Síncrotron (LNLS), Centro Nacional de Pesquisa em Energia e Materiais (CNPEM), 13083-970 Campinas, SP, Brazil

[2]Instituto de Física Gleb Wataghin, Universidade Estadual de Campinas, 13083-859, Campinas SP, Brazil

*Jeovani.brandao@lnls.br

Magnetic skyrmions with zero topological charges and non-Hall effect, are very attractive from both physical underlying mechanisms and technological point of view [1,2]. Thus, antiferromagnetic (AFM) skyrmions promise to host all the requirements to develop new magnetic memory, logic gates, and neuromorphic computing devices. Here, we show that skyrmions are stabilized in (AFM) multilayers mediated by the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction. For a range of Ru spacer layers in symmetric Pt/Co/$Ru_x$/Co/Pt heterostructures, hysteresis loops exhibit AFM coupling between bottom Pt/Co and top Co/Pt layers, leading to fully compensated magnetic moments, which are key ingredients to reduce stray fields and the skyrmion Hall effect. Magnetic force microscopic shows AFM-skyrmions at room-temperature and zero-field, and both size and density depend on the Ru thicknesses. In addition, interlayer exchange coupling (IEC) given by the RKKY and perpendicular magnetic anisotropy (PMA) follow similar oscillatory behavior across the Ru thicknesses, which determines the shape, size and density of the AFM textures. Simulations are performed and reveal that only when a sizable interfacial Dzyaloshinskii-Moriya interaction (DMI) is present in the modelling, the experimental images can be reproduced. Our findings shed light on the AFM skyrmions stability in prototype RKKY multilayers, which is a step towards for their use in devices based on antiferromagnetic spintronics.

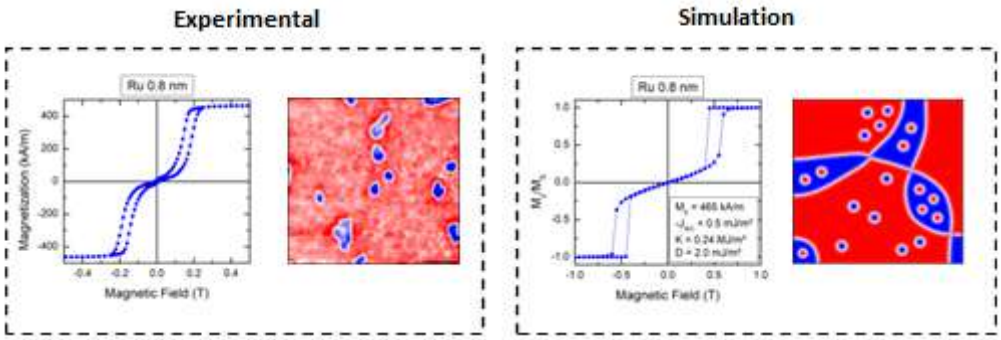
## Acknowledgements

This work was supported by the Brazilian agencies FAPESP (Project No.2012/51198-2, 2017/10581-1, and 2020/07397-7), as well CNPq (436573/2018-0), and (CAPES), grant number 88887.569085/202000. Also, we thank the (CNPEM), which provided scholarship for T. M. Batistel by means of (CNPq).

## References

[1] Brandão, J., Dugato, D. A., Puydinger dos Santos, M. V. & Cezar, J. C**. Evolution of Zero-Field Ferrimagnetic Domains and Skyrmions in Exchange-Coupled Pt/CoGd/Pt Confined Nanostructures: Implications for Antiferromagnetic Devices**. *ACS Appl. Nano Matter* **2**, 7532 (2019).

[2] J. Brandão, D. Dugato, M. Puydinger dos Santos, F. B´eron, and J. Cezar**, Tuning isolated zero-field skyrmions and spin spirals at room-temperature in synthetic ferrimagnetic multilayers** Applied Surface Science 585, 152598 (2022).



# Interface Engineering of Synthetic Antiferromagnets for Energy-Efficient Current-Induced Domain Wall Motion

*Andrea Migliorini*[1]*, *Jae-Chun Jeon*[1], *Jiho Yoon*[1], *Andriy Styervoyedov*[1], *Stuart S.P. Parkin*[1]

[1] *Max Planck Institute for Microstructure Physics, Weinberg 2, 06120 Halle (Saale), Germany*

*andrea.migliorini@mpi-halle.mpg.de

The manipulation of magnetic domain walls by electric currents, thanks to charge-to-spin conversion phenomena, has allowed the development of domain-wall-based racetrack memory devices [1]. Current-induced domain wall motion is particularly efficient in synthetic antiferromagnets with perpendicular magnetic anisotropy, due to the simultaneous occurrence of spin-orbit torque, Dzyaloshinskii-Moria interaction (DMI) and interlayer exchange coupling [2,3]. Recent progress from our group has confirmed the technological potential of racetracks, by exploring lateral junctions for improved thermal stability [4], and domain wall motion in devices that extend in the vertical direction [5].

The high domain wall velocities in synthetic antiferromagnetic racetracks could facilitate the development of ultra-fast and energy-efficient memory and logic technology. The efficiency of domain-wall-shifting operations is determined by the threshold current density necessary to displace the domain walls, as well as by the velocity at which domain walls can be moved. These properties result from the many interfacial interactions which arise in synthetic antiferromagnetic multilayers (figure 1). Therefore, understanding the role of the interfaces between the different layers, and engineering their properties, is of paramount importance to reduce the energy consumption in domain-wall-based devices.

In this work, we investigate the effects of interface engineering on the magnetic properties and domain wall motion in synthetic antiferromagnets. Firstly, we introduce engineered spin Hall multilayers in racetrack films, to increase the spin-orbit torque and consequently boost the current-induced domain wall motion. As a result, our devices show a dramatic reduction of the threshold current density, and perform three times more efficiently than racetracks with conventional spin Hall layer (e.g. Pt). In addition, we investigate the role of the termination layer in spin Hall multilayered structures, which determines the interface with the lower magnetic layer. In particular, we focus on its effect on magnetic anisotropy, interfacial DMI and spin transparency.

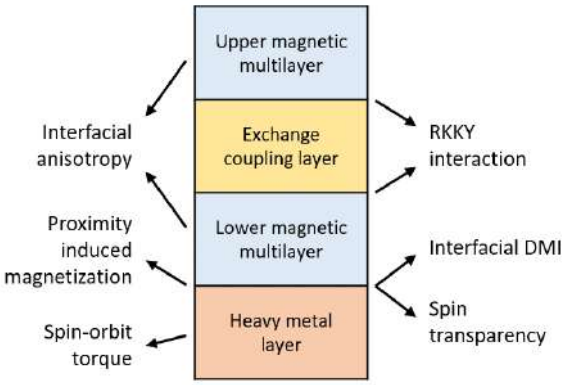



Figure 1: The role of interfaces in synthetic antiferromagnetic multilayers

Secondly, we tune the exchange coupling strength in synthetic antiferromagnetic multilayers while keeping the magnetic moment well compensated. We show that higher exchange coupling leads to enhanced domain wall speed and mobility, and demonstrate current-induced domain wall motion up to three times faster compared to conventional films. It turns out that the engineered exchange coupling layer promotes robust thermal stability in synthetic antiferromagnetic racetracks, which enables faster domain wall motion at high current density ranges, where racetracks typically suffer from thermal fluctuations due to Joule heating.

Our work clearly demonstrates that careful interface engineering in synthetic antiferromagnets can improve the efficiency of current-induced domain wall motion by an order of magnitude, which is of significant importance for lowering the energy consumption in domain-wall-based memory and logic technology.

## References

[1] S.S.P. Parkin, M. Hayashi and L. Thomas, *Science* **320**, 190–194 (2008).
[2] KS. Ryu, L. Thomas, SH. Yang and S.S.P. Parkin, *Nat. Nanotechnol.* **8**, 527–533 (2013).
[3] SH. Yang, KS. Ryu and S. S. P. Parkin, *Nat. Nanotechnol.* **10**, 221–226 (2015).
[4] J. Yoon, SH. Yang, JC. Jeon, A. Migliorini, I. Kostanovskiy, T. Ma, and S.P.P. Parkin, *Nat. Nanotechnol.* **17**, 1183–1191 (2022).
[5] K. Gu, Y. Guan, B.K. Hazra, H. Deniz, A. Migliorini, W. Zhang, and S.P.P. Parkin, *Nat. Nanotechnol.* **17**, 1065–1071 (2022).

# Driving skyrmions with high mobility in synthetic ferrimagnets

*Sougata Mallick[1*], Nicholas Figueiredo Prestes[1], Yanis Sassi[1], Fernando Gallego[1], Nicolas Reyren[1], Karim Bouzehouane[1], Sachin Krishnia[1], Vincent Jeudy[2], André Thiaville[2], Thibaud Denneulin[3], Rafal E. Dunin-Borkowski[3], Vincent Cros[1*], and Albert Fert[1]*

[1]*Unité Mixte de Physique, CNRS, Thales, Université Paris-Saclay, Palaiseau, France*

[2]*Laboratoire de Physique des Solides, Université Paris-Saclay, Orsay, France*

[3]*Forschungszentrum Jülich, ER-C for Microscopy and Spectroscopy with Electrons, Jülich, Germany*

**vincent.cros@cnrs-thales.fr; sougata.mallick@cnrs-thales.fr*

Being both, a direct signature of the topological nature of magnetic skyrmions and an issue for their implementation in renewed logic and memory devices, the so called skyrmion Hall effect (SkHE) has been the focus of many researches in the last few years [1]. In this context, investigation of ferrimagnetic systems in which two sublattices with opposing spin orientations can compensate the effects of each other is of interest. With ferrimagnets, behaviour similar to that of an antiferromagnet are obtained with the additional flexibility of tuning the properties by varying temperature, the individually detectable proportion of the constituting materials, etc. We focus on multilayers of Pt/Co/Tb by controlling the thickness of Co and Tb, as well as the numbers of repetitions to obtain a signature of antiferromagnetic coupling between the Co and Tb moments. The comparison of skyrmion velocity in Co-Tb multilayers with different Tb thicknesses is shown in Fig. 1(a). As the Tb thickness is increased, we observed a significant enhancement of skyrmion velocity simultaneously with the decrease in the net magnetization. In the best samples, we obtained velocity up to ~400 m/s for skyrmions with diameter of 160 nm. Additionally, we show that the skyrmions move in a relatively straight path due to the combination of the reduced SkHE and the edge repulsion in the narrow tracks (1-µm wide). The scaling between field induced domain wall (DW) motion and current induced skyrmion motion with prediction of DW creep, depinning, and flow is demonstrated in Fig. 1(b). In conclusion, we show that with the control of ferrimagnetic ordering in Co-Tb multilayers by varying the Tb thicknesses, we can achieve high skyrmion mobility in *DW flow* regime with reduced SkHE.

## Acknowledgements

We acknowledge French ANR grant TOPSKY (ANR-17-CE24-0025), DARPA TEE program grant (MIPR#HR0011831554), FLAG-ERA Sograph MEM (ANR-15-GRFL-0005), for their financial support.

## References

[1] A. Fert *et al*, Nat. Rev. Mat. 2 (2017) 17031

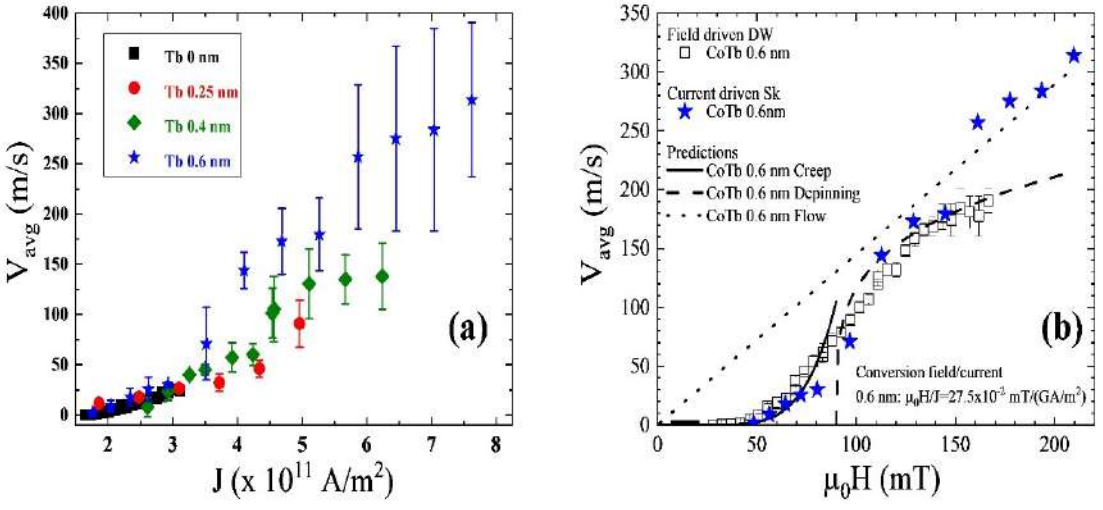



**Figure 1:** Skyrmions and domain walls velocities. (a) Current induced skyrmion velocity in Co-Tb multilayers with different Tb thicknesses. (b) Correlation between field-induced domain wall motion and current induced skyrmion motion with prediction of domain wall creep, depinning, and flow. Here we assume that the current density effect scales linearly with the one produced by a perpendicular field.

# SYMPOSIUM 13.
## DOMAIN WALLS, SKYRMIONS AND SPIN-ORBIT RELATED PHENOMENA.
## S13 POSTERS

# Ab Initio Theory of Dzyaloshinskii-Moriya Interaction via Techniques of Electron Transport

*Ilja Turek*[1]*

[1] *Institute of Physics of Materials, Czech Academy of Sciences, Zizkova 22, 61600 Brno, Czech Republic*

*turek@ipm.cz

The Dzyaloshinskii-Moriya interaction (DMI) gives rise to an important contribution to the micromagnetic energy density that is linear in the gradients of the magnetization direction of a ferromagnet. This interaction is relevant in systems with broken inversion symmetry and with non-negligible spin-orbit interaction; its contribution to the micromagnetic energy density can be quantitatively described in terms of the DMI-tensor [1, 2, 3]. In this work, we develop an ab initio theory of the DMI-tensor for itinerant ferromagnets including random alloys. In analogy to our recent formulation of the spin stiffness [4], we employ techniques of electron transport theory combined with the coherent potential approximation (CPA).

The resulting formula for the DMI-tensor contains a one-particle contribution and a two-particle contribution. The one-particle contribution includes an explicit term due to the intrinsic spin current derived earlier [2]. The full CPA-average of the two-particle contribution leads naturally to a coherent part and an incoherent part due to the disorder-induced vertex corrections. Numerical implementation of the developed formalism was done in the relativistic tight-binding linear muffin-tin orbital (TB-LMTO) method and the first applications include selected B20 compounds (FeGe), dilute magnetic semiconductors (Mn-doped GaAs), and quaternary Heusler alloys (CoMnFeSi). The obtained values and concentration trends of the DMI-tensor and of its individual contributions will be presented and discussed.

## References

[1] F. Freimuth et al., *J. Phys.: Condens. Matter* **26** (2014) 104202.
[2] T. Kikuchi et al., *Phys. Rev. Lett.* **116** (2016) 247201.
[3] S. Mankovsky et al., *Phys. Rev. B* **99** (2019) 104427.
[4] I. Turek et al., *Phys. Rev. B* **101** (2020) 134410.

# Spontaneous Square vs. Hexagonal Nanoscale Skyrmion Lattices in Fe/Ir(111)

*Mara Gutzeit[1,*], Tim Drevelow[1], Moritz A. Goerzen[1], Soumyajyoti Haldar[1], and Stefan Heinze[1,2]*

[1] *Institute of Theoretical Physics and Astrophysics, University of Kiel, Leibnizstrasse 15, 24098 Kiel, Germany*

[2] *Kiel Nano, Surface, and Interface Science (KiNSIS), University of Kiel, Germany*

*gutzeit@physik.uni-kiel.de

Spintronics resorting to non-collinear spin textures represents a rapidly growing field since the experimental discovery of magnetic skyrmion lattices [1-3]. Skyrmions have been detected in a large variety of magnetic materials even at room temperature [4,5] and several potential applications of magnetic skyrmions are currently subject of intense research [6,7].

Here, we investigate the emergence of spontaneous skyrmion lattices in an Fe monolayer in both fcc and hcp stacking on the Ir(111) surface employing first-principles calculations based on density functional theory (DFT) [8]. For fcc-Fe/Ir(111) the well-known non-collinear square nanoskyrmion lattice [3] is obtained as the magnetic ground state. However, for hcp-Fe/Ir(111) the hexagonal skyrmion lattice (Fig. 1(b)) previously proposed based on experiments [9] is energetically unfavorable with respect to a collinear arrangement of all spins (Fig. 1(c)). This leads to a hexagonal multi-Q state with nearly collinear magnetic moments which is similar to the hexagonal spin structures discovered in Fe/Rh bilayers on Ir(111) [10]. By mapping total DFT energies of a variety of complex magnetic states to an atomistic spin model we reveal the interplay of pairwise Heisenberg exchange, Dzyaloshinskii-Moriya interaction and higher-order exchange interactions to be responsible for the symmetry and the degree of collinearity of the respective spin lattice.

## References

[1] Mühlbauer *et al.*, *Science* **323** (2009) 915.
[2] X. Yu *et al.*, *Nature* **465** (2010) 901.
[3] S. Heinze *et al.*, *Nat. Phys.* **7** (2011) 713-718.
[4] C. Moreau-Luchaire *et al.*, *Nat. Nanotechnol.* **11** (2016) 444.
[5] O. Boulle *et al., Nat. Nanotechnol.* ***11*** (2016) 449.
[6] A. Fert *et al., Nat. Rev. Mater.* **2** (2017) 17031.
[7] C. Back *et al., J. Phys. D: Appl. Phys.* **53** (2020) 363001.
[8] M. Gutzeit *et al.*, arXiv: 2301.13851 (2023)
[9] K. von Bergmann *et al.*, *Nano Lett.* **15** (2015) 3280.
[10] M. Gutzeit *et al.*, *Nat. Commun.* **13** (2022) 5764.

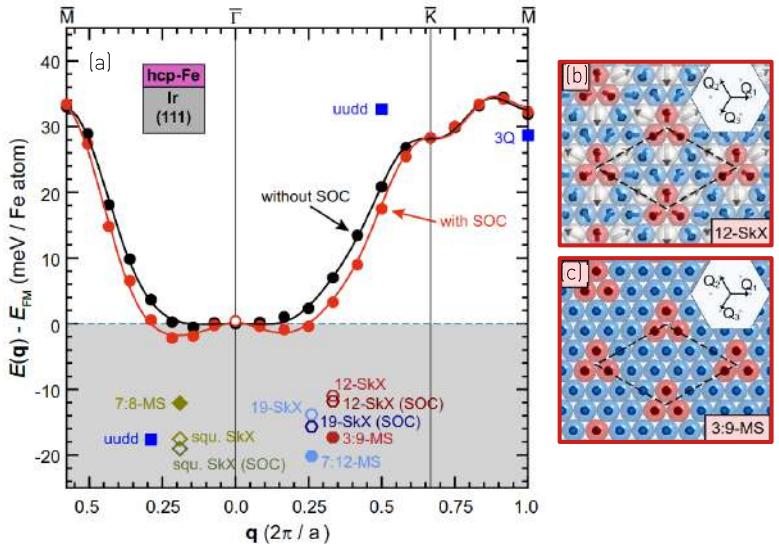



Figure 1: (a) Energy dispersion of flat cycloidal spin spirals for hcp-Fe/Ir(111) calculated via DFT, (b) sketch of the non-collinear nanoskyrmion lattice (12-SkX) based on experimental measurements [9] and (c) its respective collinear multi-Q state (3:9-MS).

# Probing the Internal Texture of Skyrmions through Spin Waves with a Quantum Sensor

*Aurore Finco[1]*, Pawan Kumar[1], Van Tuong Pham[2], Olivier Boulle[2], Joo-Von Kim[3], Vincent Jacques[1]*

[1] *Laboratoire Charles Coulomb, CNRS, Université de Montpellier, Montpellier, France*
[2] *Université Grenoble Alpes, CEA, CNRS, Grenoble INP, IRIG-SPINTEC, Grenoble, France*
[3] *Centre de Nanosciences et de Nanotechnologies, CNRS, Université Paris-Saclay, Palaiseau, France*
*aurore.finco@umontpellier.fr

NV centers are defects in diamond which can be used as quantum sensors to probe magnetism at the nanoscale when integrated in an atomic force microscope. Such a measurement relies on the spin S = 1 of the NV center: the static stray field produced by a magnetic state induces a Zeeman shift on the spin sublevels, which can be detected optically. In addition, NV centers are also sensitive to spin waves, as the magnetic noise originating from thermally activated spin waves accelerates its spin relaxation.

In the latter case, the enhanced relaxation leads to a decrease of the photoluminescence emitted by the NV center [1], which allows an easy localization of spin waves interacting with magnetic textures. We applied this approach to the study of Co-based perfectly compensated synthetic antiferromagnetic layers [2], in which we were able to observe spin waves channeled inside the domain walls [3].

We report here on a more detailed investigation of skyrmions in synthetic antiferromagnetic layers, revealing a spatial distribution of the noise contrast around their boundary. Both the noise distribution and its intensity appear to be linked to the skyrmion's internal structure.

## References

[1] M. Rollo, A. Finco, R. Tanos, F. Fabre, T. Devolder, I. Robert-Philip, and V. Jacques, *Physical Review B 103,* 235418 (2021).
[2] W. Legrand, D. Maccariello, F. Ajejas, S. Collin, A. Vecchiola, K. Bouzehouane, N. Reyren, V. Cros, and A. Fert, *Nature Materials 19,* 34–42 (2020).
[3] A. Finco, A. Haykal, R. Tanos, F. Fabre, S. Chouaieb, W. Akhtar, I. Robert-Philip, W. Legrand, F. Ajejas, K. Bouzehouane, N. Reyren, T. Devolder, J.-P. Adam, J.-V. Kim, V. Cros, and V. Jacques, *Nature Communications 12,* 767 (2021).

# Controlled Domain Wall Propagation in Pt/Co/Pt Nanostrips

*Rodrigo Guedas[1]*, Adrián Gudín[2], Paolo Perna[2], Víctor Raposo[3] and José Luis Prieto[1]*
[1] *Instituto de Sistemas Optoelectrónicos y Microtecnología (ISOM), Universidad Politécnica de Madrid, Avda. Complutense 30, 28040 Madrid, Spain*
[2] *IMDEA Nanociencia, Campus de cantoblanco, 28049 Madrid, Spain*
[3] *Departamento de física Aplicada, University of Salamanca, Plaza de la Merced, s/n, E-37008, Salamanca, Spain*
*corresponding author: rodrigo.guedas@upm.es

Materials with high Perpendicular Magnetic Anisotropy (PMA) have the potential to play an important role in future magnetic memories or logic devices. Their stability, together with the possibility of manipulating the magnetization via Spin Oribt Torques (SOTs)[1] make them key players in spintronics. A common structure used in these devices is sandwiching a thin film of a ferromagnetic material (FM) with a Heavy Metal (HM) and a Non-magnetic (NM) layer, HM/FM/NM. This structure provides a great Spin-Orbit Coupling which leads to the Spin Hall Effect (SHE)[2] in the HM/FM interface and Rashba Effect (RE)[3] as there is an inversion asymmetry in the structure.

Here we investigate the nucleation and propagation of Domain Walls (DWs) in a strip made out of Pt/(4)/Co(1)/Pt(8), at fields considerably smaller than the coercive field. DW nucleation is done via the local field and heat generated by a current line [4], together with a small external field (~200 Oe). Fig.a shows how the propagation field of the generated DWs is decreased by the action of a directional electric current via the RE and SHE with the assistance of Joule Heating in a nanostrip with very large coercivity (see inset). Fig.b provides MOKE snapshots showing the motion of a unique DW under the same conditions as those shown in figure a. Finally, realistic micromagnetic simulations are performed where the nucleation of the DW takes place by local demagnetization of the nanostrip and its propagation is driven solely by SOTs. The control of the DW nucleation and its propagation is one of the most active topics in spintronics and it can help design optimum shift registers or logic devices.

## Acknowledgements

This work was supported by projects MAT2017-87072-C4-3-P, MAT2017-87072-C4-1-9 and SA114P20. The authors acknowledge the support from Micronanofabs.

## References

[1] I. M. Miron, *Nature* **476** (2011) 189–193.
[2] E.I.Rashba, *Sov. Phys. Solid State* **2** (1960) 1109.
[3] J.E. Hirsh, *Phys. Rev. Lett.* **83** (1999) 1834–1837.
[4] O. Alejos, *Sci. Rep.* **7** (2017) 11909.

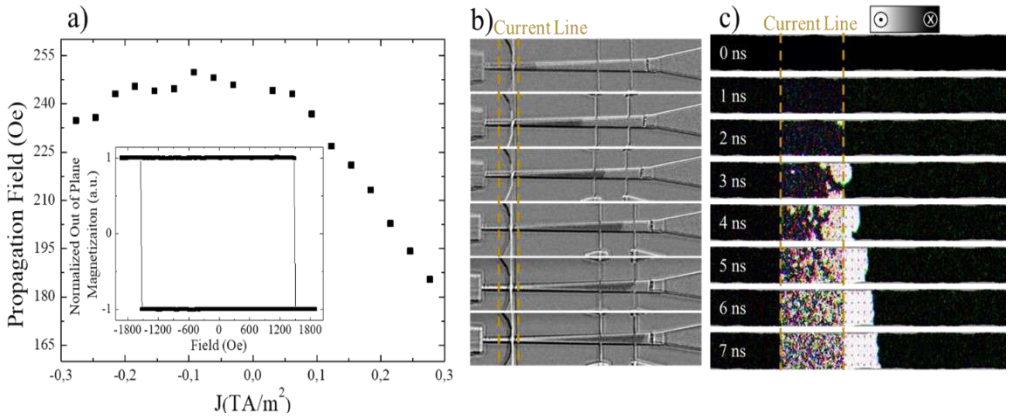



Figure: Domain Wall propagation in a Pt(4)/Co(1)/Pt(8) nanostrip. a) The propagation field is decreased by the action of an electric current density. The inset shows the hysteresis loop of this nanostrip. b) MOKE snapshots of a DW propagation in this nanostrip at a field of 200 Oe and c) Realistic micromagnetic simulations showing the nucleation and propagation of the domain wall under the same conditions of the experiment.

# Domain Wall Motion in Sputtered Terbium Iron Garnet Thin Films

*Stefano Fedel*[1]*, *Can Onur Avci*[1]
[1]**Institut de Ciència de Materials de Barcelona (ICMAB-CSIC), 08193 Bellaterra, Spain*
*sfedel@icmab.es

Magnetic domain walls (DW) motion in thin films has recently found renewed interest in spintronics thanks to the latest research on ferrimagnetic materials [1]. Especially heavy metal/insulating ferrimagnet hybrid heterostructures with perpendicular magnetic anisotropy (PMA) have been shown to exhibit suitable properties for fast and efficient current-induced DW motion [2]. Rare-earth iron garnets are distinguished by the ability to tune relevant magnetic properties such as saturation magnetization, magnetic and angular momentum compensation temperature, etc. [3]. Unlike in ferromagnetic counterparts, in ferrimagnets, the magnetic sublattices couple antiferromagnetically and prevent the net moment from precessing. This effect leads to extremely fast DW motion when the angular momentum is fully compensated [4].

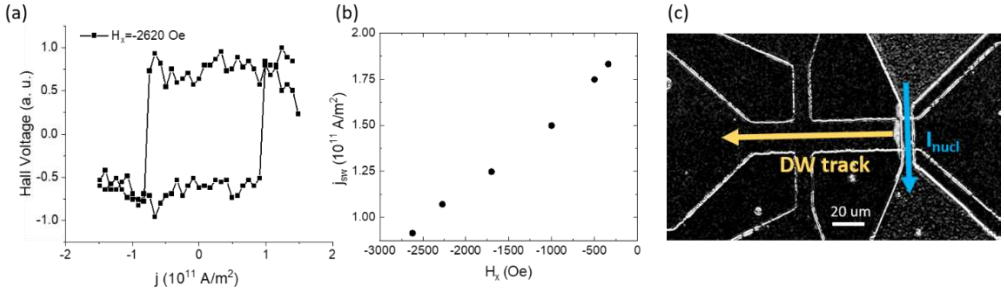
In this work, we study current-induced magnetization switching and DW motion in epitaxial Terbium Iron Garnet (TbIG) capped with Pt, both grown by magnetron sputtering. TbIG is a suitable material choice for current-induced magnetic manipulation experiments owing to its low saturation magnetization and compensation temperature close to room temperature, which can be tuned through deposition conditions [5]. The interfaces of TbIG with the substrate and Pt give rise to the Dzyaloshinskii-Moriya interaction (DMI), which stabilizes Neèl type DW. These DW can be moved by spin-orbit torques generated by the spin accumulation at the interface with Pt. First, magnetotransport and switching measurements are carried out in a Hall setup. The switching current density has been characterized as a function of different in-plane external magnetic field $H_x$, and a very low value of $j_{sw}=8\times10^{10}$ A/m$^2$ is reported for $H_x$=2600 Oe, indicating efficient spin-orbit torques and good interface quality. A home-built bright-field Kerr microscope is used to study and characterize DW motion (work in progress). This setup allows us to quantify the DW velocity under different external magnetic fields, and also to estimate the effective DMI. Moreover, temperature-depended measurements (in a range of -190ºC to 100ºC) are planned, thanks to a custom cryostat implemented in the setup.

**Acknowledgements:**

The authors acknowledge funding from the European Research Council (ERC) under the European Union's Horizon 2020 research and innovation programme (project MAGNEPIC, grant agreement No. 949052).

**References:**

[1] Kim S. K., et al. *Nat. mat.* **21(1)** (2022) 24-34.
[2] Avci C. O., et al. *Nat. nanotech.* **14(6)** (2019) 561-566.
[3] Dionne G. F. Magnetic Oxides. Springer New York, NY (2009).
[4] Caretta L., et al. *Nat. nanotech.* **13(12)** (2018) 1154-1160.
[5] Damerio S., & Avci C. O. *J. Appl. Phys*. **133(7)** (2023) 073902.




SOT switching of TbIG(7nm)/Pt(4nm): (a) Hall voltage measured for a fixed in-plane field $H_x$ during a sweep of current pulses $j$ to define the switching threshold $j_{sw}$, and (b) relationship between switching current $j_{sw}$ and $H_x$. (c) Track for DW motion in TbIG acquired with MOKE: injecting a current along the nucleation line (blue arrow) generates an inverted domain (bright area) that can be moved by SOT injecting current pulses along the DW track (yellow arrow).

# Symmetric skyrmion Hall effect in GdFeCo thin films

*Yusuke Nagata[1], Satoshi Sumi[1], Kenji Tanabe[1], and Hiroyuki Awano[1]**

[1] *Toyota Technological Institute, 2-12-1 Hisakata Tempaku, Nagoya, Aichi JAPAN*

*awano@toyota-ti.ac.jp

Magnetic skyrmions are being actively studied as future high-performance memory. Fine magnetic skyrmions exist in good ferrimagnetic crystals and can be driven by current control. However, this magnetic skyrmion has a problem in that it advances in a fixed direction different from the current direction due to the influence of the skyrmion Hall effect. Therefore, a method has been proposed in which a ferrimagnetic material is used to prevent the skyrmion Hall effect from appearing at the angular momentum compensation temperature [1]. This paper shows that the magnetic skyrmion bends to the left below the angular momentum compensation temperature and to the right above the angular momentum compensation temperature. In other words, if the angular momentum is determined, the skyrmion Hall angle is also determined to be one. However, in our ferrimagnetic GdFeCo thin film, we found a phenomenon in which two symmetrical skyrmion Hall angles occur at the same rate in spite of a single angular momentum.

A GdFeCo thin film with a thickness of 50 nm was formed on a silicon substrate with a $SiO_2$ film thickness of 350 nm by high-vacuum magnetron sputtering. In order to protect against oxidation, a 10 nm SiN layer was deposited on the underlayer and protective film of the GdFeCo layer. The size of magnetic skyrmions is about 1 μm. Fig. 1(a) shows the migration trajectories of magnetic skyrmions when a current is applied to the right side of the GdFeCo thin film with some magnetic skyrmions. As can be seen from the figure, the skyrmion hole angle is generally small. However, it can be seen that this result is greatly bent at nearly 80 degrees with respect to the current application direction. Moreover, some magnetic skyrmions are bent at about +80 degrees, and another magnetic skyrmions are bent at -80 degrees, symmetrically with respect to the electric current. The movement of any magnetic skyrmion has a skyrmion hole angle of about plus or minus 80 degrees, and movement of angles other than that is not observed. Fig. 1(b) shows the result of investigating how the skyrmion Hall angle changes with increasing current density. When the current density is increased, the skyrmion Hall angle decreases to about plus or minus 30 degrees. In this case as well, no other skyrmion Hall angles are found. Since there is no DMI supply layer like the Pt layer, the magnetic skyrmion domain wall becomes Bloch type. It is thought that 50% of the right-handed Bloch domain wall and 50% of the left-handed Bloch domain wall were generated during this magnetic skyrmion generation process, but this has not been verified.

## References

[1] Y. Hirata, et al, *Nat.Nanotech.* **14** (2019) 232-236.

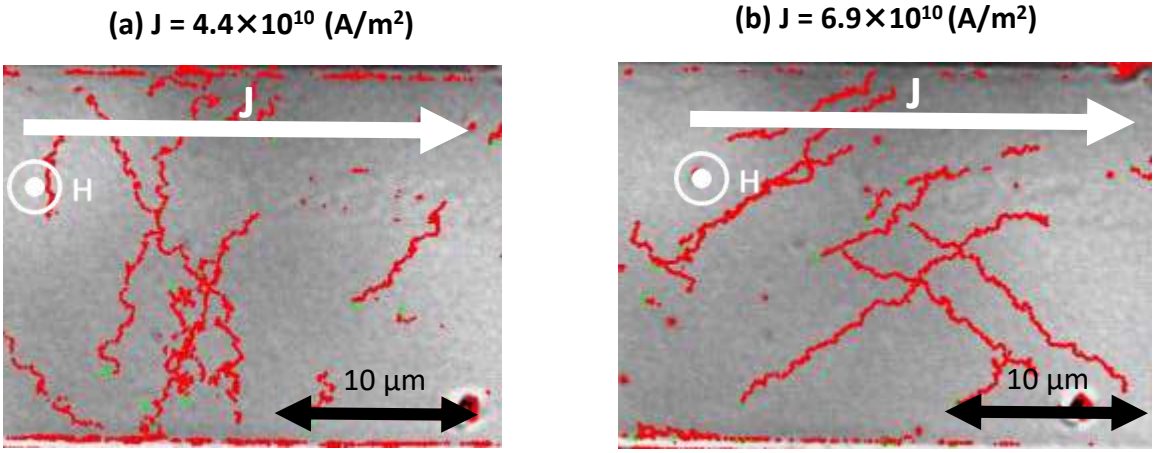



Figure: (a) A polarizing microscope photograph of the movement trajectory of magnetic skyrmions when a current (current density $4.4x10^{10}A/m^2$) is applied to magnetic skyrmions in a GdFeCo thin film. (b) is a polarizing microscope photograph of the movement trajectory of magnetic skyrmions when a current density of $6.7x10^{10}$ $A/m^2$ is applied.

# Improving Néel domain walls dynamics and skyrmion stability using $He^+$ ion irradiation

*Cristina Balan[1], Johannes W. van der Jagt[2,3], Vincent Jeudy[4], André Thiaville[4], Jan Vogel[1], Laurent Ranno[1], Dafiné Ravelosona [2, 3] and Stefania Pizzini[1]**

[1] *Univ. Grenoble Alpes, CNRS, Institut Néel, 38042 Grenoble, France*

[2] *Spin-Ion Technologies; 10 Boulevard Thomas Gobert, 91120 Palaiseau, France*

[3] *Centre de Nanosciences et de Nanotechnologies, CNRS, Université Paris-Saclay, 10 boulevard Thomas Gobert, 91120 Palaiseau, France*

[4] *Laboratoire de Physique des Solides, Université Paris-Saclay, CNRS, 91405 Orsay, France*

*stefania.pizzini@neel.cnrs.fr

Irradiation with light ions is an appealing way to finely tune the magnetic properties of thin magnetic films and in particular the interfacial perpendicular magnetic anisotropy (PMA). Early studies on Pt/Co/Pt multilayers showed that $He^+$ ions provoke short-range atomic displacements through low energy collisions. The resulting intermixing at the Co/Pt interfaces decreases the interfacial PMA, since the latter depends on the anisotropy of the chemical environment [1]. We will report the study of the effect of $He^+$ ion irradiation on the dynamics of Néel walls and the stability of skyrmions in Pt/Co/AlOx trilayers with ultrathin Co layers [2]. $He^+$ fluences up to $1.5x10^{15}$ ions/cm$^2$ strongly decrease the PMA, without affecting neither the spontaneous magnetisation nor the strength of the Dzyaloshinskii-Moriya interaction (DMI). This confirms the robustness of the DMI interaction against interfacial chemical intermixing, already predicted by theory [3]. In parallel with the decrease of the PMA in the irradiated samples, a strong decrease of the depinning field is observed. This allows the domain walls to reach large maximum velocities with lower magnetic fields with respect to those needed for the pristine films [4]. Decoupling PMA from DMI can therefore be beneficial for the design of low energy devices based on domain wall dynamics. By studying a Pt/Co/AlOx wedge sample with a gradient of Co oxidation, we demonstrate that the modifications of the magnetic properties driven by irradiation are due not only to the intermixing of the Pt/Co interface, but also to an homogeneisation of the oxidation of the Co/AlOx interface. When the samples are irradiated with larger $He^+$ fluences, the magnetisation gets close to the out-of-plane/in-plane reorientation transition, where ≈100 nm size stripe domains and skyrmions are stabilised. As the $He^+$ fluence increases, the skyrmion size decreases and the magnetic textures become more and more stable against the application of an external magnetic field.

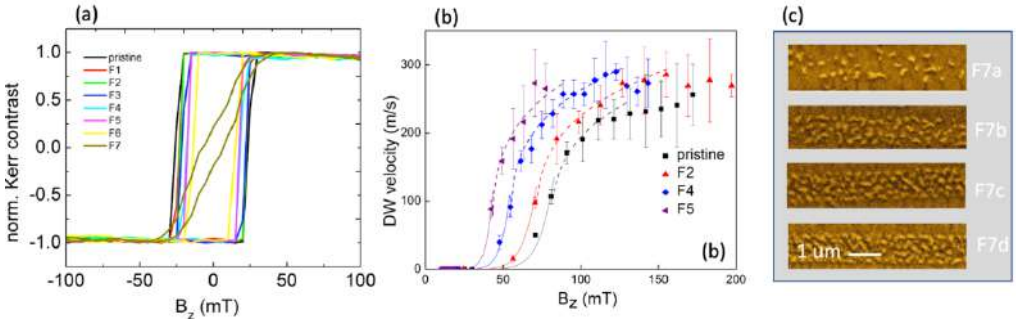



**Figure:** (a) MOKE hysteresis loops for increasing $He^+$ fluence (from F1=$2x10^{14}$ to F7=$3x10^{15}$ He+/cm$^2$ ); (b) domain wall velocity in the pristine state and after irradiation; (c) magnetic skyrmions under the application of an out-of-plane magnetic field $B_z$=5mT, in the Pt/Co/AlOx sample after the irradiation with increasing fluences F7a= $2.5x10^{15}$, F7b= $2.7x10^{15}$, F7c= $2.9x10^{15}$, F7d= $3.2x10^{15}$ ions/cm$^2$.

## References

[1] C. Chappert, H. Bernas, J. Ferre, V. Kottler, J.-P. Jamet, Y. Chen, E. Cambril, T. Devolder, F. Rousseaux, V. Mathet, H. Launois, Science, 280, 1919 (1998).

[2] C. Balan *et al.*, arXiv:2303.04543

[3] B. Zimmermann, W. Legrand, D. Maccariello, N. Reyren, V. Cros, S. Blügel, and A. Fert, Applied Physics Letters, 113, 232403 (2018).

[4] T. H. Pham, J. Vogel, J. Sampaio, M. Vanatka, J-C Rojas-Sanchez, M. Bonfim, D. S. Chaves, F. Choueikani, P. Ohresser, E. Otero, A. Thiaville, and S. Pizzini. EPL, 113, 67001 (2016).

# Domain wall chirality reversal by interfacial engineering in Pt/Co/Pt based perpendicularly magnetized systems

*Saikat Maji[1]*, Ankan Mukhopadhyay[1], Soubhik Kayal[1], P S Anil Kumar[1]*

[1] *Indian Institute of Science Bangalore, C V Raman Road, Bangalore, Karnataka, India - 560012*

*saikatmaji@iisc.ac.in

Perpendicular magnetic anisotropy (PMA) systems that involve heavy metal (HM)/ferromagnet (FM)/heavy metal (HM) tri-layer structure have immense scope for constructing high speed memory devices [1,2]. Broken inversion symmetry at HM/FM interface introduces interfacial Dzyaloshinskii-Moriya interaction (iDMI) which stabilizes chiral Neel wall that can be manipulated with spin polarized current. The current induced deterministic switching of PMA systems also realized with or without application of bias magnetic field. In this work, we have used field induced domain wall motion to estimate iDMI strength, $D_{eff}$ of sputter deposited Ta (3.5 nm)/Pt (3 nm)/Co (0.4 nm)/Au (x nm)/Pt (1 nm) thin films where x = 0, 0.3, 0.5 and 0.7 nm. The domain wall velocity of a nucleated bubble domain was estimated applying pulsed $H_z$ in presence of $H_x$. The asymmetric expansion of bubble domain reveals the chirality of domain wall of the samples. Ta/Pt/Co/Pt systems has Neel walls with right-handed chirality whereas the chirality reverses to left handed with introduction of ultrathin Au layer as shown in Fig. 1(a), (b), (c) and (d).

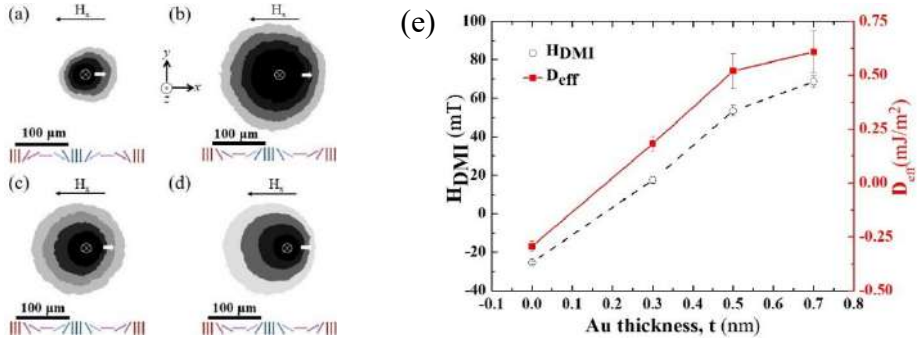



Figure 1: Asymmetric expansion of bubble domain at an in-plane magnetic field, $H_x$ = 30 mT. (a) Au (t = 0 nm), (b) Au (t = 0.3 nm), (c) Au (t = 0.5 nm) and (d) Au (t = 0.7 nm). The magnetization configuration inside the domain wall (x-z plane) is shown under each image. The domain wall velocity along x-direction is indicated with white arrow. The magnetization configuration reveals the chirality of domain wall. (e) Effective in-plane field of iDMI, $H_{DMI}$ and iDMI strength, $D_{eff}$ as a function of Au layer thickness. iDMI sign reverses with introduction of ultrathin layer of Au.

The iDMI can be manifested as an effective in-plane magnetic field ($H_{DMI}$) in PMA system. The $H_x$ dependence of the domain wall velocity has a minimum at $\mathbf{H_x = H_{DMI}}$. iDMI strength calculated using the formula $\mathbf{D_{eff} = \mu_0 H_{DMI} M_s \lambda}$ as described by Je et. al [3] and displayed in Fig. 1(e). where Ms is saturation magnetization and λ is the domain wall width. The $D_{eff}$ of symmetric Ta/Pt/Co/Pt system have very small negative value which can be attributed to the difference of roughness of bottom Pt/Co and top Co/Pt interface. $D_{eff}$ increases as the asymmetry around Co layer was introduced with insertion of Au layer. The maximum $D_{eff}$ was observed for the sample with highest degree of asymmetry (Au thickness = 0.7 nm). The $D_{eff}$ also becomes positive when Au is introduced at top Co/Pt interface. The Pt/Co and Au/Co interface has opposite polarity of iDMI [4]. These two interfaces of Ta/Pt/Co/Au/Pt effectively reverse the iDMI sign.

## Acknowledgements

We acknowledge NNFC and MNCF for sample prepareation and measurement facilities. We also thank Prof. R. A. Duine for discussion on DMI.

## References

[1] S. S. Parkin, M. Hayashi, and L. Thomas, Science 320, 190–194 (2008)
[2] A. Fert, V. Cros, and J. Sampaio, Nature Nanotechnology 8, 152–156 (2013)
[3] S. G. Je, K. J. Lee, and S. B. Choe, Physical Review B 88, 214401 (2013)
[4] H. Yang, A. Fert, and M. Chshiev, Phys. Rev. Lett. 115, 267210 (2015)

# $CrTe_2$ as a two dimensional layer for topological magnetism in complex heterostructures

*Nihad AbuAwwad,[1,2]*, Manuel dos Santos Dias[3], Samir Lounis[1,2*]*

[1] *Peter Grünberg Institute (PGI), FZJ, 42425 Julich, Germany*
[2] *Faculty of Physics, University of Duisburg-Essen, 47053 Duisburg, Germany*
[3]*Scientific Computing Department, STFC Daresbury Laboratory, Warrington WA4 4AD, United Kingdom*

* n.abuawwad@fz-juelich.de, s.lounis@fz-juelich.de

The discovery of two-dimensional (2D) van der Waals magnetic materials and their heterostructures provided an exciting platform for emerging phenomena with intriguing implications in information technology. $CrTe_2$ is a particular example that hosts complex magnetism strongly intertwined with its crystal structures [1,2]. Here, based on a multiscale modelling approach that combines first-principles calculations and a Heisenberg model, we demonstrate that interfacing this 2D layer with various Te-based layers hosting heavy or light elements enables the control of the Dzyaloshinskii-Moriya interaction and magnetic anisotropy energy of the whole heterostructure, and thereby the emergence of new magnetic phases of matter, which are of topological nature such as skyrmions and merons.

## Acknowledgements

Work funded by the Palestinian-German Science Bridge (BMBF–01DH16027) and Priority Programme SPP 2244 2D Materials Physicsof van der Waals Heterostructures of the DFG (project LO 1659/7-1).

## References

[1] AbuAwwad et al., J. Phys.: Condens. Matter 34, 454001(2022).
[2] Xian et al., Nat. Commun. 13, 257 (2022).

# Lifetime of coexisting sub-10 nm zero-field skyrmions and antiskyrmions

*Moritz A. Goerzen*[1*], *Stephan von Malottki*[1,2,3], *Sebastian Meyer*[1,4], *Pavel F. Bessarab*[2,5] *and Stefan Heinze*[1]
[1] *Institute of Theoretical Physics and Astrophysics, University of Kiel, Kiel, Germany*
[2] *Science Institute of the University of Iceland, Reykjavík, Iceland*
[3] *Thayer School of Engineering, Dartmouth College, Hanover, New Hampshire, USA*
[4] *Nanomat/Q-mat/CESAM Université de Liège, Sart Tilman, Belgium*
[5] *Department of Physics and Electrical Engineering, Linnaeus University, Kalmar, Sweden*
*goerzen@physik.uni-kiel.de

Magnetic skyrmions have raised high hopes for future spintronic devices [1,2]. For many applications it would be of great advantage to have more than one metastable particle-like texture available [3]. The coexistence of skyrmions and antiskyrmions has been proposed in inversion symmetric magnets with exchange frustration [4,5]. However, so far only model systems have been studied, an applied magnetic field was required, and the lifetime of coexisting metastable topological spin structures has not been obtained. Here, we predict that skyrmions and antiskyrmions (Fig.1a,b) with diameters below 10 nm can coexist at zero magnetic field in a Rh/Co bilayer on the Ir(111) surface – an experimentally feasible ultrathin film system [6,7]. Based on transition state theory [8] we show that the lifetimes of metastable skyrmions and antiskyrmions (Fig.1e) in the ferromagnetic ground state are above one hour for temperatures up to 75 K and 48 K, respectively. We further demonstrate that the entropic contribution to the nucleation and annihilation rates differs for skyrmions and antiskyrmions. This opens the route to thermally activated creation of coexisting skyrmions and antiskyrmions in frustrated magnets with Dzyaloshinskii-Moriya interaction.

## References

[1] A. Fert, N. Reyren, V. Cros, *Nat. Rev. Mater.* **2** (2017) 17031.
[2] C. Back et al., *J. Phys. D: Appl. Phys.* **53** (2020) 363001.
[3] B. Göbel, I. Mertig, O. A. Tretiakov, *Phys. Rep.* **895** (2021) 1.
[4] A. O. Leonov, M. Mostovoy, *Nat. Commun.* **6** (2015) 8275.
[5] S.-Z. Lin, S. Hayami, *Phys. Rev. B* **93** (2016) 064430.
[6] S. Meyer et al., *Nat. Commun.* **10** (2019) 3823.
[7] M. Perini et al., *Phys. Rev. Lett.* **123** (2019) 237205.
[8] P. F. Bessarab, V. M. Uzdin, H. Jónsson, *Phys. Rev. B* **85** (2012) 184409.

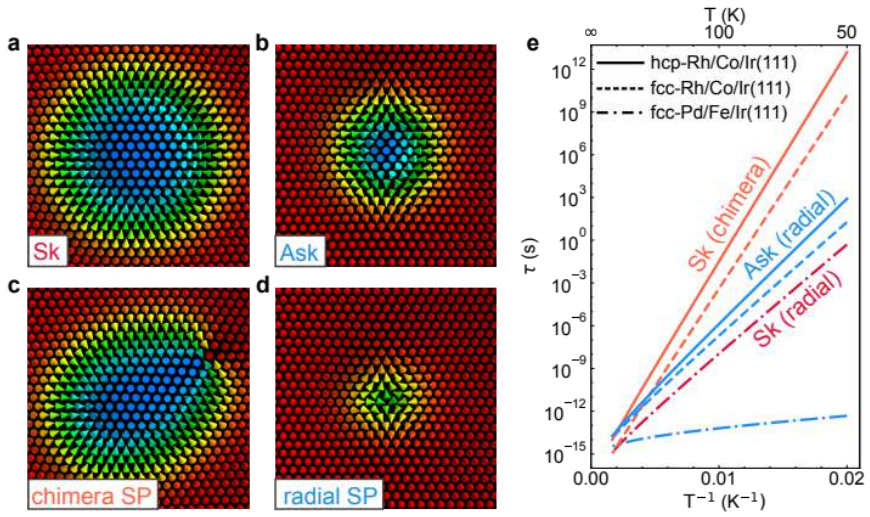



Figure 1: (a) Isolated skyrmion (Sk) and (b) isolated antiskyrmion (Ask) in fcc-Rh/Co/Ir(111) at B = 0 T and (c) respective Chimera (skyrmion) saddle point and (d) antiskyrmion saddle point. (a-d) show equally sized parts of the much larger respective simulation boxes. (e) Arrhenius plot of the lifetime of skyrmions (orange and red lines) and antiskyrmions (blue lines) in hcp-Rh/Co/Ir(111), in fcc-Rh/Co/Ir(111) at $B = 0$ T and for comparison in Pd/Fe/Ir(111) at $B = 3.9$ T.

# Control of Domain Wall Chirality in a Double Wedge System

*Capucine Gueneau[1*], Johanna Fischer[1], Charles-Elie Fillion[1], Fatima Ibrahim[1], Raj Kumar[1], Libor Vojacek[1], Stefania Pizzini[2], Laurent Ranno[2], Djoudi Ourdani[3], Mohamed Belmeguenai[3], Yves Roussigné[3], Salim-Mourad Chérif[3], Stéphane Auffret[1], Isabelle Joumard[1], Olivier Boulle[1], Gilles Gaudin[1], Mairbek Chshiev[1,4], Liliana D. Buda-Prejbeanu[1], Claire Baraduc[1], Hélène Béa[1,4]*

[1] *Univ. Grenoble Alpes, CNRS, CEA, SPINTEC, Grenoble, France*
[2] *Univ. Grenoble Alpes, CNRS, Institut NEEL, Grenoble, France*
[3] *Laboratoire des Sciences des Procédés et des Matériaux (LSPM), Villetaneuse, France*
[4] *Institut Universitaire de France (IUF), France*
*Capucine.gueneau@cea.fr

The sense of the gradual rotation of the magnetic moments within a Domain Wall (DW) is associated with the chirality. In ultrathin Heavy Metal/Ferromagnet/Metal Oxide (HM/FM/$MO_x$) heterostructures with Perpendicular Magnetic Anisotropy (PMA), interfacial Dzyaloshinskii-Moriya interaction (iDMI) generally plays an important role in the stabilization of domain walls with a given chirality [1]. This antisymmetric exchange interaction arises from the combination of spin-orbit coupling and structural inversion asymmetry at HM/FM and FM/$MO_x$ interfaces. The sign of the iDMI determines the chirality of DWs (Fig. 1a). Hence, a fine tuning of interfaces allows a control of the iDMI sign along with the chirality of the magnetic domain walls.

In this work, we present a complete study of the chirality of DWs and the iDMI sign in Ta/FeCoB/$TaO_x$ tri-layer system with a double wedge structure. The FeCoB layer is deposited with a thickness gradient perpendicular to a second gradient of Ta which is subsequently oxidized (Fig. 1b). By combining a direct iDMI measurement (Brillouin Light Scattering) with the observation of the direction of DW motion induced by spin-orbit torques (using polar Magneto-Optical Kerr Effect microscope) [2], we recently showed that the sign of the iDMI is inverted both by a change in the oxidation state of the FeCoB/$TaO_x$ interface and the FeCoB thickness [3]. In regions where iDMI is too small to be measured, we used the current-induced DW motion direction to extract the chirality over the whole PMA region (Fig. 1c). Our observation of the iDMI sign transition (red dotted line in Fig. 1c) reveals a stair-like evolution, indicating the importance of the layer coverage percentage. With the help of *ab initio* calculations we are able to untangle the mechanisms for the iDMI sign change with both oxidation state and ferromagnetic thickness. These findings may help to design future devices with a controlled value and sign of iDMI in order to optimize domain wall mobility [3].

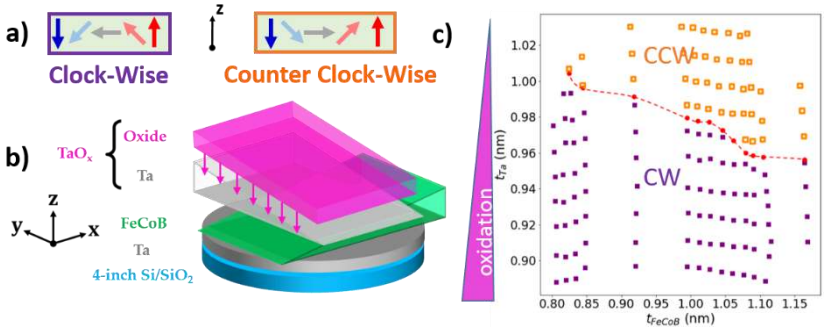



Fig.1.a) Representation of a Clock-Wise CW (left) and Counter Clock-Wise CCW (right) domain wall. b) Schematic representation of our double wedge sample. c) Map of the DW chirality with respect to FeCoB thickness ($t_{FeCoB}$) and oxidation state ($t_{Ta}$). Regions with CW (resp. CCW) DWs are represented with full purple squares (resp. empty orange squares) and iDMI sign crossover is represented by red circles. The dashed line is a guide to the eye.

## Acknowledgements

This work has been supported by ANR-19-CE24-0019 project ADMIS.

## References

[1] I. E. Dzyaloshinskii, *Exptl. Theoret. Phys. U.S.S.R.*, **46**, (1964) 1420; T. Moriya, *Phys. Rev.* **120** (1960) 91.
[2] S. Emori, et al., *Nat. Mater.* **12** (2013) 611.
[3] R. Kumar et al., *Phys. Rev. Applied.* **19** (2023) 024064.

# Time-resolved observations of a domain wall distortions induced by mechanical torsion

*Kornel Richter[2]*, Lucia Fečová[1], Rastislav Varga[2]*

[1] *Dep. of Physics, Pavol Jozef Safarik University, Kosice, 04154 Slovakia*

[2] *Centre of Progressive Materials, TIP P.J. Safarik University, Kosice 040 01, Slovakia*

*kornel.richter@gmail.com

Thin magnetic cylindrical wires attract considerable attention due to interesting features of a domain wall motion like absence of the Walker breakdown that prevents fast domain wall dynamics [1]. Amorphous glass-coated microwires are composite material with very high domain wall velocities [2]. Owing to the amorphous state of wires, the magneto crystalline anisotropy vanishes. Recently, it was shown that the axial magnetic anisotropy resulting from rapid quenching has comparable strength, leading to a tilted domain wall orientation [3]. However, it is an open question of how the tilted domain wall in cylindrical wires moves with application of mechanical torsion stresses.

In this contribution, we present direct observation of the domain wall distortions. Magneto-optical Kerr effect (MOKE) is used to visualize a surface magnetization dynamics in microwires. The domain wall is stabilized by two opposite magnetic fields, resulting in effective one-dimensional potential well. Periodic back-and-forth motion of the well is used to synchronize the domain wall position with MOKE imaging and direct time-resolved images of a surface magnetism are performed. In contrast to previous measurements, mechanical torsion is applied to the microwire. Our spatial imaging of a surface domain wall shape reveals two typical types of a DW distortions. In highly magnetostrictive alloy (e.g. FeSiB), the mechanical torsion stress results in a screw-like distortions of a DW (Fig. 1). On the other side, microwires with reduced magnetostriction exhibits almost zero distortion of a domain wall shape. In both cases, the magnetostriction plays an essential role because the mechanical distortion is always smaller than the distortion of a DW. Our results are confronted to the magnetooptical imaging by MOKE microscope.

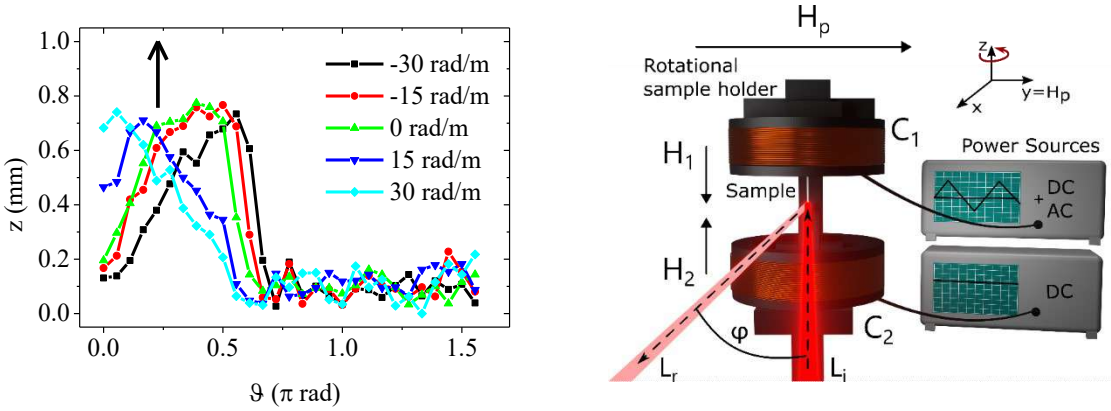



Fig. 1 (left) Snapshots of domain wall profiles for specific angles of torsion in FeSiB microwire. (right) schematic depiction of the MOKE setup used to visualize domain wall distortions

**Acknowledgements**

Part of this work was supported by the Slovak Research and Development Agency under contract No. APVV-17-0184, Slovak Research and Development Agency under contract No., APVV-20-0324 and Slovak Grant Agency VEGA 1/0195/18.

**References**

[1] M. Yan, et al., *Phys. Rev. Lett.* **104** (2010), 057201.
[2] P. Klein, et al., *J. Alloy. Comp.* **550** (2013), 31.
[3] O. Vahovsky, et al., *JMMM* **483** (2019), 266.

# Effect of Ga-Irradiation of Fe/Gd-Multilayers Hosting Skyrmions

*Timo Schmidt*[1]*, *Manfred Albrecht*[1]

[1] *Institute of Physics, University of Augsburg, Universitätsstraße 1, 86159 Augsburg, Germany*

*timo.schmidt@physik.uni-augsburg.de

The ferrimagnetic Fe/Gd antiskyrmion multilayer system, that is investigated, has been shown to be able to host magnetic spin textures like skyrmions, and trivial bubbles [1]. With the rise of scientific interest in the topic of magnetic spin textures over the last years the stabilization, but also the manipulation of these magnetic objects is of great interest for future spintronic applications. In this system the spin structures are stabilized by the magnetic dipolar interaction. Ion irradiation offers the possibility to change the structural and magnetic properties in order to influence the spin textures [2]. Using a Focus Ion Beam (FIB) system also allows to irradiate the sample locally on the nanoscale.

In principle, ion irradiation disrupts the multilayer structure and causes roughening and mixing of the individual layers. The out-of-plane (OOP) and in-plane (IP) hysteresis loops displayed in Figure 1a) show, that by this effect the OOP-anisotropy decreases with increasing irradiation intensity. The corresponding MFM-images confirm this trend. The magnetic domain structure observed in zero field before the irradiation exhibits a decreasing stripe period with increasing ion dose up to a point at which the stripe pattern is no longer observed.

The investigated Fe/Gd-system was prepared by sputter deposition on $Si/SiO_2$ substrates as well as silicon nitride membranes in order to enable imaging of the magnetic structure by Lorentz-Transmission Electron Microscopy (LTEM). These membranes were then irradiated locally in a grid geometry with Ga-ions utilizing a FIB-system. Figure 1b) shows a LTEM image aquired at an applied magnetic field of 260 mT of a membrane treated in this way. The irradiated areas forming the grid pattern are almost exclusively populated by magnetic bubbles while the unirradiated areas host skyrmions as well as bubbles.

## References

[1] M. Heigl, *et al.*, *Nat. Commun.* **12**, 2611 (2021).
[2] M. Krupinski, *et al.*, *Phys. Rev. Mater.* **5**, 024405 (2021).

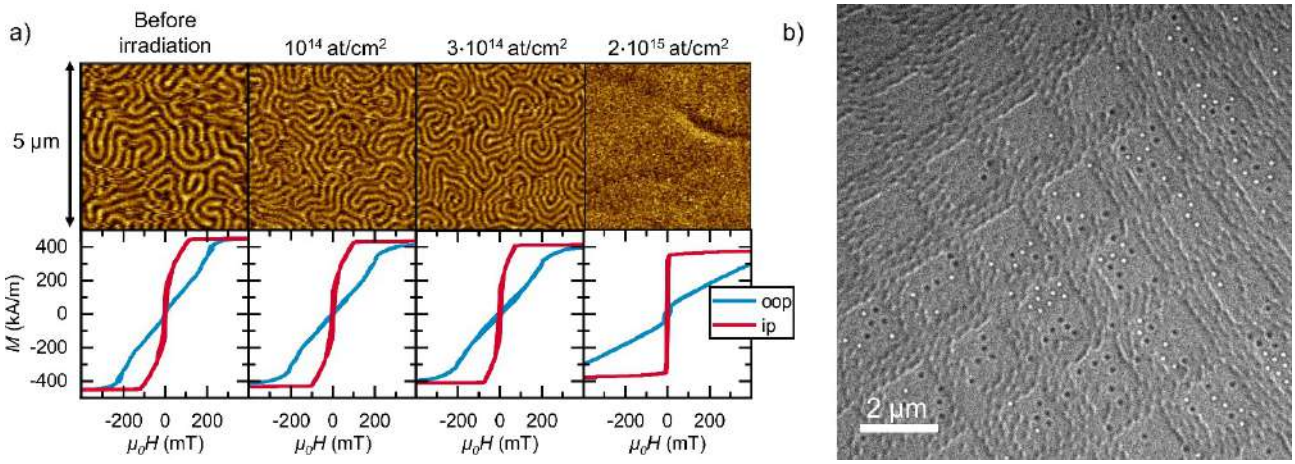



Figure 1: (a) MFM-images and M-H-hysteresis loops of a $[Fe/Gd]_{80}$-multilayer aquired for different irradiation doses. (b) LTEM image of a $[Fe/Gd]_{120}$-film locally irradiated with approx. $10^{14}$ ions/cm² in a grid pattern using a FIB system. The image was taken at 260 mT.

# Room-Temperature Skyrmion Survival in Granular Racetracks

*N. Del-Valle*, J. Castell-Queralt, L. Gonzalez-Gomez, and C. Navau*

*Departament de Física, Universitat Autònoma de Barcelona, 08193 Bellaterra, Barcelona, Catalonia, Spain*

*nuria.delvalle@uab.cat

Skyrmions can be envisioned as bits of information that can be transported along nanoracetracks. However, temperature, defects, and/or granularity can produce diffusion, pinning, and, in general, modification in their dynamics. These effects may cause undesired errors in information transport. To study the viability of these skyrmionic racetracks in real conditions (room temperature and granularity), and with feasible computational time, we use a deterministic, yet probabilistic, approach [1] to study the skyrmion dynamics in a granular nanoracetrack. First, we modeled the granularity of the system within the rigid model, computing the effective force generated over the skyrmion. Then, the Fokker-Planck equation of a skyrmion moving along the granular racetrack at room temperature is solved. From the results, we calculate some key magnitudes like the probabilities of success of the transport of information, that is, the success probability of a skyrmion traveling a given distance along the racetrack. The results are evaluated in terms of the eventual loss of skyrmions by pinning, destruction at the edges, or excessive delay due to granularity. The results could serve to establish error detection criteria and, in general, to discern the dynamics of skyrmions in realistic situations.

**Acknowledgements**

We acknowledge Spanish project PID2019-104670GB-I00 of Agencia Estatal de Investigación / Fondo Europeo de Desarrollo Regional (UE) for financial support. J. C.-Q. acknowledges a grant (FPU17/01970) from Ministerio de Ciencia, Innovación y Universidades (Spanish Government).

**References**

[1] J. Castell-Queralt, L. González-Gómez, N. Del-Valle, C. Navau. Deterministic approach to skyrmionic dynamics at nonzero temperatures: pinning sites and racetracks, *Phys. Rev. B (R)* **101** (2020) 140404.

# Demultiplexing Ferromagnetic Domain Walls: A micromagnetic study

*Luis Sánchez-Tejerina*[1]*, *Eduardo Martínez*[2], *Óscar Alejos*[1]

[1]*Departament of Electricity and Electronics, University of Valladolid, Spain*

[2]*Departament of Applied Physics, University of Salamanca, Spain*

*luis.sanchez-tejerina@uva.es

Racetrack memory is the paradigmatic proposal of a domain wall (DW) based device. Since its initial proposal [1] several works have studied how to optimize it. This optimization covers the domain size, the DW speed, the domain nucleation, or the DW motion efficiency among other features. Besides, several strategies for data processing and performing logic operations were suggested.

In this study, we numerically study the regime in which a train of DWs driven by spin-orbit torques can be demultiplexed by a Y-shape geometry on a ferromagnetic sample as shown in Figure 1(a). This geometry has been already proposed as a DW feeder [2]. In this case, we intend to operate in the opposite sense. An electric current is applied from the horizontal branch to either the upper (Figure 1(b)) or lower branch. For current densities larger than a threshold current, a DW is injected in the corresponding branch.

Remarkably, the required current is larger if an Up-Down DW is intended to be injected into the lower branch than into the upper branch. This threshold current difference becomes more relevant as the strip width is reduced. On the other hand, for wide enough strips ($w = 128$ nm) it is not possible to inject a transverse DW for large aperture angles. Another notable feature is the reduction of the asymptotic current density threshold with the width of the strip. However, this comes at the expense of larger pulse durations needed to achieve such an asymptotic limit.

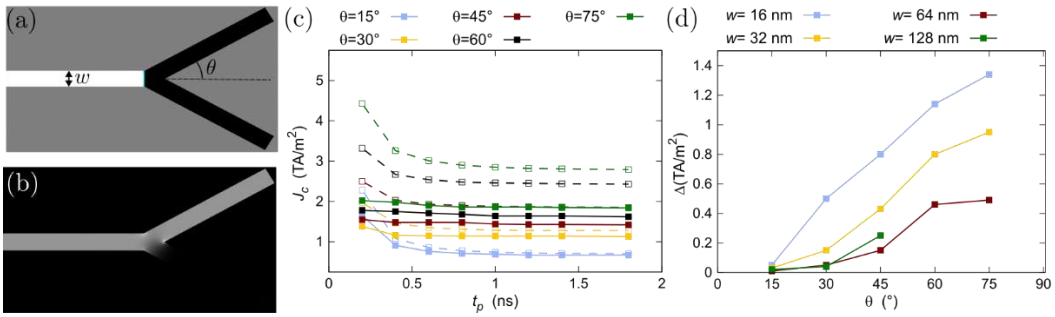



Figure 1: (a) Initial magnetization state for aperture half-angle $\theta = 30°$ and strip width $w = 32$ nm. (b) Normalized current density distribution for the case in which the electric current is injected in the upper branch calculated by COMSOL [3]. (c) Current density threshold to inject the DW in the upper branch (solid line) and lower branch (dashed line) for a strip width $w = 32$ nm. (d) Current density difference between the upper and lower branch thresholds.

In conclusion, we show that a precise control of the stimulus is needed to inject a DW towards one selected branch as the required current density for each branch differs. On the other hand, we expect such threshold difference to vanish for other materials such as antiferromagnets or ferrimagnets at the angular momentum compensation point. Additionally, we believe that this system can help to understand better DWs in 2D systems, whose theoretical studies are more scarce than the corresponding 1D systems.

## Acknowledgements

This work was supported by Projects No. PID2020117024GB-C41 funded by MCIN/AEI/10.13039/501100011033, No. SA114P20 from Junta de Castilla y León, and Project MagnEFi, Grant Agreement No. 860060, (H2020-MSCA-ITN-2019) funded by the European Commission.

## References

[1] S. S. P. Parkin, M. Hayashi, and L. Thomas. *Science*, **320**, (2008). doi:10.1126/science.1145799.

[2] D. Osuna Ruiz, O. Alejos, V. Raposo, and E. Martínez *Appl. Phys. Lett*. **121**, 102403 (2022)

[3] COMSOL Multiphysics® v. 5.6, www.comsol.com COMSOL AB, Stockholm, Sweden.

# Tuning of Room Temperature Skyrmions in Pt/Co/Ta Multilayers

*Liz Montanez[1], Valentin Ahrens[2], Laura Guasco[3], Thomas Keller[3], Markus Becherer[2], Sabine Pütter[1]**

[1] *Forschungszentrum Jülich GmbH, Jülich Centre for Neutron Science at MLZ, Garching, Germany*
[2] *Chair of Nano and Quantum Sensors, Department of Electrical and Computer Engineering, Technical University of Munich, Munich, Germany*
[3] *Max Planck Society Outstation at the Heinz Maier-Leibnitz Zentrum (MLZ), Garching, Germany*
*corresponding author email address

Magnetic skyrmions are nanoscale topological objects which are promising for next-generation information storage technologies and computing. [1,2] In magnetic multilayers, they can be stabilized at room temperature. [3-5]. Skyrmions emerge due to an interplay between several magnetic contributions. Among them the interfacial Dzyaloshinskii-Moriya Interaction (DMI) drives the spins into non-collinear orientation, while the perpendicular magnetic anisotropy (PMA) favours the out-of-plane orientation and the shape anisotropy prefers in-plane spin orientation. To study this competition of energies and the appearance of skyrmions, we have varied the Co film thickness as well as the number of repetitions in $[Pt/Co(x)/Ta]_N$ multilayers. This multilayer system was chosen because it is an established multilayer system for skyrmions and results can be compared with existing investigations, like e.g. [3,6].

Polycrystalline $[Pt(40\ Å)/Co(x)/Ta(19\ Å)]_N$ multilayers were fabricated in a molecular beam epitaxy setup by thermal deposition on oxidized Si(001) substrates with a buffer layer of 47 Å Ta and a 30 Å Pt cap layer. The Co film thickness was varied between 5 Å and 21 Å, the number of repetitions varied between 8 and 10.

Magnetic force microscopy measurements reveal the existence of skyrmions at a Co thickness between 9Å and 17 Å. The figure below gives examples with varying thickness and number of repetitions in indicated magnetic field. The density of skyrmions as well as their size varies. In remanence, stable skyrmions form only for the 17 Å Co sample with N=10 otherwise worm domains develop. Topological Hall effect measurements confirm these observations. The relationship between these findings is discussed in this contribution.

## References

[1] A. Fert, V. Cros, and J. Sampaio, *Nature Nanotech* **8**, (2013) 152.
[2] K. Raab, M.A. Brems, G. Beneke, *et al.*, *Nat Commun* **13**, (2022) 6982.
[3] S. Woo, K. Litzius, B. Krüger, M.-Y. Im, L. Caretta, K. Richter et al., Nat. Mat. 15 (2016) 501
[4] A. Soumyanarayanan, M. Raju, A. Gonzalez Oyarce, et al., *Nature Mater* 16 (2017) 898.
[5] T. Dohi, R. M. Reeve and M. Kläui, Annu Rev. Condens. Matter Phys. 13 (2022) 73.
[6] S. Zhang, J. Zhang, Y. Wen, E. M. Chudnovsky, and Y. Zhang, Comms. Phys. 36 (2018) 1.

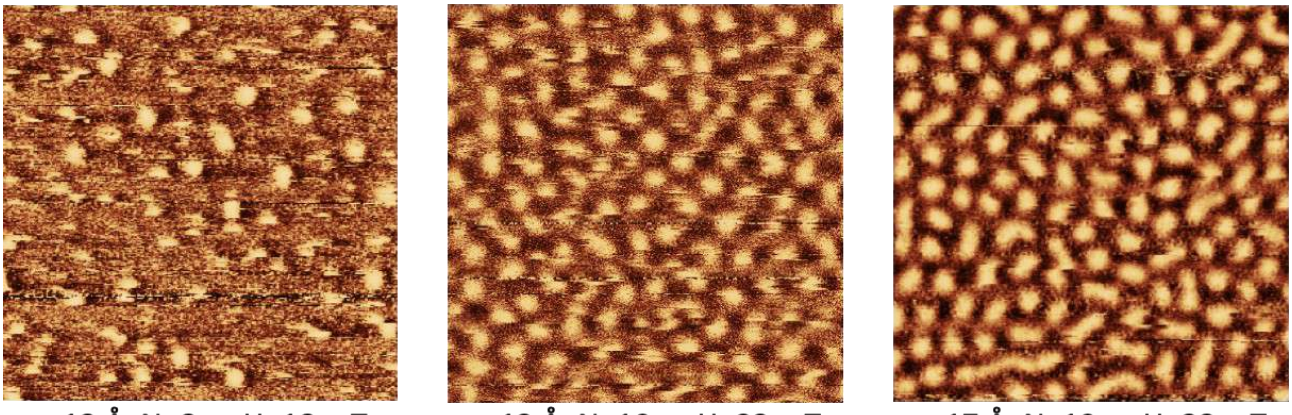



Figure: MFM images of $[Pt(40\ Å)/Co(x)/Ta(19\ Å)]_N$. The number of repetitions N, the Co thickness and the applied out-of-plane field are indicated below the images. The scan size is 3 μm x 3 μm.

# Skyrmion control for spintronic applications

*Johanna Fischer[1]*, Capucine Gueneau[1], Charles-Élie Fillion[1], Stefania Pizzini[2], Laurent Ranno[2], Stéphane Auffret[1], Isabelle Joumard[1], Olivier Boulle[1], Gilles Gaudin[1], Liliana D. Buda-Prejbeanu[1], Claire Baraduc[1], Hélène Béa[1,3]*

[1] *Univ. Grenoble Alpes, CNRS, CEA, SPINTEC, 17 Av. des Martyrs, Grenoble, France*
[2] *Univ. Grenoble Alpes, CNRS, Institut NEEL, 25 Av. des Martyrs, Grenoble, France*
[3] *Institut Universitaire de France (IUF), 103 Bd. Saint Michel, Paris, France*
*johanna.fischer@cea.fr

Magnetic skyrmions have emerged as interesting spin textures for the next generation of spintronic devices due to their promising potential in terms of high storage density and efficiency for fast computational operations [1]. Their reliable experimental manipulation is desired in order to integrate them into multichannel bit sequencers [2], transistors [3] or logic gates [4].

We have recently demonstrated the gate-voltage induced chirality reversal of skyrmions in Ta/FeCoB/TaO trilayer heterostructures with perpendicular magnetic anisotropy [5] (Figure 1a). This effect is directly related to a sign-change of the interfacial Dzyaloshinskii-Moriya interaction (iDMI) [6] that induces an inversion of the current-induced motion direction of the chiral magnetic textures [7]. The iDMI sign reversal is due to a change in the oxidation state at the top FeCoB/TaOx interface [8], caused by the electric field driven migration of oxygen ions. We stabilize trains of skyrmions on micrometric tracks and control their motion with local gate electrodes (Figure 1b). The gate voltage can tune the energy landscape and skyrmion chirality, thus changing the velocity and direction of skyrmion motion induced by the current injected in the track. Figure 1c shows a train of individual skyrmions imaged by magneto optical Kerr effect (MOKE) microscopy. The electrical tracking of moving skyrmions using extraordinary Hall effect is an interesting alternative to optical measurement and enhances the temporal and spatial resolution. These observations thus allow envisioning an optimization of the material parameters producing nanometric skyrmions that will be individually manipulated in gate controlled devices.

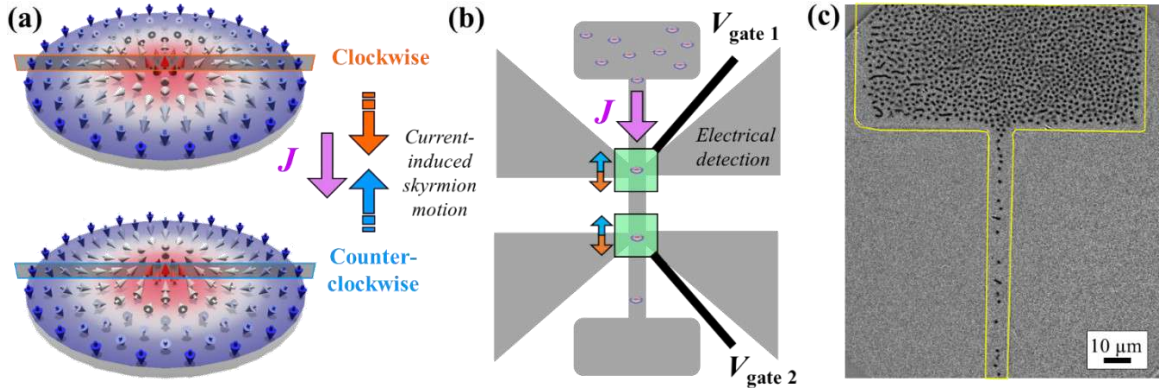



Figure 1a) Depending on their chirality, clockwise and counter-clockwise skyrmions move in opposite directions with respect to the current direction ***J***. b) Schematic Hall bar structure with a pair of gate voltage contacts for chirality control, and lateral electrodes for electrical detection. c) Skyrmions in a track (edges are in yellow), imaged by Magneto optical Kerr effect (MOKE) microscopy.

## Acknowledgements

This work has been supported by ANR-19-CE24-0019 project ADMIS.

## References

[1] A. Fert, N. Reyren and V. Cros, *Nature Review Materials* **46**, 6 (2017)
[2] R. Menezes et al., *Phys. Rev. B* **99**, 104409 (2019)
[3] X. Zhang et al., *Sci. Rep*. **5**, 11369 (2015)
[4] D. Yu et al., *Nat. Sci. Review* **9**, nwac021 (2022)
[5] C. E. Fillion et. al., *Nat. Commun*. **13**, 5257 (2022)
[6] I. E. Dzyaloshinskii, *J. Exptl. Theoret. Phys.* **46**, 960 (1964); A. Thiaville et al., *EPL* **100**, 57002 (2012)
[7] S. Emori et al., *Nat. Mater.***12**, 611 (2013)
[8] M. Arora et al., *Phys. Rev. B* **101**, 054421 (2020); R. Kumar et al., *Phys. Rev. Appl*. **19**, 024064 (2023)

# Skyrmion Formation Mechanism and Density Control in Ferromagnetic / Heavy Metal Multilayers

*Reshma Peremadathil Pradeep[1]*, Andrada-Oana Mandru[1], Hans Josef Hug[1,2]*

[1] *Empa, Swiss Federal Laboratories for Materials Science and Technology, CH-8600 Dübendorf, Switzerland*

[2] *Department of Physics, University of Basel, CH-4056 Basel, Switzerland*

*reshma.peremadathil-pradeep@empa.ch

The interplay between exchange, anisotropy, interfacial Dzyaloshinskii-Moriya interaction (DMI), magnetostatic, and Zeeman energies control skyrmion formation in thin films made of ferromagnetic and heavy metal (with strong spin orbit coupling) multilayers. Such interactions can be tuned by the thickness of the ferromagnetic layer in the system or by the substrate choice. This affects the skyrmion density and stability, which are governed by the critical material parameter $\kappa = \frac{\pi D}{4\sqrt{AK_{eff}}}$, where $D$ is the interfacial DMI constant, $A$ is the exchange stiffness and $K_{eff}$ is the effective magnetic anisotropy. Skyrmions are thermodynamically stable and can exist in large numbers for $\kappa > 1$[1]. In this work, we control the skyrmion density and formation mechanism by varying the thickness of the ferromagnetic Co layer in $[Ir(1nm)/Co(t)/Pt(1nm)]_n$ multilayers and by depositing them on different seed layers [Pt(10 nm) vs. Ta(3 nm)/Pt(10 nm)]. The samples were prepared using DC magnetron sputtering at room temperature. Magnetic system parameters, such as $K_{eff}$, saturation magnetization $M_S$, anisotropy field $H_a$, and magnetic hysteresis were obtained from vibrating sample magnetometry measurements. The domains and skyrmions were imaged in perpendicular fields with a highly sensitive magnetic force microscope operated in vacuum. Without an applied field, a stripe domain pattern, aligned along the direction of the oscillatory in-plane field used for demagnetization, is observed for all samples grown on Ta/Pt seed layers. In contrast, domains with no preferred orientation are observed for samples deposited on Pt only. For the case of samples deposited on Ta/Pt, by increasing the thickness of the Co layer, the domains (at zero field) become narrower, and a higher density of skyrmions (in an applied field) is observed. This is compatible with an increase of κ arising from an effective anisotropy approaching zero at a critical Co thickness. Furthermore, two different skyrmion formation mechanisms (shrinking and fission, see Figure) are observed [2]: for small κ, the stripe domains shorten and eventually decay into skyrmions; for larger κ, the stripe domains break up into multiple skyrmions.

## References

[1] Soumyanarayanan, A. et al. (2017). *Nat Mater.* **16** (2017), 898-904.

[2] Chen X. et al. *Adv. Sci.* **9** (2022), 2103978

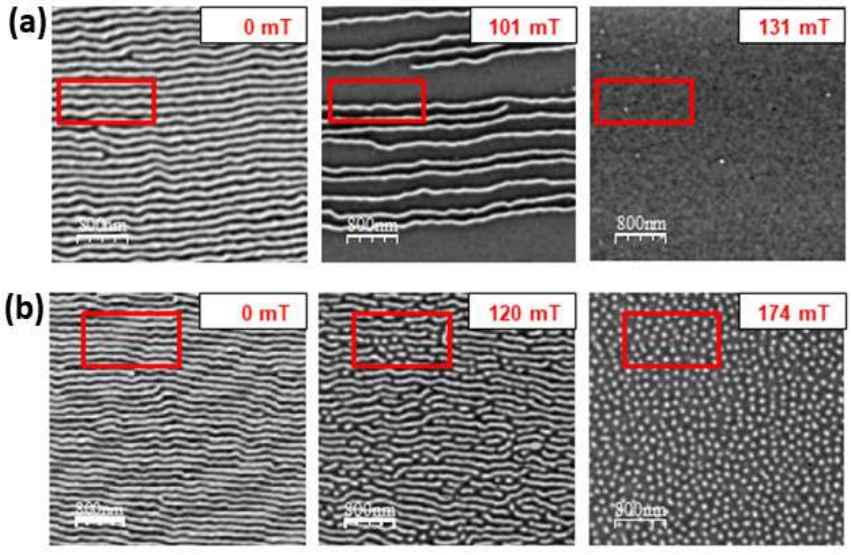



Figure: Skyrmion formation mechanism by, (a) shrinking for a system with $\kappa < 1$, (b) fission for a system with $\kappa > 1$.

# Œrsted-Field Driven Domain-Wall Motion in Core-Shell Nanotubes

*M.C. Scheuerlein[1], R. Crisp[2], L. Ngo[2], C. Thirion[3], L. Gomez Cruz[4], A. Berja[5], L. Perez[4], E. Pereiro[6], S. Finizio[7], W. Ensinger[1], J. Bachmann[2], O. Fruchart[8]*, A. Masseboeuf[8]*

[1] *Technical University of Darmstadt, Darmstadt, Germany*
[2] *Friedrich-Alexander Universität Erlangen-Nürnberg, Erlangen, Germany*
[3] *Univ. Grenoble Alpes, CNRS, Grenoble INP, Instiut Néel, Grenoble, France*
[4] *IMDEA Nanociencia, Madrid, Spain*
[5] *Instituto de Cerámica y Vidrio – CSIC, Madrid, Spain*
[6] *ALBA Synchrotron Light Source, Barcelona, Spain*
[7] *Paul Scherrer Institut, Villigen, Switzerland*
[8] *Univ. Grenoble Alpes, CNRS, CEA, Grenoble INP;SPINTEC, Grenoble, France*
*olivier.fruchart@cea.fr

Magnetic nanowires have been investigated for decades as a prototypical one-dimensional situation for the micromagnetics of domain-wall motion [1]. More recently magnetic nanotubes have been considered as well, with the inner versus outer diameter as an extra degree of freedom. Nanotubes also offer more elaborate situations such as core-shell structures. These broaden the range of available spintronic effects, which require interfaces between different materials. Here we consider nanotubes of diameter ranging from 250 to 580 nm and thickness a few tens of nm, electroless-plated in ion-track-etched polycarbonate membranes. Here we use CoNiB, a material favoring azimuthal magnetization [2]. An inner metallic shell or a full core of Cu is then added, with electroless or electroplating, respectively. The membranes are dissolved, tubes are dispersed on a highly-resistive Si wafer and single objects are contacted electrically by laser lithography. Magnetic imaging is performed under static conditions using XMCD combined with (S)TXM.

We evidence that nanosecond pulses of current set in motion the domain walls, separating domains with opposite azimuthal circulation. The direction of motion is opposite between consecutive walls, consistent with the Œrsted field as a driving force. We report domain-wall speed above 100m/s with current density around $10^{11}$ A/m , translating in a magnetic field around 10mT. This proves the concept of using nanotubes as a platform for the application of sub-ns pulses of magnetic field with strength potentially 100mT or more, which is out of reach of flat stripline geometries, and explore new physics of domain-wall motion under very strong stimulus.

**Acknowledgements**

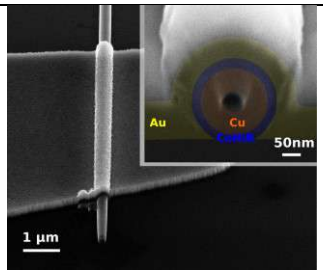
ANR-DFG project C3DS (ANR 18-CE92-0045, DFG 406700532), French RENATECH network, MISTRAL proposal 2022025738 at the ALBA Light Source.

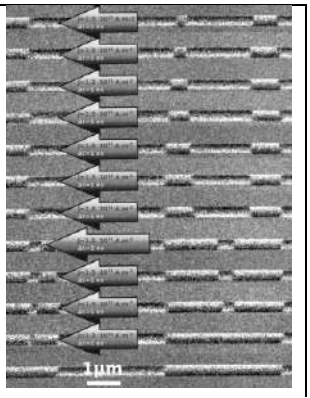
**References**

[1] M. Stano, O. Fruchart, Magnetic nanowires and nanotubes, in Handbook of magnetic materials **27**, E. Brück Ed., Elsevier (2018)

[2] M. Stano, S. Schaefer, A. Wartelle, M. Rioult, R. Belkhou, A. Sala, T. O. Mentes, A. Locatelli, L. Cagnon, B. Trapp, S. Bochmann, S. Martin, E. Gautier, J.C. Toussaint, W. Ensinger, O. Fruchart, SciPost Phys. **5** (2018) 038.

Left: NiCo tubes with 250nm diameter, filled with Cu and contacted. The inset is a cross section (using FIB) revealing the detail of the metal contact on the tube.




XMCD contrast on a contacted tube, following various ns pulses of current. The displacement versus pulse duration is used to compute the domain-wall speed.

# Developing Metallic Multilayers Hosting Different Skyrmion Types Toward Local Control via Electric Fields

*Loghman Jamilpanah [1], Artur Braun [2], Hans Josef Hug [1], Andrada-Oana Mandru[1]**

[1] *Empa, Swiss Federal Laboratories for Materials Science and Technology, Dübendorf CH-8600, Switzerland*

[2] *Department of Physics, University of Basel, Basel CH-4056, Switzerland*

*andrada-oana.mandru@empa.ch

Magnetic skyrmions are particle-like objects with a whirling structure in an otherwise uniform magnetic medium. Due to their small size and energy efficient movement, skyrmions have been proposed as information carriers for future racetrack memory applications [1]. However, their implementation in such devices requires overcoming several issues, one of which concerns maintaining a stable inter-skyrmion distance. Using two distinct solitons for encoding the information has been suggested to overcome this issue [2], as it is advantageous compared to the 'classical' case of just one skyrmion, which can cause large error rates due to the fact that it is very problematic to maintain the skyrmion distribution along the track. Two challenges that arise when approaching this suggested solution involve: 1) having the coexistence of two solitons in a single sample at room temperature and 2) the local control over the soliton types. We addressed the first challenge by demonstrating the coexistence of two distinct skyrmion types in a ferromagnetic/ferrimagnetic/ferromagnetic (FM/FI/FM) trilayer at room temperature [3]. For the second challenge, although tuning the FM layer thickness and therefore its magnetic anisotropy results in a fine tunning of the skyrmion type in these trilayers [4], the rather complex structure hinders a simple local control over the skyrmion type. Here, we demonstrate that two skyrmion types can coexist in a simplified system, i.e. a FM/FI bilayer [Fig. a)]. Our magnetic force microscopy (MFM) results [Fig. b)] show that the skyrmion type can be tuned from *incomplete* (present in the FM layer only) to *tubular* (existing in both the FM and FI layers) by decreasing the effective magnetic anisotropy of the FI layer, respectively. These results unlock the opportunity of local control of the skyrmion type via, for example, voltage-controlled magnetic anisotropy of a single FI layer within the same sample, while also opening new directions for 3D devices.

## Acknowledgements

The authors acknowledge funding support from Empa Internal Research Call.

## References

[1] A. Fert, V. Cros, and J. Sampaio, Nat. Nanotechnol. 2013 83 8, 152 (2013).

[2] F. Zheng, F.N. Rybakov, A.B. Borisov, D. Song, S. Wang, Z.A. Li, H. Du, N.S. Kiselev, J. Caron, A. Kovács, M. Tian, Y. Zhang, S. Blügel, and R.E. Dunin-Borkowski, Nat. Nanotechnol. 2018 13613, 451 (2018).

[3] A.-O. Mandru, O. Yildirim, R. Tomasello, P. Heistracher, M. Penedo, A. Giordano, D. Suess, G. Finocchio, and H.J. Hug, Nat. Commun. 11, 1 (2020).

[4] O. Yildirim, R. Tomasello, Y. Feng, G. Carlotti, S. Tacchi, P.M. Vaghefi, A. Giordano, T. Dutta, G. Finocchio, H.J. Hug, and A.O. Mandru, ACS Appl. Mater. Interfaces 14, 34010 (2022).

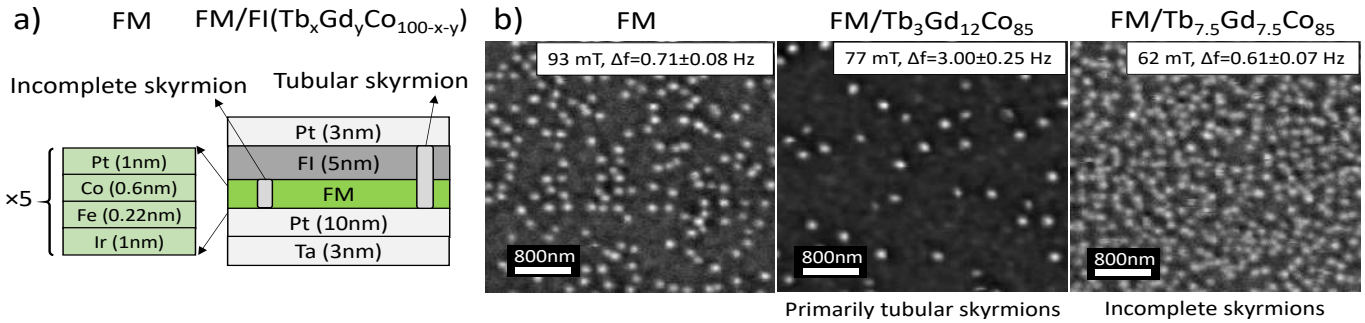



Figure: a) Sample schematics of the FM/FI bilayer (FM is an [Ir/Fe/Co/Pt] skyrmion generator layer; FI is a TbGdCo alloy). b) MFM images of the skyrmion generator layer only and of the FM/FI bilayers for a fixed FM layer and different-anisotropy FIs, showing the transition from tubular (high Δf contrast of ≈ 3Hz) to incomplete (low Δf contrast of ≈ 0.6Hz) skyrmions.

# Skyrmion Fluid and Bimeron Glass Protected by a Chiral Spin Liquid on a Kagome Lattice

*H. Diego Rosales[1,2,3] Flavia A. Gómez Albarracín[1,2,3] Pierre Pujol[4], Ludovic D. C. Jaubert[5*]*

[1] *IFLSB, CONICET, FCE, Universidad Nacional de La Plata, 1900 La Plata, Argentina*
[2] *Departamento de Física, FCE, Universidad Nacional de La Plata, 1900 La Plata, Argentina*
[3] *Departamento de Ciencias Básicas, FI, Universidad Nacional de La Plata, 1900 La Plata, Argentina*
[4] *Laboratoire de Physique Théorique, CNRS, Université de Toulouse, UPS, 31062 Toulouse, France*
[5] *CNRS, LOMA, University of Bordeaux, 33400 Talence, France*
*ludovic.jaubert@cnrs.fr

Skyrmions are of interest both from a fundamental and technological point of view, due to their potential to act as information carriers. But one challenge concerns their manipulation, especially at high temperature where thermal fluctuations eventually disintegrate them.

In this presentation, we will study the competition between skyrmions and a chiral spin liquid, using the latter as an entropic buffer to impose a quasi-vacuum of skyrmions [1]. As a result, the temperature becomes a knob to tune the skyrmion density from a dense liquid to a diluted gas, protecting the integrity of each skyrmion from paramagnetic disintegration until they vanish. With this additional knob in hand, we find at high field a topological spin glass made of zero- and one-dimensional topological defects (respectively skyrmions and elongated bimerons).

## Acknowledgements

We acknowledge financial support from CNRS IRP (COQSYS), ANR (Grant No. ANR-18-CE30-0011-01), CONICET (PIP 2021-11220200101480CO), SECyT-UNLP (I+D X893), and PICT 2018-02968.

## References

1. H. Diego Rosales, Flavia A. Gómez Albarracín, Pierre Pujol and Ludovic D. C. Jaubert, *Phys. Rev. Lett.* **130**, 106703 (2023)

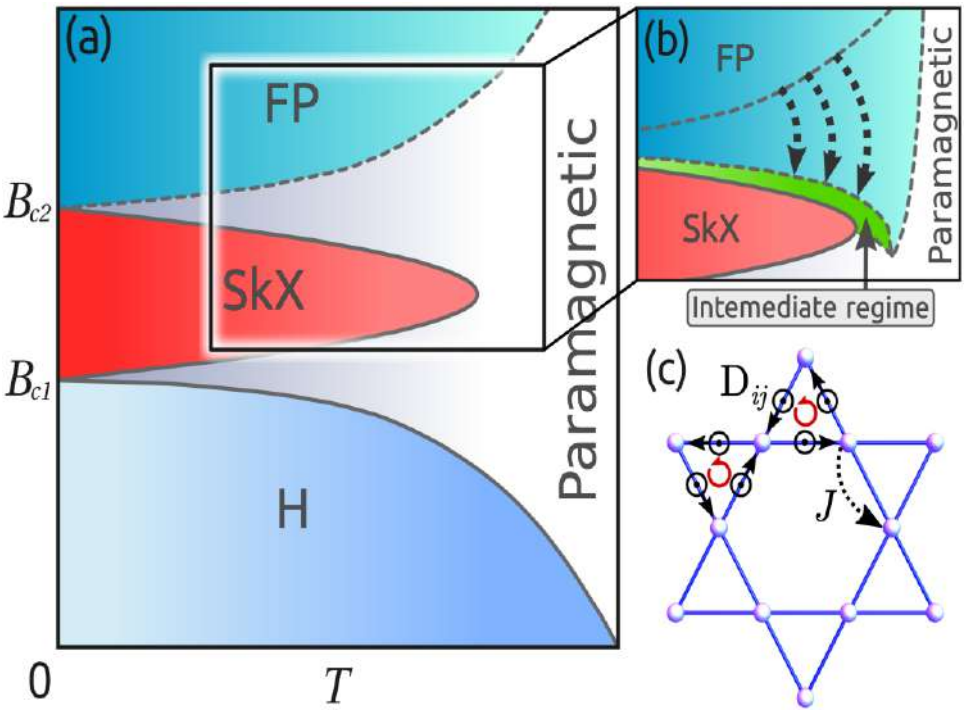



Figure: **(a)** Schematic representation of the traditional phase diagram where the magnetic field destroys the skyrmion solid (SkX) in favor of a FP regime with no skyrmions. **(b)** Illustration of our idea where the FP phase circles around the SkX phase, thus creating an intermediate regime (in green) where skyrmion density can be tuned gradually. **(c)** Kagome lattice with nearest-neighbor Heisenberg exchange and Dzyaloshinskii- Moriya interactions.

# Reversible and non-volatile control of perpendicular magnetic anisotropy in Pt/Co/MOx trilayers

*C. Balan[1], A. Fassatoui[1], J. Peña Garcia[1], H. Béa[2], J. Vogel[1], L. Ranno[1], S. Pizzini[1*]*

*[1] Univ. Grenoble Alpes, CNRS, Institut Néel, 25 Rue des Martyrs, Grenoble, France*

*[2]Univ. Grenoble Alpes, CNRS, CEA, SPINTEC, 17 Av. des Martyrs, Grenoble, France*

*stefania.pizzini@neel.cnrs.fr

The voltage control of magnetic anisotropy (VCMA) is an active field of research, as it is a promising route towards the realization of low power spintronic devices. Among a few proposed VCMA mechanisms, voltage-induced ion migration (i.e., magneto-ionics) has demonstrated to induce a large modulation of magnetic anisotropy [1]. We have studied the voltage control of the perpendicular magnetic anisotropy (PMA) in a series of Pt/Co/MOx (M=Al and Tb) trilayers [2-3]. A 10-nm-thick $ZrO_2$ layer deposited on top of the Pt/Co/MOx stacks acts as a dielectric and oxygen ion conductor layer. The application of a gate voltage allows manipulating the perpendicular magnetic anisotropy with large efficiency ($\beta > 1200$ fJ/Vm at room temperature) in a non-volatile and reversible way. The voltage-driven migration of oxygen ions towards/away from the Co/MOx interface is at the origin of the PMA variation.

By tuning the PMA, the magnetization reversal mechanism can be tuned and a variety of magnetic configurations (isolated domains, labyrinthine domains, skyrmions) can be stabilized within the Co layers. Figure 1(a) shows that in Pt/Co/TbOx, the easy magnetization axis can be switched reversibly from in-plane to out-of-plane (OOP) and back, by applying respectively a negative or a positive gate voltage. The voltage-driven switching time ($t_{sw}$) between different magnetic states, related to the oxygen ion drift velocity, depends exponentially on the applied voltage. For Pt/Co/TbOx we show that $t_{sw}$ can be varied by over 5 orders of magnitude, from several hundreds of seconds to a few milliseconds at room temperature by tuning the gate voltage amplitude from 0.5V to 6V (Figure 1(b) [3].

In a second set of experiments, we studied the effect of the gate voltage on the field-driven dynamics of chiral Néel walls in ferrimagnetic Pt/Co/Tb trilayers. The application of a negative gate voltage leads to a small decrease of the coercive field, together with a huge variation of the domain wall (DW) velocity and of the Dzyaloshinski-Moriya field (DMI field) stabilizing the chiral Néel walls. By comparing the behaviour of the domain velocities before and after gating with those of reference Pt/Co/Tb samples with different ratios between the Co and Tb thicknesses, we were able to attribute these variations to the partial oxidation of the Tb layer, and the subsequent increase of the sample magnetization and the decrease of the PMA. The velocity, which in the pristine state is only around 10 m/s for large fields, can reach 250 m/s after gating (Figure 1(c) [4].

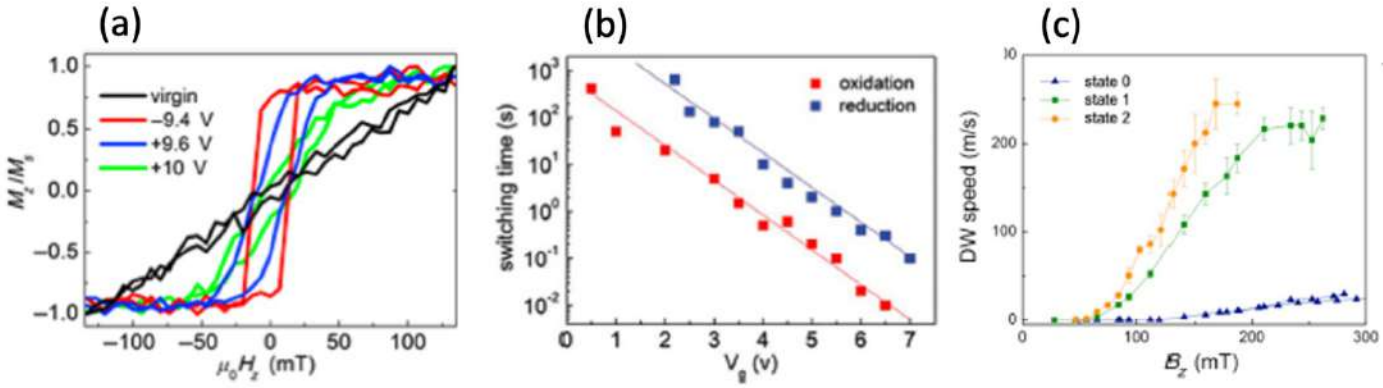



*FIGURE 1: (a) Large variation of the coercive field (proportional to the PMA) in Pt/Co/TbOx under a positive voltage; (b) Time needed to switch the magnetization from in-plane over-oxidized Co to out-of-plane magnetization with maximum PMA and vice versa as a function of the gate voltage. (c) Domain wall velocity vs. Bz field in Pt/Co/Tb trilayers, before the application of the gate voltage (state 0) and after V=-8V (state 1) and V=-10V (state 2).*

**References**

[1] C. Bi et al. Phys. Rev. Lett. 113, 267202 (2014).

[2] A. Fassatoui et al. , Phys. Rev. Appl. 14, 064041 (2020).

[3] A. Fassatoui, et al., Small. 17, 2102427 (2021).

[4] C. Balan et al. Phys. Rev. Appl. 18, 034065 (2022).

# Intrinsic Antiferromagnetic Multi-Meronic Néel Spin-Textures in Thin Films

*A. Aldarawsheh[1,2*], M.Sallermann[1,3,4], M. Abusaa[5], and S. Lounis[1,2]*

*[1]Peter Grünberg Institute & Institute for Advanced Simulation, Forschungszentrum Jülich and JARA, D-52425 Jülich, Germany*

*[2]Faculty of Physics, University of Duisburg-Essen and CENIDE, 47053 Duisburg, Germany*

*[3]RWTH Aachen University, 52056 Aachen, Germany*

*[4]Science Institute and Faculty of Physical Sciences, University of Iceland, VR-III, 107 Reykjavík, Iceland*

*[5]Arab American University, Jenin, Palestine*

*a.aldarawsheh@fz-juelich.de

The realization of topological antiferromagnetic (AFM) solitons in real materials is a major goal towards their implementation in information technology. Unlike their ferromagnetic counterparts, they are anticipated to be immune to dipolar interactions and the skyrmion Hall effect [1,2]. By utilizing density functional theory and atomistic spin dynamics, we have predicted the emergence of intricate Néel AFM vortex-antivortex structures in transition metallic thin films interfaced with Ir and Pd layers on a triangular lattice. These topological structures are intrinsic, that is they form within a single AFM material, but they differ from the previously predicted intrinsic AFM skyrmions [3]. They possess various topological charges and can combine in hexameronic, dodecameronic patterns, or even higher excited states, which may exhibit increased stability concerning an external magnetic field contingent on the electronic properties of the interfaces.

**Acknowledgements**

Workfunded bythe PGSB (BMBF-01DH16027) and DFG (SPP 2137; LO 1659/8-1).

**References**

[1] Barker, J. & Tretiakov, O. A., Physical review letters **116**, 147203 (2016).

[2] Zhang, X., Zhou, Y. & Ezawa, M., Scientific reports **6**, 1–8 (2016).

[3] A. Aldarawsheh, I. L. Fernandes, S. Brinker, M. Sallermann, M. Abusaa, S. Blügel, S. Lounis, Nat. Commun. **13**(1) (2022) 7369.

# Complementary microscopy and scattering experiments on the magnetic textures in the antiskyrmion compound $Mn_{1.4}PtSn$

M. Winter[1,2,3,4], B. Rellinghaus[2*] , M. Rahn[4], A. Sukhanov[4], D. Wolf[3], S. Schneider[2], M. Valvidares[5], C. Shekar[1], P. Vir[1], B. Achinuq[6], H. Popescu[7], G. Van der Laan[8], T. Hesjedal[6], and C. Felser[1]

[1] MPI CPfS, Dresden, Germany
[2]Dresden Center for Nanoanalysis (DCN), TU Dresden, Germany
[3]IFW Dresden, Germany
[4]IFMP, TU Dresden, Germany
[5]ALBA Synchrotron, Barcelona, Spain
[6]Clarendon Laboratory, University of Oxford, UK
[7]Synchrotron SOLEIL, Saint-Aubin, France
[8]Diamond Light Source, United Kingdom.
*bernd.rellinghaus@tu-dresden.de

Non trivial magnetic textures such as skyrmions bear the potential for applications in data storage devices and have also attracted significant attention in basic research. Recently, antiskyrmions (aSks) were observed in the tetragonal Heusler compound $Mn_{1.4}PtSn$ by Lorentz transmission electron microscopy (LTEM) [1]. The combination of anisotropic Dzyaloshinskii-Moriya and strong dipolar interactions leads to the stabilization of various magnetic structures such as spin helices, non-topological (NT) bubbles and elliptical skyrmions [2] This results in a highly complex magnetic phase diagram that depends not only on temperature, magnetic field strength and direction, but also the sample shape [3].

To better understand the interplay between the various magnetic phases we have conducted complementary experiments of resonant elastic x-ray scattering (REXS) and LTEM on *identical* lamellae of high quality single crystals of $Mn_{1.4}PtSn$ grown by the self-flux method. The FIB-cut sub 100 nm lamellae were placed on specially designed Mo disks that also fit into commercial TEM holders, and custom designed sample holders have been developed to ensure compatibility with the REXS setup. The latter was equipped with an octupole vector magnet offering 360° of freedom in magnetic field direction and fields up to 600 mT thereby giving full access to the complex phase diagram of $Mn_{1.4}PtSn$.

Our complementary approach allows us to unambiguously connect the real space magnetic textures in $Mn_{1.4}PtSn$ and the transitions between them with their corresponding scattering patterns obtained by REXS. The experiments reveal a subtle interplay between the different magnetic phases and uncover the details of the formation of antiskyrmions from the helical ground state via (intermediate) NT bubbles. Depending on the direction of the applied field, the helical ground state is found to develop into either a chiral soliton lattice (CSL) or a conical phase. Analyzing the Fourier components of the REXS patterns also provides for a detailed understanding of how the field induced phase transitions contribute to the increase or decrease of the out-of-plane magnetization as monitored by in-situ Hall effect measurements.

**Acknowledgements**
This study was supported by the International Max Planck Research School for Chemistry and Physics of Quantum Materials (IMPRS-CPQM) and by the DFG through SPP 2137 (project ID 403503416).

**References**

[1] A.K. Nayak *et al.*, *Nature.* **548**, 561 (2017)
[2] L. Peng *et al.*, *Nature Nanotechnol.* **15**, 181-186 (2020)
[3] M. Winter et al., *Commun. Mater.* **3**, 102 (2022).

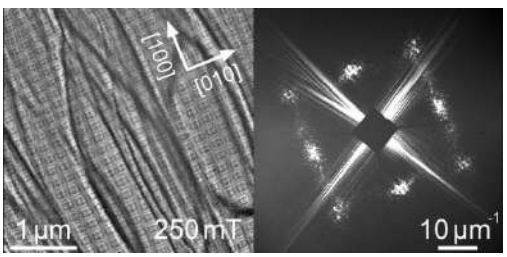



Figure: Comparing the LTEM image (left) and REXS pattern (right) of an antiskyrmion lattice obtained at a field of 250 mT from an identical lamella of $Mn_{1.4}PtSn$ allows for a direct correlation of the scattering patterns with the underlying magnetic texture. REXS data were acquired at the Diamond Light Source, UK.

# Reversal of Skyrmion Deflection in Ferrimagnets

*Léo Berges[1], Raphael Weil[1], Alexandra Mougin[1], and João Sampaio[1]**

[1] *Laboratoire de Physique des Solides, CNRS, Université Paris-Saclay, Orsay, France*

** joao.sampaio@universite-paris-saclay.fr*

Magnetic skyrmions are point-like magnetic textures with a well-defined chirality. They can be stabilized in thin-film nanostructures and efficiently moved by current-induced spin orbit torques, which has led to the proposal of many applications [1]. However, their motion is affected by a sideways gyrotropic deflection, also known as the skyrmion Hall effect, which is detrimental to some applications [2] as it reduces their forward velocity and can lead to their annihilation at the edges of the host magnetic structure. This deflection is inherent to their non-trivial topology and expected to be proportional to the material angular momentum density ($L_S$).

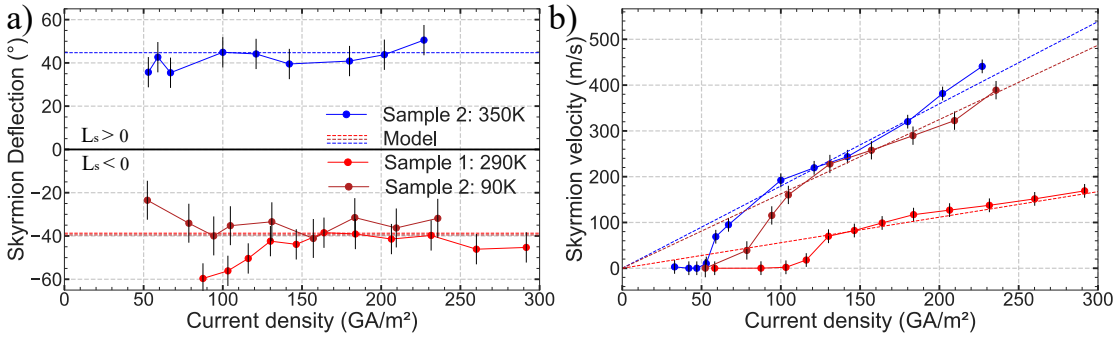



Figure: Skyrmion deflection angle (a) and velocity (b) versus applied current density for three cases: RE-dominated case in sample 1 (at 290 K), and RE- and TM-dominated cases in sample 2 (at 90 K and 350 K, respectively), showing the reversal of skyrmion deflection with the sign of angular momentum of the film.

We studied the dynamics of skyrmions in thin films of CoGd, a rare earth/transition metal (RETM) ferrimagnetic alloy, with a Pt buffer layer. In RETMs, the effective $L_S$ can be changed and reversed with temperature or alloy composition. If the RE moment is larger than the TM one, the film is 'RE-dominated' (and the effective $L_S$ is negative), otherwise, it is 'TM-dominated' (and $L_S > 0$).

We studied the skyrmion dynamics in two samples of different GdCo composition (30% and 32% Gd). In sample 1, skyrmions were stable in the RE-dominated temperature range, while in sample 2 skyrmions were stable in two temperature ranges, one RE- and the other TM-dominated. Skyrmions were driven with current pulses and their velocity and deflection were tracked using magneto-optical Kerr microscopy. Above a depinning threshold current density, the velocity increases linearly with current, up to 450 m/s (fig. b). This was reproduced quantitatively by a model with fully-determined parameters based on the Thiele equation (dashed lines in the figure) [3].

To study the effect of $L_S$ on the gyrotropic deflection, we compared the skyrmion deflection in the TM- and in the RE-dominated cases. We observed that indeed the gyrotropic deflection is negative in both RE-dominated cases and positive in the TM-dominated one. The sign, as well as the deflection angle, matched quantitatively with the model. The reversal of the gyrotropic deflection with the sign of $L_S$, by temperature and by alloy composition, demonstrates that the deflection can be tuned and eliminated in RETMs. This, along the observed low pinning and high mobility, makes GdCo a promising material for the study and applications of skyrmions.

[1] C. H. Marrows, K. Zeissler, *Appl. Phys. Lett.* **119** (2021) 250502
[2] J. Sampaio *et al.*, *Nat. Nanotech.* **8** (2013) 839
[3] L. Berges *et al.*, Phys. Rev. B **106** (2022) 144408

# Anatomy of the Dynamics of the Nucleation of Skyrmions in Nanodots via Voltage-Controlled Magnetic Anisotropy

*Pablo Olleros-Rodríguez[1]*, Adrián Gudín[1], Julio Camarero[1], Oksana Chubykalo-Fesenko[2], Paolo Perna[1]*

[1] *IMDEA Nanoscience Institute, Campus de Cantoblanco, Madrid, Spain*

[2] *Materials Science Institute of Madrid (ICMM-CSIC), Campus de Cantoblanco, Madrid, Spain*

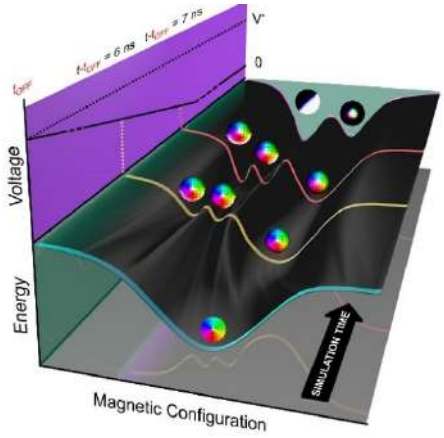
*pablo.olleros@imdea.org

Magnetic skyrmions are nanometer-size chiral spin textures that hold promise for their implementation in novel spin-orbitronic devices and for their use in novel computation paradigms sach as in neuromorphic or reservoir computing schemes.[1] The specific magnetic configuration shown by magnetic skyrmions confers to them the so-called topological protection which enlarges the energetic barriers of these stable or metastable states. The latter, confers to magnetic skyrmions a large robustness but also it hinders their nucleation as the aforementioned energetic barriers must be overcome. This can be achieved by employing external excitations as electric currents or magnetic fields or by modifying the energetic landscape making it possible to access skyrmionic states. In the recent years the exploitation of the magnetoelectric effect in multi-layered heterostructures with spinorbitronic capabilities has been boosted thanks to the evolution of materials and surface science and nanotechnology.[2] This allows for the modification of magnetic properties of the sample in a localized and ultrafast manner with a reduced power consumption and with an ease implementation in magnetoelectric devices.

In this work we analyse the nucleation of magnetic skyrmions in patterned nanodots by exploiting the voltage-controlled magnetic anisotropy (VCMA). The work has been carried out by considering a micromagnetic model in which the VCMA is introduced following theoretical and experimental reports. The results show that the probability of nucleating magnetic skyrmions is increased when following a quasi-adiabatic path in which the system is able to show always the most stable configuration during the recovery of the magnetic anisotropy. A detailed description of the nucleation process will help in the design of a reliable and universal protocol for the nucleation of magnetic skyrmions in magnetic trilayers.

## Acknowledgements

The authors acknowledge fundings received from the Regional Government of Madrid, Ministry of Science and Innovation MCIN/AEI and the European Union through projects P2018/NMT-4321 (NANOMAGCOST-CM), PID2021-122980OB-C52 (ECLIPSE-ECoSOx) and PCI2019-111867-2 (SOgraphMEM). IMDEA Nanoscience Institute is supported by the "Severo Ochoa" Programme for Centres of Excellence in R&D, (MINECO grant CEX2020-001039-S). We acknowledge NVIDIA corporation for the donation of the GPU Quadro P6000 used in the micromagnetic simulations.




## References

[1] A.Fert, N. Reyren, V. Cros, *Nat. Rev. Mat.* **2** (2017) 17031.

[2] F. Matsukura, Y. Tokura, H. Ohno, *Nat. Nanotechnol.* **10** (2015) 209.

Figure: Energetic landscape during the voltage induced nucleation of magnetic skyrmions in nanodots

# Topological Hall effect caused by magnetic skyrmions in Pd/Fe/Ir(111) film

*Adamantia Kosma[1]*, Philipp Rüßmann[2,1], Stefan Blügel[1], Phivos Mavropoulos[3]*

[1] *Peter Grünberg Institut and Institute for Advanced Simulation, Forschungszentrum Jülich and JARA,52425 Jülich, Germany*

[2] *Institute for Theoretical Physics and Astrophysics, University of Würzburg, 97074 Würzburg, Germany*

[3] *Department of Physics, National and Kapodistrian University of Athens, Panepistimioupolis 15784, Athens, Greece*

*a.kosma@fz-juelich.de

The topological Hall effect (THE) provides a way to electrically detect magnetic skyrmions. This work includes an ab-initio computational study of the THE arising from stable magnetic skyrmions [1] formed in the Fe atomic layer of the ultrathin film Pd/Fe/Ir(111) [2]. Our simulations are performed employing the non-collinear spin-density-functional theory within the full-potential relativistic Korringa-Kohn-Rostoker (KKR) Green function method combined with the semiclassical Boltzmann transport equation [3,4]. We analyze the resistivity and the Hall angle of the system, and we discuss the dependence of the THE on disorder, modelled by an additional electron scattering term. The effect of the skyrmion size on the Hall angle is also examined.

**Acknowledgements**

AK and PR acknowledge funding by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) under Germany's Excellence Strategy -- Cluster of Excellence Matter and Light for Quantum Computing (ML4Q) EXC 2004/1 – 390534769.

**References**

*[1]* D. Maccariello, W. Legrand, N. Reyren, K. Garcia, K. Bouzehouane, S. Collin, V. Cros, A. Fert, *Nat. Nanotechnology* **13** (2018).

*[2]* N. Romming, C. Hanneken, M. Menzel, J. E. Bickel, B. Wolter, K. Bergmann, A. Kubetzka, R. Wiesendanger, *Science* **341** (2013) 6146.

*[3]* https://jukkr.fz-juelich.de/.

*[4]* A. Kosma, P. Rüßmann, S. Blügel, P. Mavropoulos, *Phys. Rev. B* **102** (2020) 144424.

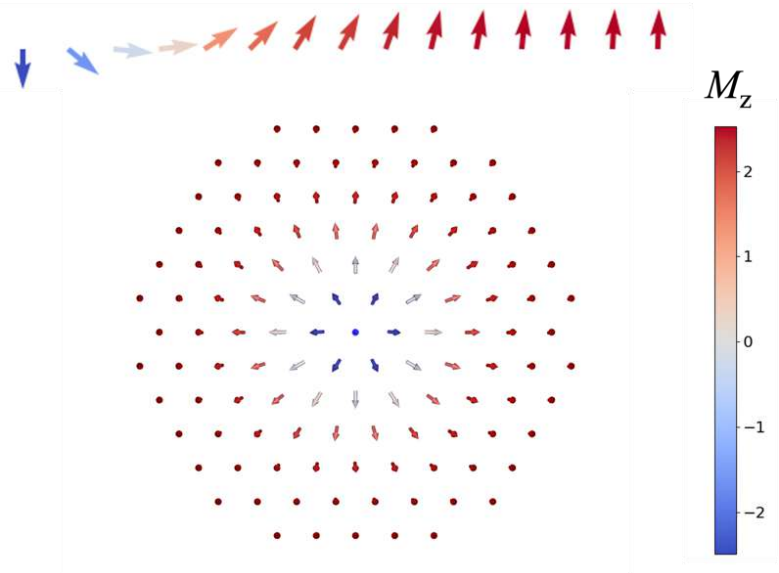



Figure: Magnetic skyrmion formed in ultrathin film Pd/Fe/Ir(111).

# Homotopies of Diverse Topological Magnetic Solitons

*Vladyslav M. Kuchkin[1]*, Nikolai S. Kiselev[2] , Filipp N. Rybakov[3,4] , Pavel F. Bessarab[1,5]*

[1] *University of Iceland, 107 Reykjavík, Iceland*

[2] *Peter Grunberg Institute and Institute for Advanced Simulation, Forschungszentrum Julich and JARA, 52425 Julich, Germany*

[3] *Uppsala University, SE-75120 Uppsala, Sweden*

[4] *KTH Royal Institute of Technology, SE-10691 Stockholm, Sweden*

[5] *Department of Physics and Electrical Engineering, Linnaeus University, SE-39231 Kalmar, Sweden*

*vkuchkin@hi.is

Continuous transformations, so-called homotopies, between different magnetic states, e.g., statically stable solitons, represent not only the fundamental interest but might also be relevant for future applications. However, the search for such homotopies can be challenging. In my talk, I demonstrate that continuous magnetic transitions can be efficiently calculated using the well-established geodesic nudged elastic band (GNEB) method [1]. In general, the outcome of the GNEB calculation represents a minimum energy path (MEP), thereby providing information about the energy barrier and rate of transition between the states. We apply this method to the 2D chiral magnetic solitons. By mapping the MEP to the third spatial direction, we found a series of reliable initial guesses for non-trivial 3D solitons – hybrid skyrmion tubes[2] (Fig.1). Moreover, we show the GNEB method helps explore the diversity of chiral magnetic solitons, which has not been known so far, so-called tailed skyrmins[3]. Finally, I will discuss the applicability of the GNEB method to hopfions, skyrmion braids, and other 3D magnetic textures.

## Acknowledgements

This work was supported by the Icelandic Research Fund (Grant No. 217750), the University of Iceland Research Fund (Grant No. 15673), and the Swedish Research Council (Grant No. 2020-05110).

## References

[1] P. F. Bessarab, V. M. Uzdin, H. Jónsson, Comput. Phys. Commun. **196** (2015) 335.
[2] V. M. Kuchkin and N. S. Kiselev, APL Materials **10**, (2022) 071102.
[3] V. M. Kuchkin, N. S. Kiselev, P. F. Bessarab, (2023) arXiv:2303.00347.

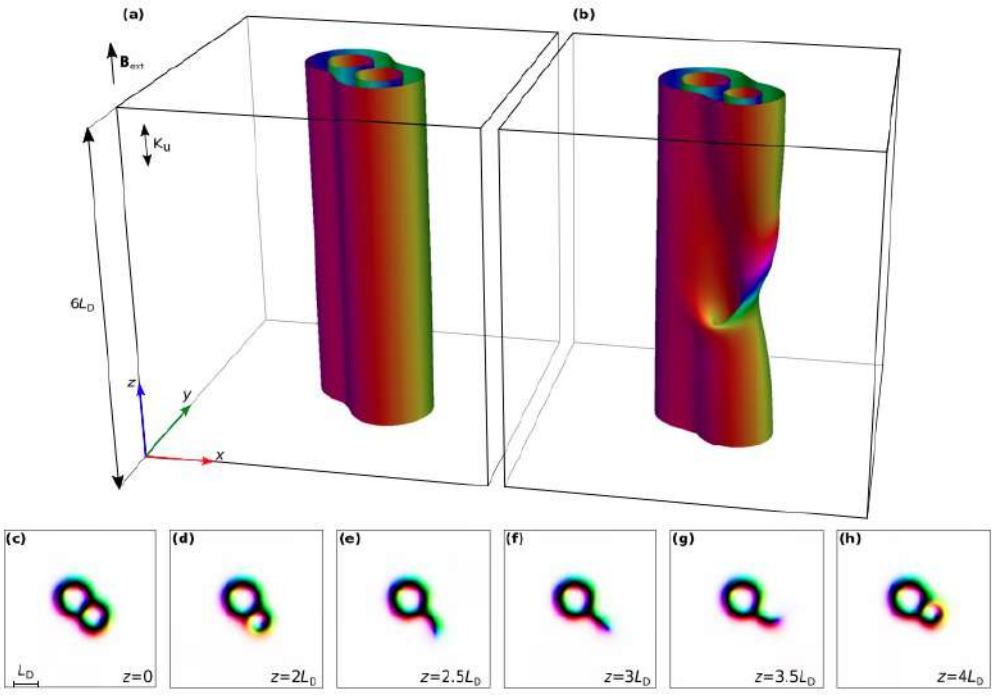



Figure 1: (a) and (b) Isosurfaces $n_z = 0$ for ordinary and hybrid skyrmion tubes. (c)–(h) Cross sections of the hybrid tube.

# Asymmetric Diode-Like Behaviour Controlled by Geometry in Current Induced Spin-Orbit Torque Driven Domain Wall Devices

*E.M. Stetco[1,2]*, T.Petrisor Jr.[1], O.A. Pop[2], I.M.Miron[3] , M.S.Gabor[1]*

[1] *Centre for Superconductivity, Spintronics and Surface Science, Physics and Chemistry Department, Technical University of Cluj-Napoca, Str. Memorandumului, 400114 Cluj-Napoca, Romania*

[2] *Applied Electronics Department, Technical University of Cluj-Napoca, Str. Memorandumului, 400114 Cluj-Napoca, Romania*

[3] *SPINTEC, UMR-8191, CNRS/CEA/Université Grenoble Alpes, Grenoble, 38054 France*

*elena.stetco@ael.utcluj.ro

Spin-orbit torques (SOTs) have earned a wide research interest being a fast, low-power, and effective technique for the manipulation of magnetic texture in spintronic devices (such as spintronic memories, magnetic oscillators, logic devices and so on) [1]. Current-induced SOT arises from the spin Hall and/or Rashba effect, where an in-plane electric current induces a spin accumulation at a heavy-metal (HM)/ferromagnetic (FM) layer interface via the spin-orbit coupling. The spin current then diffuses into the adjacent FM layer and exerts two torques on the magnetization, which can switch the magnetization and can lead to magnetic domain walls (DWs) [2] or skyrmions motion in the FM layer [3].

In this work we present an exhaustive micromagnetic and experimental study of SOT-driven DW motion, investigating the optimal parameters that allow efficient control of the dynamics of the DW by tuning the DMI, the current density, and the relative angle between the current direction and the DW magnetization. First, we performed micromagnetic simulations for a disordered stripe with an up/down DW perpendicular to the *Ox* axis direction and along the *Oy* axis direction. Our 1 μm wide and 1 nm thick Co stripe is discretized in 2 nm × 2 nm × 1 nm cells and is placed on top of a HM layer of Pt. Current pulses with different densities and widths are applied at different angles (45°, 0° and -45°) with respect to the *Ox* axis direction. In principle, the DW velocity depends on the charge current density and the projection of the DW magnetization on the charge current direction $v \propto -J_{SOT} cos(\alpha - \beta)$ [4]. For positive charge currents, when the current is applied at 45° and 0°, we observe a threshold current below which the DW is pinned by the disorder. Above the threshold current, the DW velocity increases with the current. We also observed that for the current applied along the stripe, the DW velocity is symmetric with respect to the change in the current sign, while for the current applied at 45° or -45°, there is a significant asymmetry. These results indicate that the DW propagates in one direction and the reversed motion is blocked. This is the striking feature of our results and is reminiscent of diode-like behaviour.

In order to validate the results of the micromagnetic simulations, we performed pulse current-induced DW motion experiments using polar Magneto-Optical Kerr Effect (pMOKE) microscopy. The samples were patterned using standard UV lithography technique from perpendicular magnetized Pt 4 nm/Co 1 nm/Pt 1.5 nm stacks grown by magnetron sputtering on $Si/SiO_2$ substrates. The lithography process was optimized to reduce the edge roughness, which is an important source of pinning centres for the DWs. The first lithographic step consisted in the patterning of a 10 μm wide Pt/Co/Pt. In a second step, different Co/Pt devices with geometries that allows the application of the current at different angles (45°, 0° and -45°) relative to the DW were defined on top of the 10 μm wide stripe. The pMOKE experiments confirmed the asymmetric diode-like behaviour of the DW motion in our optimized samples.

These types of devices with asymmetric diode-like DW motion have the potential to be used for the development of DW logic devices.

## References

[1] A. Manchon *et al., Rev. Mod. Phys.,* **91**, 035004 (2019).
[2] S.Emori *et al.*, *Nature Mater* **12**, 611–616 (2013).
[3] A.Fert *et al.*, Nature Nanotech 8, 152–156 (2013).
[4] A. Thiaville *et al.*, *EPL Europhys. Lett* **100**, 57002 (2012).

# Co-existing topological spin textures in Rh/Co/Ir(111) identified by saddle point searches

*Hendrik Schrautzer[1,2]*, Hannes Jónsson[1], Stefan Heinze[2], Moritz Goerzen[2] and Pavel F. Bessarab[1,3]*

[1] *Science Institute, University of Iceland, Reykjavik, Iceland*

[2] *Institute of Theoretical Physics and Astrophysics, University of Kiel, Kiel, Germany*

[3] *Department of Physics and Electrical Engineering, Linnaeus University, Kalmar, Sweden*

*hes93@hi.

Multistable magnetic systems hosting distinct co-existing topological spin textures such as skyrmions and skyrmion bags [1] are both fundamentally interesting and technologically important. However, the large number of degrees of freedom makes theoretical investigation of mutlistable magnetic systems challenging. Here, we present a methodology combining global optimization based on recursive traversing between energy minima via saddle points on the energy surface [2], and harmonic transition state theory [3]. The methodology makes it possible to predict previously unknown metastable magnetic states, identify co-existing transition mechanisms between them, and ultimately simulate long time scale finite temperature magnetization dynamics of a multistable magnetic system.

We apply the method to the highly exchange frustrated magnetic system Rh/Co/Ir(111), parametrized using the density functional theory, which is known for hosting skyrmions at zero magnetic field [4], and demonstrate a variety of co-existing topological spin textures and transition paths between them, see Fig. 1. We compute the rates of individual transition between the magnetic states and use them to simulate kinetics of the system at various temperatures. Our results demonstrate usefulness of the saddle point search-based optimization method for the characterization of complex, multistable magnetic systems.

## Acknowledgements

This work was supported by the Icelandic Research Fund (Grants No. 239435 and No. 217750), the University of Iceland Research Fund (Grant No. 15673), and the Swedish Research Council (Grant No. 2020-05110).

## References

[1] V. M. Kuchkin, *et al.*, *Phys. Rev. B* **102** (2020): 144422
[2] M. Plasencia Gutiérrez, *et al., J. Chem. Theory Comput.* **13**.1 (2017): 125-134
[3] P. F. Bessarab, *et al. Phys. Rev. B* **85**.18 (2012): 184409
[4] S. Meyer, *et al. Nat. Commun.* **10** (2019): 3823

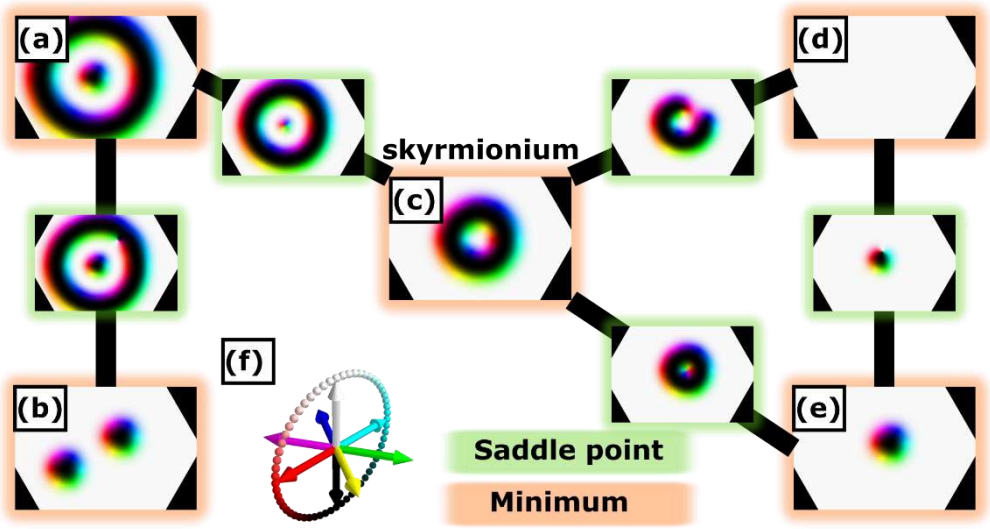



Figure 1: Application of the saddle point search method to the Rh/Co/Ir(111) system. Starting from metastable states associated with local energy minima (red, a,b,c,d,e), the method produces neighbouring energy minima by calculating saddle points on the energy surface (green). (f): Colour coding of the magnetic moment orientation.

# Current induced magnetization switching and Dzyaloshinskii–Moriya interaction in ferromagnetically coupled Co/Pt/Co trilayers

*K.Grochot*[1,2], P.Ogrodnik[3], J.Mojsiejuk[1], P.Mazalski[4], U. Guzowska[4], W.Skowroński[1], T.Stobiecki[1,2]*

[1] *Institute of Electronics, AGH University of Science and Technology, al. Mickiewicza 30, Kraków, Poland,*
[2] *Faculty of Physics and Applied Computer Science, AGH University of Science and Technology, al. Mickiewicza 30, Kraków, Poland,*
[3] *Faculty of Physics, Warsaw University of Technology, Warsaw, Poland.*
[4] *Faculty of Physics, University of Bialystok, Bialystok, Poland*
*grochot@agh.edu.pl

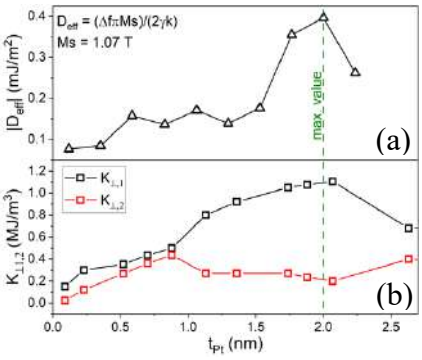



Fig. 1 (a) Effective DMI constant as a function of Pt layer thickness; (b) perpendicular anisotropy constants of the top (K1) and bottom (K2) Co layers, respectively. Green dashed line indicates the maximum value of $D_{eff}$.

Spin-orbit torque induced current magnetization switching (SOT-CIMS) provides an energy-efficient way of manipulating the magnetization in the ferromagnetic layers. We present a detailed study of Dzyaloshinskii–Moriya and spin-orbit interactions in the Ti(2)/Co(1)/Pt(0-4)/Co(1)/MgO(2)/Ti(2) (thicknesses in nanometers) patterned into micrometer-sized Hall-bar devices. Here, the Pt is used as a source of the spin current, and as a nonmagnetic spacer whose variable thickness enables the magnitude of the ferromagnetic interlayer exchange coupling (IEC) to be effectively tuned [1]. From anomalous Hall effect (AHE), anisotropic magnetoresistance (AMR) and spin Hall magnetoresistance (SMR) measurements, we found that the increase in Pt thickness ($t_{Pt}$) leads to the reorientation of Co-magnetizations from the in-plane to the perpendicular direction at $t_{Pt} \approx 1.3$ nm. Further increase in Pt thickness, over 3 nm, reduces the ferromagnetic coupling and consequently, two weakly coupled Co layers become magnetized orthogonally to each other. From analysis of the Stokes and anti-Stokes peaks spectra measured by the Brillouin light scattering (BLS), the value of effective DMI constant ($D_{eff}$) was determined as a function of Pt spacer thickness (Fig. 1). These quantities reach their highest value for Pt thicknesses of around 2 nm, where the perpendicular anisotropy is the largest [1]. Magnetic domain images obtained by the polar-magnetooptical Kerr microscopy (P-MOKE) demonstrated the skyrmion bubble domains in the region of strong coupling, which disappears with decreasing ferromagnetic coupling. The asymmetric expansion of the bubble domain indicates the counterclockwise (CCW) chirality of the Néel-type DWs (Fig. 2). The results obtained for the trilayer were compared with the Pt(4)/Co(1)/MgO bilayer system. Finally, we investigated SOT-CIMS in both systems and analyse the switching mechanism using Landau-Lifshitz-Gilbert-Slonczewski equation.

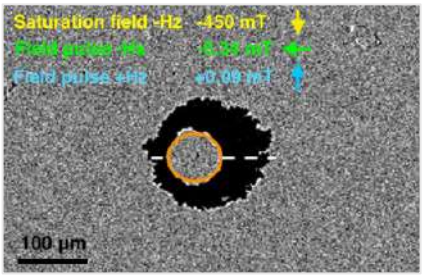



*Fig. 2 Differential P-MOKE image of bubble domain growth with an applied in-plane $H_x$ and perpendicular $H_z$ field for Co(1)/Pt(2.2)/Co(1). Orange ring indicates the initial position of the bubble domain.*

**Acknowledgements**

This work was supported by National Science Center, Poland, Grant No.: 2016/23/B/ST3/01430. KG acknowledges support for conference participation by the EU Project POWR.03.02.00-00-I004/16.

**References**

[1] P. Ogrodnik et al., *ACS Appl. Mater. Interfaces* 13, 47019 (2021)

# SYMPOSIUM 14.
## MAGNETISM IN CONFINED SYSTEMS AND 3D NANOMAGNETISM.
## S14 INVITED ORAL PRESENTATIONS

# Interplay of Œrsted Field and Curling Structures in Cylindrical Nanowires

*L. Álvaro Gómez[1,2,3*], A. Masseboeuf[1], J. Hurst[1], S. Ruiz Gómez[5], C. Fernández González[5], M. Schöbitz[1,4], A. De Riz[1], N. Mille[6], M.W. Khaliq[7], J. Bachmann[8], M.A. Niño[7], M. Weigand[10], S. Wintz[9],M. Foerster[7] L. Aballe[7], R. Belkhou[6], J.C Toussaint[4], C. Thirion[4], L. Pérez[2,3], D. Gusakova[1], O. Fruchart[1]*

[1] *Univ. Grenoble Alpes, CNRS, CEA , SPINTEC, 38000, Grenoble, France*
[2] *IMDEA Nanociencia, Campus de Cantoblanco, 28049 Madrid, Spain*
[3] *Dpto. de Fisica de Materiales, Universidad Complutense de Madrid, 28040 Madrid, Spain*
[4] *Univ. Grenoble Alpes/CNRS, Institut Néel, 38000 Grenoble, France*
[5] *Max Planck Institute for Chemical Physics of Solids, 01187 Dresden, Germany*
[6] *SOLEIL Synchrotron, L'Orme des Merisiers, Saint-Aubin, 91192, Gif-sur-Yvette Cedex, France*
[7] *ALBA Synchrotron Light Facility, CELLS, 08290, Barcelona, Spain*
[8] *Friedrich-Alexander Universität Erlangen-Nürnberg, Inorganic Chemistry, 91058 Erlangen, Germany*
[9] *Max Planck Institute for Intelligent Systems, 70569 Stuttgart, Germany*
[10] *Helmholtz-Zentrum Berlin für Materialien und Energie, 12489 Berlin, Germany*
*laualv10@ucm.es

Cylindrical magnetic nanowires are a prototypical system for exploring the physics of magnetization dynamics, and are also proposed as building blocks for three-dimensional storage devices. They host a topologically-unique Bloch-Point-Wall (BPW), which is predicted to push back the Walker limit and reach velocities in excess of 1 km/s [1]. A crucial feature of BPWs and other spin textures in nanowires, common to three-dimensional systems, is curling of magnetization induced by dipolar energy. Another key feature is the long-overlooked strong Œrsted field that arises when an electric current flows in the wire, while playing a key role by directly coupling to curling structures. Here we propose an overview of recent developments in the field, combining micromagnetic simulations, synthesis, and static and time-resolved advanced magnetic microscopy.

In a recent experiment, we reported BPW velocity above 600 m/s under a ns pulse of spin-transfer-torque [2], dynamically stabilised by the Œrsted field during motion. Simulations suggest a non-trivial underlying mechanism for the switching of circulation in case of an initially-antiparallel Œrsted field, with interplay between volume topological objects (Bloch points) and surface objects (vortex-antivortex pairs).

In search of an extra handle to control domain walls, we also considered Permalloy nanowires with short $Fe_{80}Ni_{20}$ chemical modulations, and diameter ~100 nm. Magnetization spontaneously curls at the modulations, and can be switched by a ns Œrsted field, similar to BPWs [3]. We then investigated the interplay of modulations and walls, which depends on their relative curling, and promotes various types of DW transformations (FIG.1 a-c). Finally, we provide 50 ps time-resolved experiments of both modulation and BPW switching (FIG. 1 d) that allow direct bridging with the micromagnetic simulations.

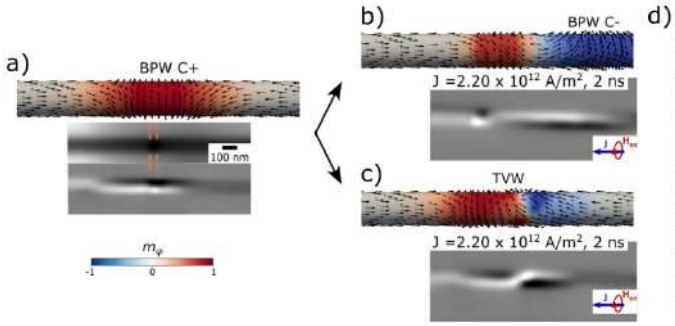



Figure 1: a) Micromagnetic simulation (top) and ptychographic reconstructed XAS and XMCD images (bottom) of a BPW pinned at a chemical modulation. b) and c) show DW transformations after the application of a 2 ns pulse of J = $2x10^{12}$ A/m$^2$ to state in a). d) Time resolved STXM imaging of BPW circulation switching.

## References

[1] R. Hertel, *J. Phys. Condens. Matter* **28**, 483002 (2016).
[2] M. Schöbitz, et al., *Phys. Rev. Lett.* **123**, 217201 (2019).
[3] L. Álvaro-Gómez, et al., *Phys. Rev. B*, **106**, 054433 (2022).

# Local and non-local chirality breaking effects in curvilinear nanoarchitectures

*Oleksii M. Volkov*[1] *

[1] *Helmholtz-Zentrum Dresden-Rossendorf e.V., Bautzner Landstr. 400, 01328 Dresden, Germany*

*o.volkov@hzdr.de

The main origin of the chiral symmetry breaking and, thus, for the magnetochiral effects in magnetic materials is associated with an antisymmetric exchange interaction, the intrinsic Dzyaloshinskii-Moriya interaction (DMI) [1,2]. The later manifests itself in magnetic materials or layer stacks with structural space inversion symmetry breaking. The DMI is responsible for the formation of non-trivial chiral and topological spin textures (e.g. skyrmions, bubbles, homochiral spirals and domain walls), that are envisioned to be utilized for prospective spintronic devices. At present, tailoring of magnetochirality is done by the selection of materials and adjustment of their composition.

Alternatively, we demonstrate that space inversion symmetry breaking of the magnetic order parameter appears in geometrically curved systems [3]. In curvilinear ferromagnets, curvature governs the appearance of geometry-induced chiral and anisotropic responses, which introduce a new toolbox to create artificial chiral nanostructures from achiral magnetic materials suitable for the stabilization of non-trivial chiral textures [4,5,6]. Moreover, curvilinear geometry also leads to the appearance of non-local chiral effects, that arise from the asymmetry of the top and bottom surfaces and existence of both in- and out-of-plane magnetization components of different parity with respect to the reflection procedure [5]. Recently, we demonstrate the existence of non-local chiral effects in geometrically curved asymmetric permalloy cap with the vortex texture. We find that the equilibrium vortex core obtain both bend and curling deformation, that are dependent on the geometric symmetries and magnetic parameters.

## Acknowledgements

This work is done in collaboration with Denys Makarov, Oleksandr V. Pylypovskyi, Attila Kákay, Jürgen Fassbender (Helmholtz-Zentrum Dresden-Rossendorf e.V., Dresden, Germany), Daniel Wolf, Axel Lubk and Bernd Büchner (Institute for Solid State Research, IFW Dresden, Dresden, Germany) and Denis D. Sheka (Taras Shevchenko National University of Kyiv, Kyiv, Ukraine). This work is financed in part via the German Research Foundation (DFG) under Grants No. MA 5144/14-1, MA 5144/22-1, MA 5144/24-1, KA 5069/1-1, KA 5069/3-1, VO 2598/1-1 and MC 9/22-1.

## References

[1] I. Dzyaloshinsky, J. Phys. Chem. Solids **4** (1958), 241.
[2] T. Moriya, Phys. Rev. Lett. **4** (1960), 228.
[3] R. Hertel, SPIN **3** (2013), 1340009.
[4] D. Makarov, et al., Adv. Mater. **34** (2021), 2101758.
[5] D. D. Sheka, et al., Commun. Phys. 3 (2020), 128.
[6] O. M. Volkov, et al., Phys. Rev. Lett. **123** (2019), 077201.

# SYMPOSIUM 14.
## MAGNETISM IN CONFINED SYSTEMS AND 3D NANOMAGNETISM.
## S14 ORAL PRESENTATIONS

# Skyrmionic 3D cocoons in aperiodic magnetic multilayers

*M. Grelier,[1] R. Battistelli,[2] S. Collin,[1] A. Vecchiola,[1] F. Godel,[1] K. Bouzehouane,[1] C. Donnelly,[3] Finizio,[5] F. Büttner,[2,6] H. Popescu,[4] N. Jaouen,[4] N. Reyren[1], V. Cros,[1,*]*

[1] *Unité Mixte de Physique, CNRS, Thales, Université Paris-Saclay, Palaiseau, France*
[2] *Helmholtz-Zentrum Berlin, Berlin, Germany*
[3] *Max Planck Institute for Chemical Physics of Solids, Dresden, Germany*
[4] *Synchrotron SOLEIL, L'Orme des Merisiers, Gif-sur-Yvette, France.*
[5] *Paul Scherrer Institut, Villigen PSI, Switzerland*
[6] *Institüt für Physik, Universität Augsburg, Augsburg, Germany*
*Vincent.cros@cnrs-thales.fr

Beyond the large number of studies on 2D topological textures such as magnetic skyrmions, a new interest has surged for more complex magnetic quasi-particles that display variations over the thickness, i.e., three-dimensional (3D) objects, leading to the discovery of new categories of topological texturese.g. truncated skyrmions [1], bobbers [2] or even hopfions [3] in magnetic multilayers.

In this work, we show how by engineering Pt/Co/Al based multilayers with variable Co thickness, we observe the signature of new textures, called skyrmionic cocoons [4] that are only present in a fraction of the magnetic layers. Interestingly, these cocoons can coexist with more standard 'tubular' skyrmions going through all the multilayer as evidenced by the existence of two very different contrasts in the magnetic force microscopy (MFM) images recorded at room temperature that can be easily correlated with the corresponding micromagnetic simulations (Fig. 1a-b). One major shortcoming of studying 3D textures is the difficulty to access information about the bulk magnetization. To this end, we also performed magneto-transport measurements as well as X-ray measurements at various synchrotrons. In Fig. 1c, an image acquired by holography, a transmission technique, clearly shows different contrasts that thus corresponds to objects with various vertical extension. Moreover, we have also obtained a reconstruction of the magnetization, measured with X-ray laminography (not shown) evidencing the different magnetic textures in our aperiodic multilayers. Their coexistence and the discovery of a novel magnetic texture are particularly interesting as they can open new paths for three-dimensional spintronics.

**Acknowledgements**

Financial supports from FLAG-ERA Sograph MEM (ANR-15-GRFL-0005), from ANR 17-CE24-0025 (TOPSKY) and ANR-20-CE42-0012-01(MEDYNA), and ANR/DFG (Topo3D) and as part of the "Investissements d'Avenir" program SPiCY (ANR-10-LABX-0035).

**References**

[1] A.O. Mandru et al. Nature communications 11,1 (2020)
[2] F. Zheng et al. Nature Nanotechnology 13, 451 (2018)
[3] N. Kent et al. Nature communications 12, 1 (2021)
[4] M.Grelier, et al. Nature Communications 13.1 (2022): 6843.
[5] M.Grelier, et al. arXiv:2302.02637

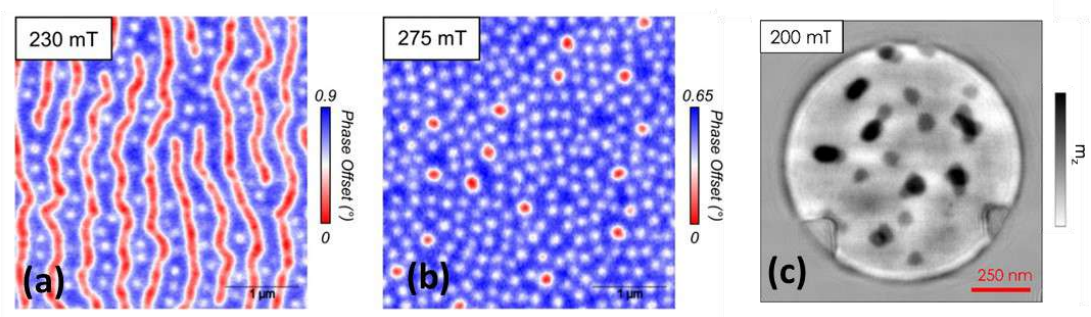



Figure: (a-b) experimental MFM phase maps displaying two types of textures (two different contrasts). (c) X-ray Fourier transform holography, a transmission technique, showing isolated cocoons (gray dots) and paired ones (black dots).

# High-Purity Ferromagnetic Nanotubes grown by Focused Electron Beam Induced Deposition

*César Magén[1,2,3*], Javier Pablo-Navarro[1,2], Luis Alfredo Rodríguez[4], Christophe Gatel[5], Ingrid-Marie Andersen[5], Etienne Snoeck[5], and José María de Teresa[1,2,3]*

[1] *Instituto de Nanociencia y Materiales de Aragón (INMA), Universidad de Zaragoza-CSIC, 50009 Zaragoza, Spain.*

[2] *Departamento de Física de la Materia Condensada, Universidad de Zaragoza, 50009 Zaragoza, Spain.*

[3] *Laboratorio de Microscopías Avanzadas (LMA), Universidad de Zaragoza, 50018 Zaragoza, Spain.*

[4] *Grupo de Transiciones de Fase y Materiales Funcionales, Departamento de Física, Universidad del Valle, A.A. 25360 Cali, Colombia.*

[5] *Centre d'Élaboration de Matériaux et d'Etudes Structurales (CEMES), 1055, Toulouse, France.*

*cmagend@unizar.es

Three-dimensional nanomagnets and curved magnetism are nowadays exciting topics in Nanomagnetism. The appearance of novel spin textures and topologically protected magnetization states may be the key to develop 3D spintronic devices for magnetic data storage, logic and sensing [1]. One of these exciting architectures are ferromagnetic nanotubes (NTs), in which the predicted core-less spin states are ideal for stable and fast domain wall motion beyond the Walker limit [2]. In this context, Focused Electron Beam Induced Deposition (FEBID) has become an exceptional nanolithography technique to produce high-quality 3D magnetic architectures.

In this work, FEBID growth [3] and thermal annealing procedures [4] have been combined to synthesize 3D cobalt nanotubes. 12-nm-thick cobalt layers have been deposited on vertical Pt-C nanowire templates with a diameter of ~70 nm, and then annealed at 450 ºC in high vacuum to produce high-purity, chemically and structurally homogeneous Co NTs (see Fig. 1a,b). Quantitative magnetic imaging of the remanent state and domain wall structure of a 3D ferromagnetic cylindrical NT has been achieved by off-axis Electron Holography (EH). Experiments carried out in an aberration-corrected Lorentz (field-free) transmission electron microscope have enabled the imaging of the remanent magnetization states. Both as-grown and thermally-annealed cylindrical NTs evidence the nucleation and pinning of a head-to-head vortex domain wall, whose structure has been confirmed by detailed micromagnetic simulations, as shown in Fig. 1c-e.

## Acknowledgements

We acknowledge the research project PID2020-112914RB-100 funded by MCIN/AEI/10.13039 /501100011033, the European Union's Horizon 2020 research and innovation program under grant agreement no. 823717-ESTEEM3, and Gobierno de Aragón through the project E13_20R.

## References

[1] A. Fernández-Pacheco *et al*. *Nature Comm.* **8** (2017) 15756.
[2] M. Staňo, O. Fruchart, *Handbook of Magnetic Materials* **27** (2018) 155.
[3] J. Pablo-Navarro, *et al. Nanotechnology* **27** (2016) 285302; *ACS Appl. Nano Mater.* **1** (2018) 38.
[4] J. Pablo-Navarro *et al. in preparation* (2023).

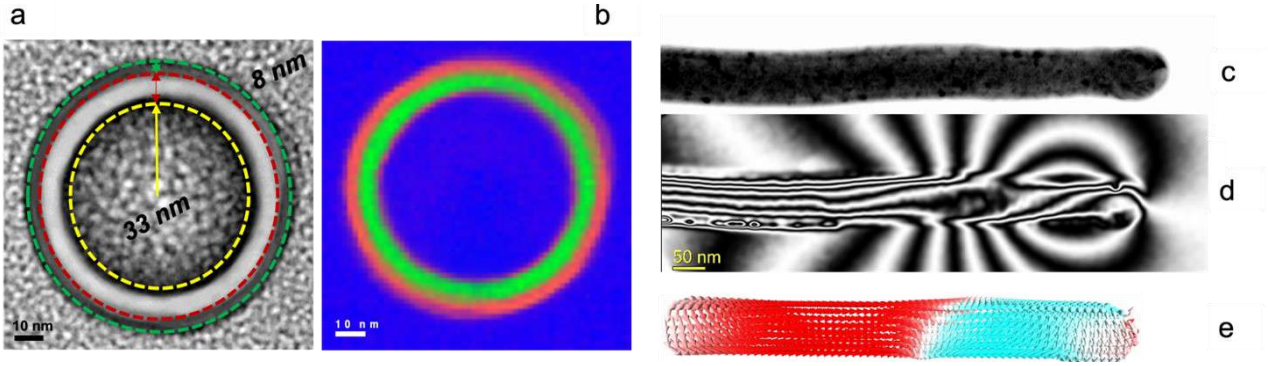



Figure 1: Structural, chemical and magnetic characterization of annealed FEBID-grown Co nanotubes. (a) Cross-sectional HAADF-STEM and (b) STEM-EELS chemical map (green = cobalt, red = oxygen, blue = carbon). (c) Bright field image. (d) Electron holography magnetic flux lines. (e) Spin configuration obtained by micromagnetic simulation.

# Influence of electrochemical surface modification on the magnetization reversal of core/shell Ni@(NiO,Ni(OH)$_2$) nanowires

*J. García[1,]*, R. Gutiérrez[1], A. S. González[1], A. I. Jiménez-Ramirez[1], Y. Álvarez[1], V. Vega[2], H. Reith[3], K. Leistner[3,4], C. Luna[5], K. Nielsch[3] and V.M. Prida[1,]**

[1] *Depto. Física, Universidad de Oviedo, C/Federico García Lorca nº 18, 33007-Oviedo, Spain*

[2] *Lab. Membranas Nanoporosas (SCTs), Universidad de Oviedo, 33006-Oviedo, Spain*

[3] *IFW-Dresden, Helmholtzstraße 20, 01069 Dresden, Germany*

[4] *Faculty of Natural Sciences, TU Chem-nitz, Strasse der Nationen 62, Chemnitz 09111, Germany*

[5] *Facultad de Ciencias Físico Matemáticas (FCFM-UANL), Nuevo León 66455, Mexico*

*garciafjavier@uniovi.es

The implementation of new groundbreaking technologies is increasingly requiring a full control of the magnetic properties of materials at the nanoscale. Besides the optimization of alloy composition, crystalline structure and geometry of the nanomaterials, magnetic heterostructures are postulated as target systems for the precise tunning of functional magnetic properties. In this regard, ferromagnetic(FM)/antiferromagnetic(AFM) coupled layers have been widely used to modify or generate unidirectional magnetic anisotropies, which is of great interest in heads of magnetic recording media based on giant magnetoresistance[1]. In addition to studying the magnetic properties and Exchange Bias effect on stacked thin films nanostructures, FM/AFM interactions have been also pursued in core/shell nanoparticles. However, core/shell cylindrical nanowires with FM core and AFM shell are investigated in a lesser extent. In this work, pure electrochemical approach has been used to fabricate core(FM)/shell(AFM) Ni@(NiO,Ni(OH)$_2$) nanowire arrays avoiding thermal oxidation procedures incompatible with integrative semiconductor technologies. Their morphological and compositional characterization have been carried out by SEM and HR-TEM techniques so as by XPS analysis. Temperature dependent magnetic hysteresis loops, thermomagnetic curves and FORC analysis, have been used to determine the influence of the electrochemical modification of Ni nanowires' surface on their magnetic properties. First of all, a magnetic hardening of nanowires along the parallel direction of applied magnetic field with respect their long axis (easy magnetization axis) has been found to be around 17% (43%) at 300K (50K). On the other hand, when field cooling (3T) the Ni@(NiO,Ni(OH)$_2$) nanowires below 100K along their easy magnetization axis, an increasing exchange bias effect has been encountered as the temperature decreases. .

## Acknowledgements

Spanish MCINN under research grant No. PID2019-108075RB-C32/AEI/10.13039/501100011033 is gratefully acknowledged for financial support.

## References

[1] C. Chappert, A. Fert, F.N. van Dau, *Nat. Mater.* **6** (2007) 813–823.

[2] J. Nogués, J. Sort, V. Langlais, V. Skumryev, S. Suriñach, J.S. Muñoz, M.D. Baró, *Phys. Rep.* **422** (2005) 65–117.

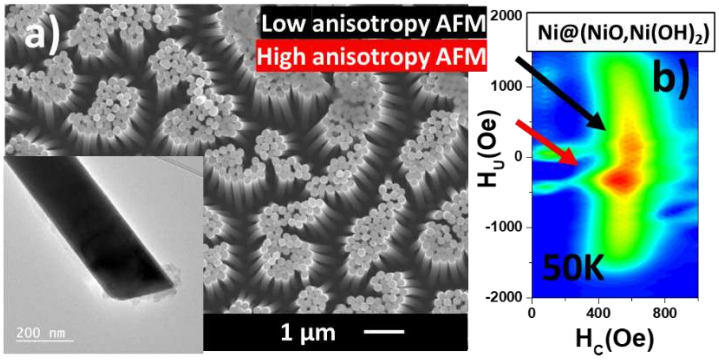



Figure 1: a) SEM image of electrochemical surface modified for Ni@(NiO,Ni(OH)$_2$) nanowires. The inset shows a high magnification HRTEM image of a single nanowire. b) FORC distribution measured at 50K shows two different magnetic behaviours ascribed to the different magnetic interactions between the core and the shell of the nanowires.

# Supermagnonic Bloch point velocities by jet propulsion effect in iron cylindrical nanowires

*Oksana Chubykalo-Fesenko[1]*, Felipe Tejo[2], Jose Angel Fernánez-Roldan[3], Konstantin Guslienko[4], Ruben Otxoa[5]*

[1] *Instituto de Ciencias de Materiales de Madrid, CSIC, 28049 Madrid, Spain*
[2] *Universidad Central de Chile, Santiago de Chile, Chile*
[3] *Institute of Ion Beam Physics and Materials Research, Helmholtz-Zentrum Dresden-Rossendorf, Germany*
[4] *Universidad del País Vasco, UPV/EHU, San Sebastian, Spain*
[5] *Hitachi Cambridge Laboratory,UK*
*oksana@icmm.csic.es

Achieving high velocities of magnetic domain walls is a crucial factor for their use as information carriers in modern spintornic applications. Unfortunately, in ferromagnetic materials the domain wall dynamics under applied fieldor current suffers from the so-called Walker breakdown phenomenon [1]. Cylindrical geometry offers possibilities to achieve very high velocities of magnetic domain walls due to the predicted absence of this effect. In our numerical modelling [2], we report very high velocity (up to 14 km/s) of the Bloch point domain wall under applied field in cylindrical magnetic nanowires of high magnetization value such as Fe. Domain wall is predicted to have a conical shape due to different magnetization propagation speed on the nanowire surface and in its center.

The obtained velocities are much higher than the ``magnonic (or relativistic) limit", estimated analyticlaly from the spinwave spectrum as being close to 2 km/s. We associate the increase of velocity to the jet-propulsion effect by a constant ejection of new Bloch points from the domain wall. The internal instabilities break the cone and give birth to a pair of domain walls which are ejected by the main comain wall. This creates a jet propulsion effect, accelerating the DW propagation along of the nanowire axis. The effect is larger in nanowires with high saturation magnetization due to its magnetostatic origin.

## References

[1] N.L.Schryer, L.R. Walker, L.R.: *J. Appl. Phys.* **45** (1974) 5406
[2] F. tejo et al arXiv:2302.12329

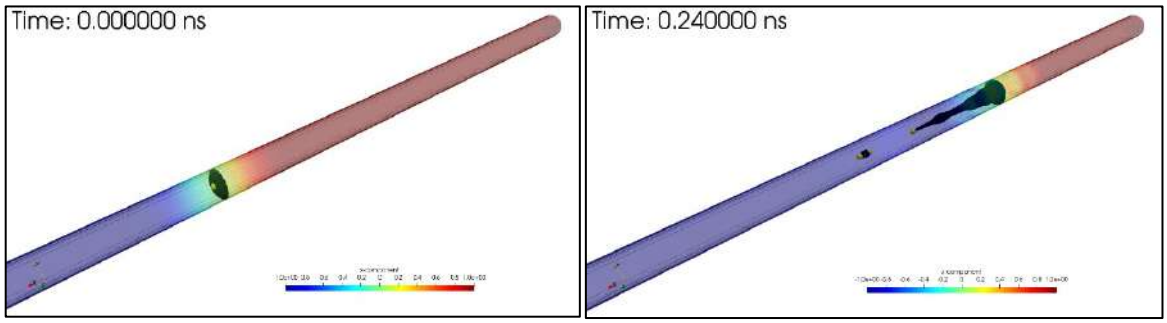



Figure: Images of domain wall dynamics in Fe cylindrical nanowire of 60 nm diameter under applied field B=60mT, showing the elongation of the Bloch point domain wall and ejection of a pair of Bloch points.

# 200 ps Current-Induced Œrsted-Field Switching of Curling in Chemically-Modulated Nanowires

*L. Álvaro-Gomez[1,2], C. Thirion[3], S. Wintz[4], M. Weigand[4], J.C. Toussaint[2], D. Gusakova[1], L. Perez[2], A. Masseboeuf[1], O. Fruchart[1]**

[1] *Univ. Grenoble Alpes, CNRS, CEA, SPINTEC, Grenoble, France*
[2] *IMDEA Nanociencia, Madrid, Spain*
[3] *Univ. Grenoble Alpes, CNRS, Grenoble INP, Instiut Néel, Grenoble, France*
[3] *Helmholz Zentrum Berlin, Germany*
*olivier.fruchart@cea.fr

Many theoretical and micromagnetic simulation reports over the past 20 years have considered domain walls and their motion in cylindrical magnetic nanowires [1]. A specific aspect in them is the existence of a unique domain wall, consisting of a Bloch point surrounded by azimuthal curling of magnetization at the wire periphery, whose velocity is predicted to be around 1km/s. Experiments have confirmed its existence and speed of several hundreds of m/s. In parallel, more elaborate situations are being explored in theory, simulation and experiments, such as smple or core-shell tubes, wires modulated in diameter or chemical composition etc.

Here we consider Permalloy $Fe_{20}Ni_{80}$ nanowires with micrometers-long segments separated by the insertion of $Fe_{80}Ni_{20}$ platelets of thickness a few tens of nanometers. Magnetostatic energy induces the occurrence of curling magnetization at the modulations, whose circulation has been shown to be switchable using 1-10 ns electrical current pulses, driven by the associated Œrsted field [2]. Here, we report time-resolved (TR) magnetic imaging using Scanning Transmission X-ray Microscopy (STXM), revaling their switching mechanism with a 50 ps time resolution. The spin texture is very sharp during the billions-averaged TR series, indicating a very reproducible process. The threshold current for switching is around $5 \times 10^{11}$ A/m$^2$. Above typically $7 \times 10^{11}$ A/m$^2$ the circulation switching occurs in less than 200 ps. This is similar to the rise time of the current pulse at the sample. This points at an ultrafast swiching mechanism, which we explain by the strength of Œrsted field and dynamic dipolar field in nanowires. When approaching the current threshold the switching time tends to diverge, consistent with expectations from theory and simulations. A comparison with micromagnetic simulations and analytical simulations is underway, with a view to provide a comprehensive picture of this ultrafast phenomenon, which we believe should be common to other curvilinear systems.

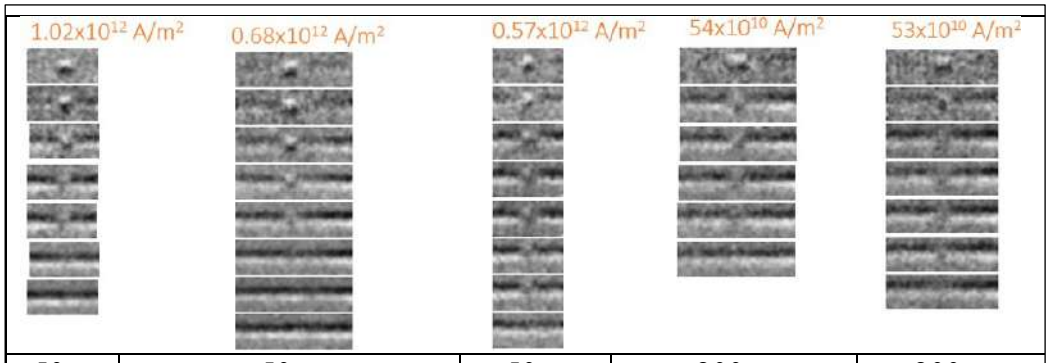
## Acknowledgements

French RENATECH network, The team of the Nanofab platform (CNRS Institut Néel).

## References

[1] M. Stano, O. Fruchart, Magnetic nanowires and nanotubes, in Handbook of magnetic materials **27**, E. Brück Ed., Elsevier (2018)

[2] L. Álvaro-Gómez, S. Ruiz-Gómez, C. Fernández-González, M. Schöbitz, N. Mille, J. Hurst, D. Tiwari, A. de Riz, I. M. Andersen, J. Bachmann, L. Cagnon, M. Foerster, L. Aballe, R. Belkhou, J. C Toussaint, C. Thirion, A. Masseboeuf, D Gusakova, L. Pérez, O. Fruchart, Phys. Rev. B 106 (2022) 054433.




TR differential magnetic images under various densities of charge current and time steps (second row). The top one is the (static) initial state, displaying curling at the modulation. Curling of magnetization in the domains during the pulse is a dynamic effect, resulting from the Œrsted field. It disappears after the pulse ends, leaving the switched circulation at the modulation (not shown here).

# Domain Wall Propagation and Pinning Induced by Current Pulses in Cylindrical Modulated Nanowires

*Cristina Bran[1*], Jose A. Fernandez-Roldan[2], Julian A. Moreno[3], Arantxa Fraile Rodríguez[4,5], Rafael P. del Real[1], Agustina Asenjo[1], Elias Saugar[1], Jorge Marqués-Marchán[1], Michael Foerster[6], Lucia Aballe[6], Jurgen Kosel[3,7], Manuel Vazquez[1], Oksana Chubykalo-Fesenko[1]*

[1] Instituto de Ciencia de Materiales de Madrid, 28049 Madrid, Spain
[2] Helmholtz-Zentrum Dresden-Rossendorf e.V., Institute of Ion Beam Physics and Materials Research, Bautzner Landstrasse 400, 01328 Dresden, Germany
[3] King Abdullah University of Science and Technology, Thuwal 23955-6900, Saudi Arabia.
[4] Universitat de Barcelona, Barcelona, 08028, Spain
[5] Institut de Nanociencia i Nanotecnologia (IN2UB) Barcelona, 08028, Spain
[6] ALBA Synchrotron Light Facility, CELLS, Barcelona, 08290, Spain
[7] Sensor Systems Division, Silicon Austria Labs, Villach 9524, Austria
*cristina.bran@icmm.csic.es

One of the most promising magnetic systems studied in the last years are the three-dimensional (3D) nanostructures with curved geometry where novel and interesting magnetization textures and dynamics are involved [1-2]. The future implementation of these nanostructures in 3D spintronic devices require the control of domain wall dynamics by means of current pulses. While this has been extensively studied in 2D magnetic strips, few reports exist in 3D cylindrical geometry, where Bloch Point domain walls are expected to have intriguing properties. Here we report this investigation in cylindrical magnetic Ni nanowires with geometrical notches. Experimental work based on synchrotron X-ray magnetic circular dichroism (XMCD) combined with photoemission electron microscopy (PEEM) indicates that large current densities induce domain wall nucleation while smaller currents move domain walls preferably against the current direction (see Figure). In the region where no pinning centers are present we found domain wall velocity of about 1 km/s. Thermal modelling indicates that large current densities temporarily raise the temperature in the nanowire above the Curie temperature leading to nucleation of domain walls during the system cooling. Micromagnetic modelling with spin-torque effect shows that for intermediate current densities Bloch Point domain walls with chirality parallel to the Oersted field propagate antiparallel to the current direction. In other cases, domain walls can be bounced from the notches and/or get pinned outside their positions. We thus find that current is not only responsible for the domain wall propagation but is also a source of pinning due to the Oersted field action [3].

## References

[1] R. Streubel et al, J. Phys. D: Appl. Phys. 2012, 49, 363001.
[2] A. Fernández-Pacheco et al., Nat Commun 2017, 8, 15756.
[3] C. Bran, J.A. Fernandez-Roldan, J. A. Moreno, et al. 2023, https://doi.org/10.48550/arXiv.2210.01480

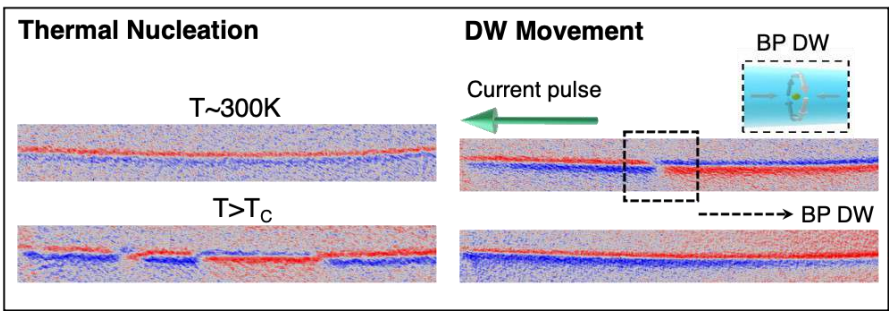



Figure: Thermal nucleation and fast current-induced Bloch Point Domain Wall (BP DW) motion in cylindrical modulated nanowires.

# Magnetic interactions in ordered cobalt three-dimensional nanonetworks

*Alejandra Ruiz-Clavijo[1], Olga Caballero-Calero[1], David Navas[2], Laura González Vivas[1], Carolina Martín-Rubio[3], Ruy Sanz[3]*, Marisol Martín-González[1]**

1. Instituto de Micro y Nanotecnología, IMN-CNM, CSIC, E-28760, Tres Cantos, Madrid, Spain (marisol@inm.csic.es)*

2. Instituto de Ciencia de Materiales de Madrid, ICMM-CSIC, 28049 Madrid, Spain

3 National Institute for Aerospace Technology (INTA), Payloads and Space Science Department, Spain (sanzgr@inta.es)*

Three-dimensional (3D) magnetic nanostructures allow the generation and control of novel effects that can give rise to the next generation of functional magnetic nanostructured metamaterials and devices[1]. This work reports some of our latest results on the synthesis and characterization of different interconnected Co nanowires forming a dense and ordered magnetic system: Co 3D Nanowire Networks (3DNN). These nanostructures were synthesized by the electrodeposition of Co for 3D ordered porous alumina templates [2-4].The route allows for control the order and some geometric features compared to other synthesis approaches [1]. The resulting 3DNN consist of hexagonally ordered nanowires with an inter-wire distance of 65 nm and a diameter of 50 nm. These nanowires are interconnected with a net of perpendicular or transversal nanowires (TNW) of around 30 nm in diameter. Meanwhile, the TNW is distributed in levels with well-defined distances between them of 450, 650, and 220 nm. The magnetic characterization consisted of hysteresis loops, first magnetization curves, and first order reversal curves at 0 (in-plane, IP) 30, 60, and 90º (out of plane, OOP) from the perpendicular NW main axis. All the characterized Co 3DNN arrays present different anisotropic magnetic responses compared to similar diameter and interwire distance Co NW arrays in porous alumina. The different responses of Co 3DNN depend on the distance between TNW. FORC measurements at different angles provided key information about the magnetic interactions in these 3D nanostructures.

## Acknowledgements

This work was supported by PID2020-118430GB-100, PID2020-115325GB-C31, RYC-2017-22820 funded by MCIN/AEI/ 10.13039/501100011033.

## References

[1] D. Bhattacharya, Z. Chen, C. J. Jensen, C. Liu, E. C. Burks, D. A. Gilbert, X. Zhang, G. Yin, K. Liu, *Nano Lett.* (2022), 22, 24, 10010–10017.

[2] J. Martín, M. Martín-González, J. Francisco Fernández, O. Caballero-Calero, *Nat. Commun.* 5, (2014) 5130.

[3] A. Ruiz-Clavijo, S. Ruiz-Gomez, O. Caballero-Calero, L. Perez, M. Martin-Gonzalez, M., *Phys. Status Solidi RRL*, 13 (2019), 1900263.

[4] A. Ruiz-Clavijo, O. Caballero-Calero, D. Navas, A. A. Ordoñez-Cencerrado, J. Blanco-Portals, F. Peiró, R. Sanz, M. Martín-González, *Adv. Electron. Mater.* 8, (2022) 2200342

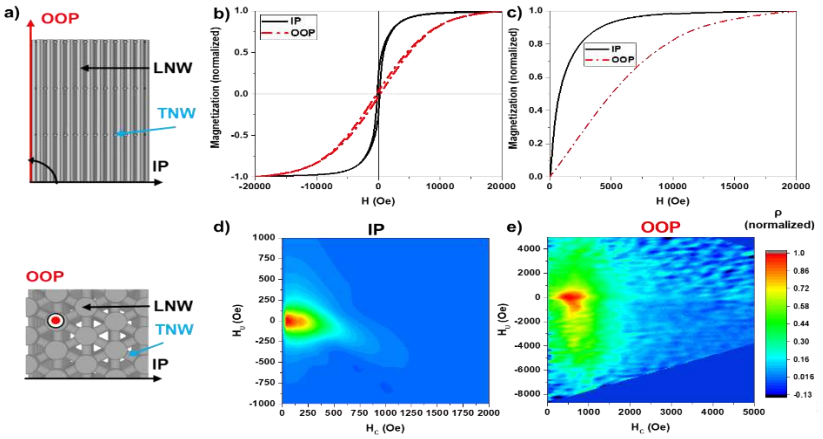



Figure: a) Scheme of the cross section and top view of Co 3DNN (LNW and TNW are denoted by arrows). For 3DNN with 200 nm between TNW b) Hysteresis loops c) first magnetization curves. d) and e) IP and OOP FORC diagrams.

# Structure and Controlled Nucleation of 3D Spin Textures in Aperiodic Multilayers

*R. Battistelli[1]*, M. Grelier[2], K. Litzius[3], M. Schneider[4], D. Metternich[1], T. Karaman[1], L.-M. Kern[4], C. Klose[4], N.Reyren[2], V. Cros[2], S. Eisebitt[4], B. Pfau[4] and F. Büttner[1,3]*

[1] *Helmholtz Zentrum Berlin, Hahn-Meitner-Platz 1, 14109 Berlin, Germany*
[2] *Unité Mixte de Physique CNRS/Thales, avenue Augustin Fresnel 1, 91767 Palaiseau, France*
[3] *University of Augsburg, Universitätsstraße 2, 86159 Augsburg, Germany*
[4] *Max Born Institute, Max-Born-Straße 2A, 12489 Berlin, Germany*
*riccardo.battistelli@helmholtz-berlin.de

Magnetic skyrmions and other chiral spin structures are subject to intense research for their interesting physics and potential future applications. 3D magnetic solitons such as bobbers [1,2] or truncated skyrmions [3] promise even richer physics and higher information density, but have not yet been studied as extensively as their 2D counterparts. A versatile platform for the study of such textures are Pt/Co/Al-based multilayers with variable thickness of the individual magnetic layers. These materials stabilize 3D magnetic cocoons, solitons localized in a fraction of the magnetic layers that can coexist with tubular skyrmions [4].

In this work, we use high resolution, synchrotron-based imaging techniques (Fourier transform holography, FTH, and Scanning transmission x-ray microscopy, STXM) to advance the study of the structure and behavior of these magnetic solitons. Owing to the exceptional resolution and contrast of FTH, we obtained detailed 2D images of the magnetic configuration of the material, clearly resolving different coexisting states (Fig. 1a) and unveiling a dipolar repulsion between worm-like domains and magnetic cocoons (Fig. 1b).

We also observed a new type of 3D feature which we name the partial worm, an isolated worm-like spin texture also present only in a fraction of the multilayer stack (Fig. 1c).

Moreover, we demonstrated the current-driven nucleation of three-dimensional cocoons from magnetic states where partial worms are present (Fig. 1c). A sistematic study of the process suggest that the nucleation is driven by thermal effects (as for skyrmions) and that cocoons are preferentially nucleated at pinning points of the material (Fig. 1d). A similar relation to pinning points was also observed after field-induced nucleation, suggesting an important relationship between magnetic cocoons and material defects.

## References

[1] Zheng, F. et al., **13**, 451-455 (2018). (10.1038/s41565-018-0093-3)
[2] Ahmed, A. S. et al., Phys. Rev. Materials **2**, 041401 (2018). (10.1103/PhysRevMaterials.2.041401)
[3] Mandru, A. et al., Nat. Commun. 11 6365 (2020). (10.1038/s41467-020-20025-2)
[4] Grelier, M. et al., Nat Commun **13**, 6843 (2022). (10.1038/s41467-022-34370-x)

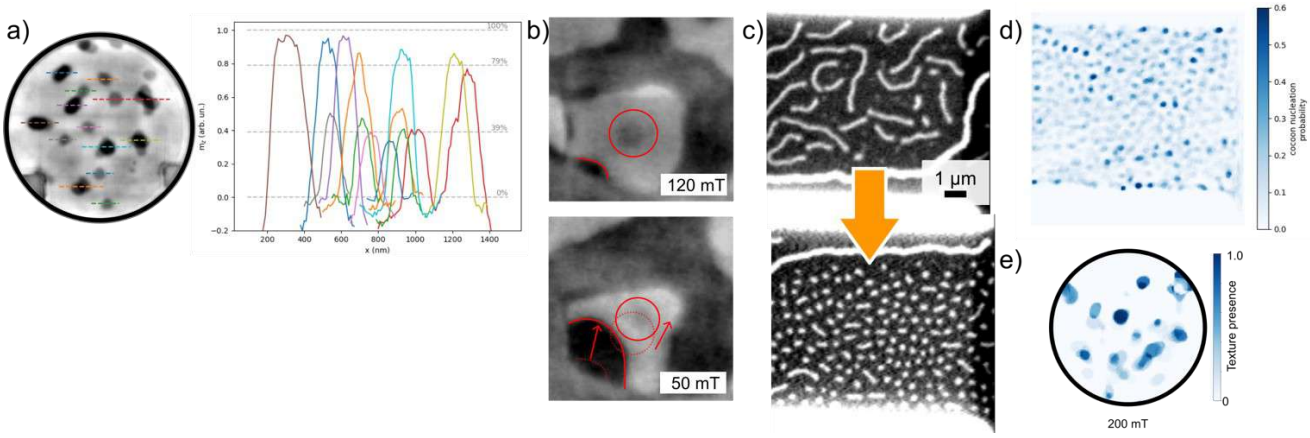



Figure 1: (a) High resolution image of a magnetic state hosting skyrmions and cocoons and relative linescans. (c) Domain-cocoon repulsion event. Red lines mark the positions of the domain wall and cocoons, while arrows highlight their motion. (c) cocoon nucleation via current pulses. (d-e) Maps of cocoon nucleation probability after current pulses and field cycling.

# Tailoring the energy landscape of domain walls with curvature

*Sandra Ruiz-Gomez[1*], Pamela Morales-Fernández[1], Claudia Fernandez-Gonzalez[1], Claas Abert[2,3], Lukas Danesi[2,3], Michael Foerster[4], Miguel Ángel Nino[4], Anna Mandziak[5], Dorota ilgocka-Ślęzakw[5], Ewa Madej[5], Markus König[1], Sebastian Seifert[1], Aurelio Hierro Rodríguez[6], Amalio Fernandez-Pacheco[7], Claire Donnelly[1]*

[1]*Max Planck Institute for Chemical Physics of Solids, 01187 Dresden, Germany*
[2]*Faculty of Physics, University of Vienna, 1010 Vienna, Austria*
[3]*Research Platform MMM Mathematics-Magnetism-Materials, University of Vienna, 1010 Vienna, Austria*
[4]*ALBA Synchrotron Light Source, CELLS, Cerdanyola del Valles, 08290, Barcelona, Spain.*
[5] *Jerzy Haber Institute of Catalysis and Surface Chemistry,, 30-239 Kraków, Poland.*
[6]*Depto. Física, Universidad de Oviedo, 33007 Oviedo, Spain*
*sandra.gomez@cpfs.mpg.de

Three-dimensional nanomagnetic systems, with novel and unconventional spin textures, represent an exciting platform to explore new magnetic phenomena, and also offers possibilities for the development of more efficient, capable and multifunctional technologies [1]. The three-dimensional geometry is predicted to have a significant influence on the dynamics of magnetic domain walls [2], soliton-like textures that form the basis of many spintronics devices proposed in recent years. In particular, through the introduction of curvature and torsion, properties such as anisotropy and chirality can be directly induced and controlled [3,4]. In this way, new physics and functionalities can be realised, from threedimensional chiral spin states [5] to ultrafast domain wall dynamics [6].

Here, we experimentally explore the influence of 3D geometry on the energetics of domain walls by introducing curvature into a magnetic nanowire system. By a careful design of the geometry of the nanostructures, grown by focused electron beam induced deposition (FEBID) [7], we are able to stabilize Bloch point domain walls and introduce well defined pinning positions. By measuring XMCD-images with soft x-ray magnetic microscopy after the applications of magnetic fields we demonstrate the control of the pinning strength by tuning the curvature of the nanostructure (Figure 1) . This insight into how control can be obtained via complex geometries will help pave the way to the next generation of 3D spintronic devices.

## References

[1] P. Fischer et al. APL Mater. 8, 010701 (2020).
[2] Robert Streubel et al 2016 J. Phys. D: Appl. Phys. 49 363001
[3] D. D. Sheka. Appl. Phys. Lett. 118, 230502 (2021)
[4] D. D. Sheka et al. Commun Phys 3, 128 (2020).
[5] A. Fernández-Pacheco et al. Nat. Comm. 8, 15756 (2017).
[6] M. Schobitz et al. Phys. Rev. Lett. 123, 217201 (2019).
[7] L. Skoric et al. Nano Lett. 20, 1 (2020).

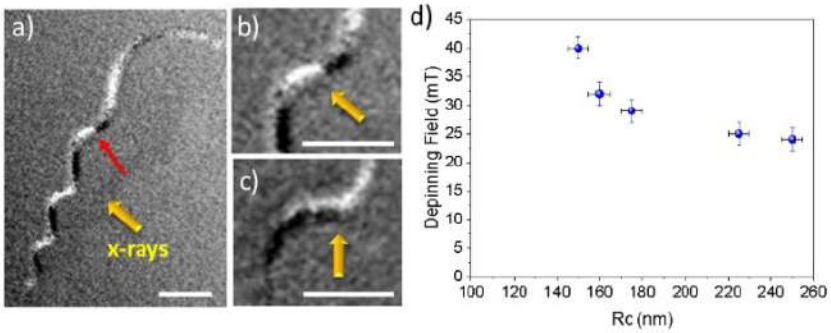



Figure1: a) XMCD image of an undulating structure after domain wall nucleation where the domain wall position is indicated with a red arrow. b) Zoom of the region with a domain wall of the XMCD showed in a). c) zoom of the same domain wall after rotating the sample to measure perpendicular to the wire. Yellow arrows mark the direction of the incoming beam. The scale bar is 1 µm in all cases. d) Depinning field as a function of radius of curvature extracted from a sequence of experiments applying magnetic fields parallel to the long axis of the structure.

# Curved Magnetism in $CrI_3$

*Alexander Edström[1*], Danila Amoroso[2], Silvia Picozzi[3], Paolo Barone[4], Massimiliano Stengel[5,6]*

[1*]*Department of Applied Physics, School of Engineering Sciences, KTH Royal Institute of Technology, AlbaNova University Center, 10691 Stockholm, Sweden*
[2]*NanoMat/Q-mat/CESAM, Université de Liège, B-4000 Liege, Belgium*
[3]*Consiglio Nazionale delle Ricerche CNR-SPIN, c/o Università degli Studi 'G. D'Annunzio', 66100 Chieti, Italy*
[4]*Consiglio Nazionale delle Ricerche CNR-SPIN, Area della Ricerca di Tor Vergata, Via del Fosso del Cavaliere 100, I-00133 Rome, Italy*
[5]*Institut de Ciència de Materials de Barcelona (ICMAB-CSIC), Campus UAB, 08193 Bellaterra, Spain*
[6]*ICREA—Instituciò Catalana de Recerca i Estudis Avançats, 08010 Barcelona, Spain*
*aleeds@kth.se

Recently, micromagnetic modeling has shown that curved magnets exhibit unusually rich phase diagrams, including chiral or topological spin states. [1,2] This is particularly relevant in 2D magnetic monolayers, such as CrI3, given their extreme flexibility and natural tendency to rippling. However, an understanding of such effects in real materials, from first principles theory, is challenging due to the breaking of translational symmetry. We have used non-collinear-spin density-functional theory to study the (flexomagnetic) coupling between magnetism and curvature in monolayer $CrI_3$. We show a crossover from a magnetization normal to the surface, to a cycloidal spin state, with increasing curvature. This cycloidal state is stabilized by curvature-induced, effective Dzyaloshinskii–Moriya contributions to the magnetic energy, and alterations of the magnetic anisotropy. While these terms remarkably appear as non-relativistic, geometric effects, in line with earlier predictions, our results reveal an unexpectedly large impact of spin-orbit coupling on their curvature dependence. [3]

Additionally, we use our results to derive a flexomagnetic coupling tensor and generalize our description for different geometries and magnetization states. This leads to predictions of new effects, such as flexomagnetoelectric/flexomagnetostrictive ones.

## Acknowledgements

We acknowledge support from the Swedish Research Council (2018-06807 and 2022-04720), ÅForsk and the Göran Gustafsson foundation. Calculations were done on resources provided by the Swedish National Infrastructure for Computing (SNIC).

## References

[1] D. Sheka, A Perspective on Curvilinear Magnetism. Applied Physics Letters **118** (2021) 230502.
[2] R. Hertel, Curvature Induced Magnetochirality. SPIN **3** (2013) 1340009.
[3] A. Edström, D. Amoroso, S. Picozzi, P. Barone, M. Stengel, Curved Magnetism in $CrI_3$. Physical Review Letters **128** (2022) 177202.

# Ferromagnetic Resonance in Gyroidal Networks

*M. Gołębiewski[1]*, R. Hertel[2], V. Vasyuchka[3], P. Pirro[3], M. Krawczyk[1], J. Llandro[4,5]*

[1] *Institute of Spintronics and Quantum Information, Faculty of Physics, Adam Mickiewicz University, Uniwersytetu Poznańskiego 2, 61-614 Poznań, Poland*

[2] *Université de Strasbourg, CNRS, Institut de Physique et Chimie des Matériaux de Strasbourg, F-67000 Strasbourg, France*

[3] *Fachbereich Physik and Landesforschungszentrum OPTIMAS, Rheinland-Pfälzische Technische Universität Kaiserslautern-Landau, Erwin-Schrödinger-Straße 56, 67663 Kaiserslautern, Germany*

[4] *Research Institute of Electrical Communication Information Devices Division Spintronics, Tohoku University, 2-1-1 Katahira, Aoba-ku, Sendai-shi, Miyagi 980-8577 Japan*

[5] *Center for Science and Innovation in Spintronics (CSIS), Tohoku University, Sendai 980-8577, Japan.*

*mateusz.golebiewski@amu.edu.pl

Topological or non-uniform geometric media may offer new possibilities for manipulating spin waves. 3D networks can facilitate interactions and collective effects in all dimensions, enabling new phenomena to be studied [1]. Gyroids are among the most promising yet largely unexplored structures in magnetism. They are defined by chiral triple junctions and periodicity along all three spatial directions (Fig. 1).

Nickel gyroid nanostructures were fabricated by thermally annealing a block copolymer template, selectively dissolving one of the gyroid-forming blocks and electrodepositing nickel into the voided right-handed gyroid network. The orientation of the gyroid network toward the static magnetic field axis appeared to be a critical factor in broadband ferromagnetic resonance measurements since we observed a strong impact of crystallography on the spectra signals as a variation in its primary intensities. Using the finite element method solver *tetmag* [2], we performed micromagnetic simulations of gyroid systems (Fig. 2) to understand better and explain the experimental results.

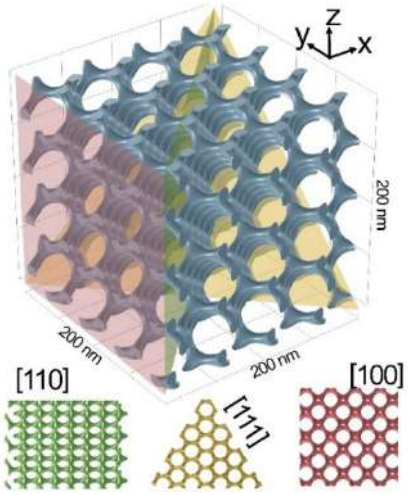



Figure 1: A geometric model of gyroid containing 4×4×4 unit cells (200×200×200 nm) was used. The three main symmetry directions of the gyroid network are shown below in the colors corresponding to the planes crossing the structure.

The findings demonstrate that geometric anisotropy can contribute significantly to the alternation in the intensity of resonance signals in rotating gyroid samples. Furthermore, ferromagnetic resonance measurements and micromagnetic simulations showed that the spin-wave spectra of nm-scale gyroids depend on the external magnetic field's orientation towards crystallography. These results offer great potential for developing 3D nanomaterials for magnonic applications.

**Acknowledgments**

The research that led to these results has received funding from the National Science Centre of Poland, project No. UMO-2020/39/I/ST3/02413, JSPS KAKENHI 21K04816, and the Graduate Program for Spintronics (GP-Spin), Tohoku University.

## References

[1] P. Fischer, D. Sanz-Hernández, R. Streubel, A. Fernández-Pacheco, *APL Materials* **8** (2020) 010701.
[2] R. Hertel, S. Christophersen, S. Börm, *Journal of Magnetism and Magnetic Materials* **477** (2019) 118-123.

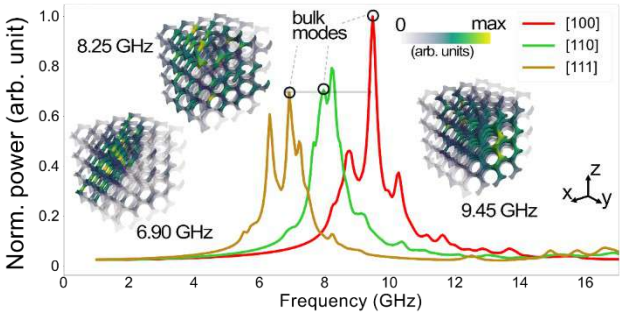



Figure 2: The resonance frequency spectra obtained from micromagnetic simulations. Different colors indicate the crystallographic direction along which the 300 mT field is applied, while the points marked with circles indicate the volumetric ferromagnetic resonance. In the order of occurrence, visualizations and values of resonant frequencies are visible as insets of the charts. The color scale corresponds to the amplitude modulus of the dynamic magnetization component.

# Three-Dimensional Nucleation Kinetics Through the First-Order Metamagnetic Phase Transition in B2-Ordered FeRh

*Jamie R. Massey[1,2], Samuel K. Treves[1,2], Andreas Apseros[1,2], Simone Finizio[2], Michael Grimes[1,2,3], Joakim Reutler[4], Claire Donnelly[5,6], Thomas Thomson[3], Laura J. Heyderman[1,2] and Valerio Scagnoli[1,2]**

[1]*Laboratory for Mesoscopic Systems, Department of Materials, ETH Zurich, 8093 Zurich, Switzerland*

[2]*Paul Scherrer Institute, 5232 Villigen PSI, Switzerland*

[3]*NEST Research Group, Dept. of Computer Science, The University of Manchester, Oxford Road, Manchester, M13 9PL, UK*

[4]*ScopeM, ETH Zurich, 8093 Zurich, Switzerland*

[5]*Cavendish Laboratory, University of Cambridge, Cambridge, CB3 0HE, UK*

[6]*Max Planck Institute for Chemical Physics of Solids, 01187 Dresden, Germany*

*valerio.scagnoli@psi.ch

B2-ordered FeRh undergoes a thermally activated first-order phase transition between antiferromagnetic (AF) and ferromagnetic (FM) order upon heating, with a transition temperature between 350 - 380 K [1], making it an ideal candidate for use in a wide range of magnetic storage architectures [2]. The evolution of magnetic domains through the transition has been well-characterized [1-5]. However, behaviours such as the domain nucleation mechanism and subsequent evolution of the magnetic domains along the sample thickness through the phase transition remain unclear [1,3-5]. Here, we present three-dimensional (3D) reconstructions of the magnetic structure through the sample volume at various temperatures through the phase transition, achieved using magnetic laminography [6-7]. Imaging cross-sections through the sample thickness reveal that domains for both types of magnetic order nucleate at the surface and travel into the bulk of the sample with changing temperature. Creating a complex 3D magnetic structure, in which the evolution of the magnetic state along the sample thickness with temperature is non-uniform. We also observe quasi-uniform FM domains at 310 K when heating, in contrast to the expected flux-closed state [1], as well as AF regions where FM domain walls are expected. Both observations can be attributed to the exchange coupling between adjacent AF and FM regions, which plays a key role determining the FM configuration in this material.

## References

[1] T. P. Almeida et al., Phys. Rev. Materials **4**, (2020) 034410.
[2] R. C. Temple et al., Phys. Rev. Materials **2**, (2018) 104406.
[3] C. Gatel et al., Nat. Comms. **8**, (2017) 15703.
[4] V. Uhlır et al., Nat. Comms. **7**, (2016) 13113.
[5] M. A. de Vries et al., Appl. Phys. Lett. **104**, (2014) 232407.
[6] C. Donnelly et al., Nature Nanotech. **15**, (2020) 356.
[7] K. Witte et al., Nano Lett. **20**, (2020) 1305.

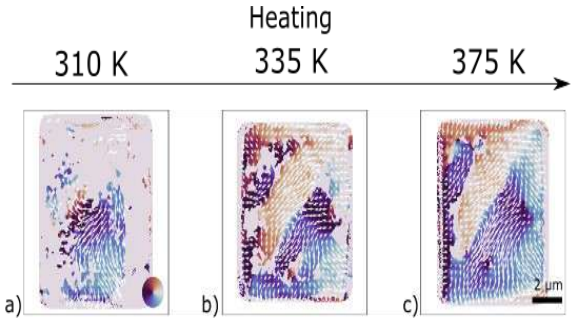



Figure: Reconstructed magnetic state when heating. (a)-(c) show the magnetic state for the top surface of the lamella. The sample area is shown in colour, which depicts the magnetization direction shown in the colour wheel in panel (a). The pink areas are those within the sample that are AF. At 310 K the sample shows a quasi-uniform FM domain, which develops into multi-striped domain at 375 K. The region between two adjacent, oppositely magnetized domains is the last to transition to being FM.

# SYMPOSIUM 14.
## MAGNETISM IN CONFINED SYSTEMS AND 3D NANOMAGNETISM. S14 POSTERS

# In-situ Modification of the Magnetocrystalline Anisotropy of Iron-rich Cobalt Ferrite by Iron Addition

*J. de la Figuera[1]*, A. Mandziak[2], S. Ruiz-Gómez[3], P. Prieto[4], J.E. Prieto[1], M. Foerster[5], L. Aballe[5]*

[1] *Instituto de Química Física "Rocasolano", 28006 Madrid, Spain*

[2] *Solaris Synchrotron, 30-392 Cracow, Poland*

[3] *Max-Planck-Institut für Chemische Physik fester Stoffe, 01187 Dresden, Germany*

[4] *Dpto. de Física Aplicada, Universidad Autónoma de Madrid, 28049 Madrid, Spain*

[5] *Alba Synchrotron Light Facility, CELLS, 08290 Barcelona, Spain*

*juan.delafiguera@csic.es

Cobalt ferrite is a ferrite with the highest magnetocrystalline anisotropy for cubic spinels. By increasing the iron content, its magnetic properties are strongly modified, up to reaching the pure iron spinel composition, magnetite. The latter is a soft ferrimagnet with a higher saturation magnetization. In the past we have shown that high-temperature oxygen-assisted molecular beam epitaxy provides epitaxial, micron-sized, crystallites of non-stoichiometric cobalt ferrite [1,2]. These crystals have a high crystalline quality and their magnetic structure has been studied in detail [3]. In this work we show how the magnetic properties of such crystals can be changed in-situ by modifying the composition. By depositing additional iron in oxygen atmosphere at high temperature, cobalt is expelled from the crystals, thus reducing their cobalt content, and therefore, their magnetocrystalline anisotropy. We map the island's magnetic domain structure by x-ray magnetic circular dichroism in photoemission microscopy (XMCD-PEEM). Azimuthal rotation of the samples provides the full vector magnetization of the crystals, allowing to observe the changes from a structure determined by magnetocrystalline anisotropy to one where the shape anisotropy plays a larger role on the same object (see Figure 1).

## Acknowledgements

This work is supported by the Grants PID2021-124585NB-C31 and TED2021-130957B-C54 funded by MCIN/AEI/10.13039/501100011033 and by "ERDF A way of making Europe" and by the "European Union NextGenerationEU/PRTR"

## References

[1] L. Martín-García, A. Quesada, C. Munuera, J.F. Fernández, M. García-Hernández, M. Foerster, L. Aballe, J. de la Figuera, *Adv. Mat.* **27** (2015) 5955.

[2] S. Ruiz-Gómez, A. Mandziak, J. E. Prieto, M. Aristu, E. M. Trapero, G. D. Soria, A. Quesada, M. Foerster, L. Aballe, J. de la Figuera, *J. Chem. Phys.* **152** (2020) 074704.

[3] S. Ruiz-Gómez, A. Mandziak, L. Martín-García, J. E. Prieto, P. Prieto, C. Munuera, M. Foerster, A. Quesada, L. Aballe, J. de la Figuera, *App. Surf. Sci.* **600** (2022) 154045.

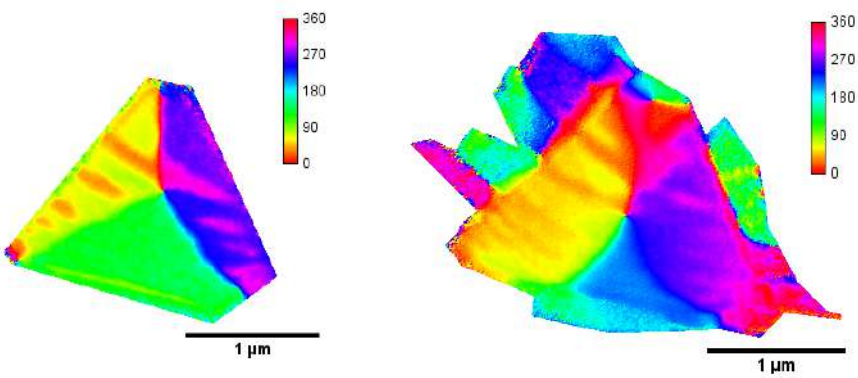



Figure: Left: In-plane magnetization of a cobalt ferrite island grown on Ru(0001). Right: in-plane magnetization after removing part of the cobalt from the island. Magnetic contrast is achieved by XMCD-PEEM at the Fe $L_3$ absorption edge.

# Topological spin textures in magnetic multilayer systems

*E.M. Jefremovas[1]*, F. Kammerbauer[1], M. Bhukta[1], M.A. Syskaki[1,2]. T. Dohi[1,3], R. Frömter[1], J. Sinova[1] and M. Kläui*

[1] *Institute of Physics, Johannes Gutenberg University Mainz, 55099 Mainz, Germany*
*2 Singulus Technologies AG, Hanauer Landstraße 103, 63796 Kahl am Main, Germany*
[3] *Tohoku University Research Institute of Electrical Communication Katahira 2-1-1, Aoba-ku 980-8577 Sendai, Japan*
*martinel@uni-mainz.de

Improving computing energy efficiency is one of the major challenges of XXI$^{st}$ century society. The current model of data reading and storage, based on charge transport, is unable to follow the exponentially growing demands. One of the most prominent alternatives to tackle this problem is taking advantage of the electron spin. This not only provides a new degree of freedom for information storage, yet it opens the way to topologically protected spin textures to be exploited as new information carriers. 2D-spin textures, known as *skyrmions*, are nanoscale size, stable even above room temperature, and require a low driving current density, enabling their implementation into (spintronic) circuits and devices [1]. Very recently, the possibility to use such magnetic solitons for non-conventional computing schemes, like reservoir computing, has been probed [2]. Nevertheless, within the neuromorphic framework, higher dimensional computational spaces are advantageous, and the limited internal degrees of freedom of skyrmions are not sufficient for complex tasks. Aiming to expand a topologically stabilized spin texture towards three dimensions, theoretical and experimental efforts have been pursued to investigate the feasibility of topological spin textures in materials [3- 5]. Among the most promising candidates, magnetic multilayer stacks, where the interplay between exchange, anisotropy, Dzyaloshinskii–Moriya and dipolar interactions can result in a stable 3D-topological spin texture, are promising [5, 6]. Notwithstanding, the role of each interaction and how to tune their interplay to obtain the desired spin configuration, still needs to be understood.

In this work, we have produced magnetic multilayers in the form $[Pt/Co_xFe_{80-x}B_{20}/Ir(Ru)]_n$, with varying layer thickness, repetitions ($n$) and magnetic composition to understand the role of each parameter in the stabilization of spin textures. Moreover, we have explored the role of lateral confinement in the stabilization of 3D topological spin textures by patterning magnetic multilayers with perpendicular magnetic anisotropy into nanodisk shape from several μm to hundred nm diameter size, aiming to relate the critical size range where 3D topological spin textures can be hosted. Indeed, a good control over the parameters needed to stabilize magnetic solitons is key to their applications, which go beyond those of skyrmions. As an example, the implementation of hopfions in neuromorphic designs, viewed as an anisotropic 3D-object whose state can be controlled, enables to modulate the scattering path in such a way that meta-learning can be realized [7].

## Acknowledgements

E.M.J. acknowledges the “Alexander von Humboldt Postdoctoral Fellowship”. This project has received funding from the European Research Council (ERC) under the European Union’s Horizon 2020 research and innovation programme (Grant No. 856538, project “3D MAGiC”)

## References

[1] A. Fert et al. *Nat. Rev. Mat.* **2**.7 (2017) 1-15.
[2] K. Raab et al. *Nat. Comm.* **13**.1 (2022) 6982.
[3] F. N. Rybakov et al. *APL Materials* **10**.11 (2022) 111113.
[4] P. Sutcliffe. *J. Phys. A: Math. and Theo.* **51**.37 (2018) 375401.
[5] M. Grelier et al. *Nat. Comm.* **13**.1 (2022) 6843.
[6] N. Kent et al. *Nat. Comm.* **12**.1 (2021) 1562.
[7] Z. Zhang et al. *Sci. Adv.* **9.6** (2023) eade7439.

# Voltage-controlled Magnetoresistance in $FeO_x$-Fe Coated Au Aerogels

*Martin Nichterwitz[1,2]*, Daniel Wolf [2], Karin Leistner[1,2]*

[1] *TU Chemnitz, Strasse der Nationen 62, 09111 Chemnitz, Germany*

[2] *Leibniz IFW Dresden, Helmholtzstrasse 20, 01069 Dresden, Germany*

*martin.nichterwitz@chemie.tu-chemnitz.de

Magnetoresistance (MR) is a key property of magnetic materials for advanced information storage or magnetic sensors. MR is mainly studied in thin film geometry, but the investigation of MR in 3D nanostructures currently becomes exciting from fundamental and application point of view.[1] At the same time, the control of MR by an external stimuli is important for application. Voltage-control of magnetism (VCM) is a promising pathway providing energy efficiency and easy applicability. In this context, magneto-ionic control based on ion migration and electrochemical reactions is an emerging route to VCM, providing advantages such as non-volatility and low voltage operation. The reversible magneto-ionic control of various magnetic properties such as magnetization, coercivity or exchange bias has been demonstrated already,[2] but only a few studies target magneto-ionic control of MR to date.

We use iron oxide/iron as magneto-ionic material, in which electrochemical switching between ferromagnetic iron and weakly magnetic iron oxides can be exploited to reversibly change the magnetism. In a recent study of sputtered $FeO_x$/Fe thin films in stripe geometry we demonstrate the magneto-ionic manipulation of MR, achieving even a voltage-controlled sign change of MR.[3] As the magneto-ionic effect relies on reactive interfaces, stronger effects are expected for higher surface-to-volume ratios. In order to realize a 3D magneto-ionic system we took advantage of a Au aerogel as conductive template and deposited a nanometer thin iron layer by self-terminating electrodeposition.[4] TEM and EELS mapping showed a 5 to 10 nm iron oxide/iron layer surrounding the Au aerogel ligaments. On these 3D $FeO_x$/Fe/Au aerogel nanostructures, we demonstrate reversible voltage-controlled magneto-ionic control of MR at room temperature. At a magnetic field of 2 T a MR of -0.04 % (-0.01 %) for the reduction (oxidation) state is achieved, representing relative MR changes between reduced and oxidized state by 300 %. Future optimization of such magneto-ionically controlled 3D nanomaterials can advance the development in low-power sensors and 3D computational or information storage devices.

**Acknowledgements**

The authors acknowledge funding from the DFG (LE 2558 2-1). The authors thank Shashank Honnali for sputtering Au substrates (IFW Dresden), and Karl Hiekel and Alexander Eychmüller for providing the Au aerogel templates (both TU Dresden).

**References**

[1] P. Fischer, D. Sanz-Hernández, R. Streubel, A. Fernández-Pacheco, *APL Mater*. **8** (2020) 010701.

[2] M. Nichterwitz, S. Honnali, M. Kutuzau, S. Guo, J. Zehner, K. Nielsch, K. Leistner, *APL Mater.* **9** (2021) 030903.

[3] M. Nichterwitz, S. Honnali, J. Zehner, S. Schneider, D. Pohl, S. Schiemenz, S.T.B. Goennenwein, K. Nielsch, K. Leistner, *ACS Appl. Electron. Mater.* **2** (2020) 2543–2549.

[4] M. Nichterwitz, K. Duschek, J. Zehner, S. Oswald, R. Heller, K. Leistner, *Electrochimica Acta* **415** (2022) 140170.

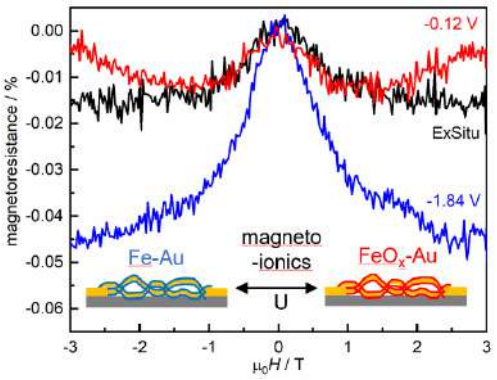



Figure: Magnetoresistance of $FeO_x$/Fe covered Au aerogel measured *in situ* during the polarization in 1 M LiOH solution at $E_{Red} = -1.84$ V (blue) and $E_{Ox} = -0.12$ V (red).

# Investigation of magnetization switching in iron oxide thin film/ silica nanoparticles heterostructures

*Mai Hussein Hamed[1,2]*, Asmaa Qdemat[1], Yifan Xu[1], Connie Bednarski-Meinke[1], Steffen Tober[1], Emmanuel Kentzinger[1], Oleg Petracic[1], and Thomas Brückel[1]*

[1] *Jülich Centre for Neutron Science JCNS-2 and Peter Grünberg Institute PGI-4, JARA-FIT, Forschungszentrum Jülich GmbH, 52425 Jülich, Germany.*

[2] *Faculty of Science, Helwan University, 11795 Cairo, Egypt.*

*m.hussein@fz-juelich.de

Magnetization switching in nanosized systems is interesting for developing novel spintronic devices. We grow $Fe_3O_4$ thin films on self-assembled $SiO_2$ nanospheres to investigate the effect of the nanospheres curvature onto the magnetization of the iron oxides ultrathin film. On the one hand, iron oxides have the ability to switch their magnetic and electrical states by varying their oxygen content. Controlling the oxygen content at the interfaces of complex heterostructures is key to controlling the switching phenomenon. On the other hand, by varying the film thicknesses down to the ultrathin regime, we manipulate the magnetization properties particularly at the interfaces[1-5]. Therefore, we aim to vary the magnetic anisotropy and thickness of thin films by the curvature of nanosphere patterned substrates achieving magnetization switching in magnetic thin films through the influence of the periodic curvature. We grow a series of $Fe_3O_4$ films with thicknesses 4, 8 and 40nm on 50nm and 200nm diameter $SiO_2$ nanospheres. The $SiO_2$ nanospheres are deposited on $SrTiO_3$ (STO) substrates using an improved variant of the drop-casting method[6]. On top of these nanospheres, we grow $Fe_3O_4$ thin films via pulsed laser deposition (PLD) that form either isolated or interconnected caps of $Fe_3O_4$ depending on the film thickness as well as on the nanosphere diameter, as observed by scanning electron microscope (SEM) and grazing-incidence small-angle X-ray scattering (GISAXS). The quality of the films on the nanospheres is investigated by atomic force microscopy (AFM) and X-ray diffraction (XRD). We investigate the magnetization reversal of these caps by MFM and SQUID by measuring magnetic hysteresis loops. The MH curves show a change at low fields for curved $Fe_3O_4/SiO_2$ in compare to the flat $Fe_3O_4$ films. The differences in MH curve must be related to the geometry of $Fe_3O_4$ thin films grown on nanosphere-patterned substrate compared to a flat substrate. We also study the effect of the oxygen content and the reduced film dimension onto the Verwey transition by measuring magnetization versus the temperature curves. In the next step, we will investigate the possible formation of a vortex state by grazing-incidence small-angle neutron scattering (GISANS), polarized neutron reflectometry (PNR) and X-ray magnetic circular dichroism (XMCD-PEEM). Planned is to detect the electric switching within the nanocaps using conductive AFM.

**References**

[1] M. H. Hamed, et.al., ACS Applied Materials & Interfaces, 11(7), 2019.
[2] M. H. Hamed, et.al., J. Mater. Chem.C, 8:1335–1343, 2020.
[3] M. H. Hamed, PhD thesis, Zugleich: Dissertation, Universität Duisburg, 2021, Jülich, 2021.
[4] M. H. Hamed, et.al., Applied Surface Science Advances, 6, 2021.
[5] M. Müller, P. Lömker, P. Rosenberger, M. H. Hamed, et.al., J. Vac. Sci. Technol., A 40, 2022.
[6] A. Qdemat, et.al., RSC Adv., 10, 2020.

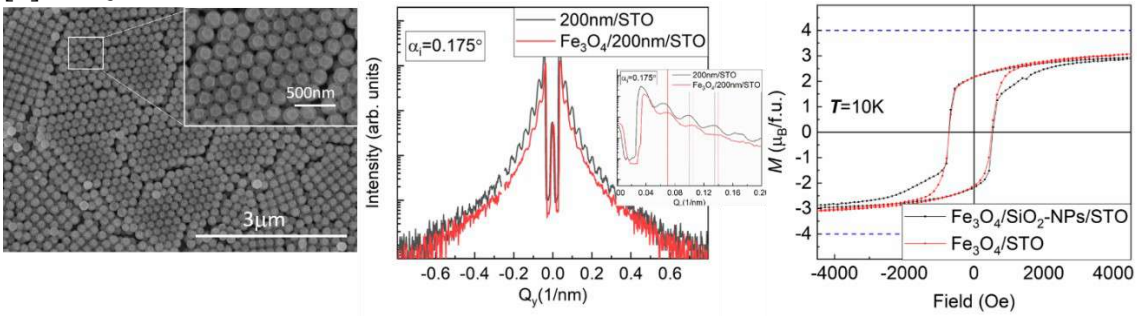



Figure: SEM (left), GISAXS pattern at an incident angle of 0.175° (middle) and the magnetic hysteresis loop at 10K (right) of 30nm $Fe_3O_4$ thin film on 200nm $SiO_2$ nanospheres on $SrTiO_3$ substrate.

# Influence of i-DMI on the spin-wave nonreciprocity of permalloy nanotubes.

*V. M. García-Suárez[1,2] and L. M. Alvarez-Prado[1,2*]*

[1]*Departamento de Fisica, Facultad de Ciencias, Universidad de Oviedo, C/ Federico García Lorca 18, Oviedo 33007, Spain*

[2]*Centro de Investigacion en Nanomateriales y Nanotecnologia (CINN), Av. De la Vega 4-6, El Entrego 33940, Spain*

*lmap@uniovi.es

3-D magnetic materials hold promising performances for computational capabilities and sensors devices [1]. The ability to tailor these magnetic properties is of paramount importance, e.g.: for reducing the footprint of the different components. We propose in this work, by means of micromagnetic simulations, a way to tune the output of the nanotube (one of the arquetypal stuctures proposed for 3D magnetism). Recent works have shown that a permalloy (Py) nanotube can emit spin waves (SW) from a local source in a non-reciprocal way [2]. If the Py nanotube is further covered by a metal possesing a high spin-orbit an interfacial-Dzyaloshinskii-Moriya exchange (i-DMI) is developed at the Py located near the nanotube's surface. This interacton bears its own source of non-reciprocity which becomes competitive/additive to the curvature-induced non-reciprocity of the nanotube (present even in the absence of i-DMI) [3]. We have adapted the free available simulator MuMax[3] [4] to include the i-DMI in line to [5] (dealing with nanodisks). Figure 1 shows that: i) the moving-forward SW's wavelenght (black) is always greater than the moving-backward SW's wavelength (red), ii) for a moderate i-DMI strength (1 mJ/m$^2$) the SW characterized by smaller wavelengths can be changed a 15% relative to its value under no i-DMI. At the same time, the other SW is hardly affected (~1.4%). It will be shown that, at high excitation frequencies, the former SWs belong to the SWs exchange-driven regime presenting long propagation lengths [6]. A detailed account of the methodology used and the changes observed in the nanotube's magnetic states, under different i–DMI strengths, will be given.

**Acknowledgements**

This study has received financial support from t he Spanish Ministry of Science and Innovation (Project No. PID2019-104604RB/AEI/10.13039/501100011033), the regional FICYT funding (Grant No. GRUPIN-AYUD/2021/51185) with the support of FEDER funds.

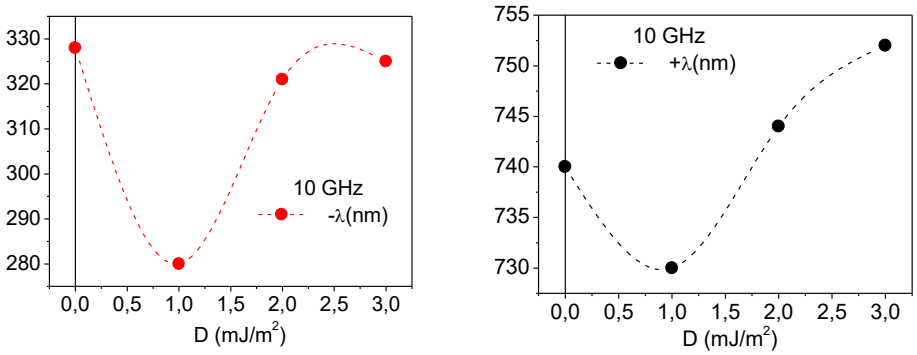



Figure 1: Change of SW wavelength with the i-DMI strength. The SW moves towards the rigth (red) and the left (black). A Py nanotube, is excited with a local 10GHz $B_{AC}$. The outer (inner) radius of the nanobe is 30nm (20 nm).

**References:**

[1] D. Raftrey et al., J. Mag. Mag. Mater. 563, 169899 (2022)

[2] L. Körber et al., Phys. Rev. B. 106, 014405 (2022) and references therein.

[3] O. Volkov et al. Sci. Rep. 8 (1), 866 (2018)

[4] B. Van de Wiele amd F. Montocello., J. Phys, D 47, 315002 (2014)

[5] J. Castell-Queralt et al., J. Mag. Mag. Mater. 549, 168972 (2022)

[6] C. Liu et al., Nat. Comm. 9, 738 (2018)

# Geometrical control of topological spin states in double-helix nanostructures

*N. Leo[1]*, J. Fullerton[2], J. Jurczyk[1], C. Donnelly[3], C. Abert[4,5], D. Raftrey[6], N. Mille[7], R. Belkhou[7], A. Sorrentino[8], L. Aballe[8], D. Sanz-Hernández[9], D. Suess[4,5], P. Fischer[6], A. Hierro-Rodriguez[10], A. Fernández-Pacheco[1]*

[1] *Instituto de Nanociencia y Materiales de Aragón, CSIC-University of Zaragoza, 50009, Zaragoza, Spain*

[2] *Glasgow University, G12 8QQ, Glasgow, United Kingdom*

[3] *Max Planck Institute for Chemical Physics of Solids, 01187, Dresden, Germany*

[4] *Faculty of Physics, University of Vienna, 1090, Vienna, Austria*

[5] *Research Platform MMM Mathematics–Magnetism–Materials, University of Vienna, 1090 Austria*

[6] *LBNL Materials Science Division, CA 94720, Berkeley, United States of America*

[7] *Synchrotron SOLEIL, 91190, Gif-sur-Yvette, France*

[8] *ALBA Synchrotron, 08290 Cerdanyola del Vallès, Spain*

[9] *Unité Mixte de Physique CNRS/Thales, 91120, Palaiseau Cedex, France*

[10] *University of Oviedo, 33007, Oviedo, Spain*

*naemi.leo@unizar.es

Magnetic chirality is a key ingredient in the formation of complex spin structures such as chiral domain walls, skyrmions and hopfions. The standard method to create magnetic chirality is to either employ particular noncentrosymmetric crystals [1] or multilayered hetero-structures without mirror symmetry [2].

Using 3D printing nanoscale methods [3], we recently demonstrated the control of magnetic chirality via 3D geometrical effects [4-6]. As shown in Fig. 1a, we can define nanoscale interfaces where two magnetic domains of opposite structural chirality are forced to meet at a nanometric volume, promoting the formation of complex 3D spin states [4].

After previously demonstrating spin-geometry coupling in these double-helix structures [5], here we will present recent results where we studied the magnetization reversal of the system under magnetic fields applied in different directions. Using synchrotron x-ray imaging and complementary micromagnetic simulations, we show how interfacing of two opposite geometrically-chiral domains and different magnetic field protocols it is possible to generate metastable complex chiral spin structures at will. This is a very attractive approach which opens a route for future experimental studies on their spin structure and unconventional dynamics [7,8].

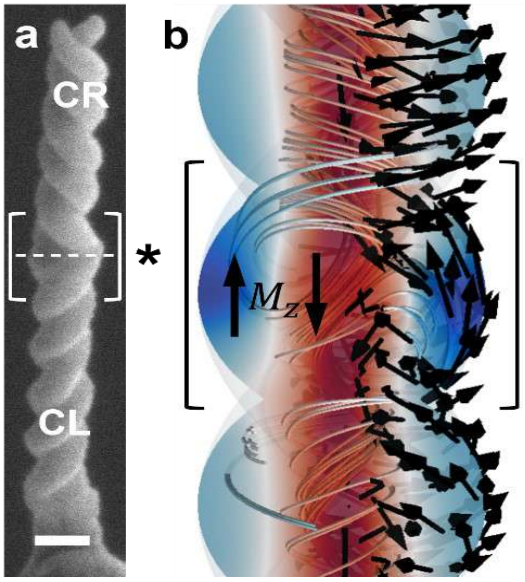



Figure 1: a SEM image of double-helix structure with a chirality interface marked by *. Scale bar measures 150 nm. b Due to coupling between magnetic and geometrical chirality, complex metastable spin textures, such as Skyrmion tubes, can be generated
by field protocols.

**Acknowledgements**

N.L., J.J. and A.F.P acknowledge support by the European Community under the Horizon 2020 Program, Contract No. 101001290 (3DNANOMAG). D.R. and P.F. acknowledge the support by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, Materials Sciences and Engineering Division, under Contract No. DE-AC02-05-CH11231 within the Non-Equilibrium Magnetism program (MSMAG).

## References

[1] S. Mühlbauer *et al.*, Science **323** 915 (2009).
[2] A. Fernández-Pacheco *et al.*, Nature Materials **18** 679-684 (2019).
[3] L. Skoric *et al.* Nano Lett. **20** 184 (2020).
[4] A. Fernández-Pacheco *et al.*, Nature Communications **8** 15756 (2017).
[5] D. Sanz-Hernández *et al.*, ACS Nano **14** 8084 (2020).
[6] C. Donnelly *et al.*, Nature Nanotechnology **17** 136–142 (2022).
[7] M.Y. Im *et al.*, Nature Communications **10** 593 (2019).
[8] L. Skoric *et al.*, ACS Nano **16** 8860–8868 (2022).

# Characterisation of 3D magnetic nanostructures using the dark-field magneto-optical Kerr effect

*Jakub Jurczyk[1]*, Naëmi Leo[1], Angelo Mottolese[1], Miguel Ángel Cascales-Sandoval[2], Luka Skoric[3], Dédalo Sanz-Hernández[4], Amalio Fernández-Pacheco[1]*

[1] *Instituto de Nanociencia y Materiales de Aragón, CSIC-University of Zaragoza, C/Pedro Cerbuna 12, 50009, Zaragoza, Spain*

[2]*SUPA, School of Physics and Astronomy, University of Glasgow, Glasgow, United Kingdom*

[3]*Cavendish Laboratory, University of Cambridge, JJ Thomson Avenue, CB3 0HE Cambridge, United Kingdom*

[4]*Unité Mixte de Physique, CNRS, Thales, Université Paris-Saclay, 91120 Palaiseau, France*

*jakub.jurczyk@unizar.es

The emerging field of 3D nanomagnetism demands for the development of novel techniques for the characterisation of 3D spin states and geometries at the nanoscale with high sensitivity and spatial resolution [1]. The complex geometry and magnetic configuration of 3D nanostructures makes probing and interpretation of these measurements particularly challenging in comparison with their 2D counterparts. One of the most frequently used methods of characterisation of planar (2D) magnetic systems, successfully applied to both nanostructures and thin films [2], is the magneto-optical Kerr effect (MOKE).

Recently, our group has extended the MOKE method to probe 3D nano-geometries via the application of dark-field MOKE [3]. In dark-field MOKE experiments, different planes of a 3D nano-geometry reflect light at different angles, allowing to detect and analyse the magneto-optical signal independently from each of them. This technique was first introduced to study the controlled injection of domain walls in 3D magnetic nanowire conduits [3], and details about the method and a thorough comparison with standard 2D MOKE have been recently covered [4].

In this contribution, we present application of dark-field MOKE for the characterisation of 3D magnetic nanostructures with different geometries and materials. Using 3D ramped nanowires as an example (see Fig. 1), we will discuss the sensitivity limits of the method, linking it with the minimum dimensions of a 3D nano-geometry that we can currently probe. Also, the experimental system that we have recently developed, specifically dedicated to the dark-field MOKE technique, will be presented. This setup, allowing to apply complex vector magnetic fields, provides high versatility to probe a range of 3D geometries.

**Acknowledgements**

This work was supported by the European Community under the Horizon 2020 Program, Contract No.101001290 (3DNANOMAG). The authors thank Pavel Strichovanec for help with sample preparation.

**References**

[1] A. Fernández-Pacheco, R. Streubel, O. Fruchart et al. *Nat Commun* **8**, (2017) 15756.

[2] D. A. Allwood et al. *J. Phys. D: Appl. Phys*. **36** (2003),2175

[3] D. Sanz-Hernández, R. F. Hamans, J-W Liao, A. Welbourne, R. Lavrijsen, and A. Fernández-Pacheco, *ACS Nano* **11** (2017), 11066-11073

[4] D. Sanz-Hernández, L. Skoric, M. Á. Cascales-Sandoval, A. Fernández-Pacheco, *J. Appl. Phys.* **133**, (2023) 043901

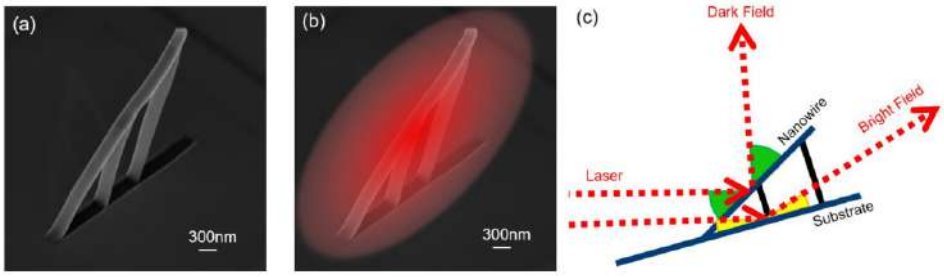



Figure a) 3D printed ramped nanowire made of Permalloy; b) illumination of the structure with a laser spot (not in scale) to perform dark-field MOKE c) schematics of dark-field MOKE detection for a nanowire grown at an angle on a substrate, with both planes leading to two distinct specular reflections (Taken from [4]).

# Skyrmions Confined in Plates as Building Blocks for Magnetic Metamaterials

*Carles Navau*, Leonardo González-Gómez, Nuria Del-Valle*

[1] *Departament de Física, Universitat Autònoma de Barcelona, 08192 Bellaterra, Barcelona, Catalonia, Spain*

*carles.navau@uab.cat

Magnetic metamaterials are studied as elements for shaping magnetic fields. They are composed by internal repetitive (magnetic) elements that results in effective (magnetic) properties than cannot be found in natural materials. Their potential applications include magnetic cloaking [1], transport of magnetic fields through predefined paths (magnetic hoses) [2], concentration and harvesting of magnetic energy at distance [3], or magnetic illusion by using effective negative-permeability systems [4], for example. All these applications have been proposed considering lineal soft or hard magnetic elements in combination with ideal perfect diamagnets (i. e, ideal superconductors). Here we present a different type of metamaterials composed by multiple ferromagnetic thin plates containing magnetization structure, i. e. a skyrmion. Since the skyrmions can be confined in plates with typical size side of few tens of nanometers, we can define and evaluate the effective permeability of such systems at scales of the order of the micrometer. We present how to evaluate the effective permeability in these systems and show results as a function of the internal structure of the ferromagnetic plates. We also discuss how these values can be tuned externally and discuss some of the potential applications one could obtain.

**Acknowledgements**

We acknowledge Spanish project PID2019-104670GB-I00 of Agencia Estatal de Investigación / Fondo Europeo de Desarrollo Regional for financial support.

**References**

[1] Experimental realization of a magnetic cloak. F Gömöry, M Solovyov, J Šouc, C Navau, J Prat-Camps, A Sanchez, Science 335 (6075), 1466-1468 (2012)

[2] Long-distance transfer and routing of static magnetic fields, C Navau, J Prat-Camps, O Romero-Isart, JI Cirac, A Sanchez, Physical Review Letters 112 (25), 253901 (2014)

[3] Magnetic energy harvesting and concentration at a distance by transformation optics. C Navau, J Prat-Camps, A Sanchez Physical Review Letters 109 (26), 263903 (2012)

[4] Magnetic illusion: transforming a magnetic object into another object by negative permeability R Mach-Batlle, A Parra, S Laut, N Del-Valle, C Navau, A Sanchez, Physical Review Applied 9 (3), 034007. (2018)

# Magnetization reversal processes of ferromagnetic nanowires modulated in geometry and composition

*J. Garcia[1]*, V. Vega[2], M. Mendez[1], J. A. Fernández-Roldán[1,3], S. González[1], Y. Alvarez[1], A. I. Jiménez[1], V. M. Prida[1]*

[1] *Depto. Física, Universidad de Oviedo, C/Federico García Lorca nº 18, 33007-Oviedo, Spain*
[2] *Lab. Membranas Nanoporosas (SCTs), Universidad de Oviedo, 33006-Oviedo, Spain*
[3] *Helmholtz-Zentrum Dresden-Rossendorf (HZDR), 01328-Dresden, Germany*
*garciafjavier@uniovi.es

The peculiar features and novel exotic phenomena displayed by three-dimensional (3D) network architectures made of arrays of nanowires (NWs) and nanotubes (NTs), lead to new possibilities for broad number of applications, such as nanodevices in nano-electronics, energy harvesting and storage systems, high-density data storage and magnetic sensors. Metallic NWs can be readily fabricated through several techniques, being one of the most versatile methods the electrochemical deposition by employing nanoporous alumina membranes as nanopatterned templates. Additionally, atomic layer deposition (ALD) is also a reliable physico-chemical deposition technique offering large area and optimal conformality of coatings, typically with metallic oxides, made on 3D nanostructured surfaces. Many works refer to the use of ALD technique for obtaining tailored 3D nanostructured networks based on template-assisted procedures, such as geometrically segmented arrays of NWs, or multilayered NTs and thin films [1]. Here we present the latest research on magnetic NWs forming building blocks of complex 3D nanostructures with modulated geometry and composition. To achieve further improvements in these research fields, a complete understanding of the magnetic properties exhibited by these 3D complex nanostructures is needed, including their magnetization reversal processes and control on the magnetic domain walls movement [2]. The magnetic properties of these 3D modulated nanostructures and their magnetostatic interactions are investigated by vibrating sample magnetometry (VSM) together with magneto-optical Kerr effect (MOKE) measurements, and first order reversal curves (FORC) analysis, allowing us to extract the individual magnetic properties of each single entity from the global magnetic behaviour of the array (see Figure 1). Furthermore, micromagnetic simulations performed by mumax[3] helped us to determine the main mechanisms that govern their magnetization reversal process. These results set an appealing strategy for the design of novel 3D magnetic storage and spintronic devices based on geometrical and compositional modulated nanowires that has not been envisaged up to know.

## Acknowledgements

Spanish MCINN under research grant No. PID2019-108075RB-C32/AEI/10.13039/501100011033 is gratefully acknowledged for financial support.

## References

[1] E. M. Palmero, M. Méndez, S. González, C. Bran, V. Vega, M. Vázquez, V. M. Prida, *Nano Research* **12** (2019) 1547-1553.
[2] J. García, V. Vega, A. Thomas, V.M. Prida, K. Nielsch, *Nanomaterials* **8** (2018) 548.

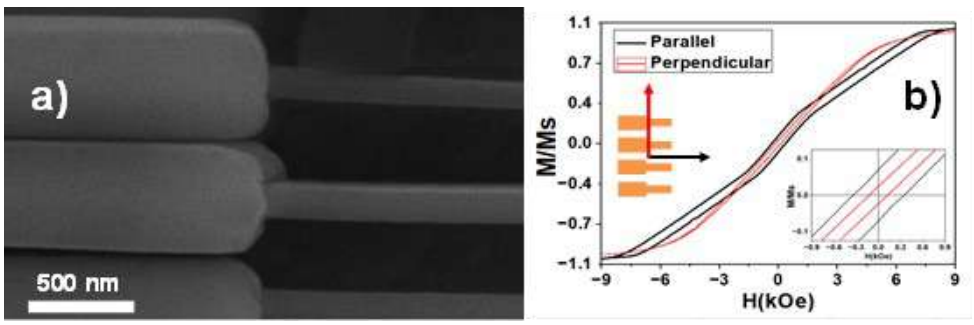



Figure 1: a) SEM image of diameter modulated FeNi nanowires synthesized by electrochemical deposition in tailored nanoporous alumina template. b) VSM room temperature hysteresis loops of ferromagnetic nanowires array shown in a), measured by applying the magnetic field along the parallel and perpendicular directions to the nanowires length axis.

# Surface Confinement of Bulk States on O/Fe

*J. Fujii[1]*, A.M. Finardi[1,2], J. Minar[3], I. Vobornik[1]*

[1] *CNR-IOM, AREA Science Park Basovizza, Trieste, Italy*

[2] *Dipartimento di Fisica, Università degli Studi di Milano, Via Celoria 16, Milano, Italy*

[3] *New Technologies-Research Centre, University of West Bohemia, Univerzitni 8, Pilsen, Czech Republic*

*jun.fujii@eletta.eu

We have investigated the electronic band structures for oxygen-adsorbed ferromagnetic Fe(001), as well as that for clean Fe(001) by means of ARPES, Spin-ARPES and DFT calculations. The Fermi surface of the oxygen adsorbed surface is significantly richer in structures compared with the calculated Fermi surface for clean Fe(001) by bulk band calculation (Fig. (a)). The spin polarization analysis by Spin-ARPES reveals that all the observed states are fully spin polarized also after the oxygen adsorption. This fact, as well as the symmetry analysis, enables us to identify the observed states. Surprisingly the Fermi surface of O/Fe(001) mimics the calculated bulk Fermi surface for clean Fe(001) at kz=0 (Figs. (b) and (c)). Interestingly many of oxygen-induced states are actually slightly observed before the oxidation as surface states of clean Fe(001). Oxygen adsorption emphasizes such surface states in photoemission, as if the oxygen adsorption confines the 3D bulk Fe electrons in the Surface, giving them 2D character. These findings are significant for full understanding of how 2D O2p electrons and 3D Fe3d electrons hybridize to form 2D O2p-Fe3d bands, and for the reason behind the enhancement of the surface magnetization of clean Fe and O/Fe is compared to the bulk.

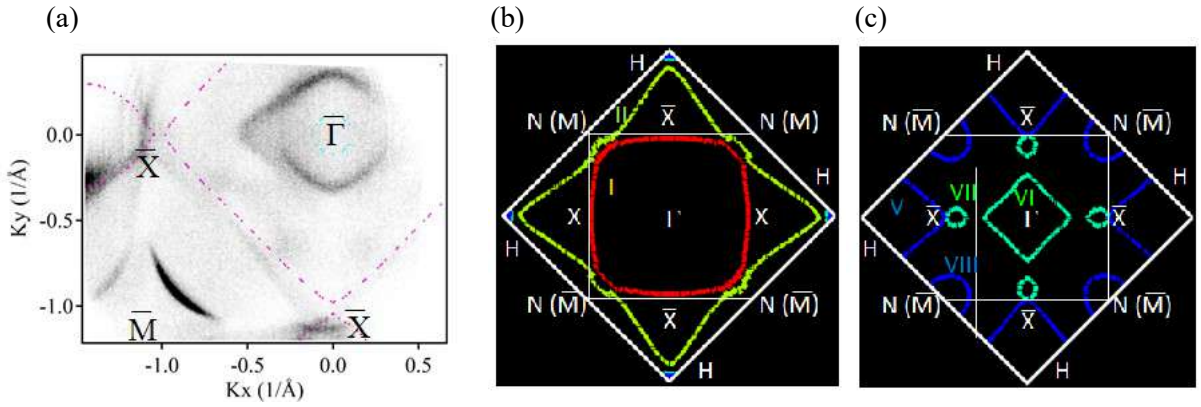



Figure: (a) Measured Fermi surface by ARPES for O/Fe(001) with hν = 50eV. Red (blue) dotted lines denote the calculated Fermi surface of majority (minority) spin bands for hν = 50eV (kz ~ 0.1 x 2π/a). Calculated Fermi surfaces for bulk Fe at kz=0 for majority spin bands (b) and minority spin bands (c).

# Complex free-space magnetic field textures induced by three-dimensional magnetic nanostructures

*Claire Donnelly[1,2], Aurelio Hierro-Rodríguez[3,4,5], Claas Abert[6], Katharina Witte[7,8], Luka Skoric[2], DédaloSanz-Hernández[2], Simone Finizio[7], Fanfan Meng[2], Stephen McVitie[3], Jörg Raabe[7], Dieter Suess[6], Russell Cowburn[2]& Amalio Fernández-Pacheco[9]*

[1] *Max Planck Institute for Chemical Physics of Solids, Dresden, Germany.*
[2]*Cavendish Laboratory, University of Cambridge, Cambridge, UK.*
[3]*University of Glasgow, Glasgow, UK.*
[4] *Universidad de Oviedo, Oviedo, Spain.*
[5] *CINN (CSIC–Universidad de Oviedo), El Entrego, Spain.*
[6] *University of Vienna, Vienna, Austria.*
[7]*Paul Scherrer Institute, Villigen, Switzerland.*
[8]*Berlin Partner für Wirtschaftund TechnologieGmbH, Berlin, Germany.*
[9]*CSIC-Universidad de Zaragoza, Zaragoza, Spain*
*claire.donnelly@cpfs.mpg.de

Three dimensional magnetic systems promise significant opportunities for applications, for example providing higher density devices [1] and new functionalities associated with complex topology and greater degrees of freedom [2]. In particular, when we pattern three dimensional nanogeometries, we gain control over the energies and behaviour of magnetic textures.

Here, I will present our recent work studying the behaviour of domain wall textures in such patterned three dimensional magnetic nanostructures [3]. By patterning complex geometries with nanoscale resolution [4], one can design structure combining curvature, chirality and high inter-structure coupling, leading to the realisation of highly coupled curvilinear systems that not only host new types of domain walls, but offer a route to controlling their behaviour.

These new capabilities open the door to complex three-dimensional magnetic structures, and their dynamic behaviour, of relevance not only for the fundamental understanding of three dimensional nanomagnetism, but also potentially for future spintronic devices.

**References**

[1] S. Parkin, M. Hayashi and L. Thomas, Science **320**, 190 (2008)
[2] A. Fernández-Pacheco et al., Nature Communications **8**, 15756 (2017).
[3] C. Donnelly and V. Scagnoli, J. Phys. D: Cond. Matt. **32**, 213001 (2020).
[4] C. Donnelly et al., Nature Nanotechnology 17, 136 (2022).
[5] L. Skoric et al., Nano Letters 20, 184 (2020).

# Magnetic Coupling in Electrodeposited Nanowires with Radial Modulation of Composition

*Lucia Gómez-Cruz[1]*, Claudia Fernández-González[2], Alba Guío[3], Alejandra Guedeja-Marrón[3], María Varela[3], Lucas Pérez[1,3], Sandra Ruiz-Gómez[2]*

[1] *IMDEA Nanociencia, Campus de Cantoblanco, 28049 Madrid,*
[2] *Max Planck Institute for Chemical Physics of Solids, 01187 Dresden, Germany*
[3] *Dpto. de Fisica de Materiales, Universidad Complutense de Madrid, 28040 Madrid, Spain*

* lugome11@ucm.es

The control of the magnetization processes along elongated structures is key for the development of novel magnetic devices [1]. In this sense, recent studies have shown that cylindrical nanowires have more complex, and exciting spin textures than their 2D counterparts, which make novel physics phenomena arise [2]. Therefore, these nanowires are suitable for building blocks of a new generation of magnetic devices. For this, the control of their magnetization processes is a must.

Electrodeposition is a versatile tool for the synthesis of cylindrical nanowires with controlled morphology and composition [3]. This technique allows the introduction of changes in composition during the growth procedure along the axial direction, which directly impacts the magnetic properties and magnetization dynamics of the magnetic nanostructures [4]. In a previous work, we show the possibility of modulating the composition along the radial direction in electrodeposited magnetic nanowires, producing multilayered structures in which the composition and thickness of each layer can be controlled independently [5]. This possibility increases the degrees of freedom to harness the magnetization during the growth.

In this work, we present a detailed study of the magnetic properties of such structures. In particular, we report on the growth and characterization of Co/Au/permalloy and Co/Au/Ni nanowires, in which the composition is varied along the radial direction. The hysteresis loops measured in arrays of nanowires, as well as micromagnetic simulations of individual nanowires, show how the coupling between layers can be controlled by carefully selecting the thickness of the Au interlayer, the thickness of the external ferromagnetic layer and the magnetic anisotropy of the external layer.

## Acknowledgements

This research was funded by MCINN/AEI/ 10.13039/501100011033 under Grants PID2020-117024GB-C43 and TED2021-130957B-C52.

## References

[1] S. S. P. Parkin, M. Hayashi, and L. Thomas, Science 320 (2008), 190
[2] M. Schöbitz et al. Phys. Rev. Lett 123 (2019) 217201
[3] S. Ruiz-Gómez, C. Fernández-González and L. Pérez. Micromachines 13, (2022) 1223
[4] S. Ruiz-Gómez et al. Nanoscale 12 (2020) 17880
[5] C. Fernández-González et al. Nanomaterials 12 (2022) 2565

# Tunable magnetic equilibrium configurations in dipolar helices

*Ot Garcés[1] and Òscar Iglesias[1]**

[1] *Dpt. Física de la Matèria Condensada and IN2UB, Universitat de Barcelona, Av. Diagonal 645, 08028, Barcelona, Spain*

*oscariglesias@ub.edu

The role that geometry and topology play in emergent order parameters has become an important topic in theoretical and experimental studies in different areas of physics [1]. A general theoretical framework to describe statics and dynamics of curved magnetic wires and surfaces has been developed recently, providing a starting point to study magnetization configurations in curvilinear structures [2]. New terms such as induced effective anisotropies and emergent Dzyaloshinskii-Moriya interactions appear as a consequence of curvature, which are responsible for magnetochiral effects, not present in the conventional cases. In this work, we have particularized the study to anti- and ferromagnetic (FM) helices, that are the simplest curves with constant κ and τ. By performing atomistic Monte Carlo simulations, we have validated the micromagnetic theoretical framework that predicts stable magnetization transitions between quasi-tangential (QT) to onion-like configurations [2]. Varying the curvature κ (or radius R) and torsion τ (or pitch p), we have obtained phase diagrams for the FM and AF cases for different kinds of magnetocrystalline anisotropies, extending them beyond the limits of application of the micromagnetic model. In a second part of the work, we will study the effects of curvature in systems where exchange determines the global magnetization direction, but the global magnetic order is dominated by dipolar interactions. This is the case of nanoparticle assemblies, colloidal magnets, magnetic beads or molecular clusters, that can be treated as magnetic macro-dipoles [3]. We will show that in dipolar helices a rich variety of equilibrium configurations can be reached by tuning the angle between consecutive dipoles. They include QT states, entwined head-to-tail magnetic helices that have a periodicity different from the generative helix and FM or AF ordered chains along the helix axis. Varying the radius or the pitch of the helix, abrupt transitions between states having zero and sizable net magnetization can be induced, which could be achieved experimentally by applying stress to the helix ends and be used as magneto-mechanic sensors.

## Acknowledgements

Work supported by Spanish MINECO (PGC2018-097789-B-I00, PID2019-109514RJ-I00), Catalan DURSI (2017SGR0598) and EU FEDER funds (Una manera de hacer Europa) also CSUC for supercomputer facilities.

## References

[1] C. Donnelly *et al. Nature Nanotechnol.* **17** (2022) 136.
[2] D. D. Sheka *et al. Small* **18** (2022) 2105219; Phys. Rev. B **92** (2015) 054417.
[3] I. Stankovic, *et al. Nanoscale* **11** (2019) 2521; A. Edström, *et al. Phys. Rev Lett.* **128** (2022) 177202.

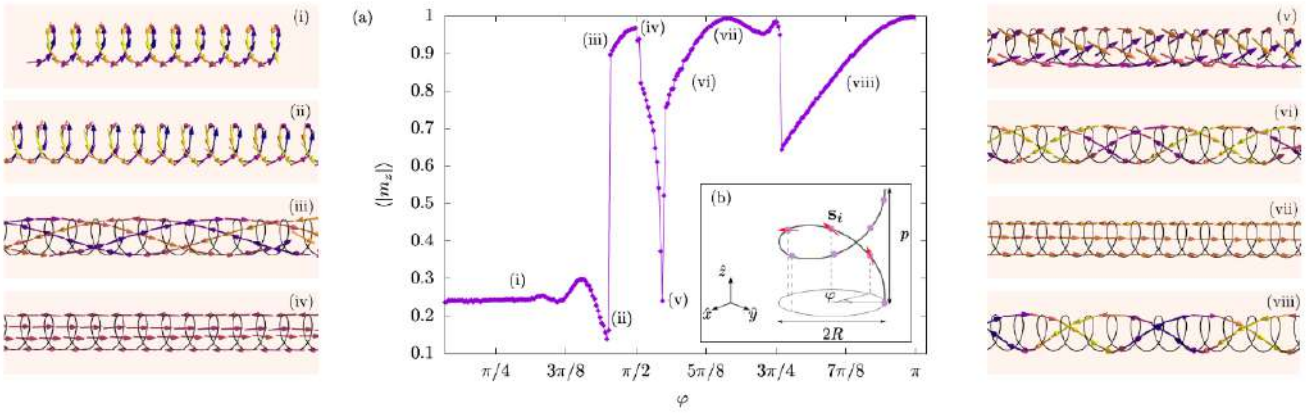



Figure: Ground states of a dipolar helix (R=1, p=π/2) and their staggered magnetization along the helix axis.

# Transition between magnetic vortex and double pole states in Py/Ru/Py diamond-shaped trilayers

*Ana Parente[1]*, H. Navarro[2], N.M. Vargas[2], P. Lapa[2], A.C. Basaran[2], E.M. González[1,3], C. Redondo[4], R. Morales[4,5], A. Munoz-Noval[1,3], I.K. Schuller[2] and J.L. Vicent[1,3]*

[1] *Department of Material Physics, Facultad de Fisica UCM, 28040 Madrid, Spain*
[2] *Department of Physics and Center for Advanced Nanoscience, University of Califronia San Diego, California 92093, United States*
[3] *IMDEA Nanociencia, 28049 Madrid, Spain*
[4] *Department of Physical Chemistry, UPV/EHU & BCMaterials, 48940 Leioa, Spain*
[5] *IKERBASQUE. Basque Foundation for Science, 48009 Bilbao, Spain;*
*aparente@ucm.es

Controlling the magnetic ground states at the nanoscale is a long-standing basic research problem and an important issue in magnetic storage technologies. [1] Here, a nanostructured material that exhibits very unusual hysteresis loops due to a transition between a magnetic vortex and double pole states is designed. [2]

Arrays of 700 nm diamond-shaped nanodots consisting of Py(30 nm)/Ru($t_{Ru}$)/Py(30 nm) (Py, permalloy ($Ni_{80}Fe_{20}$)) trilayers were fabricated by interference lithography and e-beam evaporation. In this work, it is shown that varying the Ru interlayer spacer thickness ($t_{Ru}$) the interaction between the Py layers can be modified. [2] We explain that this interaction is mainly mediated by two mechanisms: magnetostatic interaction that favors antiparallel (antiferromagnetic, AFM) alignment of the Py layers and interlayer exchange interaction that oscillates between ferromagnetic (FM) and AFM couplings.

For a certain range of Ru thicknesses, FM coupling dominates and magnetic vortices arises in the upper and lower Py layers. For Ru thicknesses at which AFM coupling dominates, the magnetic state in remanence is a double pole structure. The results showed that the interlayer exchange coupling interaction remains finite even at 4 nm Ru thickness. The magnetic states in remanence, observed by Magnetic Force Microscopy (MFM), are in good agreement with corresponding hysteresis loops obtained by Magneto-Optic Kerr Effect (MOKE) and micromagnetic simulations (Mumax[3]).

## References:

[1] E. Y. Vedmedenko *et al. Journal of Physics D: Applied Physics* **53**, 453001 (2020)
[2] A. Parente *et al. ACS Appl. Mater Interfaces,* **14,** 54961–54968 (2022)

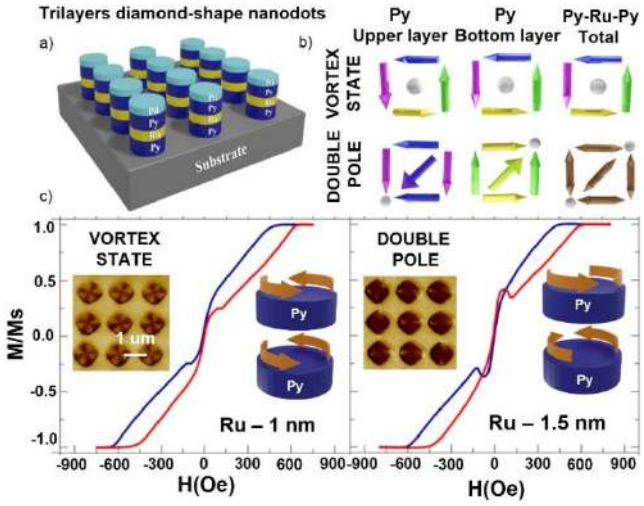



Figure: a) Schematics of the trilayer structure and the dot array. b) Micromagnetic simulations scheme of two magnetic configurations at zero external field. c) Longitudinal MOKE hysteresis loops with magnetic field parallel to the main axis of the square array in the substrate plane for samples with different Ru thickness. Insets: MFM images of the array in both states: vortex and double pole. The cartoon in the right of the loop schematizes the relative magnetization between both layers in the vortex and double pole states.

# Curvature-induced Tilt and pinning in CrOx/Co/Pt corrugated strips

*Jose A. Fernandez-Roldan*[a], *S. Shakeel*[a], *M. Quintana*[a,b], *O. Volkov*[a], *A. Polypovskyi*[a], *E. S. Oliveros-Mata*[a], *F. Kronast*[c], *M.-A. Mawass*[c], *C. Abert*[d,e], *D. Suess*[d,e], *D. Erb*[a] *and D. Makarov*[a]

[a] *Helmholtz-Zentrum Dresden-Rossendorf e. V., 01328 Dresden, Germany*
[b] *CIC nanoGUNE BRTA, E-20018 Donostia—San Sebastián, Spain*
[c] *Helmholtz-Zentrum Berlin für Materialien und Energie, 12489 Berlin, Germany*
[d] *Faculty of Physics, University of Vienna, Boltzmanngasse 5, 1090 Vienna, Austria*
[e] *Research Platform MMM Mathematics-Magnetism-Materials, University of Vienna, Oskar-Morgenstern-Platz 1, 1090 Vienna, Austria*
*j.fernandez-roldan@hzdr.de

In recent years, curvilinear nanomagnetism is attracting attention for the broad range of effects emerging in curved geometries that are appealing for the innovative developments in stretchable and magnetoelectric devices, microrobots, sensors, flexible magnetic memories and nanoelectronics [1-5].

These phenomena encompass a vast range of exchange- and Dzyaloshinskii-Moriya (DMI)- induced interactions that typically result in topological magnetization patterning in shells, chiral symmetry breaking, and pinning of domain walls [1-5]. Less attention has been paid though to the role of the curvilinear effects in the magnetization dynamics of domain walls in curved geometries [4]. From application perspectives, spin-orbit torques are appealing as an alternative way to achieve the manipulation of magnetic domain walls and magnetization [8] with the breakthrough of lower power consumption. Recent developments in ultra-thin planar asymmetric multilayered strips describe a method to extract DMI and damping estimations from the dynamical tilt of domain walls from static measurements [9]. Following a similar approach, here we provide first results in single 100 nm-wide thin periodically corrugated strips of CrOx/Co/Pt with thickness of 2 nm and average curvature of 0.06 nm-1, tailored for an enhanced exchange-induced DMI. The orientation of the corrugation is tuned from the parallel to the perpendicular direction of the strip axis in different strips.

Our results indicate that curvature plays a crucial role in the pinning and tilting of domain walls through DMI-induced and exchange-induced effects. In particular, DMI-induced anisotropy leads the pinning mechanism, while its combination with exchange-induced effects enhances the domain wall tilt. This opens a perspective for quantification and design of curvature-induced effects with application prospects in current challenges of spin-based nanoelectronics [10].

**References**

[1] Denys Makarov, et al., Advanced Materials 34, 3 (2022)

[2] Denis D Sheka, Oleksandr V Pylypovskyi et al., Small 18, 12 (2022)

[3] D. Sander et al., J. Phys. D: Appl. Phys. 50, 363001 (2017).

[4] E. Y. Vedmedenko, et al., J. Phys. D. Appl. Phys. 53, 453001 (2020).

[5] D. Makarov et al., Applied Physics Reviews 3, 011101 (2016).

[6] E. Berganza et al., Sci Rep 12, 3426 (2022)

[7] J.A. Fernandez-Roldan et al., Sci Rep 9, 5130 (2019).

[8] O. V. Pylypovskyi et al., Scientific Reports 6, 23316 (2016).

[9] O. M. Volkov et al., Phys. Rev. Applied 15, 034038 (2021)

[10] B. Dieny, et al., Nat Electron 3, 446 (2020).

# SYMPOSIUM 15.
## MAGNONICS AND MAGNETOPLASMONICS, ULTRAFAST MAGNETIZATION DYNAMICS.
## S15 INVITED ORAL PRESENTATIONS

# Magnetoplasmonics in Confined Geometries: Current Challenges and Future Opportunities

*Nicolò Maccaferri*[1,2]*

[1] *Department of Physics, Umeå University, Linneaus väg 24, 901 87, Umeå, Sweden*
[2] *Department of Physics and Materials Science, University of Luxembourg, 162a avenue de la Faïencerie, L-1511 Luxembourg, Luxembourg*
*nicolo.maccaferri@umu.se

Plasmonics represents a unique approach to confine and enhance electromagnetic radiation well below the diffraction limit, bringing a huge potential for novel applications, for instance in energy harvesting, optoelectronics, and nanoscale biochemistry. To achieve novel functionalities, the combination of plasmonic properties with other material functions has become increasingly attractive. In this talk, I will review the current state of the art, challenges, and future opportunities within the field of magnetoplasmonics in confined geometries, an emerging area aiming to merge magnetism and plasmonics to either control localized plasmons, confined electromagnetic-induced collective electronic excitations, using magnetic properties, or vice versa [1,2]. I will begin by highlighting the cornerstones of the history and principles of this research field, by focusing on the the use of localized surface plasmons to enhance magneto-optical effects in metallic systems, in the framework of active flat-optics metamaterials for light polarization control as well as for biochemical sensing. I will then try to provide a roadmap of its future development by showcasing raising research directions in hybrid magnetoplasmonic systems to overcome radiation losses and novel materials for magnetoplasmonics, such as transparent conductive oxides and hyperbolic metamaterials [3,4]. Finally, I will provide an overview of recent developments in plasmon-driven magnetization dynamics, nanoscale opto-magnetism and acousto-magnetoplasmonics. I will conclude by giving an outlook on the future of the field, showcasing new possible directions to achieve a full control of magneto-optical effects and their enhancement by using nanoscale materials, as well as drive magnetic phenomena with plasmons at the atomic and sub-femtosecond timescales.

**Acknowledgements**

We acknowledge support from the Swedish Research Council (grant n. 2021-05784), Kempestiftelserna (grant n. JCK-3122), the European Innovation Council (grant n. 101046920 ‘iSenseDNA’), the Luxembourg National Research Fund (grant n. C19/MS/13624497 ‘ULTRON’) and the European Commission (grant n. 964363 ‘ProID’).

**References**

[1] N. Maccaferri, *J. Opt. Soc. Am. B* **36** (2019) E112.
[2] N. Maccaferri, I. Zubritskaya, I. Razdolski, I.-A. Chioar, V. Belotelov, V. Kapaklis, P. M. Oppeneer, A. Dmitriev. *J. Appl. Phys.* **127** (2020) 080903.
[3] J. Kuttruff, A. Gabbani, G. Petrucci, Y. Zhao, M. Iarossi, E. Pedrueza-Villalmanzo, A. Dmitriev, A. Parracino, G. Strangi, F. De Angelis, D. Brida, F. Pineider, N. Maccaferri, *Phys. Rev. Lett.* **127** (2021) 217402.
[4] N. Maccaferri, A. Gabbani, F. Pineider, T. Kaihara, T. Tapani, P. Vavassori, *Appl. Phys. Lett.* DOI: 10.1063/5.0136941 (2023).

# Magnetization Dynamics in Transition Metal Films with Low Perpendicular Magnetic Anisotropy

*Boris Vodungbo[1]*, Emmanuelle Jal[1], Marcel Hennes[2], Renaud Delaunay[1], Julian Milano[3], Juliette Dubois[1,4], Guillaume Lambert[4]*

[1] *Sorbonne Universite, CNRS, Laboratoire de Chimie Physique—Matière et Rayonnement, LCPMR, 75005 Paris, France*

[2] *Sorbonne Universite, CNRS, Institut des NanoSciences de Paris, INSP, 75005 Paris*

[3] *Instituto de Nanociencia y Nanotecnología, INN (CNEA-CONICET). Centro Atómico Bariloche, R4802AGP, Río Negro, Argentina.*

[4] *Laboratoire d'Optique Appliquée, ENSTA Paris, CNRS, École Polytechnique, Institut Polytechnique de Paris, 828 Bd des Marechaux, 91702 Palaiseau, France*

*boris.vodungbo@sorbonne-universite.fr

Above a critical thickness, in the order of tens to hundreds of nanometers, transition metal thin films with low perpendicular magnetic anisotropy (PMA) present a stripe domains structure [1]. Compared to high PMA thin films such as Co/Pt multilayers, in which such a stripe domains structure can also be observed albeit with a lot smaller critical thickness (few nanometers), the domains are not fully magnetized out of plane and the transition between adjacent domains is not very steep. This gives rise to more complex magnetic structures, with magnetic caps at the domains extremities for example, which could be exploited for spintronics applications [2]. For these potential applications, it is fundamental to be able to control the magnetic structure. One way to realize this control is to use optical excitation of the system [3]. Moreover, using the shortest laser pulses possible, in the femtosecond range, opens up the possibility to realize ultrafast devices.

Here, we will report on the magnetization dynamics induced by an near infrared (NIR) femtosecond laser pulse in three types for Ni based thin films: pure Ni, $Ni_{1-x}Fe_x$ and $Ni_{1-x}Co_x$. To probe the nanometric magnetic structure of these systems we used high order harmonics (HHG) of NIR femtosecond laser in resonance with the Fe, Co and Ni $M_{2,3}$ absorption edges between 50 and 70 eV (about 20 nm). Indeed, resolving the magnetic structure requires very short wavelengths which cannot be reach in the NIR or visible range. At resonant energies, the magnetic domains structure acts as a diffraction grating and recording the diffracted orders on a CCD camera allow us to retrieve time-resolved informations on this structure. We have pioneered this type of experiment for the study of ultrafast magnetization dynamics in high PMA systems by following the demagnetization of Co/Pt multilayers in a transmission geometry [4]. The films we want to study here are much to thick, between 100 and 300 nm, for conducting a transmission experiment at the $M_{2,3}$ absorption edges. Fortunately, we have recently shown that it is possible to conduct similar experiments in reflection geometry and to study films of arbitrary thickness [5]. This has the advantage to enhance the sensitivity to surface structures which may arise in low PMA films.

## Acknowledgements

We are grateful for financial support received from the CNRS-MOMENTUM and Émergence–Sorbonne Université (MADAM grant) programs.

## References

[1] D. Metrio, E. Duardo, O. Legario, *Nat. Phys.* **15** (2019) 735. (Times New Roman 10 points, left-justified)N. Saito, H. Fujiwara, Y. Sugita, *Journal of the Physical Society of Japan*, **19** (1964) 1116-1125.

[2] L.-C. Garnier, M. Marangolo, M. Eddrief, D. Bisero, S. Fin, F. Casoli, M. Gloria Pini, A. Rettori, S. Tacchi, *J. Phys. Mater.* **3** (2020 ) 024001.

[3] C. D. Stanciu, F. Hansteen, A. V. Kimel, A. Tsukamoto, A. Itoh, A. Kirilyuk, Th. Rasing, *Phys. Rev. Lett.* **98** (2007) 207401

[4] B. Vodungbo, J. Gautier, G. Lambert, A. Barszczak Sardinha, M. Lozano, S. Sebban, M. Ducousso, W. Boutu, K. Li, B. Tudu, M. Tortarolo, R. Hawaldar, R. Delaunay, V. López-Flores, J. Arabski, C. Boeglin, H. Merdji, P. Zeitoun, J. Lüning, *Nature Commun.* **3** (2012) 999

[5] M. Hennes, G. Lambert, V. Chardonnet, R. Delaunay, G. S. Chiuzbăian, E. Jal, B. Vodungbo, *Appl. Phys. Lett.* **120** (2022) 072408.′

# Picosecond Spin Seebeck effect in ferromagnets and antiferromagnets

*Chiara Ciccarelli*

[1] *Department of Physics, Univesrity of Cambridge*

The longitudinal spin Seebeck effect describes the transfer of a spin current from a magnetic insulator driven by a temperature gradient. Here we present our recent studies on the longitudinal spin Seebeck effect on the picosecond timescale in both ferromagnets and antiferromagnets using THz emission spectroscopy [1]. As a function of temperature, we observed a different temperature dependence compared to DC electrical studies carried out in the same temperature range. By comparing different antiferromagnets belonging to the same family of fluoride perovskites we are able to correlate the spin transfer efficiency with the bands of the magnon spectrum.

[1] Farhan Nur Kholid, Dominik Hamara, Ahmad Faisal Bin Hamdan, Guillermo Nava Antonio, Richard Bowen, Dorothée Petit, Russell Cowburn, Roman V. Pisarev, Davide Bossini, Joseph Barker & Chiara Ciccarelli, *Nature Communications* **14** (2023) 538.

# Plasmonic Control of Spin Waves

*Sebastiaan van Dijken*

*Department of Applied Physics, Aalto University School of Science, Espoo, Finland*

sebastiaan.van.dijken@aalto.fi

Optical control of propagating spin waves in yttrium iron garnet (YIG) films enables active reprogramming of the magnonic band structure [1] and the redirection of spin-wave transport [2]. For practical devices, real-time manipulation of spin waves should be fast and confined to small areas. Here, we report optical control of propagating spin waves on the nanosecond time scale in a YIG film with plasmonic nanodisk arrays patterned on top. The YIG film is 250 nm thick and the Au nanodisks are 50 nm thick and have a diameter of 180 nm. By ordering the Au nanodisks in a square array, we intentionally utilize the excitation of collective surface lattice resonances (SLRs) to control the absorption of incident laser light. We maximize the absorption of 915 nm laser pulses by tuning the period of the nanodisk array [3]. Under optimized conditions, we demonstrate that the transmission signal between two parallel microwave antennas in broadband spin-wave spectroscopy measurements can be turned off by a 400 ns laser pulse, while a partial reduction in the spin-wave signal is attained for shorter pulses. The fast optical control of propagating spin waves is explained by efficient thermoplasmonic heating of the YIG film under SLR excitation, as confirmed by a measured reduction of the saturation magnetization and finite-difference time-domain simulations. Various prospects of local optical control of spin waves through plasmon excitation will be discussed.

**Acknowledgements**

This work is supported by the Jane and Aatos Erkko Foundation and Technology Industries of Finland through the Future Makers program

**References**

[1] M. Vogel, A. V. Chumak, E. H. Waller et al., *Nat. Phys.* **11**, (2015) 487.
[2] M. Vogel, R. Aßmann, P. Pirro et al., *Sci. Rep.* **8**, (2018) 11099.
[3] V. G. Kravets, A. V. Kabashin, W. L. Barnes, A. N. Grigorenko, *Chem. Rev.* **118**, (2018) 5912.

# SYMPOSIUM 15.
## MAGNONICS AND MAGNETOPLASMONICS, ULTRAFAST MAGNETIZATION DYNAMICS. S15 ORAL PRESENTATIONS

# Thermoplasmonic Activation of Nanomagnetic Logic Gates

*Matteo Menniti*[1,2]*, *Paolo Vavassori*[1,3]

[1] *CIC nanoGUNE BRTA, Tolosa Hiribidea 76 , 20018 Donostia/San Sebastian, Spain*
[2] *University of the Basque Country, Barrio Sarriena, 48940 Leioa, Spain*
[3] *IKERBASQUE, Basque Foundation for Science, 48009 Bilbao, Spain*
* m.menniti@nanogune.eu

Networks of interacting, bi-stable, single domain nanomagnets have been used to study collective phenomena in frustrated systems like in Artificial Spin Ices (ASIs), which are of interested not only for their ability to shed light on fundamental physics but also for their potential applications in magnonic devices, future energy efficient logic devices, and novel computing schemes [1]. The nanomagnets in these systems have the ability to spontaneously change their direction of magnetization in order to reduce their magnetostatic energy and, increasing the temperature allows to significantly accelerate this relaxation [2]. The heating of ASIs has typically been made by global heating of the entire network (*i.e.* sample) using a hotplate; however this method, apart from being slow and energy inefficient, is also lacking selectivity in the heating. By means of thermoplasmonic laser heating, however, these limitations can be circumvented and sublattices or even single elements within the network can be heated to temperatures of 100s of K at the sub-nanosecond time scale [3-4].

In these networks of interacting nanomagnets bits of digital information (1's and 0's) can be enconded in the direction of their individual magnetization. By defining input and output nanomagnets these networks can be used for creating nanomagnetic logic (NML) devices where the computation is performed by the magnetostatic interaction and the result expressed by the more favorable (relaxed) direction of magnetization of the output nanomagnet [5].

To give one prof of concept for how the nanomagnetic and thermoplasmonic properties of hybrid noble metal/ferromagnetic nanostructures can be used to design functional metamaterials we proposed a reconfigurable Boolean NML gate [6]. Here we will show the experimental implementation of such gate from the design process and nanofabrication to the magnetical and optical characterization as well as visualization of the NML by means of force microscopy.

**Acknowledgements**

Spanish Ministry of Science, Innovation and Universities for the financial support through predoctoral grant

**References**

[1] Skjærvø, S.H., Marrows, C.H., Stamps, R.L. et al. *Nat Rev Phys* **2** (2020), *p* 13–28.
[2] J. M. Porro, A. Bedoya-Pinto, A. Berger and P. Vavassori, *New J. Phys.* **15** (2013) 055012.
[3] M. Pancaldi, N. Leo, and P. Vavassori, *Nanoscale* **11** (2019), *p* 7656-7666.
[4] G. Baffou, R. Quidant, *Laser Phot. Rev*. **7** (2013), *p* 171-187.
[5] A. Imre, G. Csaba, L. Ji, A. Orlov, G. H. Bernstein, W. Porod, *Science* **311** (2006), *p* 205-208.
[6] P. Gypens, N. Leo, M. Menniti, P. Vavassori, J. Leliaert, *Phys. Rev. Applied* **18** (2022) 024014.

# Magnon-Plasmon Hybridization Mediated by Spin-Orbit Interaction

*Anna Dyrdał[1]*, Alireza Qaiumzadeh[2], Arne Brataas[2], Józef Barnaś[2,3]*

[1] *Department of Mesoscopic Physics, Faculty of Physics, Adam Mickiewicz University in Poznań, ul. Uniwersytetu Poznańskiego 2, 61-614 Poznań, Poland*

[2]*Center for Quantum Spintronics, Department of Physics, Norwegian University of Science and Technology, NO-7491 Trondheim, Norway*

[3]*Institute of Molecular Physics, Polish Academy of Sciences, ul. M. Smoluchowskiego 17, 60-179 Poznań, Poland*

*corresponding author email address

We will present a mechanism of magnon-plasmon hybridization in ferromagnetic (FM) and anti-ferromagnetic (AFM) systems [1]. This mechanism is based on the electronic spin-orbital coupling (SOC) and s–d(f) exchange interaction. The key point of the mechanism is the non-equilibrium spin density induced by the electric field associated with plasmon oscillations (via the Edelstein effect). This plasmon-induced spin density becomes coupled to the localized spins (and thus effectively to magnons) via the s–d exchange interaction. The strength of the magnon-plasmon coupling depends on the magneto-electric susceptibility of the system and on the wavevector at which the bare magnon and plasmon dispersion curves cross each other. The magnon-plasmon interaction leads then to the level repulsion (or level anticrossing).

In the case of antiferromagnets, the degeneracy of the two magnon modes is lifted in the presence of an external magnetic field, so one finds then two separate hybrid modes.

The proposed mechanism is applicable to 3D as well as 2D systems. We will focus mainly on 2D materials, where the magnon-plasmon hybridization can be easily achieved owing to low frequency of plasmons (the plasmon energy vanishes in the limit of zero wavevectors). The proposed hybridization opens a link between magnonics and plasmonics.

**Acknowledgements**

This work has been supported by the Norwegian Financial Mechanism 2014- 2021 under the Polish -- Norwegian Research Project NCN GRIEG “2Dtronics” no. 2019/34/H/ST3/00515.

**References**

[1] A. Dyrdał, A. Qaiumzadeh, A. Brataas, and J. Barnaś, “Magnon-plasmon hybridization mediated by spin-orbit interaction in magnetic materials”, arXiv:2212.06105

# Accessing sub-100nm magnons in reciprocal space by resonant small-angle x-ray scattering

*Steffen Wittrock[1,2]*, Christopher Klose[1], Korbinian Baumgaertl[3], Andrea Mucchietto[3], Michael Schneider[1], Josefin Fuchs[1], Victor Deinhart[2], Tamer Karaman[2], Niklas Schneider[1], Dirk Grundler[3], Stefan Eisebitt[1], Bastian Pfau[1], Daniel Schick[1]*

[1] *Max Born Institute for Nonlinear Optics & Short Pulse Spectroscopy, Max-Born-Str. 2A, 12489 Berlin, Germany*

[2] *Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, Hahn-Meitner-Platz 1, 14109 Berlin, Germany*

[3] *Laboratory of Nanoscale Magnetic Materials and Magnonics, Institute of Materials (IMX), École Polytechnique Fédérale de Lausanne (EPFL), 1015 Lausanne, Switzerland*

*steffen.wittrock@helmholtz-berlin.de

Miniaturization of magnonics into the exchange-dominated regime is essential in the development of future magnonic devices beyond CMOS signal processing. However, the generation and detection of spin waves of small wavelength $\lambda$ remain a challenge. Here, we demonstrate the investigation of sub-100nm-wavelength magnons by small angle X-ray scattering (SAXS), a technique that has been mainly explored in the context of static spin textures so far. Our approach closes the gap between established optical methods with resolution limited to the sub-µm range and inelastic scattering exhibiting a resolution only below the 10 nm range. Furthermore, compared to real-space methods, e.g. STXM, essential characteristics of the magnon dynamics, such as its dispersion, can be directly accessed in reciprocal space. Resonant X-ray scattering from magnons is very sensitive and gives further access to the types of excited modes, their coupling and scattering mechanisms, as well as nonlinearities. We demonstrate all these effects to appear in 100nm thick ferrimagnetic yttrium iron garnet (YIG) in which we excite propagating small-wavelength spin waves by the exploitation of ferromagnetic grating couplers (GCs) in the GHz frequency regime. The method is not limited to the magnon frequency and has the potential of establishing as a novel standard tool for the study of small-wavelength magnonics. The work is supported by SNSF via grant 197360.

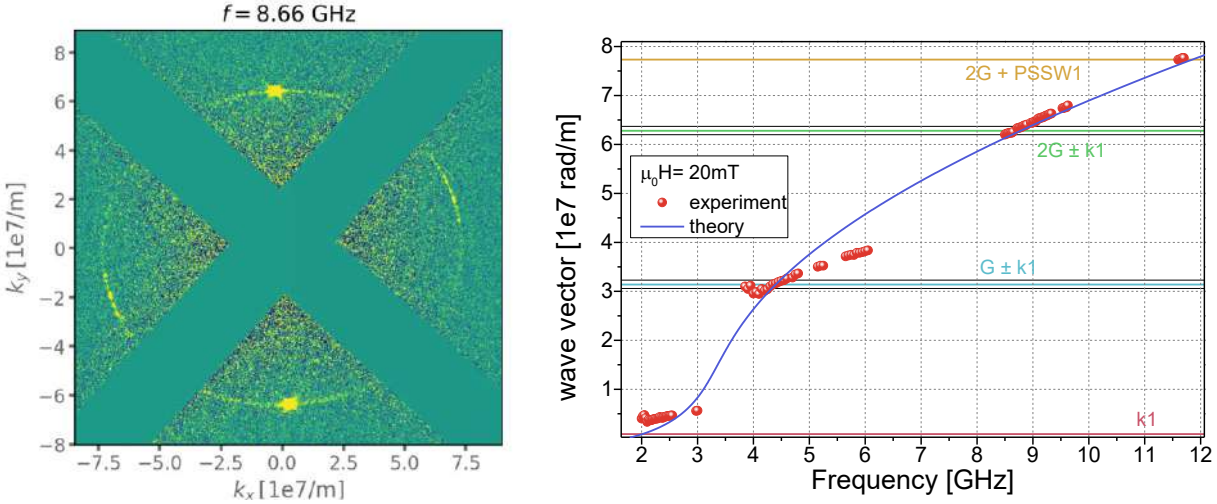



Figure: Scattering signal of a $\lambda \sim 100\,nm$ wavelength magnon (left). The bright upper/lower peaks correspond to the main propagation direction of the corresponding Damon-Eshbach (DE) spin wave mode. The azimuthal distribution of the scattering signal reveals that spin wave scattering into other modes takes place. The mode corresponding to a perpendicular propagation direction with respect to the main excited mode (i.e. backward volume mode) corresponds to a wavelength of $\sim 80\,nm$, resulting in a slightly elliptical shape of the scattering ring. Right: Directly accessed dispersion relation in the DE configuration. Marked k-vectors reflect the geometry of the grating coupler (GC), with k1 the most efficiently excited vector of the coplanar waveguide, G the reciprocal lattice vector of the GC, and PSSW the perpendicular standing spin wave mode. The data reveal that the exchange dominated modes at larger wave vector agree well with the theoretical curve, while the modes with dominant dipolar contribution are subject to a pulling or conversion effect.

# Impact of Interactions on Topological Magnonic Transport

*Konstantinos Sourounis*[1]*, *Aurelien Manchon*[1]

[1] *Aix-Marseille Universite, CNRS, CINaM, Marseille, France*

*konstantinos.sourounis@univ-amu.fr

The topological transport of magnons is a popular alternative to the electronic transport for novel spintronic applications due to the absence of Joule heating and the large range of frequencies in these systems. The magnonic transport has been investigated in various platforms (ferro- and antiferromagnets) and is characterized by anomalous mechanisms such as the thermal Hall effect and the magnon Nernst effect. The usually employed framework of Linear Spin Wave Theory, in fact, does not take into account effects as magnon lifetime damping [1], energy renormalization [2] and topological phase transitions [3] that arise purely due to magnon-magnon interactions and can alter the flow of magnons dramatically. In this work we primarily investigate the results of three and four magnon interactions in the topologically non-trivial collinear honeycomb antiferromagnet and the results of these interactions in the magnon Spin Nernst effect. If time allows the impact of magnon interactions in a noncollinear antiferromagnet will also be addressed.

**Acknowledgements**

K.S. and A.M acknowledge support from the Excellence Initiative of Aix-Marseille Universite - A*Midex, a French ”Investissements d’Avenir” program.

**References**

[1] A.L. Chernyshev and P.A. Maksimov. Damped topological magnons in the kagome-lattice ferromagnets. Physical Review Letters, 117(18):187203, 2016.

[2] A. Mook, K. Plekhanov, J. Klinovaja, and D. Loss. Interaction stabilized topological magnon insulator in ferromagnets. Physical Review X, 11(2):021061, 2021.

[3] Y.S. Lu, J.L. Li, and C.T. Wu. Topological phase transitions of dirac magnons in honeycomb ferromagnets. Physical Review Letters, 127(21):217202, 2021

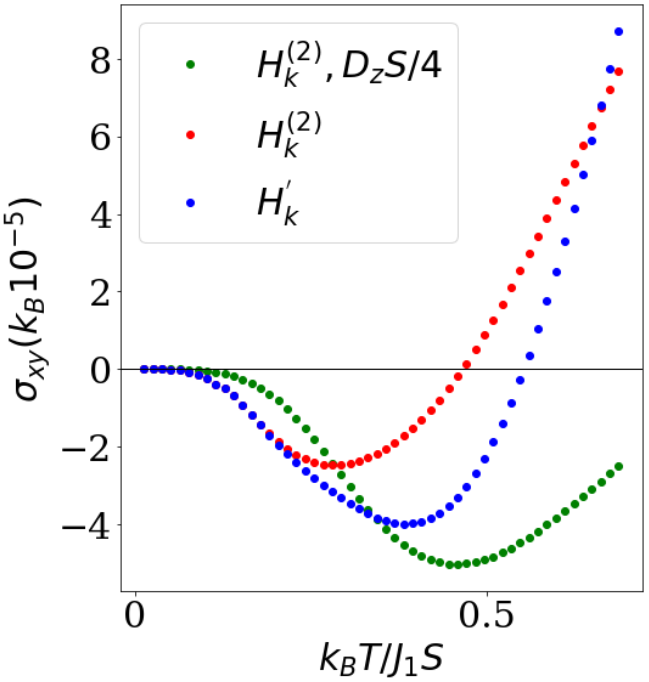



Figure: The magnon Spin Nernst conductivity is plotted along the temperature for the non-interacting model for two different values of out-of-plane DMI (green and red line) and the interacting model (blue line). The interacting conductivity matches the non-interacting one for low temperatures but as the temperature increases it approaches the negative values for a lower DMI.

# Impact of Spin Waves Dispersion on Surface Acoustic Waves and Ferromagnetic Resonance in Iron Thin Films

*Pauline Rovillain[1*], Jean-Yves Duquesne[1], Louis Christienne[1], Mahmoud Eddrief,[1]Maria Gloria Pini,[2] Angelo Rettori,[3,4] Silvia Tacchi,[5] and Massimiliano Marangolo[1]*

[1]*Sorbonne Université, CNRS, Institut des NanoSciences de Paris, UMR7588, F-75005 Paris, France*
[2]*Istituto dei Sistemi Complessi del CNR (CNR-ISC), I-50019 Sesto Fiorentino, Italy*
[3]*Dipartimento di Fisica ed Astronomia, Universita degli Studi di Firenze, I-50019 Fiorentino, Italy*
[4]*INFN, Sezione di Firenze, 50019 Sesto Fiorentino, Italy*
[5]*Istituto Officina dei Materiali del CNR (CNR-IOM), c/o Dipartimento di Fisica e Geologia, Universita di Perugia, I-06123 Perugia, Italy*
*pauline.rovillain@insp.upmc.fr

Surface Acoustic Waves (SAW) have been proposed to dynamically control Spin Waves (SW) propagation and even to generate SWs, in order to implement reconfigurable and energy efficient magnonic devices. Indeed, SAW technology is mature and widely used in today's sensors, filters and microwave circuitry, not withstanding the lack of tunability of SAW transducers. For this reason, a multitude of them are currently integrated in modern devices (e.g. mobile phones).

Recently, the so-called SAW induced Ferromagnetic Resonance (SAW-FMR) has been observed by Weiler et al. [1] in Ni thin films by exciting SAWs in the GHz and sub-GHz regime in piezoelectric media. Moreover, SAW-FMR permits to induce spin-pumping in CoPt bilayers [2,3]. The SAW-FMR interaction is often described by taking into account only the uniform FMR mode. However, this approximation is rather crude, and it misses out the wealth of modes that can be excited in a ferromagnetic (FM) material.

Here, we study SAW propagation in a Fe thin film epitaxially grown on a piezoelectric GaAs substrate. The SAW velocity and absorption show a dependence on the external magnetic field in amplitude and direction after coupling with the FMR. [4,5] The observed angle dependence can be described only by taking into account the spin wave dispersion. To interpret this dependence on the magnetic field angle, a phenomenological approach to the relative change in SAW velocity is implemented with the calculated spin wave dispersion curves. Our study permits to envisage SAW-based magnonic devices where a single IDT provides the energy needed to activate magnetization dynamics and SW propagation in hundreds if not thousands of magnonic waveguides, in a Joule heat free manner.

**Acknowledgements**

The authors acknowledge support from the Agence National de la Recherche française Grant No. ANR-22-CE24-0015 SACOUMAD and from the European Union within the HORIZON-CL4-2021-DIGITAL-EMERGING-01 Grant No. 101070536 MandMEMS.

**References**

[1] M. Weiler, *et al., Physical Review Letters* **106** (2011) 117601.
[2] M. Weiler *et al. Physical Review Letters* **108** (2012) 176601.
[3] J.-Y. Duquesne *et al. Physical Review Applied* **12** (2019) 024042.
[4] P. Rovillain *et al. Physical Review B* **102** (2020) 184409.
[5] P. Rovillain *et al. Physical Review Applied* **18** (2022) 064043.

# Topological Spin-Wave Edge Modes in a Moiré Magnonic Crystal

*Marco Madami*[a]*, *Hanchen Wang*[b], *Jilei Chen*[c], *Gianluca Gubbiotti*[d], *Haiming Yu*[b,c]

[a] *Dipartimento di Fisica e Geologia, Università di Perugia, Perugia, Italy*
[b] *Fert Beijing Institute, MIIT Key Laboratory of Spintronics, School of Integrated Circuit Science and Engineering, Beihang University, Beijing, China.*
[c] *International Quantum Academy, Shenzhen, China*
[d] *Istituto Officina dei Materiali del Consiglio Nazionale delle Ricerche (IOM-CNR), c/o Dipartimento di Fisica e Geologia, Perugia, Italy.*
*marco.madami@unipg.it

Moiré superlattices consisting of twisted layers of van der Waals materials [1] exhibit extraordinary electronic behaviours such as superconductivity and correlated topological states. The concept of moiré physics was recently applied in photonics to reconstruct photonic band structures providing novel functionalities such as magic-angle lasers. Magnons, quanta of spin waves, are collective excitations of magnetically ordered materials and can convey information without charge transport promising for low-power magnon computing. To date, moiré physics in magnonic systems has only been studied from a theoretical point of view [2]. In this work, we fabricated nanostructured moiré magnonic crystals based on low-damping yttrium iron garnet thin films. We report experimental observation of propagating spin waves at the edge of moiré unit cells using Brillouin light scattering spectro-microscopy. Spin-wave edge modes appear only around an optimal twist angle and are highly sensitive to the excitation frequency, which indicates their origin from mode hybridization with a moiré mini-flatband. Moiré spin-wave edge modes observed in this work, as the magnonic counterpart of the magic-angle electronic and photonic systems, open an emergent research direction of moiré magnonics.

## References

[1] Cao, Y. et al. *Nature* **556**, 43-50 (2018).
[2] Li, Y.-H. & Cheng, R. *Phys. Rev. B* **102**, 094404 (2020).

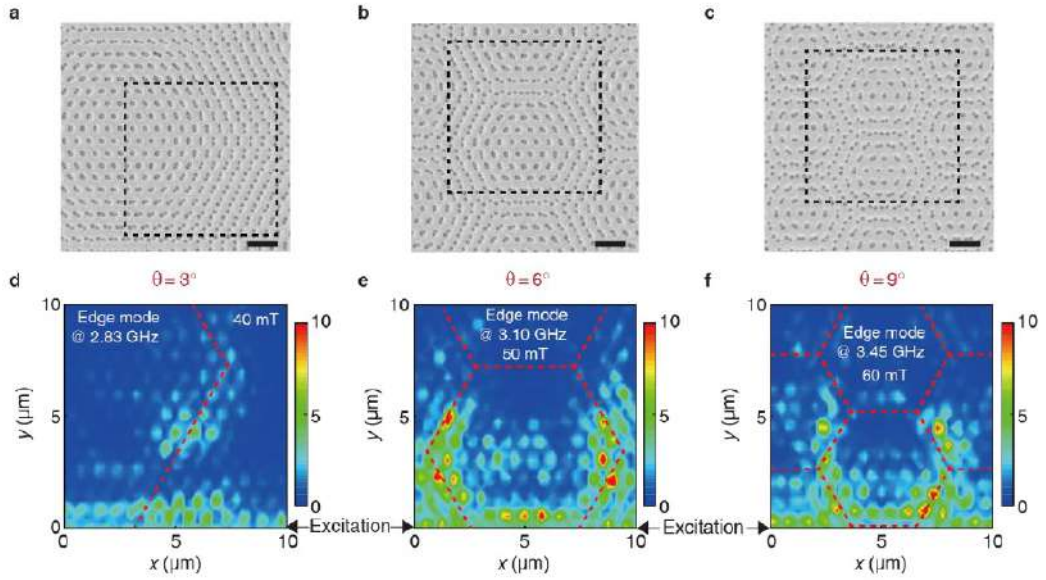



Figure: Spin-wave edge mode profiles at different twist angles. a-c, SEM images of moiré magnonic crystals with twist angles of 3°, 6°, and 9°, respectively. d-f, Spatial-resolved BLS measurement at a twist angle of 3°, 6°, and 9°, respectively.

# Nanopositioning of a single-spin sensor for imaging spin waves

*S. Kurdi[1]*‡, Brecht G. Simon [2‡], Allard J, Katan[1], Toeno van derSar[1]*

[1] *Department of Quantum Nanoscience, Delft University of Technology, 2628 CJ Delft, The*

*s.kurdi@tudelft.nl

‡These authors contributed equally to this work

Magnetometry based on nitrogen-vacancy (NV) spins in diamond has recently emerged as a powerful tool for probing spin waves [1], the elementary excitations of coupled spins in magnetically ordered materials. In this work, I will focus on how we utilize scanning NV magnetometry, in which we use a NV sensor spin that is shallowly embedded in the tip of a diamond scanning probe, to image spin waves in a thin film insulator (figure). Specifically, I will focus on demonstrating the power of controlling the NV-sample distance for probing spin waves of varying spatial frequencies.

Spin waves generate a rotating magnetic stray field with an amplitude that decays with increasing distance to the sample in which the decay length set by the spin-wavenumber. Controlling the NV-sample distance is therefore crucial for selective probing of spin-wavenumbers.

In this work, I will present two applications for controlling the NV-film distance on the sub-micron/nm scale, via lift-height measurements, to probe both coherent and incoherent spin waves:

(1) I will demonstrate that our NV probe can be used as a spin-wave filter for selective imaging of short or long wavelength spin waves that are frequency degenerate [2].

(2) I will show how microwave excitation of low-wavenumber spin waves leads to a high-density and a unidirectional incoherent magnon gas. I will demonstrate how the spatial decay of the stray fields reveals the wavenumber content of both coherently excited spin waves with a well-defined wavenumber as that of the incoherent magnon gas [3].

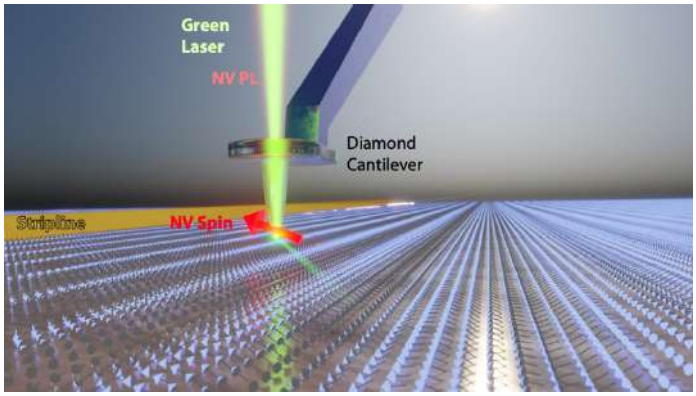



**Figure:** A diamond cantilever, with a nitrogen vacancy centre implanted ~20 nm below the tip surface, is mounted in a scanning-probe setup and is used for detecting the stray field of spin waves that are excited by a gold stripline. The nitrogen vacancy spin is initialized using a green laser and read out via spin-dependent photoluminescence (PL).

These results reveal the power of scanning NV-magnetometry as a tool for spin-wave probing. Whilst showcasing that nanoscale control over the NV-sample distance enables wavenumber-selective imaging of magnetization oscillations and open up new avenues for imaging other coherent and incoherent spin-wave modes.

## References

[1] I. Bertelli et al. *Sci. Adv.* **6** (2020) eabd3556.
[2] B. G Simon*, S. Kurdi* et al. *Nano Lett*. **22** (2022) 9198.
[3] B. G Simon*, S. Kurdi* et al. *Nano Lett*. **21** (2021) 8213.

# Compact Localised States in Magnonics

*Grzegorz Centała*[1], *Jarosław W. Kłos*[1]*

[1] *ISQI, Faculty of Physics, Adam Mickiewicz University, Poznań, Poland*

*klos@amu.edu.pl

In bipartite lattices, the sites from sublattice A have the nearest neighbours (NN) in sublattice B only. Therefore, the mods occupying one sublattice are not able to propagate (in tight-binding models with NN hopping) and the dispersion relation can by described as flat bands with zero group velocity. Such modes, localized without defects in a perfectly periodic and infinitely extended system, are called compact localized states (CLS) [1]. The well-known type of bipartite lattice is a Lieb lattice [2]. The CLS were already observed in photonic crystals based on Lieb lattices [3] but the studies on CLS in the magnonic system still need to be performed. We proposed a perpendicularly magnetized Ga-doped YIG layer as a base for a magnonic Lieb lattice where the lattice sites are mimicked by cylindrical inclusion made of YIG (without Ga-doping). We tailored the structure to observe the oscillatory and evanescent spin waves in inclusions and matrix, respectively. We calculated the dispersion relations exhibiting Dirac cones, almost touching each other at the M-point (with a very narrow gap ~15 MHz), intersected by a relatively flat band of magnonic CLS. Then, we supplemented our studies by considering the extended magnonic Lieb lattices, characterized by a larger number of weakly dispersive bands specific for CLS. The computations were performed by finite element method, using COMSOL Multiphysics.

**Acknowledgements**

The authors would like to acknowledge the support from the National Science Center – Poland grants No. 2020/39/O/ST5/02110 and 2021/43/I/ST3/00550. GC acknowledges Polish National Agency for Academic Exchange grant BPN/PRE/2022/1/00014/U/00001.

**References**

[1] J.-W. Rhim and B.-J. Yang, *Phys. Rev.* B **99** (2019) 045107.

[2] E. H. Lieb, Phys. *Rev. Lett.* **62** (1989), 120; erratum **62** (1989), 1927

[3] D. Leykam et al, *ADV PHYS-X* **3** (2018), 1473052.

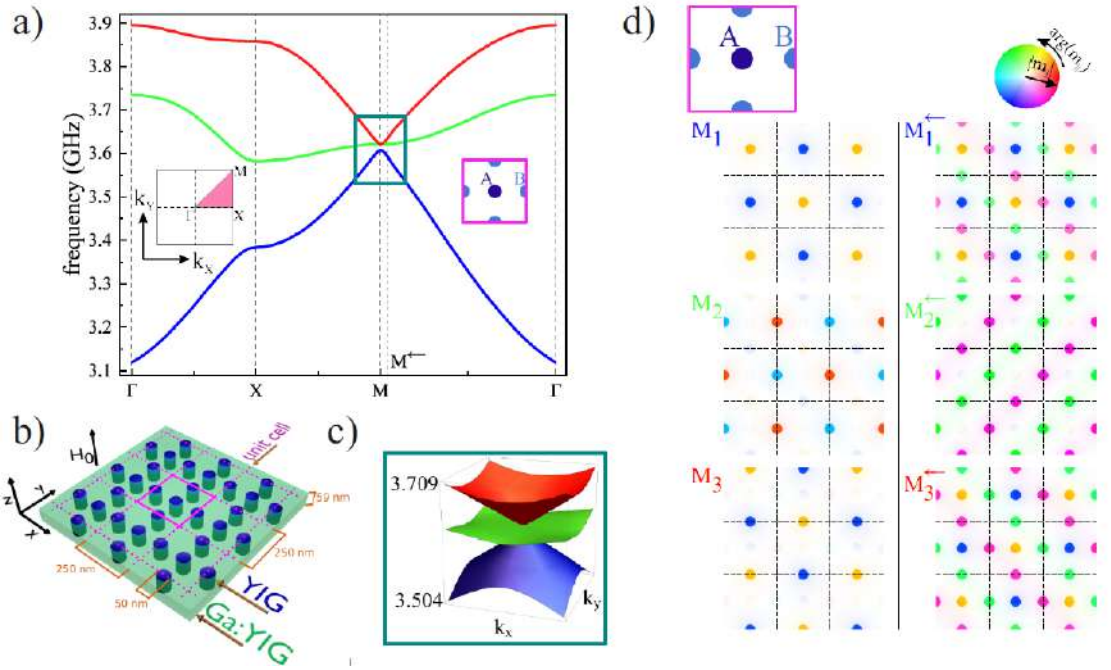



Figure: (a) Dispersion relation for (b) magnonic Lieb lattice with perpendicularly applied field (100 mT) shows (c) Dirac cones intersected by the flat band (green sheet) where (d) compact localized states ($M_2^{\leftarrow}$) are found.

# Magnon straintronics in 2D magnetic materials

*Dorye L. Esteras[1]*, Andrey Rybakov[1], Alberto M. Ruiz[1], José J. Baldoví[1]*
[1] *Instituto de Ciencia Molecular, Universitat de València, Catedrático José Beltrán 2, 46980 Paterna, Spain.*
*dorye.esteras@uv.es

2D magnetic materials expanded the scope of material science to design devices that can offer a plethora of opportunities[1]. One of the most important groups of 2D magnetic materials is $CrX_3$ (X: Cl, Br, I) where in particular $CrI_3$ was the first 2D ferromagnet in the monolayer limit[1]. Another promising group of 2D magnetic materials is the $MPS_3$ (M: Fe, Ni, Co, Mn) family, formed by van der Waals materials with excellent electronic and optical properties[2-5]. Despite all these properties make 2D magnets very interesting by themselves, the magnetism present in these materials is wide and yet not well understood, giving rise to exotic phenomena. Herein we present a theoretical methodology to analyse 2D materials, based in DFT calculations, Wannier Hamiltonians and Green functions. Using these techniques, the magnetic and exchange properties can be resolved in terms of chemical orbitals and improved with strain simulations that result in an increase of Curie temperatures and magnon propagation along the 2D materials,[6,7] thus exploring alternative platforms for spintronic devices.

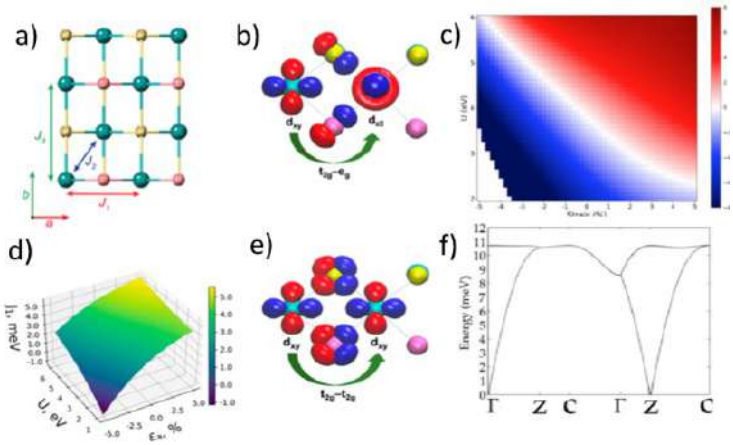



**Figure:** Strain in 2D magnets: Exchange interactions, Curie temperature and magnons modes.

## Acknowledgements

The authors acknowledge the financial support from the European Union (ERC-2021-StG-101042680 2D-SMARTiES and the Generalitat Valenciana (grant CDEIGENT/2019/022, CIDEGENT/2018/004.

## References

[1] B. Huang, G. Clark, E. Navarro-Moratalla, D. R. Klein, R. Cheng, ... & X. Xu, *Nature*, ***546*** (2017), 270-273.
[2] B. L. Chittari, Y. Park, D. Lee, M. Han, A. H. MacDonald, E. Hwang, & J. Jung, Phys. Rev. B, ***94*** (2016), 184428.
[3] M. J. Coak, D. M. Jarvis, H. Hamidov, … & J. G. Park, *Journal of Physics: Condensed Matter, 32,* (2019), 124003.
[4] M. Ramos, F. Carrascoso, … ,**D. L. Esteras,** ... & M. R. Calvo, *npj 2D Materials and Applications, **5** (*2021) 1-9.
[5] M. Ramos, … , **D. L. Esteras,** ... & M. R. Calvo, *ACS applied materials & interfaces,* **14** (2022), 33482-33490.
[6] **D. L. Esteras,** A. Rybakov, A. M. Ruiz, & J. J. Baldoví, *Nano Letters*, **22** (2022), 8771-8778.
[7] A. M. Ruiz, **D. L. Esteras**, A. Rybakov, & J. J. Baldoví, Dalton Transactions, **51** (2022), 16816-16823.

# Nonlinear longitudinal and transverse magnetoresistances due to magnon creation-annihilation

*P. Noël [1*], R. Schlitz[1], E. Karadza[1], F. Binda[1],C.-H. Lambert[1], and P. Gambardella[1]*

*1Department of Materials, ETH Zurich, CH-8093 Zurich, Switzerland*

*paul.noel@mat.ethz.ch

At finite temperature, the magnetization vector is not a fixed quantity. Quantized spin waves – magnons – excited by thermal fluctuations reduce the magnetization relative to the full alignment of all spins. It is well known that magnons can carry or excite spin currents [1] and that the magnon population in turn can be modified by spin currents in ferromagnet (FM)/normal metal (NM) bilayers [2]. The current-induced modulation of the population of magnons thus inevitably leads to a modification of the average magnetization [3]. The change of the total magnetization due to the magnon create-annihilation is depicted in the figure.

The concept of nonlinear longitudinal and transverse magnetoresistance due to magnon creation-annihilation follows naturally from this premise: the current-induced modulation of the population of magnons leads to a modification of the magnetization as depicted in the figure causing a change of the resistance. We describe the angular and field dependence of these nonlinear effects and evidence their role in the total nonlinear contribution to the transport in FM/NM bilayers. These contributions appear in both insulating and conductive FM [4]. Additionally, their angular dependence is not easily distinguishable from other current-dependent contributions due to the spin orbit torques (SOTs), unidirectional magnetoresistances and magnetothermal effects observed in FM/NM bilayers [5].

We show that the magnon creation-annhilation magnetoresistances appear prominently in $Y_3Fe_5O_{12}$/Pt (YIG/Pt) bilayers due to the small damping of YIG. They are weaker but also present in conductive ferromagnets with larger damping such as Co or CoFeB. We further show that the similar angular dependence of the magnetoresistance and SOTs contributions to the Hall voltage can strongly affect the SOT estimation when using the harmonic Hall measurement method. When the magnon contribution is not unaccounted for, both the antidamping (AD) and fieldlike (FL) torque contributions strongly deviates from their expected field dependence and a correction needs to be performed to properly estimate their value. The AD torque, the most important parameter for spin orbit torque applications, is overestimated by 30% in Pt/CoFeB, 100% in W/CoFeB and one order of magnitude in YIG/Pt when not accounting for the magnonic contribution [6]. This can explain, at least partially, the discrepancy on the torque estimation observed in the literature.

[1] Cornelissen, L. J., Liu, J., Duine, R. A., Youssef, J. B. & van Wees, B. J., Nature Phys 11, 1022–1026 (2015).
[2] Demidov, V. E. *et al.*, Phys. Rev. Lett. 107, 107204 (2011).
[3] Demidov, V. E. *et al.*, Sci Rep 6, 32781 (2016).
[4] Noël, P., Schlitz, R., Karadza, E. *et al.* (in preparation)
[5] Avci, C. O. *et al.*, Phys. Rev. B 90, 224427 (2014).
[6] Noël, P., Karadza, E., Schlitz, R. *et al.* (in preparation)

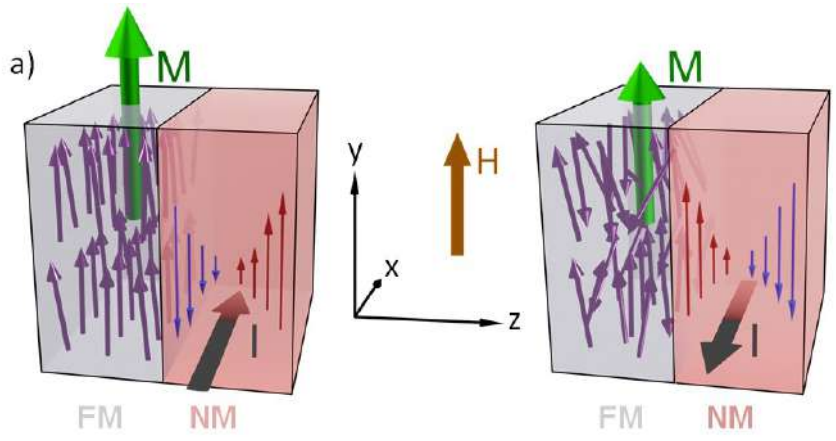



Figure: annihilation and creation of magnons due to spin accumulation in the normal metal layer. The local magnetic moments are shown in purple.

# Magnetoelastic Transducers for Spin-Wave Generation

*Daniele Narducci[1,2], Florin Ciubotaru[1], Moritz Geilen[3], Julius Balliet[3], Giacomo Talmelli[1,4], Björn Heinz[3], Burkard Hillebrands[3], Johan De Boeck[1,2], Inge Asselberghs[1], Gouri Sankar Kar[1], Philipp Pirro[3], Sebastien Couet[1], Christoph Adelmann1[1]*

*[1]IMEC, Leuven, 30001, Leuven, Belgium*

*[2] Department of Electrical engineering, ESAT, 3001, Leuven, Belgium*

*[3]Rheinland-Pfälzische Technische Universität Kaiserslautern-Landau, 67663 Kaiserslautern, Germany*

*[4]Department of Material engineering, MTM, 3001, Leuven, Belgium*

*Daniele.Narducci@imec.be

Spin-wave devices use wave interference to enable logic operations and offer a promising solution to reduce energy consumption in computational units. Energy-efficient spin-wave transducers are needed to compete with CMOS and while current-based system does not meet the energy requirements [1], magnetoelectric systems show potential for much higher power efficiency [2]. The device proposed in this study is based on magnetoelastic transducers (METs), a magnetoelectric system based on piezoelectric-magnetostrictive heterostructures where the transduction between voltage and magnetization is mediated by the strain. It consists of a PZT pillar, the piezoelectric element, embedded in a soft material and in contact with a CoFeB waveguide, the magnetostrictive element (figure 1 (a)). RF voltages are applied to the structure using top and bottom electrodes. The devices are fabricated using an extensive set of deposition and etching techniques, as well as common lift-off processes, whereas all the patterning steps were done by electron beam lithography. Here we report on the fabrication of spin-wave devices based on METs with piezoelectric pillar dimensions down to 80 × 80 nm$^2$ (figure 1 (b)) and their characterization by micro-focus Brillouin Light Scattering (BLS) spectroscopy. Figure 2 displays a BLS spectrum recorded for a device where the PZT pillar has lateral dimensions of 500 nm × 10 µm, and a CoFeB waveguide 10 µm wide. It is shown that the phonon spectra strongly depend on the geometry and size of the pillar due to quantization effects. The emission of spin waves depends on the magnetization direction with respect to the waveguide, a direction of 45° being more favourable for excitation (see Fig. 2). The magnetoelastic field and torque on the magnetization for different geometries were derived from the strain distributions obtained by device simulation in COMSOL Multiphysics software. We show that the excitation efficiency depends strongly on both the magnetization direction as well as on the strain configuration.

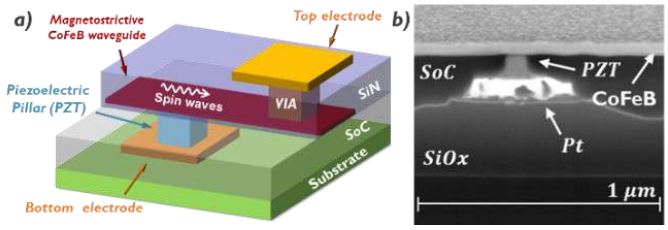



Figure 1: The magnetoelastic transducer device a) schematic b) cross section scanning electron microscopy image of a fully processed 80 by 80 nm2 device.

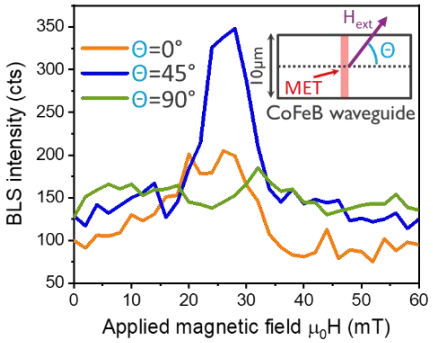



Figure 2: Spectra of spin waves emitted by a 500nm x 10 µm MET in a 10 µm wide magnetic waveguide as a function of the external magnetic field amplitude and direction with respect the CoFeB waveguide. The applied RF frequency was 6 GHz.

## Acknowledgements

This work has been funded by the European Union's Horizon 2020 research and innovation program within the FET-OPEN Project CHIRON under Grant Agreement No. 801055. D.N. and F.V. acknowledge financial support from the Research Foundation – Flanders (FWO) through Grant Nos. 1SB9121N and 1S05719N, respectively. The authors acknowledge the imec's lab support and the electron beam lithography teams.

## References

[1] F. Meng, F. Ciubotaru, S.-Y. Lee, O. Zografos, M. Gupta, V.D. Nguyen, S. Couet, G. Sankar Kar, G. De Micheli, C. Adelmann, I. Asselberghs, arXiv:2209.01999 (2022).

[2]A. Khitun, K. L. Wang, "Non-volatile magnonic logic circuits engineering", Journal of Applied Physics **110**, (2011) 034306

# Ultrafast Laser-Induced Magneto-Optical Changes in Resonant Magnetic X-Ray Reflectivity

*Wolfgang Kuch[1]*, Ivar Kumberg[1], Evangelos Golias[1], Sebastien Hadjadj[1], Rahil Hosseinifar[1], Sangeeta Thakur[1], Tauqir Shinwari[1], Ismet Gelen[1], Niko Pontius[2], Christian Schüßler-Langeheine[2], Clemens von Korff Schmising[3], and Sangeeta Sharma[3]*

[1] *Institut für Experimentalphysik, Freie Universität Berlin, Berlin, Germany*
[2] *Helmholtz-Zentrum Berlin für Materialien und Energie, Berlin, Germany*
[3] *Max-Born-Institut für Nichtlineare Optik und Kurzzeitspektroskopie, Berlin, Germany*
*kuch@physik.fu-berlin.de

The element-resolved study of ultrafast demagnetization processes is essential to understand the underlying local and nonlocal mechanisms that govern the magnetization dynamics in multi-component systems. Combining ultrashort soft-x-ray probe pulses with near-infrared pump pulses allows to follow the magnetic response of an excited system in an element-resolved way. Resonant reflectivity of soft x rays is thereby an invaluable tool for probing the magnetic response of intransparent samples via magnetic circular dichroism. However, the change of x-ray reflectivity is not simply related to the quench of the magnetization but reflects also the transient repopulation of electronic states and changes in the dielectric properties, for example caused by heat-induced strain.

We present time-resolved experimental x-ray reflectivity data of the response of Co films in Cu/Co/Mn/Cu(001) to 100 fs near-infrared laser excitations of different fluences, performed at the femtosecond slicing facility of BESSY II. Recording the reflectivity at the Co $L_3$ edge for opposite directions of sample magnetization, we separate magnetic and nonmagnetic contributions (Figure). We simulate the results by two different approaches. On the one hand, we use a purely thermal model, a time-dependent macroscopic magnetization, and heat-induced strain profiles. On the other hand, we employ time-dependent density-functional theory to calculate the transient optical response to the laser-induced excitation and from that the reflected intensities. While both methods are able to reproduce the time dependence of the magnetic signal, the nonmagnetic change in reflectivity is captured satisfactorily only in simulations of the transient optical response function and has thus to be assigned to electronic effects.

From the simulations, we derive a rule of thumb to know whether a reflected signal is dominated by the dielectric or the absorptive part of the dielectric function for given scattering conditions.

**Acknowledgements**

Financial support from the DFG (Sfb/TRR 227 “Ultrafast Spin Dynamics”) is gratefully acknowledged. We thank the Helmholtz-Zentrum Berlin for the allocation of synchrotron radiation beamtime.

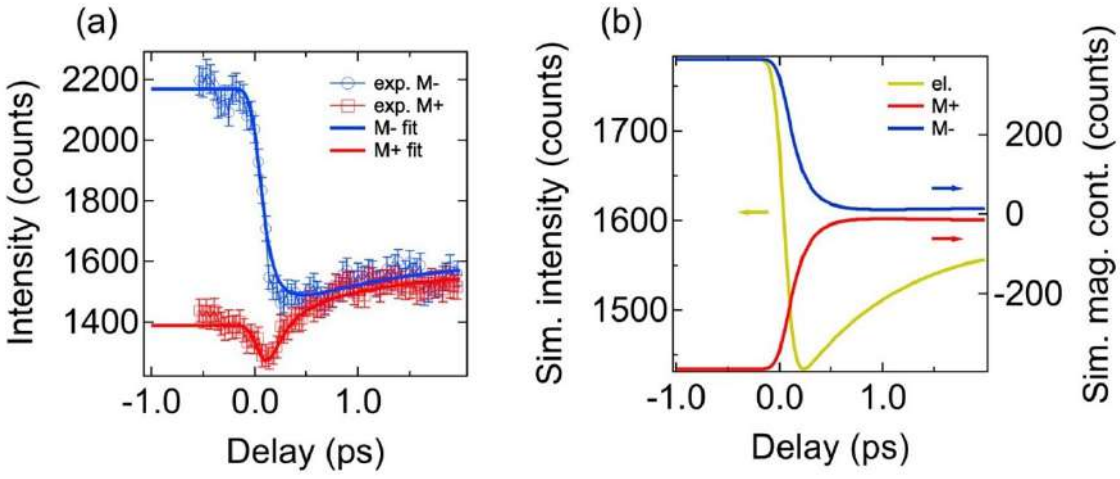



Figure: (a) Measured time traces of the resonant reflectivity at the Co $L_3$ edge for opposite magnetization directions after pumping with a 100 fs laser pulse. (b) Decomposition of the measured signal into magnetic and electronic contributions.

# Coherent Control of Photo-Magnetic Precession and Switching by Double-Shot Laser Pulses

*T. Zalewski*[1]*, *L. Nowak*[1,2], *A. Stupakiewicz*[1]

[1] *Faculty of Physics, University of Bialystok, Bialystok, Poland*

[2] *Charles University, Faculty of Mathematics and Physics, Prague, Czech Republic*

*tmkzlwsk@uwb.edu.pl

The ability to coherently control the state of magnetization in magnets with an ultrashort laser pulse has recently been the subject of extensive research [1]. This intense stimulus makes it possible to achieve ultrafast all-optical switching of magnetization. We can differentiate thermal-based switching in metals and non-thermal switching based on the photo-magnetic effect in dielectrics. The non-thermal switching mechanism originates from a precessional reversal of the magnetization driven by the photo-induced change of magnetic anisotropy. It was shown that using only a single, linearly polarized laser pulse is enough to reversibly switch the magnetization state in dielectric yttrium iron garnet (YIG:Co) films [2]. Such a switching does not require any external magnetic field and can be done at room temperature. In this condition, the magnetic anisotropy of the examined garnet is predominantly cubic, which results in four easy magnetization axes localized close to the cube diagonals of <111>-type. We recently demonstrated that by using double-shot femtosecond pulses with alternating linear polarization planes it is possible to re-write magnetization with a repetition rate of up to 20 GHz in YIG:Co film with a non-zero miscut angle. [3]

Here, using a double-shot time-resolved experiment setup, we studied the ultrafast photo-induced back-switching in YIG:Co films with pure cubic symmetry without miscut angle. The lack of miscut provides the lack of symmetry degeneracy which provides the back-switching effect in which the even number of incident pulses leaves the same magnetic state, and the odd number of pulses reverses it. No need to control the polarization of the pump beam simplifies the system at the expense of non-deterministic switching. Moreover, we show that using two temporally separated stimuli gives a field for modifying and adjusting the precession and therefore the switching trajectories. We retrieve the ultrafast magnetization dynamics by utilizing an experimental setup with three color pump and probe geometry, with the ability to trace single-shot switching dynamics with the spatiotemporal resolution obtained by time-resolved imaging. [4] The use of two phase-coherent pulses makes it possible, through their interference controlled by relative delay, to strengthen the precession amplitude, and thus to lower the switching threshold. On the other hand, impulses in the opposite phase will allow obtaining a complete freeze of precession. This effect was not observed with the sample with the non-zero miscut angle.

Finally, we discuss the temporal delay between writing-erasing sequences and their properties using the back-switching regime in YIG:Co. Modification of the effective anisotropy using the sample temperature allows tuning the recording frequency in the range of 10-40 GHz.

## References

[1] A. Kirilyuk, A.V. Kimel, and Th. Rasing, *Rev. Mod. Phys.* **82**, 2731 (2010).
[2] A. Stupakiewicz, et al., Nature **542**, 71 (2017).
[3] K. Szerenos, et al., *Phys. Rev. App.* ***12***(4), 1 (2019).
[4] T. Zalewski, A. Stupakiewicz, *Rev. Sci. Instrum.* **92**, 103004 (2021)

# Magnetoelastic Excitation Driven by Femtosecond Laser Comb

*Avinash Kumar Chaurasiya*[1 *], *Ahmad A. Awad*[1,2], *Roman Khymyn*[1], *and Johan Åkerman*[1,2]
[1]*Department of Physics, University of Gothenburg, 412 96 Gothenburg, Sweden*
[2] *Research Institute of Electrical Communication, Tohoku University, 2-1-1 Katahira, Aoba-Ku, Sendai 980-8577, Japan*
**avinash.kumar@physics.gu.se*

Recent progress in the field of magnonics and spintronics has sparked significant interest in studying phenomena related to magnons and phonons, both from a fundamental research perspective and for potential applications in information and communication technology [1-2]. Most recently, ultrafast electron diffraction studies of Ni films revealed that ultrafast interactions of spins with high-frequency phonons are decisive for the dynamics of rapidly demagnetized magnetic materials [3]. Here, we adopt a novel approach based on high repetition rate (1 GHz) femtosecond (fs) laser combs combined with micro-focused Brillouin light scattering microscopy ($\mu$BLS) to investigate the polarization characteristics of both phonons and magnons in NiFe thin films. The versatility of frequency comb enhanced $\mu$BLS [4] has proven to be an efficient way for ultrafast generation of sustained spin waves [5] and spinwave caustics [6].

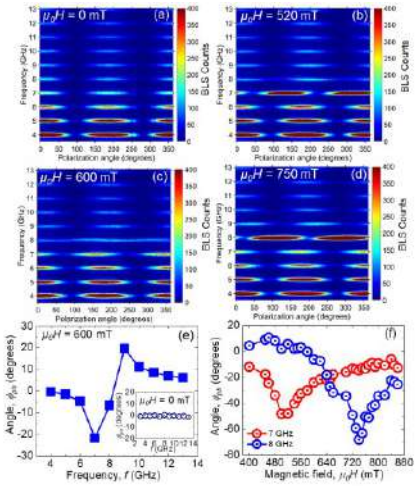



Figure 1: (a-d) Polarization maps for 20 nm NiFe thin film measured at various applied magnetic fields. (e-f) The variation of the extracted angle for phonons as a function of frequency and field magnitude respectively.

Films of permalloy ($Ni_{80}Fe_{20}$) were sputtered onto *c*-plane sapphire substrates by dc magnetron sputtering under a 3 mTorr Ar atmosphere in an ultrahigh vacuum chamber. In the $\mu$BLS experiment, the sample is excited with a 1 GHz rep-rate fs-laser of wavelength 816 nm and probed with a continuous wave laser of wavelength 532 nm. The scattered beam from the sample is passed through a polarizer and guided to the tandem Fabry-Pérot interferometer for spectral decomposition of the light. Figure 1(a-d) shows the polarization maps measured for different external magnetic fields ($\mu_0 H$) = 0, 520, 600, and 750 mT at fixed pump power of 40 mW. In the presence of the pump laser, a series of peaks, corresponding to the harmonics of the 1 GHz rep rate, appear. We model the observed BLS counts ($I$) for various frequencies by considering the contribution of both phonons and magnons which is in good qualitative agreement with a $\sin^2$ dependence of the polarization angle.

$$I = A_{ph} \sin^2(x - \phi_{ph}) + A_m \sin^2(x - \phi_m) \quad (1)$$

The respective amplitudes of the phonons and magnons ($A_{ph}$ and $A_m$) as well as the polarization angle for phonons ($\phi_{ph}$) have been extracted as a function of frequency. In stark contrast to thermally excited incoherent phonons/magnons, photons scattered from the coherently excited phonons/magnons exhibit a giant rotation of their polarization angle. The rotation is frequency and field dependent, showing a resonant behavior around the NiFe ferromagnetic resonance, where it changes sign (as shown in Fig. 1(e-f)). We observe the tunability of the magnetoelastic nature of SW modes with the magnitude of the applied magnetic field. Our study highlights the effectiveness of frequency combs for generating and investigating magnetoelastic sustain excitation in magnetic thin films at several GHz frequencies.

## Acknowledgements
We gratefully acknowledge the financial support from Swedish Research Council (VR, Grant No. 2016-05980) and the Horizon 2020 Research and Innovation Programme (ERC Advanced Grant No. 835068 “TOPSPIN”).

## References
[1] A. V. Chumak, V. I. Vasyuchka, A. A. Serga, and B. Hillebrands, *Nat. Phys*. **11** (2015) 453.
[2] J. Holanda, D. Maior, A. Azevedo, and S. Rezende, *Nat. Phys.* **14** (2018) 500.
[3] S. R. Tauchert, M. Volkov, D. Ehberger, D. Kazenwadel, M. Evers, H. Lange, A. Donges, A. Book, W. Kreuzpaintner, U. Nowak, and P. Baum, *Nature* **602** (2022) 73.
[4] A. Alemán, S. Muralidhar, A. A. Awad, J. Åkerman, and D. Hanstorp, *Opt. Express* **28** (2020) 29540.
[5] S. Muralidhar, A. Awad, A. Alemán, R. Khymyn, M. Dvornik, D. Hanstorp, and J. Åkerman, *Phys. Rev. B* **101** (2020) 224423.
[6] S. Muralidhar, R. Khymyn, A. Awad, A. Alemán, D. Hanstorp, and J. Åkerman, *Phys. Rev. Lett.* **126** (2021) 037204.

# Role of Non-Equilibrium Magnons in the Ultrafast Demagnetization of Derromagnets

*Félix Dusabirane*[1], *Kai Leckron, Hans Christian Schneider**

[1] *Physics Department and Research Center OPTIMAS, RPTU University of Kaiserslautern-Landau, Kaiserslautern, Germany*

*hc.schneider@rptu.de

We theoretically study the carrier dynamics in an itinerant ferromagnet due to electron-magnon scattering on ultrafast timescales. Our approach is based on a model band structure ("Stoner model") for the electronic single-particle states, and the electron magnon-interaction is formally obtained as coupling to a Heisenberg model, which incorporates the magnetic properties of the itinerant ferromagnet [1]. We compute the dynamics of momentum resolved electron $n_{\sigma=\pm,k}(t)$ and magnon $N_q(t)$ distributions due to electron-magnon and electron-electron scattering, which are treated at the level of Boltzmann scattering integrals. We also include an Elliott-Yafet like spin relaxation mechanism due to spin-orbit coupling [2].

We find that electron-magnon scattering leads to a pronounced non-equilibrium for magnon modes at higher energies and wave vectors that couple directly to Stoner transitions as shown in the Figure. For reasonable parameters that capture some of the properties of magnons in iron, the electronic spin-flip scattering with magnons results in a momentum dependent absorption/emission of magnons and a transient *increase* of the electron spin polarization on a timescale of a few ten femtoseconds, which is then removed by the Elliott-Yafet spin relaxation. Such an increase of the spin polarization is also found in the dM/dt model, which has been introduced to model ferromagnetic demagnetization dynamics and spin current generation [3]. As we do not make its quasi-equilibrium assumption, we can go beyond the dM/dt model and demonstrate the non-equilibrium features inherent in the demagnetization dynamics. We study the influence of magnon-phonon relaxation processes and different excitation conditions, and compare our results to recent experiments [4].

## Acknowledgements

Funded by the Deutsche Forschungsgemeinschaft (DFG) TRR 173 – 268565370 (project B03).

## References

[1] B. Woolsey and R. M. White, *Phys. Rev. B* 1 (1970) 4474.
[2] K. Leckron, S. Vollmar, H. C. Schneider, *Phys. Rev. B* **96** (2017) 140408.
[3] M. Beens, R. A. Duine, B. Koopmans, *Phys. Rev. B* **102** (2020) 054442.
[4] M. Stiehl et al., *Appl. Phys. Lett.* **120** (2022) 062410.

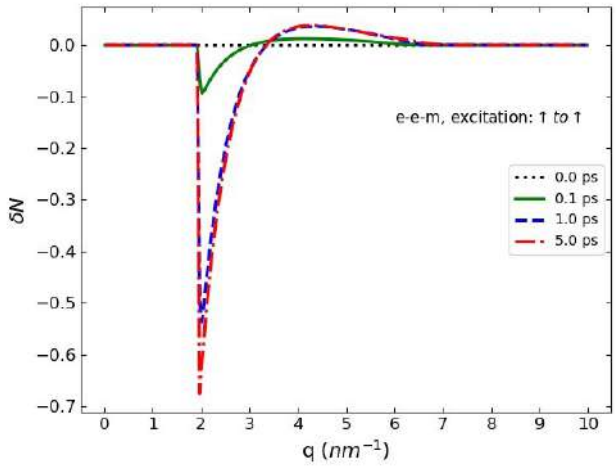



Figure: Change of magnon distribution $\delta N_q(t)$ due to electron-magnon and electron-electron scattering after excitation by instantaneous heating process. The region of $q$ values, for which electronic spin flip transitions are possible, starts around 2 nm$^{-1}$.

# Spin Polarized Hot Electron Induced Ultrafast Demagnetization dynamics in FeGd

*D. Gupta[1]*, M. Riepp[1], M. Hehn[2], N. Pontius[3], C. Schüßler-Langeheine[3], K. Holldack[3], N. Bergeard[1], C. Boeglin[1]*

[1] *Institut de Physique et Chimie des Matériaux de Strasbourg – UMR 7504, Université de Strasbourg, CNRS, 67000 Strasbourg, France*

[2] *Institut Jean Lamour, CNRS - Université de Lorraine, 54011 Nancy, France*

[3] *Instiut für Methoden und Instrumentierung der Forschung mit Synchrotronstrahlung, Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, Albert-Einstein-Strasse 15, 12489 Berlin, Germany*

*deeksha.gupta@ipcms.unistra.fr

The generation of spin currents is a key ingredient in spintronics-based memory devices such as spin transfer torque magnetic random-access memory (STT- MRAM). Since the discovery of ultrafast demagnetization [1], it is known that the magnetization in 3d ferromagnetic thin films can be quenched within ultrashort, sub-picosecond time scales using femtosecond (fs) laser pulses. Several studies have demonstrated the generation and propagation of ultrashort spin current pulses [2–4] associated with such femtosecond laser excitation, which is capable of inducing ultrafast demagnetization in an adjacent layer. However, these works are focused on ferromagnetic 3d transition metals, while recent studies suggest that antiferromagnetic materials (AFM) are more promising for magnetization control in ultrafast spintronics devices. The rare-earth transition-metal (RE-TM) alloy thin films, such as, FeGd are interesting for spintronic applications due to their antiferromagnetically coupled 3d and 4f sublattices and the possibility of manipulating the magnetization on ultrafast time scales in a controlled way [5].

To measure the dynamics of both 3d and 4f elements, we utilized the element selective time-resolved X-ray magnetic circular magnetic dichroism technique at the femtoslicing beamline at the BESSY synchrotron, Berlin. We performed pump-probe delay time scans in an external magnetic field using 60 fs IR laser pulses (800 nm) to pump the top layer of our sample and ~100 fs X-ray pulses, with photon energy tuned to resonance with the elemental core level excitations, i.e., the Fe L3 and Gd M5 absorption edges, to probe the dynamics in the bottom FeGd layer of our sample.

The sample used in this study has the following structure: FeGd/Cu/CoPt/Cu(60)/Pt spin valve structure on SiN membrane. Here, the pinned magnetic top layer CoPt/Cu (60)/Pt (6) has the purpose to generate the femtosecond spin current pulses which are then transported into the bottom FeGd thin layer.

To distinguish the effect of these fs spin currents from pure thermal demagnetization, we have performed and compared the dynamics for antiparallel and parallel alignment of the top Co/Pt and bottom Fe magnetization. We could evidence a notable acceleration of the demagnetization dynamics for the antiparallel alignment which is explained by a sizable contribution of spin-torque on sub-picosecond time scales.

## References

[1] E. Beaurepaire, J.-C. Merle, A. Daunois, and J.-Y. Bigot, *Ultrafast Spin Dynamics in Ferromagnetic Nickel*, Phys. Rev. Lett. **76**, 4250 (1996).

[2] M. Battiato, K. Carva, and P. M. Oppeneer, *Superdiffusive Spin Transport as a Mechanism of Ultrafast Demagnetization*, Phys. Rev. Lett. **105**, 027203 (2010).

[3] G. Malinowski, F. Dalla Longa, J. H. H. Rietjens, P. V. Paluskar, R. Huijink, H. J. M. Swagten, and B. Koopmans, *Control of Speed and Efficiency of Ultrafast Demagnetization by Direct Transfer of Spin Angular Momentum*, Nat. Phys. **4**, 855 (2008).

[4] C. Stamm et al., *X-Ray Detection of Ultrashort Spin Current Pulses in Synthetic Antiferromagnets*, J. Appl. Phys. **127**, 223902 (2020).

[5] I. Radu et al., *Transient Ferromagnetic-like State Mediating Ultrafast Reversal of Antiferromagnetically Coupled Spins*, Nature **472**, 205 (2011).

# Combining Tb and Co for Ultrafast Single-Shot All-Optical Switching in Ferrimagnetic Multilayers

*Julian Hintermayr[1]*, Pingzhi Li[1], Roy Rosenkamp[1], Youri van Hees[1], Junta Igarashi[2], Stéphane Mangin[2], Reinoud Lavrijsen[1], Grégory Malinowski[2], Bert Koopmans[1]*

[1] *Department of Applied Physics, Eindhoven University of Technology, P.O. Box 513, 5600 MB, Eindhoven, Netherlands*

[2] *Université de Lorraine, CNRS, IJL, F-54000 Nancy, France*

*j.hintermayr@tue.nl

All-optical switching (AOS) of magnetic bits by femtosecond laser pulses shows great promise for fast opto-spintronic memory devices [1]. The fastest (~1 ps) and most robust switching has been observed in ferrimagnetic materials containing the rare earth Gd. However, due to its electronic configuration, Gd-based materials are inherently soft, requiring large magnetic domains to store thermodynamically stable bits. This limits their suitability for high-density magnetic recording devices. More recently, Tb/Co-based multilayers with higher magnetic anisotropy have also been shown to be all-optically switchable [2]. Nevertheless, the reversal mechanism in this type of material is not understood yet, but evidence is growing that reversal takes place on much longer time scales (~100 ps) compared to systems with Gd only.

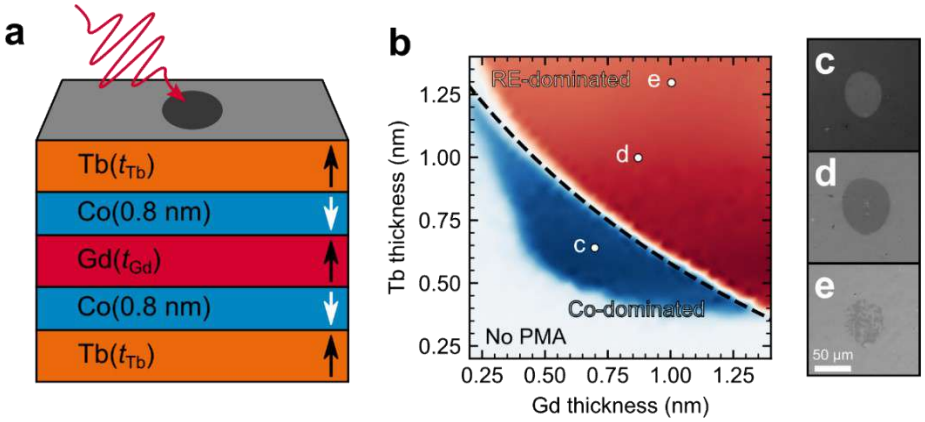



Figure 1: **a** Cartoon showing the magnetic layers within the stack with arrows indicating local spin orientations. **b** Phase diagram extracted from MOKE hysteresis loops for the sample design shown in **a**. **c, d, e** Kerr microscopy images of the magnetic state of the sample after excitation with a single fs laser pulse at the approximate thicknesses indicated in **b**.

In this work, we investigate synthetic ferrimagnetic multilayers containing both Gd and Tb (schematically shown in Fig. 1**a**) to better understand the role of the two rare-earth (RE) elements. We find perpendicular magnetic anisotropy and robust AOS in a broad range of RE thicknesses for Co and RE-dominated samples, as shown in Fig. 1 **b**. Interestingly, the critical laser fluence for AOS is independent of the amount of Tb in the system. Time-resolved MOKE measurements reveal that magnetization reversal takes place approximately within the first ps, as previously found in systems with only Co and Gd. Furthermore, we carry out Microscopic Three Temperature Model (M3TM) simulations, where – as suggested in [3] – we imitate the replacement of Gd by Tb as an increase in the spin-flip probability $a_{sf}$. We find a gradual suppression of AOS as $a_{sf}$ increases which corroborates our experimental observations.

We conclude that hybrid systems of Tb/Co and Co/Gd-based multilayers are promising candidates for future magneto-photonic memory devices.

**Acknowledgements**

This project has received funding from the European Union's Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement Nos. 861300 and 860060.

**References**

[1] A. V. Kimel, M. Li, *Nat Rev Mater* **4**, 189 (2019)
[2] L. Avilés-Felix et al., *Scientific Reports* **10**, 5211 (2020)
[3] A. Ceballos et al., *Phys. Rev. B* **103**, 024438 (2021)

# Laser induced ultrafast 4f spin dynamics at the surface of $Co_xGd_{100-x}$ ferrimagnetic alloys

M. Pacé[1], D. Gupta[1], T. Ferté[1], G. Malinowski[2], M. Hehn[2], M. Silly[3], F. Pressacco[3], F. Sirotti[4], C. Boeglin[1] and N. Bergeard[1]

[1] *Université de Strasbourg, CNRS, Institut de Physique et Chimie des Matériaux de Strasbourg, UMR 7504, F-67000 Strasbourg, France.*

[2] *Institut Jean Lamour, CNRS - Université de Lorraine, 54011 Nancy, France.*

[3] *Synchrotron SOLEIL, L'Orme des Merisiers, Saint-Aubin, 91192 Gif-sur-Yvette, France*

[4] *Physique de la Matière Condensée, Ecole Polytechnique, CNRS, 91128 Palaiseau, France*

The discovery of deterministic helicity-independent all-optical spin switching (HI-AOS) has driven intensive investigations on the laser induced ultrafast spin dynamics in ferrimagnetic rare-earth (RE) / transition metal (TM) alloys. Element- and time-resolved experiments have shown that the RE 4f and TM 3d spin dynamics occur on distinct time-scales [1], which is considered as a key ingredient for HI-AOS [2]. They have also shown that the characteristic times associate with these ultrafast dynamical processes depend on the temperature and on the alloy composition [3 - 8]. These seminal works call for further systematic investigations to establish the correlations between their static magnetic properties and their unusual laser induced ultrafast spin dynamics [9]. Such knowledge is of prime importance to identify the most appropriate materials for technological applications. For instance, previous works have revealed that amorphous RE-TM ferrimagnetic alloys show inhomogeneous in depth composition [10, 11] as well as RE segregation at interfaces [12, 13]. One can legitimately wonder whether the laser induced Gd 4f spin dynamics at the surface of the alloy is affected by these composition discrepancies in respect with that in the bulk [8]. Disparate spin dynamics at the surface and in the bulk would be detrimental regarding the downsizing of these magnetic layers to comply with the technological requirements for data storage devices.

In this work, we have investigated the laser induced ultrafast dynamics of Gd 4f spins at the surface of $Co_xGd_{100-x}$ alloys for bulk composition x = 80 and x = 65 by means of surface-sensitive time-resolved dichroic resonant Auger electron spectroscopy (TR-DRAES) [14, 15]. We have observed that the laser induced quenching of Gd 4f magnetic order at the surface of the CoGd alloys occur on a much longer time scale than that previously reported in bulk sensitive time-resolved experiments [3, 8]. We have characterized the electronic, structural and magnetic static properties at the surface and in the bulk of these alloys. These measurements have evidenced the accuracy of the bulk alloy compositions as well as the inhomogeneous composition at the surface of the alloys. Indeed, they display a strongly increased Gd content within the first layers [10 - 13]. As a consequence, we infer that the larger Gd concentration in these top layers compared to that of the nominal bulk composition allows to explain the “slower” Gd 4f demagnetization we have observed in our surface-sensitive and time-resolved measurements compared to that previously reported by “bulk-sensitive” measurements [8].

**References:**

[1] Radu et al. Nature. 472, 205 (2011)
[2] Iihama et al. Advanced Materials 30, 1804004 (2018)
[3] López-Flores et al. Phys. Rev. B 87, 214412 (2013)
[4] Ferté et al. Phys. Rev. B 96, 134303 (2017)
[5] Chen et al. New J. Phys. 21, 123007 (2019)
[6] Ren et al. ArXiv. 2012.14620v1 (2020)
[7] Ferté et al. J. Magn. Mag. Mat. 530, 167883 (2021)
[8] Ferté et al. ArXiv, 2206.10422 (2022)
[9] Atxitia et al. Phys. Rev. B 89, 224421 (2014)
[10] Hebler et al. Front. Mater. 3, 8 (2016)
[11] Haltz et al. Phys. Rev. Mat. 2, 104410 (2018)
[12] Shen et al. Jap. J. Appl. Phys. 20, L757 (1981)
[13] Bergeard et al. Phys. Rev. B 96, 064418 (2017)
[14] Beaulieu et al. Journal of Elec. Spec. 189, 40 (2013)
[15] Silly et al. Journal of synchr. Rad. 24, 886 (2017)

# Indirect optical manipulation of the antiferromagnetic order of insulating NiO by ultrafast energy transfer

*Paul Herrgen[1]*, Stephan Wust[1], Christopher Seibel[1], Hendrik Meer[2], Christin Schmitt[2], Lorenzo Baldrati[2], Rafael Ramos[3,4], Takashi Kikkawa[5], Eiji Saitoh[5,6,4,7], Olena Gomonay[2], Jairo Sinova[2,8], Yuriy Mokrousov[2,9], Hans Christian Schneider[1], Mathias Kläui[2], Bärbel Rethfeld[1], Benjamin Stadtmüller[1,2], Martin Aeschlimann[1]*

*[1] Department of Physics and Research Center, OPTIMAS, RPTU Kaiserslautern-Landau, 67663 Kaiserslautern, Germany*

*[2] Institute of Physics, Johannes Gutenberg-University Mainz, 55099 Mainz, Germany*

*[3]CIQUS, Departamento de Quımica-Fısica, Universidade de Santiago de Compostela, Santiago de Compostela, Spain*

*[4]WPI Advanced Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan*

*[5]Department of Applied Physics, The University of Tokyo, Tokyo 113-8656, Japan*

*[6]Institute for AI and Beyond, The University of Tokyo, Tokyo 113-8656, Japan*

*[7]Advanced Science Research Center, Japan Atomic Energy Agency, Tokai 319-1195, Japan*

*[8]Institute of Physics, Czech Academy of Sciences, Cukrovarnicka 10, 162 00 Praha 6, Czech Republic*

*[9]Peter Grünberg Institut and Institute for Advanced Simulation, Forschungszentrum Jülich and JARA, 52425 Jülich, Germany*

*pherrgen@rptu.de

Antiferromagnets are promising materials for future spintronic devices due to their resilience against external magnetic fields and their high frequency magnon modes up to the THz regime [1,2]. A very promising candidate is the antiferromagnetic insulator nickel oxid (NiO) with a Néel temperature of 523K that is significantly larger than room temperature [3]. Here we study the ultrafast dynamics of antiferromagnetic crystalline NiO thin films covered with a thin Pt layer after strong optical excitations. This is done by using magneto-optical birefringence in combination with a pump probe setup. Here, we have chosen pump pulses with a photon energy well below the band gap of NiO. For these excitation conditions, we uncover a fast reduction of the magnetic order parameter of NiO that is very similar to the ultrafast demagnetization of ferromagnets. Interestingly, no reduction in the magnetic order of NiO is observed for NiO films without the Pt capping layer. We conclude that the ultrafast loss of antiferromagnetic order in the Pt/NiO bilayer system is mediated by an enhanced energy transfer between the hot Pt electrons and the NiO spin system across the interface, in combination with a higher optical absorption in Pt for the used pump photon energy. To verify this results we included an additional layer of MgO between the NiO and the Pt to completely prevent the hot Pt electrons from interacting with the NiO spins. Here, the magnetic order stays constant on ultrafast timescale, but a slow and small reduction is still visible, which is due to the energy transfer via the phonon system [4].

In this way, our work lays the foundation for the ultrafast manipulation of the magnetic order of insulating and semiconducting (antiferromagnetic) materials by interface engineering and optical excitation below the optical bandgap.

## References

[1] V. Baltz, A. Manchon, M. Tsoi, T. Moriyama, T. Ono, Y. Tserkovnyak, Rev. Mod. Phys. 90, 015005 (2018)

[2] K. Olejník, T. Seifert, Z. Kašpar, V. Novák, P. Wadley, R. P. Campion, M. Baumgartner, P. Gambardella, P. Němec, J. Wunderlich, J. Sinova, P. Kužel, M. Müller, T. Kampfrath, T. Jungwirth, Sci. Adv. 4, eaar3566 (2018)

[3] W. L. Roth, J. Appl. Phys. 31, 2000 (1960)

[4] S. Wust, C. Seibel, H. Meer, P. Herrgen, C. Schmitt, L. Baldrati, R. Ramos, T. Kikkawa, E. Saitoh, O. Gomonay, J. Sinova, Y. Mokrousov, H.C. Schneider, M. Kläui, B. Rethfeld, B. Stadtmüller, M. Aeschlimann, arXiv:2205.02686 (2022)

# Multilayer 'Beyond-2D' Artificial Spin Ice Arrays for Magnonic Processing

*Jack N. Carter-Gartside*

*Imperial College, London, United Kingdom*

j.carter-gartside13@imperial.ac.uk

In this talk we'll examine the benefits of moving nanomagnetic arrays from quasi-2D, single magnetic layer systems into spin system architectures with strongly-coupled magnetic elements across all three dimensions.

As demonstrated by fantastic recent work by groups including Sam Ladak's 3D nanoarrays1 & Claire Donnelly's stunning 3D magnetic imaging2, many unexpected & exciting phenomena appear when moving beyond 2D systems. Measurement & fabrication techniques have sufficiently matured to take full advantage & the field stands at an exciting point.

While some of the experimental techniques required to accomplish fully 3D nanomagnetism are somewhat esoteric or challenging to master, benefits of leaving the 2D regime remain accessible via more traditional approaches.

Here we present a 'Beyond-2D' addition to the growing family of artificial spin systems, taking inspiration from synthetic antiferromagnets & strongly-coupling multiple layers of artificial spin ice to produce a complex dynamic system, surprisingly enriched relative to single-layer 2D arrays.

We will explore leveraging these behaviours for exploring fundamental physical phenomena such as reconfigurable activation of hybridised acoustic & optical magnon modes, magnetic vortex chirality control & functional processing including neuromorphic computing3,4.

[1] May, Andrew, et al. "Magnetic charge propagation upon a 3D artificial spin-ice." *Nature communications* 12.1 (2021): 3217.

[2] Donnelly, Claire, et al. "Time-resolved imaging of three-dimensional nanoscale magnetization dynamics." *Nature nanotechnology* 15.5 (2020): 356-360.

[3] Gartside, Jack C., et al. "Reconfigurable Training and Reservoir Computing via Spin-Wave Fingerprinting in an Artificial Spin-Vortex Ice." Nature Nanotechnology (2022)

[4] Stenning, Kilian D., Gartside, Jack C. et al. "Adaptive programmable networks for in materia neuromorphic computing." arXiv preprint arXiv:2211.06373 (2022).

# Magnon Currents Excited by the Spin Seebeck Effect in Ferromagnetic EuS Thin Films

*M. Xochitl Aguilar-Pujol[1]*, Sara Catalano[1], Carmen González-Orellana[2], Witold Skowronski[1,3], Juan M. Gómez-Pérez[1], Maxim Ilyn[2], Celia Rogero[2], Marco Gobbi[1,2,4], Luis E. Hueso[1,4] and Fèlix Casanova[1,4]*

[1] *CIC NanoGUNE BRTA, 20018 San Sebastian, Basque Country, Spain*
[2] *Centro de Física de Materiales CSIC-UPV/EHU, 20018 San Sebastian, Basque Country, Spain*
[3] *AGH University of Science and Technology, Institute of Electronics, 30-059 Krakow, Poland*
[4] *IKERBASQUE, Basque Foundation for Science, 48009 Bilbao, Basque Country, Spain*
*m.aguilar@nanogune.eu

Magnons, the collective bosonic excitations in magnetically ordered systems, allow for the transport of spin angular momentum even in insulators. The ability to control de generation, propagation, and detection of such magnon spin currents represents an asset for the progress of spintronics. However, until now, most studies have focused on $Y_3Fe_5O_{12}$ (YIG) and a few other ferri- and antiferromagnetic insulators [1], but not on pure ferromagnets. Here, we demonstrate for the first time the generation of thermal magnon curents in ferromagnetic insulator europium sulphide (EuS) [2], which exhibits a Curie temperature $T_C$ = 19 K in thin films [3]. We perform both local (LOC) and non-local (NL) transport measurements in 18-nm-thick films of EuS using Pt electrodes, with different separation distances d, as magnon injectors and detectors. We study the voltage response generated at the detector due to the inverse spin Hall effect, given that spin angular momentum is transferred through the Pt/EuS interface. We identify the magnon induced spin currents by measuring the dependence of the second harmonic component of this voltage on the in-plane angle (α) applying an external magnetc field ***B*** that orients the EuS magnetization ***M*** and determines the spin currents spin polarization. The observed LOC and NL signals indicate that magnon currents can be excited thermally by the spin Seebeck effect. By comparing the dependence of the LOC and NL singals with the temperature (2 K < $T$ < 30 K ) and magnetic field (< 9 T), we confirm the magnon transport origin of the NL signal. Finally, we extract the magnon diffusion length in the EuS films (~ 140 nm), a short value in good correspondende with the large Gilbert damping measured by ferromagnetic resonance in the same films [2].

**Acknowledgements**

We acknowledge funding by the Spanish MICINN (project No. PID2021-122511OB-I00 and Maria de Maeztu Units of Excellence Programme No. CEX2020-001038-M) and by the European Union H2020 Programme (Projects No. 965046-INTERFAST and 964396-SINFONIA). M.X.A.-P. thanks the Spanish MICINN for a Ph.D. fellowship (grant No. PRE-2019-089833). S.C. acknowledges support from the European Commission for a Marie Sklodowska-Curie individual fellowship (Grant No. 796817-ARTEMIS). W.S. acknowledges financial support from Polish National Agency for Academic Exchange (PPN/BEK/2020/1/00118/DEC/1).

**References**

[1] A. Brataas, B. van Wees, O. Klein, G. de Loubens, M. Viret, *Phys. Rep.* 885 (2020), 1-27.
[2] M. X. Aguilar-Pujol et al., *arXiv:2303.03833*.
[3] J. M. Gómez-Pérez, et al., *Nano Lett.* 20 (2020) 6815-6823.

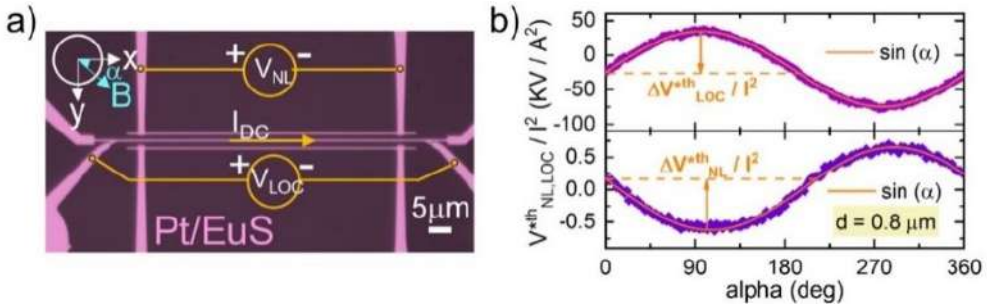



Figure: a) Optical microscope image of a device showing the measurement configuration. B) Representative LOC and NL angular dependent signal detected for thermally excited magnons at 2 K and 0.3 T.

# Magnonic Crystals Based on Stripe-Domain Pattern

*K. Szulc[1]*, S. Tacchi[2], J. Díaz[3,4], A. Hierro-Rodríguez[3,4], P. Gruszecki[1], P. Graczyk[5], C. Quirós[3,4], D. Markó[6], V. V. Fernández[3,4], M. Vélez[3,4], D. S. Schmool[6], G. Carlotti[7], M. Krawczyk[1], L. M. Alvarez-Prado[3,4]*

[1] *ISQI, Faculty of Physics, Adam Mickiewicz University, 61-614 Poznań, Poland*
[2] *Istituto Officina dei Materiali del CNR (CNR-IOM), Sede Secondaria di Perugia, I-06123 Perugia, Italy*
[3] *Departamento de Fisica, Facultad de Ciencias, Universidad de Oviedo, 33007 Oviedo, Spain*
[4] *Centro de Investigacion en Nanomateriales y Nanotecnologia (CINN), 33940 El Entrego, Spain*
[5] *Institute of Molecular Physics, Polish Academy of Sciences, 60-179 Poznań, Poland*
[6] *Université Paris-Saclay, UVSQ, CNRS, GEMaC, 78000 Versailles, France*
[7] *Dipartimento di Fisica e Geologia, Università di Perugia, I-06123 Perugia, Italy*
*krzysztof.szulc@amu.edu.pl

Magnetic thin films having weak perpendicular magnetic anisotropy (PMA), develop well-ordered stripe domains patterns [1], which can act as magnonic crystals [2]. However, strong damping of the materials with PMA significantly hinders the use of the stripe-domain systems in magnonic applications.

We present a hybrid system consisting of 64-nm-thick NdCo layer having weak PMA and 10-nm-thick low-damping soft magnetic layer (NiFe, NiFe/Co, Co/NiFe) separated by Al spacer [1,3]. Thanks to dipolar interaction between the layers, the stripe domains of NdCo film are imprinted in the soft magnetic layer (Figure 1a). Hysteresis loops measured with vibrating sample magnetometer show that using a small external magnetic field of 20-30 mT allows reversing magnetization only in soft film without changing the domain structure of NdCo layer. Interestingly, this process is fully reversible making the system reconfigurable. Spin-wave dynamics in the direction perpendicular to stripe domains were measured using Brillouin light scattering (BLS) spectroscopy. The experimental results show an asymmetric dispersion relation which differs strongly between the configurations of parallel and antiparallel orientation of magnetization in NdCo and soft layers (Figure 1b,c). Numerical simulations show that the source of the nonreciprocity lies in static magnetization configuration as well as from the interaction via dynamic stray field induced by the spin waves. Moreover, they indicate that the BLS spectra contain the fundamental soft layer mode branches coming from different Brillouin zones, pointing out a typical feature of a magnonic crystal.

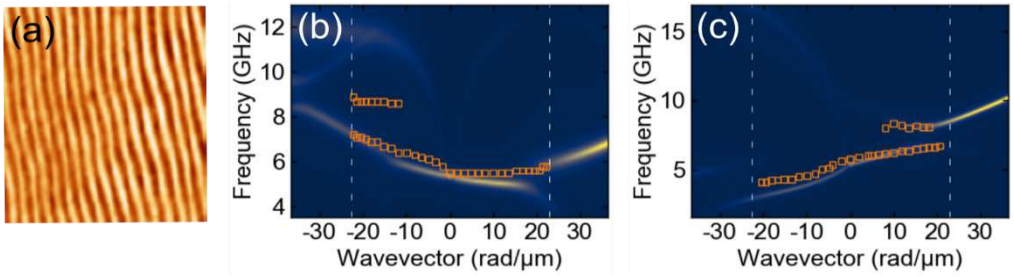



Figure 1: (a) MFM image of the stripe domains in the NdCo/Al/NiFe multilayer. Dispersion relation in the NdCo/Al/NiFe in zero external magnetic field for (b) parallel and (c) antiparallel magnetization orientation between NiFe and NdCo layer where color map represents the results of numerical simulations and orange squares—BLS results.

## Acknowledgements

This study has received financial support from the National Science Centre, Poland (projects nos. UMO-2018/30/Q/ST3/00416, UMO-2020/39/I/ST3/02413, and UMO-2021/41/N/ST3/04478), Spanish Ministry of Science and Innovation (Project No. PID2019-104604RB/AEI/10.13039/501100011033), the regional FICYT funding (Grant No. GRUPIN-AYUD/2021/51185) with the support of FEDER funds, and the European Union's Horizon 2020 research and innovation programme under grant agreement No 101007417. LMAP and ST acknowledge Italian CNR for financial support from Short Term Mobility Program.

## References

[1] D. Markó et al., *Appl. Phys. Lett.* **115** (2019) 082401.
[2] M. Krawczyk, D. Grundler, *J. Phys.: Condens. Matter* **26** (2014) 123202.
[3] K. Szulc et al., *ACS Nano* **16** (2022) 14168–14177.

# All-optical spin injection in silicon revealed by element-specific time-resolved Kerr effect

*S. Laterza[1,2]*, A. Caretta[1], R. Bhardwaj[1], and M. Malvestuto[1]*

[1] *Elettra Sincrotrone Trieste S.C.p.A., Strada Statale 14 - km 163.5 in AREA Science Park 34149 Basovizza, Trieste, Italy*

[2] *Department of Physics, University of Trieste, Via A. Valerio 2, 34127 Trieste, Italy*

*simone.laterza@elettra.eu

Spintronics aims at controlling both the charge and the spin carriers in solid-state devices. Semiconductor spintronics promises enhanced functionalities in terms of speed and energy consumption. The most promising mechanism to achieve spin currents in semiconductors is to inject spin-polarized hot electrons from a ferromagnetic film into the semiconductor substrate. Superdiffusive spin currents can be generated by the absorption of an ultrafast IR pulse by a ferromagnetic film. The excited electrons diffuse then ballistically inside the film but, as velocities and scattering times are bigger for spin majority electrons than spin minority ones, the excited electrons propagate then through interfaces as spin currents.

Silicon is an ideal material for spintronic applications due to both its role in the traditional electronics industry and its allowing long-lived spin currents. Nonetheless, the injection of spin currents in semiconductors still lacks direct evidence. To provide experimental evidence of spin injection, we studied a Ni/$Si_3N_4$/Si interface [1] using the time-resolved resonant MOKE effect [2] both at the Ni $M_{2;3}$ and at the Si $L_{2;3}$ edges. The measurements were carried out at the MagneDyn beamline [3] at the externally seeded FERMI free-electron laser.

## References

[1] S. Laterza, et al., Optica 9, 1333-1338 (2022)

[2] A. Caretta, *et al.*, Struct. Dynam. 8, 034304 (2021)

[3] M. Malvestuto *et al.*, Rev. Sci. Instrum. 93, 115109 (2022)

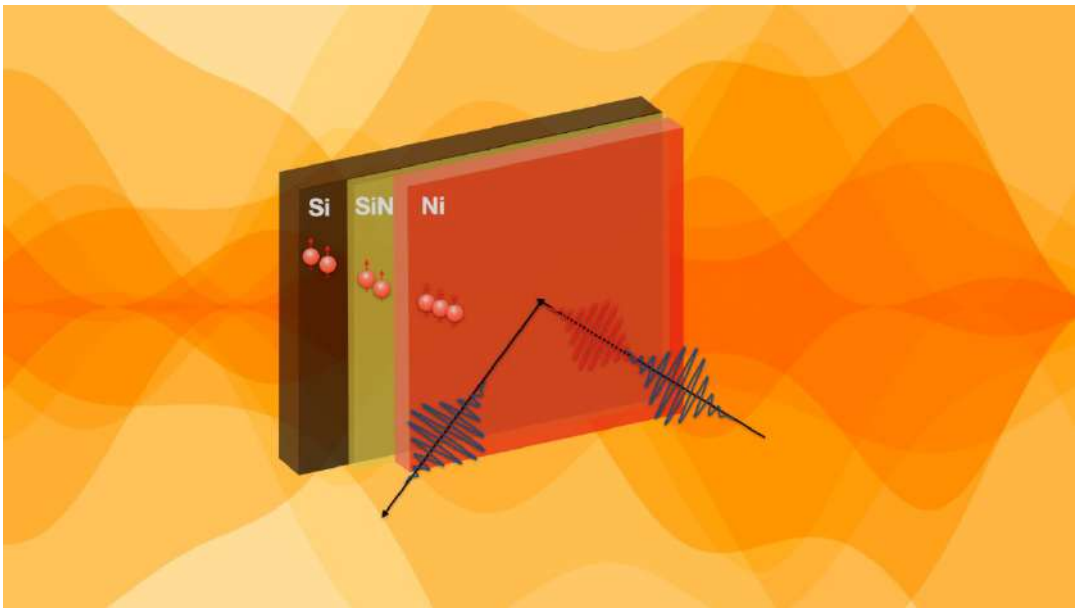



Figure 1: Pictorial representation of IR pump pulses (red wavelet) and EUV probe pulses (blue wavelets) impinging on the interface. The sample consists of a Si substrate (brown solid) passivated by an interlayer of crystalline Si3N4 (yellow solid) above which 7 nm of polycrystalline Ni have been deposited (red solid). The optical pump pulse excites spin-polarized electrons (red spheres) that are then injected in the substrate as a superdiffusive spin current.

# Ultrafast electron-phonon scattering in antiferromagnetic Dirac-semimetal

*Marius Weber[1]*, Kai Leckron[1], Baerbel Rethfeld[1], Libor Šmejkal[2], Jairo Sinova[2], Hans Christian Schneider[1]*

[1] *Department of Physics and Research Center OPTIMAS, University of Kaiserslautern-Landau, Kaiserslautern, Germany*

[2] *Institute of Physics, Johannes Gutenberg University Mainz, Mainz, Germany*

*mweber@rhrk.uni-kl.de

Systems with antiferromagnetic ordering in real space often exhibit topological features in momentum space that only recently have been investigated in a comprehensive fashion [1]. We consider here an AFM model band structure with two-dimensional *k*-space and a pronounced anisotropy in the band/spin structure of their single-particle states. For this 4-band model, which captures key properties of antiferromagnetic Dirac materials in momentum space [1], we present numerical calculations of the electron dynamics due to electron-phonon interactions [2,3]. For the excitation we implement an instantaneous heating process that simulates electronic distributions created by an ultrashort optical pulse. The dynamics after the spin-polarized excitation give rise to transient carrier distributions that are strongly influenced by the momentum-space features. A pronounced example is the occurrence of carrier distributions $f(\boldsymbol{k})$ that do not respect the symmetry of the energy landscape, even if one starts from an initial state that is essentially determined by the *k* dependence of the dispersion, i.e., $f(E(\boldsymbol{k})) = f(E)$, as shown in Figure 1. We can attribute this to the influence of band structure features on the scattering transitions. The spin-polarized excitation leads to non-trivial spin dynamics which will be analyzed in terms of the *k*-dependent site/orbital-projected spin expectation values.

## Acknowledgements

Funded by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) – TRR 173 – 268565370 (projects A08 and B03)

## References

[1] L. Šmejkal, J. Železný, J. Sinova, and T. Jungwirth; *Phys. Rev. Lett.* **118**, 106402 (2017)
[2] K. Leckron, S. Vollmar, and H. C. Schneider; *Phys. Rev. B* **96**, 140408 (2017).
[3] S. Essert and H. C. Schneider, *Phys. Rev. B* **84**, 224405 (2011).

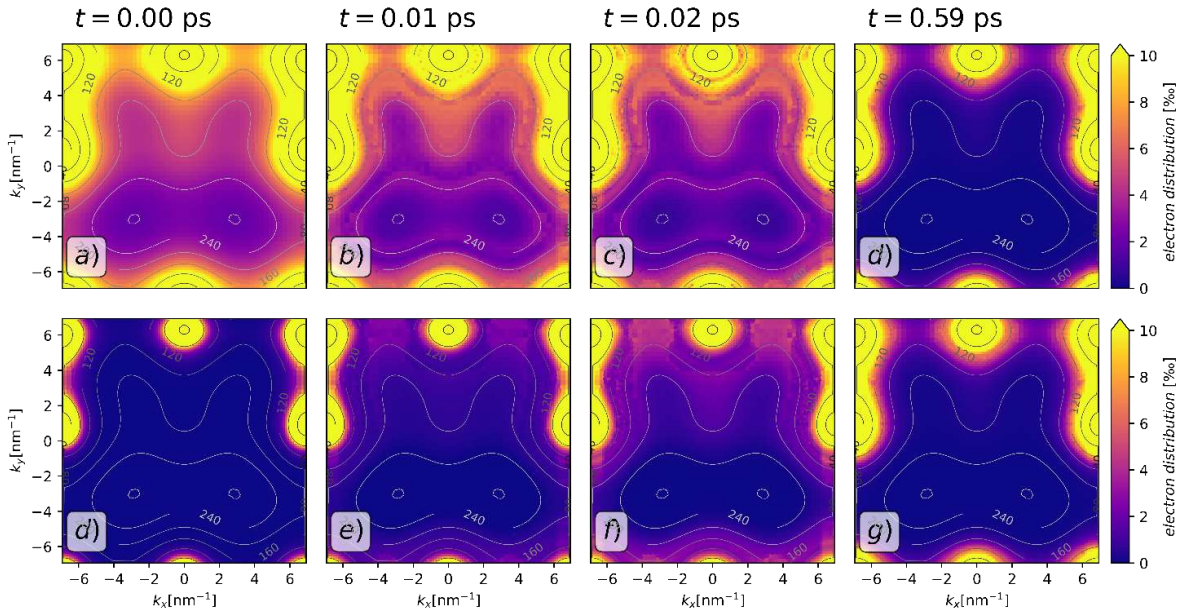



Figure 1: **k**-resolved occupation dynamics in the two upper bands, labelled as +/-, for different times from left to right. Top panel: (a) instantaneous excitation in the "+"-band, (b) shortly after excitation, (c) degenerate bands are approximately in quasiequilibrium and (d) relaxation within the upper bands. Lower panel: Corresponding dynamics in the "-" band.

# Generation of femtosecond spin-current pulses at Fe/MgO interface by quasi-static voltage

*Piotr Graczyk[1]*, Maria Pugaczowa-Michalska[1], Maciej Krawczyk[2]*

[1] *Institute of Molecular Physics, Polish Academy of Sciences, Poznań, Poland*

[2] *Faculty of Physics, Adam Mickiewicz University, Poznań, Poland*

*graczyk@ifmpan.poznan.pl

The generation of short spin-current pulses is essential for fast spintronic devices. So far, spin current pulses are generated by femtosecond laser pulses which drive spins from a ferromagnetic metal layer. This transient spin current may be used to emit terahertz electromagnetic pulse through inverse spin-Hall effect [1]. However, the need for miniaturization, simplicity and energy efficiency favours electric field control of spintronic devices over optic control. We demonstrate theoretically that the voltage-driven instability of the electronic structure at the Fe/MgO interface results in the generation of the femtosecond spin-current pulse.

We show by numeric simulations that spin-dependent screening at dielectric-ferromagnetic metal interface contributes to the spin-polarized current generation in the system subjected to the ac voltage [2]. Then, we show that spin current driven by spin-dependent screening may be used to modulate spin-wave amplitude in bilayer ferromagnetic system [3]. Finally, we combine *ab initio* calculations of electronic density of states at MgO/Fe interface with continuous model for charge transport. We show that the voltage-driven electron charge accumulation at MgO/Fe interface leads to the Stoner instability because of the electronic interface resonant states. This instability manifests itself in the spin-current and spin accumulation femtosecond pulses which are present because of the contribution of the dynamic spin-dependent potential to the spin-polarized current.

## Acknowledgements

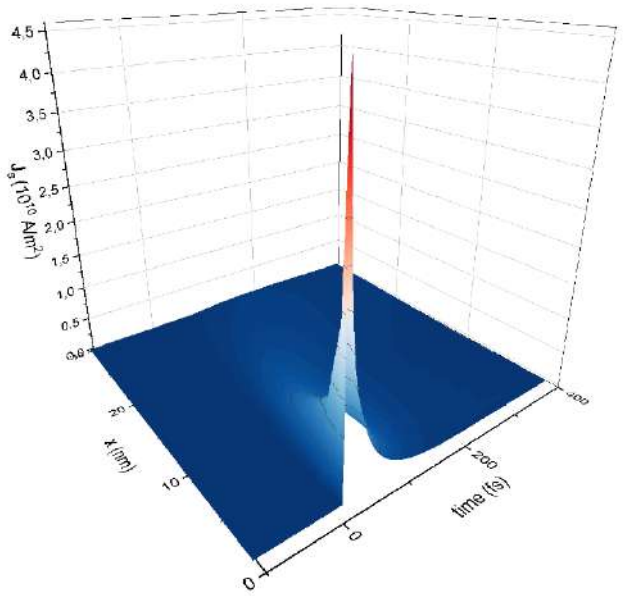



Fig. 1. The spin-current pulse in dependence on time and the distance from the Fe/MgO interface.

This work was conducted under grant no. 2018/28/C/ST3/00052 from National Science Centre in Poland.

## References

[1] T. Kampfrath *et al.*, *Nature Nanotech*, vol. 8, no. 4, Art. no. 4 2013

[2] P. Graczyk and M. Krawczyk, *Phys. Rev. B*, vol. 100, no. 19, p. 195415, 2019

[3] P. Graczyk and M. Krawczyk, *Sci. Rep.*, vol. 11, 15692, 2021

# Ultrafast Nonequilibrium Magnetization Dynamics in Heterogeneous Materials

*Christopher Seibel*[1]*, *Tobias Held*[1], *Sebastian T. Weber*[1], *Marius Weber*[1], *Martin Stiehl*[1], *Martin Aeschlimann*[1], *Hans Christian Schneider*[1], *Benjamin Stadtmüller*[1,2], *and Baerbel Rethfeld*[1]

[1] *Department of Physics and Research Center OPTIMAS, RPTU Kaiserslautern-Landau, 67663 Kaiserslautern, Germany*

[2] *Institute of Physics, Johannes Gutenberg-University Mainz, 55099 Mainz, Germany*

*cseibel@rptu.de

Ultrafast magnetization dynamics play a key role in the development of spintronic devices. The dynamics are influenced by the composition of material systems as well as by the wavelength of the optical excitation. The latter can lead to spatially inhomogeneous excitation profiles in thick nickel films [1,2] or Ni|Au heterostructures [3]. We calculate the magnetization dynamics for the Ni|Au heterostructure and compare our results to C-MOKE measurements [3]. Figure 1 shows that we can clearly reproduce the experimental trend, which we explain by a wavelength-dependent energy transfer between the layers. We attribute the remaining minor deviations between theory and experiment to effects not accounted for by the model, such as for instance transport, dynamic material properties, and nonequilibrium.

In this contribution, we investigate the influence of the nonequilibrium within and between individual subsystems created by the laser excitation using complete Boltzmann collision integrals. In addition, we present results for transient coupling parameters, such as the spin-resolved electron-phonon coupling, which themselves depend on the nonequilibrium state of the sample. Our results show that nonequilibrium magnetization dynamics and material parameters mutually influence each other and thus can hinder or enhance the interplay of relaxation processes, which influence ultrafast magnetization dynamics and transport in heterogeneous materials.

## References

[1] U. Bierbrauer *et al.*, *JOP: Cond. Mat.* **29** (2017) 244002.
[2] S. Ashok *et al.*, *Appl. Phys. Let.* **120** (2022) 142402.
[3] C. Seibel *et al.*, *Phys. Rev. B* **106** (2022) L140405.

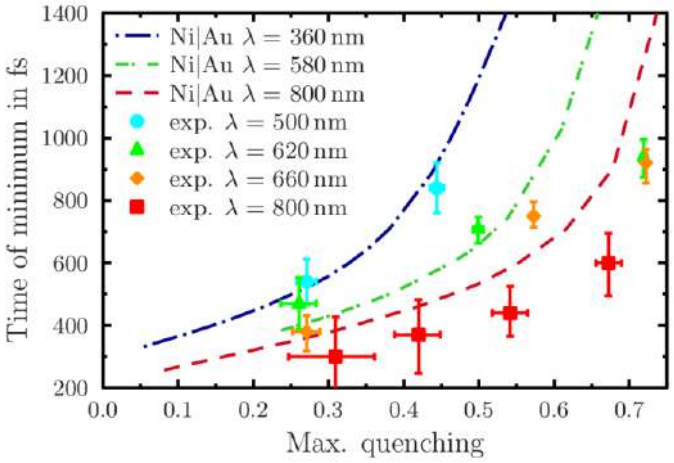



Figure 1: Comparison of calculated quenching times with experimental values for various wavelengths. Taken from Ref. [3].

# Observation of Spin Voltage and -Accumulation by Spin Resolved Femtosecond Photoelectron Spectroscopy

*Kevin Bühlmann[1], Francisco Carrión[1], Grégoire Saerens[2], Andreas Vaterlaus[1], Yves Acremann[1]**

[1] *ETH Zurich, Lab. For Solid State Physics, 8093 Zurich, Switzerland*
[2] *ETH Zurich, Institute for Quantum Electronics, 8093 Zurich, Switzerland*
*acremann@solid.phys.ethz.ch

The generation of spin current pulses by laser-driven demagnetization [1,2] links the field of ultrafast magnetism to spintronics. So far, this spin transport and its cause could only be observed indirectly. We demonstrate that femtosecond spin injection can be observed on the femtosecond time scale by spin and time resolved photoemission experiments.

We study thin, epitaxial iron films which are excited by the 800 nm pump laser beam. Photoemission by a higher harmonic source (photon energy: 21 eV) and spin resolved spectroscopy of the photo electrons leads to the measurement of the chemical potentials of the minority- and majority spins. This way, we observe the driving force of the generated spin current, which is the difference of the spin-split chemical potentials [3,4].

If we deposit a thin gold film onto an iron sample and excite the iron film through a transparent substrate, we can study spin injection. The spins which are injected from the iron layer into gold can be observed as a function of time. The spin polarization in Au rises on the femtosecond time scale and decays within < 1ps [5]. The decay time depends on the Au film thickness: in thicker Au films the spin polarization decays slower. This thickness dependence shows, that the loss of polarization is partially caused by spin transport as well as local spin flips. We can model the transport aspect by using the discharge of a "spin capacitance" postulated by Zhu et al. [6]. This spin capacitance is only visible in dynamic transport experiments. This is similar to the situation in charge-based electronics where a capacitance is only relevant if we consider dynamics.

**Acknowledgements**

This work was supported by ETH Zurich.

**References**

[1] E. Beaurepaire, J.-C. Merle, A. Daunois, J.-Y. Bigot Phys. Rev. Lett. 76, 4250 (1996)
[2] M. Battiato, K. Carva, P.M. Oppeneer, Phys. Rev. Lett. 105, 027203 (2010)
[3] A Fognini, T.U. Michlmayr, A. Vaterlaus, Y. Acremann , J. Phys.: Condens. Matter 29 214002 (2017)
[4] K. Bühlmann, G. Saerens, A. Vaterlaus, Y. Acremann, Structural Dynamics 7, 065101 (2020)
[5] K. Bühlmann, G. Saerens, A. Vaterlaus, Y. Acremann, Scientific Reports 10, 12632 (2020)
[6] Y.H. Zhu, X.X. Zhang, J. Liu, P.S. He, J. Appl. Phys. 122, 043902 (2017)

# Probing Magnons in Ferrimagnetic Garnet Thin Films: Effects of Magnetic Anisotropy, Compensation Temperature, and Gilbert Damping

*Christian Holzmann[1]*, Amit Chanda[2], Manh-Huong Phan[2], Hari Srikanth[2], Manfred Albrecht[1]*

[1] *Institute of Physics, University of Augsburg, Universitätsstraße 1, 86159 Augsburg, Germany*

[2] *Departement of Physics, University of South Florida, Tampa, Florida 33620, USA*

*christian.holzmann@physik.uni-augsburg.de

Single crystal garnets are known for their ultra-low Gilbert damping, which makes them the prime choice to study magnonics for future data manipulation, transmission, or storage devices [1]. While it is well known that yttrium iron garnet (YIG) possesses the lowest spin wave damping, the precise dependence of the magnon propagation length, a key parameter for future device design, on the garnet properties like the spin wave damping, magnetic anisotropy, or a magnetic compensation temperature is of great interest.

In this regard, the magnon propagation in garnet thin films is studied via the longitudinal spin Seebeck effect (SSE). Thermal magnons are excited by a temperature gradient perpendicular to the film plane, and converted to a measurable charge current in an adjacent Pt layer via the inverse spin Hall effect.

Gadolinium iron garnet (GdIG), thulium iron garnet (TmIG), and YIG thin films in the range of 10-250 nm thickness were prepared by pulsed laser deposition, and a Pt strip was ex-situ sputter-deposited (Fig. 1a). Epitaxial growth of garnet thin films on substrates with a slight lattice mismatch can introduce a perpendicular magnetoelastic anisotropy, allowing control of the magnetic anisotropy of the thin films by lattice mismatch (Fig. 1b) as well as strain relaxation for increasing film thickness [2,3]. Further, GdIG possesses a thickness-dependent magnetic compensation point between 200-300 K, which can cause a spin reorientation transition from an out-of-plane to an in-plane magnetic easy axis with decreasing temperature [2]. By combining measurements on three different garnets, while varying film thickness and lattice mismatch, the influence of magnetic anisotropy, Gilbert damping, and temperature on magnon excitation and propagation are studied.

The SSE measurements show a strong dependence on the magnetic anisotropy of the films, which is independently confirmed by SQUID and polar MOKE magnetometry as well as RF transverse susceptibility measurements [4,5]. Hereby, the magnon propagation length shows an inverse proportionality to the effective anisotropy field, as well as to the Gilbert damping parameter, which is measured by broadband ferromagnetic resonance. Further, a polarity change of the SSE signal is observed when crossing the magnetic compensation point (Fig. 1c), independent of other film parameters. These results hint at the presence of magnon modes with greatly varying propagation lengths. Understanding the dependence of the magnon propagation length on the magnetic properties of thin film systems is an important step for the design of future devices.

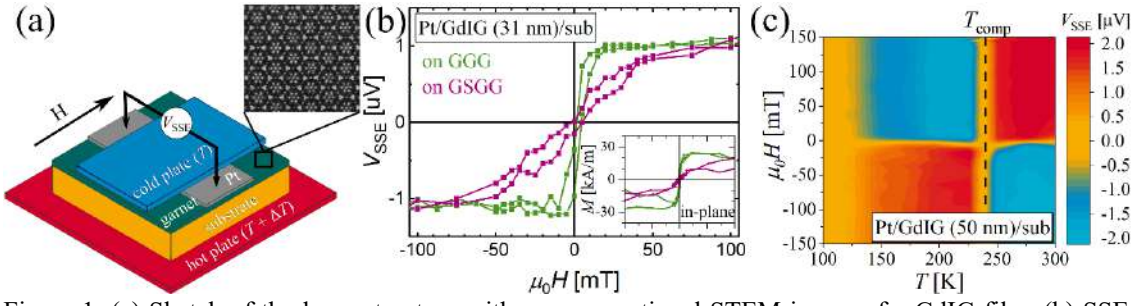



Figure 1: (a) Sketch of the layer structure with a cross-sectional STEM image of a GdIG film. (b) SSE and SQUID-VSM (inset) hysteresis loops for a 30 nm thick GdIG film grown on GGG ($Gd_3Ga_5O_{12}$) and GSGG ($Gd_3Sc_2Ga_3O_{12}$) with an applied in-plane field. (c) *H-T* SSE phase map of a 50 nm thick GdIG film.

## References

[1] Schmidt, G., Hauser, C., Trempler, P., Paleschke, M., Papaioannou, E.T., Phys. Stat. Solidi B. **257** 1900644 (2020).
[2] Holzmann, C., Ullrich, A., Ciubotariu, O., and Albrecht, M, ACS Appl. Nano Mater. **5** 1023 (2022).
[3] Ciubotariu, O., Semisalova, A., Lenz, K., and Albrecht, M., Sci. Rep.. **9** 17474 (2019).
[4] Chanda, A., Holzmann, C., Schulz, N., Seyd, J., Albrecht, M., Phan, M.-H., and Srikanth, H., Adv. Funct. Mater. **32** 2109170 (2022).
[5] Chanda, A., Schulz, N., Holzmann, C., Seyd, J., Albrecht, M., Phan, M.-H., and Srikanth, H., IEEE Trans. Magn. **58** 1500505 (2022).

# Imaging and Phase-locking of non-linear Spin-Wave Phenomena

*Rouven Dreyer[1]*, Alexander F. Schäffer[1], Hans G. Bauer[2], Niklas Liebing[1], Jamal Berakdar[1], and Georg Woltersdorf[1,3]*

[1] *Martin Luther University Halle-Wittenberg, Insitute of Physics, Von-Danckelmann-Platz 3, 06120 Halle (Saale), Germany*

[2] *Jahnstrasse 23, 96050, Bamberg, Germany*

[3] *Max Planck Institute of Microstructure Physics, Weinberg 2, 06120 Halle (Saale), Germany*

*rouven.dreyer@physik.uni-halle.de

Non-linear phenomena are a key feature in the emerging field of spin-wave based information processing since they allow to convert uniform spin-wave excitations into propagating modes at different frequencies. Over the last decades, non-linear processes such as three or four magnon scattering have been studied intensively in the limit of low modulation amplitudes and pushed magnonic devices closer towards competitive applications. Whereas the existence of non-linear magnons in the strong modulation regime has only recently been predicted for $Ni_{80}Fe_{20}$ thin films [1]. This novel class of non-linear excitations oscillates at higher half-integer multiples of the driving frequency at low bias fields. Moreover, these higher-order processes are even predicted to dominate the non-linear response in this regime. However, it is an open question under which conditions such non-linear spin waves (NLSWs) emerge coherently and how they may be used for magnonic devices.

Here, we experimentally demonstrate a class of spin waves oscillating at higher odd half-integer harmonics. We employ our super-Nyquist-sampling MOKE (SNS-MOKE) [2] technique to directly image these parametrically generated magnons in micron-sized $Ni_{80}Fe_{20}$ elements in order to determine their wave vectors [3]. The wave vectors obtained in these higher-order non-linear processes strongly differ from the well known three-magnon scattering indicating their different origin. By investigating the phase stability of these NLSW's oscillating at 3/2 of the driving frequency, we demonstrate the presence of two degenerate phase states that may be selected and stabilized by external phase-locking (as shown in Fig. 1). These results open new possibilities for applications such as spin-wave sources, amplifiers and phase-encoded information processing with magnons.

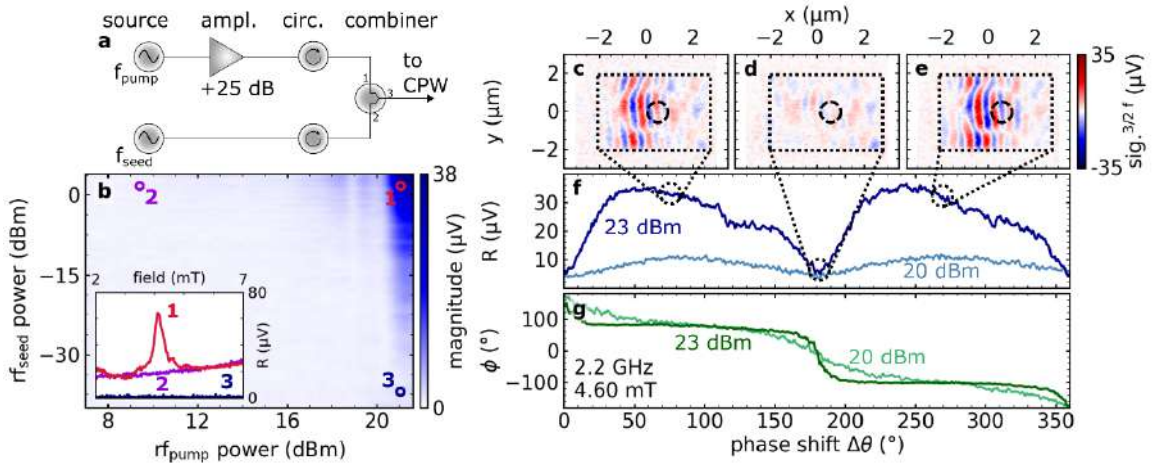



**Fig. 1**: **a** Modified excitation geometry for phase-locking experiments. **b** Power dependent stability map of NLSWs at 3/2 of the driving frequency. **c** - **e** Spatialy resolved imaging of NLSWs in different phase states within a micro-structured $Ni_{80}Fe_{20}$ element. **f** - **g** Magnitude and phase of NLSWs as a function of the seed frequencie's phase.

## References

[1] H. G. Bauer, P. Majchrak, T. Kachel, C. H. Back, and G. Woltersdorf, *Nat Commun* **6**, 8274 (2015).

[2] R. Dreyer, N. Liebing, E. R. J. Edwards, A. Müller, and G. Woltersdorf, *Phys Rev Mat* **5**, 064411 (2021).

[3] R. Dreyer, A. F. Schäffer, H. G. Bauer, N. Liebing, J. Berakdar, and G. Woltersdorf, *Nat Commun* **13**, 4939 (2022).

# Thermal laser patterning of YIG structures for magnonics

*Andrea Del Giacco*[1], *Alberto Toniato*[1], *Rouven Dreyer*[2], *Marco Madami*[3], *Stephanie Lake*[2], Frank Syrowatka[5], *Silvia Tacchi*[4], *Georg Schmidt*[2,5], *Riccardo Bertacco*[1]
*andrea.delgiacco@polimi.it

[1]*Dipartimento di Fisica, Politecnico di Milano, Milano, Italy*
[2]*Institut of Physics, Martin Luther University Halle-Wittenberg,* 0699*, Halle Germany*
[3]*Dipartimento di Fisica e Geologia, Università di Perugia, I-06123 Perugia, Italy*
[4]*Istituto Officina dei Materiali del CNR (CNR-IOM), Unità di Perugia, Perugia, Italy*
[5] Interdisziplinäres Zentrum für Materialwissenschaften, Martin-Luther-Universität Halle Wittenberg, 06099 Halle, Germany

The realization of high-quality waveguide structures for spin waves is of overwhelming importance for the integration of magnonic devices. In case of Yittrium Iron Garnet (YIG), integration of spin-wave based devices poses serious problems due to both substrate issues (so far excellent properties are obtained only on Gadolinium Gallium Garnet - GGG) and thermal budget issues, as deposition temperatures largely exceed the maximum temperature (400°C) compatible with CMOS.

In this work we describe a different approach for the fabrication of single crystal YIG structures, by means of laser induced heat. This procedure results in strongly localized crystallization of amorphous YIG films deposited at room temperature on GGG. At variance with previous works employing uniform post-annealing to obtain low-damping free-standing structures [1], our method does not expose the whole wafer to high thermal budgets, thus solving one of the bottlenecks related to YIG integration. Starting from amorphous YIG films grown by pulsed laser deposition at room temperature, a 405 nm laser with nominal spot size of 1.2 μm is employed for patterning of arbitrary shaped structures by inducing local crystallization. Micro-Raman and Electron Back Scattering Diffraction measurements confirm the crystallization of the irradiated regions for optimized laser dose and writing parameters.

To fully characterize the dynamic magnetic properties of crystallized structures in this contribution we report on Spin Wave (SW) propagation in conduits patterned with different laser doses on YIG films with 65 and 100 nm nominal thickness. Due to the different thermal expansion coefficients of YIG and GGG, in 65 nm thick films the strain induced during laser patterning leads to a morphological corrugation with wave-vector parallel to the writing direction, associated to a modulation of the local magnetic properties, as seen in the AFM and MFM images of figure 1.a and 1.b. This reflects in a non-uniform propagation of SWs for just a few microns in a 10 μm wide conduit, where Damon-Eshbach waves are excited via a microstripe antenna and observed by Brillouin Light Scattering (BLS), as reported in figure 1.c. The morphological corrugation almost disappears in structures patterned on 100 nm thick YIG and its potential impact is further minimized in narrower conduits (1 μm wide), thus leading to a continuous propagation of SWs over 10 μm (see Figure 1.d).

These results reveal the potential of our patterning approach for magnonic conduits with arbitrary shape. To further improve the SWs propagation length, we aim to tune the writing parameters in order to avoid the creation of crystal defects due to the partial overlap of the laser spots during the scanning of the targeted area. Preliminary results on single dots and magnonic conduits written with a single laser scan will be presented at the conference.

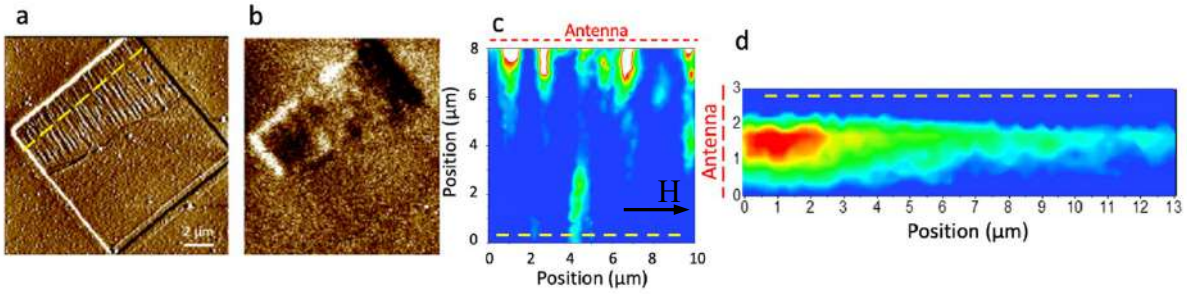



**Figure 1**: a,b) Topography and MFM signal from a square pattern with writing direction as the dashed line, from a 65 nm thick YIG film; c) and d) BLS intensity map recorded for a 10 μm wide conduit patterned on 65 nm YIG, and of a 1μm wide conduit patterned on a 100 nm thick YIG conduit, respectively.

**References**

[1] Hauser et al, Scientific reports 2016

# Spectral Content of Caustic Spin Wave Beams in Soft, Thin Ferromagnetic Films

*Alexis Wartelle[1]*, Franz Vilsmeier[2], Takuya Taniguchi[2], and Christian H. Back[2]*

[1] *Institut Jean Lamour, UMR CNRS 7198, Université de Lorraine, 54506 Vandoeuvre, France*

[2] *Fakultät fur Physik, Technische Universität München, Garching, Germany*

*alexis.wartelle@esrf.fr

The dipolar interaction induces a marked anisotropy in the dispersion relation of spin waves (SWs) in soft thin films [1]. Unusual phenomena may occur as a result; among them are caustics, often referred to as ``self-focussing'' in the context of phonon propagation [2]. This situation, where high-amplitude and low-divergence beams are generated, can be understood from the geometry of the intersection of the SW dispersion relation and a constant-frequency plane: the slowness curve. In certain conditions, the latter may comprise an inflexion point. The strong and directional SW emission results then from the stationary group velocity direction $\theta_V$ for wave-vectors in the vicinity of the one at the inflexion point, which is called a caustic point. An example of slowness curve (in blue) for a film of thickness *d* is shown in Fig.1a); a generic wave-vector $\overrightarrow{k_0}$ with wavefront angle $\varphi_0$ and group velocity $\overrightarrow{v_g}$ is highlighted. The caustic point is marked in orange on the slowness curve.

We have recently proposed a systematic numerical survey of SW caustics in soft thin films [3], based on the dispersion relation derived by the model by Kalinikos and Slavin [1]. Our investigations have revealed two cases of special interest for the field of magnonics, namely (i) caustic points of higher order *i.e.* extrema for $\theta_V$ with a quartic behaviour, (ii) and merged caustic SW beams close to the exchange-dominated regime, similarly to previous findings in perpendicular thin films [4].

The first case highlights the need for caution regarding the description of a beam's spectral content. We have shown [3] that in the case of SW caustics, one should not consider the natural wavelength defined as $\lambda_0=2\pi/k$ (with k the usual wavenumber), but rather the apparent wavelength $\lambda=\lambda_0/\cos(\theta_V-\varphi)$, corresponding to the spatial periodicity in the direction of beam propagation. We have shown that they exhibit qualitatively distinct behaviours around caustic points [3]. This is illustrated in Fig. 1b), representing the relative variations of $\lambda$, $\lambda_0$, and $\theta_V$ around the caustic point properties $\lambda_c$, $\lambda_{0,c}$, and $\theta_{V,c}$, as a function of a normalized curvilinear abscissa $s/s_M$ along the slowness curve from Fig. 1a). With this in mind, we tackle the notion of a beam's spectral content, as a first step towards a better understanding of the caustics' amplitude enhancement.

**Acknowledgements**

This work is supported by the Deutsche Forschungsgemeinschaft (German Research Foundation), Project-ID 314695032SFB 1277 (project A08).

**References**

[1] B. A. Kalinikos, A. N. Slavin, *J. Phys. C: Solid State Phys.* **19** (1986) 7013.

[2] H. J. Maris, ``Phonon Focusing'' p. 51-90 in *Nonequilibrium phonons in nonmetallic crystals*, (1986, edited by W. Eisenmenger, A. A. Kaplyanskii) Vol. 16 of ``Modern Problems in Condensed Matter Sciences'', Elsevier.

[3] A. Wartelle, F. Vilsmeier, T. Taniguchi, C. H. Back, ``Caustic spin wave beams in soft, thin films: properties and classification'', *Phys. Rev. B*, in press. arXiv preprint 2301.01220

[4] J.-V. Kim, R. L. Stamps, R. E. Camley, *Phys. Rev. Lett.* **117** (2016) 197204.

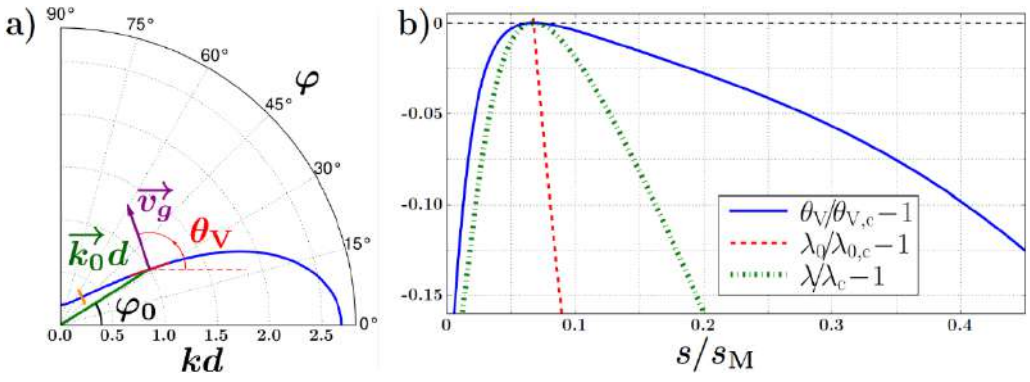



Figure 1: a) Examplary slowness curve (blue) with its caustic point shown in orange. b) Variations of the group velocity direction, the natural wavelength and the apparent wavelength with respect to their values at the caustic point.

# Imaging and Quantifying Magnetoacoustic Waves in Ferromagnetic Materials

*Marc Rovirola*[1,2]*, *M. Waqas Khaliq*[1,3], *Blai Casals*[2,4], *Michael Foerster*[3], *Miguel Angel Niño*[3], *Jens Herfort*[5], *Joan Manel Hernàndez*[1,2], *Ferran Macià*[1,2], *and Alberto Hernández-Mínguez*[5]

[1] *Dept. of Condensed Matter Physics, University of Barcelona, 08028 Barcelona, Spain*
[2] *Institute of Nanoscience and Nanotechnology (IN2UB), University of Barcelona, 08028 Barcelona, Spain*
[3] *ALBA Synchrotron Light Source, 08290 Cerdanyola del Vallès, Spain*
[4] *Dept. of Applied Physics, University of Barcelona, 08028 Barcelona, Spain*
[5] *Paul-Drude-Institut für Festkörperelektronik, Leibniz-Institut im Forschungsverbund Berlin e.V., Hausvogteiplatz 5-7, 10117 Berlin, Germany*

*marc.rovirola@ub.edu

Spin waves, which are collective excitations of magnetic order, hold great promise as a replacement for charge carriers in information transport and processing. This is due to their low power consumption and potential for integration into nanoscale devices. In addition, spin waves do not involve the movement of charged particles reducing heat dissipation and increasing overall efficiency. Surface Acoustic Waves (SAWs) provide an efficient coupling between strain and magnetization, making them an attractive option for spin wave manipulation.

In recent experiments, we used X-ray Photoemission Electron Microscopy (XPEEM) and X-ray Magnetic Circular Dichroism (XMCD) to image and measure the coupling between SAW and spin waves [1]. We studied the behavior of these coupled waves at frequencies of several GHz in different materials, including Nickel, Cobalt and a Heusler Alloy ($Fe_3Si$). Contrary to Ferromagnetic Resonance (FMR) spectroscopy experiments where only energy absorption at resonances is detected, we found a clear distinction between resonant and non-resonant spin waves [2]. Moreover, micromagnetic simulations allowed us to corroborate the results and estimate the magnetoelastic constants, which provides an idea of the SAW-spin wave coupling strength in each material.

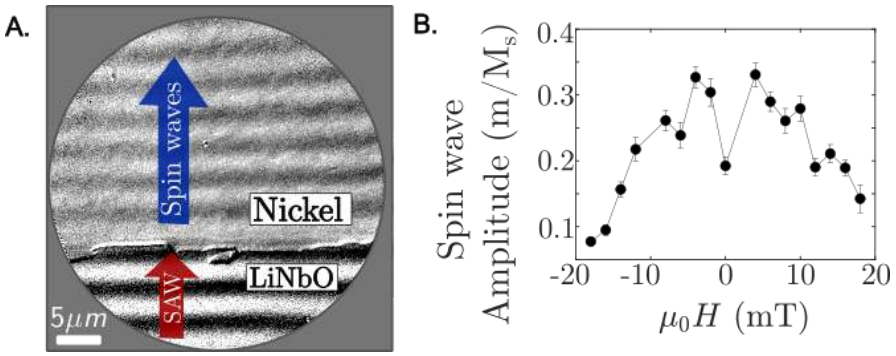



Figure: **A.** XMCD image showing SAW in LiNbO and spin waves in Nickel. **B.** Spin wave amplitude as a function of the magnetic field.

## References

[1] B. Casals, N. Statuto, M. Foerster, A. Hernández-Mínguez, R. Cichelero, P. Manshausen, A. Mandziak, L. Aballe, J.M. Hernàndez, and F. Macià, Phys. Rev. Lett. 124, 137202M (2020)

*[2]* M. Rovirola, M.W. Khaliq, B. Casals, M. Foerster, M.A. Niño, J. Herfort, J.M. Hernàndez, F. Macià, A. Hernández-Mínguez., eprint arXiv:2212.07994 (2022)

# All-inductive observation of nonlinear spin wave processes in synthetic antiferromagnet microstripes

*A.Mouhoub[1]*, C.Chappert[1], J-V.Kim[1], A.Solignac[2] and T.Devolder[1]*

[1] *Université Paris-Saclay, CNRS, Centre de Nanosciences et de Nanotechnologies, 91120 Palaiseau, France*

[2] *SPEC, CEA, CNRS, Université Paris-Saclay, 91191 Gif-sur-Yvette, France*

*asma.mouhoub@u-psud.fr

Spin waves (SW) are the eigen-excitations of magnetic order parameters. They exhibit specific properties such as nonlinearity (NL) which are potentially useful for neuromorphic computing and magnonic logic devices. Here we report on an all-inductive study of non linear processes in microstripes of synthetic antiferromagnets (SAFs). Such films exhibit two types of SW modes, known as acoustical and optical modes. The frequencies of these modes vary with the applied magnetic field. For specific configurations of the applied static magnetic field $H_{DC}$, the frequency of the acoustical magnon mode ($f_{ac}$) becomes half that of the optical magnon mode($f_{op}$), a favorable condition to investigate nonlinear phenomena in SAF [1].

We used the experimental configuration shown on Fig. 1.a. to measure inductively the nonlinear SW processes in a SAF. A RF stimulus is sent through an inductive antenna using a synthesizer to excite spin wave in an optimized $Co_{40}Fe_{40}B_{20}$/Ru/CoFeB stripe [2] acting as spin wave conduit. The response is then recorded on a spectrum analyzer using a second antenna. To investigate non-linear effects, three parameters are varied: i) the static field $H_{DC}$ chosen for each sample to ensure $f_{op} = 2f_{ac}$, ii) the microwave power arriving at the sample (-15<$P_{in}$<11 dBm), and iii) the pump frequency to excite the optical mode (10<$f_{pump}$<15 GHz). To evidence the nonlinear behaviors, an excess power spectral density is determined by substracting values recorded on the same sample but for a slightly different value of $H_{DC}$. A typical spectra is shown on Fig1.b. The doublets near $f_{pump}/2$ evidence a three-magnon process where one optical magnon at $f_{pump}$ split into two acoustical magnons at $f_{pump}/2 - \delta$ and $f_{pump}/2 + \delta$. 2). The second harmonics of the doublet around the $f_{pump}$ indicates that we are deep in the NL regime and the strong halo around $f_{pump}$ corresponds to a four-magnon scattering process, in which two optical magnons excited at $f_{pump}$ annihilate and create two new optical magnons [1,3].

For the three-magnon scattering process, we observe that the frequency of the split magnons is directly related to the input frequency and the power level applied. To investigate the threshold behavior, we performed a detailed study on the power dependence of the magnon amplitude at various input frequencies. In addition, we are developing a new methodology to investigate a time-resolved measurement of nonlinear SW.

**Acknowledgments**

We acknowledge financial support from the EOBE doctoral school of Paris-Saclay University.

**References**

[1] A. Kamimaki, et al., Phys. Rev. Applied 13, 044036 (2020)
[2] A.Mouhoub, et al., arXiv:2211.02497 (2022).
[3] H. Schultheiss, et al. Phys. Rev. B 86, 054414 (2012)

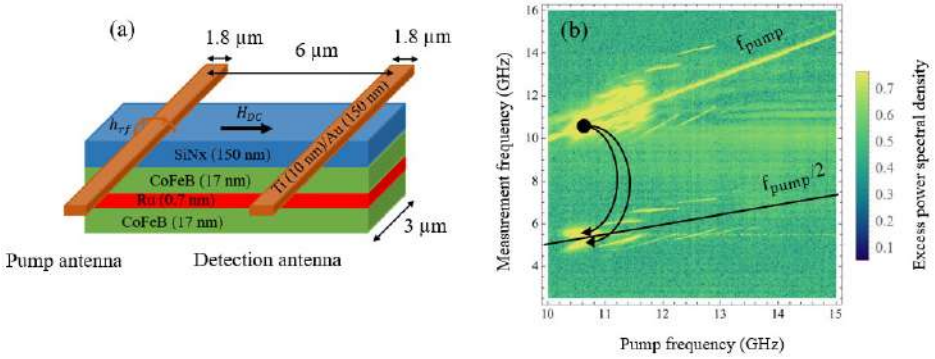



Figure 1: a) Scheme of the experimental configuration. In practice, 4 well separated $Co_{40}Fe_{40}B_{20}$ microstripes are positioned under the antennas to improve the signal-to-noise ratio. b) Spectra of excess power spectral density versus pump frequency at excitation power arriving at the sample 11 dBm and $H_{DC}$=20 mT, for a SAF with 2x17 nm of CoFeB.

# Three-dimensional spin waves unraveled in a synthetic antiferromagnet

*Davide Girardi[1*], Simone Finizio[2], Claire Donnelly[3], Guglielmo Rubini[1], Sina Mayr[2,4], Valerio Levati[1], Simone Cuccurullo[1], Federico Maspero[5,6], Jörg Raabe[2], Daniela Petti[1], Edoardo Albisetti[1]*

[1]*Dipartimento di Fisica, Politecnico di Milano; Milano, Italy.*
[2]*Swiss Light Source, Paul Scherrer Institut; PSI Villigen, Switzerland.*
[3]*Max Planck Institute for Chemical Physics of Solids; Dresden, Germany.*
[4]*Laboratory for Mesoscopic Systems, Department of Materials, ETH Zurich; Zurich, Switzerland.*
[5]*Dipartimento di Ingegneria Civile e Ambientale, Politecnico di Milano; Milano, Italy.*
[6]*Istituto di Fotonica e Nanotecnologie, Consiglio Nazionale delle Ricerche; Milano, Italy.*

*davide.girardi@polimi.it

Spin waves are one of the most interesting candidates for the development of next generation energy efficient devices for carrying and processing information. In this context, three-dimensionality is an intrinsic aspect of their rich phenomenology, both fundamentally and in view of technological applications [1]. However, the experimental visualization of propagating spin waves in three dimensions has remained elusive, due to the harsh requirement of combining nanoscale spatial resolution in 3D, and time resolution across the GHz frequency range.

Here, we experimentally image in three dimensions propagating spin waves emitted by nanoscale spin textures [2] in a CoFeB/NiFe-based synthetic antiferromagnet, revealing the wealth of three-dimensional features associated with their dynamics, spatial localization, and interference. For doing so, we exploit the Time-Resolved Soft X-Ray Laminography (TR-SoXL) technique recently developed at the PolLux beamline of the Swiss Light Source synchrotron facility [3], that allows to obtain the three-dimensional time-resolved reconstructions of the magnetization dynamics of thin samples, with nanoscale spatial resolution and sub-ns temporal resolution [4]. In particular, we reconstruct the three-dimensional precession of the magnetization associated with spin-wave propagation, and map the localization of the spin-wave modes within the plane and through the film thickness. We find that non-uniform spin-wave profiles lead to three-dimensional interference patterns, which we experimentally reveal.

This work enables a comprehensive and detailed understanding of complex spin-wave modes in three-dimensions and opens the way for manipulating and harnessing new degrees of freedom for the design of novel functions in magnonic devices.

## References

[1] G. Gubbiotti. Three-Dimensional Magnonics: Layered, Micro- and Nanostructures. *Jenny Stanford Publishing*, **2019**.
[2] E. Albisetti *et al.* Synthetic Antiferromagnets: Optically Inspired Nanomagnonics with Nonreciprocal Spin Waves in Synthetic Antiferromagnets. *Advanced Materials*, vol. 32, no. 9, **2020**.
[3] C. Donnelly *et al.* Tomographic reconstruction of a three-dimensional magnetization vector field. *New Journal of Physics*, vol. 20, no. 8, **2018**.
[4] C. Donnelly *et al.* Time-resolved imaging of three-dimensional nanoscale magnetization dynamics. *Nature Nanotechnology*, vol. 15, no. 5, **2020**.

# Magnetoelastic Interaction Of Surface Acoustic Waves With FeRh Across Its Magnetic Transition

*A.K. Vythelingum[1], G. Olivetti[1], D. Nguyen[1], E. Dandeu[1], C. Gourdon[1], L. Thevenard[1]**

[1] *Institut des Nanosciences de Paris, Sorbonne Université, CNRS, 4 place Jussieu, Paris*

*thevenard@insp.jussieu.fr

Remote excitation of ferromagnetic resonance (FMR) using piezo-generated surface acoustic waves (SAWs) and magnetoelasticity opens up perspectives of new spintronic architectures, resonant magnetization switching, and non-reciprocal acoustic propagation [1]. It involves matching the ~GHz frequencies of SAWs to the spin-wave (SW) frequencies of a ferromagnet. The extreme sensitivity of $f_{sw}$ and of the magneto-elastic torque to the angle/amplitude of the applied field can make it challenging to reach the SAW FMR regime at which $f_{SAW}=f_{sw}(B_{res})$. While most studies are carried out on polycrystalline Nickel or Co, or epitaxial Fe or $Fe_3Si$ [1,2], here we explore the magnetoelastic interaction of SAWs with FeRh. This antiferromagnet undergoes a first order transition to an FM phase with a large volume change, and could allow for : (i) both anisotropic and volume magnetostrictive effects, and (ii) an exploration of the interaction of SAWs with both FM and AF:FM domain walls, respectively at the sample's FM coercivity and AF<>FM coexistence region.

A thick ($d$=270nm) polycrystalline FeRh mesa was grown on GaAs [2], instead of the usual non-piezoelectric MgO. Interdigitated transducers were deposited to excite SAWs, whose amplitude after propagation is first measured versus field at fixed temperature $T_{set}$. As $T_{set}$ and $f_{SAW}$ rise (Figs.(a,b)), an increasingly deep drop is seen at increasingly small $B_{dip}$ ~ 0-40 mT, with attenuations of up to 42 dB/cm, much higher than the 5-12 dB/cm seen in Fe or Ni at similar frequency [3]. Contrary to what is expected from usual magnetoelastic torques [4], this peaked attenuation remains strong for fields perpendicular to $k_{SAW}$, and $B_{dip}$ is clearly hysteretic and varies only weakly with $f_{SAW}$. In order to probe the influence of the AF phase on the SAW, its amplitude is then measured versus temperature at set field $B_{set}$ (Fig.(d)). At low frequencies, a clear, temperature-hysteretic, attenuation coincides with the middle of the transition (Fig.(c)), at which we expect a large number of AF:FM domains. At the highest $f_{SAW}$, this effect seems overshadowed by the FM phase resonance first mentioned, but is recovered by applying $B_{set} >> B_{dip}$ (889MHz, B=500G curve in (d)). These novel features will be discussed in the frame of a SAW interacting with spin-wave modes – as usually described – or with FM and AF:FM domain-walls, whose dynamics have recently been probed by pump-probe measurements [5].

This work thus presents the first field-dependent observations of SAWs interacting with FeRh. Among others, its polycrystalline nature allows for the presence of numerous domains in order to explore exciting new magneto-acoustics physics involving both multidomain FM and mixed FM/ AF phases.

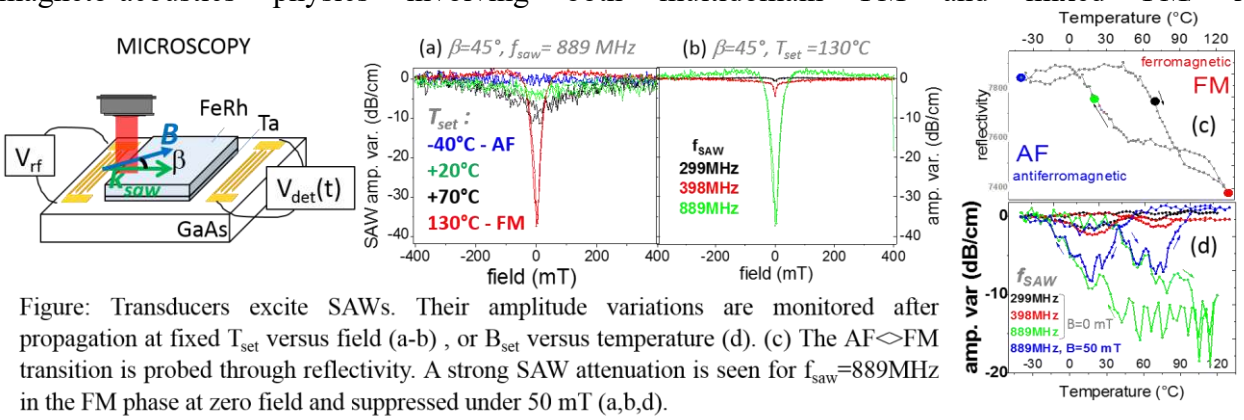



Figure: Transducers excite SAWs. Their amplitude variations are monitored after propagation at fixed $T_{set}$ versus field (a-b) , or $B_{set}$ versus temperature (d). (c) The AF<>FM transition is probed through reflectivity. A strong SAW attenuation is seen for $f_{saw}$=889MHz in the FM phase at zero field and suppressed under 50 mT (a,b,d).

## References

[1] Delsing *et al*, *Journal of Physics D: Applied Physics* , **52** 353001 (2019)
[2] Nguyen *et al*, JEMS2023 abstract
[3] Duquesne *et al*, *Phys. Rev. App.* **12** 024042 (2019), Weiler *et al*, Phys. Rev. Lett **106**, 117601 (2011)
[4] Xu *et al Sci. Adv.* **6**, eabb1724 (2020), Küß *et al Phys. Rev. App.* **15** 034046 (2021)
[5] Gourdon *et al*, JEMS2023 abstract

# Spectroscopy and Microscopy of Propagating Spin-Waves in a Synthetic Anti-Ferromagnet by Brillouin Light Scattering.

*Jean-Paul Adam*[1*], *Florian Millo*[1], *Sokhna-Méry Ngom*[1], *Asma Mouhoub*[1], *Joo-Von Kim*[1], *Claude Chappert*[1], *Aurélie Solignac*[2], *and Thibaut Devolder*[1]

[1] *Université Paris-Saclay, CNRS, Centre de Nanosciences et de Nanotechnologies, 91120, Palaiseau, France, 91120 Palaiseau, France*

[2] *SPEC, CEA, CNRS, Université Paris-Saclay, 91191 Gif-sur-Yvette, France*

*jean-paul.adam@universite-paris-saclay.fr

Here we study propagating spin-waves in a symmetric SAF made of CoFeB (17 nm thickness) and Ruthenium (0.7 nm thickness) layers patterned in 5 µm wide stripe. Spin-waves are excited by an r.f. magnetic field emitted by a single wire antenna – see Figure 1-b. We report on their measurement using micro-focused Brillouin Light Scattering using a 532 nm wavelength LASER focused on the sample with a lateral size of about 350 nm in diameter. Localised spectroscopy allows to identify both acoustical and optical spin-waves with an increased sensitivity to the acoustical mode since the magnetization dynamics in both CoFeB layers are excited '*in phase*' with a constructive contribution to the optical signal. This spectroscopy evidences a complex structure of the acoustical mode composed of a main band with parallel modes at both higher and lower frequencies than the main acoustical mode. In order to gain more details about theses modes, we performed BLS intensity mapping at fixed static magnetic field and r.f. pumping magnetic field. For excitation frequencies matching the main mode, we could observe uniform like propagating spin-waves – see Figure 1c right panel – while for the other modes, the modes are strongly non uniform with nodes in the width – see Figure 1c left and central panels – as well as of directional spin-wave beams propagating not along the magnetic stripe – see Figure 1c central panel. We calculated numerical dispersion relations and used micromagnetic simulations to explain these behaviours which cannot be accounted for by simple demagnetizing contributions but by the large non-reciprocities of the dispersion relation in SAF[1].

**Acknowledgements**

This work was supported by the French National Research Agency (ANR) [as part of the Investissements d'Avenir program (Labex NanoSaclay, reference: ANR-10- LABX-0035), SPICY, and research contracts No. ANR-20-CE24-0012 (MARIN) and No. ANR-20-CE24-0025 (MAXSAW)].

**References**

[1] R.A Gallardo *et.al.* Physical Review B **104**, 174417 (2021)

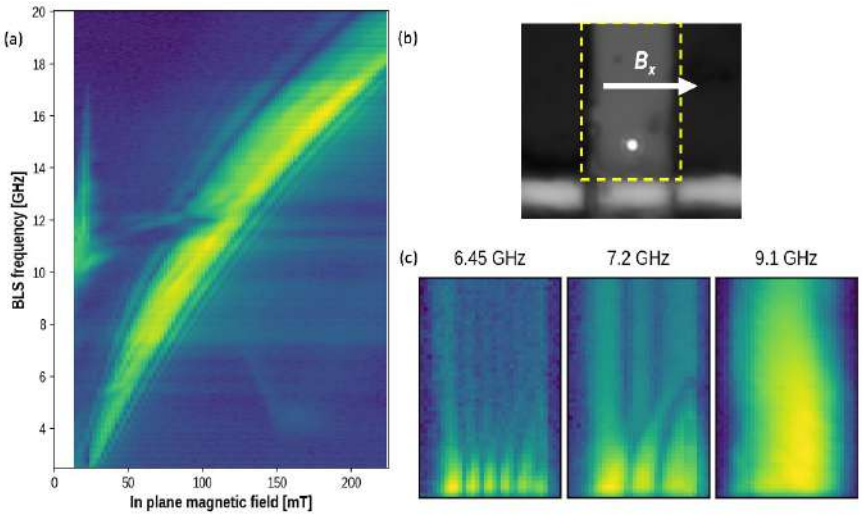



Figure 1. (a) BLS intensity as a function of the applied magnetic field applied transversely to the SAF stripe at the laser spot position represented in the optical imaging (b). (c) BLS intensity maps recorded for two different r.f. excitation frequencies under a transverse magnetic field of 75 mT.

# SYMPOSIUM 15.
## MAGNONICS AND MAGNETOPLASMONICS, ULTRAFAST MAGNETIZATION DYNAMICS. S15 POSTERS

# Spin Wave Hybridization in a Magnonic Crystal Formed by Antidot Lattice with Perpendicular Magnetic Anisotropy

*M. Moalic*[1,*], *M. Zelent*[1], *M. Krawczyk*[1]

[1] Institute of Spintronics and Quantum Information, Faculty of Physics, Adam Mickiewicz University, Poznan, ul. Uniwersytetu Poznańskiego 2, Poznan, PL-61-614 Poland

*matmoa@amu.edu.pl

Magnonic crystals (MCs) have demonstrated a lot of potential as a way to control the propagation of spin waves (SWs). Having the ability to create and control SWs could lead to the creation of magnonic devices that are more space efficient than optical devices and more energy efficient than current electronics. In this research, we study a MC created in a thin film made up of 8 repetitions of Co (0.75nm) and Pd (0.9nm) bilayers for a total thickness of 13.2 nm [1]. This particular combination of a ferromagnetic layer and a heavy metal layer results in a strong perpendicular magnetic anisotropy (PMA) which is interesting as it makes the SW dispersion isotropic. Periodically throughout this thin-plane film, nanodots were etched out using a 10nm wide focused ion beam producing a pattern of antidots. This process not only removed some material, but also damaged the area around each antidot, creating a ring around the antidots where the magnetic properties, notably the PMA have been modified. Due to this, the magnetization at the antidot's edges is almost in-plane. As shown in Fig.1, the ground state of a circular antidot is magnetized in its edge ring in a vortex-like configuration. Through micromagnetic simulations, we analyse the dynamic coupling between edge localised and bulk modes in the film. At first, we limit our analysis to non-propagating SWs and we modify the exciting field as well as the strength of the global external static magnetic field which is oriented out-of-plane and we analyze the SW modes that exist in the rim or in the bulk as shown in Fig.2. Next we show the dynamic coupling between rims and bulk, demonstrating collective behavior on the lattice and promising magnonic applications.

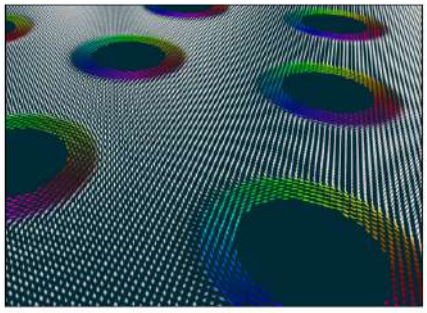


Figure 1: Magnetization in the magnonic crystal.

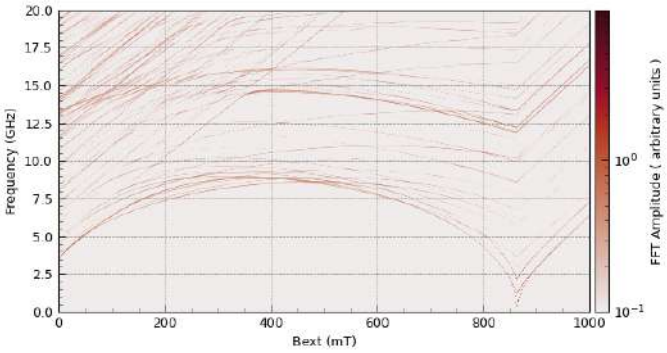



Figure 2: Resonnance spectra of spin waves in the MC depending on the external field.

The authors acknowledge funding from the Polish National Science Centre, project No. UMO2020/37/B/ST3/03936.

## References

[1] S. Pan, S. Mondal, M. Zelent, R. Szwierz, S. Pal, O. Hellwig, M. Krawczyk, and A. Barman, “Edge localization of spin waves in antidot multilayers with perpendicular magnetic anisotropy”, Physical Review B 101, 014403 (2020).

# Nonequilibrium Magnons from Hot Electrons in Antiferromagnetic Systems

*Marion M. S. Barbeau[1], Mikhail Titov[2], Mikhail I. Katsnelson[2], Alireza Qaiumzadeh[1]**

[1] *Center for Quantum Spintronics, Department of Physics, Norwegian University of Science and Technology, NO-7491 Trondheim, Norway*

[2] *Radboud University, Institute for Molecules and Materials, 6525 AJ Nijmegen, The Netherlands*

*alireza.qaiumzadeh@ntnu.no

We describe a nonthermal magnon activation mechanism in antiferromagnetic (AFM) systems via locally equilibrated spin-unpolarized hot electrons excited by an ultrafast intense laser pulse. We employ a quantum kinetic equation that takes into account a direct electron-magnon scattering channel in either bulk AFM metal or at the interface of the AFM/normal-metal heterostructure. The mechanism is responsible for the nonequilibrium population of AFM magnon modes on a subnanosecond timescale, which are formed shortly after the local thermalization of hot electrons by Coulomb interactions. Nonequilibrium magnon populations can be additionally manipulated by applying an external magnetic field. Our work paves the way toward spin dynamics control in AFM systems via the ultrafast manipulation of out-of-equilibrium magnon excitations [1].

**Acknowledgements**

This project has been supported by the Norwegian Financial Mechanism Project No. 2019/34/H/ST3/00515, “2Dtronics”; and partially by the Research Council of Norway through its Centers of Excellence funding scheme, Project No. 262633, “QuSpin”.

**References**

[1] M. M. S. Barbeau, M. Titov, M. I. Katsnelson, A. Qaiumzadeh, *arXiv:2209.03469*.

# Electrical Detection of Parametrically Excited Spin Waves in Cubic Anisotropic Materials.

*Shoki Nezu[1]*, Thomas Scheike[2], Hiroaki Sukegawa[2] and Koji Sekiguchi[3]*

[1] *Graduate School of Engineering Science, Yokohama National University, 79-5 Tokiwadai, Hodogaya, Yokohama, Kanagawa 240-8501, Japan*

[2] *National Institute for Materials Science, 1-2-1 Sengen, Tsukuba, Ibaraki 305-0047, Japan*

[3] *Faculty of Engineering, Yokohama National University, 79-5 Tokiwadai, Hodogaya, Yokohama, Kanagawa 240-8501, Japan*

*nezu-shoki-nv@ynu.jp

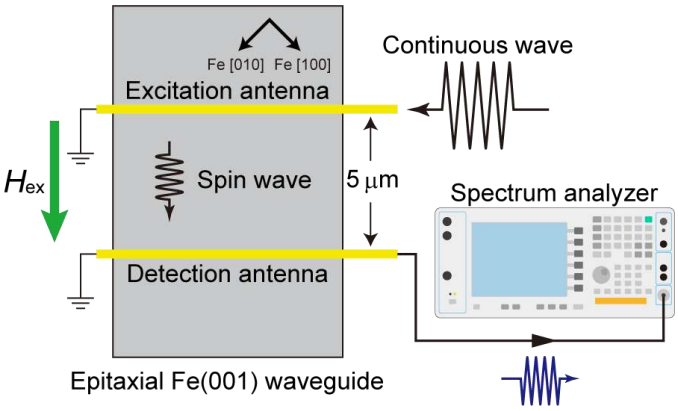
Recently, propagation properties of magnetostatic spin waves have been investigated in epitaxial iron (Fe) thin films, which has the cubic anisotropy, indicating that they are promising materials for the spin-wave devices [1, 2]. In this study, we investigated the propagation characteristics of parametrically excited spin-waves in epitaxial Fe thin films, in order to develop a spin-wave amplification system and quantum devices.

A schematic illustration of an experimental setup is shown in Fig. 1. A 25-nm-thick epitaxial Fe(001) film was fabricated by RF sputtering and the Fe film was then patterned into a 180 μm × 110 μm rectangle waveguide using a lift-off and Ar ion milling techniques. In the presence of an external magnetic field $H_{ex}$ in the direction of the hard magnetization axis, spin waves were parametrically excited by applying continuous waves of $f_p$ = 9.0 GHz to an excitation antenna placed on the waveguide. The pumped spin waves were subsequently detected by a spectrum analyser via a detection antenna.

Figure 2 depicts the pumped spin-wave amplitudes as a function of input power. The external magnetic field was fixed at $\mu_0 H_{ex}$ = 76 mT, which corresponds to the FMR frequency $f_p/2$. The solid line represents the fitting curve obtained by a function of $a\sqrt{P - P_{th}}$, where $a$ and $P_{th}$ are a fitting coefficient and a threshold of an input power, respectively. The threshold power was deduced to be $P_{th}$ = 0.32 mW, which is approximately 80% of the value reported in a prior study on the bulk acoustic wave resonator with a ZnO/YIG/GGG/YIG/Pt structure [3]. The parametric excitation at low power levels results from the reduction of the internal magnetic field due to the cubic anisotropy of the Fe film.

## References

[1] K. Sekiguchi, S-W. Lee, H. Sukegawa, *et al.*, *NPG Asia Mater.* **9**, e392 (2017).
[2] S. Nezu, T. Scheike, H. Sukegawa, *et al.*, *AIP Adv.* **12**, 035320 (2022).
[3] S. G. Alekseev, S. E. Dizhur, N. I. Polzikova, *et al.*, *Appl. Phys. Lett.* **17**, 072408 (2020).



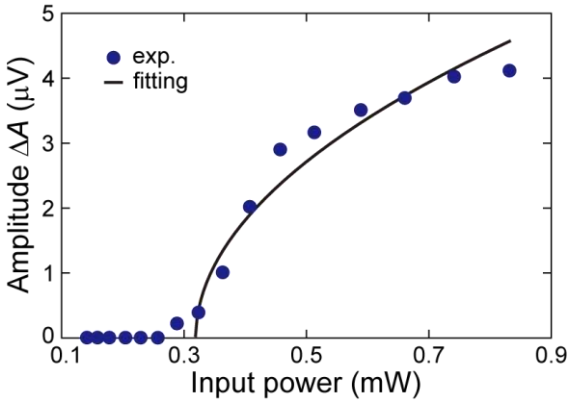




Fig. 1 : Schematic illustration of an experimental setup.
Fig. 2: Input power dependence of parametrically excited spin wave amplitudes at $\mu_0 H_{ex}$ = 76 mT

# Nonreciprocal spin waves in symmetric synthetic antiferromagnetic: BLS measurements and theoretical framework

*F. Millo[1]*, J. -P. Adam[1], J. -V. Kim[1], R. L. Seeger[2], A. Solignac[2], T. Devolder[1]*
[1]*Centre de Nanosciences et de Nanotechnologies (C2N), CNRS, Université Paris-Saclay, France*
[2]*SPEC, CEA, CNRS, Université Paris-Saclay, France*
*florian.millo@universite-paris-saclay.fr

Spin waves (SWs) are elementary magnetic excitation exhibiting nonreciprocity (NR) and nonlinear behaviour [1]. NR SWs in synthetic antiferromagnet (SAF) i. e. ferromagnetic bilayer with opposite directions of the static magnetization of layers that can be stabilized by the RKKY interaction via a non-magnetic spacer layer [2], have growing interest in applications such as magnonic diode, spin wave emitters.

In this work, we study the NR behaviour of SWs in symmetric SAF system with strong dipole-dipole interaction. We compare experimental measurements with micromagnetic simulations [3] and analytical framework based on Dynamic Matrix theory [4] and focus in the small wavevector regime (linear regime [2]). The system [Figure. 1(a)], annealed to reduce anisotropy, is studied by Brillouin Light Scattering (BLS) where we observe the optic and acoustic mode of SWs [Figure. 1(b)].

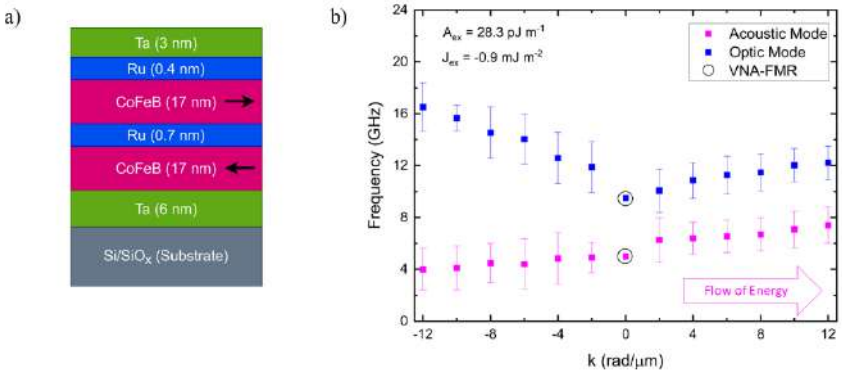



Figure. 1: a) Geometry. b) Dispersion relation of SWs in SAF where wavevector $(\vec{k})$ is parallel with applied field 25 mT. The error bars are the FWHM of stokes and anti-stokes peaks of BLS spectra.

Two situations were studied by BLS; $\vec{k} \parallel \vec{H}$ and $\vec{k} \perp \vec{H}$, where the former shows the largest SW NR and the latter shows the reciprocal behaviour of the system with a group velocity of optic mode close to 0 km/s as expected from theory and micromagnetic simulations. The comparison between BLS measurements, micromagnetic simulations, and analytical framework gives a good qualitative and quantitative agreement allowing us to understand the system.

**Acknowledgements**

The authors acknowledge the French National Research Agency (ANR) under contract No. ANR-20-CE24-0025 (MAXSAW). This work was supported by a public grant overseen by the ANR as part of the "Investissements d'Avenir" program (Labex NanoSaclay, reference: ANR-10-LABX-0035, Project SPICY).

**References**

[1] S. Demokritov – Spin wave confinement, 2nd edition. Pan Stanford (2019).
[2] Verba et al. Physical Review Applied 12, 054061 (2019).
[3] A. Vansteenkiste et al, AIP Advances 4, 107133 (2014).
[4] F. C. Nortemann et al. Physical Review B, Vol 47:18 (1993).

# Hybridized Propagating Spin Waves in a CoFeB/IrMn Bilayer

*Hanchen Wang[1,2], Jinlong Wang[1]*, Jilei Chen[3], Jean-Philippe Ansermet[4], Haiming Yu[1,2]*

[1] *Fert Beijing Institute, MIIT Key Laboratory of Spintronics, School of Integrated Circuit Science and Engineering, Beihang University, Beijing 100191, China*

[2] *International Quantum Academy, Shenzhen 518055, China*

[3] *Shenzhen Institute for Quantum Science and Engineering (SIQSE), and Department of Physics, Southern University of Science and Technology (SUSTech), Shenzhen 518055, China*

[4] *Institute of Physics, École Polytechnique Fédérale de Lausanne (EPFL), 1015, Lausanne, Switzerland*

*by2141004@buaa.edu.cn

Control over the coherent coupling between quasiparticles is a key issue in realizing information manipulation in quantum technology [1]. Magnons, the quasiparticle of spin waves, are the low-energy collective excitations of magnetic moments with long-range order [2]. The hybridization between multiple magnon modes in antiferromagnets and ferrimagnets provides a platform for a coherent control and engineer spin dynamics, of which the coupling strength can be tuned via interlayer dipolar or interfacial exchange interactions in heterostructure systems [3]. However, detailed experimental investigations for coherent control of the coupling strength of hybridized propagating spin waves are still rare within single layer material.

In this work[4], we report the propagating spin waves hybridized between first-order and quasiuniform modes in a $Co_{20}Fe_{60}B_{20}$ thin film capped by $Ir_{25}Mn_{75}$. The anticrossing gaps are observed at room temperature both in the reflection and transmission spectra, where the coupling strength can be tuned by varying the value of the in-plane wave vector at which the dispersion curves cross. The key mechanism behind this feature is theoretically ascribed to the dipole-dipole interaction by a model which accounts for many features of our experimental results. The strong coupling with a cooperativity up to 2.0 is achieved with taking the dissipation rates of two coupled branches into account. A reference CoFeB sample without IrMn reveals that the interfacial pinning effect also plays a role in the magnon-magnon hybridization. Our results provide experimental insight for hybridized magnonics on a magnetic system widely used in spintronics and may foster the field of magnonics in information processing.

## Acknowledgements

The authors acknowledge support from the National Key Research and Development Program of China, Grants No. 2017YFA0206200 and No. 2016YFA0300802, the NSF China under Grants No. 12074026, No. 12104208, and No. U1801661, and the 111 Talent Program B16001.

## References

[1] Y. Tabuchi, S. Ishino, A. Noguchi, T. Ishikawa, R. Yamazaki, K. Usami, Y. Nakamura, *Science* **349** (2015) 405.
[2] P. Pirro, V. I. Vasyuchka, A. A. Serga, and B. Hillebrands, Advances in coherent magnonics, *Nat. Rev. Mater.* **6** (2021) 1114.
[3] J. L. Chen, C. P. Liu, T. Liu, Y. Xiao, K. Xia, G. E. W. Bauer, M. Z. Wu, H. M. Yu, *Phys. Rev. Lett.* **120** (2018) 217202.
[4] H. C. Wang, W. Q. He, R. D. Yuan, Y. Z. Wang, J. L. Wang et al, Hybridized propagating spin waves in a CoFeB/IrMn bilayer. *Phys. Rev. B*. **106** (2022) 064410.

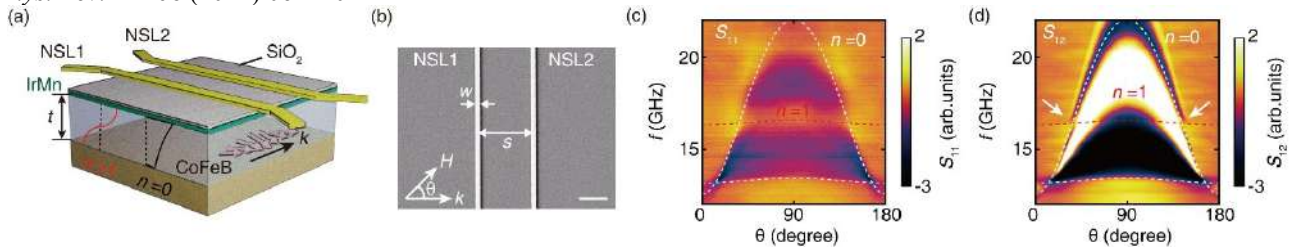



Figure: (a) Illustrative diagram of the device structure. The measured sample is composed of a CoFeB layer and an IrMn layer with $SiO_2$ capping. (b) The SEM image of the NSL antennas, where the scale bar is 1 μm and the angle between the external magnetic field and wave vector direction is defined as θ. (c),(d) Angle-dependent reflection (dBmag) and transmission (Imag) spectra, where the red and white dashed lines are the theoretical fittings of the first-order and quasiuniform modes, respectively.

# Optimization of spin-wave feedback structure

*Masashi Iwaba*[1]* *and Koji Sekiguchi*[1]

[1] *Yokohama national university, Tokiwadai 79-5, Yokohama 240-8501, Japan*

*iwaba-masashi-vg@ynu.jp

Spin wave has been intensively studied in recent years because of its virtue of ultra-low energy propagation. However, if we simply generate large amplitude spin-waves with high power RF radiation to compensate the magnetic damping, the system easily becomes chaos and lose phase information of spin-waves [1]. For the realization of integrated spin-wave circuit, the spin-wave linear amplifier is essential. In this study, we suggest that the feedback waveguide with loop ring should be a possible candidate of linear amplifier.

We measured spatial distributions of spin-waves in feedback structures using a micro-focused Brillouin light scattering (BLS) spectroscopy [2]. The feedback waveguides were made of permalloy film [3], and the film thickness and width of waveguide were 50 nm and 3 μm, respectively. As shown in Figs. 1(a) ~ 1(c), the spin-waves were excited by dual antennas; one antenna was set at edge of main path of waveguide, and the other was set at the middle position of loop ring structure. The line lengths of loop ring structure were changed to be $L_R$ = 2 μm, 4 μm, and 10 μm, respectively. The BLS measurement points were in the centre line of waveguide as shown in the inset of Fig.1(d).

The spatial distributions of spin-waves were shown in Fig. 1(d). For different three structures, the spin wave intensities in range $0 < x < 18$ μm were almost same values. The feedback waveguides with $L_R$ = 2 and 4 μm indicated by red and green data points show larger intensities in the range $22 < x < 27$ μm, while the spin-wave intensity in the feedback waveguide with $L_R$ = 10 μm was rapidly decreased. These results mean that the spin-wave feedback structure favours to the small $L_R$. At distance $x$ = 35 μm, the spin-wave intensity keeps 2.3 times larger value with the optimized structure, indicating that the feedback ring could be the basis of linear amplifier.

## Acknowledgements

This work was partly supported by the Grants-in-Aid for Scientific Research (20H05652 and 22J15058).

## References

[1] A. B. Ustinov, A. V. Kondrashov, I. Tatsenko el al., *Phys. Rev. B* **104** (2021) L140410.
[2] T. Sebastian, K. Schultheiss, B. Obry el al., *Frit. Phys.* **3** (2015) 00035.
[3] K. Sekiguchi, K. Yamada, S. M. Seo, et al., *Appl. Phys. Lett.* **97** (2010) 022508.

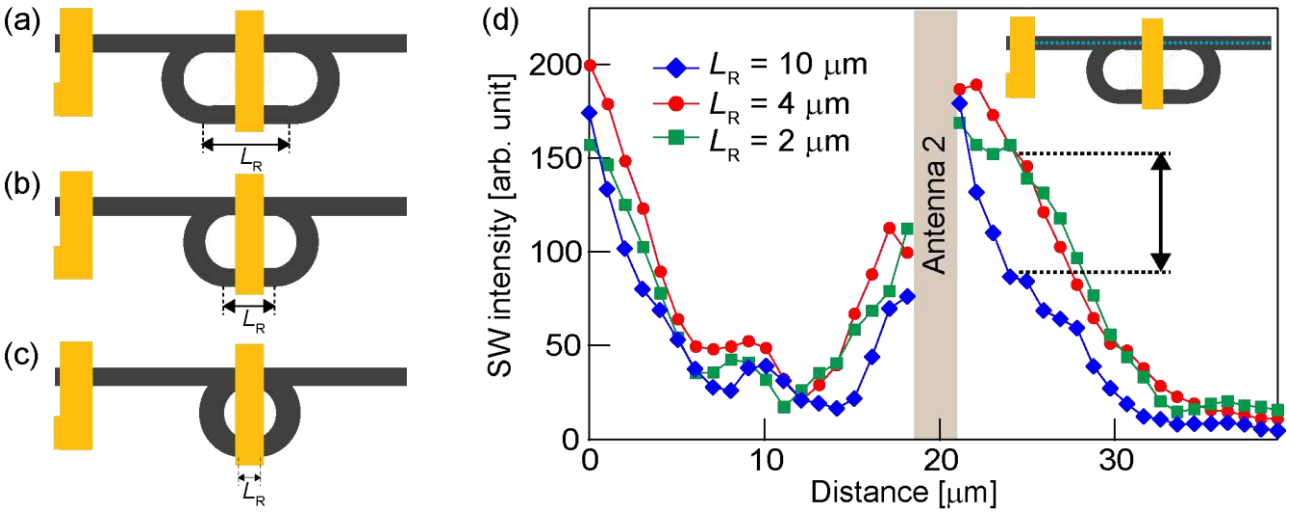



Figure1: Feedback structures with two ring arms and different line length $L_R$. (a), (b), (c) The lengths were $L_R$ = 10 μm, $L_R$ = 4 μm, and $L_R$ = 2 μm, respectively. (d) Spatial distributions of spin wave intensity in main path detected by micro-focused Brillouin light scattering spectroscopy.

# Modelling a 3-Port Network in Cavity Magnonics for Nonreciprocal RF Devices

*Maxime Ardisson[1,2*], Vincent Castel[1], Romain Lebrun[2], Isabella Boventer[2]*
*[1]IMT Atlantique, Technopole Brest-Iroise, CS 83818, 29238 Brest Cedex 3, France*
*[2]Unité Mixte de Physique CNRS, Thales, Université Paris-Saclay, Palaiseau 91767, France*
*maxime.ardisson@imt-atlantique.fr

Over the last decade, cavity magnonics has attracted a lot of attention. The study of the associated quasiparticle of cavity magnonics, cavity magnon polaritons which refer to strongly coupled magnon-photon system offers new possibilities of sensing and quantum information processing [1,2] as well as a new interesting platform for disruptive RF applications. In the realm of signal transmission, nonreciprocity allows the enhancement of the communication channels capacities as well as the protection of the transmission quality. Our goal is to build new types of magnon based microwave non-reciprocal device. For this purpose, we use the possibility to tune and combine the regimes of dissipative and coherent coupling control leading to PT symmetry breaking [3] in cavity magnonics devices.

We develop a non-reciprocal device consisting of a three ports 3D re-entrant cavity coupled with a ferromagnetic sample (see sketch in Figure 1. (a)). The cavity is equipped with two cavity ports and an additional third port used as a direct drive for the magnons [4].

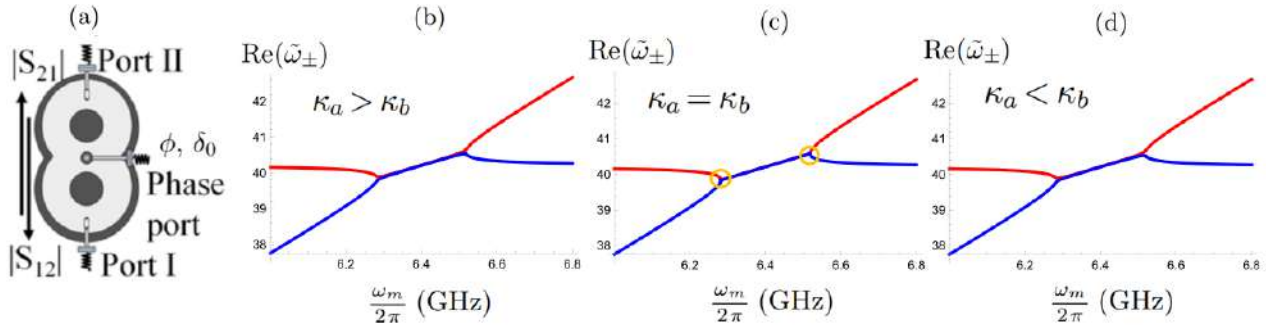



Figure 1. (a) sketch of the 3-port re-entrant cavity setup. (b), (c) and (d) calculated real part of the eigenfrequencies of the system showing level attraction and exceptional points (encircled in orange in (c)) for matched damping condition where $\kappa_a$ and $\kappa_b$ stand for the damping ratios of the cavity and the magnon respectively.

By adjusting the amplitude ratio $\delta_0$ and phase shift $\Phi$ between the cavity input port and the magnon port, the coupling strength can be either purely real, complex or purely imaginary, allowing us to tune the regime of the system from coherent to dissipative at will (see Figure 1. (b) and (d)).

As displayed in Figure 1. (c), our modelled system exhibits exceptional points for matched damping condition [3] which is a signature of PT symmetry breaking. Following the approach described in [5] we then expect both theoretically and experimentally to achieve controlled and compact non-reciprocal behaviour by controlling the two tuning parameters $\delta_0$ and $\Phi$ in our system.

## References

[1] D. Lachance-Quirion, Y. Tabuchi, A. Gloppe, K. Usami, Y. Nakamura, *Appl. Phys. Express* **12** (2019) 070101.
[2] B. Z. Rameshti, S. V. Kusminskiy, J. A. Haigh, K. Usami, D. Lachance-Quirion, Y. Nakamura, C.-M. Hu, H. X. Tang, G. E. Bauer, and Y. M. Blanter, *Physics Reports* **979** (2022) 1–61.
[3] Yi-Pu Wang, J.W. Rao, Y. Yang, Peng-Chao Xu, Y.S. Gui, B.M. Yao, J. Q. You, C.-M. Hu, *Phys. Rev. Lett.* **123** (2019) 127202.
[4] I. Boventer, C. Dörflinger, T. Wolz, R. Macêdo, R. Lebrun, M. Kläui, M. Weides, *Phys. Rev. Research* **2** (2020) 013154.
[5] L. Zhong, C. Zhang, B. M. Yao, *AIP Advances* **12** (2022) 085323.

# Frequency Multiplication by Collective Nanoscale Spin-Wave Dynamics

*Chris Koerner[1]*, Rouven Dreyer[1], Hans G. Bauer[2], Niklas Liebing[1], and Georg Woltersdorf[1,3]*

[1] *Martin Luther University Halle-Wittenberg, Insitute of Physics, Von-Danckelmann-Platz 3, 06120 Halle (Saale), Germany*

[2] *Jahnstrasse 23, 96050, Bamberg, Germany*

[3] *Max Planck Institute of Microstructure Physics, Weinberg 2, 06120 Halle (Saale), Germany*

*chris.koerner@physik.uni-halle.de

Frequency conversion is a process where an input frequency is converted into higher output frequencies. This process is vital for many electronic devices. Devices based on the propagation and interaction of spin waves are a promising alternative to conventional electronics. They utilize spin waves as a carrier of information instead of the electron's charge which promises less energy dissipation and increased energy efficiency. In ferromagnetic materials the characteristic frequency of these excitations is in the gigahertz (GHz) range. This makes the approach competitive with modern electronics for applications like spin wave based computing. However, the interface of magnonic devices with conventional electronics is challenging and often inefficient. Thus, it is preferable to transfer all important functionalities including frequency conversion to the magnonic system.

Here, we probe the magnetic excitations in a soft magnetic material, NiFe, by optical methods locally. We use diamond NV-centers as probes for rf magnetic fields. We find that upon MHz rf excitation a six octave spanning frequency comb is generated inside the magnetic material [1]. Furthermore, we employ Super Nyquist Sampling MOKE microscopy [2] to image the generated spin waves and show the coherent nature of the emerging frequency comb. We attribute the effect to self-synchronized spin-wave assisted switching effects in the sample on the micrometer scale which lead to phase-locked spin-wave emission in the GHz range. Our micromagnetic models very well reproduce the observed comb generation upon MHz- excitation. The discovered frequency multiplication process opens exciting perspectives for spintronic applications, such as all-magonic mixers or coherent on-chip GHz sources.

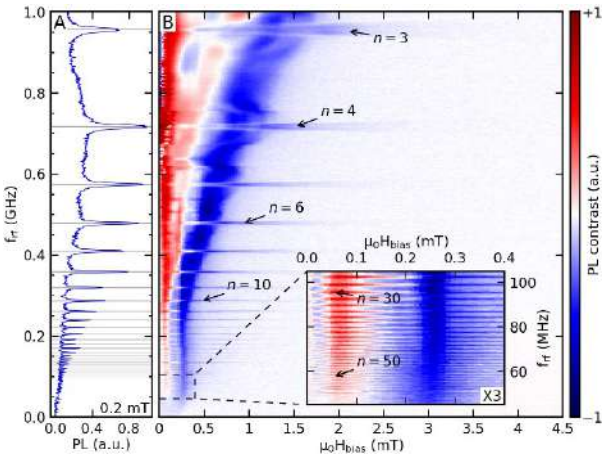



**Fig. 1**: Frequency comb in NiFe (sharp lines) measured by NV-center microscopy. Excitation frequency (y-axis) and static bias field (x-axis) are varied. The NV-centers are sensitive to a fixed frequency of 2.8 GHz, thus the spacing of the comb lines is inversely proportional to the harmonics mode number *n*. Up to the 50th harmonic of the driving frequency are detected.

## References

[1] C. Koerner, R. Dreyer, M. Wagener, N. Liebing, H. G. Bauer, and G. Woltersdorf, "Frequency multiplication by collective nanoscale spin-wave dynamics", Science 375, 1165–1169 (2022).R. Dreyer, N. Liebing, E. R. J. Edwards, A. Müller, and G. Woltersdorf, *Phys Rev Mat* **5**, 064411 (2021).

[2] R. Dreyer, N. Liebing, E. R. J. Edwards, A. M¨ uller, and G. Woltersdorf, "Spin-wave localization and guiding by magnon band structure engineering in yttrium iron garnet", Phys. Rev. Mater. 5, 064411 (2021).

# Tuning the magnon transport property in $Y_3Fe_5O_{12}$ with molecule

*Peisen Yuan[1]*, Sara Catalano[1], Witold Radoslaw Skowronski[1],Roger Llopis[1], Felix Casanova[1] and Luis Hueso Arroyo[1,2]*

[1] *CIC nanoGUNE (Nanoscience Cooperative Research Center),20018 Donostia-San Sebastian, Spain*
[2] *IKERBASQUE (Basque Foundation for Science), 48013 Bilbao, Spain*
*y.peisen@nanogune.eu

Magnonics has been an emerging research field of modern magnetism, especially for the investigation of transportation and manipulation of spin information in magnetic insulators, supplying the advantages of new information technology with low-cost and high-speed signal transmission, storage and process. It is known that the contact of the organic molecules and magnetic atoms or even normal non-magnetic metals will form the hybrid state at the interface within a very narrow interfacial region, which could obviously affect the magnetic properties for both the metal and molecular sides or the whole system [1-3]. Therefore, tailoring the magnetic properties of magnetic insulators by forming the hybrid interface with molecules will be one important way to further manipulate the magnon transport behavior. Here, we demonstrate that the interfacial interaction between organic molecule of cobalt phthalocynine (CoPC) and the magnetic insulator of $Y_3Fe_5O_{12}$ (YIG) could improve the transport property of magnon current, which is related to the interfacial hybridization effect. The device with YIG/CoPC interface shows lager amplitudes of nonlocal signals for both electrically and thermally excited magnon currents. By measuring the field-dependent nonlocal signal and ferromagnetic resonance (FMR) spectra in YIG/CoPC structure, we found that the interfacial effect could change the magnetic anisotropy property of YIG with exhibiting larger saturation field. Moreover, the device with the CoPC overlayer could obtain longer electrically magnon diffusion length due to this interfacial hybridization effect reduced Gilbert damping, which is verified by the FMR measurement. Our result could give us insights to utilize the molecular functionalization to tune the magnon transport in magnetic insulators through the interfacial hybridization effect.

**Acknowledgements**

This work was supported by the "Juan de la Cierva" grant from the Spanish Ministry.

**References**

[1] M. Cinchetti, V. A. Dediu, L. E. Hueso. *Nat. Mater.* **16** (2017), 507.
[2] K. Bairagi, A. Bellec, V. Repain, C. Chacon, Y. Girard, Y. Garreau, J. Lagoute, S. Rousset, R. Breitwieser, Y. C. Hu, Y. C. Chao. *Phys. Rev. Lett.* **114** (2015) 247203.
[3] C. Barraud, P. Seneor, R. Mattana, S. Fusil, K. Bouzehouane, C. Deranlot, P. Graziosi, L. Hueso, I. Bergenti, V. Dediu, F. Petroff. *Nat. Phys.* **6** (2010), 615.

# Magnetooptical Investigation of Nonreciprocal Phonon-Magnon Interaction

*Yannik Kunz[1]*, Michael Schneider[1], Moritz Geilen[1], Philipp Pirro[1], Matthias Küß[2], Manfred Albrecht[2] and Mathias Weiler[1]*

[1] *Fachbereich Physik and Landesforschungszentrum OPTIMAS, Rheinland-Pfälzische Technische Universität Kaiserslautern-Landau, 67663 Kaiserslautern, Germany*

[2] *Institut für Physik, Universität Augsburg, 86159 Augsburg, Germany*

*ykunz@rptu.de

The coupling of surface acoustic waves (SAWs) with spin waves (SWs) intrinsically breaks the time-inversion symmetry. The resulting nonreciprocity can be exploited for applications such as miniaturized microwave isolators. SAWs can be efficiently excited and detected by interdigital transducers. Therefore, the magnetic field dependent SAW transmission induced by the coupling of SAWs with SWs is commonly detected via electrical methods [1].

However, for the investigation of magnetoelastic interactions with spatial resolution, magnetooptical measurement methods are needed. We employed microfocused Brillouin light scattering spectroscopy and frequency-resolved magneto-optical Kerr effect spectroscopy [2] to map the spatial dependence of the phonon-magnon-coupling in a $LiNbO_3/Co_{40}Fe_{40}B_{20}(10\,nm)/Si_3N_4(5\,nm)$-structure.

By taking advantage of the spatial resolution of our measurement methods, the increased SAW absorption caused by the coupling with spin waves is observed and characterized by a magnetic field dependent decay rate. The nonreciprocal nature of the SAW and SW coupling is identified by different magnitudes of the absorption rate for inverted field directions. The coherent conversion of phonons to magnons is shown by an increase in the magnon population, peaking with the maximum phonon intensity and vanishing with the decaying SAW as shown in Figure 1.

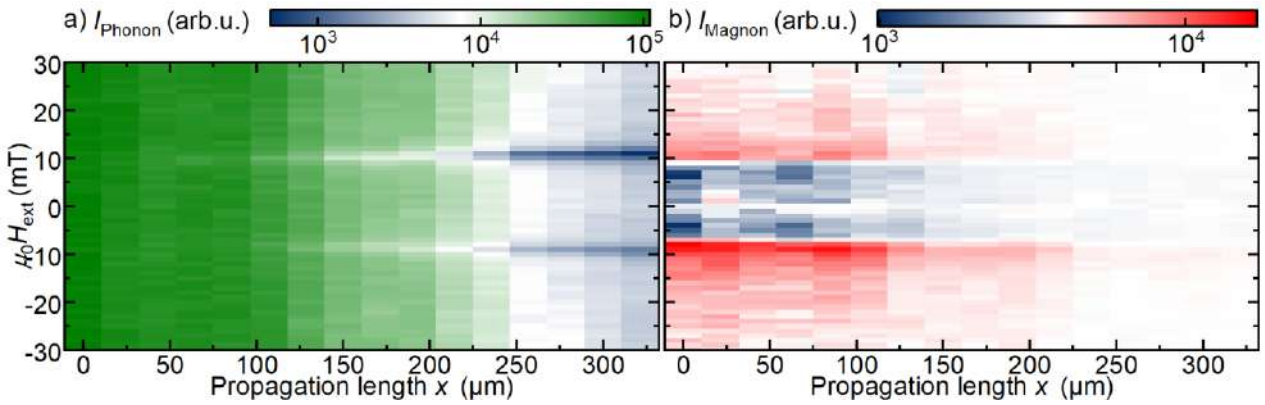



Figure 1: Spatial map of phonon (a) and magnon (b) propagation in a $Co_{40}Fe_{40}B_{20}$ film as a function of external in-plane magnetic field. The SAW is incident from the left (x = 0) and absorbed during propagation through the $Co_{40}Fe_{40}B_{20}$ film. Magnons are most efficiently generated at x = 0 where SAW amplitude is maximal.

## Acknowledgements

We acknowledge the funding by DFG via project No. 492421737.

## References

[1] M. Küss, M. Heigl, L. Flacke, A. Hörner, M. Weiler, M. Albrecht, A. Wixforth, *Phys. Rev. Lett.* **125** (2020) 217203.

[2] L. Liensberger, L. Flacke, D. Rogerson, M. Althammer, R. Gross, M. Weiler, *IEEE Magn. Lett.* **10** (2019) 5503905.

# Shaping Caustic Spin Wave Beams in an Extended Thin Film

*Loic Temdie[1,2,*], Dinesh Wagle[3], M. Taghipour Kaffash[3], Vincent Castel[1,2], H. Majjad[4], R. Bernard[4], Yves Henry[4], Daniel Stoeffler[4], Matthieu Bailleul[4], M. Benjamin Jungfleisch[3], Vincent Vlaminck[1,2]*

*[1]IMT Atlantique, Microwave Dpt., CS 83818, 29238 Brest, France*

*[2]Lab-STICC (UMR 6285), CNRS, Technopole Brest-Iroise, 29238 Brest Cedex 3, France*

*[3]University of Delaware, Dept. of Physics and Astronomy, Newark, Delaware, USA*

*[4]IPCMS – Institut de Physique et Chimie des materiaux de Strasbourg, France*

**contact : loic.temdie-kom@imt-atlantique.fr*

The processing and directional control of information by spin waves is a key challenge for developing of magnonic devices. In the case of in-plane magnetized thin films, for which the dispersion relations are anisotropic, the group and phase velocities do not coincide. This leads to a caustic-like effect in the spin wave propagation. So far, studies of caustic spin wave beams have only been carried out in geometries where a narrow spin wave conduit reaches an extended plane, the junction acting as a diffractive source [1,2].

In this communication, we demonstrate the excitation of two directional caustic spin waves beams in an extended thin film from a nano-constricted *rf* waveguide. Extending our near-field diffraction model to in-plane spin wave modes [3,4] allows us to reveal the importance of the shape and sharpness of the constriction from which the caustic beam is emitted, and the frequency/field conditions for the existence of the caustic beams over the broad microwave spectrum. These predictions are confirmed with micromagnetic simulations and micro-focused Brillouin light scattering (BLS) measurements on nanostructured coplanar waveguides patterned on the top of 200 nm thin yttrium iron garnet (YIG) films, as shown in Fig. 1. Furthermore, the field and frequency dependence of these caustic beams reveal a non-reciprocal asymmetry in intensity between the two beams, which we attribute to a sign change of the perpendicular component of the wavevector with respect to the bias field direction. Our findings have important implications for developing of switchable spin-wave splitters, passive spin-wave frequency-division demultiplexers, and magnonic interferometry.

This work was supported by the French ANR project MagFunc, the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering under Award DE-SC0020308, and the Transatlantic Research Partnership, a program of FACE Foundation and the French Embassy.

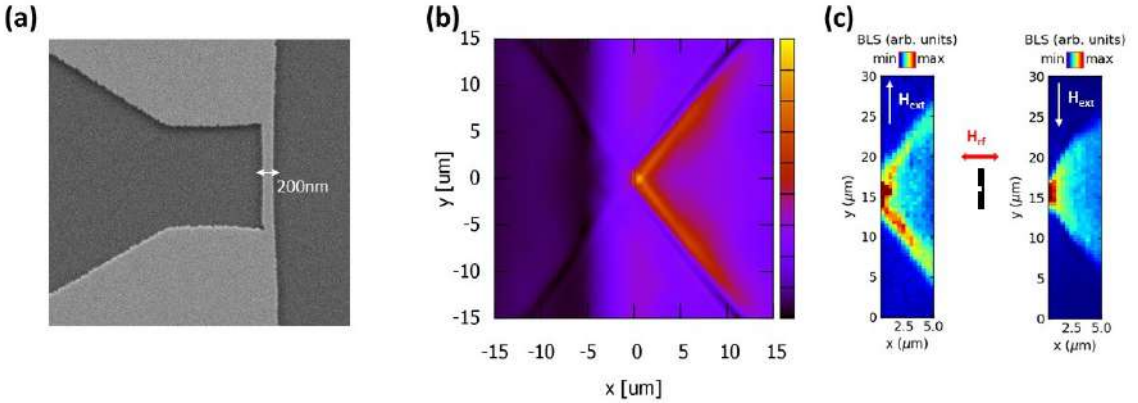



Figure 1: (a) Scanning electron micrograph image of a 200nm wide constriction. (b) Near-field diffraction simulation, and (c) BLS measurement of spin-wave caustic at f=7.5GHz and $\boldsymbol{\mu_0 H_{ext}}$ =187mT applied along y-axis.

## References

[1] T. Schneider, A. A. Serga, A. V. Chumak, C. W. Sandweg, S. Trudel, S. Wolff, M. P. Kostylev, V. S. Tiberkevich, A. N. Slavin, and B. Hillebrands, Phys. Rev. Lett. **104**, 1 (2010)

[2] F. Heussner, M. Nabinger, T. Fischer, T. Brächer, A. A. Serga, B. Hillebrands and P. Pirro, Phys. Status Solidi Rap. Rrev. Lett. **12**, 1800409, (2018)

[3] V. Vlaminck, L. Temdie, V. Castel, M. B. Jungfleisch, D. Stoeffler, Y. Henry, and M. Bailleul, "Spin wave diffraction model for perpendicularly magnetized films", J. Appl. Phys. **133**, 053903 (2023)

[4] N. Loayza, M. B. Jungfleisch, A. Hoffmann, M. Bailleul, and V. Vlaminck, Phys. Rev. B **98**, 144430 (2018)

# Spin Wave Resonance Probed via Acoustoelectric Current in a Metallic Thin Film

*Hiroki Matsumoto[1*], Takuya Kawada[1], Masashi Kawaguchi[1], and Masamitsu Hayashi[1]*

[1] *Department of Physics, The University of Tokyo, Tokyo, Japan*

*hmatsumoto@g.ecc.u-tokyo.ac.jp

Surface acoustic wave (SAW) on a ferroelectric substrate can drive a spin wave in a ferromagnetic thin film deposited on the substrate [1][2]. The phonon-magnon coupling in the SAW device will provide a new functionality for conventional magnonic devices. For example, nonlinear higher harmonic generation of spin waves was experimentally demonstrated by applying SAWs with the power of up to ~1 W to magnetic semiconductors [3] and metals [4], meaning that parametric magnon pumping, which has been studied in a $Y_3Fe_5O_{12}$ film on a $Ga_3Gd_5O_{12}$ substrate [5], can be achieved by the nonlinear magnetoacoustic dynamics. Furthermore, parametric amplification of spin waves with SAW application was numerically studied very recently [6].

To observe parametric magnons, the magneto-optical detection scheme [3] and Brillouin light scattering [4] have often been employed, where elaborate time- and space-resolved measurements are needed. On the other hand, scattering matrix measurement using vector network analyzer (VNA) [1][2] is commonly used to study SAW-spin wave conversion because it is simple and fast. SAW power is transferred to the spin waves and the power reduction is detected from the change in the transmission. The method, however, cannot be applied to probe nonlinear magnetoacoustic process because the detectable SAW frequency is limited by the pitch of the IDT fingers. To investigate nonlinear magnetoacoustic effects, a simple experimental way to sense SAW power conversion in a wide frequency range is required.

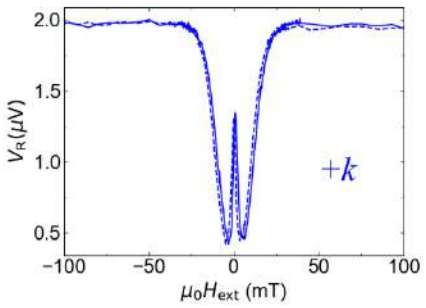



Fig: The external magnetic field dependence of the DC voltage in a metallic thin film placed on the right side of the ferromagnetic thin film. SAW propagates from the left to the right.

In this work, we experimentally demonstrate DC detection of acoustically-driven spin wave resonance in a SAW device with a ferromagnetic thin film. Instead of probing SAW-spin wave conversion from IDT with VNA, we use a metallic thin film deposited on the SAW delay line as a detector. Previous studies showed that a direct current is generated in a conductive film on a piezoelectric substrate when SAW traverses the film, which is known as acoustoelectric current and was reported to be proportional to the SAW power. We use the characteristics of the acoustoelectric current to detect the SAW power reduction associated in SAW-spin wave conversion.

**Acknowledgements**

We thank T. Funato and M. Matsuo for fruitful discussion. This work was partly supported by JSPS KAKENHI (Grant Nos. 20J20952) from JSPS, and JSR Fellowship, the University of Tokyo.

**References**

[1] M. Weiler, L. Dreher, C. Heeg, H. Huebl, R. Gross, M.S. Brandt, S.T.B. Goennenwein, *Phys. Rev. Lett.* **106** (2011) 117601.
[2] L. Dreher, M. Weiler, M. Pernpeintner, H. Huebl, R. Gross, M.S. Brandt, S.T.B. Goennenwein, *Phys. Rev. B* **86** (2012) 134415.
[3] M. Kraimia, P. Kuszewski, J.-Y. Duquesne, A. Lemaître, F. Margaillan, C. Gourdon, L. Thevenard, *Phys. Rev. B* **101** (2020) 144425.
[4] M. Geilen, R. Verba, A. Nicoloiu, D. Narducci, A. Dinescu, M. Ender, M. Mohseni, F. Ciubotaru, M. Weiler, A. Müller, B. Hillebrands, C. Adelmann, P. Pirro, *arXiv*:2201.04033 (2022).
[5] A.A. Serga, A.V. Chumak, B. Hillebrands, J. Phys. D: Appl. Phys. **43** (2010) 264002.
[6] M. Mohseni, A. Hamadeh, M. Geilen, P. Pirro, *arXiv*:2302.10614 (2023).

# Magnon-Phonon Coupling in Polycrystalline Metallic Thin Films

*M. Müller[1,2], J. Weber[1,2], F. Engelhardt[3,4,5], V.A.S.V. Bittencourt[6], T. Luschmann[1,2,7], M. Cherkasskii[5], S.T.B. Goennenwein[8], S.Viola Kusminskiy[5,3], S. Geprägs[1], R. Gross[1,2,7], M. Althammer[1,2,]*, H. Huebl[1,2,7]*

[1] *Walther-Meißner-Institut, Bayerische Akademie der Wissenschaften, Garching, Germany*
[2] *Technische Universität München, TUM School of Natural Sciences, Physik-Department, Garching, Germany*
[3] *Max Planck Institute for the Science of Light, Erlangen, Germany*
[4] *Department of Physics, University Erlangen-Nuremberg, Erlangen, Germany*
[5] *Institute for Theoretical Solid State Physics, RWTH Aachen University, Aachen, Germany*
[6] *ISIS (UMR 7006), Université de Strasbourg, Strasbourg, France*
[7] *Munich Center for Quantum Science and Technology (MCQST), München, Germany*
[8] *Department of Physics, University of Konstanz, Konstanz, Germany*
*matthias.althammer@wmi.badw.de

Magnetoelastic coupling between excitation modes of the spin system (spin waves) and the lattice (phonons) is of interest from a fundamental perspective and can enable mode hybridization. For quantum sensing and transduction protocols, excitation exchange between the magnetic and elastic systems is of importance, but typically requires strong coupling between the magnetic and elastic modes. Here, we present our current results on coupling the magnetization dynamics of a Kittel mode in a ferromagnetic thin film to a high-overtone bulk acoustic resonator. We show that the typically weak coupling affects the magnetization dynamics of the magnetic layer and can thereby be characterized with high sensitivity using ferromagnetic resonance spectroscopy [1]. In our experiments, we investigate the magnetoelastic coupling of polycrystalline metallic thin films deposited on silicon and sapphire substrates via DC sputter deposition as a function of microwave excitation frequency and substrate material by performing broadband ferromagnetic resonance spectroscopy. Utilizing a model based on coherent magnetoelastic coupling and the phononic properties of the substrate material, we obtain a full description of the observed changes in ferromagnetic resonance as a function of microwave frequency. Furthermore, we will discuss the implications of this model for the phononic transport of angular momentum.

**Acknowledgements**

We gratefully acknowledge financial support from the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) under Germany's Excellence Strategy – EXC-2111 – 390814868.

**References**

[1] R. Schlitz et al., Phys. Rev. B **106**, 014407 (2022).

# Spin-wave dynamics in $Co_2MnSi$ Heusler magnonic crystals consisting of square antidot lattices

*Marco Madami[1], S. Mantion[2], N. Biziere[2], Silvia Tacchi[3]**
[1] *Dipartimento di Fisica e Geologia, Università di Perugia, I-06123 Perugia, Italy*
[2] *CEMES-CNRS, Université de Toulouse, 29 rue Jeanne Marvig, Toulouse, 31055, France*
[3] *Istituto Officina dei Materiali del CNR (CNR-IOM), Sede Secondaria di Perugia, c/o Dipartimento di Fisica e Geologia, Università di Perugia, I-06123 Perugia, Italy*
**tacchi@iom.cnr.it*

Co-based Heusler alloys represent an interesting class of materials for application in the field of magnonics, due to their unique properties. For example $Co_2MnSi$ is characterized by high Curie temperature, low damping and high saturation magnetization, which enables to operate at frequencies in the GHz range at remanence. However, only a few studies about magnonic crystals (MCs) made of Co-based Heusler alloys are present in literature. In this work we have investigated spin wave (SW) dynamics in MCs, consisting of $Co_2MnSi$ antidots (ADs) arrays, formed by square holes, having a side of 200 nm and arranged into square matrix having a period of 0.8 μm and 1 μm. The samples have been fabricated from a 50 nm thick $Co_2MnSi$ film, by e-beam lithography followed by Ar+ ion beam etching (IBE). Micro-focused Brillouin light Scattering (micro-BLS) has been used to map the spatial profile of the spin-wave modes supported by the ADs lattices. All micro-BLS measurements have been performed exciting SWs by means of an inductive antenna and applying an external magnetic field, H, parallel to the side of the square matrix and to the antenna. For both the investigated samples we have experimentally observed an extended mode propagating in the channels of continuous $Co_2MnSi$ film comprised between the square holes and a localized mode located in the regions between adjacent holes (see Fig.1). BLS measurements have been compared to MuMax micromagnetic simulations, obtaining a good agreement between the experimental results and the calculated frequencies and spatial profiles.

**Acknowledgements**
This work is performed using HPC resources from CALMIP (Grant No. 2022-p1554). Financial support from the italian Ministery of University and Research through the PRIN-2020 project entitled "The Italian factory of micromagnetic modeling and spintronics", (cod. 2020LWPKH7) is kindly acknowledged.

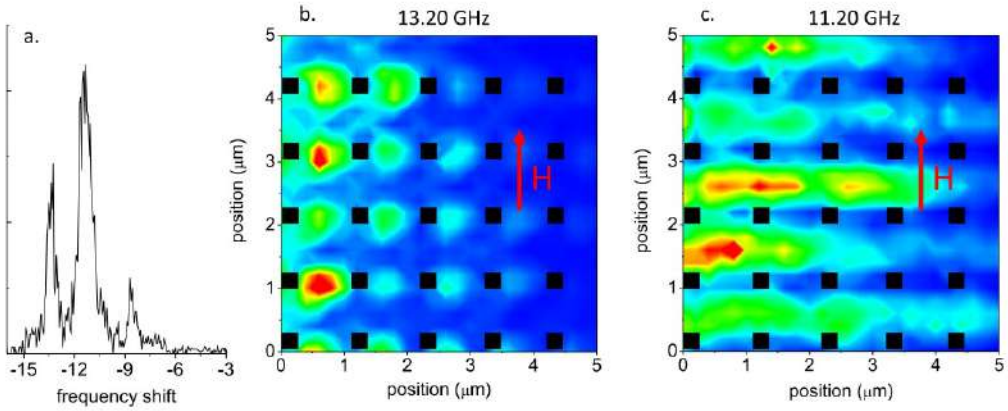



**Fig.1** Micro-BLS measurements of the sample having a period of 1μm performed applying a magnetic field of H=1 kOe. (a) Micro-BLS spectrum. (b) and (c) Areal maps of BLS intensity, recorded from the antenna edge (positioned on the left side of the graphs) at excitation frequencies of 13.20 GHz and 11.20 GHz, respectively.

# MOKE and VSM-FORC magnetometry of SAF/non-magnetic/stripe-domain textured magnet multilayers for reconfigurable spin wave transport.

*J. Díaz[1,2], D. Martínez-Blanco[1], M. Vélez[1,2], J. Rodríguez-Fernández[1], V. V. Fernández[1] and L. M. Alvarez-Prado[1,2*]*

[1]*Departamento de Fisica, Facultad de Ciencias, Universidad de Oviedo, C/ Federico García Lorca 18, Oviedo 33007, Spain*

[2]*Centro de Investigacion en Nanomateriales y Nanotecnologia (CINN), Av. De la Vega 4-6, El Entrego 33940, Spain*

*lmap@uniovi.es

Hybrid magnetic multilayers for reconfigurable spin wave transmission are made of two magnetic layers separated by a non-magnetic spacer to ensure magnetic dipolar coupling. One of the magnetic layers is in charge of the spin wave transport properties, whereas the other one has a reconfigurable magnetic texture. Its stray field is imprinted in the spin-wave transmitter changing its spin-wave dispersion. One of the most promising magnetic textures in this kind of spin-wave devices is that of stripe domains due to its regular periodicity, acting as a magnonic crystal [1]. Ferromagnetic resonance (FMR) and Brillouin scattering (BLS) proved strong hysterical and asymmetric features in this system [1,2]. We propose to enhance the resonant energies of the transmitted spin-waves and the configurability of the system by substituting the transmitter layer, made of a 10 nm thick permalloy (Py) layer, by a synthetic antiferromagnet made of a 15 nm thick Py/Ruthenium/Py multilayer, which was deposited on top of a 65 nm thick NdCo layer capped by 5 nm thick layer of ambient oxidized silicon. The understanding of the system requires a fine characterization of the magnetic interactions involved between layers and within the layers. For this purpose, we characterized the system using the transverse magneto-optic Kerr effect (T-MOKE), which is sensitive to the top layer (the spin wave transmitter in this case) and Vibrating Sample Magnetometry (VSM), which is bulk sensitive. The onset of the interlayer and intralayer interactions was characterized using minor loops and First Order Reversal Curve (FORC) measurements. Samples with different antiferromagnetic exchange energies were investigated. A clear and expected modification in the coercivity of the spin-wave transmitter layer is observed as the AF interaction is increases. A detailed information of the different interaction and coercive fields that appear in the system are obtained by FORC and the minor loops, demonstrating the relevant role of the in-plane component of the magnetization of the magnetic textured layer in the hysteretic and asymmetric effects observed in the spin-wave transmission in these systems.

**Acknowledgements**

This study has received financial support from the Spanish Ministry of Science and Innovation (Project No. PID2019-104604RB/AEI/10.13039/501100011033), the regional FICYT funding (Grant No. GRUPIN-AYUD/2021/51185) with the support of FEDER funds.

**References:**

[1] D. Markó et al., Appl. Phys. Lett. 115, 082401 (2019)
[2] K. Szulc et al. ACS Nano 16 (9), 14168–14177 (2022)

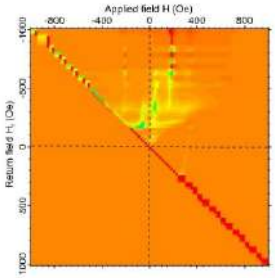



Figure 1. FORC measurement of a thin film made of NdCo (65 nm)/Si (5 nm)/Py (10 nm)/Ru (0.5 nm)/Py (5 nm)

# The Spin-wave Symmetry Analysis in Confined Rectangular $Ni_{80}Fe_{20}$ Microstrips under Uniform MW Excitation

*Santa Pile[1*], Andreas Ney[1], Kilian Lenz[2], Ryszard Narkowicz[2], Jürgen Lindner[2], Sebastian Wintz[3], Johannes Förster[3], Sina Mayr[4], Markus Weigand[5]*

[1]*Johannes Kepler University Linz, Linz, Austria*
[2]*Helmholtz-Zentrum Dresden-Rossendorf, Dresden, Germany*
[3]*Max Planck Institute for Intelligent Systems, Stuttgart, Germany*
[4]*Paul Scherrer Institut, Villigen PSI, Switzerland*
[5]*Helmholtz-Zentrum Berlin Für Materialien Und Energie, Berlin, Germany*
* santa.pile@jku.at

The design of a microstructure can affect the internal field distribution and, therefore, the spin-wave (SW) behaviour. That potentially can be used for information transport and processing [1]. In general, the confinement of the structure leads to the quantization of SW k-vectors in the direction of confinement [2]. Under the uniform excitation only SW eigenmodes with an odd number of nodes are expected. This results in a symmetric interference pattern. Changes in the geometry of the structure, such as a presence of an additional rectangular microstrip, can cause the "breaking" of the symmetry [3].

In this work the asymmetry quantification by an asymmetry parameter (AP) of SW dynamics in confined rectangular microstrips is suggested and applied to the TR-STXM [4] results and micromagnetic simulations. 30 nm thick $Ni_{80}Fe_{20}$ microstrips with nominal rectangular 1x5 $\mu m^2$ cross section were researched using microantenna-based FMR [5-7], TR-STXM and MuMax3 [8] micromagnetic simulations. AP indicates a deflection of a central profile of an interference pattern from the mirror-symmetric state, which in turn is an indicator that there is an asymmetry in the interference pattern itself. A mirror-symmetric profile here is a profile, which is invariant under a reflection about the line in its center. AP for a profile consisting of normalized data values $\{x_n\}_{n=1}^{N}$ is calculated as follows:

$$AP = \frac{1}{[N/2]} \sum_{n=1}^{[N/2]} |x_n - x_{N-n+1}|,$$

where one half of the profile is subtracted from the other its half point by point, and then the mean value of the absolute values of all differences is taken. If profile is symmetric, that results in $AP = 0$. In this work profiles of the out-of-plane component of the dynamic magnetization are analyzed [7]. The frequency of the uniform MW magnetic field was fixed at 9.43 GHz, while the external static magnetic field was varied from 250 mT to 0 mT. The results show a higher asymmetry for a strip in easy axis orientation when a second perpendicular microstrip is placed at the distance of 2 µm, compared to the case when a single microstrip is analysed.

## Acknowledgements

We thank the Helmholtz-Zentrum Berlin for the allocation of synchrotron radiation beamtime.We also thank T. Feggeler and H. Stoll for their help during the TR-STXM measurements and A. Halilovic for valuable contributions to the lithography process. Additionally, we would like to thank M. Bechtel for technical support at the beamline. The authors would like to acknowledge financial support from the Austrian Science Fund, Project No. ESP 4 and project No. I-3050 during the early stage of the work. S.M. would like to acknowledge funding from the Swiss National Science Foundation (SNSF) under Grant No. 172517.

## References

[1] A. V. Chumak et al., *IEEE Transactions on Magnetics* **58** (2022) 1–72.
[2] C. Bayer et al., *Phys. Rev. B* **72** (2005) 064427.
[3] P. Clausen et al., *Appl. Phys. Lett.* **99** (2011) 162505.
[4] H. Stoll et al., *Front. Phys.* **3** (2015) 26.
[5] R. Narkowicz et al., *J. Magn. Reson* **175** (2005) 275–284.
[6] S. Pile et al., *Appl. Phys. Lett.* **116** (2020) 072401.
[7] S. Pile et al., *Phys. Rev. B* **105** (2022) 094415.
[8] A. Vansteenkiste et al., *AIP ADVANCES* **4** (2014) 107133.

# Angular Dispersions of the Main-Dipole, Standing-Waves and Zero-Field FMR Modes in Thin Permalloy Films

*Lucy Quinn[1], Niclas Teichert[1], Brian Shortall[1] and P. Stamenov[1]**

[1] *School of Physics and CRANN, Trinity College, Dublin 2, Ireland*

*stamenop@tcd.ie

Magnetisation, anisotropy and exchange stiffness are three paramaters of critical importance for both the practical device applications of magnetic thin films and the micromagetic simulation and optimisation of structures. While all three properties can be assessed by simple or more involved forms of bulk magnetometry, in principle, the sensitivity drops at small film thickness and alternatives based on FMR gain advantages. In essentially dielectric films, where both conductivity-related and resonance damping losses are low, such as YIG, large number of standing waves modes are observable, which aids the extraction of exchange stiffness [1]. In metallic systems, especially in ones with large anisotropy and damping, often a single additional mode (not the main dipole one) is observable, which makes parameter extraction more difficult [2].

Here we extend the approach of explicit fitting of multiple standing wave resonance modes, with field applied normal to the surface [3] to the complete interpretation of all available resonance modes and their angular dispersions. This is illustrated with examples of Py (80:20) films prepared by DC magnetron sputtering in a Shamrock deposition tool with base pressure of $10^{-7}$ mBar, at thickness below 30 nm. The key measurements are performed in a Bruker EMX 300 X-band spectrometer, equipped with an electromagnet ($\mu_0 H_{max} = 0.9$ T) and a resonance cavity ($Q > 7000$), with supporting a rotationally symmetric cylindrical mode.

For sufficiently thin films (the figure shows data collected on a 15 nm film), three distinct classes of modes can be measured and modelled: the main dipole mode, dispersing in a negative, demagnetisation-limited fashion, with the highest resonance induction reaching 0.6 T and peaking close to the polarisaiton value, close to 90° or essentially normal to the film surface. The three standing wave resonance modes, which are clearly resolved $n$ = 0, 1 and 2, are dispersing in a sinusoidal fashion, with breath of the dispersions varying approximately quadratically, as $\sim n^2$. The $n$ = 0 mode dispersion is small, but clearly negative, while the ones for $n$ = 1 and 2 are strongly positive. The zero-field resonance mode has a dispersion, which is dominated by a $\cos^2$ term. The apparent amplitudes of all modes depend on both their damping factors, which scale close to linearly in field and essentially quadratically as a function of $n$, for the standing wave ones. This effective approach allows for the complete modelling of the modal dispersions, as illustrated on the figure, and the accurate extraction of the critical parameters ($M_s$ = 1.035 A/m, $A_{ex}$ = 0.15 pJ/m, $N_{eff}$ = 0.98).

**Acknowledgements**

Financial support from SFI under the MANIAC programme is gratefully acknowledged.

## References

[1] S. Klingler, A. V. Chumak, T. Mewes et al, *J. Phys. D: Appl. Phys.* **48** (2015) 015001.
[2] C. Antoniak, J. Lindner, K. Fauth *et al*, *Phys. Rev. B* **82** (2010) 064403.
[3] P. Talagala, P. S. Fodor, D. Haddad *et al*, *Phys. Rev. B* **66** (2002) 144426.

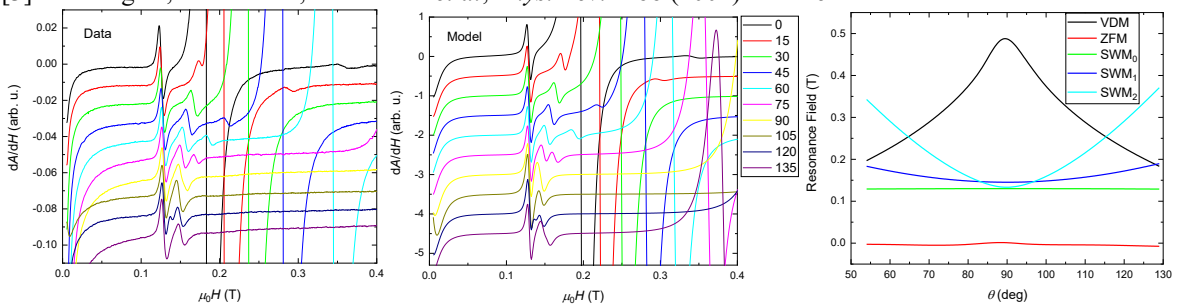



Figure: Experimentally measured X-Band FMR spectra for a 15 nm Py film (left panel) and their corresponding model analogues (middle panel). Note that only the lower field region is shown and the main dipole mode is out of scale, to promote the visibility of the much smaller in amplitude standing wave and zero-field resonance modes. The individual dispersions for all modes observed are exhibited on the (right panel). All angles are indicated with respect to the substrate (at 90 degree the applied field is normal to the film).

# Controlling magnon transport by drift currents

*Saül Vélez*

[1] *Condensed Matter Physics Center (IFIMAC), Instituto Nicolás Cabrera, and Departamento de Física de la Materia Condensada, Universidad Autónoma de Madrid, E-28049 Madrid, Spain*

*saul.velez@uam.es

Magnetic insulators (MIs), such as rare earth garnets ($R_3Fe_5O_{12}$; R=Y,Tm,..), have attracted a lot of interest because of their low Gilbert damping and high-frequency dynamics. Interestingly, unlike charge currents, spin currents can couple and propagate through MIs, making possible to realize spintronic devices based on these materials.

In this talk, we will discuss how we can exploit the charge to spin conversion phenomena in heavy metals for electrically injecting and detecting magnon currents carrying spin information in ferrimagnetic garnets such as $Y_3Fe_5O_{12}$ (YIG) and $Tm_3Fe_5O_{12}$ [1,2]. We will also see how we can induce magnon drift currents controlling the transport by exploiting antisymmetric chiral interactions and thermal gradients [3,4]. In particular, we will demonstrate that the interfacial Dzyaloshinskii-Moriya interaction naturally occurring at the garnet/oxide substrate and garnet/metal interfaces [5] can modulate the magnon propagation length with measured values up to 6% in YIG thin films [3]. Finally, we will show that large drifts can also be induced by thermal gradients.

## Acknowledgements

S.V. acknowledges support by IFIMAC through the Maria de Maeztu research Grant No. CEX2018-000805-M, by the Spanish Ministry of Science and Innovation via Grant No. PID2021-122980OA-C53 and by the Comunidad de Madrid through the Atracción de Talento Grant No. 2020-T1/IND20041.

## References

[1] J. M. Gómez-Pérez et al. Phys. Rev. B **101**, 184420 (2019)
[2] J. Gao et al., Phys. Rev. Res. **4**, 043214 (2022).
[3] R. Schlitz et al. Phys. Rev. Lett. **126**, 257201 (2021).
[4] S. De-la-Peña et al., J. Phys: Cond. Matt. 34, 295801 (2022).
[5] S. Vélez et al., Nat. Comm. **10**, 4750 (2019) and Nat. Nanotech. **17**, 834 (2022).

# Strain in Ferrimagnets of the MnRuGa Family. Quasi-static Magnetisation Dynamics, Magnonic, Phononic and Combined Excitations

*Jack O'Brien[1], Ümit Dağlum[1], Maria Stamenova[1] and Plamen Stamenov[1]**

[1] *School of Physics and CRANN, Trinity College, Dublin 2, Ireland*

*stamenop@tcd.ie

Low- and zero-moment, highly spin-polarized Mn-based ferrimagnets, such as $Mn_3Ga$, $Mn_3Al$, $Mn_2AlGa$ and the prototypical $Mn_2RuGa$ (MRG) family have been investigated both theoretically and experimentally, as of their potential applications in THz resonance devices [1] relying on either MTJs and CPP transport [2] or Hall bar type structures [3], and using both STT and/or SOT. Ultra-fast depolarisation and magnetisation all-optical switching, using single pulses, have also been demonstrated [4]. Understanding, control and optimisation of the phenomenology requires that an effective spin-lattice model is constructed, within a divide-and-conquer strategy, substantially cheaper to compute dynamical properties in the time domain compered to state-of-the-art time-dependent density functional theory (TD DFT).

Here we use *ab initio* (DFT) calculated parametrisation of the exchange integrals and anisotropy components, for the perfectly ordered structures. We take three approaches: (i) direct integration of the equations of motion for effective classical spins; (ii) approximation of frozen magnons and phonons; and (iii) Monte Carlo dynamics: for recovering the phonon and magnon dispersion relations, the quasi-static magnetisation curves (rotational torque curves and hysteresis). Distance-dependent long-range exchange couplings; 2- and 4-fold anisotropy; Zeeman coupling; elastic coupling coefficients; are all taken into account.

The effects of compressive and tensile strain can be traced (see figure) for the phonon, magnon and combined excitations. The system exhibits crossing of the in- and out-of-phase magnon branches with clear increment in both effective masses for compressive strain. The combined elastic-magnetic excitations can be fully mapped, allowing for the purposeful engineering of the anisotropy gaps and dispersions crossings in ultra-thin layers of off-compensated ferrimagnets, thus opening the road to the calculation (prediction) and potential practical exploitation of magnetic dipole emission in the 0.3 – 3 THz bands.

**Acknowledgements**

Financial support from SFI under the MANIAC and ATMOST programmes is gratefully acknowledged.

**References**

[1] R. E. Troncoso, K. Rode, P. Stamenov, J. M. D. Coey and Arne Brataas, *Phys. Rev. B* **99** (2019) 054433

[2] A. Titova, C. Fowley, E. Clifford *et al*, *Sci. Reports* **9** (2019) 4020

[3] N. Thiyagarajah, Y.-C. Lau, D. Betto, K. Borisov *et al*, *Appl. Phys. Lett.***106** (2015) 12402

[4] C. Banerjee, N. Teichert, K. E. Siewierska *et al* , *Nature Comm.* **11** (2020) 1-6

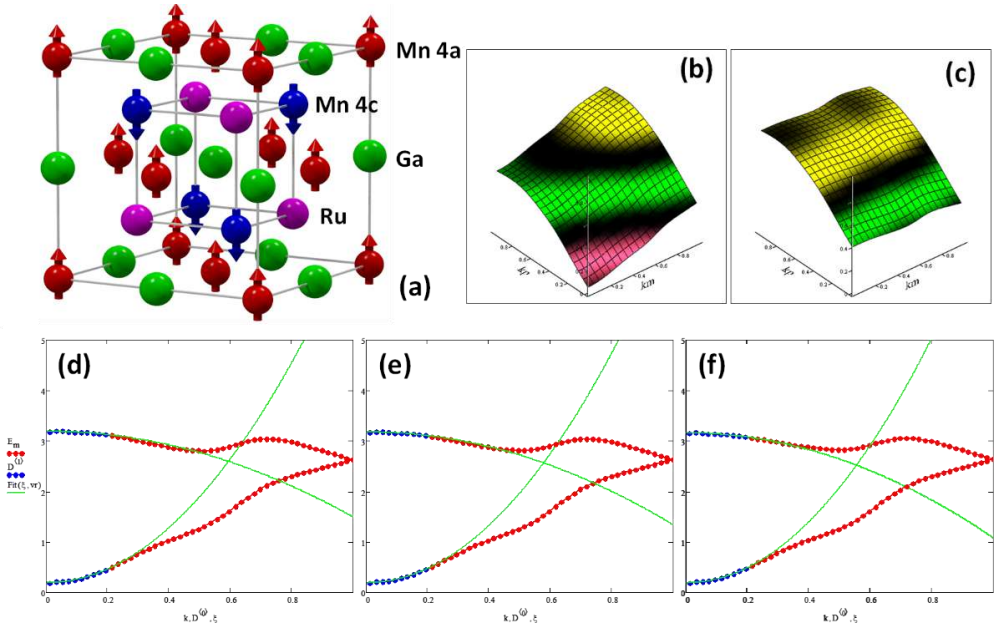



Figure: Fig. 1 (a) The structure of MRG. The energy for arbitrary momenta, for the in-phase modes (b) and out-of-phase modes (c). (d-f) The in-phase (acoustic) and out-of-phase (optical) magnon dispersions along the <001> axis, for *c*-axis lattice parameter of 6.10 Å, 6.18 Å (experimental), and 6.30 Å. The relative change in the magnon $m_{eff}$ is ~ -15 %, for the antiphase (AF) magnonic branch and ~ -7 %, for the in-phase (FM) one, for a 3.3 % lattice expansion.

# Ultrafast Demagnetisation of 3d Ferromagnets in Heat Conserving Three-Temperature Model

*M. Pankratova[1*], I. P. Miranda[1], D. Thonig [2,1], M. Pereiro[1], E. Sjöqvist[1], A. Delin [3], O. Eriksson[1], and A. Bergman[1]*

[1] *Department of Physics and Astronomy, Uppsala University, Box 516, SE-75120 Uppsala,*
[2] *School of Science and Technology, Örebro University, SE-701 82, Örebro, Sweden*
[3] *Department of Applied Physics, School of Engineering Sciences, KTH Royal Institute of Technology, AlbaNova University Center, SE-10691 Stockholm, Sweden*
*maryna.pankratova@physics.uu.se

The discovery of ultrafast demagnetization by Beaurepaire and coauthors [1] around thirty years ago allowed the manipulation of magnetization using laser pulses on picosecond timescales. Their work started a new and quickly growing research field with numerous promising applications in all-optical magnetization switching, magnetic data storage, and spintronics. Following Beurepaire's work on nickel, ultrafast demagnetization was studied in other 3d ferromagnets, in particular, many theoretical and experimental investigations have been focused on the magnetization dynamics of cobalt and iron. In a recent work [2] the ultrafast magnetization dynamics of nickel, iron, and cobalt were studied and compared, using the same experimental setup and sample thicknesses for all three metals. It was shown that demagnetization scales linearly with laser fluence and, moreover, the demagnetization rate of nickel is the largest followed by iron and cobalt. The authors connect this linear dependence with a reduction of interatomic exchange.

Our work is focused on the ultrafast demagnetization of nickel, iron, and cobalt and, in particular, on their demagnetization reduction with laser fluence in the heat-conserving three-temperature model (HC3TM) proposed in Ref.[3]. We have previously applied this theory to simulate the ultrafast demagnetization of nickel [3]. In Ref.[3] we showed that HC3TM describes the demagnetization of nickel on subpicosecond timescales better than Beaurepaire's three-temperature model. In addition, in contrast to Beaurepaire's three-temperature model, HC3TM does not rely on heat-transfer coefficients. These coefficients are hard to estimate and, therefore, make the direct comparison of simulations with experimental observations very challenging [3].

In this work, we perform atomistic spin-lattice dynamics simulations to study ultrafast demagnetization dynamics of iron, nickel, and cobalt using HC3TM with parameters, such as exchange interaction, spin-lattice coupling, and Gilbert damping, obtained from ab-initio simulations. Our results show that using HC3TM we can correctly, on a quantitative level, reproduce ultrafast magnetization dynamics of iron and cobalt, especially on subpicosecond timescales. The positions of the magnetization minima in these simulations are in agreement with experimental data. In addition, we show the importance of lattice dynamics in studying ultrafast processes, even when spin-lattice coupling itself is not significant. Finally, we demonstrate that by using HC3TM one obtains a linear scaling of the demagnetization amplitude with laser fluence, a finding also observed in experiments, even without considering the reduction of interatomic exchange. Moreover, we show that the demagnetization rate is the highest for nickel and the smallest for cobalt, which is in agreement with experimental results [2]. We confirm that the rate is dependent on Gilbert and lattice damping values, however, the linear trend remains unchanged for realistic damping values. Overall, the HC3TM provides qualitatively accurate ultrafast demagnetization dynamics for iron, cobalt, and nickel and results in a linear scaling of demagnetization with laser fluence, as observed experimentally [2].

**References**

[1] E. Beaurepaire, J.-C. Merle, A. Daunois, and J.-Y. Bigot, Phys. Rev. Lett., **76**, (1996), 4250.
[2] P. Scheid, J. Hohlfeld, G. Malinowski, A. Bergman, O. Eriksson, S. Lebegue, and S. Mangin, arXiv preprint arXiv:2301.01055, (2023).
[3] M. Pankratova, I. P. Miranda, D. Thonig, M. Pereiro, E. Sjöqvist, A. Delin, O. Eriksson, and A. Bergman, Phys. Rev. B, **106**, (2022), 174407.

# Ultrafast Magnetization Dynamics in Arrays of Dipolar-Coupled Permalloy Nanostructures

*S. Parchenko[1,2,3], D. Pecchio[1,2]*, K. Hofhuis[1,2,4], S. Sahoo[1,2], V. Scagnoli[1,2] and L. Heyderman[1,2]*

[1]*Laboratory for Mesoscopic Systems, Department of Materials, ETH Zurich, 8093 Zurich, Switzerland*

[2]*Laboratory for Multiscale Materials Experiments, Paul Scherrer Institute, 5232 Villigen PSI, Switzerland*

[3]*European XFEL GmbH, Holzkoppel 4, 22869 Schenefeld, Germany*

[4]*Laboratory for Nano and Quantum Technologies, Paul Scherrer Institute, 5232 Villigen PSI, Switzerland*

*davide.pecchio@psi.ch

Laser-induced quenching of magnetization in metallic multilayers has attracted considerable interest since the first demonstration of sub-picosecond demagnetization in nickel thin films [1]. Despite more than two decades of research on this subject, the proposed microscopic mechanisms underpinning ultrafast demagnetization are not yet fully established [2]. Ascertaining the physical origin of such processes would pave the way to an exquisite control of the magnetization dynamics induced by ultrashort laser pulses, potentially resulting in a faster and more efficient storage of information.

As most of the experiments conducted so far have been carried out on magnetic thin films, little attention has been paid to the role played by the dipolar coupling and shape anisotropy in the ultrafast demagnetization of arrays of magnetic nanostructures. Nevertheless, scaling the magnetic material to nanometer sizes has been shown to play a significant role in their magnetization dynamics in response to an ultrafast photoexcitation [3].

Using a femtosecond laser-based time-resolved magneto-optical Kerr effect setup, we show that the optically induced magnetization suppression in arrays of parallel-oriented permalloy nanostructures can substantially differ from that of unpatterned thin films. For the thin films, there is an increase in the Kerr signal preceding the demagnetization of the material (black points in Fig. 1a), which can be ascribed to the optically induced spin transfer (OISTR) between the minority bands of Ni and Fe sublattices [4]. In contrast, the efficiency of this mechanism is highly reduced in the arrays of nanostructures (red points in Fig. 1a). Moreover, an unexpected behaviour is seen in the magneto-optical Kerr rotation as a function of permalloy thickness (see Fig. 1b). For the continuous films, as expected, the Kerr rotation decreases as the thickness of permalloy films decreases. In contrast, an unexpected saturation of the Kerr rotation is seen for the arrays of nanomagnets, pointing to the existence of a threshold. By comparing the thickness dependence for nanostructures with different dimensions, we argue that the threshold originates from a transition to the superparamagnetic state of the nanostructures.

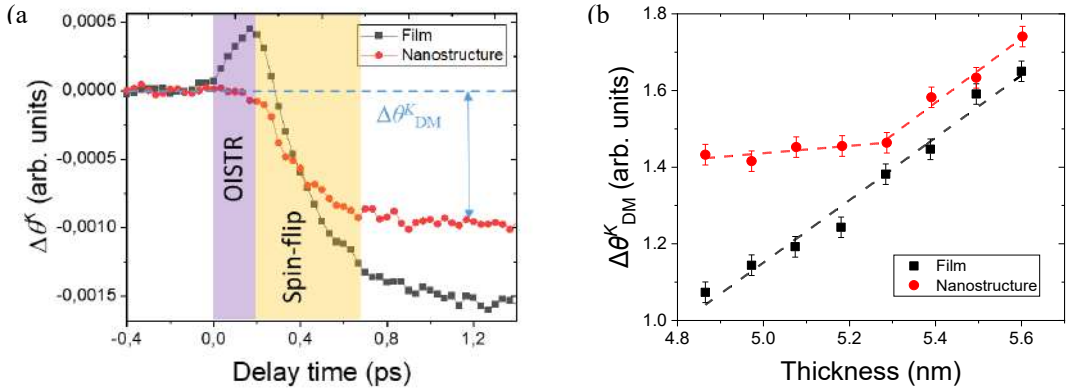



Figure 1: (a) Time-resolved changes of Kerr signal for a thin film and an array of nanostructures. (b) Change in the Kerr signal after the demagnetization process $\Delta\theta^K_{DM}$ for the thin film and array of nanostructures as a function of thickness.

## References

[1] E. Beaurepaire *et al.*, *Physical review letters*, **76** (1996), 4250–4253.
[2] A. V. Kimel and M. Li, *Nature Reviews Materials* **4** (2019), 189.
[3] J. Y. Bigot and M. Vomir, *Annalen der Physik* **525** (2013), 2.
[4] M. Hofherr *et al.*, *Science Advances* **6**, (2020) 8717.

# Nonreciprocal Transmission of Surface Acoustic Waves in Synthetic Antiferromagnets

*M. Küß[1]*, S. Glamsch[1], M. Hassan[1], Y. Kunz[2], A. Hörner[1], M. Weiler[2], and M. Albrecht[1]*

[1] *Institute of Physics, University of Augsburg, 86135 Augsburg, Germany*

[2] *Fachbereich Physik and Landesforschungszentrum OPTIMAS, Rheinland-Pfalzische Technische Universitat Kaiserslautern-Landau, 67663 Kaiserslautern, Germany*

* matthias.kuess@physik.uni-augsburg.de

Surface acoustic waves (SAWs) have made their way into many everyday devices, thanks to the greatly reduced wavelength of SAWs compared to free-space microwaves of the same frequency. These "nano earthquakes" can be efficiently launched and detected on piezoelectric substrates with periodic metallic gratings in the MHz- to GHz-range. However, SAWs are in general propagating reciprocally, which means that SAW propagation does not change under inversion of the propagation direction and limits the usage of SAWs for reciprocal devices.

Since spin waves (SWs) are known to show a pronounced nonreciprocal behavior, magnetoelastic coupling of SAWs with SWs is a straightforward approach to obtain nonreciprocal transmission of surface waves. Besides the nonreciprocity of the SAW-SW coupling mechanism itself [1,2], the SW dispersion relation can be nonreciprocal. For example, the interfacial Dzyaloshinskii-Moriya interaction (DMI) in an ultrathin ferromagnetic/heavy metal bilayer causes SW nonreciprocity and thus induces nonreciprocal SAW propagation [2].

In our recent study, we investigate the SAW-SW interaction in a synthetic antiferromagnet (SAF) composed of two ferromagnetic layers with different thicknesses separated by a thin nonmagnetic spacer layer. Because of interlayer dipolar coupling fields, the optical SW mode shows a large nondegenerate dispersion relation for oppositely propagating SWs. Due to SAW-SW interaction, we observe nonreciprocal SAW transmission in the piezoelectric/SAF hybrid device. The equilibrium magnetization directions of both ferromagnetic layers are manipulated by an external magnetic field to set a ferromagnetic, canted, or antiferromagnetic configuration. This has a strong impact on the SW dispersion, its nonreciprocity, and SAW-SW interaction [3]. The experimental results are in agreement with a phenomenological SAW-SW interaction model.

## References

[1] L. Dreher, M. Weiler, M. Pernpeintner, H. Huebl, R. Gross, M. S. Brandt, and S. T. B. Goennenwein, Phys. Rev. B **86**, 134415 (2012).

[2] M. Küß, M. Heigl, L. Flacke, A. Hörner, M. Weiler, M. Albrecht, and A. Wixforth, Phys. Rev. Lett. **125**, 217203 (2020).

[3] M. Küß, M. Hassan, Y. Kunz, A. Hörner, M. Weiler, and M. Albrecht, Phys. Rev. B **107**, 024424 (2023).

# Soft X-rays PEEM studies of magnetization dynamics driven by GHz Surface Acoustic Waves

*M. W. Khaliq[1], S. Ruiz-Gómez[1,2], M. Rovirola[3], B. Casals[3], F. Macià[3], J.M. Hernàndez[3], A. Hernández-Mínguez[4], L. Aballe[1], M.Á. Niño[1], M. Foerster[1]*

[1]*ALBA Synchrotron Light Facility, CELLS, E-08290 Cerdañola del Vallés, Barcelona, Spain*
[2]*Max Planck Institute for Chemical Physics of Solids. 01069 Dresden, Germany*
[3]*Department of Condensed Matter Physics, University of Barcelona, Barcelona-08028, Spain*
[4] *Paul Drude Institute for Solid State Electronics, Berlin-510117, Germany*

*corresponding author email address

Magnetic excitations in thin films with spatial variations at the nano- and microscale are interesting in the development of devices for high-speed and low-power signal processing compatible with existing technology. Surface Acoustic Waves (SAWs) can excite and control magnetization dynamics using electric signals and magneto-elastic coupling, and are thus a current-free alternative to magnetic fields. We have developed a method based on synchrotron X-ray Photoemission electron microscopy (XPEEM) and SAW synchronized to the X-ray pulses, allowing stroboscopic measurements of dynamic effects [1, 2], using XMCD and XMLD as magnetic contrast mechanisms. We will give an overview of the most relevant and recent results obtained: long range magneto-acoustic waves in ferromagnets [3], and the determination of the delay time in magnetic domain wall motion [1]. More recently, excitation of magneto-acoustic waves in antiferromagnetic CuMnAs has been demonstrated using linear dichroism (XMLD) (see figure below). A new sample stage connection [4] allows the application of GHz electric signals, which now opens the path to observe by XPEEM the SAW driven magnetic excitations approaching the intrinsic ferromagnetic resonance (FMR) [5].

## References

[1] M. Foerster et al., Nature Comm. 8, 407 (2017).
[2] M. Foerster et al., J. Synchroton. Rad. 26, 184 (2019).
[3] B. Casals et al., Phys. Rev. Lett. 124, 137202 (2020).
[4] M. Khaliq et al., Ultramicroscopy (2023) (under review).
[5] M. Weiler et al., Phys. Rev. Lett. 106, 117601 (2011).

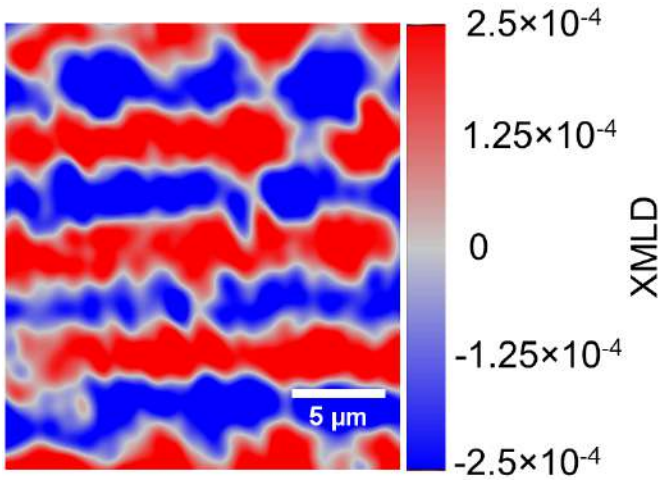



Figure : Visualization of a magneto-acoustic wave in antiferromagnetic CuMnAs obtained by stroboscopic XMLD-PEEM. The driving 500 MHz surface acoustic waves (SAW) are excited in the GaAs substrate using interdigitated transducer electrodes (IDTs).

# Analyzing and Predicting Electromagnetic Interference in Magnetic Microwire Signatures

*M. P. Marin Palacios[1,2], L. Cebada Almagro[1], R. P. del Real[2,3], X. Zhang[4], B. Lejeune[4], O. Fitchorova[4,5], A. Varma[6], R. Sundaram[6], L. H. Lewis[4,5]**

[1] *Department of Materials Physics, Complutense University of Madrid, Madrid, Spain.*
[2] *Instituto de Magnetismo Aplicado, UCM-ADIF, Madrid, Spain*
[3] *Instituto de Ciencia de Materiales de Madrid, Cantoblanco, Madrid, Spain.*
[4] *College of Engineering, Northeastern University, Boston, USA*
[5] *George J. Kostas Research Institute for Homeland Security, Northeastern University, Boston, USA*
[6] *Khoury College of Computer Sciences, Northeastern University, Boston, USA*

*corresponding author email address: lhlewis@northeastern.edu

Glass-coated amorphous magnetic microwires have the capability to act as antennae, emitting modified electromagnetic (EM) radiation when excited by incident GHz radiation [1]. While the EM signature of an individual magnetic microwire is complicated by magnetic, strain, thermal, and compositional effects, the collective response of multiple proximal microwires is yet more complex, challenging, and likely invalidating theoretical treatment of the resultant spectra [2]. Addressing this issue, a machine learning approach was applied to 225 unique sets of EM spectra measured from arrangments of multiple amorphous magnetic microwires. The response distribution of unseen microwire spectral data was reproduced to a high confidence level of 90%, with a mean square error of less than 0.01. The research applied an auto-encoder-inspired deep neural network architecture to describe the high-frequency response of two-dimensional arrangements of amorphous magnetic microwires. These microwire arrangements were interrogated to produce a set of complex spectra that were then utilized to obtain a parametrization of the response function, reducing the challenge of maximizing unique signatures to a geometric problem in parameter space. This proof-of-concept result illuminates a novel approach for scanning technologies incorporating glass-coated amorphous magnetic microwires, with the eventual goal of realizing hidden machine-readable authentication of objects and assets.

## Acknowledgments

Regional Government of Madrid under Project S2018/NMT-4321 NANOMAGCOST-CM, Northeastern University

## References

[1] D. Archilla, *et al.* (2019) *Nanomaterials,* 11(4), 920.

[2] M. Vazquez (2007). Handbook of Magnetism and Advanced Magnetic Materials.

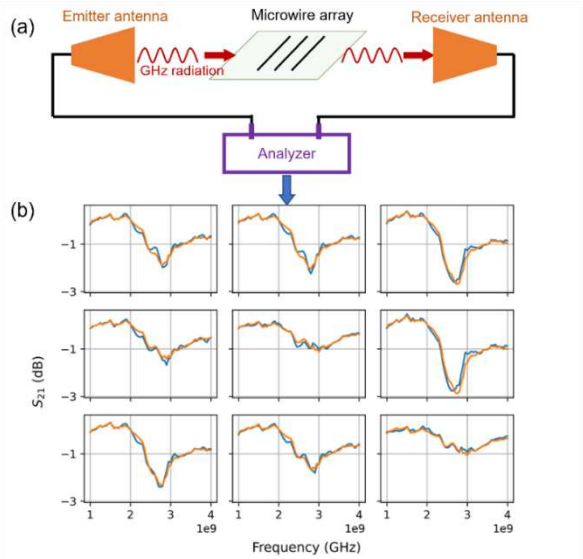



**Figure**. (a) Schematic of the microwire EM response (scattering coefficient, $S_{21}$) measurement setup; it incorporates two antennae that separately emit and receive GHz radiation to and from a two-dimensional arrangement of microwires. (b) Actual responses (blue traces) and machine learning predicted responses (orange traces) for the frequency-dependent scattering coefficient data originating from various unseen magnetic microwire arrangements.

# Reconfiguring Magnonic Devices via Permanent Micro-Magnets

*Maria Cocconcelli[1]*, Federico Maspero[2], Andrea del Giacco[1], Giulia Pavese[1], Silvia Tacchi[3], Marco Madami[4], Moritz Geilen[5], Björn Heinz[5], Philipp Pirro[5], Riccardo Bertacco[1]*

[1] *Dipartimento di Fisica, Politecnico di Milano, Milano, Italy*

[2] *Dipartimento di ingegneria civile ambientale, Politecnico di Milano, Italy*

[3] *Istituto Officina dei Materiali del CNR (CNR-IOM), Unità di Perugia, Perugia, Italy*

[4] *Dipartimento di Fisica e Geologia, Università di Perugia, Perugia, Italy*

[5] *Rheinland-Pfälzische Technische Universität Kaiserslautern-Landau and Landesforschungszentrum OPTIMAS, Kaiserslautern, Germany*

*maria.cocconcelli@polimi.it

Emerging communication protocols, such as 5G, are spreading rapidly and call for the development of new technologies that can ensure strong bandwidth selectivity and high-frequency data transmission [1]. In this context, magnonics offers the possibility of meeting these needs thanks to the flexibility of spin excitations [2], which would allow the possibility of processing, encoding and transporting RF signals with low dissipation.

However, a key challenge in realizing commercial magnonic technology is the integration with traditional electronics. In fact, to operate and reconfigure magnonic elements, a bias magnetic field is required, which until now has usually been implemented through space-consuming and power-hungry components that are difficult to integrate into commercial devices.

To overcome these difficulties we are developing tunable magnonic devices where the bias field is provided by permanent micromagnets integrated on micro-actuators. By properly designing the magnet and varying the distance between the magnet and magnonic conduits, fast field tunability can be achieved within compact and energy efficient RF components.

In this contribution we first describe a process for the integration of SmCo micromagnets on top of silicon based micro-actuators [Figure 1a)]. Then, we study the effect of the proximity of a micromagnet on the frequency of a spin-wave mode propagating in YIG by measuring with Brillouin Light Scattering the frequency shift in correspondence of the magnet position. [Figure 1b)]. A sizable frequency shift for a distance of about 6 microns between the micromagnets and the YIG film has been observed due to the local bias field produced by the magnet.

These activities are carried out within the European M&MEMS project [3], whose goal is to provide a new generation of devices well suited for microwave signal processing and radio frequency communications.

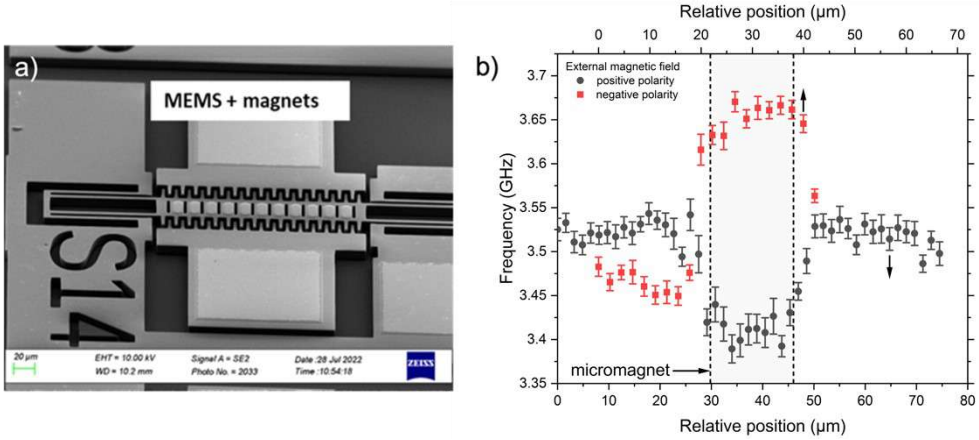



Figure 1: (a) SEM image of micro-actuator with an array of permanent magnets on top; (right) BLS measurement of the frequency shift of a spin-wave mode propagating in YIG in correspondence of the position of a SmCo magnet (area between dashed lines).

## References

[1] https://www.rfwireless-world.com/Terminology/5G-NR-Frequency-Bands.html

[2] P. Pirro, V. I. Vasyuchka, A. A. Serga, and B. Hillebrands, *Nat. Rev. Mater.* **6**, 1114 (2021).

[3] https://mandmems.eu/ (EU Project 101070536 — MandMEMS)

# Exploring of the plasmonic's benefits for magneto-optics

*Dany Zakharia[1], Nathalie Lidgi-Guigui[1], Yves Roussigné[1], Claire Goldmann[2], Raphael Weil[2], Eloi Haltz[1]**

[1] *LSPM-CNRS, UPR3407, Sorbonne Paris Nord University, Villetaneuse, France*

[2] *Université Paris-Saclay, CNRS, Laboratoire de Physique des Solides, 91405 Orsay, France*

*eloi.haltz@univ-paris13.fr

Magneto-optics (MO) consists in the modification of the state of a light beam interacting with a magnetic system. By measuring these variations, it is then possible to study the static and dynamic magnetic properties of such systems. One can cite the archetypal magneto-optical Kerr effect (MOKE) probing magnetic orders or Brillouin scattering (BLS) giving access to the dispersion relation of spin-waves. However, the optical nature of these approaches leads to intrinsic limitations in terms of signal and resolution. To overcome these limitations, we propose to take advantage of the plasmonic properties of metallic nano-objects. Surface plasmons are the collective excitations of the electronic cloud of these nano-objects that selectively and locally enhance the optical response of the systems [1]. Although plasmonic enhancement has demonstrated great effectiveness for chemical and photonic studies, its application and benefits for MO in thin film systems still need to be explored [2,3].

In this experimental study, we measure and interpret the plasmonic enhancement of gold nanoparticles for MOKE and BLS experiments. Gold nanoparticles were chemically synthesized to have their plasmon resonate at 532nm (the wavelength commonly used for MOKE and BLS experiments; as shown in Figure 1(b)) and spread on NiFe thin films (as shown in Figure 1(a) and (c)). We performed experiments to measure the evolution of the MOKE-induced rotation and ellipticity of a reflected light beam (shown in Figure 1(d)). We also quantitatively measured the magnitude of the light/spin-wave interaction using Brillouin spectroscopy measurements (see Figure 1(e)). By doing this with and without nanoparticles, we were able to quantify how the plasmon enhancement of NPs affects the light/magnetization interaction. We performed these experiments for different NP spreads (changing the NP density or promoting cluster formation) and for different wavelengths (matching or not the plasmon resonance). Finally, we have developed an effective model that describes all the experimental results obtained with a good agreement considering only a reduced number of parameters (as plotted in Figure 1(d)).

These experimental and theoretical results demonstrate that this new use of plasmonic enhancement of these techniques offers a strong improvement for MO.

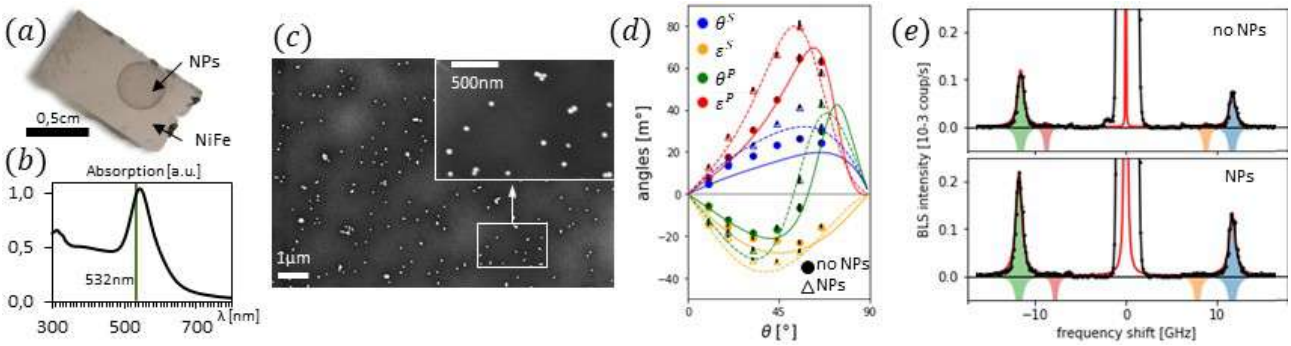



Figure 1: (a) Image of NPs spreaded on a $Si/SiOx_{(100nm)}/NiFe_{(10nm)}/Al_{(10nm)}$ thin film. (b) Absorption spectrum of the NPs with a plasmon resonance at ~532nm. (c) Electronic image of the NPs organization on the sample. (d) Variation of the MOKE-induced rotation ($\theta$) and ellipticity ($\varepsilon$) *versus* the incident angle of a 532mn light beam, *S* or *P* polarized, with or without NPs. The lines show the theortical results obtained with our effective model. (e) BLS intensity *versus* the frequency shift of the backscattered light for a 20° beam incidence, with and without NPs. Green and blue peaks show the contribution of the spin-waves.

## References

[1] M. Garcia *et al.*, *J. Phys. D: Appl. Phys.* **44** (2011) 283001.
[2] T. Loughran *et al.*, *OPTICS EXPRESS* **26** (2018) 4738.
[3] G. Armelles, *et al.*, *OPTICS EXPRESS* **16** (2008) 16104.

# Combining intersite and interlayer spin transfer in ultrafast magnetization dynamics

*Martin Anstett[1]*, Simon Häuser[1], Jonas Hoefer[1], Laura Scheuer[1], Philipp Pirro[1], Burkard Hillebrands[1], Benjamin Stadtmüller[1,2] and Martin Aeschlimann[1]*

*[1]Department of Physics and Research Center OPTIMAS, RPTU Kaiserslautern-Landau, Erwin-Schroedinger-Str. 46, 67663 Kaiserslautern, Germany*
*[2]Institute of Physics, Johannes Gutenberg University Mainz, Staudingerweg 7, 55128 Mainz, Germany*
*anstett@rptu.de

Optical excitations with femtosecond light pulses offer the intriguing opportunity to control material properties on ever-shorter timescales down to the duration of the optical excitation itself. In magnetic alloys on insulating substrates, the so-called optically induced spin transfer (OISTR) effect enables such an extremely fast manipulation of the magnetic subsystems on the sub-100 fs timescale by optical redistribution of spin carriers between the sublattices [1, 2]. On conducting substrates, however, the pure optically induced changes of the magnetic order are further influenced by (ballistic) spin transport that alters the magnetization on a very similar timescale [3, 4].

Here, we aim to reveal the mutual interplay of OISTR and spin transport on ultrafast timescales. To study those effects, time-resolved Kerr spectroscopy with fs-XUV radiation in transversal geometry (TMOKE) can be used. More specifically, we investigate the ultrafast demagnetization of a thin $Fe_{20}Ni_{80}$ alloy on a non-magnetic gold substrate. We will report on how the optically induced magnetization dynamics are altered by the spin-dependent charge transport into the Au substrate.

As an outlook, we present our new ultra-high vacuum (UHV) TMOKE setup which will enable us to explore interlayer OISTR in low-dimensional heterostructures that are only stable under UHV conditions.

## References

[1] Dewhurst et al., Nano Lett. **18** (2018) 1842-1848.
[2] Hofherr et al., Sci. Adv. **6** (2020) eaay8717.
[3] Battiato et al., Phys. Rev. Lett. **105** (2010) 027203.
[4] Hofherr et al., Phys. Rev. B **96** (2017) 100403(R).

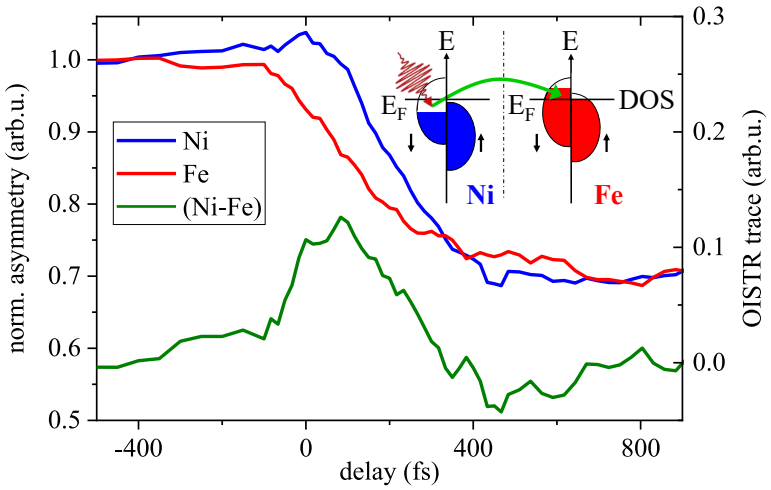



Figure: Time-resolved trace of the spin polarization of Ni (blue line) for energies under $E_F$ and of Fe (red line) for energies over $E_F$ in Py/MgO (without metallic substrate). The so called OISTR-trace corresponds to the difference between the Ni and Fe dynamics (green line, right axis). Inset: Sketch of the optical inter-side spin transfer of minority electrons of Ni into Fe within the Ni and Fe-like density of states (DOS) in Py.

# Intrinsic Energy Flow in Ultrafast Dynamics of Electrons, Phonons and Spins in $Fe_3GeTe_2$

*T. Griepe[3], Y. W. Windsor* [1,2], V.Taylor[2], D. Zahn[2], T. Pincelli[2], H. Jung[2], S.-E. Lee[2], H. Seiler[2], C. Schüßler-Langeheine[4], N. Pontius[4], Sukanya Ghosh[5], U. Atxitia[3], R. Ernsthofer[1], Laurenz Rettig[2]*

[1] *Fritz-Haber-Institut Berlin, Faradayweg 4-6, 14195 Berlin, Germany*

[2] *Technische Universität Berlin, Hardenbergstraße 36, 10623 Berlin, Germany*

[3] *Instituto de Ciencia de Materiales de Madrid, C. Sor Juana Inés de la Cruz 3, 28049 Madrid, Spain*

[4] *Helmholtz-Zentrum Berlin für Materialien und Energie, Hahn-Meitner-Platz 1, 14109 Berlin, Germany*

[5] *Uppsala University, 752 36 Uppsala, Sweden*

*windsor@fhi-berlin.mpg.de

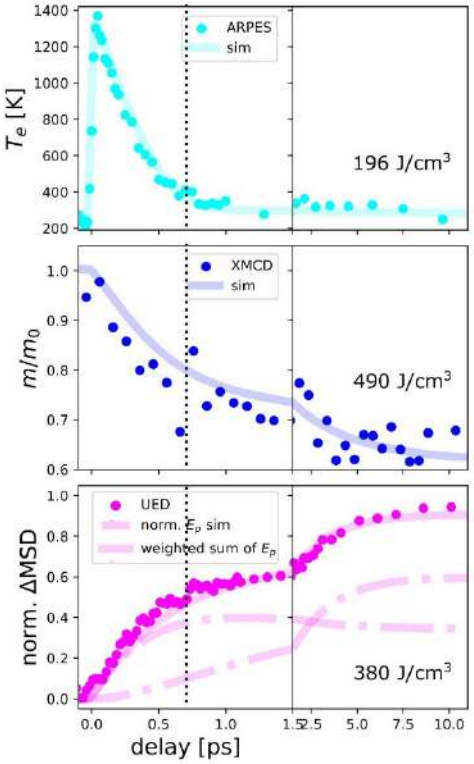



Figure 1: Ultrafast dynamics of electrons, magnetization and lattice, respectively. The dashed line highlights different dynamics before and after electron relaxation. $E_p=C_pT_p$ is the energy of phonon-subsystems. Numbers denote absorbed energy in experiments and simulations

Twodimensional (2D) van der Waals ferromagnets have gathered a lot of interest over the last years due to their low dimensionality and easy manipulation of their magnetic state. Recently ultrafast magnetization dynamics (UMD) in the compound $Fe_3GeTe_2$ have been observed [1], but complete understanding of the undelying processes involving energy and angular momentum flow are absent. Usually, only one of the subsystems (electrons, lattice, spins) is measured, leaving many free fit parameters for modeling. In this work, we present data from all three subsystems measured on an ultrafast timescale and give quantitative interpretation to the data by a four temperature model based on an extended microscopic three temperature model [2,3]. Electron temperature dynamics are measured by time- and angle-resolved photo-emission spectroscopy, magnetization dynamics by transmission X-ray magnetic circular dichroism, and lattice dynamics by ultrafast electron diffraction experiments, where the change in Mean Square displacement (ΔMSD) is recorded. Importantly, all data were collected on samples from the same production batch and measured under similar conditions. With our model, we reveal that the lattice reacts on two intrinsic timescales within the first tens of picoseconds (see Figure). We do this in a simplified fashion by assuming two internally thermalized phonon groups I and II, of which only I is coupled to the electron system while II only couples to I. With this modeling process we give a complete, comprehensive parameter study for all three subsystems in direct comparison to experiments and reveal the importance of non-thermal lattice excitations in this compound. This gives rise to more detailed investigation of lattice dynamics and angular momentum transfer to specific phonon modes in UMD of 2D van der Waals materials.

## Acknowledgements

TRR227 projects A09 A03 A10, DFG's emmy noether program under Grant No. RE 3977/1
Funding was also received from the Max Planck Society
European Research Council (ERC) under the European Union's Horizon 2020 research and innovation program (Grant Agreement No. ERC-2015-CoG-682843. V.C.A.T. acknowledges financial support from the Alexander von Humboldt Foundation. T. G. and U.A. acknowledge support by Grant No. RYC-2020-030605-I funded by MCIN/AEI/10.13039/501100011033 and by "ESF Investing in your future."

**References**

[1] T. Lichtenberg et al. *2D Mater.* **10** 015008 (2022)
[2] B. Koopmans et al. *Nature Mater* **9**, 259–265 (2010)
[3] T. Griepe and U. Atxitia *Phys. Rev. B* **107** L100407 (2023)

# Spin noise and ultrafast magnetic switching in antiferromagnetic $Sm_{0.7}Er_{0.3}FeO_3$

*Julius Schlegel[1]*, Tobias Dannegger[1], Martin Evers[1], Andreas Donges[1], Marvin A. Weiss[1], Takayuki Kurihara[2], Sebastian T. B. Goennenwein[1], Ulrich Nowak[1]*

[1] *Department of Physics, University of Konstanz, 78457 Konstanz, Germany*
[2] *ISSP, The University of Tokyo, Tokyo, Japan*
*julius.schlegel@uni-konstanz.de

In recent years, spin noise spectroscopy has emerged as an experimental tool to investigate material properties in solid state physics [1]. Here, we investigate spin noise from a theoretical point of view in an antiferromagnetically ordered orthoferrite.

The material of interest is $Sm_{0.7}Er_{0.3}FeO_3$, which undergoes a second-order magnetic reorientation transition at approximately room temperature [2]. On the basis of a classical spin model following the stochastic Landau-Lifshitz-Gilbert equation of motion we numerically compute the time evolution of a four-sublattice spin system, resembling the subsystem of iron atoms in $Sm_{0.7}Er_{0.3}FeO_3$, on a timescale from picoseconds up to one nanosecond and investigate the thermal fluctuations of the magnetization. Because of the competing anisotropies the second order reorientation transition is realized. Moreover, a Dzyaloshinskii-Moriya interaction, mediated by the oxygen atoms, induces a canting of the magnetic moments, leading to a finite magnetization and making the system a weak ferromagnet.

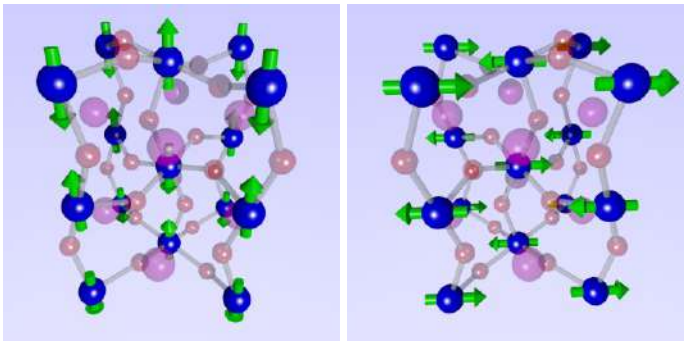


Figure: Reorientation transition of the magnetic moments of the iron atoms in $ErFeO_3$ from the low temperature regime (left) to the high temperature regime (right).

By Fourier-transforming the time dependent magnetization it is found that the spectral noise power density comprises resonances. With the help of linear spin-wave theory these resonances can be assigned to the magnetic eigenmodes of the system. As the reorientation transition also affects the eigenmodes, the characteristic appearance of the resonances in the different crystallographic components of the spectral power density indicates, in which magnetic phase the system is. The autocovariance, calculated via the Wiener-Khinchin theorem, unveils critical fluctuations at the critical temperatures of the reorientation transition in the time domain and thus marks the reorientation transition in the noise of the magnetization. In addition, the system shows random telegraph noise on picosecond timescales close to the critical temperatures of the reorientation transition due to its two energetically equivalent paths.

We compare the theoretical findings to spin noise measurements, underlying a novel probe-probe technique with two consecutive femtosecond laser pulses [3]. With the help of the Faraday effect the fluctuations of the magnetization are then analyzed. We find good agreement regarding the eigenfrequencies in the noise spectra and the critical fluctuations at the critical temperatures. Moreover, an enhancement of the spectral power density in the gigahertz regime close to the critical temperatures of the reorientation transition is observed, which can be ascribed to the ultrafast random switching process of the magnetization found in the simulations.

## Acknowledgements

We acknowledge the funding by the *Deutsche Forschungsgemeinschaft* via the SFB 1432 (Project B06).

## References

[1] Valerii S. Zapasskii, Adv. Opt. Photon. **5**, 131-168 (2013)
[2] Gabriel Fitzky et al., Phys. Rev. Lett. **127**, 107401 (2021).
[3] Marvin A. Weiss et al., arXiv:2301.02006 (2023)

# Pt-Transition Metal Synthetic Ferrimagnets for All Optical Switching

*Jade N. Scott[1]*, Maciej Dabrowski[2], William R. Hendren[1], Colin M. Forbes[1], Andreas Frisk[3], David M. Burn[3], David G. Newman[2], Connor R. J. Sait[2], Paul S. Keatley[2], Alpha T. N'Diaye[4], Thorsten Hesjedal[5], Gerrit van der Laan[3], Robert J. Hicken[2], Robert M. Bowman[1]*

[1]*School of Mathematics and Physics, Queen's University Belfast, Belfast, UK*
[2]*Department of Physics and Astronomy, University of Exeter, Exeter, UK*
[3]*Diamond Light Source, Harwell Science and Innovation Campus, Didcot, UK*
[4]*Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, USA*
[5]*Department of Physics, Clarendon Laboratory, University of Oxford, Oxford, UK*

Synthetic ferrimagnets (SFI), two ferromagnetic (FM) layers with different magnetic moments exchange coupled antiferromagnetically via the RKKY interaction across a non-magnetic spacer (NM), are of particular interest as a feasible media for all optical switching (AOS) [1], [2], which has been demonstrated to be a robust, ultrafast and efficient process with which to write data [3]. SFi are especially promising for applications of data storage as the properties of the constituent layers can be tailored independently to tune the AOS mechanism.

Design of the magnetic properties of SFi structures requires knowledge and control of the magnetization, M(T), and anisotropy, K(T), of each FM layer, and the strength of the exchange coupling between them, $J_{ij}(T)$. We show it is possible to engineer the magnetic properties of series of compensating $Ni_3Pt$/Ir/Co SFis and Ni/Pt multilayer based SFi, fabricated by magnetron sputtering and exhibiting perpendicular magnetic anisotropy (PMA). The tuneability of these structures (through variation of deposition conditions and layer thicknesses) provides an ideal system with which to investigate the effect of M(T), K(T) and $J_{ij}(T)$ on reversal mechanisms within SFis, with the added benefit of distinct magnetic species allowing element and layer specific characterization.

Initial pump/probe experiments have revealed that these Ni, Pt, Ir and Co, SFis exhibit multi-pulse, helicity independent AOS [4], up to a temperature of 330K in the Ni/Pt multilayer samples. Our results also reveal the importance of anisotropy, usually assumed to play a less important role [5] in AOS, and that a spin current generated during demagnetization is integral to the switching process.

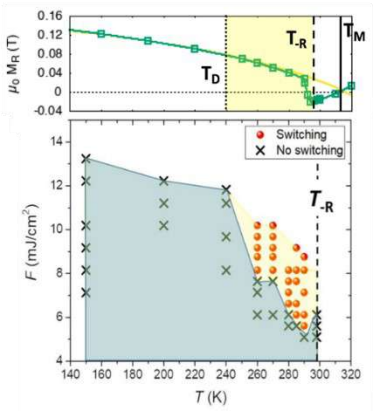



Figure 1 Temperature dependent behaviour of a SFi. Three distinct temperature regions can be observed. Switching behaviour as a function of sample temperature and fluence (using $\sigma^-$ polarization, a pump spot size diameter of 90 µm, pulse duration of 1ps, and 1MHz repetition rate), compared to the squid remanence measurement, measured by saturating and removing the field at every temperature point, of a $Ni_3Pt$(10nm)/Ir(0.5nm)/Co(1nm) SFi

## Acknowledgements

The authors acknowledge the Engineering and Physical Sciences Research Council (EPSRC) under Grant Numbers EP/P021190/1, EP/P020151/1, and EP/P02047X/1, the support of the Royal Academy of Engineering under the Research Chairs and Senior Research Fellowships Scheme and Seagate Technology (Ireland) under SOW #00077300.0. D.G.N.

## References

[1] S. Mangin et al, *Nature Materials*, 13, 286-292 (2014)
[2] J-W Liao et al, *Adv. Sci*, 6, 1901876 (2019)
[3] A.V. Kimmel and M. Li, *Nat. Rev. Mater,* 4, 189-200 (2019)
[4] M. Dabrowski et al, *Nano. Lett,* 21, 9210-9216 (2021)
[5] J. Barker et al, *Sci. Rep,* 3, 3262 (2013)

# Ballistic or Diffusive transport? Signature in Kerr response of magnetized films

*Sanjay Ashok*[1], Jonas Hoefer[1], Martin Stiehl[1],*
*Hans Christian Schneider[1], Martin Aeschlimann[1], Baerbel Rethfeld[1], Benjamin Stadtmueller[2]*
[1] *Department of Physics and OPTIMAS Research Center, RPTU Kaiserslautern-Landau, Germany*
[2] *Department of Physics, Johannes Gutenberg University, Mainz, Germany*
*sanjay.ashok@rptu.de

The influence of spin-resolved charge and heat transport in ultrafast demagnetization is an open problem. Due to the rich application of these phenomena in the design and fabrication of ultrafast spintronic devices, it is essential to understand the importance of diffusive and ballistic transport mechanisms on ultrafast timescales.

In this contribution, we theoretically address the differences between these transport mechanisms and propose an experimental method to distinguish between them. We first calculate the ultrafast magnetization dynamics in a thick nickel layer using the thermodynamic μT model [2], which includes diffusive transport mechanisms [1]. Our results show that gradients in the magnetization persist on timescales of picoseconds, well beyond typical demagnetization times. We assume that ballistic transport quickly leads to a homogeneous energy distribution within the film, so that the resulting magnetization dynamics are independent of depth.

We then theoretically simulate the probe angle dependent dynamics of the complex magneto-optical Kerr effect (CMOKE) [3], from which the Kerr-rotation dynamics as well as the Kerr-ellipticity dynamics are obtained for both considered transport cases. Our results show a pronounced sensitivity of the probe angle dependent Kerr-ellipticity dynamics to the presence of spatial gradients. CMOKE experiments on a 40-nm nickel film show the same signature, characteristic of spatial gradients of magnetization within the information-depth [3]. We conclude that ballistic energy transport plays a minor role in this sample.

Our results demonstrate the power of the CMOKE method, together with appropriate theoretical modeling, to study the role of transport in the ultrafast response of thick magnetic materials.

**Acknowledgements**

This work was funded by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) – TRR 173 - 268565370 Spin+X (Projects A08 and B03).

**References**

[1] Ashok et al., Appl. Phys. Lett. 120, 142402 (2022)
[2] Mueller and Rethfeld, Phys. Rev. B 90, 144420 (2014)
[3] Kuch et al., Magnetic Microscopy of Layered Structures, Springer, ISBN 978-3-662-44531-0 (2015)

# Cavity-mediated coupling of terahertz magnons and vibrational modes of molecules

*M. Białek*[1]*, *W. Knap*[2]

[1] *Institute of High Pressure Physics, Polish Academy of Sciences, Warsaw, Poland*

*marcin.bialek@unipress.waw.pl

In the regime of strong light-matter coupling, polariton modes are formed that are hybrid light-matter excitations sharing properties of both, an electrodynamic cavity mode and a matter mode. In the recent decade, magnon-polaritons were intensively researched using ferromagnetic materials in the microwave range, with potential applications for quantum technology and sensors. Exploring antiferromagnetic resonance (AFMR) rises magnon-polariton frequencies into the terahertz (THz) range [1]. In this range there are many dielectric excitations like phonons, vibrational modes of molecules, plasmons in two-dimensional electron gas, etc, which are characterized with higher light-matter coupling rates than those of magnetic excitations because of their high dipole moments. Recently, we reported cavity-mediated coupling of magnons in two distant slabs of antiferromagnets [2].

Here, we are investigating AFMR in yttrium ferrite $YFeO_3$ (YFO) owing to its low spin damping and temperature-dependent frequency above room temperature. We report on coupling of its quasi-antiferromagnetic mode to a vibrational mode of α-lactose. Our experimental setup consists of parallel-plane slabs of both materials, placed next to each other at a well controlled gap, forming a tunable Fabry-Perot type cavity. Frequency of AFMR was controlled by temperature of a YFO crystal. We used time-domain spectrometer to measure reflection spectra, collected as a function of YFO temperature and distance between the slabs. Frequencies of cavity modes are controlled by changing the gap between the crystals. Thus, as a function of distance, we observed narrow avoided crossings of cavity modes with AFMR and much broader avoided crossings with the vibrational mode of lactose at 0.53 GHz. At some distances between the slabs and YFO temperatures, we observed such polariton modes that are simultaneously coupled to the vibrational mode of lactose and the AFMR in the YFO crystal. Such states are hybridized magnon-vibrational modes that share properties of both matter excitations.

**Acknowledgements**

Partial funding from the European Union's Horizon 2020 research and innovation programe under the Marie Skłodowska-Curie grant agreement No. 847639 and from the Ministry of Education and Science of Poland is acknowledged. This work was also partially supported by the "International Research Agendas" program of the Foundation for Polish Science, co-financed by the European Union under the European Regional Development Fund (No. MAB/2018/9).

**References**

[1] M. Białek, J. Zhang, H. Yu and J-.Ph. Ansermet, Phys. Rev. Appl. 15, 044018 (2021)
[2] M. Białek, W. Knap and J-.Ph. Ansermet, arXiv.2212.01129

# Tuning of Effective Anisotropy Field for Time-Resolved Photo-Magnetic Reversal in YIG:Co Films

*L. Nowak*[1*, 2], *T. Zalewski*[1], *A. Frej*[1], *A. Stupakiewicz*[1]

[1] *Faculty of Physics, University of Bialystok, Bialystok, Poland*

[2] *Charles University, Faculty of Mathematics and Physics, Prague, Czech Republic*

*l.nowak@uwb.edu.pl

Magnetization state control by a single ultrashort laser pulse has recently been the focus of scientific interest since it offers extremely short switching times, does not require an external magnetic field, and can be operated at room temperature. Additionally, selective and reversible photo-magnetic switching in Co-doped yttrium iron garnet films (YIG:Co) can be obtained. This recording was characterized by a switching time of about 20 ps. Moreover, because of its resonant character the write–read events are accompanied by an unprecedentedly low heat load [1]. Microscopically, in this mechanism incident linearly polarized pump pulse excites strongly anisotropic garnet ions, generating a photo-induced contribution to the effective field of magnetic anisotropy [2]. This strong and short-living addition, driven by an external light pulse can trigger the precession or even switch the magnetization. However, while the dynamic is described by the LLG equation, other contributions to the effective field of anisotropy, both external and internal, clearly affect the trajectory of the magnetization movement.

The heat load threshold of photo-magnetic switching has been found for different thicknesses of garnet films by means of magnetic domain imaging. For our experiments, we used Co-doped yttrium iron garnet thin films (YIG:Co). Comparative results were obtained from a batch of 2-8 μm-thick YIG:Co single-crystal films fabricated from liquid phase epitaxy on (001)-oriented $Gd_3Ga_5O_{12}$ substrate without miscut angle, which were later chemically etched to the set thickness. In these garnets, cubic magnetic anisotropy dominanates uniaxial grown-induced anisotropy contrubution. Here we investigate the effects of the garnet on magnetic anisotropy and photo-magnetic dynamics. Moreover, we examine the possibility to use an electric field to modify the anisotropy in garnet films. With the sample patterned with gold-plated electrical contacts, we apply the electric field affecting the laser-induced magnetization dynamics. Combining ultrafast laser pulse and pulse of electrical field illustrates the high potential of the magnetic dielectrics for cold ultrafast magnetic recording.

## Acknowledgments

We acknowledge support from the grant European Union's Horizon 2020 Research and Innovation Programme under the Marie Skłodowska-Curie grant agreement No 861300 (COMRAD).

## References

[1] A. Stupakiewicz et al., Nature **542**, 71 (2017).
[2] A. Frej et al., arXiv:2211.08048

# Influence of Metal/Organic-Molecules Interface on Magnetic Anisotropy in Co Thin Films: A Time-Resolved MOKE Investigation

*J. Strohsack[1], G. Jecl[1], H. Zhao[1], A. Shumilin[1], V. V. Kabanov[1], T. Mertelj[1,2,*], M. Benini[3], R. Rakshit[3], V.A. Dediu[3], M. Rogers[4], S. Ozdemir[4], O. Cespedes[4], F. Mertens[5], U. Parlak[5], M. Cinchetti[5]*

[1]*Department for complex matter (F7), Jožef Stefan Institute, 1000 Ljubljana, Slovenia*

[2]*CENN Nanocenter, Jamova 39, 1000 Ljubljana, Slovenia*

[3] *ISMN-CNR, Via Piero Gobetti 101, Bologna, Italy*

[4] *School of Physics and Astronomy, University of Leeds, Leeds, LS2 9JT, United Kingdom*

[5] *Technische Universität Dortmund, Otto-Hahn-Straße 4, Dortmund, Germany*

*tomaz.mertelj@ijs.si

We investigated influence of metal/organic-molecules interfaces on the magnetic anisotropy in thin polycrystalline Co thin films grown by electron-beam evaporation. To assess the magnetic anisotropy we measured temperature and magnetic field dependence of the femtosecond-laser induced coherent spin waves by means of the time-resolved magneto-optical Kerr effect (MOKE) spectroscopy [1]. We compare the effects of interfacing the Co films to a nonmagnetic metal (Al), metalorganic complexes tris(8-hydroxyquinoline)gallium ($Gaq_3$) and M-phthalocyanines (M=Cu, Co) as well as Buckminster-fullerene ($C_{60}$) molecules.

In general, the transient MOKE signals were found to exhibit damped coherent spin wave oscillations (CSWO) with frequencies up to several tens of GHz. Detailed analysis of the spin-wave temperature and magnetic field dependences allowed us to compare the influence of different molecular interfaces.

We found that the thin Co films interfaced with molecular layers display strong hardening of the CSWO frequency at low $T$ with a rather sharp transition in the 150 K - 200 K range, depending on the molecular species. Interfaces with different molecules show qualitatively similar behavior despite different molecular shapes. The hardening is attributed to increase of the interface induced anisotropy due to the hybridization between the molecular-orbitals and the Co *d*-orbital derived interface states. [2]

Strong damping of the CSWO observed in the molecular-layers interfaced Co films is attributed to the dephasing due to the molecular layer disorder.

**Acknowledgements**

We acknowledge the support of EC project INTERFAST (H2020-FET-OPEN-965046).

**References**

[1] M. van Kampen *et al., Phys. Rev. Lett.* ***88*** (2002) 227201.
[2] T. Moorsom *et al., Phys. Rev. B*. ***101*** (2020) 060408.

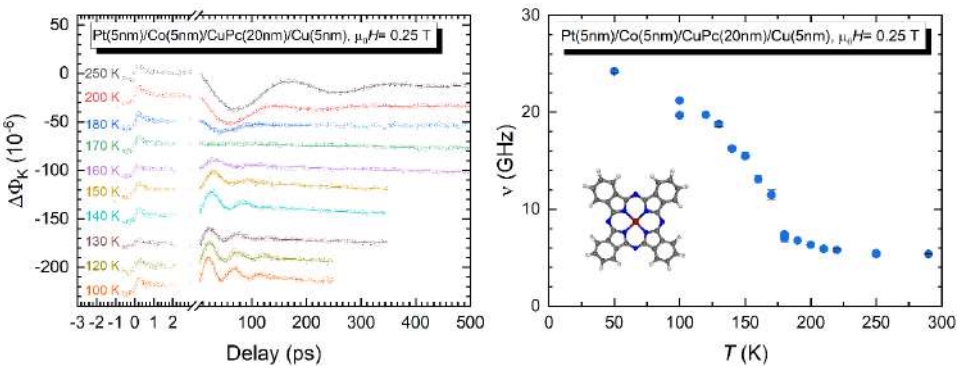



Figure: Coherent spin-wave oscillations (left) and the temperature dependence of their frequency (right) in a Cu-phthalocyanine capped Co film

# SYMPOSIUM 16.
## MICROMAGNETIC AND MULTISCALE MAGNETIC MODELLING AND SIMULATIONS.
## S16 INVITED ORAL PRESENTATIONS

# Micromagnetic modelling from 1D to 3D to efficiently simulate nanostructured magnetic materials and devices

*Alessandra Manzin*[1]*, *Riccardo Ferrero*[1], *Marta Vicentini*[1]

[1] *Istituto Nazionale di Ricerca Metrologica (INRIM), Strada delle Cacce 91, 10135, Torino, Italy*

*a.manzin@inrim.it

Computational micromagnetic modelling is a powerful tool to address the design of nanostructured magnetic materials and devices for applications in health, energy, automotive, ICT and metrology sectors, as well as to guide the interpretation of novel magnetic phenomena at the nanoscale [1]. However, the numerical implementation of efficient micromagnetic solvers is a very complex task, especially for the simulation of several-micron-sized magnetic nanostructures or large-scale arrays of nanomagnets. The main reason of such complexity is the multiscale nature of the problems to be investigated, which involve both long-range magnetostatic interaction and rapidly decaying exchange interaction.

Several strategies have been implemented to speed-up micromagnetic simulations, such as fast-multipole and FFT-based techniques for the magnetostatic field computation. At the same time, efforts have been made towards multiscale modelling, coupling atomistic models to micromagnetic calculations [2]. In the last decade, many parallel micromagnetic solvers have also been developed, able to run on massively parallel architectures based on graphical processing units (GPUs) [3].

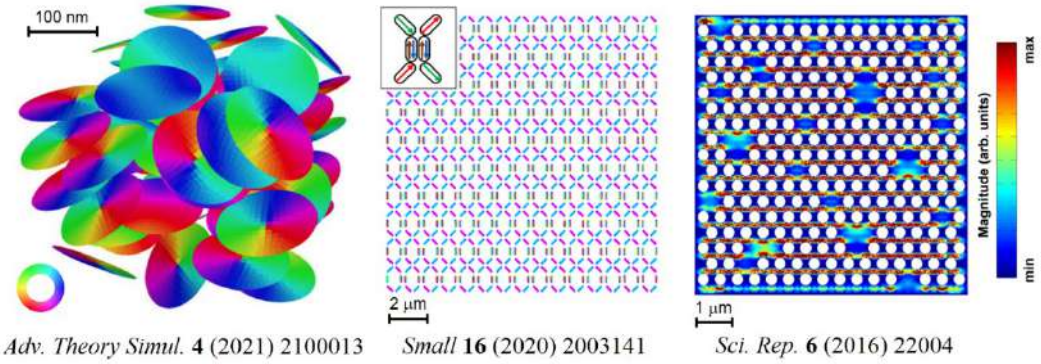
To perform reliable micromagnetic simulations in an efficient way, we have combined GPU-accelerated computing with non-standard numerical techniques (e.g. multipole expansion approaches for magnetostatic field calculation, finite-difference methods for exchange field calculation on irregular grids, domain symmetry exploitation, geometric integration based on Cayley transform). In this talk, we will present the implemented strategies, including hybrid 1D-3D, 2D-3D or totally 3D modelling approaches [4, 5], and their application in nanomagnetism, from nanostructured magnetic materials (magnetic nanoparticle clusters, nanopatterned thin films, spin-ice systems) to magnetoresistive and spintronic devices, also in array configuration.

## Acknowledgements

This work is funded by the Italian Ministry of University and Research under Project PRIN 2020LWPKH7 IT.

## References

[1] C. Abert, *Eur. Phys. J. B* **92** (2019) 120.
[2] S. C. Westmoreland et al., *Scripta Materialia* **148** (2018), 56-62.
[3] J. Leliaert, J. Mulkers, *J. Appl. Phys.* **125** (2019) 180901.
[4] A. Manzin, R. Ferrero, *J. Magn. Magn. Mater.* **492** (2019) 165649.
[5] R. Ferrero, A. Manzin, *J. Magn. Magn. Mater.* **518** (2021) 167409.




*Adv. Theory Simul.* **4** (2021) 2100013 *Small* **16** (2020) 2003141 *Sci. Rep.* **6** (2016) 22004

Figure: An excursus of applications of the micromagnetic modelling tools developed at INRIM (left: magnetic nanodisks for hyperthermia; centre: artificial spin-ice; right: ferromagnetic resonance in magnonic crystals).

# Relativistic Elastic Collision Among Antiferromagnetic Domain Walls

*Rubén M. Otxoa[1,2]*, O. Chubykalo-Fesenko[3], Gen Tatara[4]*

*[1, *] Hitachi Cambridge Laboratory, J. J. Thomson Avenue, Cambridge CB3 0HE, United Kingdom, UK*

*[2]Donostia International Physics Center, San Sebastián 20018, Spain*

*[3] Instituto de Ciencia de Materiales de Madrid, CSIC, c/Sor Juana Ines de la Cruz, 3 Spain*

*[4] RIKEN Center for Emergent Matter Science (CEMS) and RIKEN Cluster for Pioneering Research (CPR), 2-1 Hirosawa, Wako, Saitama, 351-0198 Japan*

*ro274@cam.ac.uk

Soliton-based information storage, processing and computing is a very appealing technological proposal since these objects are fast, robust to perturbations and preserve their shapes during collisions. Here we discuss the possibility for its realisation, based on spintronics of antiferromagnetic domain walls. The applications of antiferromagnetic spintronics are recently attracting huge attention. Antiferromagnets are abundant, insensitive to external fields and their spin-current manipulation is energy efficient. Over the last few decades different names have been coined to describe localized stable solutions for the dynamical equations of magnetization. For instance, domain walls are called kinks or solitons interchangeably. The name soliton is reserved to a restricted solitary waves which have the remarkable property of emerging as intact entities after a head-on collision with their shapes and velocities unaltered. Magnetic domain walls are often referred to as solitons disregarding the strict mathematical definition requiring the above scattering property. Here we demonstrate the conditions of elastic and inelastic scattering for spin-orbit torque-induced dynamics of relativistic domain walls on the technologically relevant $Mn_2Au$ antiferromagnetic material. We show that even domain walls with opposite winding numbers can experience elastic scattering and we present the corresponding phase diagram as a function of the spin-orbit field strength and duration. The elastic collision requires minimum domain walls speed, which we explain assuming an attractive potential created by domain wall pair. On the contrary, when the domain walls move at lower speeds, their collision is inelastic and results in a dispersing breather.

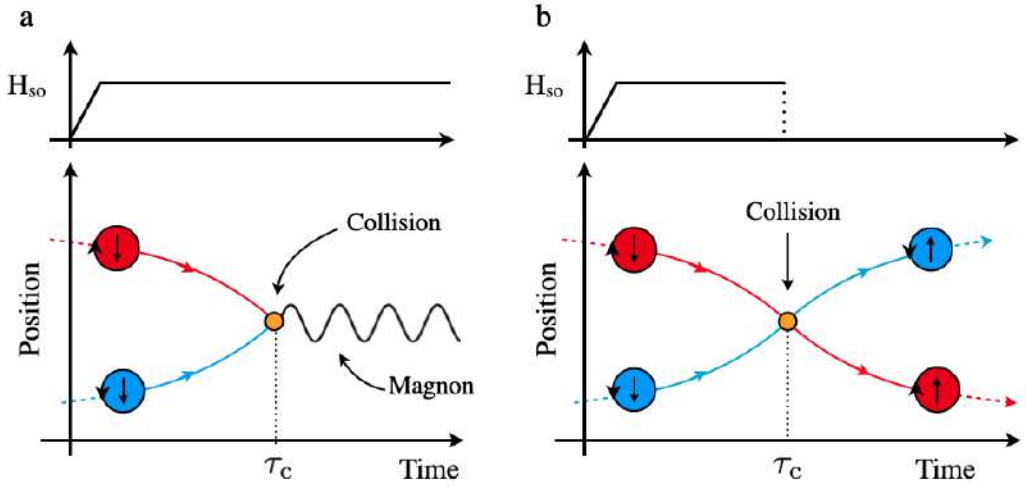



Figure: a Schematic illustration of two magnetic textures with opposite winding number driven by a spin-orbit (SO) field which collide while the SO-field is present. In such a condition the inelastic collision gives rise to the excitation of breather. b Schematic illustration of two magnetic textures with opposite winding number driven by a SO-field which collide elastically in the absence of the SO-field.

## References

[1] R. Otxoa, U.Atxitia, P.E.Roy and O. Chubykalo-Fesenko, "Giant localised spin-Peltier effect due to ultrafast domain wall motion in antiferromagnetic metals" Comm. Phys. **3**, 31 (2020)

[2] R. M. Otxoa, R. Rama-Eiroa, P. E. Roy, G. Tatara, O. Chubykalo-Fesenko, and U. Atxitia., "Topologically-mediated energy release by relativistic antiferromagnetic solitons" Phys Rev Res. 3, 043069 (2021)

[3] R. Otxoa, G. Tatara, U. Atxitia and O. Chubykalo-Fesenko "Tailoring Elastic Scattering of Relativistic Antiferromagnetic Domain Walls for Collision-based Computing" arXiv:2212.03126 (2022)

# SYMPOSIUM 16.
## MICROMAGNETIC AND MULTISCALE MAGNETIC MODELLING AND SIMULATIONS.
## S16 ORAL PRESENTATIONS

# Minimum energy paths of magnetic skyrmions crossing grain boundaries

*Rhodri Mansell*[1]*, *Hannes Jónsson*[2], *Sebastiaan van Dijken*[1]

[1] *NanoSpin, Department of Applied Physics, Aalto University School of Science, P.O. Box 15100, FI-00076 Aalto, Finland*

[2] *Science Institute and Faculty of Physical Sciences, University of Iceland, VR-III, 107 Reykjavík, Iceland*

*rhodri.mansell@aalto.fi

Magnetic skyrmions have been widely studied for possible use in devices. The most studied material systems for technological applications are sputtered multilayers, which can contain material grains that affect the local magnetic properties. The interactions between skyrmions and magnetic grains has been studied in dynamic micromagnetic simulations e.g. for thermally-activated and current-driven skyrmions [1]. However, such simulations cannot be used to study large energy barriers, where skyrmions may be stable for timescales inaccessible with dynamic simulations. Here, we use atomistic simulations combined with the geodesic nudged elastic band method [2] to determine the minimum energy path of a skyrmion crossing an idealized grain boundary. The simulations make use of the *Spirit* software [4], using parameters derived from a well-studied Fe/Pd on Ir(111) system [3]. We create a grain boundary by reducing the exchange coupling, moment and/or Dzyaloshinskii-Moriya interaction in a single line across the simulation (Fig. 1a) consisting of 8700 spins. The simulation is initialized by setting a chain of 121 images of the skyrmion with initial positions uniformly straddling the boundary. The system is then evolved to find the minimum energy path. Snapshots of the skyrmions where the grain boundary is formed by a 40 % reduction in the local magnetic properties, are shown in Fig. 1b-e. The energy of the skyrmion at each position is shown in Fig. 1f with the energy terms decomposed. The total energy shows that, for the set of parameters used here, the boundary forms a barrier which is dominated by the changes in the DMI energy. Further, we study the effect of changing the boundary properties and the overall DMI in the simulation, showing that the boundary can also act as a pinning site for skyrmions or cause their annihilation depending on the chosen parameter values.

## Acknowledgements

We acknowledge the provision of computing facilities by the Aalto Science-IT project.

## References

1. Y. Zhou, *et al.,* Phys. Rev. B **104**, 144417 (2021); J.-V. Kim et al., Appl. Phys. Lett. **110**, 132404 (2017)
2. P.F. Bessarab, V.M. Uzdin and H. Jónsson, Comp. Phys. Commun. **196**, 335 (2015).
3. N. Romming et al., Science **341**, 636-639 (2016).
4. G.P. Müller, *et al.*, Phys. Rev. B **99**, 224414 (2019)

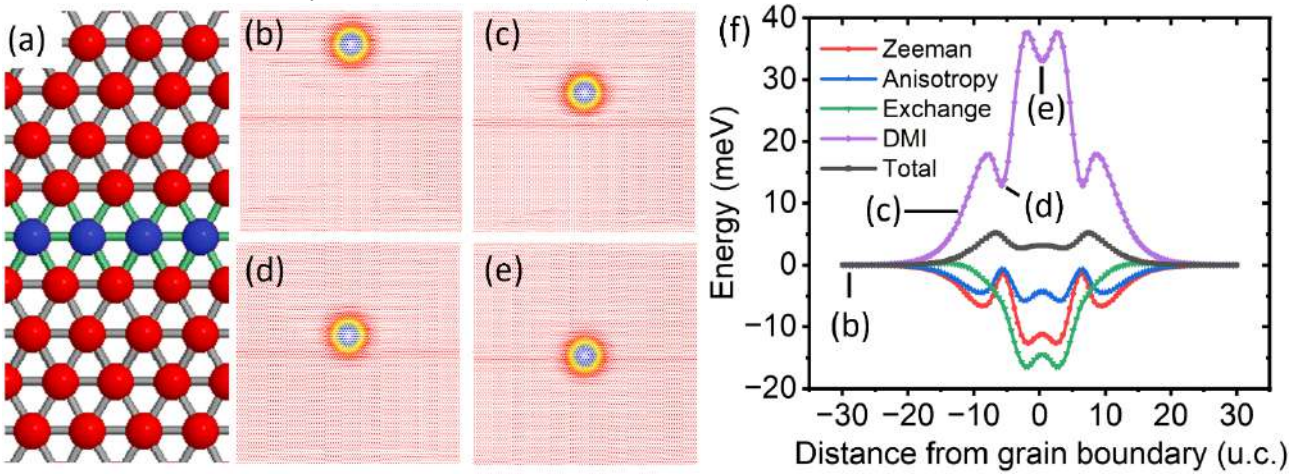



Figure 1. (a) Schematic of part of the lattice used in the simulations. The blue atoms and green bonds mark where the magnetic properties are altered compared to the rest of the lattice (red atoms / grey bonds). (b)-(e). The spin configurations for various skyrmion positions. The images show the entire simulation. Spins pointing up out-of-plane are red, those down out-of-plane are blue. The grain boundary runs horizontally across the centre of the simulation. (b) Magnetic energy terms as a function of skyrmion position for a minimum energy path across a boundary where the exchange constant, DMI and moment of the central atom are all reduced by 40 % compared to the rest of the lattice. The energy reference point is taken at 30 unit cells away from the grain boundary. The position of each snapshot is marked.

# Accelerating The Simulations of Spintronic Oscillators Using An Unconventional Data-Driven Approach

*Simon DE WERGIFOSSE[1]*, Chloé CHOPIN[1], and Flavio ABREU ARAUJO[1]*

[1] *Université catholique de Louvain, Place Croix du Sud 1, Louvain-la-Neuve, Belgium*

*simon.dewergifosse@uclouvain.be

Spintronic oscillators have attracted great attention recently, namely for prospective use as hardware neurons within artificial neural networks [1]. In this regard, either micromagnetic simulations or analytical models based on the Thiele equation approach (TEA) are employed to study these devices theoretically. However, none of these methods is both fast and accurate, which limits their use for simulation-intensive applications like in neuromorphic computing.

Here, micromagnetic simulations are compared to the Thiele equation approach [2]. Fair agreement is shown in the resonant regime, while only qualitative predictions can be obtained for steady-state oscillations (see Fig. 1). Moreover, we show the significant impact of the Ampère-Oersted field on the dynamics of the oscillator. However, the purely analytical model remains qualitative, and finding suitable mathematical expressions to describe the oscillator's dynamics accurately is impractical [3].

To overcome this theoretical limitation, a data-driven method [4] has been developed on top of our updated analytical model [2]. Indeed, using a limited set of micromagnetic simulations, we were able to make our model quantitative by adjusting the damping and gyrotropic terms in TEA. As the discrepancies with micromagnetism are absorbed, our semi-analytical model is capable of predicting the steady-state regime of the oscillator with the same accuracy compared to micromagnetic simulations (see Fig. 1). Furthermore, the transient regime is also accurately rendered, allowing to solve the full dynamics of the oscillators.

This data-driven model is more than two billion times faster than micromagnetic simulations, for an equivalent accuracy. Such accelerating factor opens the way to high-throughput simulation campaigns, for example in the framework of neuromorphic spintronics.

## Acknowledgements

Computational resources have been provided by the Consortium des Équipement de Calcul Intensif, funded by the F.R.S.-FNRS. F.A.A is a Research Associate and S.d.W. is a FRIA grantee, both of the F.R.S.-FNRS.

## References

[1] J. Torrejon, M. Riou, F. Abreu Araujo, *et al.*, *Nature* **547** (2017) 7664.
[2] F. Abreu Araujo, C. Chopin, and S. de Wergifosse, *Sci. Rep.* **12** (2022) 10605.
[3] S. de Wergifosse, C. Chopin, and F. Abreu Araujo, *arXiv preprint* (2022) 2206.13438.
[4] F. Abreu Araujo, C. Chopin, and S. de Wergifosse, *arXiv preprint* (2022) 2206.13596.

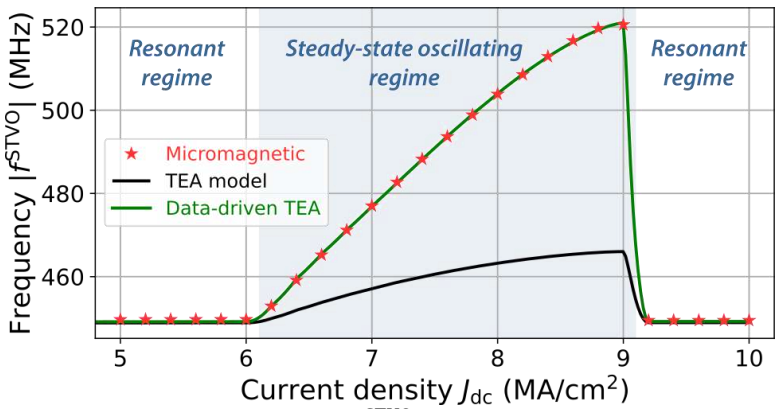



Figure 1: Comparison of the oscillator gyrotropic frequency $f^{STVO}$ with respect to the current density $J_{DC}$ between our improved Thiele equation approach (TEA) model, our data-driven TEA and micromagnetic simulations.

# On the numerical accuracy of the demagnetization tensor

*Rasmus Bjørk*[1]*, *Massimiliano d'Aquino*[2]

[1] *Department of Energy Conversion and Storage, Technical University of Denmark, Kgs. Lyngby, Denmark*

[2] *Department of Electrical Engineering and ICT, University of Naples Federico II, Naples, Italy*

*rabj@dtu.dk

Computing the demagnetization field is important in both micromagnetism and magnetostatic applications. The magnetic field, $\mathbf{H}$, from a geometrical object that is homogeneously magnetized can be expressed as

$$\mathbf{H}(\mathbf{r}) = -\nabla\phi_{\mathrm{M}}(\mathbf{r}) = -\nabla\frac{1}{4\pi}\oint_{S'}\frac{\hat{\mathbf{n}}(\mathbf{r}')\cdot\mathbf{M}(\mathbf{r}')}{\parallel \mathbf{r}-\mathbf{r}' \parallel}\mathrm{d}S'$$

where $\mathbf{r}$ is the radius vector to the point where the field is to be known, $\mathbf{r}'$ and $S'$ are the radius vector and the surface of the object generating the field, $\hat{\mathbf{n}}$ is the surface normal and $\mathbf{M}$ is the magnetization. This equation can be written as the product of a tensor, i.e. the demagnetization tensor, $\mathbb{N}$, and the magnetization as

$$\mathbf{H}(\mathbf{r}) = -\mathbb{N}\cdot\mathbf{M}$$

The demagnetization tensor is known for several geometrical objects or tiles, such as a prism [1], tetrahedron [2] and cylindrical tile [3,4]. The magnetic field generated by these tiles can be used for a fast and accurate calculation of the demagnetization field in micromagnetic simulations, as is done in the MagTense micromagnetic framework [5]. However, the demagnetization tensor has previously been found to be numerically inaccurate at distances far away, but potentially also close to, specific tiles [6].

Here we present an analysis of the accuracy of the demagnetization tensor compared to the dipolar field, as this is what the magnetic field should approximate at distances far from any tile. We computed the median error on the surface of 121 spheres with radius varying uniformly in logspace. Shown in Fig. 1 is the normalized fractional error of the field compared to the dipolar field, as computed using the four different expressions for the demagnetization tensor referenced above. It is seen that the demagnetization tensor correctly calculates the field out to several thousand tile radii, for both single and double precision computations..

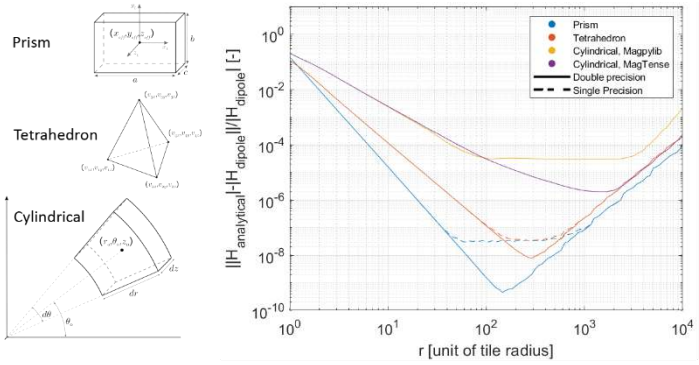



Figure: The geometries considered and the error as function of the distance, r, from the tile considered.

## Acknowledgements

R. Bjørk wish to acknowledge the Independent Research Fund Denmark, grant "Magnetic Enhancements through Nanoscale Orientation (METEOR)", 1032-00251B, and the Poul Due Jensen Foundation project on Browns paradox in permanent magnets, Project 2018-016, for sponsoring this work.

## References

[1] A. Smith, et al.: J. Appl. Phys., 107 (2010) 103910.
[2] K. K. N., A. R. Insinga and R. Bjørk: IEEE Magnetics Letters, 10 (2019) 1–5.
[3] K. K. Nielsen and R. Bjørk: J. Magn. Magn. Mater., 507 (2020) 166799.
[4] F. Slanovc, M. Ortner, M. Moridi, C. Abert and D. Suess: J. Magn. Magn. Mater., 559 (2022) 169482.
[5] R Bjørk and K. K. Nielsen: MagTense, doi.org/10.11581/DTU:00000071 (2019)
[6] D. Chernyshenko and H. Fangohr: J. Magn. Magn. Mater., 38 (2015) 440–445.
[7] M. Ortner and L. G. C. Bandeira: Magpylib. SoftwareX (2020).

# Lumped-Element Circuit Modeling of Spin-Wave Transducers

*Róbert Erdélyi[1], Ádám Papp[1]*, Philipp Pirro[2], György Csaba[1]*

[1] *Faculty of Information Technology and Bionics, Pázmány Péter Catholic University, Práter st. 50/A, Budapest, Hungary*

[2] *Fachbereich Physik and Landesforschungszentrum OPTIMAS, Technische Universität Kaiserslautern, Kaiserslautern, Germany*

*papp.adam@itk.ppke.hu

Efficient conversion between the electric (microwave) and spin-wave domains is one of the most pressing problems in the way of commercially viable magnonic devices. Using antennas based on conducting wires to excite spin waves via Oersted fields and pick them up via induction is the most straightforward way for longer-wavelength spin waves. To incorporate spin-wave devices into RF circuitry one needs to have a circuit model of the spin-wave device. We are modeling the spin-wave loading on the antenna via an impedance connected in series (Figure 1a) [1-2]. Oersted fields are calculated numerically for an estimated current distribution in the wires. These fields are applied in a micromagnetic simulation to generate spin waves, and subsequently induced EMF (electromotive force) in the antenna is calculated. The complex ratio of the current and voltage (EMF) on the antenna constitutes the equivalent impedance of the spin-wave loading. Besides, the antenna has a self-inductance and Ohmic resistance, these can be estimated using standard electromagnetic methods.

On the pickup side we are assuming the same antenna model, with an additional voltage source representing the induced EMF by the transmitted spin waves. In addition, a direct electromagnetic coupling between the two antennas can be represented by a mutual inductance, calculated similarly to the self-inductance above. The circuit model allows us to estimate the performance of the full magnonic device, including insertion loss and total transmitted power, as well as determining the effect of parasitic couplings and losses. Depending on the application, a matching network might be necessary to match the typically low impedance spin-wave transducer to, e.g., a 50 Ω system. Our model takes into account all relevant couplings between the magnetic and electrical domains and in most cases substitutes a fully-coupled electromagnetic—micromagnetic solver.

In typical nanoscale magnonic systems the power transferred to the magnonic domain is typically a very small portion of the power available on the line. In general, increasing the amount of magnetic material in connection with the transducer (thicker, wider spin-wave medium) will increase the loading on the antenna, however, this goes against the general requirement of downscaling. In addition, the magnetic configuration will strongly influence the transmission efficiency of the transducer [3]. Our model supports extension with machine learning for the optimization of power-transmission efficiency of spin-wave transducers.

## Acknowledgements

The authors acknowledge support from the European Union within the HORIZON-CL4-2021-DIGITAL-EMERGING-01 Grant No. 101070536 MandMEMS.

## References

[1] Connelly, D.A., Csaba, G., Aquino, H.R.O. et al. *Sci. Rep.* **11** (2021) 18378,
[2] Vanderveken, F., Tyberkevych, V., Talmelli, G. et al. *Sci. Rep.* **12** (2022) 3796,
[3] M. Kostylev, A.A. Stashkevich, *J. Magn. Magn. Mater.* **484** (2019) 329-344.

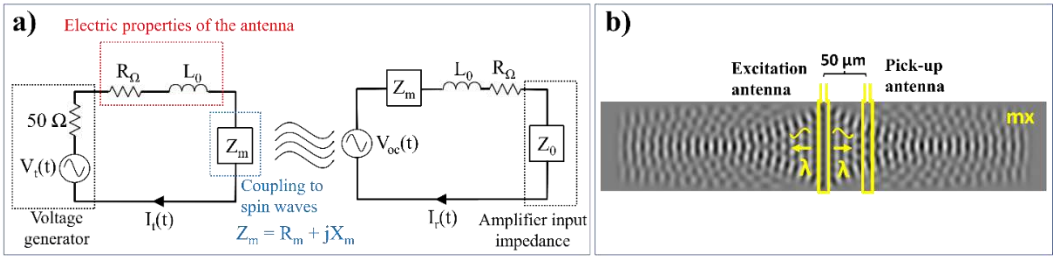



**Figure 1**: a) Lumped-element circuit model of the spin wave device. b) Spin waves generated in a *500µm x 100µm x 1µm* YIG stripe calculated via a micromagnetic simulation and position of the excitation and the pick-up antenna.

# Inelastic Scattering Of Spin-Wave Beams On Localised Spin Waves And Goos-Hänchen For Scattered Beams

*Krzysztof Sobucki[1] *, Julia Kharlan[1,2], Roman V. Verba[2], Igor L. Lyubchanskii[3], Maciej Krawczyk[1], Paweł Gruszecki[1]*

[1] *ISIK, Faculty of Physics, Adam Mickiewicz University, Poznań, Poland)*
[2] *Institute of Magnetism of NAS of Ukraine and MES of Ukraine, Kyiv, Ukraine*
[3] *Faculty of Physics, V. N. Karazin Kharkiv National University, Kharkiv, Ukraine*
*krzsob@amu.edu.pl

A spin wave (SW) is a fascinating research object and a potential information carrier. One of the properties of SWs is that their dynamics allows for easy access to nonlinear phenomena, e.g., confluence and stimulated splitting processes. Here, we examine the interaction between an incident SW beam and SW modes localized in the vicinity of the ferromagnetic film's edge. During nonlinear interactions between the incident SW beam and the localized modes, new beams with shifted frequencies appear due to confluence and stimulated splitting processes, see Fig 1. Here, we use two different approaches to obtain localized SWs, i.e., (i) using the demagnetizing field about the film's edge [1] and (ii) placing a ferromagnetic strip directly over the edge of the film. In the second approach, we employ a magnonic Gires-Tournois interferometer [2,3], which influences the localized mode properties depending on the interferometer's geometry and, thereby, allows access the modes that differ in quantization by the strip's width. The result is two additional primary beams whose frequencies are lowered and raised relative to the incident beam by the frequency of the localized mode resulting from the stimulated splitting and confluence processes, respectively. In our investigation, we compare the efficiency of the inelastic scattering processes for both types of mode's localization and investigate the lateral displacement of inelastically scattered SW beams' along the interface, see the inset in Fig 1. This phenomenon is an SW analog to the Goos-Hänchen effect for inelastically scattered beams [4]. In both cases of mode localization, we show that the stimulated splitting process has higher efficiency than the confluence process. Moreover, we demonstrate that the lateral shift of the inelastically scattered beams depends on the edge mode frequency. Our research provides a better understanding of nonlinear processes involving SW and may open new avenues for the development of magnonic systems using nonlinear SW dynamics.

The research leading to these results has received funding from the Polish National Science Centre projects No. 2019/35/D/ST3/03729 and 2022/45/N/ST3/01844.

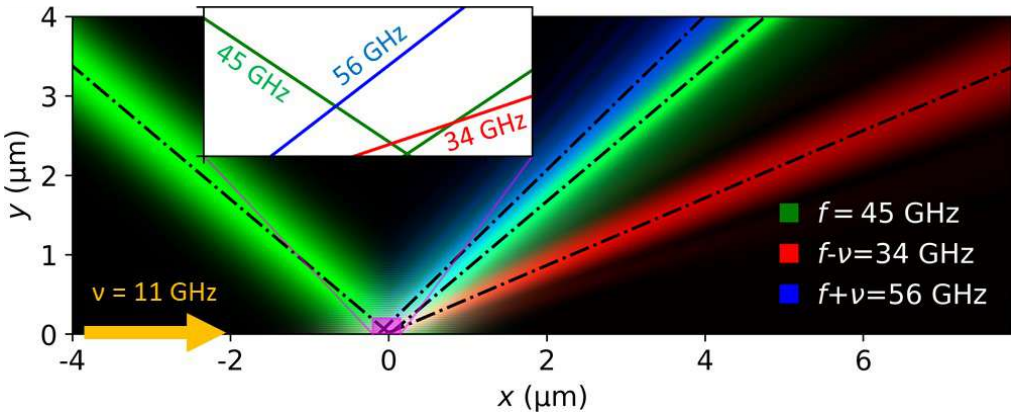



Figure 1: Example of the inelastic scattering of a SW beam (frequency 45 GHz, green color) on an edge mode (frequency 11 GHz, orange color). The inelastic scattering results in creation of two new SW beams with new frequencies, 32 GHz (stimulated splitting process, red color) and 56 GHz (confluence process, blue color). The new beams are also shifted spatially at the interface as shown in the inset.

## References

[1] P. Gruszecki, et al., Phys. Rev. Applied 17, 044038 (2022)
[2] K. Sobucki, et al., Sci. Rep. 11, 4428 (2021); K. Sobucki, et al., IEEE Trans. Magn. 58, 1300405 (2022)
[3] K. Sobucki et al. arXiv:2302.11507 (2023)
[4] Y. S. Dadoenkova, et al., Optical Materials Express 12, 717, 2022.

# Micromagnetic Simulations Explore the Link between Microstructure and Performance of Permanent Magnets

*Andrea Roberto Insinga*[1]*, *Rasmus Bjørk*[1]

[1] *Department of Energy Conversion and Storage, Technical University of Denmark, Anker Engelunds Vej, Kgs. Lyngby, Denmark*

*aroin@dtu.dk

Developing permanent magnet materials with increased performance is one of the main objectives of research within the area of magnetism. In particular, emphasis is placed on increasing the coercivity and the maximum energy product of magnets, since reducing the content of rare-earth elements without decreasing these properties has so far proven challenging [1]. Since the theory predicts a much higher coercivity than what is observed experimentally, a discrepancy known as Brown's paradox, it is expected that this parameter can be improved by studying the magnetization reversal process at the microscale.

Micromagnetic simulations are employed to investigate the relation between the microstructure of the materials and their magnetic behavior, in order to inform the manufacturing process. The open source magnetic simulation framework MagTense [2] has been developed at the Technical University of Denmark for this purpose. In this work we report the results of our systematic investigation about the impact that different geometrical qualities of the microstructure have on the performance of the magnet.

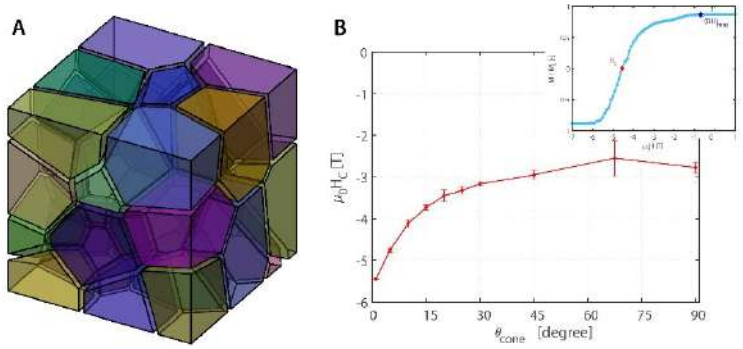



Figure: A) Microstructure generated by Voronoi tessellation. B) Coercivity as function of the misorientation of the easy axes of the grains (Inset: Hysteresis curve from which the coercivity is calculated).

The microstructure of permanent magnets is characterized by crystal grains of a hard magnetic phase separated by a region occupied by a soft magnetic phase. We generated the microstructures by using Voronoi tessellations, which allow us to specify the number of grains and the size of the intergrain region. An example of microstructure is shown in Figure A. We also considered the effect of the misorientation between the easy axis of the different crystal grains by generating directions according to random distributions parameterized by a standard deviation $\theta_{cone}$. The corresponding results are shown in Figure B, where the coercivity is plotted as function of the parameter $\theta_{cone}$, showing the decrease of coercivity associated with the easy axis misorientation. The inset shows one example of the magnetic hysteresis curves from which the coercivity has been calculated.

Besides the effect of misorientation and the geometrical parameters describing the microstructure, we also consider the effect of the different magnetic parameters such as the anisotropy constant and the exchange interaction constant characterizing the hard and soft magnetic phases. Compared to previous studies, our investigation follows a more systematic procedure, where all the main effects are analyzed one by one. Thanks to this approach, we are able to shed light on which of these effects play a more significant role in reducing the coercivity with respect to the theoretical prediction.

## Acknowledgements

This work was supported by the Poul Due Jensen Foundation project on Browns paradox in permanent magnets, Project 2018-016.

## References

[1] H. Sepehri-Amin, S. Hirosawa, and K. Hono, *Handb. Magn. Mater.* **27** (2018) 269–372.
[2] R. Bjørk, E.B. Poulsen, K.K. Nielsen, and A.R. Insinga, *J. Magn. Magn. Mater.* **535** (2021) 168057.
[3] R. Bjørk, and A.R. Insinga, *J. Magn. Magn. Mater.* **571** (2023) 170510.

# Simulating Magnetization Reversal in Synthetic Antiferromagnetic Disk Particles with Mechanical Rotation

*Santiago Helbig[1]*, Claas Abert[1], Pedro A. Sánchez[1,2], Inge K. Herrmann[4,5], Hans J. Hug[4], Sofia S. Kantorovich[1], Dieter Suess[1]*

[1] *Faculty of Physics & VDSP, University of Vienna, Kolingasse 14, 1090 Vienna, Austria*
[2] *Physics Departments, University of the Balearic Islands, Cra. De Valldemossa km 7.5, 07122 Palma de Mallorca, Spain*
[3] *Empa, Überlandstrasse 129, 8600 Dübendorf, Switzerland*
[4] *Department of Mechanical and Process Engineering, ETH Zürich, Sonneggstrasse 3, 8092 Zürich, Switzerland*
*santiago.helbig@univie.ac.at

Magnetic hyperthermia therapy is a promising cancer treatment modality that involves injecting magnetic nanoparticles into the tumor site and applying an alternating magnetic field to generate heat. Typically, superparamagnetic single-domain iron oxide nanoparticles are used. Their heating power is however limited by the small area hysteresis and their low saturation magnetization. Synthetic antiferromagnetic disk particles (SAFDP) offer an alternative. Due to their size and shape, the disk particles could dissipate heat over a larger surface area. However, SAFDP with in-plane magnetic materials so far did not show hysteretic M(H)-loops, while SAFDP with perpendicular anisotropy show hysteresis but contain expensive materials and their coercive fields are too large. In-plane SAFDPs with optimized M(H) loops were designed by Hug and Suess [1].

We present simulation results that show the behavior of such magnetic particles. We use a micromagnetic approach [2] (Figure 1) in combination with mechanical rotation [3]. The in-plane anisotropy axis and the surrounding viscous fluid limit the motion of the disk to a rotation around its axis of least resistance and thus can be reduced to a two-dimensional problem. At a frequency of 10 MHz the SAFDP show no significant increase in energy dissipation when simulated with mechanical rotation, but the M(H)-loops (Figure 2) show differences due to the additional mechanical relaxation process even at high frequencies, which is crucial to understand in order to account for field adjustments and necessary realignment of the particles. Within the talk we discuss the magnetic and mechanical relaxation processes responsible for heat dissipation depending on field strength, frequency and particle shape.

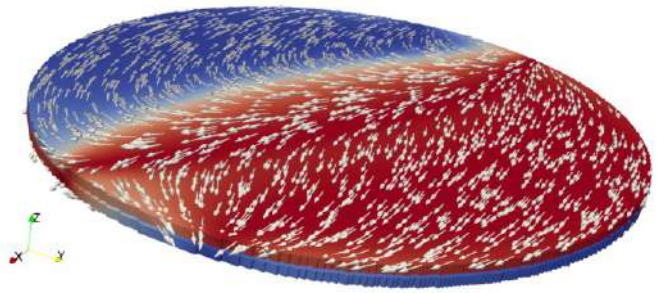


Figure 1: The transient state of the SAFDP during magnetic reversal. It shows multiple domains and the top and bottom layers with opposing magnetization indicated by the color coding.

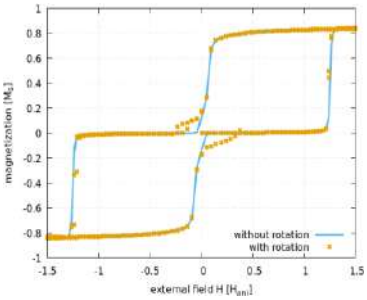



Figure 2: Comparison of the M(H)-loops for a fixed and a disk that can also rotate mechanically at 10 MHz.

## Acknowledgments

The authors wish to thank the "FWF - Der Wissenschaftsfonds" for funding under the Project No. P 33748 and the Vienna Scientific Cluster (VSC) for providing the necessary computational resources. We acknowledge financial support by the Vienna Doctoral School in Physics (VDSP).

## References

[1] H. J. Hug and D. Suess (2021) Patent No. PCT/EP2021/083572.
[2] F. Bruckner, S. Koraltan, C. Abert, D. Suess, arXiv preprint (2023) arXiv:2302.08843.
[3] S. Helbig, C. Abert, P. Sánchez, S. Kantorovich, D. Suess, Phys. Rev. B 107 (2023) 054416.

# Estimating the heat spatial distribution of complex nanoparticle aggregates for magnetic hyperthermia

*Javier Ortega-Julia,[1, 2] Jonathan Leliaert,[3] Daniel Ortega[1,2,4*]*

[1] *University of Cádiz, Condensed Matter Physics Dept., Puerto Real, Spain*
[2] *Institute of Research and Innovation in Biomedical Sciences of Cádiz (INiBICA), Cádiz, Spain*
[3] *Department of Solid State Sciences, Ghent University, Ghent, Belgium*
*3| IMDEA Nanoscience, Madrid, Spain*

*daniel.ortega@uca.es

Magnetic nanoparticles (MNP) tend to form aggregates when injected in living tissues, which alters their response to external alternating magnetic fields in magnetic hyperthermia treatments. Understanding and predicting the heat released by these MNP aggregates is central to treatment planning in clinical magnetic hyperthermia.

We propose and validate an equation that can be used to resolve the individual heat dissipation of interacting MNP at nonzero temperature. After assessing this equation for different model systems, we have found that the proportion of heat dissipated in each individual MNP tends to become more uniformly distributed for larger applied fields [1]. Harnessing the developed methodology, we next performed an *in silico* analysis to investigate the heat released by MNP aggregates featuring different size and fractal geometry factors [2]. By digitally mirroring aggregates seen in biological tissues, we found that the average heat released per particle stabilizes starting from moderately small aggregates, facilitating the estimates for their larger counterparts. Additionally, we studied the heating performance of particle aggregates over a wide range of fractal parameters. We compared this result with the heat released by non-interacting MNP to quantify the reduction of heating power after being instilled into tissues.

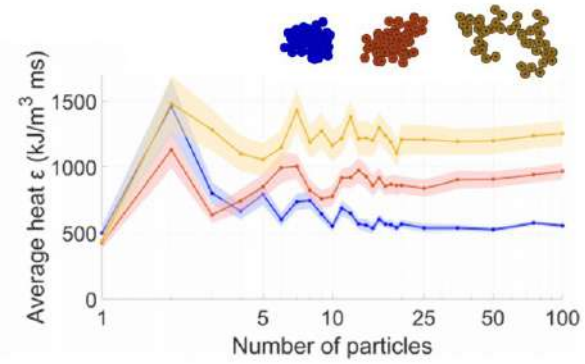



**Figure 1.** Evolution of the heat dissipated per particle as a function of the number of particles in aggregates with different shapes.

Our results can be used to estimate the expected heating *in vivo* based on the experimentally determined MNP properties, thus addressing one of the most longstanding challenges in hyperthermia, which is to achieve a homogeneous therapeutic temperature distribution in the target region during a treatment.

**Acknowledgements**

We acknowledge support from FWO (fellowship No. 12W7622N); PID2020-117544RB-I00 and RED2018-102626-T funded by MCIN/AEI/ 10.13039/501100011033, and grant RYC2018-025253-I funded by MCIN/AEI/10.13039/501100011033, and the European Commission (NoCanTher GA No. 685795).

**References**

[1] J. Leliaert, J. Ortega-Julia, D. Ortega, *Nanoscale* **13** (2021) 14734
[2] J. Ortega-Julia, D: Ortega, J. Leliaert, arXiv preprint 2207.14551

# Modeling the Inverse-Faraday-Effect as a Driving Stimulus of Domain-Wall Dynamics Within a High-Temperature Micromagnetic Formalism

*Paul-Iulian Gavriloaea[1]*, Rubén Otxoa[2], Oksana Chubykalo-Fesenko[1]*

[1] *Instituto de Ciencia de Materiales de Madrid, CSIC, Cantoblanco, 28049 Madrid, Spain,*
[2] *Hitachi Cambridge Laboratory, Hitachi Europe Limited, Cambridge CB3 0HE, UK*
*paul.gavriloaea@csic.es

The displacement of ferromagnetic domain walls (DWs) can be achieved in a variety of ways: using fields, currents, thermal gradients etc. With the continuous development of the magneto-optics research field, [1] the use of ultrafast circularly-polarised laser stimuli has also been demonstrated capable of inducing a deterministic DW displacement with the Inverse-Faraday effect (IFE) playing one of the most important roles. [2]

While the IFE is traditionally assumed to govern the magnetisation dynamics based on an effective magneto-optical field acting along the light-propagation axis, [2-3] recent ab-initio studies describe the effect both in terms of an helicity-dependent magnetic moment or torque. [4-5] Here we employ high-temperature micromagnetics described by the Landau-Lifshitz-Bloch equation [6] with the aim of investigating the role of the IFE in driving DW dynamics in the three aforementioned pictures. Inducing a magnetisation modulus gradient across an $180^0$ Néel wall, we first show that pure longitudinal relaxation leads to DW displacement towards the reduced magnetisation region similarly to the Spin-Seebeck effect, but disregarding any additional field or thermal gradients. This method is compared with the field and torque actuation IFE schemes, investigating the DW displacement and velocity as a function of intrinsic damping and electron temperature.

Our work demonstrates the three IFE pictures qualitatively describe identical DW dynamics and showcases the final displacement is proportional to the electron temperature while an increase in intrinsic damping leads to an increase of the DW velocity without affecting the final displacement. Finally, we show how in the presence of the Dzyaloshinskii-Moriya interaction, it is possible to augment the DW motion and achieve larger as well as faster DW displacements even under the application of one laser pulse.

**Acknowledgements**

This project has received funding from the European Union's Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement No 861300 COMRAD.

**References**

[1] A.V. Kimel, M. Li, *Nat. Rev. Mater*. **4** (2019) 189–200.
[2] V. Raposo, R. Guedas, F. García-Sánchez , M.A. Hernández, M. Zazo, E. Martínez, *Appl. Sci.* **10** (2020) 1307.
[3] K. Vahaplar, A. M. Kalashnikova, A. V. Kimel, S. Gerlach, D. Hinzke, U. Nowak, R. Chantrell, A. Tsukamoto, A. Itoh, A. Kirilyuk, Th. Rasing, *Phys. Rev. B* **85** (2012) 104402.
[4] R. John, M. Berritta, D. Hinzke, C. Müller, T. Santos, H. Ulrichs, P. Nieves, J. Walowski, R. Mondal, O. Chubykalo-Fesenko, J. McCord, P. M. Oppeneer, U. Nowak, M. Münzenberg, *Sci. Rep.* **7** (2017) 4114.
[5] F. Freimuth, S. Blügel, Y. Mokrousov, *Phys. Rev. B* **94** (2016) 144432.
[6] U. Atxitia, O. Chubykalo-Fesenko, N. Kazantseva, D. Hinzke, U. Nowak, R. W. Chantrell (2007) *Appl. Phys. Lett.* **91** (2007) 232507.

# Tuning Magnetic Relaxation Processes via Nanoparticle Organization

*A. Kuncser[1*], C. Radu[1], N. Iacob, I. D. Vlaicu[1], G. Schinteie[1]*
*[1]National Institute of Materials Physics, Atomistilor 405A, Magurele, Romania*
*andrei.kuncser@infim.ro

Deeper perspectives on the magnetic relaxation phenomena fundamentals in magnetic nanoparticles (MNP) systems can be particularly interesting in the field of magnetic hyperthermia as a promising supporting therapy against various types of cancer [1].

In the case of magnetic hyperthermia, the heating up to 45 C of the tumoral tissue is accomplished using the power dissipated by MNPs under the influence of an oscillating magnetic field [2]. The mechanisms of power dissipation from the MNP to the environment is related to the magnetic relaxation phenomena inside the nanoparticles [3]. The present work explores the influence of MNP morphology and organization (e.g. spheres, stripes) on the magnetic relaxation constant and uniaxial anisotropy of individual entities within the system. This is accomplished by computational methods, involving both micromagnetic simulations (using Object Oriented Micromagnetic Framework) and in-house designed software for data manipulation. An elementary proof of concept is provided in Fig1.

## Acknowledgements

*Financial support of* Romanian Ministry of Research and Innovation *through project PN-III-P1-1.1-TE-2021-0273, contract TE86/2022 is highly acknowledged*

## References

[1] A. Wlodarczyk at al., Nanomaterials. **12**(11) (2022) 1807
[2] A. Skumiel, J. Magn and Magn Mat. 553 (2022) 169294
[3] A. Kuncser, N. Iacob, V. Kuncser, B.J. Nano. **10** (2019) 1280-1289

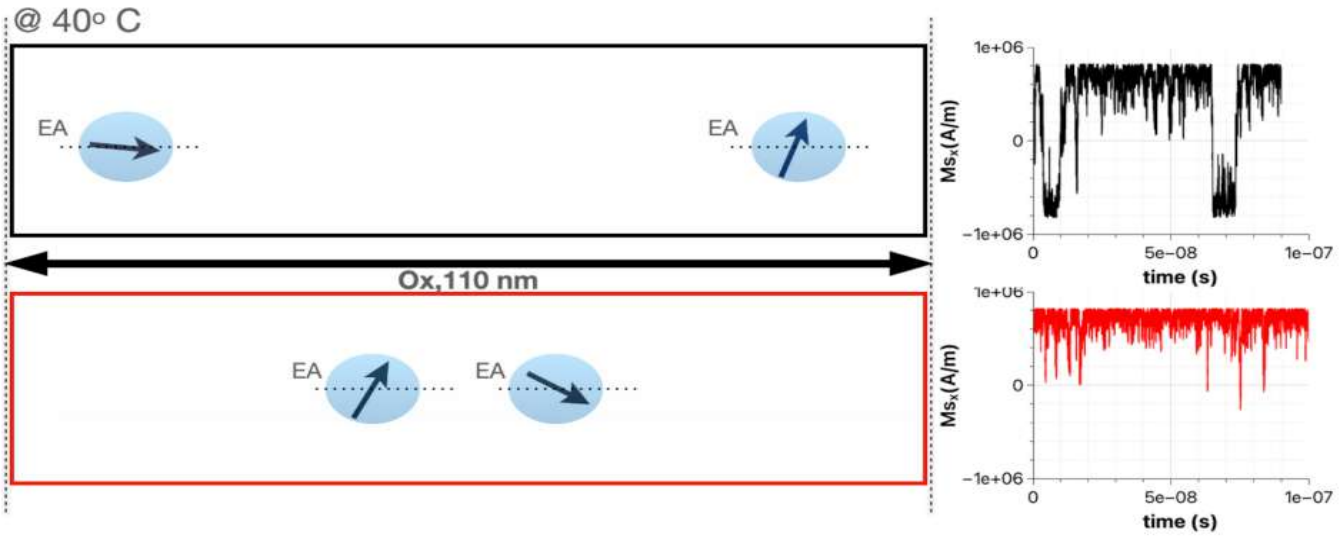



**Fig. 1:** Thermal fluctuations of a $Fe_3O_4$ MNP magnetic moment as affected by the presence of another identical MNP.

# Phonon and magnon jets above the critical current in nanowires with planar domain walls

***Maria Stamenova*** [1*], ***Plamen Stamenov*** [1] ***and Tchavdar Todorov*** [2]
[1] *School of Physics and CRANN, Trinity College Dublin, Ireland*
[2] *Centre for Quantum Materials and Technologies, Queen's University Belfast, UK*
*ctamenom@tcd.ie

The spontaneous emission of phonons and the corresponding Joule heating has long been considered as the main stability-limiting mechanism in nanoscopic conductors under large current densities. However, stimulated emission of phonons is also predicted to play a role [1]. In spintronic devices in this regime there is a further possibility for non-trivial interplay between the lattice and magnetic excitations.

Here we report on time-dependent non-adiabatic open-boundary simulations coupling electronic, lattice and spin degrees of freedom in magnetic atomic wires, which feature a narrow domain wall [2]. Above the critical current density of $10^{12}$-$10^{13}$ A/m$^2$, violent phonon and magnon jets are generated, which absorb almost fully the electron momentum flux and dramatically increase the resistance of the nanowire. This poses a fundamental limit on the currents that can be passed with impunity through electronic and spintronic devices. At the same time it opens up, for instance, an exciting possibility for generating intense magnonic currents for spintronic applications.

**Acknowledgements**

We gratefully acknowledge funding from SFI Grants No. 18/SIRG/5515 and 18/NSFC/MANIAC.

**References**

[1] T. N. Todorov, B. Cunningham, D. Dundas, and A. P. Horsfield, Mater. Sci. Technol. **33**, 1442 (2017).
[2] M. Stamenova, P. Stamenov, and T. N. Todorov, submitted.

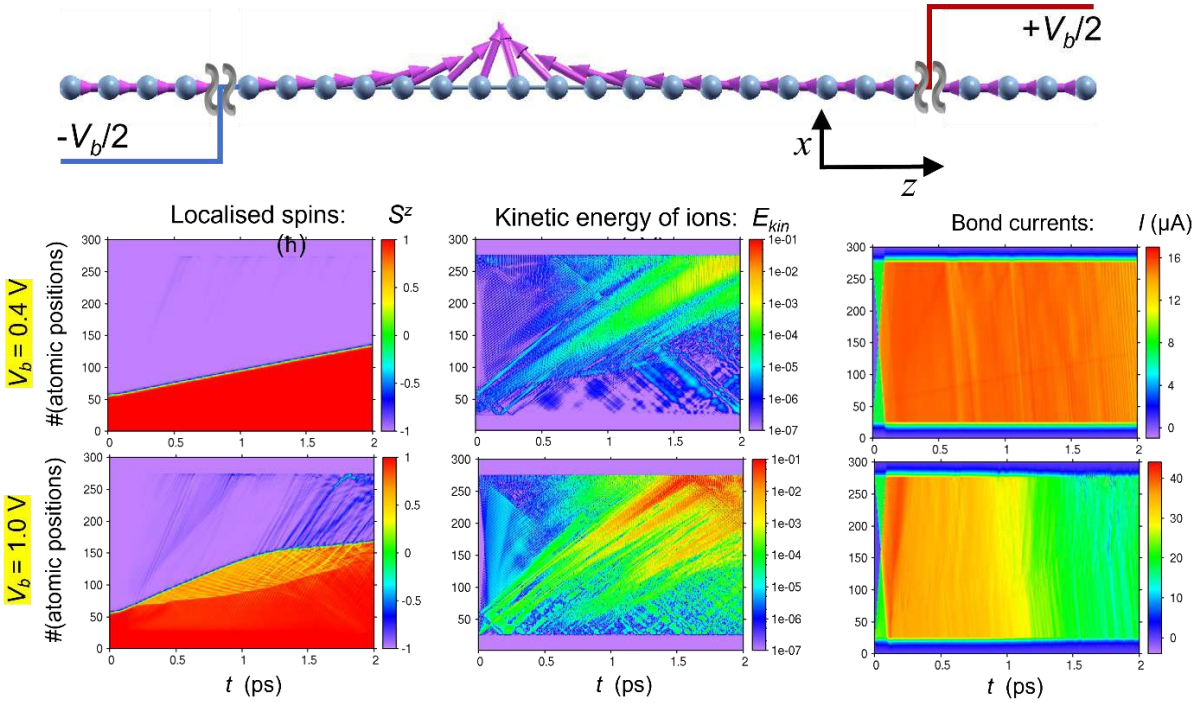



Figure: A schematic of the atomic wire bearing a narrow planar domain wall and the open boundary conditions. Heatmaps for two applied bias voltages (in rows), across the current-density threshold, showing (by columns) the atomically-resolved dynamical evolution of the longitudinal (z) component of the localised spins, the kinetic energy of the ions and the bond currents in the nanowire.

# Exchange Bias Effect in Compensated Ferrimagnets

*I. Fita,[1] E. E. Zubov,[2] R. Puzniak[1], and A. Wisniewski[1]**
[1] *Institute of Physics, Polish Academy of Sciences, Aleja Lotnikow 32/46, PL-02668 Warsaw, Poland*
[2] *Kyiv Academic University, 36 Acad. Vernadsky Blvd., UA-03142 Kyiv, Ukraine*
*wisni@ifpan.edu.pl

Rare-earth orthoferrites $R$FeO$_3$, where $R$ is a rare-earth ion, are narrow-gap multiferroics and are of considerable interest in the creation of multifunctional devices with magnetoelectric coupling ([see, e.g.[1]). These compounds meet such requirements as the natural abundance of the constituent chemical elements and the relative cheapness of their synthesis. A fairly large amount of research on the physical properties of orthoferrites in recent years is due to the presence of the phenomena of magnetic moments terahertz frequency dynamics, negative magnetization, exchange bias (EB), spin switching, and magnetocaloric effect induced by magnetic and thermal treatment.

The exchange-bias effect in single crystals of compensated ferrimagnets $R$FeO$_3$ ($R$ = Nd, Sm, Er) is discussed. In these compounds, the weak ferromagnetic (FM) moment results from the canted antiferromagnetic (AFM) ordering of Fe spins below $T_N \approx 700$ K due to the Dzyaloshinskii-Moriya interactions, while the opposite compensating paramagnetic moment of $R$ spins appears owing to a strong AFM coupling between 4$f$ and 3$d$ ions within the unit cell. Due to this mechanism, the Er, Nd, and Sm orthoferrites exhibit a specific $T_{comp}$ at which the two opposite moments cancel each other, so the net magnetization vanishes, and below $T_{comp}$ the FM moment is aligned oppositely to the moderate applied magnetic field, demonstrating a negative magnetization. It was found that the magnetization hysteresis loops of these ferrimagnets are analogously exchange biased around their compensation temperatures $T_{comp}$ [2]. Interestingly, in spite of very different $R$-Fe interactions, $T_{comp}$ values, and spin-reorientation temperatures, the EB field similarly emerges and diverges upon approaching $T_{comp}$ and changes sign with crossing $T_{comp}$. In addition, SmFeO$_3$, with a complicated AFM order caused by the nonequivalent Fe spin configuration, shows EB also at temperatures far above $T_{comp}$ and its sign alters from negative to positive with increasing cooling field.

In order to explain EB in these compounds, a mean-field theory approach for the representative ErFeO$_3$ orthoferrite was proposed [3]. The general case of two sublattices, antiferromagnet with exchange anisotropy and $R$-Fe interactions, was considered. It was shown that small applied magnetic field is a source of additional anisotropy, resulting from canting of sublattice moments. This anisotropy leads to an imbalance of free energy for two different types of magnetic domains, causing a spin jump near the $T_{comp}$. The proposed model makes it possible to reproduce magnetization reversal and main features of EB effect in ErFeO$_3$.

ErFeO$_3$ exhibits another specific phenomenon: *temperature* driven spin switching and exchange bias [4]. The EB manifests itself as the temperature shift of the hysteresis loops $M$ vs $T$, which occurs upon successive cooling and heating in a weak magnetic field. The $M(T)$ loops limiting the region of coexistence of negative and positive magnetization are shifted towards lower or higher temperatures, depending on the sign of the applied magnetic field, which causes the unidirectional EB anisotropy. The EB anisotropy energy, which contributes to the energy barrier for switching spins to an equilibrium state, determines the shift in the switching temperature $T_{sw}$.

A model of nonequilibrium thermodynamics of ErFeO$_3$ was proposed. The quantum-mechanical effects in arbitrary magnetic fields related both to the direction of a general quantization axis and an influence of anisotropic exchange interactions on the magnetic structure, spin-reorientation phase transition, spin reversals, and hysteresis were considered. Based on recent experimental data on temperature-induced spin switching, a possible mechanism for the exchange bias of magnetic hysteresis loops is proposed [5].

## References

[1] D. G. Oh, J. H. Kim, H. J. Shin, Y. J. Choi, N. Lee, Sci. Rep. **10**, 11825 (2020).
[2] I. Fita, A. Wisniewski, R. Puzniak, E. E. Zubov, V. Markovich, G. Gorodetsky, Phys. Rev. B **98**, 094421 (2018).
[3] E. E. Zubov, V. Markovich, I. Fita, A. Wisniewski, R. Puzniak. Phys. Rev. B **99**, 184419 (2019).
[4] I. Fita, R. Puzniak, E. E. Zubov, P. Iwanowski, and A. Wisniewski, Phys. Rev. B **105**, 094424 (2022).
[5] E. E. Zubov, I. Fita, R. Puzniak, and A. Wisniewski, unpublished.

# SYMPOSIUM 16.
## MICROMAGNETIC AND MULTISCALE MAGNETIC MODELLING AND SIMULATIONS. S16 POSTERS

# Theory of three-wave scattering of bulk and edge spin waves

*J. Kharlan[1,2*], P. Gruszecki[1], R. Verba[2], K.Sobucki[1] ,M. Krawczyk[1]*

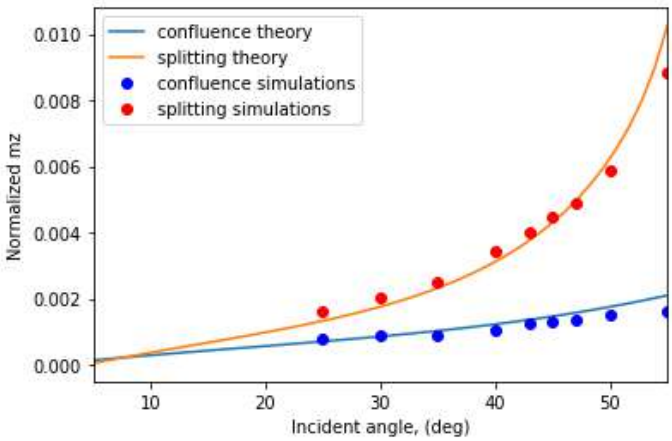
[1] *Faculty of Physics, ISQI, Adam Mickiewicz University, Poznan, Uniwersytetu Poznańskiego 2, Poznan, Poland*

[2] *Institute of Magnetism of NAS of Ukraine and MES of Ukraine, 36-b Acad. Vernadskogo Ave., Kyiv, Ukraine*

*yuliia.kharlan@amu.edu.pl

Multimagnon scattering processes play an important role in magnetization dynamics in magnetic nanostructures, constituting, in particular, an additional dissipation channel, as well as allowing for signal processing with a frequency change. Here we present a combined theoretical description supported by micromagnetic simulation results of inelastic scattering of bulk and edge spin waves (SWs) in the in-plane magnetized semi-infinite ferromagnetic film. Although this topic was considered in Ref. [1], the previous theoretical approach failed to explain obtained in the simulations significant difference between confluence and splitting efficiencies, because didn't take into account the influence of SW group velocity, a phase shift between an incident and reflected beams, the ellipticity of dynamical magnetization precession, etc.

Using vector Hamiltonian formalism [2] for calculations of three magnon coefficients and Hamilton motion equations for describing magnetization dynamic, we have developed a strict, but quite simple analytical model and derived efficiency of the 50 GHz SW beam scattering on edge SWs at frequency 12 GHz depending on the incident beam angle (we assumed external magnetic field applied perpendicularly to the film edge and angle counted from the edge normal). As seen in Fig.1, the magnetization amplitude of the resulting wave for the stimulated splitting process is much larger than for confluence one, and both processes' efficiencies are increasing with the incident angle. The theoretical calculations are in perfect agreement with the simulations.




It should be noted, that three-magnon efficiency is determined by the range of factors (three-magnon coefficient, group velocity, incident angle, film thickness, edge mode depth length), which opens possibilities to control inelastic scattering efficiency and, consequently, is of great importance due to the various applications in magnonics.

Fig. 1 Comparison between the results of micromagnetic simulations (empty circles) with theoretical calculation (solid lines) for the angular dependence of normalized magnetization z-component of inelastically scattered waves. Blue circles and blue line correspond to the confluence process, and red circles and red line correspond to the splitting process, respectively.

## Acknowledgements

The authors acknowledge the support from the grant of the National Science Center – Poland, No. 2021/43/I/ST3/00550.

## References

[1] P. Gruszecki, K.Y. Guslienko, I.L. Lyubchanskii, M. Krawczyk, arXiv:2106.09954 [cond-mat.mes-hall] 18 Jun 2021.

[2] V. Tyberkevych, A. Slavin, P. Artemchuk, and G. Rowlands. Vector Hamiltonian Formalism for Nonlinear Magnetization Dynamics. arXiv:2011.13562 [cond-mat.mtrl-sci].

# Micromagnetic Simulations of Domain Walls in Coupled Synthetic Antiferromagnetic Nanowires with In-Plane Magnetization

*Jhon Chiliquinga*[1]*, *Maria J. Benitez*[1]

[1]*Departamento de Física, Escuela Politécnica Nacional, Quito, Ecuador*

*jhon.chiliquinga@epn.edu.ec

The study of the structure and dynamics of magnetic domain walls (DW) in nanowires has been of great interest in the last years due to their possible applications in sensors, racetrack memories and logic devices [1]. Much work up to now has focused on Permalloy single layers, showing well defined domain walls [2]. However, the walls supported in such nanowires are extended, having dimensions of the width of the wire, and requiring high current densities to set them in motion, which are inefficient for technological devices. It has been shown that a synthetic antiferromagnet (SAF) system can simplify the wall structure and the current densities needed to set these walls in motion are much lower than the values reported for other systems [4]. Therefore, a deeper understanding of the DW structure of these systems is relevant.

In this work, using Object Oriented MicroMagnetic Framework (OOMMF), we studied two DWs structures, parallel and antiparallel (fig. a, b), in Co/Ru/Co SAF-trilayer nanowires, depending on the wire geometry. The geometrical independent variables were the width of the SAF (w) and its top layer thickness (t); while the length (5000 nm), the bottom layer thickness (4 nm), and the spacer layer thickness (1 nm) were held constant.

We find that in all cases, the relative wall width in the parallel DW configuration was narrower than the antiparallel one (fig. c). When varying the SAF width, Δ/w tends to decrease with increasing w. Furthermore, with antiparallel DWs we found that for equally sized Co layers the relative wall width decreases faster with w than the case where the top layer is thicker.

**References**

[1] Parkin, S., Yang, SH. Memory on the racetrack. *Nature Nanotech* 10, (2015) 195–198.
[2] Y. Nakatani, A. Thiaville, J. Miltat, *J. Magn. Magn. Mater*. 290 (2005) 750–753.
[3] Yamaguchi, A. et al. *Phys. Rev. Lett.* 92 (2004) 077205.
[4] Lepadatu, S., Saarikoski, H., Beacham, R. et al. *Sci Rep* 7 (2017) 1640.

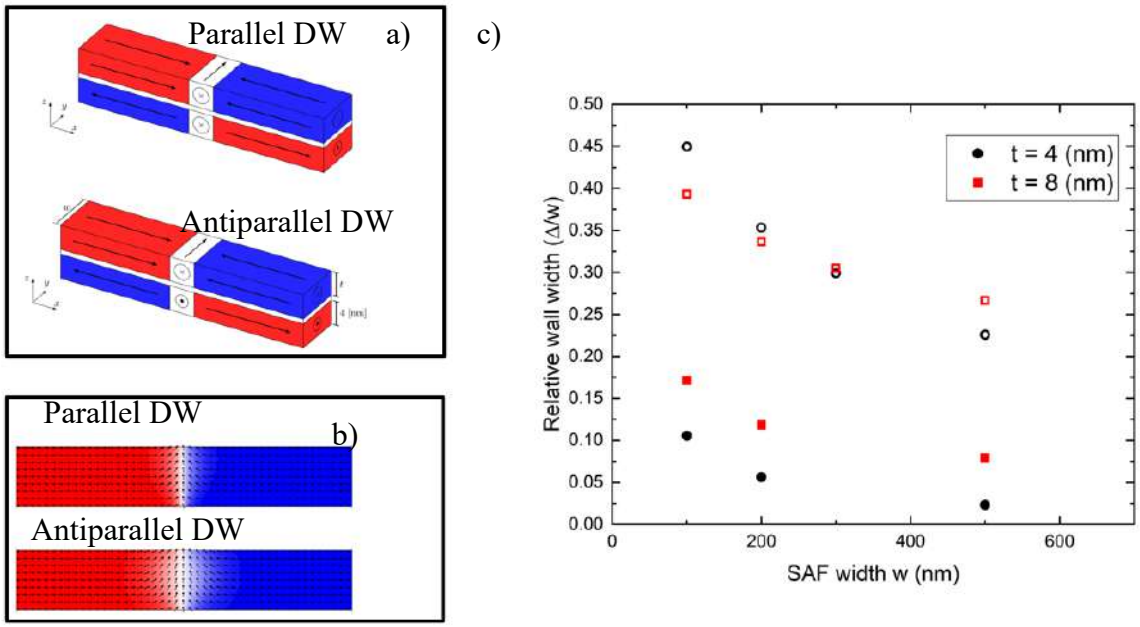



Figure: a) Initial magnetization scheme. Arrows represent a specific direction of magnetization in a certain domain. b) DWs in the upper layer of a 100 nm wide Co/Ru/Co SAF-trilayer nanowire with 8 nm thick Co top layer, simulated using OOMMF. c) Relative wall width (Δ/w) dependence on SAF width w. The filled dots correspond to the parallel DW and the hollow ones to the antiparallel DW.

# Dynamic Hysteresis and Specific Loss Power of Magnetite Nanoparticles: Viscosity, Temperature and Orientation Effects

*J. Restrepo[1]*, J. C. Zapata[1], and N. Roa-Motta*

[1] *Group of Magnetism and Simulation G+. Universidad de Antioquia. A.A. 1226, Medellín, Colombia*

*johans.restrepo@udea.edu.co

In this work, a computational analysis of magnetite single-domain magnetic nanoparticles in colloidal suspension in water above and below freezing temperature is carried out, in order to study the response of magnetization to time-dependent alternating external magnetic fields. In our model, a Hamiltonian is considered that includes Zeeman interactions and the uniaxial magnetocrystalline anisotropy energy, and the Landau-Lifshitz-Gilbert stochastic differential equation is solved together with a torque equation. In this way it is possible to simulate the Néel rotation [1] mechanisms of the magnetization and that of Brown [2] of the easy axis of the nanoparticle, respectively. Both equations include a white noise stochastic term associated with thermal bath. The Voger-Fulcher-Tamman (VLT) model was also used, which takes into account a temperature-dependent viscosity term for the liquid phase of water. The results allowed to obtain the hysteresis cycles (see Figure 1) and with them the specific loss power, which is a relevant quantity in magnetic hyperthermia. Finally, an analysis of the influence of the orientation of the particles, the temperature of the medium, the frequency of the applied magnetic field and the size of the nanoparticles suspended in the medium was made.

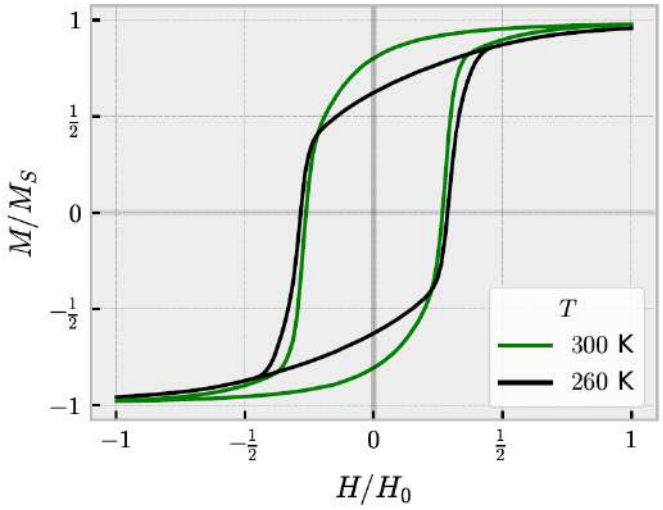



Figure 1. Hysteresis loops for two different temperatures of the magnetic fluid above and below the freezing point of water.

## Acknowledgements

Financial support was provided by the CODI-UdeA 2020-34211, 2022-51311, 2022-51312 and 2022-51330 projects.

## References

[1] Néel, L. Ann Geophys., 5, 99, (1949).

[2] Brown, W.J., Phys. Rev., 130, 1677, (1963).

# Inhomogeneous Magnetization in Packed Beds

*Nikka Mosleh[1]*, Christian R.H. Bahl[1], Rasmus Bjørk[1]*

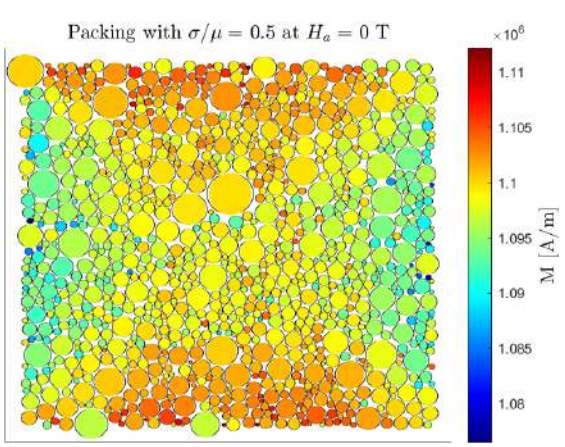
[1]*Department of Energy Conversion and Storage, Technical University of Denmark, Anker Engelunds vej 301, 2800 Kgs. Lyngby, Denmark*

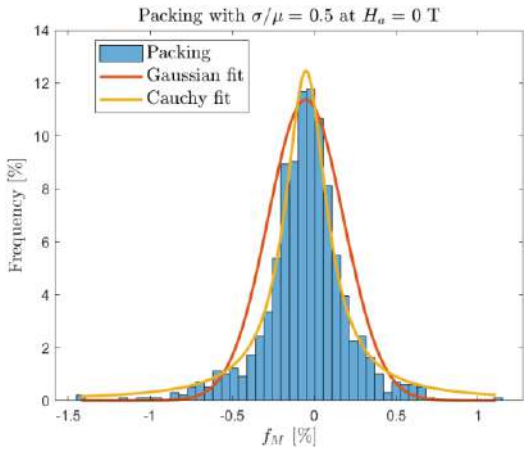
*nikkam@dtu.dk

Magnetic particles are almost always found in assemblies, i.e in clusters. This clustering or packing will affect the magnetic properties of both the entire collection of particles, but also their individual properties, depending on their location and relative size compared to the packing. The goal of this work is to investigate the importance of the size distribution of magnetic grains, and whether or not this has an effect on the magnetic properties of a resulting magnet. Here we do this by numerically investigating the influence of packing infinitely long cylindrical magnetic particles. i.e. a 2D-system. Each magnetic particle has a defined hysteresis curve and we characterize the overall hysteresis curve of the packing. We study how the average magnetization of the packings depend on the applied field for packings consisting of particles of different sizes, which have been random closely packed. The radii of theparticles follow a lognormal size distribution, and the packings are distinguished by the standard deviation of this distribution.

We examine how the demagnetizing field affects a packing of magnetic particles differently than an equivalent solid geometry. We do this by considering the difference between the average magnetization within a magnetic particle in the packing and the average magnetization of the corresponding area of a solid sample.

Blow the average magnetization, *M*, within each particle in the packing following a lognormal distribution with parameters being the mean $\mu = 1$ and standard deviation $\sigma/\mu = 1$ (i.e. the natural logarithm of the radii are normally distributed with the corresponding mean and standard deviation) is shown in the figure to the left. In the right figure a histogram of the relative difference $f_M = (M_{packing} - M_{solid})/|M_{solid}|$ between the average magnetization of a magnetic particle in the packing and of the corresponding area of a solid sample is shown. As can be seen, some particles are more and some less magnetic compared to a solid sample. A Gaussian fit and a Cauchy fit of the histogram are also shown.




Doing this for a range of different packings distinguished by different standard deviations in the particle size distribution reveals that the packings have a relative difference in average magnetization with a spread that ranges from approximately 0% to 25%, i.e. the spread in the distribution in the right figure. The average magnetization deviates the most from that of a solid sample when the spread of the particle size distribution is large and when the applied field is close to the coercivity of the magnetic particles.

**Acknowledgements**

The authos wish to acknowledge the Independent Research Fund Denmark, grant “Magnetic Enhancements through Nanoscale Orientation (METEOR)”, 1032-00251B, for sponsoring this work.

# First-principles study of perpendicular magnetic anisotropy in CoFeB/MgO/CoFeB magnetic tunnel junctions

*Justyn Snarski-Adamski[1]*, Ján Rusz[2], Mirosław Werwiński[1]*

[1] *Institute of Molecular Physics, Polish Academy of Sciences, M. Smoluchowskiego 17, 60-179 Poznań, Poland*

[2] *Department of Physics and Astronomy, Uppsala University, Box 516, SE-751 20 Uppsala, Sweden*

*justyn.snarski-adamski@ifmpan.poznan.pl

Magnetic tunnel junctions with perpendicular anisotropy are an intensively developed basic element for applications in spintronic devices [1]. Today's junctions have a nominal thickness of CoFeB magnetic layers and an insulating MgO spacer in the range of 1.0 nm, or just a few atomic monolayers [1]. This device thickness makes 1:1 modeling possible using a first-principles calculation method, where materials are modeled at the atomic scale taking into account a complete quantum-mechanical description of their electron structure. In the work presented here, within the framework of density functional theory (DFT) and FPLO code [2], we model CoFeB/MgO/CoFeB trilayers with a magnetic film thickness of about 0.8 nm (five atomic monolayers) depending on the thickness of the non-magnetic spacer of about 0.8, 0.5 and 0.2 nm (five, three and one atomic monolayers). We thus continue and extend our earlier theoretical consideration of the Fe/MgO interface [3]. Figure shows the model with five-monolayer thicknesses of the MgO spacer. The calculations performed make it possible to determine the type of coupling versus thickness relationship between two magnetic films (parallel or antiparallel coupling) and the direction of easy magnetization in the magnetic films (in plane or perpendicular to plane), thus defining the magnetic anisotropy of the system. Furthermore, determining the magnetic moment and electric charge on individual atoms in the film makes it possible to detail the properties of the CoFeB/MgO interface and the CoFeB surface. The presented results provide a better understanding of the magnetic tunnel junctions developed so far and enable the development of improved devices.

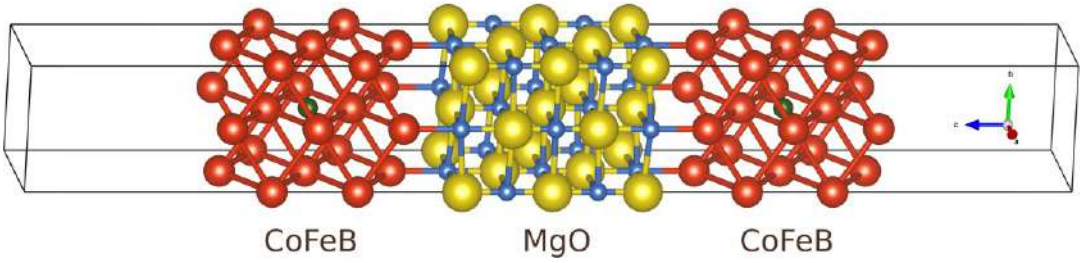



Figure: The considered CoFeB/MgO/CoFeB magnetic tunnel junction with a five atomic monolayers of MgO. Yellow, blue, red, and green balls represent Mg, O, Fe/Co, and B atoms, respectively.

## Acknowledgements

We acknowledge the financial support of the National Science Centre Poland under the decisions DEC-2019/35/O/ST5/02980 and DEC-2018/30/E/ST3/00267 (PRELUDIUM BIS 1 and SONATA BIS 8).

## References

[1] S. Ikeda, K. Miura, H. Yamamoto, K. Mizunuma, H.D. Gan, M. Endo, S. Kanai, J. Hayakawa, F. Matsukura, H. Ohno, A perpendicular-anisotropy CoFeB–MgO magnetic tunnel junction, Nature Mater. 9 (2010) 721–724.

[2] K. Koepernik, H. Eschrig, Full-potential nonorthogonal local-orbital minimum-basis band-structure scheme, Physical Review B. 59 (1999) 1743–1757.

T. Thersleff, S. Muto, M. Werwiński, J. Spiegelberg, Y. Kvashnin, B. Hjörvarsson, O. Eriksson, J. Rusz, K. Leifer, Towards sub-nanometer real-space observation of spin and orbital magnetism at the Fe/MgO interface, Sci. Rep. 7 (2017) 44802.

# Spin Waves in Ferrimagnets at and around the Angular Magnetization Compensation Temperature: A Micromagnetic Study

*Luis Sánchez-Tejerina[1]*, David Osuna Ruiz[2], Eduardo Martínez Vecino[3], Luis López-Díaz[3], Óscar Alejos[1]*

[1] *Department of Electricity and Electronics, University of Valladolid, Spain*

[2] *Department of Electrical, Electronical and Communications Engineering. Public University of Navarra, Spain*

[1] *Department of Applied Physics, University of Salamanca, Spain*

*luis.sanchez-tejerina@uva.es

Magnon propagation in antiferromagnetic (AFM) and/or ferrimagnetic (FiM) insulators has drawn attention recently due to the benefits as opposed to their ferromagnetic (FM) counterparts, such as higher fundamental frequency and insensitivity to external magnetic field perturbations. Spin wave (SW) propagation in these materials connects magnonics with spintronics, since spin current is mainly transported by magnons.

Here, SW propagation along a FiM strip with out-of-plane magnetization is studied by means of micromagnetic simulations. The FiM material is considered as formed by two antiferromagnetically coupled sublattices, each having its own temperature dependent saturation magnetization. Two critical temperatures can be defined for such systems: that of magnetization compensation ($T_M$ ) and that of angular momentum compensation ($T_A$), both different due to distinct Landé factors for each sublattice. SWs in the strip are promoted by a spin current injected at one of its edges. This current is modulated as to determine the dispersion curves shown in Figure 1.

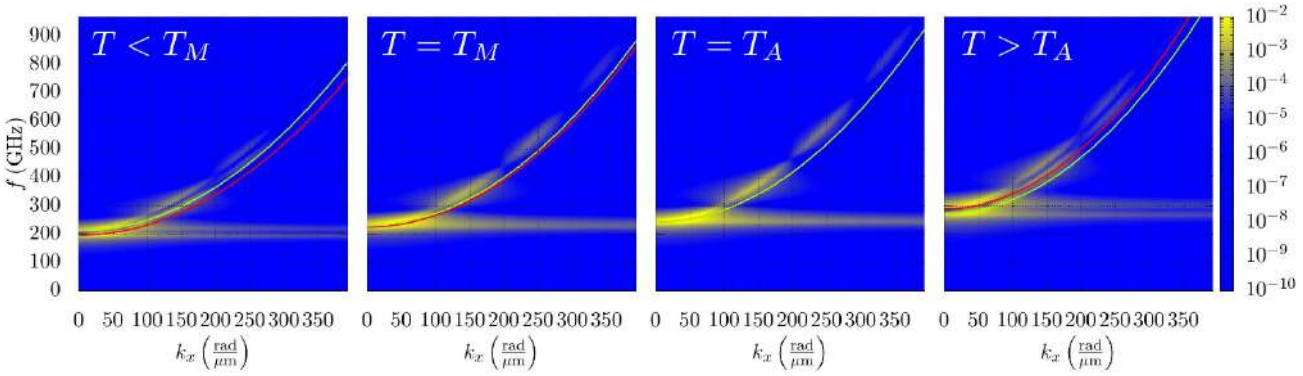



Figure 1: Dispersion curves for a FiM at different temperatures, including the relevant cases of both magnetization andangular momentum compensation.

From the figure, the exchange-dominated character of the SWs can be verified, in the form of a $k^2$-dependent mode frequency. Solid curves show the fitting to the data from Kalinikos and Slavin's theory [1], when each sublattice magnetization is modeled as of separate effective FM. These curves reveal that SWs travel with different phase velocities through each sublattice except at $T_A$. As a result, the in-plane components of the Neel vector describes elliptic trajectories where the orientation of their axes is found to be position dependent whereas their eccentricity is determined by the temperature. Consequently, different positions are subjected to different torques, not only by the amplitude fading but because this reorientation effect. Therefore, fine control of magnetization by means of SWs in FiM materials must take into account the reported effect.

**Acknowledgements**

This work was supported by Projects No. PID2020117024GB-C41 funded by MCIN/AEI/10.13039/501100011033, No. SA114P20 from Junta de Castilla y León, and Project MagnEFi, Grant Agreement No. 860060, (H2020-MSCA-ITN-2019) funded by the European Commission.

**References**

[1] B. A. Kalinikos and A. N. Slavin, *Journal of Physics C: Solid State Physics* **19**, (1986) 7013–7033.

# Spin and orbital Edelstein effect in a bi- and trilayer system with Rashba interaction

*Sergio Leiva M.*[1]*, *Jürgen Henk*[1], *Ingrid Mertig*[1] , *Annika Johansson*[2]

[1] *Martin Luther Universiy Halle-Wittenberg, Von-Danckelmann-Platz 1, 06120 Halle, Germany.*

[2] *I Max Planck Institute of Microstructure Physics, Weinberg 2, 06120 Halle, Germany.*

*sergio-tomas.leiva-montecinos@physics.uni-halle.de

The spin Edelstein effect has proven to be a primising phenomenon to generate spin polarization from a charge current in systems without inversion symmetry. In recent years, current-induced orbital magnetization, also called the orbital Edelstein effect, has also been predicted for several systems with broken inversion symmetry [1-6].

In the present work, we calculate the current-induced spin and orbital magnetization for a bilayer and a trilayer system with Rashba interaction for the interface and the free-standing slab configurations. We use the modern theory of orbital magnetization [7] and the Boltzmann transport theory. We found a significantly larger orbital than spin effect, with a strong dependence on the model parametersm such as effective mass and spin-orbit coupling per layer. This dependence allows us to enhance and even revert the sign of the orbital effect.

**Acknowledgements**

This project has received funding from the Eurpean Union's 2020 research and innovation programme under the Marie skłodowska-Curie grant agreement No 955671.

**References**

[1] T. Yoda, T. Yokoyama, S Murakami , Sci. Rep. **5** (2015) 12024.
[2] D. Go *et.at,* Sci. Rep. **7** (2017) 46742.
[3] T Yoda, T Yokoyama, S Murakami, Nano Lett., **18** (2018) 916.
[4] L. Salemi, B. Marco, AK Nandy, P. M. Oppeneer, Nar. Commun. **10** (2019) 5381.
[5] D. Hara, MS Bahramy, S Murakami, Phys. Rev. B, **102** (2020) 18404.
[6] A. Johansson, B Göbel, J Henk, M Bibes, I Mertig, Phys. Rev. Research, **3** (2021) 013275.
[7] T. Thonhauser, D Ceresoli, D Vanderbilt, R Resta, Phys. Rev. Lett. **95** (2005) 137205.

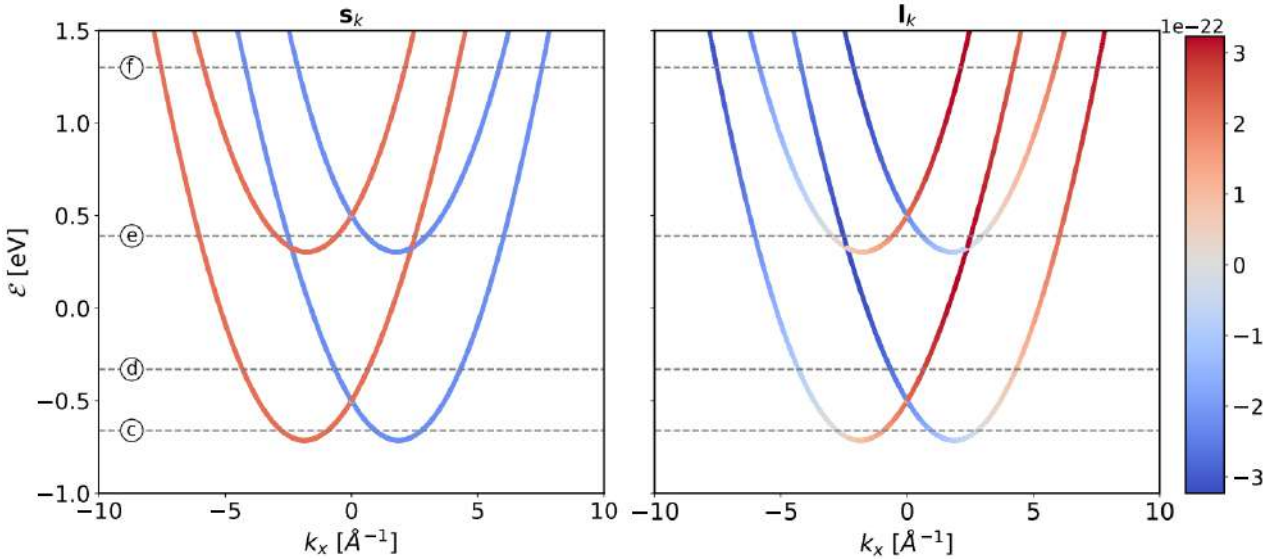



Figure: Band structure of the Rashba bilayer model. In color, the y component of the spin (left) and orbital (right) expectation values are shown.

# CMTJ: Simulation Package for Multilayer Spintronic Devices

*J. Mojsiejuk[1], S. Ziętek [1*], M. Czapkiewicz[1], K.Grochot[1,2], and W.Skowroński[1]*

[1] *AGH University of Science and Technology, Institute of Electronics, al. Mickiewicza 30, 30-059 Kraków, Poland*

[2] *AGH University of Science and Technology, Institute of Electronics, al. Mickiewicza 30, 30-059 Kraków, Poland*

*zietek@agh.edu.pl

Operation of spintronic devices relies on multiple material parameters. Some of them, for example: magnetic anisotropy, coupling energy or damping can be tuned with various engineering methods. In the recent years there is a growing need to search for optimal values of various material parameters in spintronic devices. This problem can become non-trivial due to, among other issues, the computational complexity, large number of parameters or lack of automatisation in the process.

We combine well known macromagnetic and free-energy based models (Smit-Beljers [1,2]) along with procedures for computing experimental quantities such as magnetoresistance, magnetic hysteresis loops, magnetisation dynamics and current-induced switching into a single Python package [3]. Thanks to the optimised code, the package is capable of performing multiple parameter sweeps in parallel, Bayesian optimisation or can be used to simulate large quantities of data for neural network training.

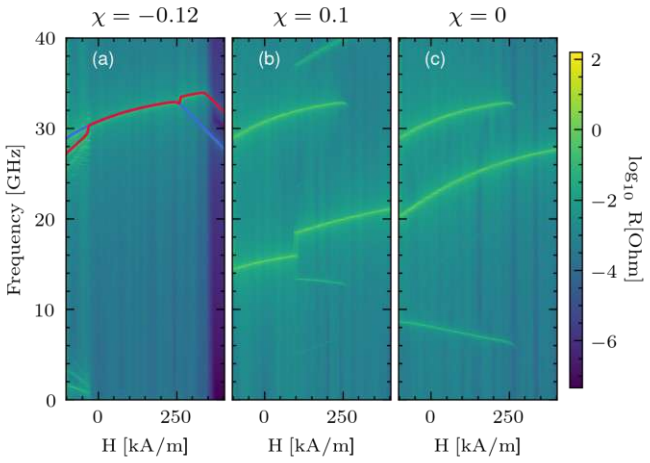



*Figure 1 a) electric synchronisation of two negatively coupled MTJs. b-c) the same MTJs but with positive and zero coupling respectively.*

In the spirit of modular development we expose additional layers of abstraction to the user which allow for constructing stack of electrically coupled devices, as demonstrated with Fig.1 where two slightly different magnetic tunnel junctions (MTJs) synchronise to a common resonance mode via the electric synchronisation model based on [4].

**Acknowledgements**

W.S acknowledges grant No. 2021/40/Q/ST5/00209 from the National Science Centre, Poland and „Excellence initiative – research university" for the AGH University of Science and Technology. K.G recognises grants: No. 2021/40/Q/ST5/00209 and 2016/23/B/ST3/01430.

**References**

[1] Baselgia, L. et al., Phys. Rev. B 38, 2237–2242 (1988).
[2] Rodríguez-Suárez, R. L. et al., Phys. Rev. B 71, 224406 (2005).
[3] Mojsiejuk, J., et al., Preprint at http://arxiv.org/abs/2207.11606 (2022) (in press, NPJ Computational Materials)
[4] Taniguchi, T., et al., Appl. Phys. Express 11, 013005 (2018).

# Quantification of Coercivities and Interactions in Ferromagnetic Systems from FORC Measurements

*José Carlos Martínez-García*, Montserrat Rivas*

[1] *Departamento de Física de la Universidad de Oviedo & IUTA, Campus de Gijón, Asturias, España*

*jcmg@uniovi.es

First Order Reversal Curve (FORC) method has received considerable attention as a powerful approach to identify microscopic details if hysteretic systems thorugh macrocopic measurements. It has been widely employed for the characterization of a variety of magnetic and other hysteretic systems. However, a coherent framework to interpret the features of a FORC diagram and extract quatitative information is still lacking in spite the efforts of numerous groups during the lasts decades.

A large number of references [1-3] can be found in the scientific literature, in which it is stated that FORC distribution provide a means to estimate the intrinsic distributions of coercivities and interactions fields within a magnetic system and, therefore, provide information on composition and domain state. Some works propose particular methods to determine the mentioned instrinsic distributions, most of them consisting in the projections on particular axis of the FORC diagram, or the extraction from it of cross sections along certain directions.

But all these methods relies in the Preisach model of hysteresis, identifying FORC diagram and Preisach plane. As is well known [4], Preisach hysteresis model requires two conditions: (1) the *wiping-out property* is accomplished by real magnetic systems, and is related to the ability of erasing the magnetic memory by appliying a saturating magnetic field; (2) the *congruency property* would require, for a magnetic system, that all the minor loops between two fixed values of the magnetic applied field must be congruent, that is, must have the same shape. But in real magnetic systems, this condition usually fails. And this is why the methods of obtaining the distributions of coercivity and interactions, which would be valid for the Preisach plane, are not for the FORC diagram.

The present work describes a way to extract, from a complete set of FORC measurements, the coercivity distribution and the internal interactions of a magnetic system, using procedures not related to the Preisach plane, but to some reasonable hypotheses about reversible and irreversible magnetization processes.

**Acknowledgements**

This work is supported by the Principality of Asturias under Project SV-PA-21-AYUD/2021/57470, and the UITA/Gijón Council under Project SV-22-GIJON-15.

**References**

[1] C. R. Pike, *Phys. Rev. B* **68** (2003) 104424.
[2] M. Winklhofer, G. T. Zimanyi, *J. Appl. Phys.* **99** (2006) 08E710.
[3] S. Ruta, O. Hovorka, PW. Huang et al, *Sci. Rep.* **7** (2017) 45218.
[4] I. D. Mayergoyz, G. Friedman, *IEEE Trans. Magn.* **34** No. 1 (1988) 212–217

# Multi-scale spin dynamics in MTJs based on ferrimagnetic $Mn_3Ga$

*Umit Daglum[1]*, Carenza Cronshaw[2], Richard Evans[2] and Maria Stamenova[1]*

[1] *School of Physics and CRANN, Trinity College Dublin, Dublin 2, Ireland*

[2] *School of Physics, Engineering and Technology, University of York, York, UK*

*dalumm@tcd.ie

A low-moment ferrimagnet, the tetragonal ($DO_{22}$) Mn3Ga exhibits a large uniaxial anisotropy, a high spin-polarization and a low Gilbert damping, which makes it a good candidate for a range of spintronic applications. For instance, it has been proposed as a promising functional material for spintronic oscillators in the THz range [1]. Recent first-principles spin-transport calculations [2] indicate significant spin-transfer torques (STT) in magnetic tunnel junctions (MTJs) based on $Mn_3Ga$ and MgO as spacer. The STTs appear staggered, but show a strong oscillatory position-dependence away from the MgO interface – hence, any macro-spin models will not be adequate in modelling the resulting current-induced dynamics in the ferrimagnet.

Here we employ and extend the multi-scale method, developed in Ref. [3], which combines *ab initio* calculations of STT with large-scale time-dependent simulations using atomistic spin dynamics [4]. We supplement the method with junction-specific (accounting for the bilateral lateral strain of $Mn_3Ga$ in the junction with MgO) long-range distance-dependent Heisenberg couplings (extracted from *ab intio* spin-spiral calculations) and anisotropy constants.

**Acknowledgements**

We gratefully acknowledge funding from SFI Grant No. 18/SIRG/5515.

**References**

[1] S. Mizukami, *et al*., Phys. Rev. Lett. **106** (2011), 11720.
[2] M. Stamenova, *et al.*, Phys. Rev. B **103** (2021), 094403 (2021); M. Stamenova, et al., AIP Advances **13** (2023), 025016.
[3] M. Ellis, *et al*., Phys. Rev. B **96** (2017), 224410.
[4] R. F. L. Evans, *et al*., J. Phys.: Condens. Matter **26** (2014), 103202.
[5] K. Rode, *et al.*, Phys. Rev. B **87** (2013), 18.

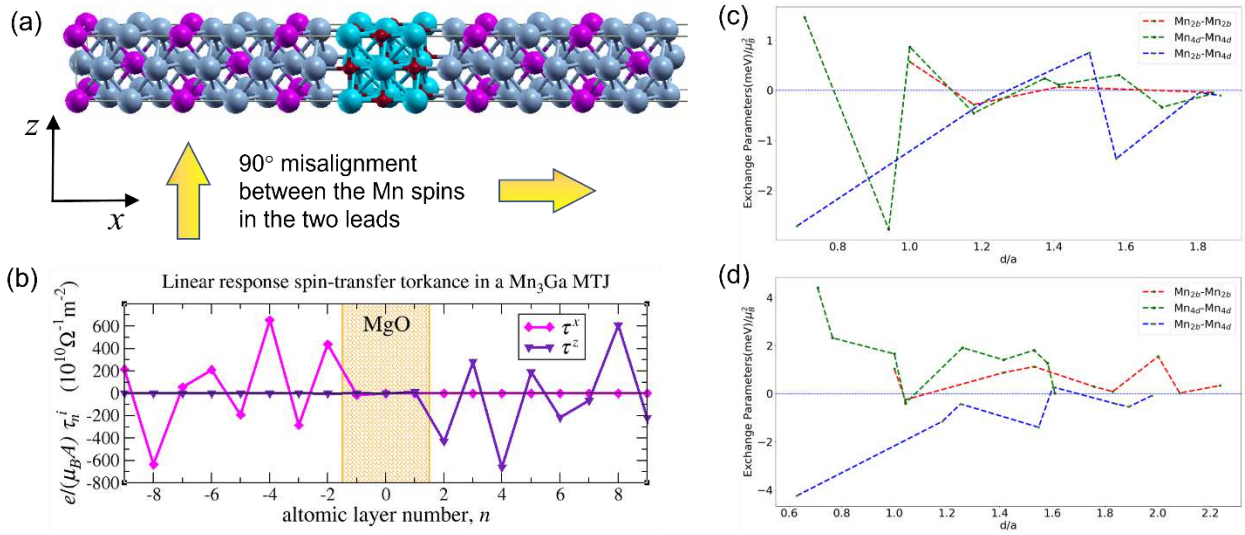



Figure: (a) Schematic of a $Mn_3Ga$ junction with MgO spacer and the orientation of the Mn spins in the two leads used to calculate (b) the atomically resolved spin-transfer torkance (in linear response ballistic transport). Distance-dependent Heisnberg parameters for tetragonal $Mn_3Ga$ from first-principles spin-spiral calculations with lattice constants from (c) experiment [5] and (b) strained in plane to match MgO.

# Modelling of rotational losses for magnetic steel sheets through micromagnetism

*Amil Ducevic[1]*, Klaus Roppert[2], Manfred Kaltenbacher[2] and Dieter Suess [1]*

*[1] Universität Wien, Universitätsring 1, Vienna, Austria*

*[2] Technische Universität Graz, Rechbauerstraße 12, Graz, Austria*

*amil.ducevic@univie.ac.at

Rotational losses are defined as the energy that is required to rotate a ferromagnet quasi-statically through an angle of 360° in a magnetic field $\boldsymbol{H}$. The losses are determined by the torque $\boldsymbol{T}$ exerted by a sample during rotation, and can be computed as one half the enclosed area by the torque $\boldsymbol{T}$ versus the rotation angle $\alpha$,

$$\frac{1}{2}\oint T(\alpha)d\alpha = \frac{1}{2}\oint \mu_0 H m_z d\alpha \quad (1)$$

where $m_z$ is the magnetization perpendicular to the rotating field. and $\mu_0$ is the magnetic permeability of vacuum.

These losses can provide information on irreversible magnetization process and therefore the anisotropies in the sample. To understand these loss mechanisms is relevant to numerous applications, for instance in induction motors and transformers, where rotating fields will induce rotational losses which can become dominant under certain conditions. This shows that studying and understanding the origin of these losses, could help improve the magnetic performance of such devices.

Numerical micromagnetism is an optimal candidate for modelling these losses. To understand this on a fundamental level, we started by studying the rotational losses of a Stoner Wolfarth particle. Figure 1 shows the losses $P$ as a function of the magnitude of the rotating field $H$ in units of the anisotropy field $H_a=2Ku/Js$. It is evident that the losses are zero until $H_a/2$, where they reach a maximum, and then decline until at $H_a$ they are zero again. This behaviour is very well understood[1]. Up until $H_a/2$ the particle turns reversibly and therefore there are no losses, which is evident from the Stoner-Wolfarth model. If the field exceeds $H_a$, the magnetization will always point towards the applied field, which again means that the losses vanish.

In order to bring the simulation closer to reality of magnetic steel sheets, we also employ a model in which we simulate a grain-oriented softmagnetic sample with periodic boundary conditions[2]. The entire simulation cell is shown in figure 2. Since the softmagnet usually consists of exchange coupled grains we have chosen to divide the simulation box in equally sized cubic grains. Each grain has different easy axis, which are randomly chosen so that the direction are uniformly distributed on the unit sphere. This work focuses on trying to understand the simplest models, in order to be able to explain the behaviour of the more complex full softmagnetic simulation.

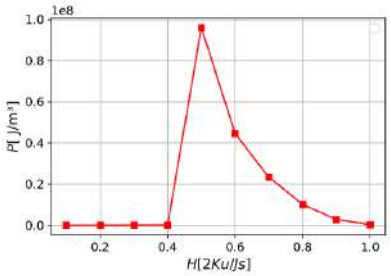



Figure 1: Simulated Losess P(H) for Stoner Wolfarth model

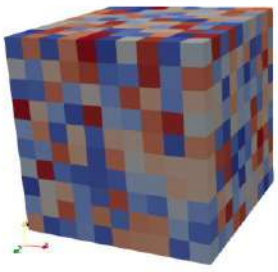


Figure 2: Simulation geometery with grains

## References

[1] Keller, R., & Schmidbauer, E. (1999). Magnetic hysteresis properties and rotational hysteresis losses of synthetic stress-controlled titanomagnetite (Fe2. 4Ti0. 6O4) particles—I. Magnetic hysteresis properties. *Geophysical journal international*, *138*(2), 319-333.

[2] Bruckner, F., Ducevic, A., Heistracher, P., Abert, C., & Suess, D. (2021). Strayfield calculation for micromagnetic simulations using true periodic boundary conditions. *Scientific Reports*, *11*(1), 9202.

# SYMPOSIUM 17.
## NOVEL MAGNETIC TECHNIQUES, INSTRUMENTATION AND METROLOGY.
## S17 INVITED ORAL PRESENTATIONS

# Frequency multiplication and spin wave generation by nonlinear magnetization dynamics.

*Georg Woltersdorf*[*1], Rouven Dreyer[1], Chris Koerner[1]*

[1] *Martin Luther University Halle-Wittenberg, Von Danckelmann Platz 3, Halle, Germany*

*georg.woltersdorf@physik.uni-halle.de

Frequency multiplication and frequency conversion are important processes in modern electronics in which harmonics of the input frequency are generated in nonlinear electronic circuits. Devices based on the propagation and interaction of spin waves represent a promising alternative to conventional electronics. However, the characteristic frequency of spin waves is in the gigahertz range, and thus cannot be easily combined with conventional electronics. We investigate the magnetic excitations in magnetic materials using optical methods and show that excitation at frequencies in the megahertz range can cause magnetic switching processes on the micrometer scale, and these lead to coherent spin wave emission in the gigahertz range [1]. Moreover, at high modulation amplitudes, we demonstrate a new class of nonlinear spin waves that oscillate with half-integer harmonics of the excitation. Imaging of these parametrically generated spin waves enables measurement of the wave vectors and determination of the coherence properties [2]. In addition, we demonstrate the existence of two degenerate phase states, each of which can be selected by the phase position of an external signal source. The frequency multiplication process occurring within the magnetic medium covers more than six octaves and opens new perspectives for applications such as fully magnetic mixers or "on-chip" signal generators. The phase-coherent excitation and control of half-integer harmonic spin waves is highly interesting for applications such as amplifiers and phase-encoded spin-wave-based information processing.

**Acknowledgements**

H2020-EU Project: MANNGA, DFG CRC227 Project: B02

**References**

[1] C. Koerner, R. Dreyer, M. Wagener, N. Liebing, H.G. Bauer, G. Woltersdor, *Science* **375**, (2022) 1165.
[2] R. Dreyer, A. F Schäffer, H. G. Bauer, N. Liebing, J. Berakdar, and G. Woltersdorf, *Nat. Commun.* **13**, (2022) 4939
[3] R. Dreyer, N. Liebing, E.R.J. Edwards, A. Müller, G. Woltersdorf, Phys. Rev. Mat. **5**, (2021) 064411.

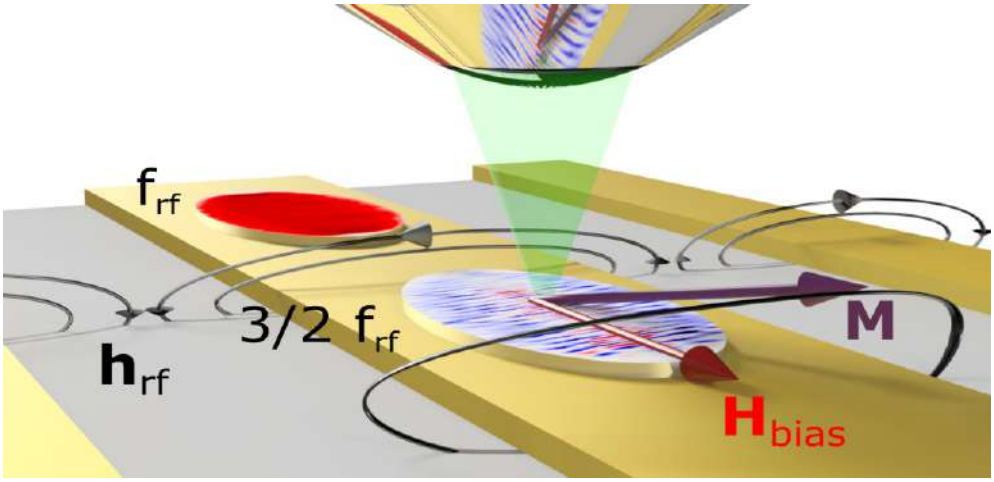



Figure: Magneto-optical measurement of the non-linear excitation of half-integer spin waves in a structure made of NiFe. The excitation is performed by the microwave field of a waveguide structure. Imaging is performed using magneto-optic super Nyquist sampling [3].

# Scanning thermoelectric microscopy of nanoscale soliton systems

*C. Barton[1]*, R. Puttock[1], E. Saugar[2], P. Klapetek[3], A. Fernández Scarioni[4], P. Freitas[5], K Zeissler[1,6], A. Sola[7], V. Basso[7], M. Pasquale[7], H. W. Schumacher[4], T. Ostler[8], O. Chubykalo-Fesenko[2], C.H. Marrows[6] and O. Kazakova1[1]*

[1]National Physical Laboratory, Teddington, United Kingdom.
[2]Instituto de Ciencia de Materiales de Madrid (ICMM-CSIC), Madrid, Spain.
[3]Czech Metrology Institute, Brno, Czech Republic.
[4]Physikalisch-Technische Bundesanstalt, Braunschweig, Germany.
[5]Instituto de Engenharia de Sistemas e Computadores (INESC-MN), Lisbon, Portugal.
[6]School of Physics and Astronomy, University of Leeds, Leeds LS2 9JT, United Kingdom.
[7]Istituto Nazionale di Ricerca Metrologica,10135, Turin, Italy.
[8]Sheffield Hallam University, Sheffield, United Kingdom.
*craig.barton@npl.co.uk

The interplay between thermal gradients and magnetic topological soliton systems represents an exciting frontier in the understanding of novel thermo-magnetic interactions at the nanoscale [1,2]. Thermally driven non-equilibrium spin polarised charge currents and their interaction with the non-trivial topology of skyrmions is central in understanding phenomena such as the topological Nernst effect in chiral magnetic systems within the context of spin caloritronics [3]. In this talk we will address aspects of scanning thermoelectric microscopy (SThEM), Fig 1 (a), where, we will present a novel thermal scanning probe technique that allows us to locally investigate the thermoelectric response from the magnetic domain structure in YIG [4] to nanomagnetic topological solitons: domain-walls and skyrmions. By building up a localised picture of the resulting spatially resolved electric field we can build an understanding of the underlying spin configuration.

We will discuss experimental and modelling results which elucidate to the local response from a pinned domain wall system in an ultra-thin magnetic layer, Fig 1(b). We demonstrate that the domain-wall introduces an additional contribution to the total measured signal, Fig 1(c), which can only arise if we assume a Néel type domain-wall [5]. We will also report on local thermoelectric measurements of skyrmionic systems where we have investigated a set of device structures based on chiral magnetic multilayers, Fig 1(d).

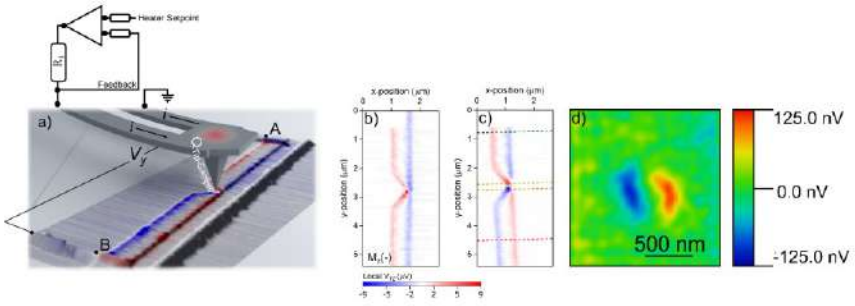



**Fig. 1. (a)** Schematic representation of the sThEM measurement setup used in this work showing the heated AFM probe used to apply the local thermal gradient. The electric field is probed along the device (y) and the integrated response is mapped pixelwise. **(b-c)** sThEM micrographs for saturated and domain wall pinned configurations, respectively. **(d)** sThEM micrograph of a skyrmion nucleated in a chiral magnetic multilayer.

## References

[1] Jakub Zázvorka, et al, Thermal skyrmion diffusion used in a reshuffler device Nature Nanotechnology, 14, 658–661 (2019).
[2] Zidong Wang et al, Thermal generation, manipulation and thermoelectric detection of skyrmions, Nature Electronics, 3, 672–679 (2020).
[3] Alexander Fernández Scarioni, Craig Barton, et al , Thermoelectric Signature of Individual Skyrmions, Phys. Rev. Lett. 126, 077202, (2021).
[4] Alessandro Sola, Craig Barton, et al, Phys. Rev. Applied 14, 034056, (2020).
[5] Puttock, Barton, et al., Sci. Adv. 8, eadc9798 (2022).

# Temperature Imaging Using a Custom Thermal-Magnetic Particle Imaging (T-MPI) System

*C.L. Dennis*, E. Lima Correa, B. dos Santos Correa, F Abel, E. Rus, T Moffat, M.-A. Henn, M. Hu, M. Donahue, A. Biacchi, A. Hight Walker, T. Bui, K. Quelhas, S. Woods, W. Tew*

*National Institute of Standards and Technology (NIST), 100 Bureau Drive, Gaithersburg, MD 20899, USA*

*cindi.dennis@nist.gov

Temperature affects every physical system, every chemical reaction, and every biological process. Although imaging within materials and living things with X-rays and magnetic resonance is now routine, temperature measurement is limited to a 2D surface for infrared imaging, or is an invasive macroscale measurement such as with traditional contact thermometers (e.g., thermocouples). Here, we present our work developing a general, remote, and non-optical method to measure and regulate temperature (through heating) throughout a volume. As with all thermometers, the temperature is measured by a change in a physical property with temperature – specifically, here, the change in the magnetic response of a magnetic nano-object (or collection of magnetic nano-objects [MNOs]) to a change in temperature. This magnetic response is read using the fundamental idea behind magnetic particle imaging: that MNOs respond non-linearly to an applied magnetic field close to zero, and respond linearly at fields near saturation. Therefore, multiple orthogonal magnetic field gradients can be applied to a 3D volume, resulting in only a small region with a magnetic field near zero. The magnetic response of this region is non-linear and easy to determine. However, significant improvements to the signal and thermosensitivity of magnetic nano-objects were necessary for microscale spatial resolution and milli-Kelvin temperature resolution. This was achieved through control of the Curie temperature [1] and exchange coupling between anti-ferromagnetic and ferrimagnetic/ferromagnetic materials [2]; initial results on controlled interactions are also promising. Thermosensitivities of up to 10 were achieved around room temperature, as compared to the platinum resistance thermometer (PRT) value of 1 (see Figure). To achieve traceability, PRTs serve as the transfer artifacts and are traceable to the International Temperature Scale (ITS-90) via NIST reference standard-PRTs. These are individually calibrated and then integrated into a custom hybrid liquid/gas mediated thermostat with a coaxial geometry. The coaxial design achieves the required low uncertainty of ± 10 mK overall with a demonstrated temperature uniformity of ± 5 mK that is immune to changes in the common mode temperature inside the thermostat of at least 100 mK (see Figure). Finally, we will discuss our initial efforts to demonstrate T-MPI in a practical application, specifically polymer 3D printing.

## References

[1] E.D. Rus, E.L. Corrêa, E. Groopman, T. Williamson, H. Hijazi, L. Kasei, C.L. Dennis and T.P. Moffat, *J. Electrochem Soc*., **169** (2022) 092518

[2] F.M. Abel, E.L. Correa, A.J. Biacchi, T.Q. Bui, S.I. Woods, A.R. Hight Walker, and C.L. Dennis, *ACS Applied Materials and Interfaces*, (published 2023). https://doi.org/10.1021/acsami.2c19673

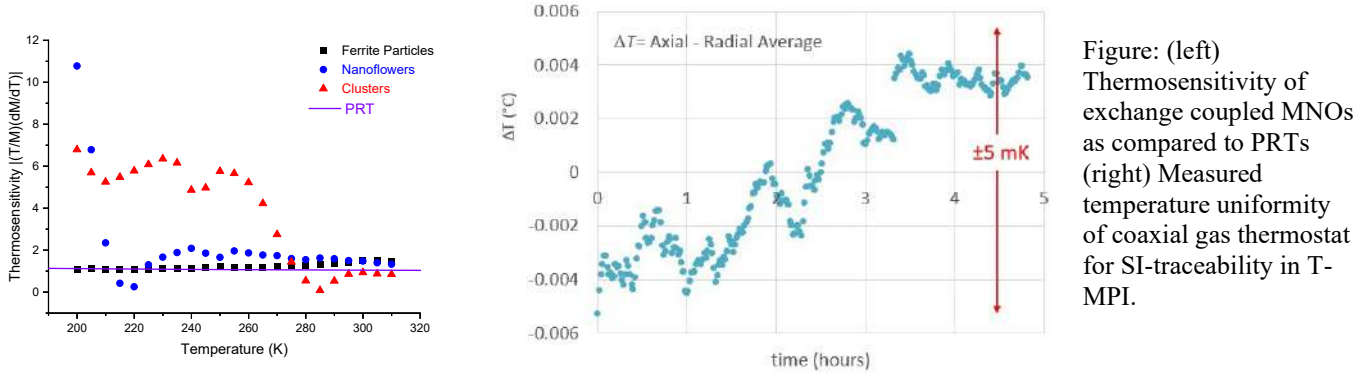



Figure: (left) Thermosensitivity of exchange coupled MNOs as compared to PRTs (right) Measured temperature uniformity of coaxial gas thermostat for SI-traceability in T-MPI.

# SYMPOSIUM 17.
## NOVEL MAGNETIC TECHNIQUES, INSTRUMENTATION AND METROLOGY.
## S17 ORAL PRESENTATIONS

# Periodogram-based detection of unknown frequencies in time-resolved scanning transmission X-ray microscopy

*Simone Finizio[1]*, Joe Bilko Bailey[1,2], Bart Olsthoorn[3],and Jörg Raabe[1]*

[1] *Paul Scherrer Institut, 5232 Villigen PSI, Switzerland*

[2] *Institut de Physique, EPFL, 1015 Lausanne, Switzerland*

[3] *Nordita, KTH Royal Institute of Technology and Stockholm University, SE-106 91 Stockholm, Sweden*

*simone.finizio@psi.ch

Time-resolved X-ray microscopy is a powerful technique utilized for the investigation of dynamical processes in condensed matter systems, thanks to the possibility of combining high spatial (sub 100 nm) and temporal (sub 100 ps) resolutions. Examples of such techniques are time-resolved photoemission electron microscopy, and time-resolved scanning transmission X-ray microscopy (TR-STXM), both of which have been used for the study of a rich ensemble of dynamical processes such as e.g. dynamics of magnetic domain walls, magnonics, switching processes, and the dynamics of topological solitons such as magnetic vortex cores and skyrmions.

TR-STXM is a transmission X-ray microscope where spatially local time-dependent variations in the X-ray absorption cross section of a sample are recorded with high spatial resolution. STXM imaging is made possible by the use of a Fresnel zoneplate to focus the X-ray beam onto a sub-30nm spot on the surface of an X-ray transparent sample, and by recording, in a time-resolved fashion, the intensity of the beam transmitted across the sample. An image is then obtained by scanning the sample by means of a piezoelectric scanner, and performing the measurement of the transmitted beam intensity for each pixel of the image. TR-STXM imaging is performed using a high bandwidth avalanche photodiode (APD), so that it is possible to resolve in time X-ray photons generated by neighboring bunches of the electron beam in the synchrotron light source.

Currently, pump-probe TR-STXM imaging is performed with signals either directly locked to the master clock of the synchrotron light source [1], or by measuring the time difference between the arrival time of an X-ray photon at the APD detector and a marker synchronized with the pump signal used to excite the sample [2]. However, this limits the ensemble of dynamics that can be investigated to those locked to an external excitation signal, excluding processes such as e.g. the parametric excitation of magnons [3], and spin-torque vortex oscillations [4].

In this contribution, we employ the recently-commissioned setup that allows for the measurement of the arrival time of X-ray photons at the APD detector [2] to reconstruct TR-STXM images of periodic dynamical processes oscillating at *a-priori* unknown frequencies, unlocked to any of the available external timing signals [5]. This method is based on the Schuster periodogram [5,6], allowing for the reconstruction of a power spectrum from a sparcely sampled dataset. By performing a set of additional computations with minimal assumptions on the behaviour of the dynamical process, the phase of the excitation can also be reconstructed with reasonable quality [5]. In this work, we present the proof-of-concept measurments performed with this method, and a comparison between the “standard” TR-STXM imaging and the Schuster periodogram approaches, where we observe that the quality of the Schuster periodogram reconstruction is comparable to that achievable by the “standard” method [5].

## Acknowledgements

This work was performed at the PolLux (X07DA) beamline of the Swiss Light Source. The PolLux endstation was financed by the German Bundesministerium für Bildung und Forschung through contracts 05K16WED and 05K19WE2. The authors acknowledge funding from the European Research Council through the ERC HERO-810451 grant.

## References

[1] A. Puzic *et al.*,*Synchrotron Radiation News* **23** (2010), 26
[2] S. Finizio, S. Mayr, and J. Raabe, *J. Syn. Rad.* **27** (2020), 132
[3] S. O. Demokritov *et al.*, *Nature* **443** (2006), 430
[4] N. Locatelli *et al.*, *Nature Materials* **13** (2013), 11
[5] S. Finizio, J. Bailey, B. Olsthoorn, and J. Raabe, *ACS Nano* **16** (2022), 21071
[6] J. T. VanderPlas, *The Astrophysical Journal Supplement Series* **236** (2018), 16

# Correlating the Hall effect with magnetic textures in $Mn_{1.4}PtSn$ by in-situ TEM

*Bernd Rellinghaus[1]*, Darius Pohl[1], Sebastian Schneider[1], Yejin Lee[2], Dominik Kriegner[3,4], Sebastian Beckert[3], Praveen Vir[5], Moritz Winter[1,5], Claudia Felser[5], and Andy Thomas[2,3]*

[1] *Dresden Center for Nanoanalysis (DCN), cfaed, TU Dresden, 01062 Dresden, Germany*
[2] *Leibniz Institute for Solid State and Materials Research, IFW Dresden, 01062 Dresden, Germany*
[3] *Institute of Solid State and Materials Physics, TU Dresden, 01062 Dresden, Germany*
[4] *Institute of Physics, Academy of Sciences of the Czech Republic, 162 00 Praha 6, Czech Republic*
[5] *Max Planck Institute for Chemical Physics of Solids, 01187 Dresden, Germany*
*bernd.rellinghaus@tu-dresden.de

Besides their nanoscopic size, the interest in skyrmions, antiskyrmions, and related topologically protected magnetic textures is based on the potential to motion them by external electrical stimuli and to detect their presence and/or motion electrically utilizing the Hall effect. Accordingly, materials that host such skyrmionic objects are of growing interest due to their potential for data storage and manipulation – and likewise does their Hall signature. Since emerging magnetic fields related to the presence of skyrmions give rise to additional, "topological" contributions to the Hall effect, it is essential to disentangle them from the anomalous Hall effect. This drives the need to correlate Hall effect measurements with the underlying magnetic textures using high-resolution magnetic imaging techniques such as transmission electron microscopy (TEM) based Lorentz-TEM. However, in (anti)skyrmion hosts with a strong susceptibility to dipole-dipole interactions, the stability of skyrmionic phases depends strongly on the demagnetization field and sample geometry. Hence, a correlation of properties determined from samples of the same material, however, with different shapes are problematic.

We have therefore devised a novel measurement setup that allows us to measure the anomalous Hall effect and concurrently acquire high-resolution images of the underlying magnetic textures on the *identical* sample in-situ in a transmission electron microcope using Lorentz-TEM. In proof-of-concept measurements on a thin Ni bar, we have not only provided evidence for the feasibility of the method, but have also highlighted the added value of detailed structural and chemical nanoanalysis for the interpretation of the Hall data [1].

We have then applied this in-situ approach to a lamella of a $Mn_{1.4}PtSn$ single crystal and measured the Hall voltage $U_H$ as function of the magnetic field provided by the objective lens of the microscope (see Fig.). With increasing field, the ground state helical phase develops into a chiral soliton lattice (CSL), thereby enhancing the out-of-plane component of the magnetization and likewise $U_H$. At fields of 300 mT, the CSL domains break up on the expense of saturated areas. This accelerates the saturation process that is then decelerated again by the formation of non-topological bubbles and antiskyrmions that "resist" further increasing fields. Upon reducing the field from saturation, $U_H$ drops exactly where the material re-enters the CSL phase and transforms back to the helical ground state towards zero fields. These findings are supported by model calculations and resonant elastic x-ray scattering experiments. Noteworthy, the observed magnetic textures explain the measured Hall voltage without any need for additional contributions such as a topological Hall effect.

## Acknowledgements

This research is funded by the German Research Foundation (DFG) within priority program SPP 2137 (project ID 403503416). M.W. thanks the Max Planck Society for funding through IMPRS-CPQM.

## References

[1] D. Pohl et al., arXiv:2302.06127 [cond-mat.other]

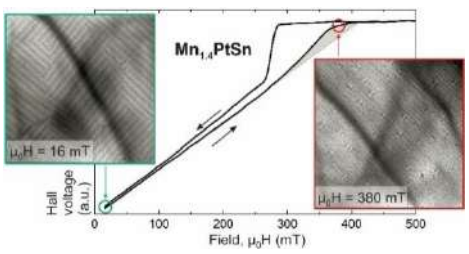



Figure: In-situ measured Hall voltage as function of an external magnetic field provided by the objective lense of the TEM (central plot). Two Lorentz-TEM images acquired simultaneously reveal the fingerprint stripe pattern of the helical phase close to zero field (green frame, left) and the presence of both non-topological bubbles and anti-skyrmions at fields close to saturation (red frame, right).

# Magnetic imaging by X-ray ptychography with a single polarisation

*Marisel Di Pietro Martinez[1,2]*, Alexis Wartelle[1,3,4], Nicolas Mille[5], Stefan Stanescu[5], Rachid Belkhou[5], Vincent Favre-Nicolin[4], Guillaume Beutier[1]**

[1] *Univ. Grenoble Alpes, CNRS, Grenoble INP, SIMaP, Grenoble, France*
[2] *Max-Planck-Institute for Chemical Physics of Solids, Dresden, Germany*
[3] *Institut Jean Lamour, Université de Lorraine, Nancy, France*
[4] *ESRF - The European Synchrotron, Grenoble, France*
[5] *Synchrotron Soleil, Saint-Aubin, France*
*guillaume.beutier@grenoble-inp.fr

Polarised X-rays are a fantastic probe of magnetic textures in ferromagnetic materials [1]. The usual contrast mechanism is X-ray Magnetic Circular Dichroism [1] (or X-ray Magnetic Circular Birefringence [2]), which is obtained as the difference between two measurements performed with circularly polarised X-rays of opposite helicity. The two measurements are used to separate the magnetic contrast from the charge contrast.

Here we demonstrate a method based on X-ray ptychography [3] which allows simultaneous imaging of the charge and magnetism with a single polarisation (linear polarisation). Ptychography is a high-resolution imaging technique that combines information from direct space and reciprocal space [3]. As it is based on redundancy of information, it can provide more information than a single image of the sample [4]. Here we use this property to obtain simultaneously two images of the sample [4]: the charge density and a projection of the magnetic texture. Several samples with magnetic domains were tested and the results compared with images obtained more classically using X-ray Magnetic Circular Dichroism.*

This approach could be particularly interesting when circular polarisation is not available or when circular dichroism is not sufficient to disentangle charge and magnetism, for instance at large scattering angles, and could thus be applied to antiferromagnetic textures in Bragg geometry.

**Acknowledgements**

We ackowledge Soleil for providing synchrotron beamtime on beamline HERMES.

**References**

[1] P. Fischer, J. Phys. D: Appl. Phys. **50**, 313002 (2017).
[2] M. Di Pietro Martinez *et al*, Phys. Rev. B (accepted), arXiv:2212.10183 (2023).
[3] P. Thibault *et al*, Science **321**, 379 (2008).
[4] P. Thibault & A. Menzel, Nature **494**, 68–71 (2013).

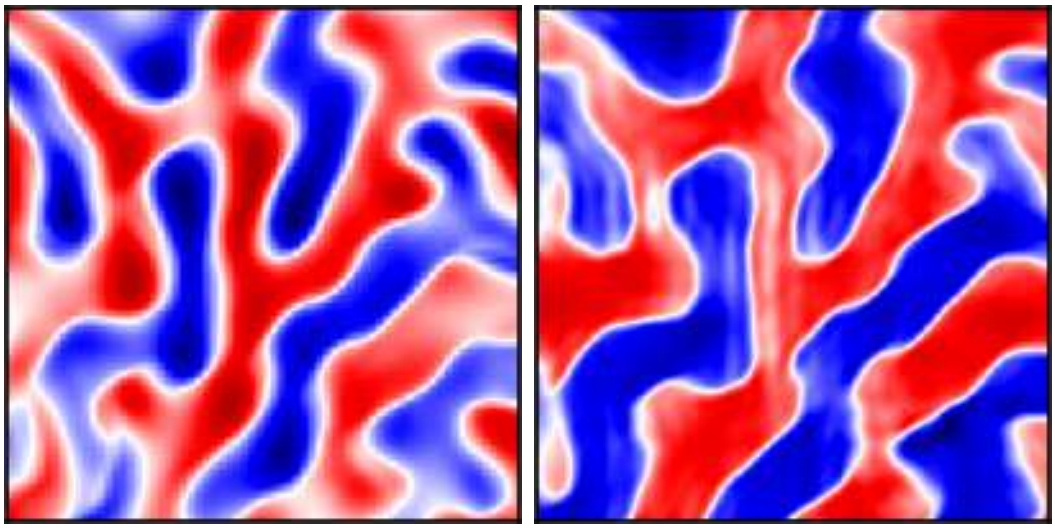


Figure: Magnetic domains in a Co/Pt multilayer imaged by ptychography with linear polarisation (left) and circular dichroism (right). The field of view is 1x1 μm².

# Vector Magnetic Imaging of 3D Objects Using Fourier Transform Holography

*Marisel Di Pietro Martínez[1], Alexis Wartelle[2,3]*, Carlos Herrero Martínez[4], Farid Fettar[5], Florent Blondelle[5], Jean-François Motte[5], Claire Donnelly[1], Luke Turnbull[6], Feodor Ogrin[7], Gerrit van der Laan[8], Horia Popescu[9], Nicolas Jaouen[9], Flora Yakhou-Harris[3], Guillaume Beutier[4]*

[1] *Max Planck Institute for Chemical Physics of Solids, Noethnitzer Str. 40, 01187 Dresden, Germany*
[2] *Institut Jean Lamour, UMR CNRS 7198, Université de Lorraine, 54506 Vandoeuvre, France*
[3] *ESRF, The European Synchrotron, 71 Avenue des Martyrs, CS40220, 38043 Grenoble Cedex 9, France*
[4] *Université Grenoble Alpes, CNRS, Grenoble INP, SIMaP, 38000 Grenoble, France*
[5] *Université Grenoble Alpes, CNRS, Grenoble INP, Institut Néel, 38042 Grenoble, France*
[6] *Department of Physics, Durham University, Durham, DH1 3LE, UK*
[7] *School of Physics and Engineering, University of Exeter, Stocker Road, Exeter EX4 4QL, UK*
[8] *Diamond Light Source, Harwell Science and Innovation Campus, Didcot OX11 0DE, UK*
[9] *Synchrotron SOLEIL, Saint Aubin, BP 48, 91192 Gif-sur-Yvette, France*
*alexis.wartelle@esrf.fr

Advances in three-dimensional (3D) fabrication techniques have highlighted the need for techniques capable of probing ferromagnetic samples with novel geometries while retaining high spatial resolution. The complexity of configurations available to magnetization as vector field in 3D objects [1] furthermore calls for vector tomographic approaches in order to recover the sample's complete magnetic state. Several X-ray-based techniques employing X-ray resonant magnetic scattering [3] combine both aspects [2], which grants them remarkable efficiency in 3D experimental studies.

Here, we present an extension of Fourier Transform Holography [4] where dual-axis tomography allows us to reconstruct the complete domain pattern of a 800 nm thick Fe/Gd multilayer [5]. Our sample design, shown in Fig.1 a), uses a pair of orthogonal reference slits in a HERALDO approach [6]. The field of view is defined by a 5 µm wide opening milled into an opaque Ti\Au multilayer. The slit length allows them to function under tilt, which grants sensitivity to two components of magnetization. Within the object window, the reconstructed domains match very well the patterns observed with Magnetic Force Microscopy (MFM), illustrated in Fig. 1b). However, our approach also retrieves finer details from the 3D magnetic configuration, as can be seen in Fig. 1c). All components of magnetization are recovered with a resolution below 80 nm [5].

**Acknowledgements**
We acknowledge SOLEIL and the ESRF for providing synchrotron beamtime. We acknowledge the Agence Nationale de la Recherche, CNRS, the Max Planck Society, the Marie Skłodowska-Curie Actions, and NATO for funding. The authors acknowledge Laurent Cagnon (Institut Néel) for the EDX measurements.

**References**

[1] C. Donnelly *et al. Nat. Phys.* **17** (2021) 316.
[2] C. Donnelly, V. Scagnoli, *J. Phys.: Condens. Matter* **32** (2020) 213001.
[3] J. Stöhr, H. C. Siegmann, ``Magnetism: From Fundamentals to Nanoscale Dynamics'' (2006), Springer Berlin Heidelberg.
[4] A. Scherz *et al.*, *Phys. Rev. B* **76** (2007) 214410.
[5] M. Di Pietro Martínez *et al., Phys. Rev. B* (2023) in press; arXiv preprint 2212.10183.
[6] M. Guizar-Sicairos, J. R. Fienup, *Optics Express* **15** (2007) 17592.

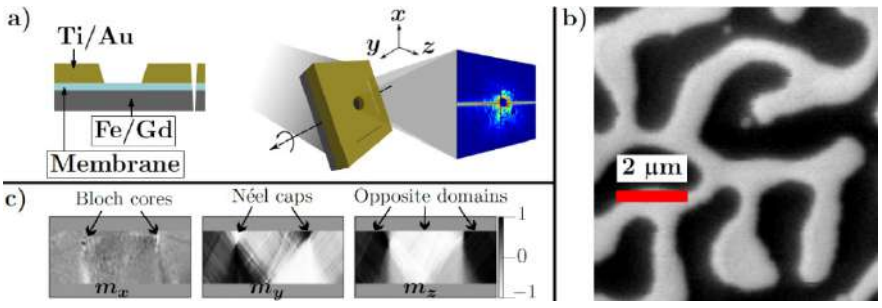



Figure 1: a) Schematic representation of the sample design in cross-section view (left) and during experiment, at a tilt (right) such that only one slit diffracts; see scattering pattern at the right. b) MFM image of the investigated Fe/Gd multilayer, revealing the imaged worm domains. c) Slices (2.4 µm in length) through our vector reconstruction, showing the three components of magnetization. Fine features from the sample volume (not accessible with MFM) are revealed.

# Time-Resolved Soft X-ray Ptychographic Imaging of Spinwaves

*Tim A. Butcher[1], Nicholas W. Phillips[1], Manuel Langer[1], Sina Mayr[1,2], Sebastian Wintz[3], Simone Finizio[1], and Jörg Raabe[1]*

[1] *Paul Scherrer Institut, 5232 Villigen PSI, Switzerland*

[2] *Laboratory for Mesoscopic Systems, Department of Materials, ETH Zurich, 8093 Zurich, Switzerland*

[3] *Max-Planck-Institut für Intelligente Systeme, 70569 Stuttgart, Germany*

*tim.butcher@psi.ch

Time-resolved scanning transmission X-ray microscopy (TR-STXM) is a powerful technique for the investigation of a plethora of dynamical processes in condensed matter, including the dynamics of topological magnetic solitons such as vortex cores and skyrmions, the dynamics of magnetic domains and domain walls, magnetisation switching processes and magnonics. However, it is limited by the achievable spatial resolution, which is ultimately determined by the lithographical fabrication of the X-ray Fresnel zoneplate used to focus the X-ray beam onto the sample [1].

X-ray ptychography is an imaging technique, which exploits the advantages of both STXM and coherent diffractive imaging, resulting in both amplitude and phase information as well as the possibility to achieve spatial resolutions limited by the wavelength of the probing X-rays and independently from the X-ray optical elements used to shape the beam. This overcomes the main limiting factor of STXM imaging regarding the spatial resolution of the images [2].

X-ray ptychographic imaging is performed by acquiring diffraction patterns from a sample illuminated by a coherent X-ray beam at different points, where the distance between neighbouring points is chosen to guarantee an overlap between the illumination spots. This provides sufficient over-determination of information to allow an iterative algorithm to reconstruct a complex image of the object [2]. For TR imaging, the main limitation of X-ray ptychographic imaging is given by the 2D X-ray detector with which the 2D diffraction patterns are acquired. Due to the lower bandwidth of 2D X-ray detectors if compared to the point detectors used in TR-STXM, it is impossible to resolve X-ray photons generated by neighbouring bunches in time, which prohibits the use of the specialised pump-probe protocol used in TR-STXM [3,4]. Nonetheless, as a synchrotron light source is an intrinsically pulsed photon source with a repetition rate of 500 MHz (at the Swiss Light Source), TR imaging at frequencies locked to the repetition rate or one of its harmonics is possible through the “standard” pump-probe protocol [5].

Here we present TR ptychographic imaging of the spinwave emission and propagation in a magnetic vortex state stabilised in a ferromagnetic microstructured element excited by means of an oscillating magnetic field generated by a microantenna patterned on top of the ferromagnetic microstructure. Different excitation frequencies are investigated, allowing us to present a first proof-of-concept of TR ptychography in the soft X-ray energy range utilising strong X-ray magnetic circular dichroism (XMCD) from the transition metal L-edge.

## References

[1] B. Rösner *et al.*, *Optica* **7** (2020), 1602
[2] F. Pfeiffer, *Nat. Photonics* **12** (2018), 9
[3] S. Finizio, S. Mayr, and J. Raabe, *J. Synchrotron Radiat.* **27** (2020), 132
[4] A. Puzic *et al.*, *Synchrotron Radiat. News* **23** (2010), 26
[5] C. Donnelly *et al.*, *Nat. Nanotechnol.* **15** (2020), 356

# Imaging hidden magnetic domains in Fourier space

*P. Mazalski[1*], S. Jarausch[2], M. Vogel[1], W. Dobrogowski[1], A. Wawro[3], A. Maziewski[1], J. McCord[2]*

[1] *Faculty of Physics, University of Bialystok, Bialystok, Poland*
[2] *Institute for Materials Science, Kiel University, Kiel, Germany*
[3] *Institute of Physics, Polish Academy of Sciences, Warsaw, Poland*
*piotrmaz@uwb.edu.pl

The analysis of magnetic domain structures (DS) in combination with applied micromagnetics provides a unique opportunity to extract magnetic properties from DS analysis. Magnetooptical (MO) microscopy [1] is one of the most prominent techniques for the visualization of DS within magnetic materials. Magnetooptics offers the ability to probe the dynamic magnetization response using an illumination source, providing flexible temporal resolution down to the femtosecond time scale. Temporal resolution for imaging magnetization dynamics using stroboscopic imaging techniques is standard in MO microscopy. Yet, fast single-shot imaging is limited by the brilliance of the illumination source with nanosecond time-resolution being possible with lab-based magnetooptical microscopy setups [2]. We extend the imaging modes to fast single-shot imaging below the noise limit to proof the existence and to characterize "invisible" DS in sub-nm thick magnetic films.

Fluctuating DS have been reported in ultrathin magnetic films (Pt/Co/Os/Pt [3], Pt/CoFeB/Ru/Pt [4]). Here, we study fluctuating DS in W/Co wedge ($0 < d_{Co} < 2$ nm)/Pt layers deposited on sapphire. With decreasing Co thickness ($d_{Co}$), a spin reorientation transition from in-plane to out-of-plane occurs at $d_{Co} = 1.52$ nm. With smaller $d_{Co}$ the spontaneous magnetization reduces and at $d_{Co} \leq 0.58$ nm, a transition to the (super)-paramagnetic state takes place. Due to the effective reduction of the magnetic energies, thermal (energy) effects then come into play.

In this context, we focus on the analysis and the demonstration of existence of rapidly fluctuating out-of-plane DS near the critical thickness. For the study of the fluctuating DS, single-shot imaging down to the microsecond time scale is used.

Figure 1(a) presents imaging data spanning only 2.9 pm of effective $d_{Co}$ thickness, where a strong decrease of DS visibility with decreasing $d_{Co}$ is observed. To extract the DS parameters, we have used local image thresholding enhancement, which is based on random forest type machine learning methods and a set of features developed for the obtained data. For the thinner $d_{Co}$, where the spontaneous magnetization drops, we proof the existence of the DS by single-shot imaging using flexible pulsed LED and diode laser sources with exposure times down to 3 µs. To overcome the brilliance limitations of single-shot imaging on fast timescales, multiple apparently noisy images were transformed and averaged in Fourier space, allowing the determination of the domain period dependence on thickness. DS characteristics are extracted directly from the apparent featureless images (see Fig. 1(b)) proofing the existence of the hidden DS.

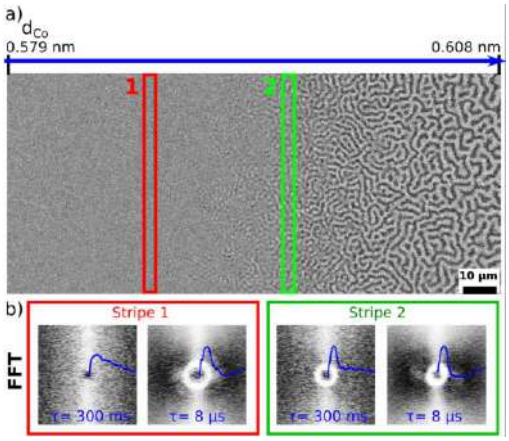



Figure 1 (a) DS of a Co wedge taken at an exposure time of $\tau = 40$ ms. With decreasing $d_{Co}$ the MO contrast of the DS. (b) Exemplary series of averaged Fast Fourier Transformed (FFT) single-shot images taken from two positions for exposure times of $\tau = 300$ ms and $\tau = 8$ µs. The contrast was enhanced by subtraction of the previously fitted 1/f and broadband noise. DS characteristics such as the domain period of the "invisible" DS become apparent.

## Acknowledgements

This work was supported by the Polish NSC 2020/37/B/ST5/02299 and by the German DFG Mc9/19-1.

## References

[1] J. McCord, *J. Phys. D: Appl. Phys.* **48** (2015) 333001.
[2] A. Kimmel, *et al.*, *J. Phys. D: Appl. Phys.* **55** (2022) 463003.
[3] R. Mansell, *et.al.*, *Phys. Rev. B* ***106*** (2022) 054413.
[4] R. Tolley, S.A. Montoya, E.E. Fullerton, *Phys. Rev. Mat.* **2** (2018) 044404.

# Direct Imaging of Nanoscale Domain Wall Oscillations in Landau Structures

*Balram Singh[1], Rachappa Ravichankar[1], Jorge A. Otálora[2], Ivan Soldatov[1], Rudolf Schäfer[1], Daniil Karnaushenko[3], Volker Neu[1*], Oliver G. Schmidt[3]*

[1] *Leibniz IFW Dresden, 01069 Dresden, Germany*

[2] *Departamento de Fisica, Universidad Católica del Norte, Casilla 1280, Antofagasta, Chile*

[3] *Research Center for Materials, Architectures and Integration of Nanomembranes (MAIN), Chemnitz University of Technology, 09126 Chemnitz, Germany*

*v.neu@ifw-dresden.de

AC-excited domain wall processes are the heart of modern domain wall (DW) based microscale devices such as giant-magneto impedance sensors [1] and magnetophoretic circuits [2]. However, it remains a major challenge for magnetic microscopies to follow the DW trajectory and amplitude while it is in motion.

Here we report an imaging approach to investigate DW dynamics with nanoscale spatial resolution [3] employing conventional magnetic force microscopy (MFM) -- a powerful technique that is, however, largely unexplored concerning imaging of time-dependent domain processes. The method is based on a quantitative assessment of the time-averaged MFM phase shift signal and its description by the locally varying dwell time function of an oscillating DW. With this technique, we quantify the oscillations of a 180° Nèel domain wall in a patterned permalloy rectangle as a function of external magnetic field strength, frequency, magnetic structure size, thickness, and strain-induced anisotropy – down to a resolution of 60 nm. We understand the behaviour of the excited DW behavior as a forced damped harmonic oscillator with restoring force being influenced by the geometry, thickness, and anisotropy of the $Ni_{81}Fe_{19}$ structure.

This approach offers new possibilities for the analysis of DW motion at elevated frequencies and at a spatial resolution of well below 100 nm in various branches of nanomagnetism.

## References

[1] D. Karnaushenko *et al.* Self-Assembled On-Chip-Integrated Giant Magneto-Impedance Sensorics. *Adv. Mater.* **27** (2015) 6582.
[2] A. Sarella *et al*. Two-dimensional programmable manipulation of magnetic particles on-chip. *Adv. Mater.* **26** (2014) 2384.
[3] B. Singh *et al.* Direct imaging of nanoscale field-driven domain wall oscillations in Landau structures. *Nanoscale* **14** (2023) 13667.

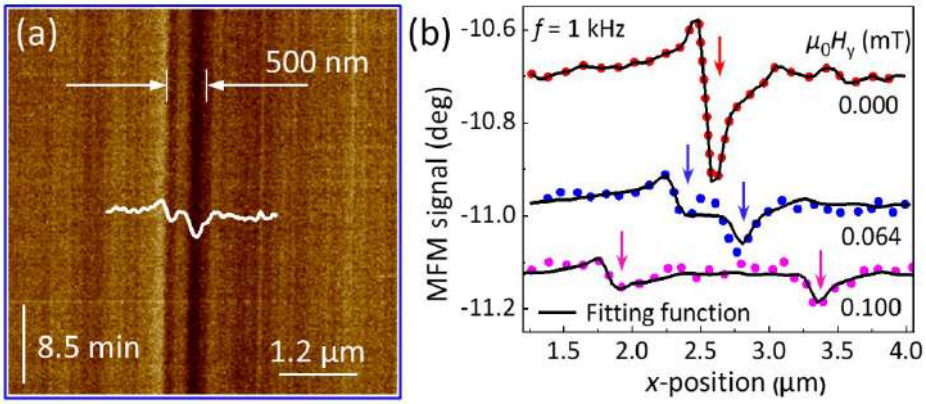



Figure: (a) Time-averaged imaging of the DW oscillating with a frequency of 1 kHz, reveiling a peak-to-peak amplitude of 500 nm. (b) Measured intensity profiles of a static DW and the time-averaged DW oscillating at 1 kHz (dotted lines). The convolution (solid lines) of the MFM tip – DW interaction with the dwell-time function explains well the experimentally measured profiles.

# Simultaneous Magnetic Field and Field Gradient Mapping of Hexagonal MnNiGa by Quantitative Magnetic Force Microscopy

*Norbert H. Freitag[1], Christopher F. Reiche [2], Volker Neu[1], Parul Devi[3], Ulrich Burkhardt[3], Claudia Felser[3], Daniel Wolf[1], Axel Lubk[1], Bernd Büchner[1], and Thomas Mühl[1]**

[1] *Leibniz Institute for Solid State and Materials Research IFW Dresden, 01069 Dresden, Germany*

[2] *Department of Electrical and Computer Engineering, University of Utah, Salt Lake City, UT-84112, USA*

[3] *Max Planck Institute for Chemical Physics of Solids Dresden, 01187 Dresden, Germany*

*t.muehl@ifw-dresden.de

Magnetic force microscopy (MFM) is a scanning microscopy technique that is commonly employed to probe the sample's magneto-static stray fields via their interaction with a magnetic probe tip. In this work, a quantitative, single-pass MFM technique is presented that maps one magnetic stray-field component and its spatial derivative at the same time [1]. This technique uses a special cantilever design and a special high-aspect-ratio magnetic interaction tip that approximates a monopole-like moment. Experimental details, such as the control scheme, the sensor design, which enables simultaneous force and force gradient measurements, as well as the potential and limits of the monopole description of the tip moment are discussed.

To demonstrate the merit of this technique for studying complex magnetic samples it is applied to the examination of polycrystalline MnNiGa bulk samples. In these experiments, the focus lies on mapping and analyzing the stray-field distribution of individual bubble-like magnetization patterns in a centrosymmetric [001] MnNiGa phase. The experimental data is compared to calculated and simulated stray field distributions of 3D magnetization textures, and, furthermore, bubble dimensions including diameters are evaluated. The results indicate that the magnetic bubbles have a significant spatial extent in depth and a buried bubble top base.

## Acknowledgements

This project was supported by the Deutsche Forschungsgemeinschaft (DFG) (Grant No. MU 1794/13-2) and the European Research Council (ERC) under the Horizon 2020 research and innovation program of the European Union (grant agreement no. 715620).

## References

[1] N. H. Freitag, C. F. Reiche, V. Neu, P. Devi, U. Burkhardt, C. Felser, D. Wolf, A. Lubk, B. Büchner, and T. Mühl, *Commun. Phys.* **6**, 11 (2023).

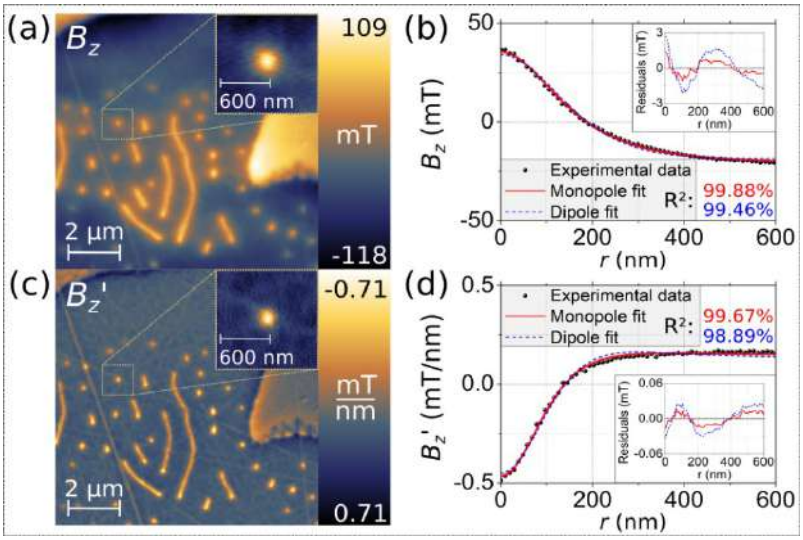



Figure: (a) $B_z$ map of an [001] oriented MnNiGa crystallite at a perpendicular field of −270 mT. In the inset, a magnified image of an individual bubble is shown. (b) An angularly averaged $B_z$ profile for the same bubble is shown together with point monopole and dipole fits. Residues are provided in the inset. The monopole model fits better. (c) and (d) present the same again but now based on $B_z'$ data.

# Novel Methodology to Determine the Magnetic Anisotropy of Magnetic Nanoparticles in Colloidal Suspensions

*David Cabrera[1,2], Takashi Yoshida[3], Mohamed M. Shams[1], Gorka Salas[1,4], Alberto Bollero[1], María del Puerto Morales[5], Julio Camarero[1,6] and Francisco J. Teran[1,4]*

[1] *iMdea Nanociencia, Campus Universitaria de Cantoblanco, 28049 Madrid. Spain.*
[2] *School of Pharmacy and Bioengineering, Keele University, Guy Hilton Research Centre, Thurnburrow Drive, ST4 7QB, Stoke on Trent, UK.*
[3] *Dpt. of Electrical Engineering, Kyushu University, Fukuoka 819-0385, Japan.*
[4] *Nanobiotecnología (iMdea Nanociencia), Unidad Asociada al CNB-CSIC, 28049 Madrid, Spain*
[5] *Instituto de Ciencia de Materiales de Madrid-CSIC, Cantoblanco, 28049 Madrid, Spain*
[6] *Dpto Física de la Materia Condensada, Universidad Autónoma de Madrid, 28049 Madrid, Spain.*
* francisco.teran@imdea.org

The potential of magnetic nanoparticles for acting as active agents in catalysis, magnetic particle imaging or magnetic hyperthermia grounds on their superparamagnetic behaviour under alternating magnetic fields (AMF). In spite of the application potential of this magnetic phenomenon, the identification of fingerprints specifically related to the transition from unblocked to blocked magnetic states at room temperature under AMF still remains a challenge. Here, we report an experimental and theoretical study to determine the effective magnetic anisotropy from iron oxide nanoparticles (IONPs) in colloidal suspensions at room temperature [1]. The experimental methodology is based on magneto-optical measurements of IONP suspensions based on Faraday effect under alternating magnetic fields in a six decades frequency range from hundreds of mHz to kHz with field intensities up to 40 kA/m. Our measurements demonstrate a room temperature transition from unblocked to blocked magnetic states in magnetic suspensions under alternating magnetic fields. The transition is characterized by AC anhysteretic (unblocked state) magnetization cycles at low frequencies and AC hysteretic (magnetically blocked state) magnetization cycles beyond an onset frequency ($f_{onset}$) value which depends on nanocrystal size (see Figure). Thus, $f_{onset}$ values vary from 13 kHz for 12 nm IONPs to 30 Hz for 22 nm IONPs. Our experimental observations are predicted by a theoretical model based on a modified Landau-Liftshitz-Gilbert equation that explains the experimental results in terms of the magnitude of the effective magnetic anisotropy barrier ($\Delta E_{eff} = K_{eff} \cdot V$). Thus, an empirical expression is proposed to determine the effective magnetic anisotropy ($K_{eff}$) from $f_{onset}$. Narrow IONP size distribution, and the negligible contribution of Brownian mechanism to relaxation process benefit the good agreement in the non-Linear Response Regime between the $K_{eff}$ values obtained from our model and those obtained by Blocking temperature via Zero Field Cooling/Field Cooling measurements. Our results provide alternative methodologies to determine experimental parameters. At the same time, numerical simulations significantly improve the understanding and the description of superparamagnetic behaviour in magnetic suspensions.

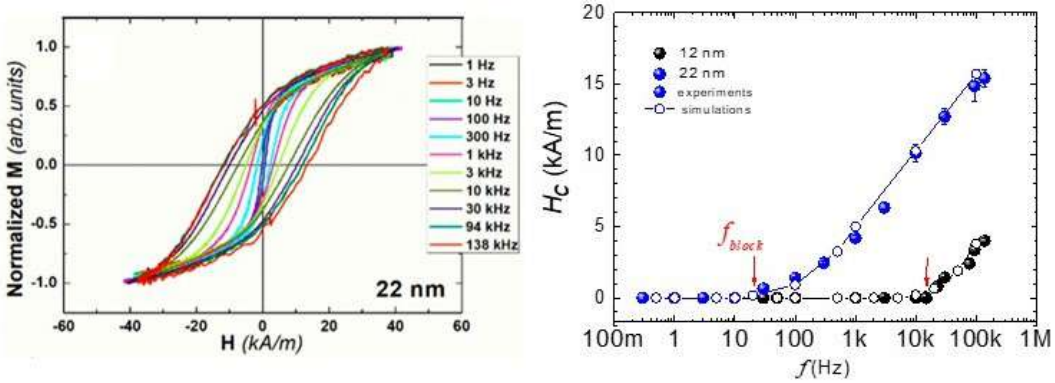



**Figure:** Left) Frequency dependence of AC magnetization cycles of 22 nm IONP; Right) Experimental (filled dots) and theoretical (empty dots) frequency dependence of coercivity obtained for 12nm (black dots) and 22 nm (blue dots) IONP sizes ($H$= 40 kA/m). $f_{onset}$ values are highlighted with red arrows.

## References

[1] D. Cabrera et al., *Nanoscale Nanoscale.***14** (2022) 8789.

# X-ray nanothermometry to monitor the local temperature of nanoparticles under hyperthermia conditions

*R. López-Méndez[1], G. R. Castro[2,3], J. Reguera[4], C. Castellano[5], J. Camarero[1,6], M. A. García[7], C. Wilhelm[8], A. Muñoz-Noval[9], A. Espinosa[3,1*]*

[1] *IMDEA Nanociencia, c/ Faraday, 9, 28049 Madrid, Spain*
[2] *Spanish CRG beamline at the European Synchrotron (ESRF), B.P. 220, F-38043 Grenoble, France*
[3] *Instituto de Ciencia de Materiales de Madrid, ICMM-CSIC, 28049 Madrid, Spain*
[4] *BCMaterials, Basque Center Centre for Materials, Applications and Nanostructures, UPV/EHU Science Park, 48940 Leioa, Spain*
[5] *Dipartimento di Chimica, Università degli Studi di Milano, via Golgi 19, 20133 Milano, Italy*
[6] *Departamento de Física de la Materia Condensada and Instituto 'Nicolás Cabrera', Universidad Autónoma de Madrid, 28049 Madrid, Spain*
[7] *Departamento de Electrocerámica, Instituto de Cerámica y Vidrio, ICV-CSIC, Kelsen 5, 28049 Madrid, Spain*
[8] *Laboratoire Matière et Systèmes, Complexes MSC, UMR 7057, CNRS & University Paris Diderot, 75205, Paris Cedex 13, France*
[9] *Dpto. Física Materiales, Facultad CC. Físicas, Universidad Complutense de Madrid, 28040 Madrid, Spain*
*ana.espinosa@csic.es

The influence of temperature on biological events, metabolic rates, and homeostatic processes in cells is significant, making it a crucial parameter to consider. Non-invasive thermal nanotherapies utilize magnetic and photo-thermal nanomaterials to generate localized heat, which effectively ablates tumors and contributes to the fight against cancer. These nanomaterials serve as efficient therapeutic agents. To achieve effective therapeutic outcomes while preserving healthy tissue, optimal heating efficiency in the intratumoral environment is crucial for thermal nanotherapies. This involves accounting for cellular confinement effects that impact the final heat-generating performance. [1,2]. Interestingly, therapeutic effects can be achieved through localized nanoparticle heating, even without any noticeable macroscopic increase in temperature. This study explores the heating efficiency of nanoparticles under the influence of magneto- and photo-thermal effects in conjunction with other therapeutic methods. [3,4]. We also present the use of the extended X-ray absorption fine structure (EXAFS) analysis as a direct and *in situ* nanothermometric probe to determine the local temperature at the core of nanoparticles upon laser photo-excitation, revealing significant nanothermal gradients [5].

**Acknowledgements**

This work was supported by the Comunidad de Madrid (2018-T1/IND-10360 Talento, 2018-T1/IND-1005 Talento, 2018/NMT-4321), AECC project Ideas Semilla 2019, Spanish Ministry of Economy and Competitiveness (FPI SEV-2016-0686-20-1, SEV-2016-0686, RYC2020-029282-I, PID2021-127033OB-C21, PID2021-126323OA-I00, RED2018-102626-T).

**References**

[1] E. Cazares-Cortes, A. Espinosa, J.-M. Guigner, A. Michel, N. Griffete, C. Wilhelm, and C. Ménager, *ACS Appl. Mater. Inter*. **9**, (2017) 25775.
[2] J. G. Ovejero *et al.*, *Nano Letters* **21** (2021) 7213.
[3] A. Espinosa *et al.*, *Adv. Funct. Mater.* **28** (2018) 1803660.
[4] A. Espinosa, J. Reguera, A. Curcio, Á. Muñoz-Noval, C. Kuttner, A. Van de Walle, L. M. Liz-Marzán, and C. Wilhelm, *Small* **16** (2020) 1904960.
[5] A. Espinosa, G. R. Castro, J. Reguera, C. Castellano, J. Castillo, J. Camarero, C. Wilhelm, M. A. García, and A. l. Muñoz-Noval, *Nano Letters* **21** (2021) 769.

# Evaluation of the heating capacity at the nanoscale

*Jorge Marqués-Marchán[1], Miriam Jaafar[2], Alvaro Gallo-Cordova[1], M. del Puerto Morales[1] and Agustina Asenjo[1]**

[1] *Instituto de Ciencia de Materiales de Madrid, CSIC, 28049 Madrid (Spain)*
[2]*Dpto. Física de la Materia Condensada and IFIMAC, UAM, 28049 Madrid (Spain)*
* aasenjo@icmm.csic.es

Among the numerous applications of the magnetic nanoparticles for in-vivo biomedical therapies, the magnetic particle hyperthermia is one of the most active research area. A key question is the relationship between the morphology of the nanoparticle and the heating capacity. Moreover, to achieve the evaluation of this properties at the nanoscale is still an onging topic.

Measuring dissipation in scanning force microscopy relies on extracting physical information from either tip or sample by recording variations in the cantilever oscillation [1]. Non-conservative interactions between the tip and the sample give rise to a decrease in the oscillation amplitude of the lever, as some energy is transferred in every oscillation cycle [2]. In the case of magnetic materials, due to the hysteretic processes at the tip apex (or in the sample).

In this work, we analyse the information extracted from the dissipation in Magnetic Force Microscopy (MFM) in different systems. The dissipative maps in Magnetic Dissipation Force Microscopy (MDFM) have been used to distinguish between Néel and Bloch domain walls [3], to detect magnetic nanoparticles [4] or to obtain 2D maps of the sample stray field [5]. In addition, the MDFM mode allows to improve the lateral resolution, to obtain unexpected negative gradients of dissipation [6] or to distinguish artefacts [7].

The MFM arises a unique technique not only to obtain information about the magnetic configuration of different nanostructures, but also to characterize biomagnetic materials of great interest in biomedicine [8]. One open question, the evaluation of the heating capacity of the magnetic nanoparticles used in hyperthermia treatment is addressed in this work. We have used MDFM mode to evaluate the dissipation of energy at the nanoscale in flowers-like magnetic nanoparticles [9]. The flower-like nanoparticles present a magnetic behaviour close to superparamagnetic state at room temperature. Thus, the magnetic moment can be reoriented due to the stray field of the MFM tip.

**References:**

[1] W. Denk and D. W. Pohl, Appl. Phys. Lett. 59 (1991) 2171
[2] N. F. Martínez and R. García, Nanotechnology 17 (2006) S167–S172
[3] P. Grütter et al., Appl. Phys. Lett. 71 (1997) 279-281
[4] B. Torre et al., Scientific Reports, 1(1), 1–8(2011)
[5] Ó. Iglesias-Freire et al., Appl. Phys. Lett., 102, 022417 (2013)
[6] M. Jaafar, …, J. J. Sáenz and A. Asenjo, Nanoscale, 8(38), 16989–16994(2016).
[7] M. Jaafar and A. Asenjo, Appl. Sci., 11(22), 10507(2021)
[8] C. Moya et al., Nanoscale, 7, 8110 (2015)
[9] H. Gavilán et al., Part. Syst. Charact. 34 (2017)

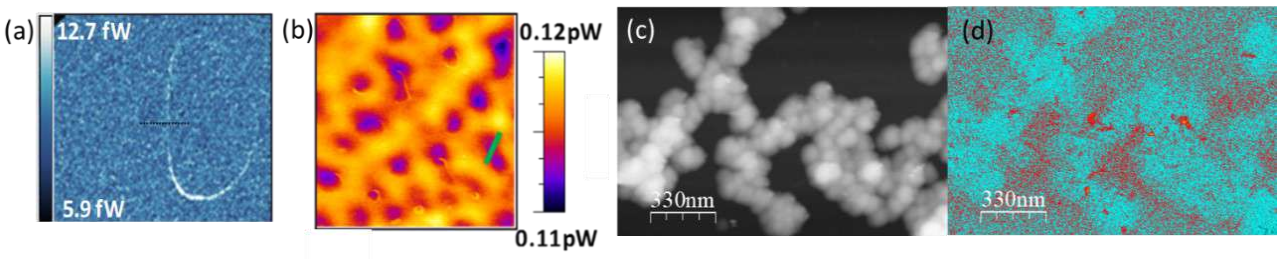



Figure: Dissipation images corresponding to (a) Saturated Co disc [6], (b) CoPt multilayer [7] and (d) flower-like nanoparticles. (c) is the corresponding topographic image of the flower-like nanoparticles

# Dynamic susceptibililty compact apparatus to measure the concentration of carbon nanotubes

*Myriam Pannetier Lecoeur[1*], Claude Fermon[1], Aurelie Solignac[1], Léa Boutet[1], Jonathan Bouillonnec[2], Mathieu Pinault[2], Martine Mayne[2].*
[1] *SPEC, CEA, CNRS, Université Paris-Saclay, CEA Saclay 91191 Gif-sur-Yvette Cedex, France*
[2] *Université Paris-Saclay, CEA, CNRS, NIMBE,CEA Saclay 91191 Gif-sur-Yvette Cedex, France*
*myriam.pannetier-lecoeur@cea.fr

Carbon nanotubes (CNTs) are widely used for their mechanical, electrical properties, in particular for application in aeronautics, construction…Usually, small amount (few percents) is dispersed within a matrix and brings the targeted properties. Therefore, mass concentration is an important information, as well as inhomogeneity. To access their content and dispersion, one may use Scanning Electron Microscopy (SEM), but it gives access to local information and requires to extract some small sizes samples.

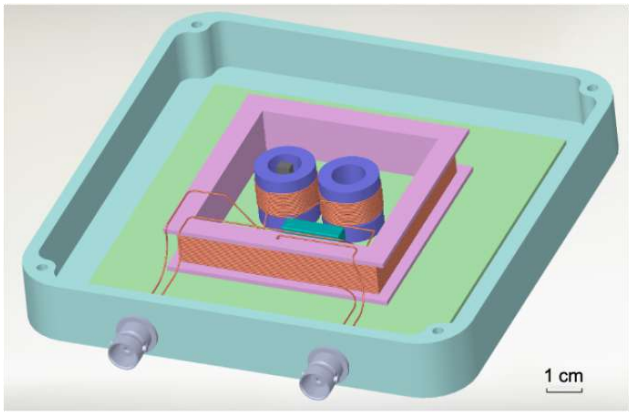
CNTs themselves are diamagnetic [1] but as their synthesis method can include ferrocene as metal precursor [2]; one may use the ferromagnetism of the iron to access the CNT concentration. As so, Vibrating Sample Magnetometry (VMS), by measuring the magnetic moment, can lead to the sample CNTs concentration [3], whenever the matrix containing the CNT is free of other ferromagnetic materials. VSM requires a large magnet-instrument and sample size is also limited.

We have developed an approach where we access the CNTs content by detecting the ferromagnetic signature of the iron used during the CNTs synthesis by dynamic susceptibility [4]. We designed and fabricated a table-top compact apparatus which detects the magnetic susceptibility of the samples with a varying excitation frequency. This dynamic susceptometer allows accessing the ferromagnetic signature of the ferrocene, and so of the CNTs. We demonstrated, by comparing top VSM measurements, that it allows detecting contents of few percents of CNTs in cement or polymer based matrices, and can be a quantitative way to determine the global CNTs content of different composite materials as well as homogeneity.

This technique could be extended to other type of composite materials containing one ferromagnetic element.

## References

[1] J. Heremans *et al.* (1994) https://doi.org/10.1103/PhysRevB.49.15122
[2] Pinault et al., (2005), https://doi.org/10.1021/nl051472k
[3] B.C. Satishkumar et al. (2002) https://doi.org/10.1016/S0009-2614(02)01082-5
[4] Patent pending - FR2114295 (2021).



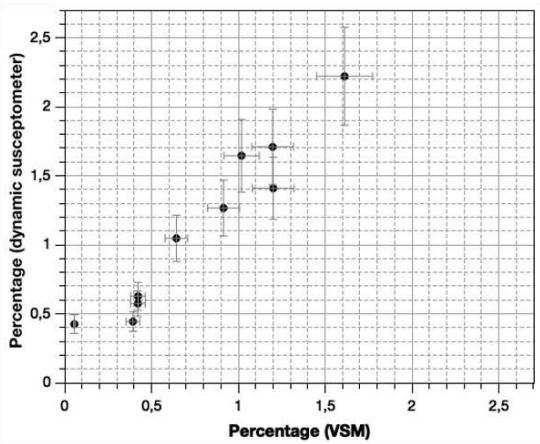




*Figure: (Left) Schematic of the dynamic susceptometer module (without its electronics). It comprises a square excitation loop (pink) within which are located two round detection coils (blue). (Right) Calculated CNT percentage of samples with an CNT content from 0 to 2%, obtained from the dynamic susceptometer and from the VMS measurements calibrated on 100% samples. Error bars for both type of measurements are given (grey bars).*

# Magnetic Force Microscopy: a tool to analyze magnetic properties of core-shell nanoparticles

*Miriam Jaafar[1,2]*, Gurvinder Singh[3], Cesar Tejera[4], Jorge Marqués -Marchán[4], Agustina Asenjo[4], Silvia Gallego[4], David Martínez- Martín [3]*

[1] *Departamento de Física de la Materia Condensada and Condensed Matter Physics Center (IFIMAC). Universidad Autónoma de Madrid 28049, Madrid, Spain*

[2] *Instituto Nicolás Cabrera, INC Cantoblanco, Madrid, Spain*

[3] *Faculty of Engineering and Nano Institute. University of Sydney, NSW 2006 Australia*

[4]*Instituto de Ciencia de Materiales de Madrid (CSIC), Spain*

*miriam.jaafar@uam.es

Despite decades of advances in magnetic imaging, obtaining direct, quantitative information with high spatial resolution remains an outstanding challenge. The imaging technique most widely used for local characterization of magnetic nanostructures is the Magnetic Force Microscope (MFM), which is indeed a very active topic of investigation [1,2]. Advantages of MFM include relatively high spatial resolution, simplicity in operation as well as sample preparation, and the capability to applied in situ magnetic fields to study magnetization process [3]. In this work we study the combination of hard and soft magnetic materials, characterized respectively by their large and weak magnetic anisotropies that enables interesting applications in a wide diversity of areas: permanent magnets, magnetic recording media, microwave absorption, ferrofluids or biomedical solutions. We create create a novel synthetic strategy to efficiently design magnetic alloy nanoparticles (10-20 nm) by virtue of a multi-shell onion structure type in which hard magnetic materials (cobalt ferrite, barium ferrite, and strontium ferrite) and soft magnetic materials (manganese ferrite, iron oxide) are carefully layered. These unique structures enable strong multiple exchange couplings at the interface of hard and soft magnetic layers, enhancing the nanoparticle magnetic properties [4] and reaching truly ferromagnetic behaviour. In that sense, the developement of novel modes and additional functionalities [5,6,7] in MFM are crucial to be widely used in experiments in such emerging areas of magnetism at the nanoscale.

**Acknowledgements**

This work has been financially supported through the projects of the Spanish Ministry of Science, Innovation, and Universities, PID2021-122980OA-C53 (FEDER), TED2021-130957B-C55 of the Spanish Ministry of Science and Innovation and within the framework of the agreement with the Autonomous University of Madrid: Excelencia para el Profesorado Universitario.

**References**

[1] O Kazakova, R Puttock, C Barton, H Corte-León, M Jaafar, V Neu, A Asenjo, Journal of Applied Physics 125, (2019) 060901
[2] V.G. Gisbert, C. A. Amo, M. Jaafar et al. Nanoscale 13 (2021) 2026-2033
[3] E. Berganza, M. Jaafar, et al. Nanoscale, 12 (2020) 18646-18653
[4] C. Tejera-Centeno and S. Gallego, J. Phys. D: Appl. Phys. 54 (2021) 264001
[5] M.Jaafar, A.Asenjo, Appl. Sci. 11 (2021) 10507
[6] M.Jaafar, J. P. Navarro, E. Berganza et al, Nanoscale, 12 (2020) 10090 –1009
[7] M.Jaafar, O. Iglesias- Freire, P. García Mochales, J.J. Saenz, A. Asenjo, Nanoscale, 8 (2016) 16989-16994

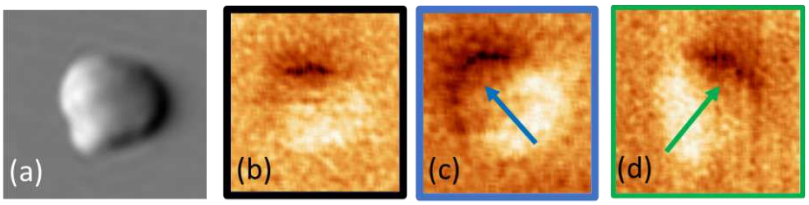



Figure: (a) Topography image of an individual multishell (4 layers) nanoparticle (~ 20 nm size); MFM images acquired at room temperature at different magnetic fields (b) remant state (c) + 700 Oe (d) -700 Oe.

# New versatile instruments to measure element-specific and macroscopic hysteresis at ID12 of the ESRF

Alex Aubert[1*], Gabriel Gomez[2,3], Konstantin Skokov[1], Fabrice Wilhelm[2], Heiko Wende[3], Andrei Rogalev[2], Oliver Gutfleisch[1] and Katharina Ollefs[3]

[1]Functional Materials, TU Darmstadt, 64295 Darmstadt, Germany, *alex.aubert@tu-darmstadt.de*
[2]European Synchrotron Radiation Facility, 38043 Grenoble, France
[3]Faculty of Physics, University of Duisburg-Essen, 47057 Duisburg, Germany
*alex.aubert@tu-darmstadt.de

With the increasing demand for new high technology and energy-efficient devices, multi-stimuli functional materials with strong interplay between their structural, magnetic, and electronic subsystems appear at the forefront of research in material science. However, to properly characterized such samples, it is highly appealing to develop a novel experimental approach which combines element-specific and macroscopic measurements performed under strictly the same conditions (e.g., temperature and magnetic field) on a single sample. We have recently implemented two new instruments at the beamline ID12 of the European Synchrotron Radiation Facility (ESRF), which will be introduced in this presentation. These instruments offer the ESRF users a unique possibility to measure under strictly the same experimental conditions the element-specific X-ray absorption spectroscopy (XAS)/ X-ray magnetic circular dichroism (XMCD), high-resolution XRD simultaneously with the measurement of various macroscopic properties (magnetization, volume changes, magnetocaloric properties, resistivity), all as a function of magnetic field (up to 17 T) and temperature (5–325 K) [1].

To demonstrate the potential and features of these scientific instruments, two case studies will be presented: (1) FeRh, which has a first-order anti-ferromagnetic to ferromagnetic transition around room temperatura (2) HoCo2, which exhibits a first-order ferrimagnetic to paramagnetic transition. These two cases demonstrate new horizons for studying the physics of magnetic materials, where the interplay between the magnetic, structural, and electronic subsystems of the solid is essential.

**Acknowledgements**

This work was supported by BMBF (Projekt05K2019) & DFG (405553726-TRR 270).

**References**

[1] Aubert A. et al. (2022). IEEE Transactions on Instrumentation and Measurement, 71, 1-9. Doi: 10.1109/TIM.2022.3157001

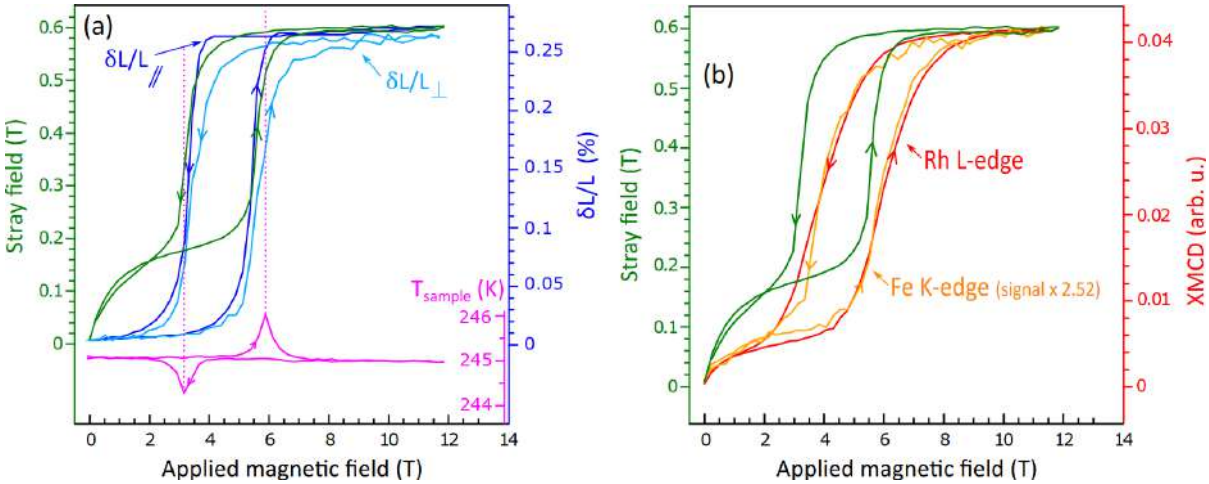



Fig. 1 Field dependence of the stray field at the surface of the FeRh sample, with (a) its temperature and magnetostriction data measured simultaneously and (b) with XMCD(H) curves either at the Fe K-edge (scaled by a factor of 2.52) or at the Rh L2-edge. These measurements are performed at 245 K to observe the transition from antiferromagnetic to ferromagnetic.

# SYMPOSIUM 17.
## NOVEL MAGNETIC TECHNIQUES, INSTRUMENTATION AND METROLOGY.
## S17 POSTERS

# Estimation of TbCo composition from a local-minimum-state image captured by MOKE microscope using machine learning

*Kenji Tanabe*[1]*, *Shiori Kuno*[1], *and Hiroyuki Awano*[1]

[1] *Toyota Technological Institute, Nagoya, Aichi, Japan*

*tanabe@toyota-ti.ac.jp

Recently, machine learning has allowed significant progress in material science. In spintronics, the estimation of some characteristics from a magnetic structure image has been studied as a new attempt of the application of machine learning. Its study is achieved by convolution neural network (CNN), which is a technique of image recognition in informatics. Magnetic structures are well-known to be deeply related to magnetic parameters. Although a complicated magnetic structure, which often appears in an as-grown magnetic film, is considered random by human eyes, it is influenced by some magnetic energies such as magneto-static energy. Thus, the characteristics of the parameters are probably hidden in the random magnetic structure. Previous studies have reported estimation of the parameters from magnetic domain images that were captured by high-resolution microscopes [1-2]. It, however, takes a long time to take an image. If the parameters can be estimated from the quickly captured image, the time required to estimate parameters can be greatly reduced and the speed of research and development may greatly increase in spintronics. In previous studies, the magnetic parameters are estimated from a magnetic state where the total magnetic energy is globally minimized[1-2]. It is known that there are numerous magnetic states where the energy is locally minimized because the magnetic material has hysteresis, and they often appear in the magnetization process. It is, however, unclear whether or not the parameters can be estimated from the local-minimum-energy state because the local-minimum-energy magnetic state depends on not only the parameters but also the magnetic process.

In this study, we have shown that TbCo composition is estimated from a local-minimum-state image captured by the MOKE microscope using CNN. Using the MOKE microscope is easy for most of the researchers in spintronics, and the time required to take an image is shorter than one second. We have proposed the ac demagnetizing method combined with a MOKE technique to control local-minimum-energy magnetic states and have generated several local-minimum-energy magnetic states from a single sample by its method. We have succeeded in the estimation of the TbCo composition from the generated local-minimum-energy state. Our results show that the magnetic parameters are deeply related to not only global-minimum-energy magnetic states but also local-minimum-energy magnetic states. This approach may estimate all magnetic parameters from any magnetic states that appear in experiments and considerably simplify experimental processes in spintronics.

## Acknowledgements

This work was partially supported by the Nagai science and technology foundation and the Konica Minolta science and technology foundation.

## References

[1] H. Y. Kwon et al., Science Advances **6**, eabb0872 (2020).
[2] M. Kawaguchi et al., npj Computational Materials **7**, 20 (2021).

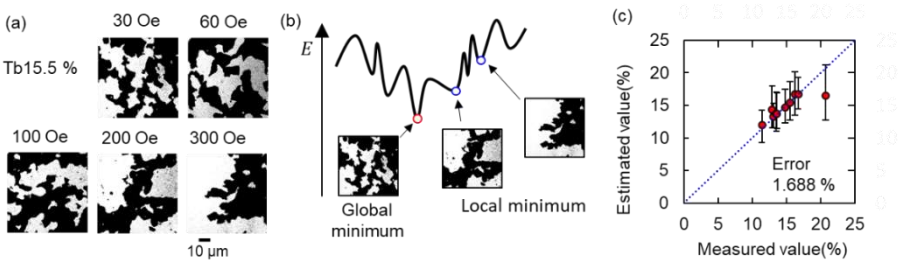



Figure: (a) Magnetic images having different domain sizes, which appear in a same sample. (b) Schematic diagram of global- and local-minimum states. (c) Estimation results of Tb composition.

# Spectral dependence of QMOKE anisotropy in FeRh

*Z. Sadeghi*, V. Wohlrath, P. Němec, E. Schmoranzerová, J. Kimák, P. Kubaščík, and T. Ostatnický*

*Faculty of Mathematics and Physics, Charles University, Praha, CZ-121 16, Czech Republic*

*Zeynab.sadeghi@matfyz.cuni.cz

An effective tool for the characterization of thin magnetic films is the magneto-optic Kerr effect (MOKE), which can provide information about fundamental magnetic properties, such as magnetic anisotropy or domain structure. MOKE linear-in-magnetization is only applicable in ferromagnets (FM) and certain canted antiferromagnets (AFM). In contrast, the quadratic-in-magnetization MOKE (QMOKE) is also employable for the fully compensated AFM [1]. We revealed that QMOKE signal exhibits a remarkable dependence on the polarization of incident light. This anisotropy is apparent in the amplitude and shape of the QMOKE signal. Our contribution describes how to measure the anisotropy of QMOKE comprehensively.

To demonstrate our approach, we selected a 20 nm thin layer of FeRh, prepared on the MgO substrate. FeRh is particularly interesting as a potential candidate for spintronic devices owing to its specific magnetic ordering, having FM phase above and a compensated AFM phase below the ordering temperature of 380 K [2]. We used an in-house built MOKE setup to measure the QMOKE anisotropy of the sample at different wavelengths in its FM state (at T= 420 K). We illuminated the sample with a wide-spectral range light source (supercontinuum laser, 460-1600 nm) under near-normal incidence. We detected polarization rotation ($\Delta\beta$) of the light reflected from the sample while applying a magnetic field of 50 mT under variable angles ($\varphi_H$ =0 to 360 deg) using home-made vectorial electromagnet. The method is described in detail in [3]. In order to study the impact of different polarization of incident light ($\beta$) on the polarization rotation $\Delta\beta$, we measured the QMOKE signal $\Delta\beta(\varphi_H)$ for different initial polarizations $\beta$. An example of the QMOKE signal, obtained at wavelength 810 nm is shown in Fig.1 (a). Clearly, there is a remarkable change in the amplitude of the MO signal for different $\beta$, indicating a strong anisotropy of the magneto-optical response. Since the QMOKE is known to be strongly wavelength-dependent [4], we measured the full spectrum of the QMOKE anisotropy (see Fig.1 (b)). The obtained spectral dependence is rather similar to that of QMOKE in thin Fe films [5]. Moreover, by measuring QMOKE, we are also able to extract the magnetic anisotropy of FeRh.

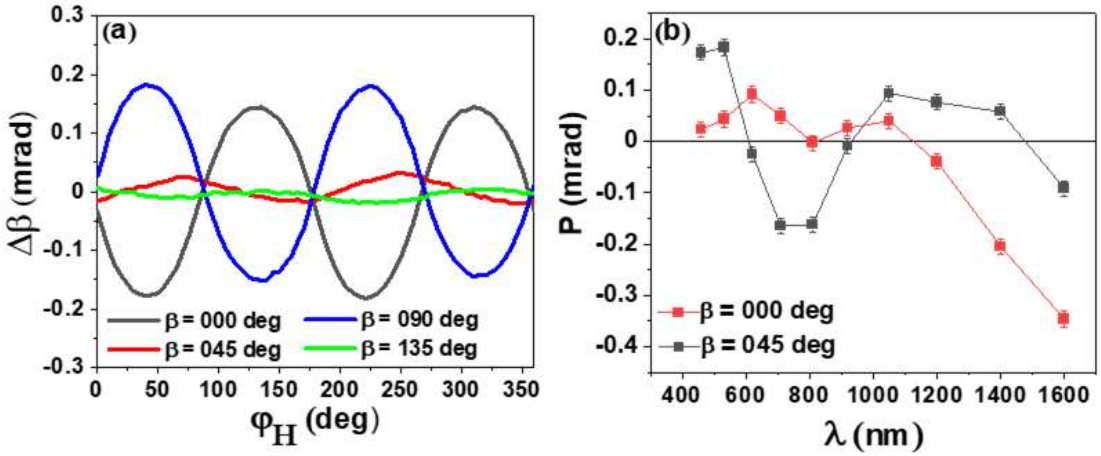



Figure 1: (a) Polarization rotation ($\Delta\beta$) as a function of the external field angle ($\varphi_H$) at $\lambda$ = 810 nm for different polarization orientations $\beta$, which indicates large anisotropy. b) Spectral dependence of QMOKE amplitude for polarizations $\beta$ =0° and 45°. Temperature T = 420 K, applied field $\mu_0 H$= 50 mT.

## References

[1] P. Němec, M. Fiebig, T. Kampfrath, and A. V. Kimel, Nature Physics 14, 229 (2018).
[2] V. Saidl et al., New J. Phys. 18, 083017 (2016).
[3] V. Wohlrath et al., in preparation.
[4] N.Tesařová, et al., Physical Review B 89, 085203 (2014).
[5] R. Silber et al., J. Physical Review B 100, 064403 (2019).

# Materials Informatic for Magnetic Material Discovery

*Nicola A. Morley[1]*, Richard Rowan-Robinson[1], Coyun Oh[1], Sophia Carpio[1] and Zhaoyuan Leong[1]*

[1] *Department of Materials Science and Engineering, University of Sheffield, Sheffield, UK, S1 3JD*

*n.a.morley@sheffield.ac.uk

The climate emergency has established the need for sustainability within existing and new technologies, which is driving a demand for material innovation. New materials need to be economically sourced from abundant elements, whilst still obtaining the functional characteristics of the existing leading materials. Functional Magnetic Materials (FMMs) are central to new green technologies and an excellent example of a sector with great industrial demand for innovation. FMMs are categorised as either hard/permanent magnets, or soft magnets, both of which find a wide selection of applications: wind turbines, electrical machines, power electronics and 6G technology. Different magnetic properties such as coercive field (large for hard magnets and small for soft magnets) and saturation magnetisation (high for both) are required for different applications. At the present, existing hard magnets, i.e. NdFeB and SmCo, consist of elements that are on the critical list, while soft magnets are limited due to processing costs and eddy losses.

The need for innovation is clear: by improving the material properties of FMMs, industries can fully capitalise on the aforementioned engineering advances in green technologies, thus saving money and benefiting the environment. Traditional methodologies for material discovery, whereby existing material compositions are tweaked and optimised, have proved too slow and costly, taking over a decade to reach deployment; such an approach is not practical or sufficient to address the current material challenges. Material informatics will pave the way for discovering new FMMs that overcome these existing problems, by using data-driven solutions to reduce the use of natural resources and expensive experiments. The figure below shows the digital approach we have been taking for FMM discovery.

Our research has focused on using Natural Language Processing (NPL) to data mine open access papers to create a FMM database. This has been achieved by combining the linear approach NPL, which searches for defined compositions and parameters within papers, with semantic networks, to allow the compositions related parameters to be correctly linked together [1]. In doing this we are able to data mine papers, which contain more than one composition and magnetic parameter for the database. Having created this database, machine learning (ML) algorithms are trained on it, which are then used to observe trends within the data, along with predicting compositions with specific magnetic parameters. These compositions are then fabricated and characterised using high throughput techniques, including combinatorial sputtering, XRD, FMR and MOKE magnetometry. This allows us to check the results from the ML, quickly and cheaply, along with discovering new functional magnetic materials.

## Reference

[1]https://figshare.shef.ac.uk/collections/Feasibility_study_to_assess_Natural_Language_Processing_for_Functional_Magnetic_Materials/6373155

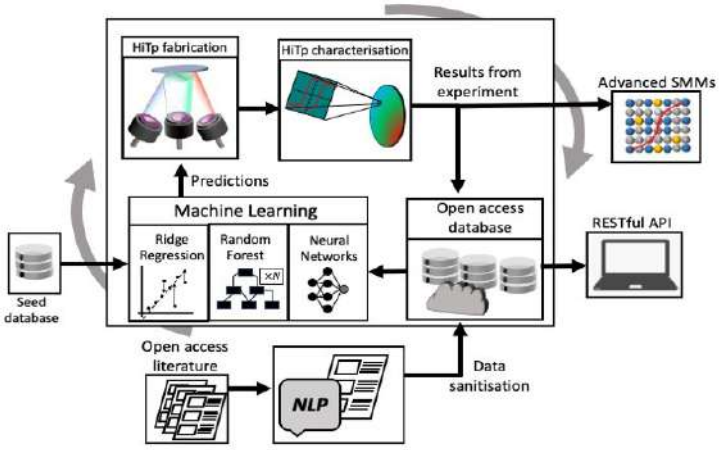



Figure: Schematic of how materials informatics works for magnetic materials

# Multi-Parameter Analyser for Simultaneous Conversion X-ray and Backscatter Mössbauer Spectroscopy

*Jack O'Brien[1,*], Hasan Ahmadian Baghbaderani[2,1], Frederico O. Keller[3], Nora Dempsey[3], Laura H Lewis[4], Plamen Stamenov[1,2,†]*

[1] *School of Physics and CRANN, Trinity College, University of Dublin, Dublin 2, Ireland*
[2] *Microsystems Centre, Tyndall National Institute, University College Cork, Cork, Ireland*
[3] *Univ. Grenoble Alpes, CNRS, Institut NEEL, 38000 Grenoble, France*
[4] *Department of Chemical Engineering, Department of Mechanical and Industrial Engineering, Northeastern University, Boston, MA 02115, USA*
[*]obriej25@tcd.ie [†]stamenop@tcd.ie

We present a method for the acquisition and analysis of $^{57}$Fe backscatter Mössbauer spectra with detection across a continuous energy range that includes the 14.4 keV γ-rays and characteristic 6.4 keV x-rays. We utilise purpose-built MPAs (multi-parameter analysers) constructed on the basis of commercial ADCs and custom high-speed digital latches. Our system allows for the simultaneous registration of Doppler modulation velocities and photon energies with up to 4K and 8K digital channel resolution, respectively, and a maximum acquisition speed with these resolutions of 100 MHz. Every detector event is recorded amd optimal discrimination of the γ-ray events, the conversion x-ray events, and the high-energy non-resonant fluorescent photons, is performed post-acquisition, in contrast to most systems which record at a single narrow energy window per detector. The processed data is subsequently fit using a full Hamiltonian model for the nuclear energy levels of $^{57}$Fe [1]. There are a number of advantages of our combined acquisition and analysis method, for example: the shorter absorption length of the 6.4 keV x-rays allow for greater surface sensitivity compared to the 14.4 keV γ-rays [2], the non-resonant fluorescent photons at higher energies provide an intrinsic route for robust background correction, and Fe-containing samples of arbitrary atomic structure and morphology can be investigated non-destructively without the need for any surface preparation.

Four examples of systems we have investigated are presented to demonstrate the utility of our method: oxidation on the surface of a millimetric α-Fe plate, 12 µm thick amorphous CoFeB-based ribbons with ultra-soft magnetisation [3], 5 µm thick anisotropic Nd-Fe-B films sputtered on Si substrates with varying deposition conditions [in prep], and a 6 mm thick sample from the nickel-rich ungrouped iron meteorite NWA 6259, to investigate the presence of FeNi in the difficult-to-synthesise, $L1_0$ phase, tetrataenite [4].

**Acknowledgements**

The Authors would like to acknowledge the financial support of the SFI under grants 12/RC/2278 (AMBER) and 17/NSFC/5294 (MANIAC), the EC under Grant Agreement No. DLV-737038 (TRANSPIRE), and the U.S. Department of Energy, Office of Basic Energy Science, under Award Number DE SC0022168.

**References**

[1] C. J. Voyer and D. H. Ryan, *Hyperfine interactions*, **170** (2006) pp. 91-104.
[2] I. Fleischer et al., *Journal of Geophysical Research: Planets*, **113** (2008), p. E06S21.
[3] H. A. Baghbaderani et al., *Journal of Alloys and Compounds*, **924**, (2022) pp. 166366.
[4] L. H. Lewis, *Journal of Physics: Condensed Matter*, **26.6**, (2014) pp. 064213.

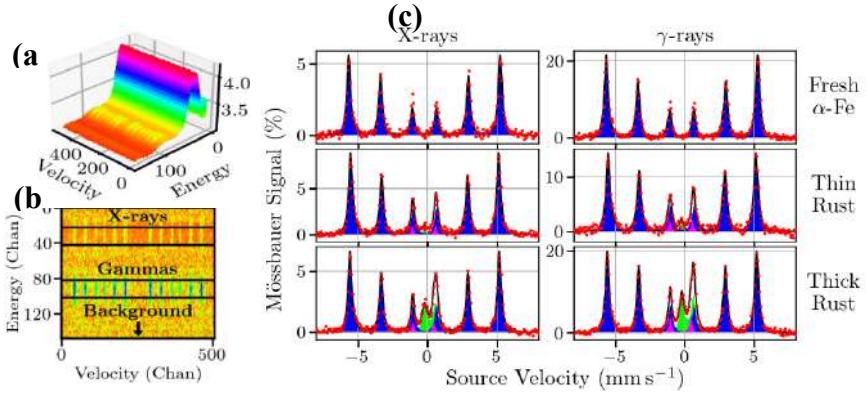



Figure: (a) Raw data matrix of the acquisition, zoomed to resonant energies, (b) data normalised for each velocity channel, with resonant peaks clearly visible and (c) the resulting data and fit for the iron plate with increasing surface oxidation, the blue, magenta and lime contributions are from pure α-Fe, β-FeOOH and an unidentified Fe(II) product, respectively.

# Quantitative high sensitivity Magnetic Force Microscopy in Vacuum

*Christopher Habenschaden[1]*, Sibylle Sievers[1], Andrea Cerreta[2], Hans Werner Schumacher[1]*

[1] *Physikalisch-Technische Bundesanstalt, Bundesallee 100, 38116 Braunschweig, Germany*

[2] *Park Systems Europe GmbH, Schildkrötstraße 15, 68199 Mannheim, Germany*

*christopher.habenschaden@ptb.de

Magnetic force microscopy (MFM) is a well-established technique in scanning probe microscopy (SPM) allowing the imaging of magnetic samples with spatial resolution of tens of nm and stray fields down to the mT range. However, as commonly device-specific phase or frequency deviation is used in publications, it lacks comparability between measurements. This issue can be overcome by characterizing the magnetic stray field of the tip by using magnetic reference samples. As a result, the calculation of sample stray fields in A/m is possible, yielding quantitative magnetic force microscopy (qMFM) measurements.

Spatial resolution and field sensitivity can be pushed to several nm and the hundred µT range by measuring in vacuum conditions. This results from the higher cantilever quality factors $Q$ in vacuum, directly leading to an increase in measurement signal. However, with increasing signal amplitude, non-linear behavior must be considered. Additionally, advanced feedback techniques are required for stable operation [1].

We here present an implementation of a phase-locked loop-based (PLL) signal detection [2] that overcomes non-linearities into a Park Systems NX-Hivac SPM. This allows transfer function based quantitative and stable magnetic force microscopy also in vacuum without an artificial reduction of $Q$. The gain in resolution and sensitivity is discussed and measurements on samples like stripe domain samples and nanopatterned as well as skyrmion hosting multilayer stacks are shown and analyzed to demonstrate the feasibility of the PLL approach. Furthermore, it will be shown that the enhanced sensitivity also enables the sensing of large samples with magnetic features in the mm range.

## Acknowledgements

This project was supported by the Federal Ministry of Economics Affairs and Climate Action within the TransMeT project *"Realisierung eines quantitativen Magnetkraftmikroskopie-Messverfahrens gemäß IEC TS 62607-9-1 mit einem kommerziellen System"*.

## References

[1] J. Schwenk, M. Marioni, S. Romer, N. R. Joshi, and H. J. Hug, "Non-contact bimodal magnetic force microscopy", Appl. Phys. Lett. **104**, 112412 (2014); https://doi.org/10.1063/1.4869353

[2] H. Seo, D. Goo and G. Jung, R. Stomp and J. Wei, "Park NX-Hivac: Phase-lock Loop for Frequency Modulation Non-Contact AFM", Park Systems Atomic Force Microscopy Application note **#21**.

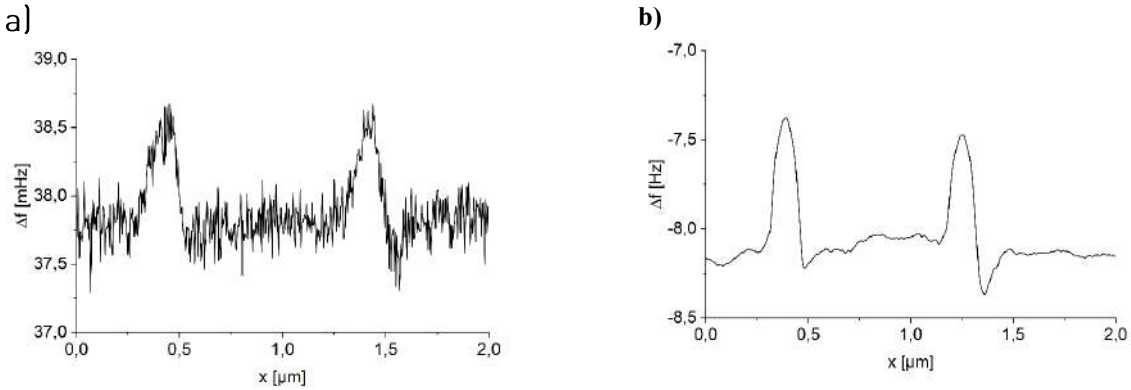



**Figure:** MFM line scan of a nanostructured magnetic multilayer sample $[Co(1.0\ nm)/Ru(1.4\ nm)/Pt(0.6\ nm)]_{10}$ in **a)** ambient contions ($p \approx 10^3$ mbar, $Q \approx 200$) and **b)** high vacuum conditions ($p \approx 10^{-6}$ mbar, $Q \approx 31$ k). Plotted raw data has been not post processed after lock-in measurement. Drastically improved signal to noise ratio (SNR) in vacuum is self-evident, allowing advances in spatial resolution and field sensitivity.

# New Perspective on Planar Inductive Sensors: Radio-Frequency Refractometry for Highly Sensitive Quantification of Magnetic Nanoparticles

J.L. Marqués[1*], J.C. Martínez-García [1], M. Salvador [1], P. Fernández-Miaja [2,]
A. García-Arribas [3], M. Rivas [1]
1*Department of Physics, University of Oviedo, Gijón, Spain*
2*Department of Electrical Engineering, University of Oviedo, Gijón, Spain*
3*Department of Electricity and Electronics, University of the Basque Country, Leioa, Spain*
*marquesjose@uniovi.es

We have developed a method to quantify the mass of nano-disperse materials with high sensitivity and low-cost with applications in medical, food safety and industrial environments. In this work, we provide a method to quantify the presence of magnetic and dielectric nanomaterials, which employs inductive sensors working at radio-frequency. The main difference between our method compared to previous ones is the sensitivity to the magnetic permeability and the electric permittivity, which expands the applicability scope of the technique and causes its sensitivity improvements.

Our method uses the self-resonant frequency (SRF) of inductors allowing high sensitivity with low cost and enabling the development of portable devices. Starting with the equivalent impedance model [2] and Lichtenecker's mixing formulas [3], we define an analytic simplified model to describe the relation between nanomaterial mass and variations of the SRF of the inductor[4]. Our method measures the refraction index of the nano-tag, making the technique able to quantify any nanomaterial like gold, latex or MNPs.

We verified the model and simplifications against electromagnetic simulations and experimental measurements. These verifications demonstrate the validity of the model, the value of magnetic nanoparticles as nano-tags for precise quantification, and the advantages of the SRF measuring method for low-cost, high-sensitivity systems.

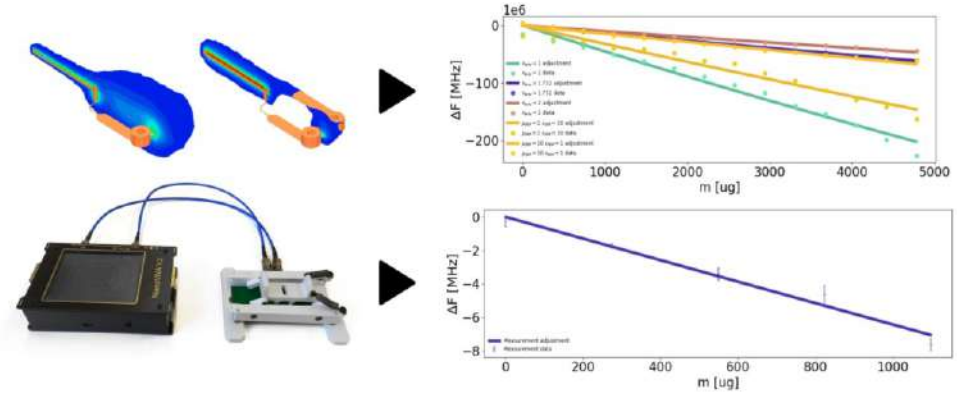



Figure 1. Simulations and experimental setups, data, and model adjustment results.

## Acknowledgements

This work was partially founded by the Ministry of Science and Innovation of the SpanishGovernment through grant PLEC2022-009490, the University Technological Institute of Asturias(IUTA), under grant SV-22-GIJON-18, and the Government of the Principality of Asturias underprojects FICYT/IDI/2021/000100 and FICYT/IDI/2021/000273.

## References

[1] Moyano, A. et al., 2020. Magnetic Lateral Flow Immunoassays. Diagnostics.
[2] Qin Yu. et al., 2002. RF Equivalent Circuit Modeling of Ferrite-Core Inductors and Characterization of Core Materials.
[3] A.V. Goncharenko. et al., 1999. Lichtenecker's equation: applicability and limitations.
[4] Marqués, J. et al., New Perspective on Planar Inductive Sensors: Radio-Frequency Refractometry for Highly Sensitive Quantification of Magnetic Nanoparticles.

# Electromagnet End Station for X-ray Magnetic Circular Dichroism

*Peter Bencok[1]*, Raymond Fan[1], Paul Steadman[1]*

[1] *Diamond Light Source, Harwell Science and Innovation Campus,Didcot, United Kingdom*

*peter.bencok@diamond.ac.uk

We report on state of an electromagnet end station for x-ray magnetic circular dichroism (XMCD). This end station has been installed on the absorption branch of the Beamline for Advanced Dichroism Experiments (BLADE). At the Diamond Light Source (DLS), BLADE is a soft x-ray beamline optimized for studies of magnetic materials using x-ray magnetic circular dichroism.

The absorption branch has been originally equipped with the superconducting high field magnet that can provide the field up to 14T and cool samples down to 3K. The operation of the high field magnet is however time demanding and it suffers from the remanent field of about 20 mT so when small magnetic field is applied, the magnetisation cannot be determined accurately. Hence, the need for a complementary end station which would provide accurate small field and high throughput.

The electromagnet end station is built around commercial coil from GMW Associates providing the field up to 1.9T. The sample can be cooled with a flow of liquid helium using vertically mounted cryostat Janis ST-400. The base temperature is about 15K. Samples can be cleaved in-situ or annealed to the temperature of 500K.

The electromagnet system has been successfully tested and routinely used since 2021. An example of experimental result will be presented [1]. It is open to users via standard DLS proposal route.

## References

[1] B. Achinuq et al., *Phys. Status Solidi RRL* **16**(2022), 2100566

Figure: Image of the electromagnet end-station connected at the end of the absorption branch of I10.

# Mapping the magnetic response of materials on a local scale using magneto-resistive sensors

*Aurélie Solignac[1*], Wanissa Benmessaoud[1], Julien Moulin[1], Fawaz Hadadeh[1], Andrin Doll[1], Elodie Paul[1], Myriam Pannetier-Lecoeur[1], Natalia Segeeva-Chollet[2], Caude Fermon[1]*

[1] *SPEC, CEA, CNRS, Université Paris-Saclay, CEA Saclay, 91191 Gif sur Yvette Cedex, France*
[2] *CEA LIST, 91191 Gif-sur-Yvette, France*
*aurelie.solignac@cea.fr

The characterization of the magnetic properties of materials at the local scale is important for applications such as in situ monitoring, non-destructive testing or nanometrology. Indeed, for some materials that exhibit a magnetic response and in particular steels, mechanical and magnetic properties are correlated via the microstructure. The measurement of magnetic properties at the local scale could therefore allow access to the mechanical properties of materials in a non-destructive way and a better understanding of their microstructure. In order to obtain additional contrasts, it is possible to use the mapping of the frequency response to the application of an alternating magnetic field (magnetic susceptibility).

Two magnetic mapping tools, at two scales, submillimeter and submicrometer, are developed in the laboratory by combining magnetoresistive magnetic sensors [1] and a scanning system. The use of the giant and tunnel magnetoresistance effect, based on spin electronics, allows the development of very sensitive magnetic sensors whose size can be submicron. These sensors allow to detect magnetic fields emitted by the sample surface in a quantitative way, with a detectivity of the order of $nT/\sqrt{Hz}$, in a topography-decolored way, and on a large frequency range (DC to the hundred MHz).

In order to achieve micrometer and submicrometer resolution, the GMR sensor is integrated into a flexible cantilever (Figure 1 left) and combined with a local probe microscope. On a larger scale, the tool working with sub-millimeter resolution is composed of a 3D probe integrating GMR sensors mounted in Wheatstone bridge and gradimeter to eliminate temperature and environmental drift, and positioned on a pyramidal support (Figure 1 right), and a motorized stage. The three components of the field can be extracted and allow a 3D mapping of the fields emitted by the material surface.

## Acknowledgements

We acknowledge the following organisms for funding: the CEA for the internal funded projects MIMOSA and CALM and the PhD Grant “Phare Amont-Aval”, the Swiss National Science Foundation (SNSF) for a mobility fellowship (project 165238) to A. Doll, the ANR funding through grant n°ANR-17-CE19-0021-01, the regional French Labex NanoSaclay and Palm for the funded projects CALM and CARTE.

## References

[1] C. Fermon and Marcel Van de Voorde Nanomagnetism: applications and perspectives, Wiley VCH (2017)
[2] J. Moulin, PhD thesis 2020; https://theses.hal.science/tel-02976807v1
[3] F. Hadadeh, A. Solignac, M. Pannetier-Lecoeur, N. Sergeeva-Chollet and C. Fermon, IEEE Sensors Journal V19, I22 (2019); https://hal.archives-ouvertes.fr/hal-02316945

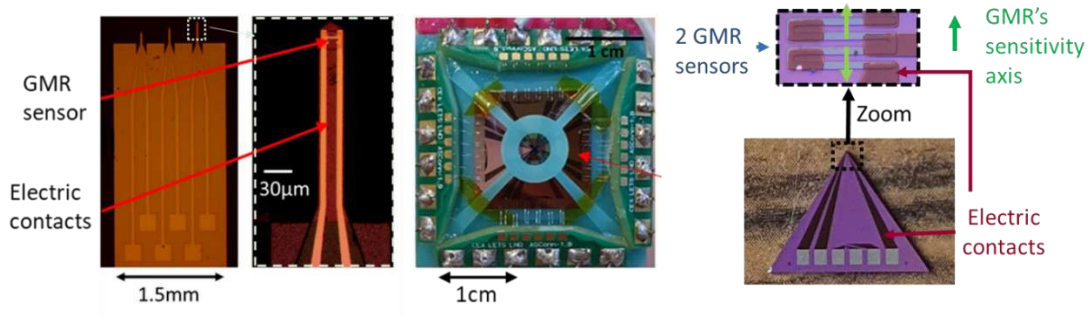



Figure: (Left) Atomic force microscope (AFM) support composed of 3 flexible cantilivers (1µm Si3N4) on which are integrated a GMR sensor. (Right) 3D probe composed of 4 GMR dies positioned on a pyramidal support.

# Multi-Port Sample Carrier System for All-Electrical Characterisation of Thin-Film Magnonic Devices

*Travis Gustafson,[1]*, Vedran Furtula[1], Ferran Macià , Thomas Tybell[1], Erik Wahlström[1]*

*[1] Norwegian University of Science and Technology, Høgskoleringen 1, Trondheim, Norway*

*[2] University of Barcelona, Martí i Franquès, 1-11, Barcelona, Spain*

*travis.gustafson@ntnu.no

Implementing a sample carrier system to characterize spin waves on thin films put demands on signal integrity, wide bandwidth, optimized interface connections and multiple layouts per sample. We will present detailed results from our design of an improved device carrier for wideband electrical interfacing (<17 GHz) of mixed signal magnonic devices. The device is designed for all electrical measurements of mixed surface acoustic waves and spin waves.

The focus will be the verification of a compact, 16-port chip carrier and enclosure compatible with material requirements for measurement setups that include an applied magnetic field and variable temperature. The design premise accommodates commercially available direct cable interface contacts and a mechanical enclosure for both shielding and thermal control. The contacts are connected to a reusable chip carrier printed-circuit-board (PCB), designed to provide a controlled, repeatable sample chip environment. For signal integrity and broad band impedance, CST software simulations are used and compared to control measurements. The chip carrier PCB is designed for commercial reproduction. The sample chips are produced in-house, utilizing a thin film growth and lithography techniques.

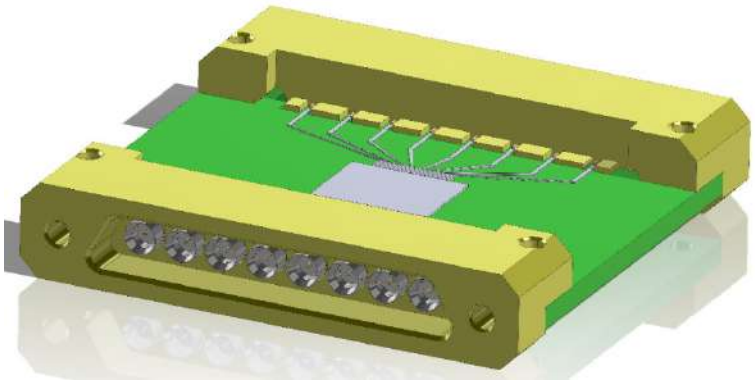


Figure 1: Sample (gray) connected to the PCB (green) via ribbon bonding, and then connected to two compact 8-port Mini-D RF Edge Launch Connectors (gold)

From this design we will present simulations and experimental results. We will address effects of different bonding methods (wire/ribbon) and circuit layout designs of the chip carrier and the sample lithography. We will exemplify the design through presenting results from studies of all-electrical measurements of coupling between spin-waves generated by wide-band antennas [1] and surface acoustic waves on $LiNbO_3$ [2]. We will present results on signal integrity, correlation with simulation, as well as preliminary results of intermixing of signals. Results will be presented from thin film magnetic materials in surface acoustic wave substrates. Prototypical materials such as Fe and Ni films will provide baseline measurements, leading toward measurement of $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) thin films deposited on the surface acoustic wave material $LiNbO_3$.

## References

[1] Singh, Suraj, et al. "Propagation properties of spin wave in Co2FeAl Heusler alloy ultrathin films." *Materials Research Express* 8.8 (2021): 086101.

[2] Casals, Blai, et al. "Generation and imaging of magnetoacoustic waves over millimeter distances." *Physical review letters* 124.13 (2020): 137202.

# Ontologies for Materials Science: Beyond Databases and Electronic Laboratory Notebooks

*L. Caron*[*1,2], *T. Samanta*[1], *C. Taake*[1], *M. Blum*[3] *and B. Ell*[3]

[1] *Faculty of Physics, Bielefeld University, 33501 Bielefeld, Germany*

[2] *Helmholtz-Zentrum Berlin für Materialien und Energie, 12489 Berlin, Germany*

[3] *Faculty of Technology and CITEC, Bielefeld University, 33619 Bielefeld, Germany*

*lcaron@uni-bielefeld.de

As materials scientists we have always used digital means to acquire and analyse our data, at least ever since computers became available. One point which was slower to change was how we record our thoughts and make our notes during the experiments themselves: we still heavily rely on the good old paper notebook.

However, in the past three decades computers and digital mobile devices have become ubiquitous not only in laboratories but also in our daily lives. This led to a gradual shift in how we produce and handle data. Many of us have experimented or made the full switch to electronic laboratory notebooks (ELN). There are many advantages to ELNs. They are easy to search, copy, archive and share. You can easily collaborate with and monitor other scientists work [1]. In many cases this switch is strongly encouraged by funding agencies' requirements on data management.

However, ELNs have limitations when it comes to machine automated data processing at accessing all information within the data. But what if your digital book could do more than just store data and aid you research?

This is where ontologies come in. Ontologies are a structured representation of knowledge that follows a standardized (domain specific) schema. They enable the modelling of relationships between concepts or things (entities), making it easier to retrieve data using a powerful query language named SPARQL.

When using a database of experiments, you may e.g. query which/how many samples you have substituted Mn for Cr. From an ontology however, you can query what was the mechanism used to reduce the volume of your parent compound, which is a much more complex question than a database can answer since it models not only values but also the knowledge of how you got to your results.

Overall, ontologies are a crucial step towards making our data FAIR - Findable, Accessible, Interoperable, Reusable. It is not a matter of choosing between ELNs or Ontologies. For instance, integrating ontological concepts into ELNs can enhance their functionality and expand the range of data that can be processed and analysed.

In this presentation I will talk about the challenge of designing and implementing an ontology for magnetocaloric materials in the project DiProMag [2] as part of Germany's Material Digital initiative [3]. What advantages ontologies bring versus simple databases? How building ontologies actually changes the way we conceptualize and record our experiments? These are questions I will strive to answer as ontologies are the natural evolution of how we manage our data.

## Acknowledgements

This work was carried out in the framework of the Joint Lab BiBer and was supported by the BMBF joint project DiProMag FKZ 13XP5 120B.

## References

[1] R. Kwok, *Nature* **560**, 269-270 (2018)

[2] https://www.dipromag.de/

[3] https://www.materialdigital.de/

# Wave Vector Dependence of the Relaxation Time of Exchange Spin Waves

*Loic Temdie[1,2,*], Vincent Castel[1,2], Carsten Dubs[3], Hicham Majjad[4], Romain Bernard[4], Yves Henry[4], Matthieu Bailleul[4], Vincent Vlaminck[1,2]*

*[1]IMT Atlantique, Microwave Dpt., CS 83818, 29238 Brest, France*

*[2]Lab-STICC (UMR 6285), CNRS, Technopole Brest-Iroise, 29238 Brest Cedex 3, France*

*[3]INNOVENT E.V. Technologieentwicklung, Prussingstrasse 27B, Germany*

*[4]IPCMS Institut De Physique Et Chimie Des Materiaux De Strasbourg, France*

**contact : loic.temdie-kom@imt-atlantique.fr*

The field of magnonic constitutes a promising ground for the integrating of non-reciprocal microwave components such as circulators, isolators, and phase shifters [1]. Recently, unidirectional transmission of exchange spin waves down to 60 nm wavelength was achieved using the chiral coupling between ferromagnetic nanowires with thin YIG films [2].

In this communication, we implement this method to study the wave vector dependence of the relaxation time of dipole-exchange spin waves over a broad range of wavevectors k = [0-100 rad.µm$^{-1}$]. We performed spin wave spectroscopy in the Damon-Eshbach configuration on several devices consisting of a w=200 nm width, t=60 nm thick, and a=400 nm lattice spacing permalloy ($Ni_{80}Fe_{20}$) nanowires arrays on top of a 55 nm thick YIG film (see Fig. 1-a). The frequency-field mapping of the transmission spectra for these devices reveals two types of non-reciprocal transmission (see Fig. 1-b). At higher frequency, we attribute the unidirectional transmission to the excitation of high wavevectors ($k_{NW}$) from the nanowires arrays. At lower frequencies, the partial non-reciprocity observed comes from the wavevectors ($k_{CPW}$) directly excited by the *rf* field of the antenna. We fit the separation distance dependence (D) of the amplitudes, and the oscillation frequencies of theses transmission spectra in order to extract the attenuation length $L_{att}$ (k, H), and the group velocity $V_g$ (k, H) for all wavevectors [4]. The obtained relaxation times defined as $\tau$ (k, H) = $L_{att}$ (k, H) /$V_g$ (k, H) (see Fig. 1-c) show that it decreases as the wavevector and the bias field increase. We obtain a good agreement with theoretical expressions of the relaxation time derived from Kalinikos-Slavin formalism. This study benchmarks the propagation properties of exchange spin waves in the context of phenomenological Gilbert damping.

This work was supported by the French ANR project “MagFunc” and the Département du Finistère through the project “SOSMAG”.

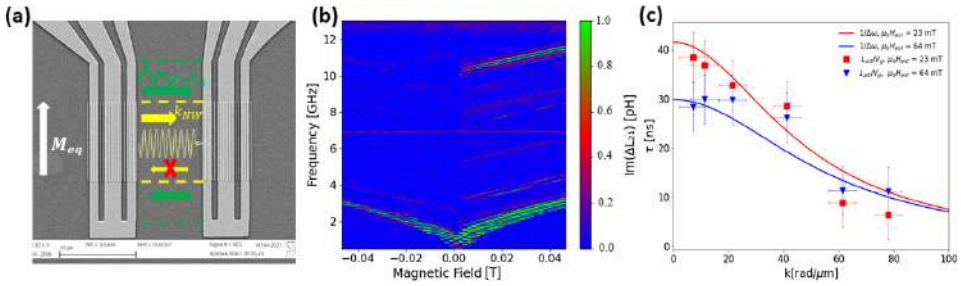



Figure 1: (a) Scanning electron micrograph image of $Ni_{80}Fe_{20}$ NWA. (b) Mapping of imaginary part of transmission spectra $\Delta L_{21}$. (c) Comparison of measured relaxation time with analytical expression for 23mT and 64mT in plane applied field.

**Reference:**

[1] A. Barman et al., J. Phys. : Condens. Matter **33**, 413001 (2021).

[2] J. Chen, et al., Phys. Rev. B **100**, 104427 (2019).

[3] L. Temdie, V. Castel, C. Dubs, G. Pradhan, J. Solano, H. Majjad, R. Bernard, Y. Henry, M. Bailleul, and V. Vlaminck, “High wave vector non-reciprocal spin wave beams”, AIP Advances **13**, 025207 (2023).

[4] N. Loayza, M. B. Jungfleisch, A. Hoffmann, M. Bailleul, and V. Vlaminck, “Fresnel diffraction of spin waves”, Physical Review B 98, 10.1103/physrevb.**98**.144430 (2018).

# Complete Thermodynamic Characterization of a Magnetocaloric Material Through Magnetometry

*C. Pereira[1], R. Almeida[1], C.Amorim[2], J. S. Amaral[2], J. P. Araújo[1], J. H. Belo[1]**

[1] *IFIMUP, Departamento de Física e Astronomia da Faculdade de Ciências da Universidade do Porto, Rua do Campo Alegre 687, 4169-007 Porto, Portugal*

[2] *Departamento de Física and CICECO, Universidade de Aveiro, 3810-193 Aveiro, Portugal*

*jbelo@fc.up.pt

Nowadays more than ever, it is important to look for greenhouse gas emissions free technologies to implement in the refrigeration industry. Magnetic refrigeration is a very promising candidate since this technology is based on the magnetocaloric effect of solid-state materials around room temperature which do not emit greenhouse gases. Measuring and comparing thermodynamic properties of such magnetocaloric materials is a crucial tool towards the continuous improvement of magnetic refrigerants.

The magnetocaloric effect is expressed by two quantities. The first one is the entropy change of the material upon the variation of the field $H$ under isothermal conditions, $\Delta S_{iso}$, and can be calculated indirectly from isothermal magnetization versus field curves using a commercial superconducting quantum interference device (SQUID) [1]. The second one is the temperature change induced by the application of the field $H$ under adiabatic conditions, $\Delta T_{ad}$. This last quantity can be obtained directly by measuring the temperature of the sample, however, it can be challenging to develop a setup for such measurements in adiabatic conditions. Nevertheless, recently, it has been shown that this quantity can also be obtained directly through time-dependent magnetometry, namely using a SQUID device [2]. With this last approach, a $\Delta T_{ad}(T)$ curve profile was obtained using Gadolinium (a benchmark magnetocaloric material) under a 1 T field change. The adiabatic temperature change can also be measured indirectly by using thermodynamic equilibrium measurements of magnetization and using the following expression derived from Maxwell relations [2],

$$\Delta T_{ad}(T_i)_{\Delta H} = \int_{H_i}^{H_f} \frac{T_i}{C_p(T_i,H)\left(\frac{\partial M(T_i,H)}{\partial T}\right)_H} dH$$

In this work, by solving this equation in order to $C_p(T_i,H)$ and using the direct measurement of $\Delta T_{ad}(T_i)_{\Delta H}$ through magnetometry for small incremental changes $\Delta H$ up to 1 T under adiabatic conditions, we were able to estimate the temperature and field dependent heat capacity of the material at a given temperature. Ensuring adiabaticity during these measurements is also challenging, which motivates studying additional setups with improved thermal insulation and/or faster magnetic field applications. Our current experimental procedure of increasing the field incrementally has improved on the previously reported method, reducing the underestimation of $\Delta T_{ad}(T_i)_{\Delta H}$ without compensation techniques. We are currently exploring these different approaches to estimate the heat capacity $C_p(T_i,H)$ of Gadolinium using time-dependent magnetometry and have obtained values within 48% of the reported values for Gadolinium [3]. This approach will allow a full characterization of a magnetocaloric material ($\Delta S_{iso}$, $\Delta T_{ad}$, $C_p$) using a single SQUID magnetometer, enabling accelerated progress towards new, competitive, and industry-ready materials.

**Acknowledgments:** This work was developed within the scope of the following projects financed by EEA grants via the project FBR_OC1_85 and by the Portuguese national funding agency for science, research and technology (FCT): UIDB/50011/2020 and UIDP/50011/2020 (CICECO), and PTDC/EME-TED/3099/2020 (IFIMUP). J.H.B. also thanks FCT for the projects PTDC/FISMAC/31302/2017 and CERN/FISTEC/0003/2019 and for his contract DL57/2016 reference SFRH-BPD-87430/2012.

## References

[1] Quantum Design North America, SQUID Magnetometer - Quantum Design MPMS 3.

[2] R. Almeida, C.Amorim, J. S. Amaral, J. P. Araújo, J. H. Belo, Direct Measurement of the Magnetocaloric Effect through Time-Dependent Magnetometry, Physical Review Applied **18**, 024081 (2022).

[3] S. Y. Dan'kov, A. M. Tishin, V. K. Pecharsky, and K. A. Gschneidner, Magnetic phase transitions and the magnetothermal properties of gadolinium, Phys. Rev. B **57**, 3478 (1998).

# The BOREAS Beamline at ALBA – Scientific Opportunities and Recent Technical Developments

*M. Valvidares[1], P. Gargiani[1], J. Herrero-Martín[1]*, D. Pérez-Salinas[1], J. Llobet[1], A. Carballedo[1], A. Enrique[1], X. Fariña[1], J. Moldes[1], J. R. Molinero[1], X. Serra[1], N. Serra[1]*

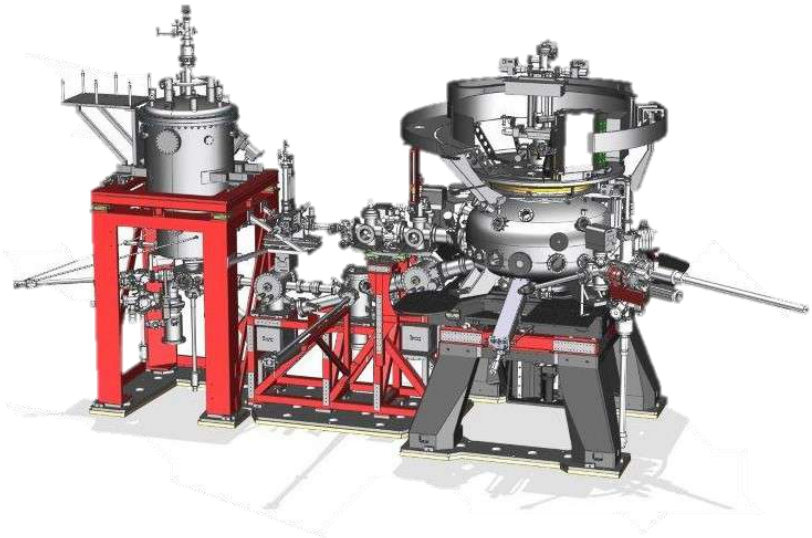
[1] *ALBA Synchrotron, Carrer de la Llum 2-26, 08290 Cerdanyola del Vallès, Spain*

*jherrero@cells.es

BL29- BOREAS at the ALBA Synchrotron Light Facility is a beamline for soft x-ray polarization-dependent spectroscopic investigations on advanced basic and applied materials [1]. It counts on two endstations: MaReS – designed for RMXS and reflectometry but nowadays also offering GISAXS, holography and CDI capabilities – and HECTOR – devoted to XAS, XLD and XMCD studies [2].

An APPLE II undulator and a monochromator based on three variable line spacing plane gratings generate a high photon flux from about 0.1 up to 4 keV with a sharp energy resolution and full polarization control. The beamline provides a rather broad photon energy range (approx. 0.1 to 4 keV) with full polarization control and variable focusing for experiments at the two endstations. Both MaReS and HECTOR implement high magnetic field (6 and 2 Tesla max., respectively) and low temperature capabilities (4-370 K and 18-400 K, respectively). This allows atomic-level investigations on magnetic and electronic properties of quantum materials, among which ferromagnets, antiferromagnets, superconductors, ferroelectrics, multiferroics or topological insulators. The samples under study may have the form of isolated atoms, nanoparticles and powders, organic molecules, Van-der-Waals layers, thin films or bulk specimens.

The beamline being operative since mid 2012, along the past few years we have largely enhanced the instrumentation available with notably: i) a state-of-the-art Scanning Tunneling Microscope, ii) a full surface preparation technique chamber with Molecular Beam Evaporation, LEED/Auger, ion sputtering and high temperature annealing, iii) an Argon-filled Glove box. All capabilities are now completed, fully commissioned and interconnected to the x-ray endstations via an iv) ultra-high vacuum radial distribution chamber.



## References

[1] A. Barla, J. Nicolás, D. Coco, M. Valvidares, J. Herrero-Martín, P. Gargiani, J. Moldes, C. Ruget, E. Pellegrin, S. Ferrer, *J. Synchr. Rad.*. **23** (2016)

[2] https://www.cells.es/en/beamlines/bl29-boreas

# Broadband Femtosecond Ellipsometry for The Study of Magnetic Materials

*Martin Zahradník[1]*, Shirly Espinoza[1], Mateusz Rebarz[1], Martin Veis[2], Jakob Andreasson[1]*

[1] *ELI Beamlines Facility, The Extreme Light Infrastructure ERIC, Za Radnicí 835, 25241 Dolní Břežany, Czech Republic*

[2] *Charles University, Faculty of Mathematics and Physics, Ke Karlovu 3, Prague 2 12116, Czech Republic*

*martin.zahradnik@eli-beams.eu

Optical techniques that allow the investigation of ultrafast magnetization dynamics are typically using the magneto-optical (MO) effects, such as MO Kerr and Faraday effect. These techniques are usually limited to single-wavelength operation, however, a development of a broadband femtosecond Faraday effect spectrometer has recently also been demonstrated [1]. Acquisition of the transient response in a broad spectral region combines advantages of ultrafast pump-probe experiments with conventional spectroscopy. Here, we would like to present a broadband femtosecond instrument, which is using optical ellipsometry [2] instead of the MO effects. The instrument was developed at ELI Beamlines and proved to be a powerful tool in investigation of ultrafast properties in magnetic materials.

ELI Beamlines is a part of the pan-European research network of user facilities ELI-ERIC, providing the international community with leading high-power, high-intensity, and short-pulsed laser systems. The presented broadband femtosecond pump-probe ellipsometry has recently attracted considerable attention of the user community. Using a femtosecond driving laser and a whitelight supercontinuum, the technique allows acquisition of the dynamic evolution of optical properties in a spectral range from 350 to 750 nm, with time resolution of 100 fs, and with the possibility of tuning the pump excitation between 250 nm and 2.5 µm.

Similar to conventional ellipsometry, the ultra-fast beamline is especially suitable for, though not limited to, characterization of thin films of solid state materials. The wide application potential and versatility of the presented technique was demonstrated in a number of various material systems. After initial experiments on bulk semiconductors (Ge, Si, InP), mapping the dynamic evolution of critical points [3], the instrument was further used for probing the dynamics of hot charge carriers [4] and birefrigence in thin films of ZnO [5]. Most recently we reported on photo-induced insulator-to-metal transition in thin films of cobaltites [6], or ultra-fast changes in absorption onset of zincblende GaN.

Here we would like to emphasize that the instrument can successfully serve also for investigation of ultrafast properties in magnetic materials. We demonstrate it on the example of Ni-Mn-Ga, a ferromagnetic shape memory alloy, whose spectral transient response was systematically studied with respect to its martensitic transformation.

## Acknowledgements

We acknowledge the support of the LM2018141, MEYS – Large research infrastructure project ELI Beamlines, and the ADONIS (Grant No. CZ.02.1.01/0.0/0.0/16-206 019/0000789) project from the European Regional Development Fund.

## References

[1] J. Sutcliffe, J. O. Johansson, *Rev. Sci. Instrum.* **92** (2021) 113001.
[2] S. Richter, et al., *Rev. Sci. Instrum.* **92** (2021) 033104.
[3] S. Espinoza, et al., *Appl. Phys. Lett.* **115** (2019) 052105.
[4] S. Richter, et al., *New J. Phys.* **22** (2020) 083066.
[5] O. Herrfurth, et al., *Phys. Rev. Research* **3** (2021) 013246.
[6] M. Zahradník, et al., *Phys. Rev. B* **105** (2022) 235113.

# Magnetization experiment to help the understanding the origin of Mars moons

*Sergio Fernández Romero [(*1)], M. A. Rivero Rodriguez [(1)], Alberto López Escolano [(1)], Marcos Gutiérrez del Olmo [(2)], Pascal Rosenblatt [(3)], Benoit Langlais [(3)] and Marina Díaz Michelena [(1)]*

[1] *Space Magnetism Area of Space Systems Subdirectorate, INTA, Madrid, Spain*
[2] *Metallic Materials Area of Aeronautical Systems Subdirectorate, INTA, Madrid, Spain*
[3] *Laboratoire de Planétologie et Géosciences, UMR 6112, CNRS, Nantes Université - Université d'Angers - Le Mans Université, Nantes, France*

* fdezrs@inta.es

Space magnetism is a discipline that seeks to answer big questions about the past and present of different planets, satellites and other bodies. In the case of rocky bodies, the magnetic bearings are capable to record the magnetic conditions that existed when they were created. This property allows us to see through a window in time and compare present and past magnetic field levels to get answers about the geological evolution of these bodies.

The purpose of this paper is to present an experiment aimed at knowing the possible magnetization of a rocky body during the cooling that it suffered in the process of its formation. We have focused on the possible origin of the satellites of Mars for which two hypotheses are considered asteroid capture and giant impact.

A giant impact on Mars would be capable of detaching a significant amount of material from the planet itself. Upon impact, material at a temperature of more than 1500 ºC would be ejected at a distance of several radius of the planet. This material would cool down and follow an elliptical trajectory that could cover areas located several times the radius of the planet.

For such scenario, if the impact took place in the former stage of the planet the magnetic field of Mars has been published to be approximately twice that of Earth [1]. In that case, the magnetic field at distances 5.5 times the radius of Mars would be about 600 nT, increasing and decreasing as it moves in the elliptical orbit [2].

To simulate this scenario, some samples of volcanic rock are going to be heated to 1000 ºC in an industrial furnace GALTERMIC Model ATMD200P1200. The supply of Ar gas is necessary throughout the process and it is carried out inside the furnace by means of a flowmeter. The temperature of the experiment as well as the duration of the experiment are controlled by a programmer. Afterwards, during the cooling, the rock will be exposed to a rotating magnetic field within a set of triaxial Helmholtz coils that simulates the scenario. Three different power supplies will be used to compensate the Earth's magnetic field in DC and, simultaneously, introduce a 10 Hz signal that simulates the movement of the body in the elliptical orbit. The magnetic field levels are calibrated prior to the experiment using a fluxgate magnetometer and the rock temperature will be monitored with a type K thermocouple (TT-K-1-0041) of 300 mm in length and 1.5 mm in diameter.

The experiments will be carried out during the months of March and April 2023. And the final measurements will include an in-depth analysis of the magnetic properties, both in bulk and by layers, that the volcanic rocks have acquired after cooling, being exposed to environmental conditions that would simulate the possible origin of a satellite on Mars.

## Acknowledgements

This work has been funded by the Spanish Programme for Research, Development and Innovation under the grant of reference PID2020-119208RB-I00: MINOTAUR.

## References

[1] Mittelholz, et al., 2020, JGR Planets, DOI: 10.1029/2020JE006505
[2] Wei Wang, et al., 2017, Advances in Space Research, DOI: 10.1016/j.asr.2017.05.015.

# Magnetic Domain Wall Ferromagnetic Resonance Confirmed By Magnetotransport Measurements And Kerr Microscopy

*Laura Fernández-García[1]*, Sandra Ruiz Gómez[2], Rubén Guerrero[3], José Luis Prieto[4], Claudio Aroca[4], Manuel Muñoz[1]*

[1] *Consejo Superior de Investigaciones Científicas - Instituto de Tecnologías Físicas y de la Información (CSIC – ITEFI), Serrano Str. 144, Madrid, Spain*

[2] *Max Planck Institute for Chemical Physics of Solids, Nöthnitzer Str. 40, Dresden, Germany*

[3] *Instituto Madrileño de Estudios Avanzados en Nanociencia (IMDEA-Nanociencia), Faraday Str. 9, Madrid, Spain*

[4] *Instituto de Sistemas Optoelectrónicos y Microtecnología - Universidad Politécnica de Madrid (ISOM-UPM), Ciudad Universitaria, Madrid, Spain*

manuel.munoz@csic.es

In spintronic applications, the generation and manipulation of magnetic domain walls have been an important research field since its proposal as the key element in magnetic data storage structures [1]. Nowadays, these systems are studied, not only for their information storage capabilities, but for its potential application to process such information at a small computational cost [2, 3], and for their ability to generate microwave signals [4]. In this work we study the ferromagnetic resonance of a permalloy nanostrip with a gold microantenna sputtered on top, measured by the stripline ferromagnetic resonance tehcnique [5]. The behavior of the microstrip resonance with the radiofrequency stimulus and the applied magnetic field fits the Kittel's equation. Additionally, a new resonance is observed between 4 Oe and 9 Oe for an applied radiofrequency signal between 2.0 GHz to 3.0 GHz. Throughout magnetoresistance measurements, this new resonance at low magnetic fields was found to correspond to the formation and pinning of a magnetic domain wall. The presence of the domain wall was confirmed by Kerr microscopy. Our results show a distinctive RF emission caused by the DW whose frequency increases linearly with the external field, which can be very important for applications in computing and communications.

**Acknowledgements**

K-space Neural computation with magnEtic exciTations H2020-FETOPEN-2018-2020
Ingeniería de Deformación de Ondas de Espín PID2020-117024GB-C44

## References

[1] S. S. P. Parkin, M. Hayashi, L. Thomas, *Sci.* **320** (2008) 190.
[2] F. Riente, G. Turvani, M. Vacca, M. Graziano, *IEEE Trans. Emerg. Topics Comput.* **10** (2022) 1216.
[3] S. A. Siddiqui, S. Dutta, A. Tang, L. Liu, C. A. Ross, M. A. Baldo, *Nano Lett.* **20** (2020) 1033.
[4] S. Dhull, A. Nisar, B. K. Kaushik, *Proc.* **11470** (2020) 114703Y-1.
[5] S. S. Kalarickal, P. Krivosik, M. Wu, C. E. Patton, M. L. Schneider, P. Kabos, T. J. Silva, J. P. Nibarger, *J. Appl. Phys.* **99** (2006) 093909.

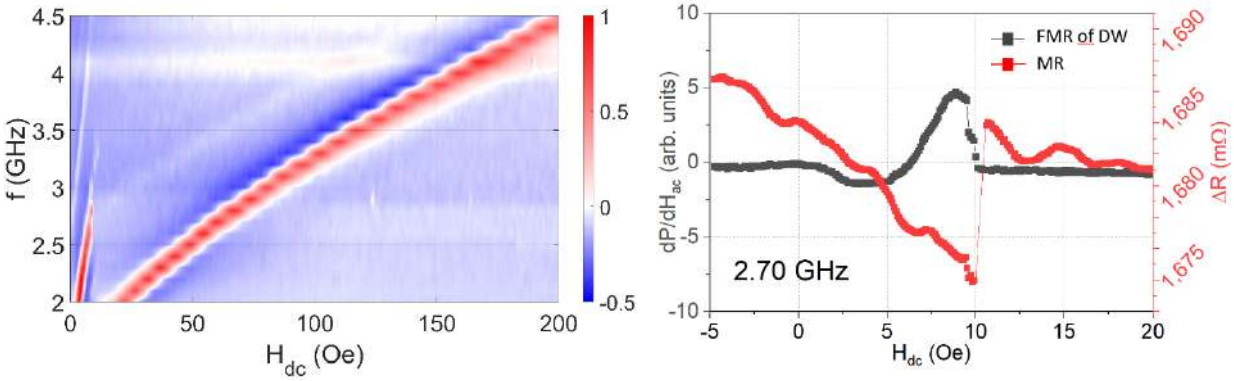



Figure: Normalized measurement of the derivative of the ferromagnetic resonance signal from 2.00 GHz to 4.50 GHz at 0 dBm and the shape anisotropy axis of the microstrip oriented at 0º with the applied magnetic field (left). Derivative of ferromagnetic resonance of the domain wall in grey line, and simultaneous magnetoresistance measurements in red representation at 2.70 GHz (right).

# The MagneDyn beamline at the FERMI free electron laser

*Marco Malvestuto,[1, 2, 3, *] Antonio Caretta,[1] Richa Bhardwaj,[1] Simone Laterza,[1, 3] Fulvio Parmigiani,1, 3 Alessandro Gessini,[1] Matteo Zamolo,[1] Fabio Galassi,[1] Rudi Sergo,[1] Giuseppe Cautero,[1] Miltcho B. Danailov,[1] Alexander Demidovic,[1] Paolo Sigalotti,[1] Marco Lonza,[1] Roberto Borghes,[1] Adriano Contillo,[1] Alberto Simoncig,[1] Michele Manfredda,[1] Lorenzo Raimondi[1] and Marco Zangrando[1, 2]*

*1)Elettra-Sincrotrone Trieste S.C.p.A. Strada Statale 14 - km 163.5 in AREA Science Park 34149 Basovizza, Trieste, Italy*

*2)CNR-IOM Istituto Officina Dei Materiali, Consiglio Nazionale Delle Ricerche, Area Science Park, Strada Statale 14, km 163.5 Trieste, Basovizza, 34149, Italy*

*3)Department of Physics, University of Trieste, Via A. Valerio 2, 34127 Trieste, Italy*

*marco.malvestuto@elettra.eu

The scope of this communication is to outline the main marks and performances of the MagneDyn beamline [1] at the Free Electron Laser FERMI which was designed and built to perform ultrafast magnetodynamic studies in solids. Open to users since 2019, MagneDyn operates with variable circular and linear polarized femtosecond pulses delivered by the externally laser-seeded FERMI free-electron laser (FEL). The very high degree of polarization, the high pulse-to-pulse stability, and the photon energy tunability in the 50-300 eV range allow to perform advanced time-resolved magnetic dichroic experiments at the K-edge of light elements, e.g. carbon and at the M- and N-edge of the 3d-transition-metals and rare earth elements, respectively. To this end two experimental end-stations are available. The first is equipped with an in-situ dedicated electromagnet, a cryostat, and an extreme ultraviolet (EUV) Wollaston-like polarimeter[2,3]. The second, designed for carry-in users instruments, hosts also a spectrometer for pump-probe resonant X-ray emission and inelastic spectroscopy experiments with a sub-eV energy resolution. A Kirkpatrick–Baez active optics system provides a minimum focus of ~20x20 $\mu m^2$ FWHM at the sample. A pump laser setup, synchronized with the FEL-laser seeding system, delivers sub-picosecond pulses with photon energies ranging from the mid-IR to near-UV for optical pump-FEL probe experiments with a minimal pump-probe jitter of few femtoseconds. The overall combination of these features renders MagneDyn a unique state-of-the-art tool for studying ultrafast magnetic phenomena in solids.

## References

[1] Malvestuto, M. *et al.* The MagneDyn beamline at the FERMI free electron laser. *Rev Sci Instrum* **93**, 115109 (2022).

[2] Caretta, A. *et al.* A novel free-electron laser single-pulse Wollaston polarimeter for magneto-dynamical studies. *Struct Dynam-us* **8**, 034304 (2021).

[3] Laterza, S. *et al.* All-optical spin injection in silicon investigated by element-specific time-resolved Kerr effect. *Optica* **9**, 1333 (2022).

# 13th JEMS

## Joint European Magnetic Symposia

27th August to 1st September 2023

MADRID