Proceedings of the European Conference "Physics of Magnetism '99", Poznań 1999

MAGNETIC BEHAVIOUR AND ELECTRONIC STRUCTURE OF THE R_2PdSi_3 (R = Ce, Nd, Tb–Er) COMPOUNDS

A. Szytuła[a], M. Hofmann[b], B. Penc[a], M. Ślaski[c], Subham Majumdar[d], E.V. Sampathkumaran[d] and A. Zygmunt[e]

[a]Institute of Physics, Jagiellonian University, Reymonta 4, 30-059 Kraków, Poland
[b]Berlin Neutron Scattering Centre, Hahn–Meitner Institute
14109 Berlin-Wannsee, Germany
[c]School of Physics and Space Research, University of Birmingham, Birmingham, UK
[d]Tata Institute of Fundamental Research
Homi Bhabha Road, Colaba, Mumbai 400005, India
[e]W. Trzebiatowski Institute of Low Temperatures and Structural Research
Polish Academy of Sciences, Okólna 2, 50-950 Wrocław, Poland

The magnetic properties including the magnetic structure were determined by magnetometric and neutron diffraction measurements of the polycrystalline samples of R_2PdSi_3 (R = Ce, Nd, Tb–Er). In Ce_2PdSi_3 the Ce moments order below 2.5 K. The Nd magnetic moments in Nd_2PdSi_3 exhibit ferromagnetic ordering below 17 K. In Tb_2PdSi_3, a complex situation is observed: in the temperature range from 1.5 K to (T_N = 25 K), there is a coexistence of a ferromagnetic spiral (about 75% of the magnetic moment) and a spin-glass (about 25%). For the Dy, Ho, and Er compounds, below T_N (7.5, 8, and 7 K respectively), sinusoidally modulated structures are observed. The valence band and the core level photoemissions of 4*d* of rare-earth metal, 3*d* Pd, 2*s* and 2*p* Si were investigated by X-ray photoemission spectroscopy measurements.

PACS numbers: 71.20.–b, 75.30.Cr, 75.30.Kz, 79.60.–i

1. Introduction

The ternary rare-earth (R) compounds [1] have attracted considerable attention [1]. Recently, we have been paying special attention to the R_2PdSi_3 [2]. X-ray diffraction data [2] indicate that these compounds with R = Pr, Nd, Gd, Tb, Dy, Ho, Er, Tm, and Y crystallize in AlB_2-derived hexagonal structure. Pr_2PdSi_3 does not show any magnetic ordering temperature down to 4.2 K. Nd_2PdSi_3 is ferromagnetic with a Curie temperature of 16 K. The heavy rare-earth compounds (R = Gd–Er) show magnetic ordering temperatures in the region between 8 and

21 K [2]. New magnetic data [3–6] reveal small anomaly in the temperature dependence of the magnetic susceptibility of Ce_2PdSi_3 near 7 K [3], two ordering temperatures at 40 and 10 K for Eu_2PdSi_3 [4], one at 21 K for Gd_2PdSi_3 [5], 23 for Tb_2PdSi_3 and 8 K for Dy_2PdSi_3 [6]. To resolve these data we performed new magnetic, neutron diffraction and photoemission experiments for the R_2PdSi_3 samples (R = Ce, Nd, Tb, Dy, Ho, and Er). With these data magnetic and crystal structures and electronic structure of these compounds are determined.

2. Experiment and results

The preparation of the samples were described in [5]. The X-ray diffraction patterns confirm that the compounds crystallize in a AlB_2-derived hexagonal structure. The magnetic susceptibility and magnetization of polycrystalline samples were determined with a Cryogenics S100 SQUID susceptometer that operates in magnetic fields up to 1000 Oe and with an Oxford Instrument VSM 12 T magnetometer in magnetic fields up to 120 kOe between 4.2 and 300 K. For Ce_2PdSi_3 compound additional measurements were performed at low temperatures down to 1.8 K. For all the compounds the neutron-diffraction measurements were carried out in the BER II reactor at the Hahn–Meitner Institut in Berlin. The neutron wavelength was 2.44 Å. The observed neutron-diffraction intensities were processed using the FULLPROF program [7].

Photoemission measurements were performed using commercial LHS10 SPECS spectrometer with hemispherical energy analyzer using following radiation sources: non-monochromatized Mg K_α ($h\nu = 1253.6$ eV), monochromatized and non-monochromatized Al K_α ($h\nu = 1486.6$ eV). The total energy resolution was about 0.8 eV for Ag $3d$ peaks with Mg K_α radiation and 0.55 eV for monochromatized Al K_α radiation. The spectrometer was calibrated using the Cu $2p$, Ag $3d$ and Au $4f$ core-level photoemission spectra. Measurements were made at room temperature in high vacuum (2×10^{-9} mbar).

The temperature dependences of the magnetization and magnetic susceptibility for R_2PdSi_3 compounds at low temperatures give the following results:

— for Ce_2PdSi_3 below $T = 2.5$ K a small maximum,

— for Nd_2PdSi_3 ferromagnetic ordering below 16 K,

— for Tb_2PdSi_3 a sharp peak at 23 K and a second broad peak between 10 and 18 K,

— for other compounds at 7.5 K (R = Dy) and at 8 K (R = Ho and Er) a maximum is observed and shows antiferromagnetic ordering.

The neutron diffraction data of R_2PdSi_3 compounds, at low temperatures (below T_N) shows the presence of the following magnetic structures:

— sine modulated with the propagation vector $\boldsymbol{k} = (0, 0, 0.208(1))$ for R = Ce. The Ce magnetic moments equal to 1.14(10) μ_B lie in the a–a plane,

— collinear ferromagnetic with the Nd magnetic moment equal to 1.97(9) μ_B parallel to the c-axis for R = Nd,

— for Tb_2PdSi_3 compound, the majority nearly 75% ferromagnetic spiral with the propagation vector $\boldsymbol{k} = (0.486(1), 0, 0.174(2))$ and spiral axis parallel to the c-axis. Nearly 25% of the Tb magnetic moments form a spin glass.

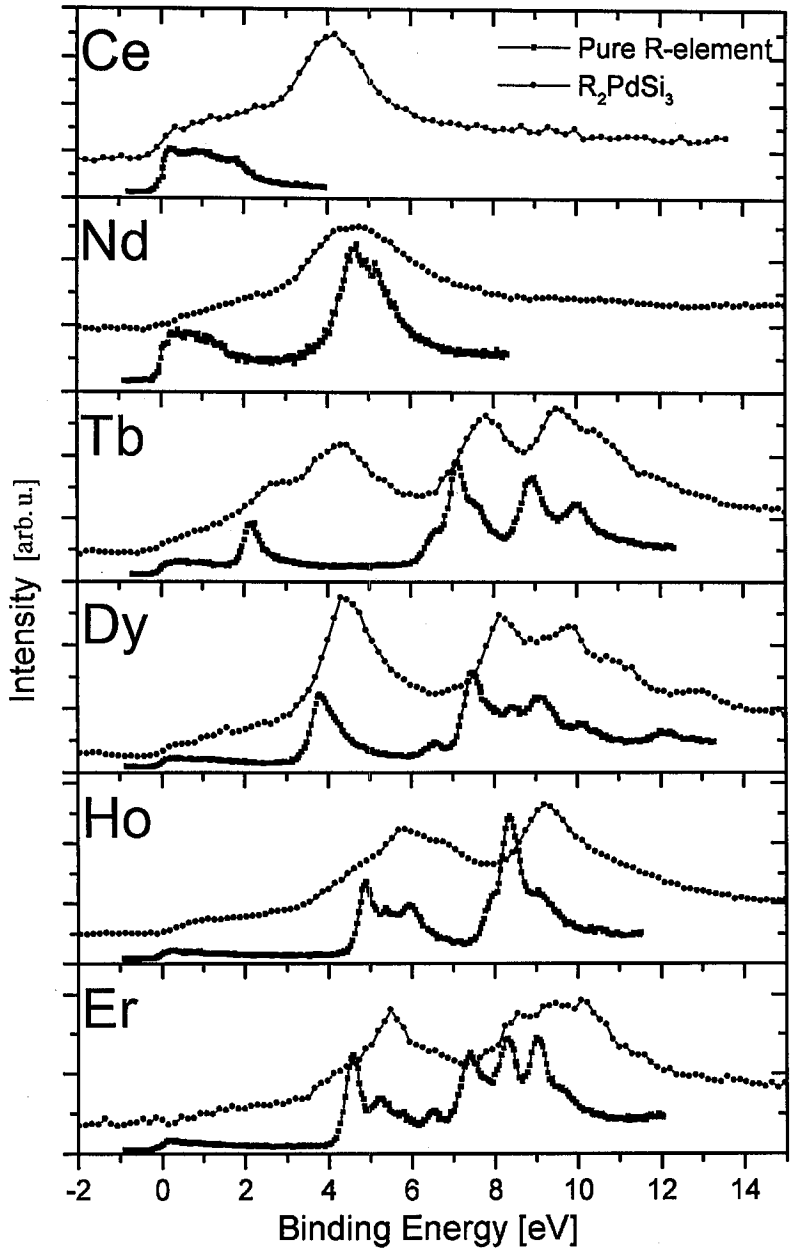

Fig. 1. XPS valence band spectra for R_2PdSi_3 compounds and adequate spectra for metallic rare earths from Ref. [8].

For the other compounds the sine modulated structures are observed. In Dy_2PdSi_3 magnetic order is described by the propagation vector $\boldsymbol{k} = (0.174(8), 0.1618(7), 0)$ and the magnetic moment equal to $4.50(17)\ \mu_B$ parallel to the c-axis.

In Ho_2PdSi_3 magnetic structure with $\boldsymbol{k} = (0.1387(2), 0.0066(3), 0)$ with the Ho magnetic moment equal to 8.27(10) μ_B and parallel to the c-axis. In Er_2PdSi_3 magnetic order is described by the two propagation vectors: $\boldsymbol{k}_1 = (0.1434(2), 0, 0)$ and $\boldsymbol{k}_2 = (0.112(2), 0.118(2), 0)$. Er magnetic moment is parallel to the c-axis.

The X-ray photoemission (XPS) spectra of valence bands (see Fig. 1) indicate that these are dominated by contribution of the rare-earth $4f$ states. The Pd $4d$ state near 4 eV are observed (see XPS band for Ce_2PdSi_3) while Si states near 9 eV are not observed. Except the cerium compound, for other compounds the intensity on the Fermi level is small. The peak near the Fermi level in Ce_2PdSi_3 compound is connected with the Ce $4f$ band.

Analysis of the core levels Si $2s$, Si $2p_{3/2}$, Si $2p_{1/2}$, Pd $3d_{5/2}$, Pd $3d_{3/2}$, and rare earth $4d$ give the following results:

— the energy of Si $2s$, Si $2p_{1/2}$, and Si $2p_{3/2}$ levels does not change for all compounds except Ce_2PdSi_3, which is small on 0.6 eV. Also the value of $\Delta E = E(2p_{1/2}) - E(2p_{3/2})$ for cerium compound is smaller than other,

— the values of $E(\text{Pd } 3d_{5/2})$ and $E(\text{Pd } 3d_{3/2})$ for Ce_2PdSi_3 are smaller than those for other compounds, while the values of $\Delta E = E(\text{Pd } 3d_{3/2}) - E(\text{Pd } 3d_{5/2})$ are constant for all compounds,

— we observed the wide-spread multiplet structure of $4d$ XPS in all compounds. The observed for all compounds multiplet structure is similar to the clean rare-earth metals, and we concluded that the rare-earth ions keep the 3+ valency in the measured compounds. The least results are in good agreement with the results of the magnetic measurements at high temperatures, which give the values of the effective magnetic moment near to the R^{3+} ion values.

References

[1] See, A. Szytuła, in: *Handbook of Magnetic Materials*, Vol. 6, Ed. K.H.J. Buschow, Elsevier, Amsterdam 1991, p. 85.

[2] P.A. Kotsanidis, J.K. Yakinthos, E. Gamari-Seale, *J. Magn. Magn. Mater.* **87**, 199 (1990).

[3] R. Mallik, E.V. Sampathkumaran, *J. Magn. Magn. Mater.* **164**, L13 (1996).

[4] R. Mallik, E.V. Sampathkumaran, M. Strecker, G. Wortmann, P.L. Paulose, Y. Ueda, *J. Magn. Magn. Mater.* **185**, L135 (1998).

[5] R. Mallik, E.V. Sampathkumaran, M. Strecker, G. Wortmann, *Europhys. Lett.* **41**, 315 (1998); R. Mallik, E.V. Sampathkumaran, P.L. Paulose, H. Sugawara, H. Sato, *Pramana J. Phys. (India)* **51**, 505 (1998).

[6] R. Mallik, E.V. Sampathkumaran, P.L. Paulose, *Solid State Commun.* **106**, 169 (1998).

[7] J. Rodriguez-Carvajal, *Physica B* **192**, 55 (1993).

[8] P.A. Cox, J.K. Lang, Y. Baer, *J. Phys. F, Metal Phys.* **11**, 113 (1981).