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

MAGNETIC ORDERING OF $RAgSb_2$ COMPOUNDS

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The magnetic structures of the polycrystalline samples of $RAgSb_2$ (R = Ce, Pr, Nd, Tb–Tm) are determined. All compounds crystallize in the tetragonal $ZrCuSi_2$-type of crystal structure and except $CeAgSb_2$ they are antiferromagnets with a Néel temperature of 11 K for R = Tb and 2 K for Tm. $CeAgSb_2$ below $T_C = 9.5$ K is ferromagnet with the magnetic moment equal to $0.33\mu_B$ and parallel to c-axis. In $NdAgSb_2$ the sine modulated magnetic ordering described by the wave vector $\boldsymbol{k} = (0.5435,0,0)$ is observed. Below T_N a collinear antiferromagnetic structure with the magnetic $(a, a, 2c)$ for R = Pr and $(2a, a, c)$ for R = Tb, Dy, Ho, and Er is observed. For Ho-compound with an increase in temperature the coexistence of two structures is observed: commensurate collinear and incommensurate sine modulated. In $TmAgSb_2$ the coexistence of the two phases is observed at $T = 1.4$ K. With an increase in temperature only the incommensurate phase stays.

PACS numbers: 75.30.Cr, 75.30.Kz, 75.50.Ee

1. Introduction

In the framework of our systematic studies of the magnetic properties of the rare-earth intermetallic compounds we present results of the neutron diffraction measurements of $RAgSb_2$ compounds, where R = Ce–Nd, Tb–Tm in this work. Recently we have reported on the structure and magnetic properties of ternary rare-earth $RAgSb_2$ compounds. X-ray powder diffraction data indicate that these compounds crystallize in the tetragonal $ZrCuSi_2$-type structure (space group P4/nmm) [1]. $RAgSb_2$ compounds, with R = Pr, Nd, Sm, Gd, Dy, Ho, Er, and Tm are antiferromagnets with the Néel temperatures lower than 13 K [1]. $CeAgSb_2$ is a ferromagnet with $T_C = 12$ [1] or 10 K [2]. In this work we present the results of the neutron diffraction measurements undertaken in order to determine the crystal and magnetic structures of $RAgSb_2$ compounds (R = Ce, Pr, Nd, Tb–Tm).

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2. Experimental and results

Measurements were carried out on polycrystalline samples, as in Ref. [4]. The examination of X-ray pattern confirmed the main tetragonal phase with a small concentration of other phases. The neutron powder diffraction experiments were performed at Saclay (Orphée reactor, Laboratoire Léon Brillouin) with "G4.1" 2-axis diffractometer $\lambda = 2.4249$ Å. The neutron data have been analysed via the Rietveld technique, using the FULLPROF [3] program. The neutron diffraction data above the Néel temperature indicate that these compounds crystallize in the $ZrCuSi_2$-type structure [4]. In the diffraction pattern of $CeAgSb_2$ at $T = 1.4$ K only two small intensity peaks of the magnetic origin are observed. The presence of these peaks, indexed by the crystal unit cell, indicate the ferromagnetic ordering with the magnetic moment equal to $0.33(12)\mu_B$ and parallel to the c-axis. The obtained value of the Ce magnetic moment agrees with the magnetic data [2]. The temperature dependence of the magnetic peak intensity of the reflection gives a Néel temperature of 9.5 K. In the diffraction pattern of $PrAgSb_2$ measured at $T = 1.4$ K the additional peaks of the magnetic origin are observed. These peaks are indexed in the $(a, a, 2c)$ unit cell. Consequently a simple antiferromagnetic scheme can be adopted, in which the magnetic moments localized on the Pr^{3+} ions form ferromagnetic basal planes with the following sequence $++--$ along c-axis. The Pr^{3+} magnetic moment equal to $2.95(10)\mu_B$ forms the angle of 45° with c-axis (see Fig. 1a). A different distribution of the magnetic peaks is observed for $NdAgSb_2$ at $T = 1.4$ K. All the observed magnetic Bragg peaks can be indexed as satellite pairs associated with $h+k+l = 2n$ and the wave vector $k = (0.5435, 0, 0)$. The Nd magnetic moments equal to $2.26(10)\mu_B$ and parallel to the b-axis form the sine modulated structure (see Fig. 1b). The temperature dependence of the magnetic peak intensities gives a Néel-temperature of 5.3 K for $PrAgSb_2$ and 3.2 K for $NdAgSb_2$. The magnetic peaks on the patterns for $TbAgSb_2$, $DyAgSb_2$, $HoAgSb_2$, and $ErAgSb_2$ are indexed in the $(2a, a, c)$ unit-cell. Consequently, a simple antiferromagnetic ordering scheme can be adopted, in which the magnetic moments localized on the R^{3+} ions form the following magnetic structure. In the magnetic unit cell there are four rare-earth moments at the following sites: S_1 $(\frac{1}{8}\ \frac{1}{4}\ z)$, S_2 $(\frac{3}{8}\ \frac{3}{4}\ \bar{z})$, S_3 $(\frac{5}{8}\ \frac{1}{4}\ z)$, and S_4 $(\frac{7}{8}\ \frac{3}{4}\ \bar{z})$. The four-collinear magnetic structure is possible: $A = S_1 - S_2 - S_3 + S_4$, $C = S_1 + S_2 - S_3 - S_4$, and $G = S_1 - S_2 + S_3 - S_4$. The best fit with the experimental results is obtained for the A model for all three compounds. The magnetic moments of Tb atoms equal to $7.55(15)\mu_B$ are parallel to b-axis ($R_M = 5.0\%$) (see Fig. 1c), the Dy moment equal to $8.02(18)\mu_B$ is parallel to b-axis ($R_M = 10.9\%$), the Ho magnetic moment equal to $6.52(12)\mu_B$ lies in b–c plane and forms the angle $\varphi = 13.50°$ with b-axis ($R_M = 6.15\%$), whereas the Er moment equal to $7.45(15)\mu_B$ is parallel to c-axis ($R_M = 5.8$ %) (see Fig. 1d). The temperature dependence of the magnetic peak intensities gives a Néel temperature equal to 11 K for $TbAgSb_2$, 9.7 K for $DyAgSb_2$, 5.5 K for $HoAgSb_2$, and 3.9 K for $ErAgSb_2$. For $HoAgSb_2$ with an increase in temperature additional peaks appear. These peaks are indexed by the propagation vector $k = (k_x, 0, k_z)$ and corresponding to the sine modulated structure with the magnetic moment parallel to b-axis (see Fig. 1e). With an in-

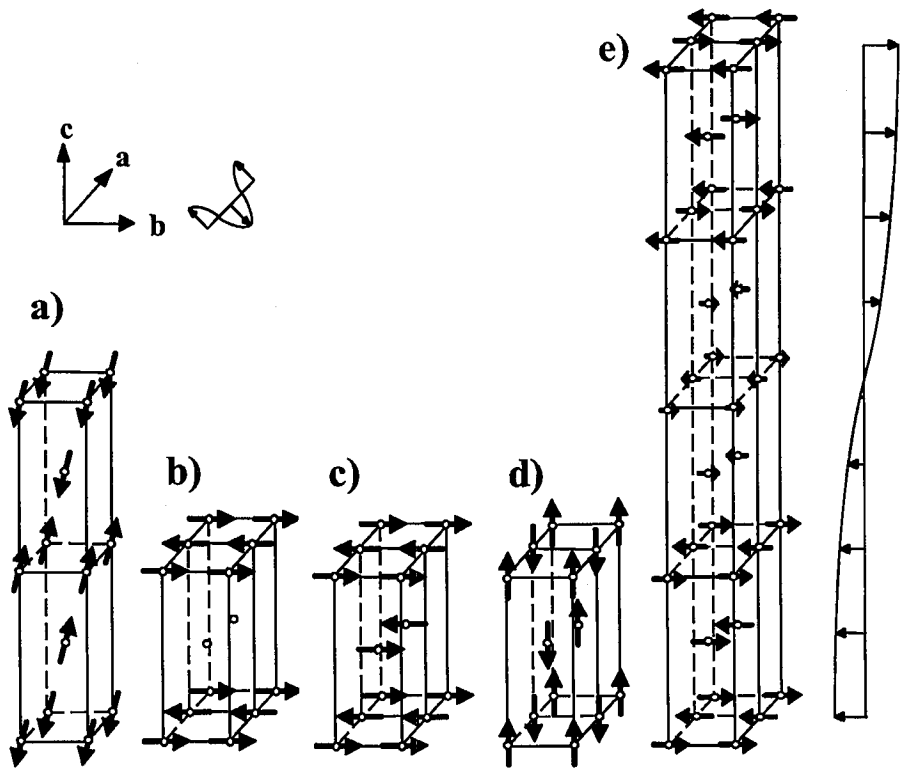

Fig. 1. Magnetic structures of (a) $PrAgSb_2$, (b) $NdAgSb_2$, (c) $TbAgSb_2$, (e) $ErAgSb_2$, and (d) $HoAgSb_2$ (modulated phase).

crease in temperature the decrease in intensities of the peaks corresponding to the collinear structure and the increase in the sine modulated one are observed. The peaks observed at $T = 1.4$ K on the neutron diffraction pattern of $TmAgSb_2$ are indexed by two wave vectors $\boldsymbol{k} = [0.5, 0, 0]$ and $\boldsymbol{k} = [0.5, 0, 0.084]$. The first group of the peaks corresponding to the collinear antiferromagnetic structure whereas the second to the sine modulated one. With the increase in temperature only the peaks connected with the sine modulated structure are observed. For both phases the magnetic moments are parallel to c-axis. The temperature dependence of the peak intensity of $000^{\pm}$ reflection gives a Néel temperature of 2 K. Near the Néel temperature in the temperature range of 1.8–2.2 K the diffusion peaks connected with the short range ordering near $2\theta = 16°$ are observed.

3. Discussion

The stability of the observed magnetic ordering scheme can thus be considered as being due to the interactions via conduction electrons (RKKY model) and the magnetocrystalline anisotropy caused by the influence of the crystalline electric field (CEF) of the $4f$ electrons. RKKY-type exchange interactions favour a modulated magnetic ordering while the magnetocrystalline anisotropy favours a uniaxial magnetic ordering. In the RKKY theory, the critical temperature of the magnetic ordering T_C, T_N is proportional to the de Gennes factor $(g_J - 1)^2 J(J+1)$ [5]. The

compared experimental data of the Curie T_C or Néel T_N temperatures in $RAgSb_2$ compounds with the theoretical prediction give the following conclusions:

– the de Gennes function is not obeyed in compounds containing light rare-earth atoms,

– in the case of heavy rare-earth atoms, the Néel temperatures points follow in principle the de Gennes function.

The second factor which influences the magnetic ordering is the interaction of the crystalline electric fields with the multiple moments of the R atom electrons at a site of a crystal lattice of $4/nmm$ point symmetry. It is given by the Hamiltonian: $H_{CEF} = B_2^0 O_2^0 + B_4^0 O_4^0 + B_4^4 O_4^4 + B_6^0 O_6^0 + B_6^4 O_6^4$, where O_n^m are the Stevens operators, B_n^m are CEF parameters defined by Hutchings [6]. The determined values for RT_2Si_2 compounds, where R is a heavy rare-earth, T is $3d$, $4d$, and $5d$ electrons atoms, indicate that B_2^0 parameter is dominant. At a site of the tetragonal point symmetry the magnetic moment is parallel to c-axis if $B_2^0 < 0$ and it is perpendicular to c-axis if $B_2^0 > 0$ [7]. For RAg_2Si_2 compounds, where R = Tb, Dy, and Ho, the B_2^0 parameters are positive, whereas for R = Er–Yb, they are negative [8]. The determined from the neutron diffraction data direction of the magnetic moments in $RAgSb_2$ compounds indicates the negative parameter for R = Ce, Er, and Tm, and positive for R = Nd, Tb, and Dy. While observing $HoAgSb_2$ and $TmAgSb_2$ compounds, the change of the magnetic structure from collinear at low temperatures to sine modulated near the Néel point is noticed in a large number of rare-earth intermetallic compounds and is interpreted in terms of the realistic mean field model which takes into account the periodic exchange field and CEF effects [9].

Acknowledgements

This work was supported by the Committee for Scientific Research (Poland) within research grant 2 P03B 024 13.

References

[1] O. Sologub, H. Noël, A. Lithe-Jasper, P. Rogl, O. Bodak, *J. Solid State Chem.* **115**, 441 (1995).

[2] Y. Muro, N. Takeda, M. Ishikawa, *J. Alloys Compds.* **257**, 23 (1997).

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

[4] G. André, F. Bourée, M. Kolenda, B. Leśniewska, A. Oleś, A. Szytuła, to be published.

[5] P.G. de Gennes, *J. Phys. Radium* **23**, 510, 630 (1962).

[6] M.T. Hutchings, in: *Solid State Physics*, Eds. F. Seits, D. Turnbull, Vol. 16, Academic Press, New York 1964, p. 227.

[7] A. Szytuła, J. Leciejewicz, in: *Handbook on the Physics and Chemistry of Rare Earths*, Eds. K.A. Gschneider, L. Eyring, Vol. 12, North Holland, Amsterdam 1989, p. 133.

[8] K. Łątka, *Rep. INP*, 1443/PS, Institute of Nuclear Physics, Kraków (Poland) 1989.

[9] D. Gignoux, D. Schmitt, *Phys. Rev. B* **48**, 12682 (1993).