Journal of Magnetism and Magnetic Materials 177–181 (1998) 1147–1148

ELSEVIER

Journal of magnetism and magnetic materials

Low-temperature magnetisation and specific heat in antiferromagnetic rare-earth germanides of the type R_3Ge_4

Y. Janssen[a], E. Brück[a], C.H. de Groot[a], F.E. Kayzel[a], F.R. de Boer[a], K.H.J. Buschow[a,*], O. Oleksyn[b], P. Schobinger-Papamantellos[b]

[a] *Van der Waals-Zeeman Institute, University of Amsterdam, Valckenierstraat 65, 1018 XE Amsterdam, Netherlands*
[b] *Laboratorium für Kristallographie, ETHZ, CH-8092 Zürich, Switzerland*

Abstract

We have studied the magnetic properties of R_3Ge_4 compounds with R = Dy and Er by means of specific heat measurements. Both compounds show specific heat anomalies indicative of a two-step magnetic ordering upon cooling. The results are discussed together with neutron diffraction data.

Keywords: Antiferromagnetism; Specific heat; Magnetic phase transition; Rare-earth germanides

1. Introduction

The compounds of the type R_3Ge_4 (R = Tb, Dy, Ho, Er, Tm) have the orthorhombic Er_4Ge_3 type structure [1]. The structure comprises two R sites (R_1 at 8f and R_2 at 4c) and three Ge sites. The R-atoms form trigonal prisms centred by Ge atoms and stacked along the (shortest) *a* axis. Within the (0, *y*, *z*) plane, adjacent prisms along *b* or *c* are shifted by $x = 1/2$.

The type of magnetic ordering displayed by these isomorphous R_3Ge_4 compounds is of substantial interest because it opens the possibility to follow the influence of the R-component on the competing intrasublattice interactions of the two sites I_{11}, I_{22} and the intersublattice interaction I_{12}, which may lead to complex ordering phenomena.

Recently, we studied the magnetic ordering of the Er_3Ge_4 compound [2]. The two Er-sublattices order simultaneously below $T_N = 7.3$ K with a triangular antiferromagnetic moment arrangement of the two sublattices within the (0, *y*, *z*) plane (space group Sh_{57}^{391}). Adjacent prisms display different chiralities. At 4 K, a reorientation of the Er_1 moments sets in without any change in symmetry. The projection of the magnetic structure along [1 0 0] is displayed in Fig. 1.

A different type of magnetic ordering has been observed in Dy_3Ge_4. This compound displays a two-step antiferromagnetic ordering below $T_N = 19$ K. The two Dy sites (Dy_1 and Dy_2) apparently order independently with two distinct order parameters associated with the same wave vector $q = (0\ 1\ 0)$. Each sublattice has a uniaxial antiferromagnetic moment arrangement but the preferred moment directions of the two sublattices are mutually perpendicular. The Dy_1 moments order below T_N and point along the shortest *a* axis. The Dy_2 moments order only below $T_i = 6$ K and point in the *c* direction (Fig. 1). The magnetic space group is Sh_{11}^{55}.

In the present study, we have used specific heat measurements in order to obtain more information on the magnetic ordering phenomena in these two

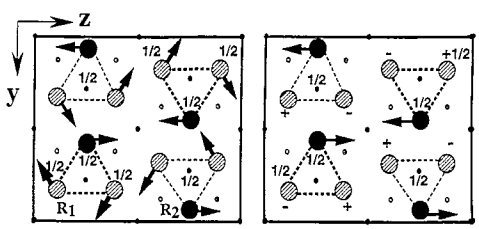

Fig. 1. Schematic representation of the magnetic structure of Dy_3Ge_4 (right) and Er_3Ge_4 (left), showing the canted-moment arrangement of the R moments when viewed along [1 0 0].

* Corresponding author. Tel.: + 31 20 5255714; fax: + 31 20 5255788; e-mail: buschow@phys.uva.nl.

0304-8853/98/$19.00
PII S0304-8853(97)00733-6

compounds. The sample preparation was the same as described before [2]. The specific heat measurements were made on a home-built calorimeter. The samples were mounted with one flat surface on a sapphire plate and fixed with n type apiezon. The temperature was monitored by means of a RuO_2 thermometer below 22 K and by a Pt thermometer at higher temperatures.

2. Experimental results and discussion

The results of the specific heat measurements obtained for Er_3Ge_4 are shown in Fig. 2a. The low-temperature behaviour, reflecting the magnetic ordering phenomena, is shown in more detail in the inset. The sharp peak observed at 6.9 K corresponds closely to the Néel temperature observed by magnetic measurement and neutron diffraction [2]. It was derived from a detailed study of the magnetic intensities of the neutron diffraction diagrams measured at various temperatures below T_N that the magnetic ordering at this temperature involves primarily the Er_1 moments while the magnetic ordering of the Er_2 moments is a more gradual one. It was also derived from the neutron diffraction data that the moments of the Er_2 site are confined to the c direction. The moments of the Er_1 site are oriented in a direction that makes an angle $\phi_b = 31^c$ with the b axis just below T_N. This angle increases slightly with decreasing temperature but does so in a jump-like manner below about 4 K. Simultaneously there is a small jump-like increase in the moment value of the Er_2 site. In the specific heat data this magnetic rearrangement is seen to give rise to an additional contribution appearing as a shoulder at the low-temperature side of the main peak.

Results of specific heat measurements made on Dy_3Ge_4 are shown in Fig. 2b. The low temperature behaviour is shown in more detail in the inset. The high temperature peak corresponds to the onset of magnetic ordering observed by neutron diffraction ($T_N = 19$ K). A more pronounced peak occurs at 5.2 K. A detailed neutron diffraction study of the thermal behaviour of the magnetic intensities has indicated that the magnetic ordering of the two sublattices occurs in two steps [3]. The Dy_1 moments order below T_N and are aligned along the a axis, the moment value increasing smoothly with decreasing temperature. Magnetic ordering of the moments at the Dy_2 site occurs at much lower temperatures, below $T_i = 6$ K. This ordering is reflected in the specific-heat data by the low-temperature peak. The neutron data have also shown that the preferred direction of the Dy_2 sublattice moments is along the c direction and hence is perpendicular to the direction of the Dy_1 moments. The different ordering temperatures and different thermal behaviour of the two sublattices reflect the fact that the moments at the two Dy sites (Dy_1 and Dy_2) order independently with two distinct order parameters associated

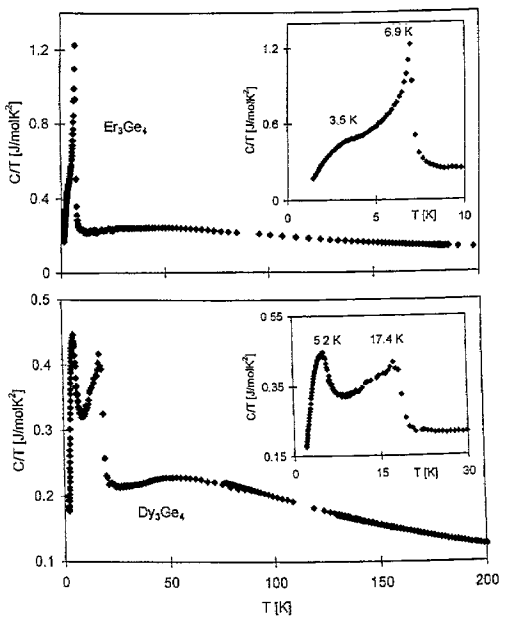

Fig. 2. Temperature dependence of the specific heat C_p/T of Er_3Ge_4 (a) and Dy_3Ge_4 (b). The insets show the low temperature data in an expanded temperature scale.

with the same wave vector $q = (0\ 1\ 0)$. Apparently, the two sublattices in Dy_3Ge_4 are more decoupled than in the case of Er_3Ge_4. At $T_i = 6$ K, an anomaly occurs in the temperature dependence of the lattice constants, a and c. The presence of the separate magnetic ordering of the Dy_2 moments and the fact that it is accompanied by magneto-elastic effects are probably the reason that the low-temperature peak in Dy_3Ge_4 is more pronounced than in Er_3Ge_4.

We have used a Debye function with a Debye temperature of 250 K for estimating the phonon contribution to the specific heat. The magnetic entropy derived from the magnetic contribution to the specific heat and evaluated up to 50 K falls considerably below the corresponding values of $R \ln(2J + 1)$ for Dy_3Ge_4 and Er_3Ge_4. This indicates substantial crystal-field splitting of the $2J + 1$ manifold. A more accurate analysis of crystal-field effects, using specific heat data of Lu_3Ge_4 for estimating the phonon contribution is in progress.

References

[1] O.Ya. Oleksyn, O.I. Bodak, J. Alloys Compounds 210 (1994) 19.
[2] P. Schobinger-Papamantellos, O. Oleksyn, C. Ritter, C.H. de Groot, K.H.J. Buschow, J. Magn. Magn. Mater. (1997) in press.
[3] P. Schobinger-Papamantellos, O. Oleksyn, C. Ritter, K.H.J. Buschow, J. Alloys Compounds 252 (1997) 53.