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Magnetic properties of RCr_6Ge_6 compounds

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Abstract

Compounds of the composition RCr_6Ge_6 have been prepared and investigated by X-ray diffraction. The crystal structure was identified as the MgFe_6Ge_6 type. The magnetic properties of the compounds have been investigated through measurements of the magnetization as a function of applied field and temperature. In the compounds with $\text{R} \equiv \text{Dy}, \text{Ho}, \text{Er}$, no indication of magnetic ordering was found for temperatures down to 5 K. A modest ordering was found in TbCr_6Ge_6 . Magnetization measurements on YCr_6Ge_6 revealed a very small saturation moment of about $0.2 \mu_B$ on the Cr ions. In high magnetic fields there is a tendency towards an anti-parallel arrangement of the Tb and Cr moments.

1. Introduction

Recently, results of various investigations have been reported on the magnetic properties of RT_6Ge_6 compounds with $\text{T} \equiv \text{Mn}, \text{Cr}$ [1–4]. These compounds crystallize in the hexagonal MgFe_6Ge_6 structure (space group $P6/mmm$, Pearson symbol $hP13$), and consist of a layered arrangement of T ions separated by R and Ge ions.

The compounds with $\text{T} \equiv \text{Mn}$ show a wide variety of magnetic phenomena, often dominated by the interplay between the R–Mn and the antiferromagnetic interlayer Mn–Mn interaction [2]. High-magnetic-field measurements revealed a quite substantial R–Mn interaction, comparable in strength with interactions determined earlier on R–Fe and R–Co intermetallics.

The low value of the magnetization at low temperatures and fields, however, raised questions about the nature of the R–R interaction and the magnetic structure of the R moments. As preliminary studies on GdCr_6Ge_6 [3] could not fully clarify the magnetic behaviour of the R moments, we performed further experiments on the other RCr_6Ge_6 compounds with $\text{R} \equiv \text{Y}, \text{Tb}, \text{Dy}, \text{Ho}, \text{Er}$.

2. Experimental details

The polycrystalline samples used in the present investigation were prepared by arc-melting and subsequent annealing at 1073 K for 3 weeks in vacuum. After this treatment, the specimens were checked by X-ray dif-

fraction, and found to be approximately single phase (MgFe_6Ge_6 structure), except for the YCr_6Ge_6 sample in which a small amount of an unknown phase was detected.

Measurements of the magnetization as a function of magnetic fields up to 5.5 T, and as a function of temperature in the range between 1.8 K and 350 K, were performed on a SQUID magnetometer.

The magnetization of TbCr_6Ge_6 was measured as a function of applied field up to 35 T in the high-field installation of the University of Amsterdam [5].

3. Results and discussion

The lattice parameters for the RCr_6Ge_6 compounds, which were deduced from the X-ray data, are listed in Table 1. The decrease of the lattice parameters with increasing R-atom number, due to the lanthanide contraction, is clearly recognizable. The lattice parameters of the Y compound are close to the lattice parameters

TABLE 1. Lattice parameters and effective moments

Compound	<i>a</i> (Å)	<i>c</i> (Å)	μ_{eff} ($\mu_B/\text{f.u.}$)
YCr_6Ge_6	5.167	8.273	–
TbCr_6Ge_6	5.167	8.275	10.2
DyCr_6Ge_6	5.162	8.272	11.0
HoCr_6Ge_6	5.158	8.269	10.7
ErCr_6Ge_6	5.150	8.258	11.1

of the Tb compound, as in many other R-3d intermetallics.

For the RCr_6Ge_6 compounds with $\text{R} \equiv \text{Dy}, \text{Ho}, \text{Er}$, measurements of the magnetization M as a function of temperature give no indication of magnetic ordering between 5 K and 350 K, and suggest a paramagnetic state in this temperature range. The measurement on ErCr_6Ge_6 presented in Fig. 1 illustrates this clearly and is representative for the compounds with $\text{R} \equiv \text{Dy}, \text{Ho}$. Plots of the inverse susceptibility (defined here as $\chi^{-1} = \mu_0 H/M$) as a function of temperature between 5 K and 350 K, and taken at a field of 3 T, confirm the paramagnetic state, as they show Curie-Weiss behaviour even at temperatures far below room temperature (Fig. 2). Values of the effective moments per formula unit deduced from the linear part of the curves pictured in Fig. 2 are given in Table 1. In all cases, the effective moment exceeds the effective free-ion moment of the R-component, although the differences remain within 10%. This could be taken as an indication for a small Cr-moment in the RCr_6Ge_6 compounds.

The possibility of a magnetic moment on the Cr ions led us to the investigation of the YCr_6Ge_6 compound. As Y is non-magnetic, this compound offers the possibility of a more detailed study of the Cr moments. Figure 3 shows the magnetization as a function of temperature between 1.8 K and 55 K. Even down to 1.8 K there is no indication for magnetic ordering. The inverse susceptibility *vs.* temperature ($\chi^{-1} = \mu_0 H/M$) does not show Curie-Weiss behaviour at low temperatures, and tends to become linear only at temperatures above 200 K (see Fig. 4). Due to the presence of a small amount of a second phase in the sample used in the present investigation, the true origin of this non-Curie-Weiss behaviour is not clear. In Fig. 5 measurements of the magnetization as a function of applied fields up to 5.5 T are presented. The magnetic isotherms were obtained at 1.8 K and 5 K. Both curves in Fig. 5 show a tendency towards saturation at low fields already. This observation is quite surprising in view of the low saturation magnetization, and the fact that Fig.

Fig. 1. Magnetization *vs.* temperature for ErCr_6Ge_6 ($B = 3$ T).

Fig. 2. Inverse susceptibility *vs.* temperature for various RCr_6Ge_6 compounds with $\text{R} \equiv \text{Dy}, \text{Ho}, \text{Er}$. The measurements were all performed in a magnetic field of 3 T.

Fig. 3. Magnetization *vs.* temperature for YCr_6Ge_6 at an applied magnetic field of 0.1 T.

Fig. 4. Inverse susceptibility *vs.* temperature for YCr_6Ge_6 , measured in a magnetic field of 3 T.

Fig. 5. Magnetic isotherms for YCr_6Ge_6 measured at 1.8 K (higher curve) and 5 K (lower curve).

3 suggests only very weak magnetic interactions in this compound. An explanation for the saturation behaviour could be a strongly field-dependent Cr moment. Both curves pictured in Fig. 5 suggest a saturated Cr moment of approximately $0.2 \mu_B$ in this compound.

For TbCr_6Ge_6 the situation is slightly different and more like that found in GdCr_6Ge_6 [3]. Measurements of the magnetization as a function of temperature in different applied fields (0.1 T, 1 T, 5.5 T), reveal magnetic ordering at temperatures below 40 K (Fig. 6). Most likely, this magnetic ordering is enhanced by the intersublattice interaction, which plays a much more important role in GdCr_6Ge_6 and TbCr_6Ge_6 than in the other compounds investigated (Gd and Tb have comparatively high spin moments). A Mössbauer study on GdCr_6Ge_6 [3] revealed a negative value for the electric field tensor element V_{zz} . Generally one may expect a relation between V_{zz} and A_2^0 of the type $A_2^0 = -\omega V_{zz}$, where ω has been found for a number of ternary R-3d compounds to be equal to about 46 [6]. According to this relationship a negative V_{zz} value would correspond to a positive A_2^0 value. For the Tb moments, which correspond to a negative Stevens factor α_1 , a positive A_2^0 value contributes to an easy axis anisotropy [7]. It is therefore likely that in TbCr_6Ge_6 the crystal field tends to align the Tb moments along the c axis, thereby

Fig. 6. Magnetization *vs.* temperature for TbCr_6Ge_6 in three different applied fields of 0.1 T (lower curve), 1 T (middle curve) and 5.5 T (upper curve) respectively.

Fig. 7. Inverse susceptibility *vs.* temperature for TbCr_6Ge_6 . (Measurement taken in a field of 3 T).

Fig. 8. Magnetic isotherms for TbCr_6Ge_6 at 4.2 K (main figure) and 5 K (inset).

favouring a sharper magnetic ordering than in GdCr_6Ge_6 .

A peculiar feature of the measurements presented in Fig. 6 is the pronounced increase of the Curie temperature with increasing applied field B , from a

Fig. 9. Magnetic isotherms at 5 K for DyCr_6Ge_6 (a), HoCr_6Ge_6 (b) and ErCr_6Ge_6 (c).

value of about 11 K for $B=0.1$ T to a value close to 30 K at $B=5.5$ T. It is possible that the TbCr_6Ge_6 compound is also sensitive to field-induced contributions to the Cr moment, a suggestion mentioned earlier in connection with the YCr_6Ge_6 compound. This would explain the strong applied field dependence of the Curie temperature.

At higher temperatures, the inverse susceptibility *vs.* temperature for TbCr_6Ge_6 between 5 K and 350 K shows Curie-Weiss behaviour, indicative of the paramagnetic state (Fig. 7). The effective moment deduced from the linear part of Fig. 7 is given in Table 1.

To investigate the type of magnetic ordering which occurs in TbCr_6Ge_6 , magnetic isotherms were obtained at 5 K and 4.2 K and in fields up to 5.5 T and 35 T respectively. As can be seen in Fig. 8, the magnetization shows a tendency towards saturation at low fields already, although a slight differential susceptibility remains, even at high fields. The magnetic moment per formula unit remains significantly below the free-ion Tb moment. This observation suggests a ferrimagnetic (anti-parallel) alignment between the Tb and Cr moments.

The magnetic isotherms at 5 K for the compounds with $R \equiv \text{Dy}, \text{Ho}, \text{Er}$ are plotted in Fig. 9. As in the case of TbCr_6Ge_6 , these isotherms show a tendency towards saturation at low fields already. An interpretation of these curves is, however, more difficult than in the case of TbCr_6Ge_6 , because no clear indication of magnetic ordering was obtained for these materials from the temperature dependence of the magnetization.

4. Conclusions

As may be inferred from the experimental data, magnetic ordering is almost absent in the RCr_6Ge_6 compounds discussed in this paper, with an exception for TbCr_6Ge_6 , where the ordering can easily be influenced by the application of an external magnetic field. The Cr ions possess a very small magnetic moment, which is revealed by magnetization measurements on YCr_6Ge_6 . At higher field strengths, the Tb and Cr moments tend to order in an anti-parallel configuration.

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