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DOI
10.1016/0925-8388(94)90769-2

Publication date
1994

Published in
Journal of Alloys and Compounds

Citation for published version (APA):
Magnetic properties of RCr₆Ge₆ compounds

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(Received August 18, 1993)

Abstract

Compounds of the composition RCr₆Ge₆ have been prepared and investigated by X-ray diffraction. The crystal structure was identified as the MgFe₆Ge₆ 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 R = Dy, Ho, Er, no indication of magnetic ordering was found for temperatures down to 5 K. A modest ordering was found in TbCr₆Ge₆. Magnetization measurements on YCr₆Ge₆ revealed a very small saturation moment of about 0.2 μₜₜ 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 RT₆Ge₆ compounds with T = Mn, Cr [1–4]. These compounds crystallize in the hexagonal MgFe₆Ge₆ 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 T = 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 GdCr₆Ge₆ [3] could not fully clarify the magnetic behaviour of the R moments, we performed further experiments on the other RCr₆Ge₆ compounds with R = Y, Tb, Dy, Ho, 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 diffraction, and found to be approximately single phase (MgFe₆Ge₆ structure), except for the YCr₆Ge₆ 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 WbCr₆Ge₆ 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 RCr₆Ge₆ 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
of the Tb compound, as in many other R-3d intermetallics.

For the RCr₆Ge₆ compounds with R = Dy, Ho, 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 ErCr₆Ge₆ presented in Fig. 1 illustrates this clearly and is representative for the compounds with R = Dy, 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 RCr₆Ge₆ compounds.

The possibility of a magnetic moment on the Cr ions led us to the investigation of the YCr₆Ge₆ 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.
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\textsubscript{6}Ge\textsubscript{6}, the situation is slightly different and more like that found in GdCr\textsubscript{6}Ge\textsubscript{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\textsubscript{6}Ge\textsubscript{6} and TbCr\textsubscript{6}Ge\textsubscript{6} than in the other compounds investigated (Gd and Tb have comparatively high spin moments). A Mössbauer study on GdCr\textsubscript{6}Ge\textsubscript{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 \( \omega \) 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 \( \alpha \), a positive \( A_2^0 \) value contributes to an easy axis anisotropy [7]. It is therefore likely that in TbCr\textsubscript{6}Ge\textsubscript{6} the crystal field tends to align the Tb moments along the c axis, thereby favouring a sharper magnetic ordering than in GdCr\textsubscript{6}Ge\textsubscript{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
To investigate the type of magnetic ordering which occurs in TbCr₆Ge₆, 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=Dy, Ho, Er are plotted in Fig. 9. As in the case of TbCr₆Ge₆, 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 TbCr₆Ge₆ 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 RCr₆Ge₆ compounds discussed in this paper, with an exception for TbCr₆Ge₆, 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 YCr₆Ge₆. At higher field strengths, the Tb and Cr moments tend to order in an anti-parallel configuration.

References