ELSEVIER

Journal of Magnetism and Magnetic Materials 157/158 (1996) 639–640

journal of magnetism and magnetic materials

Susceptibility of $(Gd,La)Mn_2(Ge,Si)_2$ compounds in the magnetically ordered regime

J.H.V.J. Brabers [a,b], C.H. de Groot [a,b,*], K.H.J. Buschow [a], F.R. de Boer [a]

[a] *Van der Waals–Zeeman Institute, University of Amsterdam, 1018 XE Amsterdam, The Netherlands*
[b] *Philips Research Laboratories, Prof. Holstlaan 4, 5656 AA Eindhoven, The Netherlands*

Abstract

The susceptibility of $ThCr_2Ge_2$-type compounds with the composition $(Gd,La)Mn_2(Ge,Si)_2$ has been investigated experimentally at temperatures at which the Mn-sublattice is still magnetically ordered, and explained in terms of a simple model. The temperature behaviour of the susceptibility can be explained as a result of the interplay between the temperature dependent antiferromagnetic Mn–Mn interaction and the net R–Mn interaction acting on the Mn-sublattice due to polarisation of the (paramagnetic) R-sublattice. There is good agreement between model calculations and experiment, enabling the determination of reliable estimates for the Mn–Mn and R–Mn interaction.

Keywords: R–Mn intermetallics; Exchange interactions; Antiferromagnetism

1. Introduction

In the past, a lot of research has been devoted to the magnetic properties of $ThCr_2Si_2$-type compounds of the RMn_2Ge_2 composition [1], which consist of a layered arrangement of Mn-ions seperated by R- and Ge-ions. A special interest was taken in the role of the (anti)ferromagnetic Mn-sublattice in the magnetic behaviour of these compounds. It was revealed that, like in other R–Mn compounds, the Mn–Mn interlayer interaction very strongly depends on the Mn–Mn interlayer distance and that thermal expansion has a large effect on the sign and magnitude of this interaction [2]. It was also found that only lowest order variations of the unit-cell parameters (or volume) have to be taken into account for an appropriate description of the mean-field interlayer interaction parameter n_{Mn-Mn} in relation to the unit-cell dimensions and a linear relation between n_{Mn-Mn} and unit-cell dimensions may be considered.

Due to the strong dependence of n_{Mn-Mn} on the unit-cell dimensions, n_{Mn-Mn} becomes strongly temperature dependent due to thermal expansion. At temperatures above 100 K, the unit-cell parameters of many RMn_2Ge_2 compounds vary linearly with temperature (T) [1,2]. Therefore, the relation between n_{Mn-Mn} and T at these temperatures may be treated also as linear.

As an illustration showing the role of the temperature dependence of the Mn–Mn interlayer interaction on the magnetic properties of antiferromagnetic $(Gd,La)Mn_2(Ge,Si)_2$ compounds, the inverse susceptibility of a few representatives of these series has been investigated theoretically and experimentally. An interesting aspect of these investigations is the paramagnetic contribution of the R-sublattice in the temperature range under investigation, although it complicates the analysis and interpretation of the χ^{-1} versus T behaviour. In case of a pure antiferromagnet, χ^{-1} is simply equal to the mean-field coupling constant related to the antiferromagnetic interaction (n_{Mn-Mn}), and its temparature dependence reflects the thermal expansion of the material. Contrary to this, the inverse susceptibility of an antiferromagnetic sublattice interacting with another sublattice, as in the compounds under investigation, is generally quite different. Under special conditions however, the experimental χ^{-1} curves can be interpreted well and give information on various materials parameters.

2. Results and discussion

According to a model oulined in Ref. [4], the susceptibility of a collinear antiferromagnet subjected to an R–3d exchange-field caused by a polarised paramagnetic R-sublattice can be expressed as:

$$\chi^{-1} = \frac{n_{3d-3d} - \chi_R n_{R-3d}^2}{1 - 2\chi_R n_{R-3d} + \chi_R n_{3d-3d}}.$$

A remarkable feature of this equation is that, like in the case of a collinear antiferromagnet, the magnetisation and

* Corresponding author. Fax: +31-40-2744-282; email: grootc@natlab.research.philips.com.

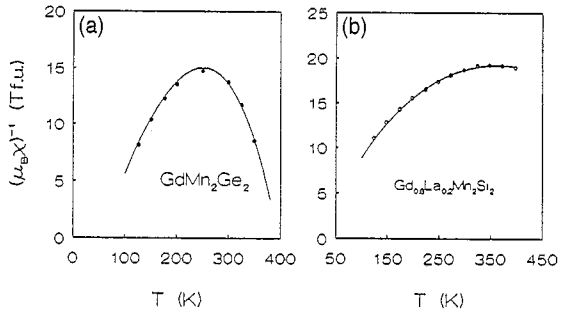

Fig. 1. Inverse susceptibility of $GdMn_2Ge_2$ (a) and of $Gd_{0.8}La_{0.2}Mn_2Si_2$ (b) as a function of temperature.

the susceptibility are independent of the 3d (Mn) sublattice moments. Both quantities depend on the values for the different parameters n_{3d-3d}, n_{R-3d}, and χ_R only. At sufficiently high temperatures, the mean-field parameter n_{3d-3d} is given by the equation: $n_{3d-3d} = a + bT$ (with $b < 0$) [2]. Provided that the concentrations of the specific R-ions in a material (N_R) are known, $\chi_R(T)$ can be easily obtained, so that according to Eq. (1) χ^{-1} can be described in terms of three (adjustable) parameters: a, b, and n_{R-3d}.

Fig. 1a and Fig. 1b represent χ^{-1} versus T measurements obtained on two different samples of nominal composition $GdMn_2Ge_2$ and $Gd_{0.8}La_{0.2}Mn_2Si_2$ (the susceptibility is defined here as $M/\mu_0 H$, with the magnetisation M expressed in μ_B/f.u.). The samples were prepared by arc-melting and annealed at 800°C for one week. The $ThCr_2Si_2$ structure of the samples was verified by X-ray diffraction. The χ^{-1} curves were obtained from magnetisation (M) versus field (B) measurements for each temperature, yielding linear M–B curves. Both figures show a parabola-shaped curve, which can be explained as follows. At low temperatures, the susceptibility of the R-sublattice is still quite high and causes a substantial R–Mn exchange field acting on the Mn-sublattice. As an overall result the susceptibility of the sample is enhanced by this mechanism and consequently χ^{-1} significantly lowered. With increasing temperature however, the R-sublattice susceptibilty decreases. Hence, the susceptibilty decreases and χ^{-1} increases. At very high temperatures, the R-sublattice susceptibilty is no longer relevant and the susceptibility is determined almost solely by the susceptibility of the Mn-sublattice, which increases with temperature due to the decrease of n_{Mn-Mn} with T. In this temperature regime, the inverse susceptibility is almost equal to n_{Mn-Mn}. The overall result of the interplay between the applied field and the R–Mn and the Mn–Mn exchange interactions is a χ^{-1} versus T curve showing a single maximum like the curves in Fig. 1a and Fig. 1b.

The full curves in Fig. 1a and Fig. 1b represent calculations of χ^{-1} versus T based on Eq. (1) and a least squares fitting procedure using a, b, and n_{R-Mn} as adjustable parameters. These calculated curves reproduce the experimental data quite well, thereby corroborating the validity of the model outlined in Ref. [4]. With the first number referring to Fig. 1a and the number in brackets to Fig. 1b, the parameter values obtained from the least squares fitting procedure are $\mu_B \cdot a = 41.9$ (48.5) Tf.u., $\mu_B \cdot b = -0.093$ (-0.058) Tf.u. K^{-1} and $\mu_B \cdot n_{R-Mn} = 11.9$ (8.8) Tf.u. The n_{R-Mn}-values can be related to the spin–spin coupling constant J_{R-Mn} through a simple mean-field analysis [3] yielding $J_{R-Mn}/k_B = 4.0$ (2.0) K. It is interesting to mention that the value of 4.0 K for $GdMn_2Ge_2$ is close to the value reported in Ref. [5]. This again confirms the relevance of the model and shows that the parameter values obtained from its application to the experimental data can be considered as realistic.

Concluding, we can say that the temperature behaviour of the susceptibility of the compounds under investigation is understood fairly well as a phenomenon dominated by the distance dependent Mn–Mn interlayer interaction. The model outlined here may be applicable to other R–Mn intermetalllics as well.

References

[1] A. Szytula, in; Ferromagnetic Materials, Vol. 5, ed. K.H.J. Buschow (North-Holland, Amsterdam, 1991).
[2] J.H.V.J. Brabers, A.J. Nolten, F. Kayzel, S.H.J. Lenczowski, K.H.J. Buschow and F.R. de Boer, Phys. Rev. B 50 (1994) 16410.
[3] R. Verhoef, Thesis, University of Amsterdam (1990).
[4] J.H.V.J. Brabers, Thesis, University of Amsterdam (1995).
[5] N. Iwata, K. Hattori and T. Shigeoka, J. Magn. Magn. Mater. 53 (1986) 318.