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Physica B 211 (1995) 102–104

PHYSICA B

Antiferromagnetic interactions in $Nd_6Fe_{12}Ga_2$-based compounds

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Abstract

It has been reported that antiferromagnetic coupling may exist between light-rare-earth and transition-metal ions in $Nd_6Fe_{11}Ga_3$-type of compounds. In the present paper, the magnetic properties of $Nd_{6-x}La_xFe_{12}Ga_2$ compounds with $x = 0, 1, 2, 3$ have been studied by measuring the free-powder high-field magnetization at 4.2 K and by measuring the temperature dependence of the magnetization in the temperature range 4.2–600 K. The Néel temperatures found above room temperature in the M versus T curves only slightly decrease with increasing La content. The M versus B curves at 4.2 K exhibit several metamagnetic transitions. The transition fields increase first and then decrease slowly with increasing La content. By extrapolating the high-field magnetization to infinite field values, values of the Fe moments have been derived by assuming that the Nd moment has the free-ion value.

1. Introduction

It has been reported that in compounds of the type $R_6Fe_{11+x}M_{3-x}$ (R = Pr, Nd, Sm; M = Ga, Al, Sn, B, etc., and $x = 1, 2, 3$) the possibility of antiferromagnetic coupling exists between rare-earth and transition-metal sublattices [1]. All these compounds crystallize in the tetragonal $La_6Co_{11}Ga_3$ structure [2]. Fig. 1 shows the crystal structure of this type of compounds. As there are several different crystallographic sites for both the Fe and the R atoms, the underlying magnetic structures are not fully understood. Although some Mössbauer experiments have been done recently, the results of the magnetic measurements cannot fully be explained with the results from the latter measurements [3–5]. In previous studies [1, 6], we have reported on the influence of Fe sublattices on the magnetic properties and shown that increase of the Fe content strongly enhances the antiferromagnetic interaction in the $R_6Fe_{11+x}Al_{3-x}$ and $R_6Fe_{11+x}Ga_{3-x}$ (R = Nd, Pr) systems. Partial substitution of B for Ga in Nd-based compounds results in a change of the room-temperature anisotropy from easy

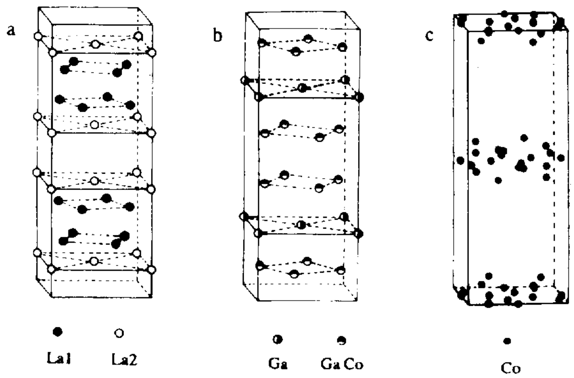

Fig. 1. Crystal structure of $La_6Co_{11}Ga_3$: (a) R sublattices; (b) Ga and GaCo sublattices; (c) Co sublattices.

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0921-4526/95/$09.50
SSDI 0921-4526(94)00955-4

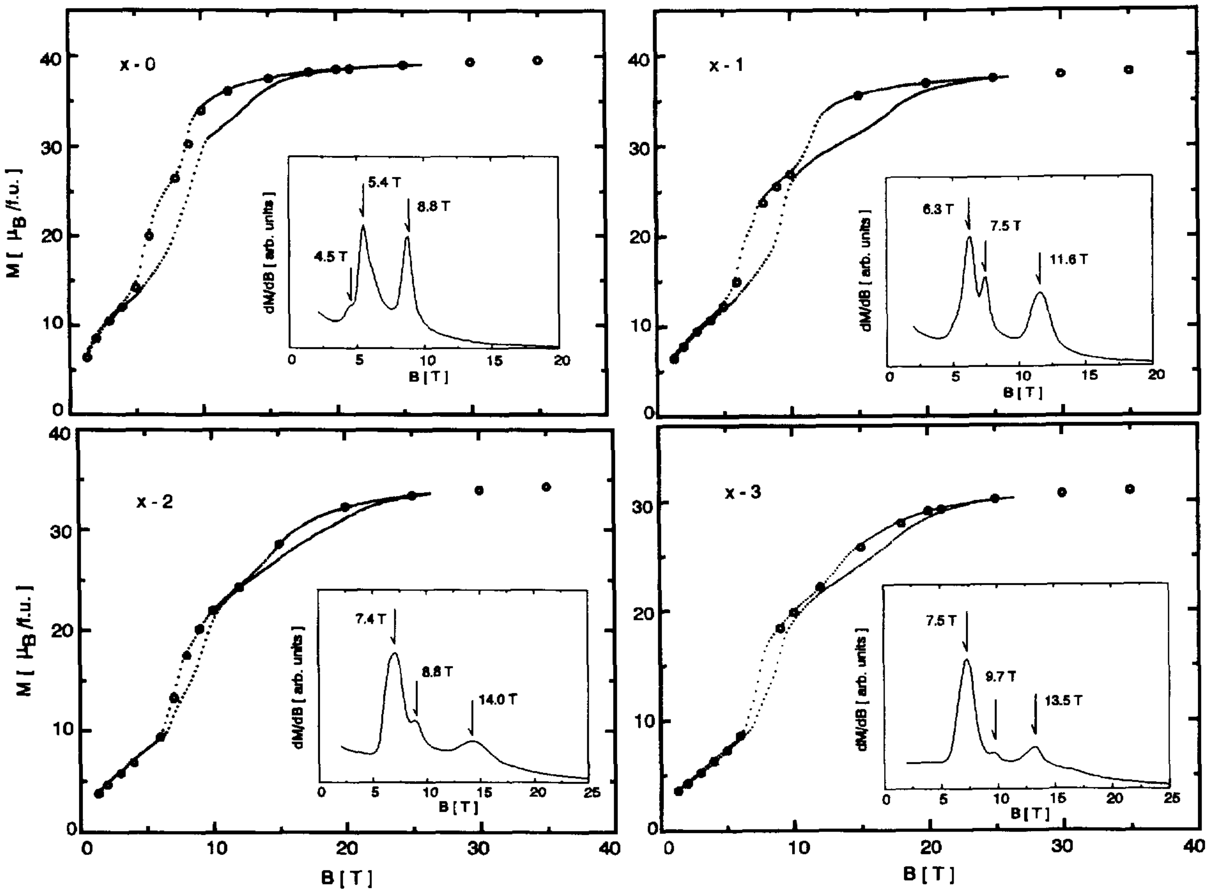

Fig. 2. Free-powder magnetization of $Nd_{6-x}La_xFe_{12}Ga_2$ with $x = 0, 1, 2, 3$ measured at 4.2 K. The circles stand for the step-like-field measurements and the dotted lines represent continuous-field measurements. The inserts give the field dependence of dM/dB for the curves measured in decreasing fields.

plane to easy axis; simultaneously, the Curie temperature increases [7]. In this paper, we focus on the influence of the Nd sublattices on the magnetic properties of $Nd_6Fe_{12}Ga_2$ compounds by studying the high-field magnetic properties of $Nd_{6-x}La_xFe_{12}Ga_2$ compounds with $x = 0, 1, 2, 3$.

2. Experimental

$Nd_{6-x}La_xFe_{12}Ga_2$ samples with $x = 0, 1, 2, 3$ have been prepared by arc melting the pure elements. An annealing treatment was carried out at 600°C for four weeks in a quartz tube filled with pure argon. All the samples were characterized by X-ray diffraction experiments which showed them to be single phase within the accuracy of the experiment.

Magnetization measurements have been done at the University of Amsterdam High-Field Facility on free-powder samples at 4.2 K in fields up to 35 T. The temperature dependence of the magnetization was measured on free-powder particles and polycrystalline bulk samples in a field of 1 T and in the temperature ranges 4.2–300 K and 300–600 K by using a SQUID magnetometer and an extracting-sample magnetometer, respectively.

3. Results and discussions

In order to be able to study the antiferromagnetic interactions in this type of compounds, free-powder magnetization measurements were performed because FOMP transitions are not observable in the free-powder magnetization [1].

Table 1
Composition dependence of the transition field and the amplitude of the transition

	$x = 0$	$x = 1$	$x = 2$	$x = 3$
M_s (μ_B/f.u.)	39.8	38.5	35.0	31.2
M_{Fe} (μ_B/f.u.)	1.7	1.8	1.8	1.8
T_N (K)	23/374	358	352	350
ΔM (μ_B/f.u.)	6	6	6	5
B_{cr} (T)	8.8	11.6	14.0	13.5

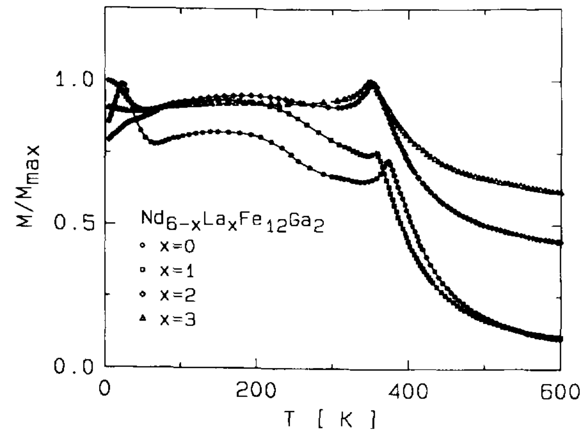

Fig. 3. Temperature dependence of the reduced magnetization of $Nd_{6-x}La_xFe_{12}Ga_2$ compounds with $x = 0, 1, 2, 3$.

The free-powder high-field magnetization curves of $Nd_{6-x}La_xFe_{12}Ga_2$ compounds are shown in Fig. 2. The inserts show the dependence of dM/dB on the external field. All these compounds show magnetic transitions, the transition fields increasing with increasing La content. There are three jumps in each magnetization curve, two of them being very pronounced. Considering the transition which occurs at the highest field only, we can estimate the magnetization changes at this transition. The results are given in Table 1 together with the transition fields. One can see that the transition field strongly depends on the rare-earth sublattice. For $x \leqslant 2$, the magnetization jump at the transition does not change with x, while the critical field of the transition strongly increases with increasing x. For $x > 2$, the magnetization jump at the transition tends to decrease but the critical field is almost independent of the La content. This may indicate that there is some preference for La to occupy one of the two rare-earth sites in this type of compounds.

By extrapolating the magnetization to infinite field, the average magnetic moment of Fe atoms can be derived by assuming that Nd contributes its free-ion moment. From the results shown in Fig. 2, one finds the values $1.7\mu_B$/Fe for $x = 0$ and $1.8\mu_B$/Fe for $x = 1$–3. It is interesting to note that these Fe moments are substantially larger than the Fe moments found in the Laves-phase compounds RFe_2 ($1.5\mu_B$/Fe) for which the relative R and Fe concentration is similar to that of the presently studied compounds.

Fig. 3 shows the temperature dependence of the reduced magnetization (M/M_{max}) of the compounds measured in the range 4.2–600 K. Néel temperatures can clearly be distinguished, which confirms that the transitions in the magnetization curves shown in Fig. 2 involve changes from antiferromagnetic to ferromagnetic magnetic-moment arrangements. One would expect that the high-temperature peaks correspond to ordering of the Fe sublattices. These peaks appear to decrease only slightly with increasing La content. Because the compounds contain many inequivalent crystallographic sites it is difficult to predict from which sublattice the magnetic transitions shown in Fig. 2 and the transitions shown in Fig. 3 originate. It is reasonable to assume that the low-temperature peak in the $M(T)$ curve of $Nd_6Fe_{12}Ga_2$ is due to ordering of the Nd sublattice. The effect of La substitution is seen in Fig. 3 to have a much larger influence on the low-temperature peak than on the high-temperature peak of $M(T)$ curve. It seems as if the magnetic transition has moved to lower temperature and has become less sharp. Both these features can be expected when part of the Nd moments is replaced by the nonmagnetic La atoms. However, to understand the magnetic properties of this type of compounds, neutron-diffraction measurements are required.

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