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Possible ferrimagnetic coupling in light-rare-earth transition-metal intermetallic compounds

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The possibility of ferrimagnetic coupling between the magnetic moments of the light-rare-earth (Nd, Pr) ions and of the transition-metal (Fe) ions in $Nd_6Ga_3Fe_{11}$- and $Pr_6Ga_3Fe_{11}$-based compounds is deduced from the high-field magnetization curves measured at 4.2 K on magnetically aligned and on free-powder samples and from the temperature dependence of the magnetization. The ferrimagnetic coupling is very weak: at 4.2 K, the rare-earth moments and the transition-metal moments can be forced to ferromagnetic alignment by application of an external magnetic field of about 4 T. Generally, in intermetallic compounds, the magnetic coupling is ferromagnetic between light-rare-earth and transition-metal ions and ferrimagnetic between heavy-rare-earth and transition-metal ions. The present results suggest that in some crystal structures ferrimagnetic coupling also may exist in the compounds formed by light-rare-earth and transition-metal elements.

I. INTRODUCTION

The compounds $Nd_6Ga_3Fe_{11}$ and $Pr_6Ga_3Fe_{11}$ crystallize in the $La_6Ga_3Co_{11}$-type of structure with space group $I4/mcm$.[1] The crystal structure of $La_6Ga_3Co_{11}$ is shown in Fig. 1. The rare-earth atoms occupy two different crystallographic sites, $La1$ and $La2$. The Ga atoms occupy one site and share another site with Co. It can be seen from Fig. 1 that the crystal entails a stacking of layers of Co, rare-earth, and Ga atoms.

Magnetization transitions at low field strengths have been found at low temperatures in the $R_6Ga_3Fe_{11}$ compounds with R=Nd, Pr, Sm.[2] A previous investigation[3] has shown that the transition fields in $Nd_6Ga_3Fe_{11}$ and $Pr_6Ga_3Fe_{11}$ almost do not change when B is substituted for Ga; however, the magnetic anisotropy of $Nd_6Ga_3Fe_{11}$ is influenced remarkably, leading to a spin-reorientation transition in $Nd_6Ga_{2.5}B_{0.5}Fe_{11}$ at about 130 K.[3] These results suggest that the low-field transitions are not connected with the magnetic anisotropy. The aim of the present work is to obtain information about the origin of these transitions in $Nd_6Ga_3Fe_{11}$ and $Pr_6Ga_3Fe_{11}$.

II. EXPERIMENTAL PROCEDURE

The compounds $Nd_6Ga_{3-x}B_xFe_{11}$ and $Pr_6Ga_{3-x}B_xFe_{11}$ (x=0, 0.5) were prepared by melting together appropriate amounts of pure Nd, Pr, Ga, Fe, Fe-B alloy (20% B) in an arc furnace in a purified-argon atmosphere. To reach homogeneity, the ingots were inverted and melted twice. Wrapped in Mo foil, the as-cast ingots were annealed at 700 °C for 2 weeks in argon atmosphere. By x-ray diffraction, the annealed samples were found to possess the $La_6Ga_3Co_{11}$-type structure and to contain small amounts of α-Fe.

Magnetically aligned samples were prepared by mixing fine powder with a resin-doped epoxy solution at room temperature and by letting the mixture solidify in a mold with a cylindrical hole in an external magnetic field of 0.8 T. High-field magnetization curves at 4.2 K were measured in the high-field installation at the University of Amsterdam with the field applied parallel and perpendicular to the alignment direction of the samples. The free-powder magnetization was measured on samples consisting of fine powder particles which could freely rotate in the sample holder under the influence of the applied external field. The temperature dependence of the magnetization was measured on free-powder samples between 4.2 and 260 K in a field of 3 T in an extracting-sample magnetometer.

III. RESULTS AND DISCUSSION

The magnetization curves of $Nd_6Ga_3Fe_{11}$ and $Nd_6Ga_{2.5}B_{0.5}Fe_{11}$, measured parallel and perpendicular to the alignment direction, are shown in Fig. 2. In the dotted magnetization curves recorded with increasing field, the transitions earlier reported[3] to occur in low fields (below 5 T) are vaguely distinguishable, but they are very clearly visible in the dM/dB curves shown in Fig. 3. In the perpendicular

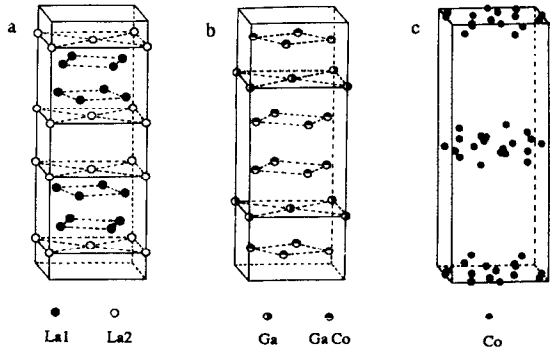

FIG. 1. Crystal structure of $La_6Ga_3Co_{11}$.

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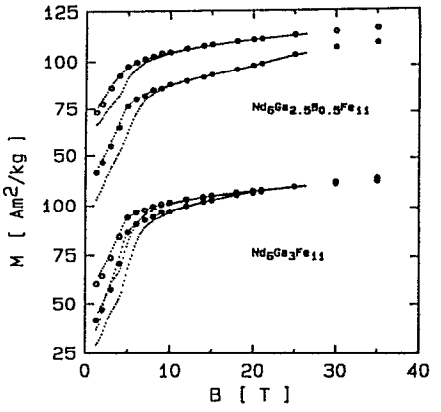

FIG. 2. Magnetization curves at 4.2 K of $Nd_6Ga_3Fe_{11}$-based compounds for the field applied parallel (○) and perpendicular (●) to the alignment direction. The dotted lines correspond to data obtained by means of a triangular continuous-field pulse in which the field increases linearly with time up to 26 T and after that decreases linearly.

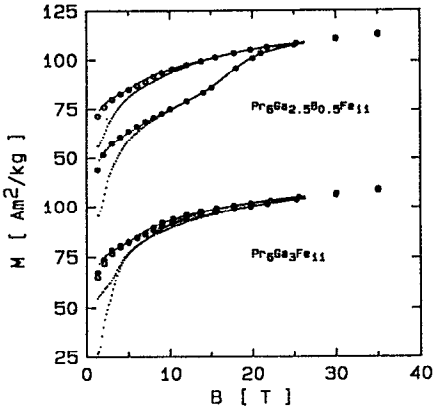

FIG. 4. Magnetization curves at 4.2 K of $Pr_6Ga_3Fe_{11}$-based compounds for the field applied parallel (○) and perpendicular (●) to the alignment direction. The dotted lines correspond to data obtained by means of a triangular-field pulse in which the field increases linearly with time up to 26 T and after that decreases linearly.

magnetization of $Nd_6Ga_{2.5}B_{0.5}Fe_{11}$ there is also a transition around 20 T. It is also seen in Fig. 3 that the critical fields of the low-field transitions hardly depend on the substitution of B for Ga. However, the magnetic anisotropy of $Nd_6Ga_3Fe_{11}$ is considerably enhanced upon substitution of B for Ga.

In $Pr_6Ga_3Fe_{11}$ (Fig. 4), the magnetic anisotropy is very small (<5 T), the parallel and perpendicular magnetization curves coinciding in relatively low fields. Similar to $Nd_6Ga_3Fe_{11}$, the substitution of B for Ga results in a higher magnetic anisotropy. Also in $Pr_6Ga_3Fe_{11}$ and $Pr_6Ga_{2.5}B_{0.5}Fe_{11}$, transitions are found at low fields in both the parallel and the perpendicular magnetization curves (Fig. 5). The critical fields of these transitions are again nearly independent of B substitution, as in the case of the $Nd_6Ga_3Fe_{11}$-based compounds. For $Pr_6Ga_{2.5}B_{0.5}Fe_{11}$, additional transitions are found both in the parallel magnetization curve (at 9 T) and in the perpendicular magnetization curve (at 17 T). All magnetization curves of both the $Nd_6Ga_3Fe_{11}$- and the $Pr_6Ga_3Fe_{11}$-based compounds display a high differential susceptibility even in the highest applied fields indicating a noncollinear structure to be present. Assuming that the rare-earth moments and the Fe moments are nearly parallel at 35 T and that Nd and Pr have the free-ion moment values of $3.20\mu_B$ and $3.27\mu_B$, respectively, we can derive for both $Nd_6Ga_3Fe_{11}$ and $Pr_6Ga_3Fe_{11}$ an Fe moment of about $1.7\mu_B$ from the values of the parallel magnetization at 35 T. This value for the Fe moment is in good agreement with Mössbauer results.[2]

The experimental results presented above show that substitution of B for Ga in $Nd_6Ga_3Fe_{11}$ and $Pr_6Ga_3Fe_{11}$ hardly affects the low-field transitions in these compounds. In the Nd compounds, the transition fields are slightly higher than in the Pr compounds. Of substantial interest is the fact that

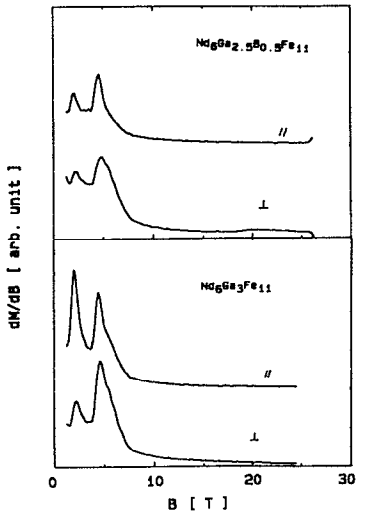

FIG. 3. Field dependence of dM/dB of $Nd_6Ga_3Fe_{11}$-based compounds at 4.2 K for the increasing-field part of the continuous-field 26 T pulse.

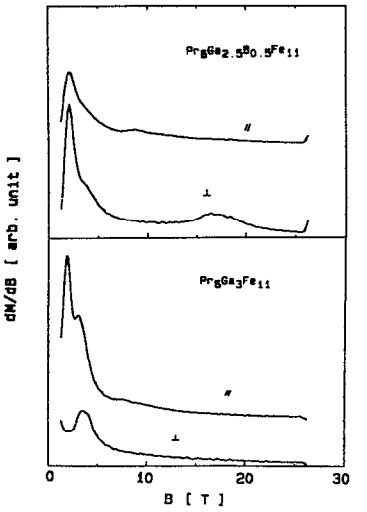

FIG. 5. Field dependence of dM/dB of $Pr_6Ga_3Fe_{11}$-based compounds at 4.2 K for the increasing-field part of the continuous-field 26 T pulse.

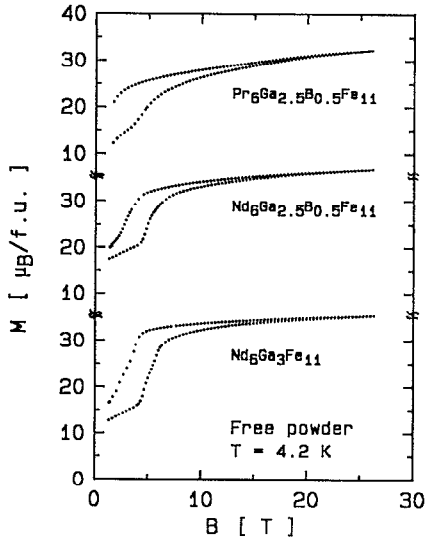

FIG. 6. Free-powder magnetization of $R_6Ga_{2.5}B_{0.5}Fe_{11}$ (R=Nd, Pr) compounds at 4.2 K.

the transitions are observed in both the parallel and perpendicular magnetization curves. Because there are at least two transitions in each magnetization curve, it is unlikely that all these transitions are first-order magnetization transitions (FOMP) as observed in $Nd_2Fe_{14}B$.[4] Even more important in this respect is the fact that the transitions appear in both the parallel and the perpendicular magnetization curves. In the case of FOMP transitions one would also have the contradiction that the anisotropy fields of the compounds are strongly enhanced by substitution, whereas the critical fields of the low-field transitions do not change. These low-field transitions are therefore not to be associated with the overall anisotropy of the compounds.

To further investigate the nature of the observed transitions we performed free-powder magnetization measurements on $Nd_6Ga_3Fe_{11}$, $Nd_6Ga_{2.5}B_{0.5}Fe_{11}$, and $Pr_6Ga_{2.5}B_{0.5}Fe_{11}$ with the idea that transitions originating from discontinuous rotation of the total magnetization vector as occurs in a FOMP should not be observable in free-powder magnetization curves. Figure 6 shows that the magnetization jumps at about 20 and 17 T in $Nd_6Ga_{2.5}B_{0.5}Fe_{11}$ (Figs. 2 and 3) and $Pr_6Ga_{2.5}B_{0.5}Fe_{11}$ (Figs. 4 and 5), respectively, have completely disappeared in the free-powder magnetization proving these transitions to be of the FOMP type. Only at low fields are transitions observed in the free-powder data, which are of different origin and may be associated

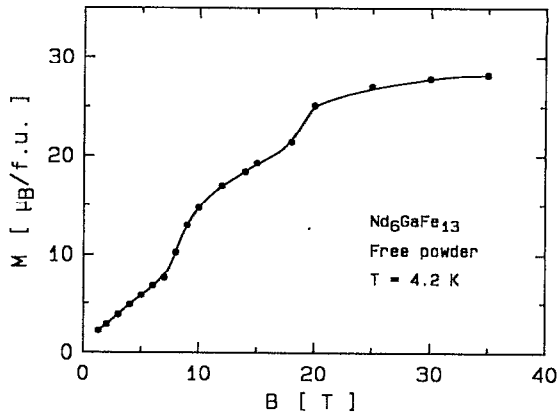

FIG. 7. Free-powder magnetization curve at 4.2 K of Nd_6GaFe_{13}. The line drawn is a guide to the eye.

with transitions from a ferrimagnetic spin structure to a ferromagnetic spin structure.

In order to obtain more information about the possible type of ferrimagnetic spin structure in these compounds, we measured the low-temperature magnetization of compounds of the type $Nd_6M_{3-x}Fe_{11+x}$ (M=Al or Ga) and found[5] that the transition fields strongly increase when Ga or Al are replaced by Fe. As an example, the result for $Nd_6Fe_{13}Ga$ is shown in Fig. 7. The substituted Fe atoms can be expected to occupy the Co positions in the GaCo layers in Fig 1(b), which separate the R and the Co layers. In this case, the substituted Fe ions will very likely influence the strength of the interaction between the moments in the R and the Fe layers. The result in Fig. 7 shows that this interaction is negative, which makes it tempting to conclude that in these compounds the Nd-Fe and the Pr-Fe interaction may be of the ferrimagnetic type. This would be the first example of compounds in which the moment configuration deviates from the ferromagnetic alignment of light-rare-earth moments with respect to the transition-metal moments, which has been explained by Campbell.[6]

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