Magnetoresistance effects at the metamagnetic transition in $R_6Fe_{14-x}Al_x$ (R=Nd, La)

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The $La_6Co_{11}Ga_3$-type intermetallic compounds $Nd_6Fe_{10}Al_4$, $La_6Fe_{10}Al_4$, and $La_6Fe_9Al_5$ have been studied by magnetization and electrical resistance measurements in applied fields up to 9 T. All compounds were found to be antiferromagnetic, i.e., no residual magnetization was found, in contrast to most compounds of the $R_6Fe_{14-x}M_x$ type. A metamagnetic transition was observed for all samples at a critical field smaller than 3 T, and a ~3%–5% resistivity drop was found to occur concomitantly. Both the magnetization (M) and the magnetoresistance (MR) exhibited a large hysteresis for the Nd compound, but the MR vs M relationship was history independent in the upper part of the magnetization curve. The metamagnetic transition field exhibited an anomalous temperature dependence for $Nd_6Fe_{10}Al_4$ which was ascribed to crystal-field effects. © *1999 American Institute of Physics.* [S0021-8979(99)34608-9]

I. INTRODUCTION

In recent years, electrical resistivity measurements have become an important complementary tool in the study of magnetic phase transitions. When the transition is from an antiferromagnetic (AF) state to a ferromagnetic (F) state, a resistivity drop usually occurs, one which is often large enough to warrant a giant magnetoresistance (GMR) designation. A long known example is the FeRh alloy, in which an AF→F transition can be driven by an applied field and a large resistivity decrease is observed.[1,2] Similar effects have been reported recently for intermetallic compounds such as $La(Fe_xAl_{1-x})_{13}$,[3] UNiGa,[4] $Ce(Fe_{1-x}Ru_x)_2$,[5] Ce_2Fe_{17},[6] and Gd_2In.[7] The excess resistivity of the AF phase can be ascribed to the opening of extra gaps in the Fermi surface (superzone effects) or to spin-dependent scattering. The latter is believed to be essential to explain the large resistivity drop associated to magnetic switching in antiferromagnetically coupled artificial multilayer structures, to which the GMR term was first applied.[8] Therefore, it is not surprising that naturally occurring systems with layered magnetic structures have attracted attention in the search for new GMR effects: the FeRh alloy itself has been pointed out as a natural magnetic multilayer.[9] In this context, the $R_6Fe_{14-x}Al_x$ compounds stand out as candidates for such studies.

The structural and magnetic properties of $R_6Fe_{13}M$ compounds, where R is a light rare earth and M=Ge, Si, Cu, Au, Ag, Sn, or Sb, have been extensively investigated.[10–15] These compounds crystallize as an ordered variant of the $La_6Co_{11}Ga_3$-type tetragonal structure. For M=Al or Ga, higher M concentrations are preferred and a solubility range exists, especially for Al. All compounds in this family feature AF couplings among Fe moments (and, most likely, between R moments as well), leading to antiferromagnetic or ferrimagnetic ordering. The magnetic structure has not been determined so far, but it has been suggested by various authors[13,14] that it may consist of ferromagnetic Fe slabs with alternating spin orientations, separated from each other by R and M atoms, with the distance between successive Fe slabs being as large as 10 Å. This model is attractive for its similarity with artificial multilayers.

Many compounds of the $R_6Fe_{14-x}M_x$ family exhibit slope changes in their magnetization curves, typical of metamagnetic transitions. The sharpest transitions are seen for M=Al and $x \geqslant 3$, irrespective of R being a magnetic ion (e.g., Nd) or not (La).[15] We have found that these transitions are accompanied by resistivity drops of 3%–5%, and in this article we report our first results.

II. EXPERIMENTAL DETAILS

Three compounds were studied in this work: $R_6Fe_{10}Al_4$ with R=Nd and La, and $La_6Fe_9Al_5$. Samples were prepared by arc melting and annealed in argon atmosphere at 750 °C for 4 weeks. All samples had the required tetragonal structure and no impurity phases could be detected in x-ray diagrams, but the La-based samples contained small amounts of α-Fe and La (or La_3Al), as revealed by their Mössbauer spectra and by the onset of superconductivity at $T \sim 5$ K. Attempts to prepare $Nd_6Fe_9Al_5$ have been unsuccessful. A $\sim 1 \times 1 \times 5$ mm^3 prism was cut from each button to be used in both the magnetization and the electrical resistance measurements. Magnetization measurements were made with a vibrating sample magnetometer mounted on a 9 T superconducting solenoid. The electrical resistance was measured with an ac bridge with the magnetic field applied longitudinally. Mössbauer spectra were taken on powder absorbers in order to characterize the samples for impurity phases, as well as to determine the Néel temperature by measuring hyperfine splitting as a function of temperature. The relevant physical parameters are given in Table I.

III. RESULTS AND DISCUSSION

Magnetization versus applied field curves, measured at $T = 4.2$ K up to 9 T, are shown in Fig. 1. The metamagnetic transition can be clearly identified as a kink in all the curves.

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TABLE I. Lattice constants and magnetic ordering temperatures of the compounds studied.

Sample	a (nm)	c (nm)	T_N (K)
$Nd_6Fe_{10}Al_4$	0.816(1)	2.306(1)	239(2)
$La_6Fe_{10}Al_4$	0.825(1)	2.381(1)	174(5)
$La_6Fe_9Al_5$	0.827(1)	2.388(1)	120(5)

The magnetization increases linearly with applied field up to the transition for the three samples studied, i.e., these compounds are antiferromagnetic. This is a remarkable result, since all similar compounds studied so far (mostly those of the $R_6Fe_{11}M_3$ type) exhibit nonlinear behavior in this region, reflecting a residual magnetization. In a recent paper, de Groot *et al.*[14] reported a spontaneous magnetization of about 2 μ_B/f.u. in many different compounds of this structure. Since this result was independent of the R element, it was concluded that the R sublattice is perfectly antiferromagnetic. The large difference between the $R_6Fe_{10}Al_4$ magnetization for R=Nd and La in Fig. 1 (corresponding to 14.4 μ_B/f.u. at 9 T) is clearly due to the contribution of Nd moments; thus, one effect of the applied field is to break up Nd–Nd antiferromagnetic couplings. It follows that the residual magnetization at low fields, observed in other compounds, must be due to an uncompensated moment in an otherwise AF Fe sublattice. In a previous paper[15] we have shown that the residual magnetization tends towards zero when going from $Nd_6Fe_{11}Al_3$ to $Nd_6Fe_{10}Al_4$, and this result is confirmed here for the Al-rich La compounds. A simple explanation is to assume that the additional Al atom just replaces the one Fe atom whose moment (of magnitude $\sim 2\ \mu_B$) is uncompensated at the 6–11–3 composition.

Magnetoresistance curves are shown in Figs. 2 and 3 for $Nd_6Fe_{10}Al_4$ and $La_6Fe_9Al_5$, respectively. The resistivity at low temperatures was estimated to be in the 180–200 $\mu\Omega$ cm range, a more accurate determination being difficult because of the smallness of our samples.

As seen in Figs. 2 and 3, at every temperature the metamagnetic transition is mirrored by a magnetoresistance curve of similar shape. For the Nd-based compound a significant hysteresis is observed at low temperatures. Since the hysteresis is much smaller for the La-based compound, it is obviously related to the crystal-field induced anisotropy of the Nd ions.

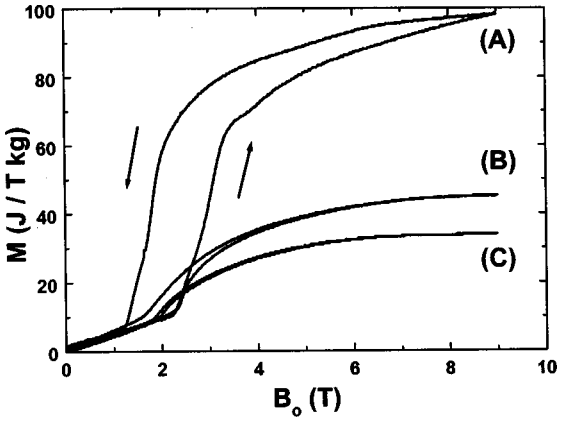

FIG. 1. Magnetization vs applied field for $Nd_6Fe_{10}Al_4$ (A), $La_6Fe_{10}Al_4$ (B), and $La_6Fe_9Al_5$ (C). The arrows indicate increasing and decreasing fields.

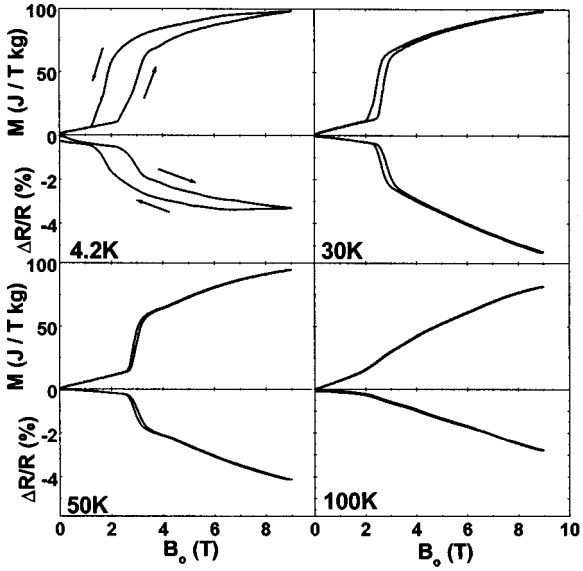

FIG. 2. $Nd_6Fe_{10}Al_4$ magnetization and magnetoresistance vs applied field at the temperatures indicated.

If the applied field B_0 is eliminated between the M vs B_0 and the $(\Delta R/R)$ vs B_0 data, a two-valued $(\Delta R/R)$ vs M function should result, corresponding to the increasing/decreasing field branches. The result of this operation for $Nd_6Fe_{10}Al_4$ at 4.2 K is shown in Fig. 4. Surprisingly, a significant portion of the curves merge together in such a way that for $M \geqslant 70$ J/T kg the magnetoresistance seems to depend on the magnetization only, irrespective of magnetic history. Further analysis shows that the data closely follow a M^2 law in this range. This result could be an indication that the upper part of the magnetization curve [see Fig. 1 (A)]

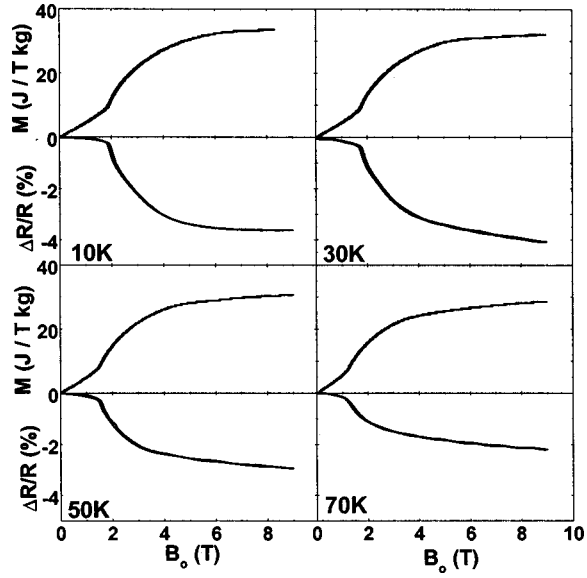

FIG. 3. $La_6Fe_9Al_5$ magnetization and magnetoresistance vs applied field at the temperatures indicated.

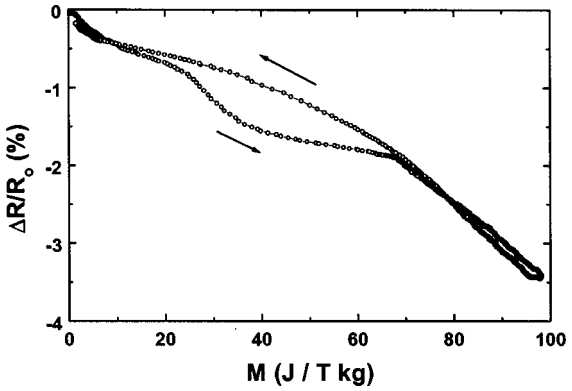

FIG. 4. $Nd_6Fe_{10}Al_4$ magnetoresistance vs magnetization at $T=4.2$ K. The arrows indicate increasing and decreasing fields.

corresponds to magnetic moment rotation towards the applied field, while the spin flips leading from an AF- to a *F*-like state have already occurred at lower fields.

The metamagnetic transition field B_c was determined from the upward knee of the *M* vs *B* curves (i.e., at the first d^2M/dB^2 maximum), so far as the transition was perceptible. The B_c temperature dependence for the three compounds studied is shown in Fig. 5. For the La compounds, B_c smoothly decreases at increasing temperatures and tends to zero for $T \rightarrow T_N$. Qualitatively, this is the expected behavior of the exchange field in an antiferromagnet. For $Nd_6Fe_{10}Al_4$, B_c is larger than for the corresponding La compound, because the Nd crystal-field anisotropy provides an extra energy (via Nd–Fe exchange coupling) that must be overcome to turn the Fe spins over. In addition, however, a B_c *increase* is seen at low temperatures, which is quite unusual. It has been inferred from point-charge calculations[16] that the two rare-earth sites in this structure (8*b* and 16*l*) have A_{20} crystal-field coefficients of opposite signs, which could lead to canted spin structures for the R sublattice. It thus may be

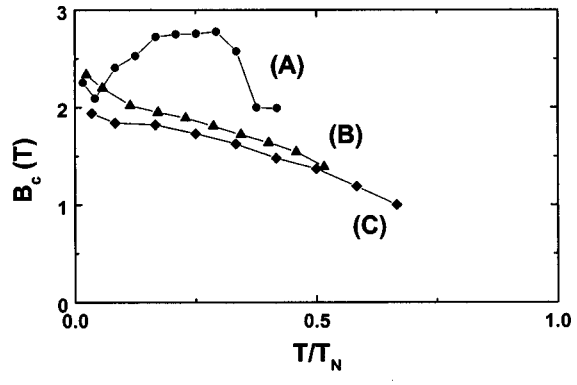

FIG. 5. Metamagnetic transition field vs reduced temperature for $Nd_6Fe_{10}Al_4$ (A), $La_6Fe_{10}Al_4$ (B), and $La_6Fe_9Al_5$ (C) (measured at increasing field).

conjectured that the magnetic ground state includes noncollinear Nd moments (most probably located at 16*l* sites), whose local anisotropy rapidly weakens with increasing temperature, making them smoothly rotate into collinearity with the Fe moments. In this way, their contribution to the total exchange field seen by Fe spins would increase with temperature. A temperature-driven spin reorientation has recently been detected by Mössbauer spectroscopy in $Nd_6Fe_{13}Si$,[17] and it would be interesting to search for a similar phenomenon in Al compounds.

In summary, the magnetization and electrical resistance of the intermetallic compounds $Nd_6Fe_{10}Al_4$, $La_6Fe_{10}Al_4$, and $La_6Fe_9Al_5$ have been measured in applied fields up to 9 T. All compounds were found to be antiferromagnetic. A metamagnetic transition was observed for all samples at a critical field smaller than 3 T, and a ~3%–5% resistivity drop was found to occur concomitantly. The metamagnetic transition field exhibited an anomalous temperature dependence for $Nd_6Fe_{10}Al_4$ which was attributed to crystal-field effects. The possible occurrence of a temperature-driven spin reorientation in this compound, as well as magnetovolume effects at the metamagnetic transition, are currently being investigated.

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