The magnetoresistance in GdNi: Magnetic-polaronic-like effect near Curie temperature and low temperature sign reversal

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Abstract

The results of magnetoresistance ($\Delta\rho/\rho$) measurements in GdNi, in the temperature range 4.2 to 300 K are reported. The sign of $\Delta\rho/\rho$ above the Curie temperature (T_C= 70 K) is negative and its magnitude in a magnetic field of 80 kOe grows with decreasing temperature below 150 K with a peak value of about -20% at T_C. These features, qualitatively resembling those in giant magnetoresistance systems (manganates), are attributed to the formation of some kind of magnetic polarons induced by Gd. The magnetoresistance changes sign below 12 K which is attributed to subtle band structure effects.

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Introduction

The observation[1] of giant and colossal magnetoresistance in perovskites, $La_{1-x}A_xMnO_3$ (A= Ca, Sr, Ba), near the semiconductor-metal transition temperature has generated much interest in this family of compounds. There have been lively discussions in recent literature on the relative importance of double exchange[2] and Jahn-Teller electron-phonon coupling mechanisms[3] for the increase in resistivity drop with the application of magnetic field (H) at the Curie temperature (T_C). Very recently,[4] large magnetoresistance in amorphous magnetic rare earth (R) alloys due to the formation of a dense concentration of magnetic polarons has been reported. In this context, we considered it worthwhile to probe the magnetoresistance ($\Delta\rho/\rho$) behavior of *normal* (where the double exchange or Jahn-Teller mechanisms are not expected to operate) ferromagnetic intermetallic compounds near T_C. At this point, it may be stated that we have recently reported interesting $\Delta\rho/\rho$ behavior in several rare-earth intermetallic compounds across the Néel temperature[5] as well as in paramagnetic alloys[6] (in addition to ferromagnetic and antiferromagnetic Ce systems). As a continuation of our programme in this direction, we report here the results of our magnetoresistance study on a ferromagnetic Gd alloy, viz., GdNi (T_C= 70 K)[7−12]. The magnetoresistance above T_C exhibits a temperature dependence similar to that noted in La manganates, as if there is a formation of magnetic-polarons (presumably induced in the Ni 3d orbital, which is otherwise nonmagnetic) by Gd 4f magnetism. In addition, we also note an interesting feature at low temperatures, viz., a positive contribution dominating below 12 K. The extension of present studies to a few La and Y substituted alloys of GdNi also enabled us to identify the influence of Gd sublattice dilution, crystal structure and the chemical pressure effects on the observed features.

Results and Discussion

The series RNi presents an interesting scenario both from the crystal structure and magnetism point of view. While La, Ce, Pr , Nd, Gd and Tb compounds of this series have been reported to crystallize in the CrB-type orthorhombic structure, the heavy rare earth members (Dy to Tm as well as Y) form in the FeB-type orthorhombic structure.[7−10] We have recently shown[11,12] that the exchange interaction is stronger in the FeB structure than in the CrB structure. While it is known that Ni does not possess any magnetic moment in light R members of the series RNi, an additional moment beyond the value expected for trivalent Gd was found in GdNi, in the para as well as in the ferromagnetic states, induced by Gd presumably in Ni 3d band. On the basis of the magnetization behavior, we have proposed[11,12] the coexistence of localised magnetism (from Gd 4f orbital) and induced itinerant magnetism in the Ni 3d band in the magnetically ordered state.

The alloys chosen for the present study, GdNi, $Gd_{0.75}R_{0.25}Ni$ and $Gd_{0.5}R_{0.5}Ni$ (R= Y and La), were the same as those investigated previously.[12] The polycrystalline samples were rectangular in shape (about 2mm $\times$ 2mm cross section; voltage leads separation about 2-4 mm). The magnetoresistance, $\Delta\rho/\rho[= \{\rho(H) - \rho(0)\}/\rho(0)]$, data were obtained as a function of temperature in the range 4.2 - 300 K in the presence of an external magnetic field (H) of 80 kOe and also as a function of H at select temperatures in the longitudinal mode. We have employed a constant current typically of 10 mA and we notice that the results are reproducible with higher excitation currents (e.g., 50 mA). The voltages were measured to an accuracy of 10 nV. A conducting silver epoxy was used to make the electrical contacts of the leads with the samples. The values of the resistivity for all the alloys at 300 K are the same, typically 200 $\mu\Omega$ cm, within the estimated error of about 40%. This error arises due to uncertainties in measurement of the separation between voltage leads, caused by spread of the silver paste. Due to this reason, only the resistance data is presented in the figures.

The temperature dependence of ρ in an applied field of 80 kOe as well as in zero field, along with the temperature dependence of $\Delta\rho/\rho$ obtained from this data, are shown in Fig. 1 for GdNi. Since the value of $\Delta\rho/\rho$ above 200 K is negligible, the data are shown only below this temperature. The value of $\Delta\rho/\rho$, though small, is negative around 140 K. Its magnitude progressively grows as T_C is approached from higher temperatures, exhibiting a maximum (about -20%) at T_C. This is followed by a continuous fall in magnitude as the temperature is lowered further. The temperature dependence of $\Delta\rho/\rho$ observed here mimics the behavior reported in La manganates in bulk form.[13] Though the magnitude of $\Delta\rho/\rho$ at T_C is not as large as in La manganates, it is still large for a metal, considering that the Lorentz force on the conduction electrons is generally known to cause a positive value of $\Delta\rho/\rho$ of less than a few percent. While the behavior in La manganates and the amorphous magnetic rare earth silicon alloys[1–4] is related to the metal-insulator transition at T_C, that in GdNi cannot be due to such a factor, as it remains metallic in the entire temperature range of investigation. As in the case of $GdNi_2Si_2$ (Ref. 5), the negative value suggests that there are spin fluctuations in the Ni 3d band induced by Gd 4f magnetic moment, that are suppressed by the application of the magnetic field. The measured effective moment is noticeably larger than that expected for trivalent Gd ion and we have proposed[12] that the induced moment (in other words, some kind of polaron) above T_C is of a localised type. The localised magnetic moment proposed to be from Ni 3d orbital was deduced[12] to be close to $3\mu_B$. The increasing magnitude of $\Delta\rho/\rho$ with a *negative sign* as T_C is approached from higher temperatures indicates a gradual increase in the suppression of spin fluctuations (from the polarons) by the applied field. To understand the origin of polaron formation, we propose the existence of short range (of the order of mean free path of the conduction electrons) magnetic ordering in Gd sublattice. The heat capacity results[8,12] provide evidence for this proposal in the sense that the magnetic contribution to heat capacity has been shown to exhibit[8,12] a continuous upturn over a wide temperature range before long range magnetic order sets in. The Ni 3d orbital is more prone to polarisation by this short range order, as demonstrated[5] by a comparative study of $GdCu_2Si_2$ and $GdNi_2Si_2$; the short range order does not seem to polarise the Cu 3d band as the magnetoresistance before long range magnetic order sets in is positive in this Cu alloy. Apparently, the 5d and 6s electrons do not contribute significantly to the polaron formation, as otherwise one should have seen negative $\Delta\rho/\rho$ above T_N in $GdCu_2Si_2$ as well. Thus, there is some justification for our proposal that the polarons are constituted mainly by

Ni 3d orbital. The present results also emphasize that the short range magnetic order (from Gd), not necessarily long range order, is sufficient to cause this polaronic-like effect. Well below T_C, the polarization cloud apparently becomes itinerant, coupling ferromagnetically with the Gd 4f moment;[12] as a result the (localised) polaronic contribution is turned off below T_C, as proposed by Millis et al[3] for $La_{1-x}Sr_xMnO_3$. Therefore, below T_C, this prominent source for negative contribution to $\Delta\rho/\rho$ diminishes.

An interesting feature observed here is the change in the sign of $\Delta\rho/\rho$ below 12 K (see Fig. 1). To get a better view of the sign and non-monotonic variation of the magnitude of $\Delta\rho/\rho$ with temperature, we present $\Delta\rho/\rho$ as a function of field at various temperatures in Fig. 2. In a field of 60 kOe, the values of $\Delta\rho/\rho$ are about -7%, -15%, -7%, zero, and 12% at 95, 50, 20, 12 and 4.2 K respectively. These values are well outside the experimental error of $\pm1\%$ (obvious from the scatter in the values as a function of field). For instance, for GdNi, at 4.2 K, the observed voltage difference for H= 0 and H= 60 kOe is typically 1μV, which is far above the sensitivity of the nanovoltmeter; thus the magnetoresistance values are reliable within the experimental error. In the paramagnetic state, for instance, at 95 K, it is clearly negative (presumably varying as H^2) as expected for spin fluctuating systems. The positive contribution however appears at 4.2 K. The positive peak in the low field data for 20 K is of no significance considering an error of $\pm1\%$. The observation of positive sign of magnetoresitance at 4.2 K with a large magnitude at higher fields is quite interesting and reveals the existence of two competing contributions to $\Delta\rho/\rho$. These two contributions appear to be of nearly equal magnitude at 12 K, as the values are close to zero at all fields at this temperature. The competition between these interactions is visible as a shoulder in $\Delta\rho/\rho$ around 25 K (Fig. 1). The sign crossover and large magnitude have been found by us at low temperatures well below T_C in other ferromagnetic systems (from Mn sublattice) like $SmMn_2Ge_2$ (Ref. 14) and $LaMn_2Ge_2$ (Ref. 15). It is also possible that the negative contribution from domain walls is frozen around 25 K, as a result of which the positive contribution from conduction electrons dominates. It is of interest to explore whether the application of the field causes an unusual effect of inducing gaps in some portions of the Fermi surface, resulting in changes in the band structure. The data on La and Y substituted alloys presented below appear to favor this view, though more work is required to completely understand this interesting aspect of $\Delta\rho/\rho$.

The results on the La and Y substituted alloys, $Gd_{0.75}La_{0.25}Ni$ (T_C= 52 K), $Gd_{0.5}La_{0.5}Ni$ (T_C= 35 K), $Gd_{0.75}Y_{0.25}Ni$ (T_C= 55 K) and $Gd_{0.5}Y_{0.5}Ni$ (T_C= 65 K) (Refs. 11, 12), are shown in Fig. 3. While the CrB-type structure is maintained in the former three alloys, the last one crystallizes in the FeB type structure.[10–12] While La expands the lattice, Y compresses the lattice. Thus the data on these alloys is useful to identify the influence of crystal structure and chemical pressure effects on the observed features. The common feature in all these alloys is that $\Delta\rho/\rho$ peaks at T_C. The magnitude decreases with the decrease in the Gd concentration, rendering support to our idea that the observed $\Delta\rho/\rho$ effect is Gd induced (polaronic-like) effect; apparently La substitution is more effective compared to Y in depressing the magnitude of $\Delta\rho/\rho$. The temperature below which the polaronic-like effect dominates appears to be about $2T_C$, as in the case of GdNi. There are some qualitative differences in the low temperature range (<12 K). It is apparent that, in all these substituted alloys, the value of $\Delta\rho/\rho$ remains negative even below 12 K. We have also slightly changed the Gd/Ni composition by about 5% and the $\Delta\rho/\rho$ remains negative (not shown here) down to 4.2 K. These findings suggest that the low temperature positive $\Delta\rho/\rho$ behavior in stoichiometric GdNi may be related to subtle band structure effects at the Fermi level (E_F), which get modified by the shift of E_F by the unit cell volume changes.

Conclusion

In summary, we have reported here a gradual increase of negative magnetoresistance below twice of Curie temperature till the onset of long range magnetic ordering in GdNi. We attribute this to some kind of magnetic polaronic contribution. Thus, this article reports polaronic-like effects in the magnetoresistance in a ferromagnetic rare earth *intermetallic* compound, that is, a material in which neither double-exchange nor Jahn-Teller mechanisms are expected to be operative. The findings suggest that the contribution of spin fluctuations from magnetic polarons may have to be considered in the giant magnetoresistance systems as well. In addition, for $T << T_C$, positive values are noted in this compound.

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Figure Captions

FIG. 1. Electrical resistance as a function of temperature for GdNi in the presence and in the absence of a magnetic field of 80 kOe. The magnetoresistance ($\Delta\rho/\rho$) obtained from this data are also shown.

FIG. 2. The magnetoresistance ($\Delta\rho/\rho$) as a function of magnetic field at various temperatures for GdNi. The lines (except for 12 K) through the data points serve as a guide to the eye. The experimental error in the values at low temperatures ($\leq$ 20 K) is larger ($\pm$1%) than at higher temperatures due to smaller values of resistance. Though the low field peak for 20 K is not genuine due to this reason, we have drawn a line through the data points for this temperature as well for the sake of clarity.

FIG. 3. Electrical resistance as a function of temperature for $Gd_{1-x}R_xNi$ (R= Y and La; x= 0.25 and 0.5) in the presence and in the absence of a magnetic field of 80 kOe. The magnetoresistance ($\Delta\rho/\rho$) obtained from this data are also shown.

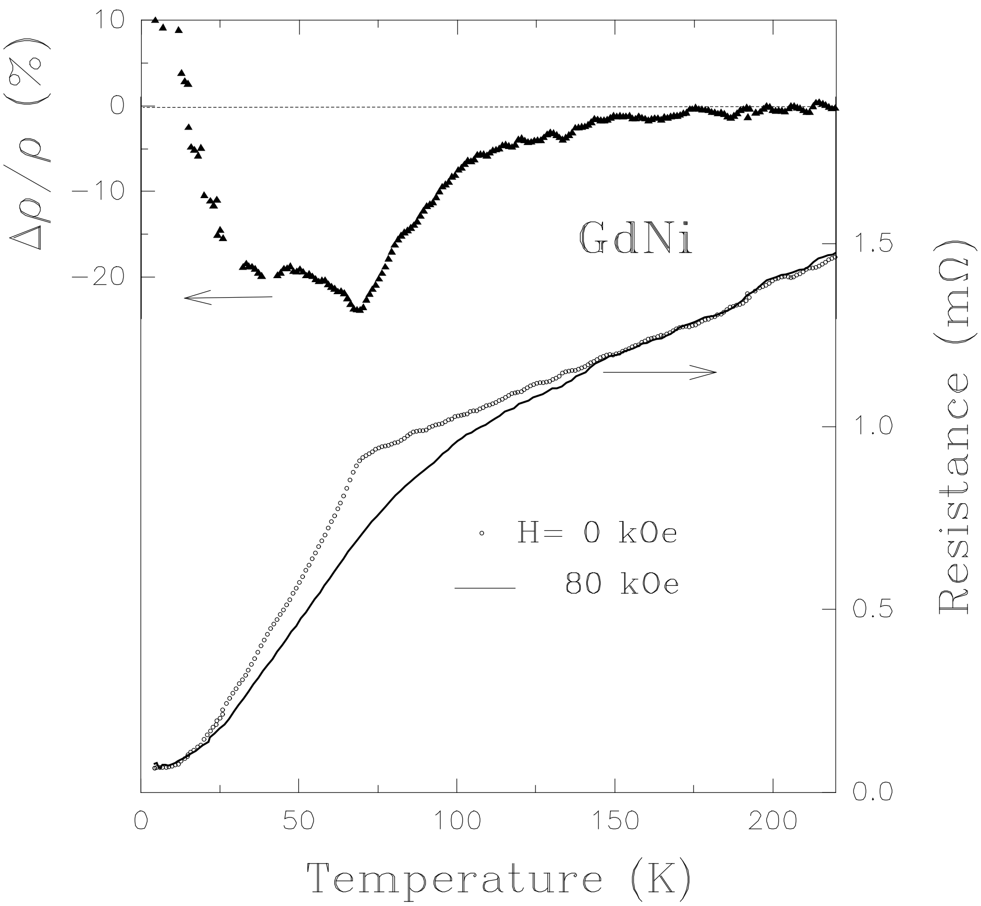

Fig 1 : Mallik et. al.

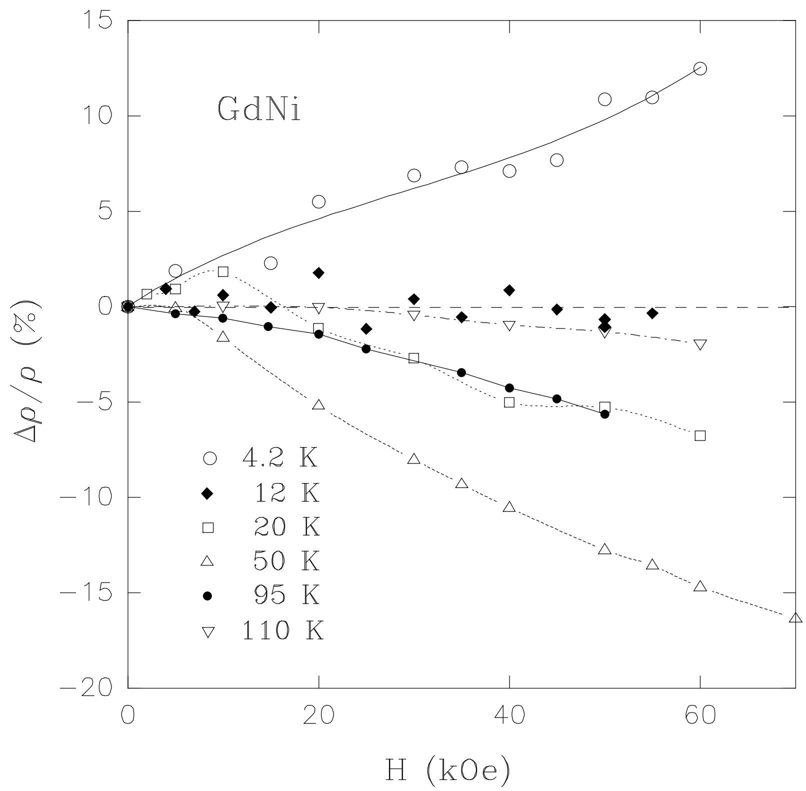

Fig 2 : Mallik et. al.

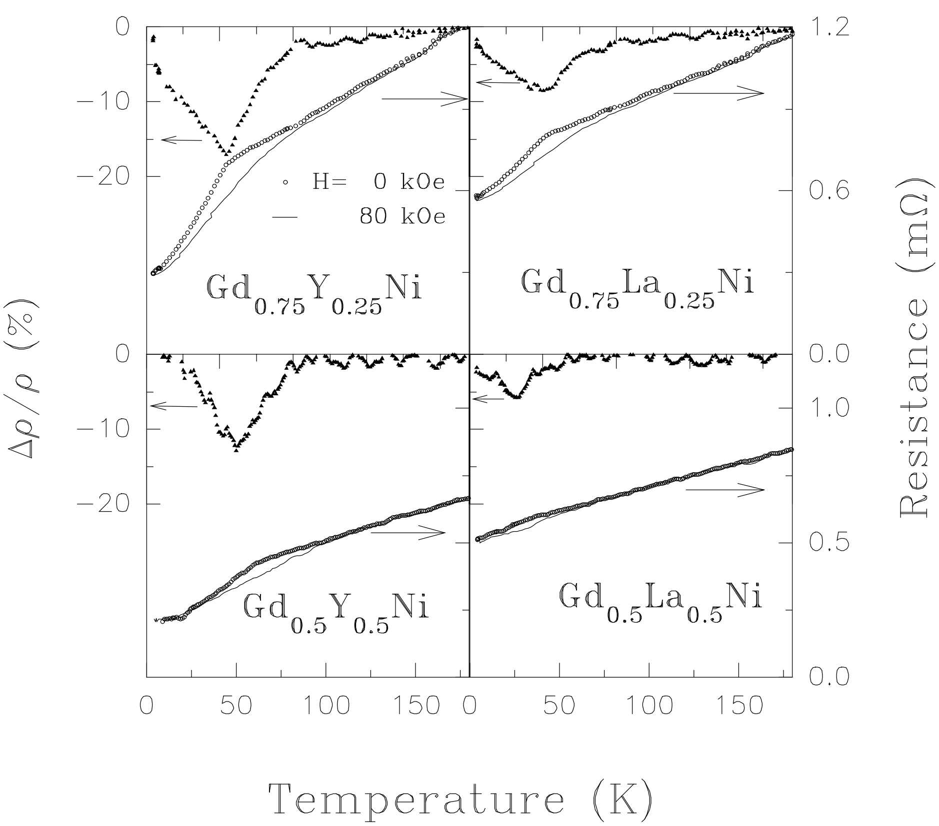

Fig 3 : Mallik et. al.