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Anomalous magnetoresistance in antiferromagnetic polycrystalline materials \( R_2Ni_3Si_5 \) (\( R=\text{rare earth} \))

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Magnetoresistance (MR) studies on polycrystalline \( R_2Ni_3Si_5 \) (\( R=\text{Y, rare earth} \)) which order antiferromagnetically at low temperatures, are reported here. MR of the Nd, Sm, and Tb members of the series exhibit positive giant magnetoresistance, largest among polycrystalline materials (85%, 75%, and 58% for \( \text{Tb}_2\text{Ni}_3\text{Si}_5 \), \( \text{Sm}_2\text{Ni}_3\text{Si}_5 \), and \( \text{Nd}_2\text{Ni}_3\text{Si}_5 \), respectively, at 4.4 K in a field of 45 kG). These materials have, to the best of our knowledge, the largest positive GMR reported ever for any bulk polycrystalline compounds. The magnitude of MR does not correlate with the rare earth magnetic moments. We believe that the structure of these materials, which can be considered as a naturally occurring multilayer of wavy planes of rare earth atoms separated by Ni–Si network, plays a role. The isothermal MR of other members of this series (\( R=\text{Pr}, \text{Dy}, \text{Ho} \)) exhibits a maximum and a minimum, below their respective \( T_N \)’s. We interpret these in terms of a metamagnetic transition and short-range ferromagnetic correlations. The short-range ferromagnetic correlations seem to be dominant in the temperature region just above \( T_N \).

We had earlier reported interesting phenomena in several members of the series \( R_2Ni_3Si_5 \). Our work had earlier revealed that Ce and Eu ions in \( \text{Ce}_2\text{Ni}_3\text{Si}_5 \) and \( \text{Eu}_2\text{Ni}_3\text{Si}_5 \), respectively, exhibit valence fluctuation behavior.\(^1\)\(^2\) Lu\(_2\)Ni\(_3\)Si\(_5 \) becomes a superconductor below 2 K (Ref. 3) and \( \text{Tb}_2\text{Ni}_3\text{Si}_5 \) exhibits a double magnetic transition with \( T_{N1} = 19.5 \) K and \( T_{N2} = 12.5 \) K.\(^4\) Considering these interesting phenomena, we studied magnetic properties of other members of the series. We found that \( \text{Pr}_2\text{Ni}_3\text{Si}_5 \), \( \text{Nd}_2\text{Ni}_3\text{Si}_5 \), \( \text{Sm}_2\text{Ni}_3\text{Si}_5 \), \( \text{Gd}_2\text{Ni}_3\text{Si}_5 \), \( \text{Dy}_2\text{Ni}_3\text{Si}_5 \), and \( \text{Ho}_2\text{Ni}_3\text{Si}_5 \) undergo antiferromagnetic transition at 8.7, 9.5, 11.1, 14.7, 9.5, and 6.5 K, respectively.\(^4\)\(^6\) The \( \chi(T) \) of \( \text{Dy}_2\text{Ni}_3\text{Si}_5 \) also shows a signature of a second magnetic transition around 4 K (lower inset of Fig. 1). Unlike other members of this series, \( \chi(T) \) of \( \text{Ho}_2\text{Ni}_3\text{Si}_5 \) exhibits a tail below its \( T_N \) (Fig. 1). We believe that this may be arising from a change in magnetic structure at low temperatures. The likelihood of such a situation is also indicated by the hysteresis (not exactly that of ferromagnetism) seen in isothermal magnetization at 6 K, which is just below magnetic ordering.\(^7\) A significant magnetic entropy seen from specific heat measurements below 4 K also supports this view.\(^8\) Here we present our longitudinal magnetoresistance (MR) results on polycrystalline \( R_2Ni_3Si_5 \) in the temperature range 4.4–100 K and in the magnetic field range 0–45 kG. Some aspects of this work have been reported in our very recent publications.\(^7\)\(^9\)

Since magnetic measurements of all the materials show the materials to order antiferromagnetically, one expects the MR of the materials to be positive.\(^10\) The MR of \( R_2Ni_3Si_5 \) (\( R=\text{Nd,Sm,Tb} \)) has been found to exhibit such a behavior, although the magnitude of MR is anomalously large.\(^9\) At 4.4 and a field of 45 kG, the magnetoresistance of \( \text{Nd}_2\text{Ni}_3\text{Si}_5 \), \( \text{Sm}_2\text{Ni}_3\text{Si}_5 \) and \( \text{Tb}_2\text{Ni}_3\text{Si}_5 \) is 58%, 75%, and 85%, respectively.\(^9\) Figure 2 (left) shows the MR behavior of \( \text{Nd}_2\text{Ni}_3\text{Si}_5 \). For a bulk polycrystalline antiferromagnetic material, these values are quite high, and to the best of our knowledge, the largest positive GMR reported ever for any bulk polycrystalline compounds. We also find that the magnetoresistance values do not scale with the magnetic moment of the rare earth.\(^9\) We believe that the structure of these materials, which can be considered as a naturally occurring multilayer of wavy planes of rare earth atoms separated by Ni–Si network [Fig. 2 (right)] plays a role.

![Fig. 1. Magnetic susceptibility and its inverse of \( \text{Ho}_2\text{Ni}_3\text{Si}_5 \). Insets shows the expanded region near the magnetic ordering temperature of \( \text{Ho}_2\text{Ni}_3\text{Si}_5 \) and \( \text{Dy}_2\text{Ni}_3\text{Si}_5 \).](image)
In contrast, MR of other magnetic members of this series \( R_2 Ni_3 Si_5 \) (R=Pr,Dy,Ho) exhibit anomalous behavior in some other way. Figure 2 (middle) shows the MR behavior of \( Dy_2 Ni_3 Si_5 \). Below their respective \( T_N \) ’s, the isothermal MR correspond to metamagnetic transition. The peak in MR disappears above \( T_N \), although the minimum still persists. At temperatures well above \( T_N \), MR exhibits a monotonic behavior w.r.t. magnetic field.

The observed minimum in the field dependence of MR may be arising from two competing contributions: A term linear in magnetic field and another, a negative term due to a ferromagnetic component. For example, a ferromagnetic component has been seen in the structurally related compound \( U_2 Ru_{0.65} Rh_{0.35} Si_5 \), where neutron diffraction study have shown that the magnetic moments are arranged ferromagnetically in a plane, whereas these planes are coupled antiferromagnetically.11

We find a significant linear component in the MR of these materials which is an important observation. In the nonmagnetic analogue, \( Y_2 Ni_3 Si_5 \) has an unusually large linear component,7,9 and we expect it to be present in all the compounds. We believe that this linear term, arises from the \( RNiSi_3 \) blocks \( (RNi_3Si_5 \) structure can be considered to be consisting of \( RNiSi_3 \) and \( RNiSi_3 \) type blocks12). A positive linear MR has earlier been observed in \( PrCu_2 Si_2 \).13 We believe that this linear term could be a general feature of \( RM_2X_5 \) type of compounds.

The temperature dependence of MR of these compounds (in a constant high field), exhibit a pronounced minimum at the magnetic ordering temperature of the corresponding compound. Above \( T_N \), the MR exhibit a broad maximum, which may be due to the presence of short range ferromagnetic correlations in these materials. Precursor effects seen in the magnetic entropy in our specific heat measurements on these compounds, also support this conjecture.8

As we have suggested above, for \( T>T_N \), short range ferromagnetic correlations exist in these materials. This effect is more prominent for those having R=Pr,Dy,Ho. To understand the MR in this regime, we adopt the following approach assuming that short range ferromagnetic correlations could be treated in the same manner as in superparamagnetic systems. Berkowitz et al.14 and Xiao et al.15 showed that in certain heterogeneous alloys, in thin film form, where ferromagnetic particles are embedded in a metallic matrix and forming nanocrystalline materials, can produce large negative MR which is proportional to square of magnetization of the material. Helmolt et al. found this to hold good for the system, \( Au_{20}Co_{20}B_{10} \),16 where they assume that the small ferromagnetic clusters have a superparamagnetic behavior (i.e., where the magnetic energy in an applied field is comparable to thermal energy17,18) and showed that MR can be expressed as proportional to the square of Langevin function.16

We attempt to apply the same approach in our case of \( R_2 Ni_3 Si_5 \) (R=Pr, Dy, Ho) by assuming that the short range ferromagnetic interactions present in our case also could be treated as in a superparamagnetic system. From the \( M vs H \)
function with power proved fit we replaced the quadratic Langevin function by a satisfactory fit to the observed MR. In order to get an im-
linear term. However, this relation, i.e., square of Langevin
As mentioned earlier, the field dependence of MR also has a middle
is positive.

The above expression gives a better fit to the data
Fig. 2 middle). Using the value of \( \mu \) obtained from the magnetization
curve, we find that just above the transition temperature, the value of \( n \) is nearly 4/3. As the temperature is increased, the value of \( n \) gets reduced and tends toward 1. At present, we do not know the reason for value of \( n \) being different from 2 and its temperature dependence.

We may also like to comment on the temperature dependence of the resistivity of these antiferromagnetic materials. The resistivity, \( \rho \), of these compounds exhibit normal metallic behavior. As a typical example, \( \rho(T) \) of \( \text{Tb}_2\text{Ni}_3\text{Si}_5 \) is given in the inset of Fig. 3. \( \rho(T) \) of \( \text{Tb}_2\text{Ni}_3\text{Si}_5 \) exhibits change of slope at \( \sim 19.5 \text{~K} \) and at \( \sim 13 \text{~K} \) (Fig. 3) which correspond to \( T_N^1 \) and \( T_N^2 \), respectively, as observed in the \( \chi(T) \) measurements. Below \( T_N^1 \), we find that the resistivity can be fit to the expression: 

\[
\rho(T) = \rho_0 + a T^2 + \beta T \left[ 1 + \frac{2T}{\Delta} \right] \exp \left( -\frac{\Delta}{T} \right). 
\]

Here the second term originates from the spin fluctuations coming from Fermi-liquid consideration, and the third term is due to electron spin wave scattering in an antiferromagnetic material having an energy gap \( \Delta \). The fit results: \( \rho_0 = 3.35 \mu\Omega \text{~cm} \), \( a = -6 \times 10^{-3} \mu\Omega \text{~cm} \text{~K}^{-2} \), \( \beta = 1843 \mu\Omega \text{~cm} \text{~K}^{-1} \) and \( \Delta = 119 \text{~K} \). The resistivity rises much faster in the temperature range 12–13 K than predicted by the model, indicating the presence of some additional scattering mechanism in this region. In the temperature range 13–19.5 K, a linear fit yields \( \rho(T) = 0.53T \mu\Omega \text{~cm} \text{~K}^{-1} \), whereas a better fit can be obtained as \( \rho(T) = 4 \mu\Omega \text{~cm} + 0.17T^2 \mu\Omega \text{~cm} \text{~K}^{-2} \) (Fig. 3).

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\[\text{FIG. 3. Expanded region of resistivity of } \text{Tb}_2\text{Ni}_3\text{Si}_5 \text{ near the transition tempera-} \] 

\[\text{tures. Below } T_N^1, \text{ the solid line represents a fit with Eq. (2). In the temperature range, } T_N^1 - T_N^2, \text{ the solid line represent a quadratic behavior. Inset shows the resistivity in the temperature range 4.2–300 K.}\]