Magnetoresistance anomalies in the Gd-based alloys

E. V. Sampathkumaran and I. Das
Tata Institute of Fundamental Research, Homi Bhabha Road, Bombay 400 005, India
(Received 29 November 1994)

The magnetoresistance ($\Delta\rho/\rho$) behavior across magnetic ordering temperatures in the Gd alloys, $GdCu_2Si_2$, $Gd_{1-x}Y_xNi_2Si_2$, $GdPt_2Si_2$, and $GdPd_2In$, is presented. In all of these alloys, except $GdCu_2Si_2$, $\Delta\rho/\rho$ is negative in the paramagnetic state and its magnitude grows as the respective magnetic ordering temperatures are approached. On the other hand, $\Delta\rho/\rho$ is positive in the case of $GdCu_2Si_2$ both above and below the Néel temperature. The findings noted for the Gd alloys containing Ni, Pt, and Pd are interesting, presumably arising from spin fluctuations from the transition-metal d bands.

In the case of Ce, Yb, and U-based alloys, the observation of large magnetoresistance ($\Delta\rho/\rho$) varying quadratically with the application of magnetic field (H) has been usually attributed to the f-ion Kondo effect arising from the proximity of the f level to the Fermi level.[1] However, very few attempts have been made to probe the $\Delta\rho/\rho$ behavior in the paramagnetic state of materials containing well-localized f electrons and also to look for the spin fluctuation effects from the transition-metal d bands. In this article, we present the results of $\Delta\rho/\rho$ measurements on Gd-containing materials. Besides various observations, the main finding is that $\Delta\rho/\rho$ in some of these Gd alloys somewhat mimics the behavior noted for Kondo systems, presumably arising from the spin fluctuation effects from the transition-metal d bands polarized by Gd f electrons.

The alloys chosen for our investigation are $GdCu_2Si_2$, $GdNi_2Si_2$ (both crystallizing in the $ThCr_2Si_2$-type tetragonal structure, Ref. 2), $GdPt_2Si_2$ ($CaBe_2Ge_2$-type tetragonal structure, Ref. 3), and $GdPd_2In$ (Heusler-type cubic structure, Ref. 4). All these compounds, except $GdPd_2In$, order antiferromagnetically below $T_N=14.5$, 14.5, and 10 K, respectively;[5] the magnetic ordering in $GdPd_2In$ below about 7 K is ferromagneticlike,[6] the exact nature of which is yet to be resolved. In addition, the alloys, $Gd_{1-x}Y\ Ni_2Si_2$ ($x=0.2$ and 0.4) were also investigated to probe the Gd-sublattice dilution effects on the observed behavior.

The samples were prepared by arc melting stoichiometric amounts of constituent elements. The samples were characterized by x-ray diffraction and were found to be single phase. The longitudinal $\Delta\rho/\rho\{[\rho(H)-\rho(0)]/\rho(0)\}$ measurements in the temperature interval 4.2–40 K were carried out in zero field as well as in 50 kOe; in addition, the data were obtained as a function of H up to 70 kOe at 5 and 20 K.

The results of the electrical resistance (ρ) and $\Delta\rho/\rho$ measurements are shown in Figs. 1–4. Initial results on an older $GdNi_2Si_2$ sample have been reported earlier[7] and the rise in ρ noted below 20 K earlier is not prominent in

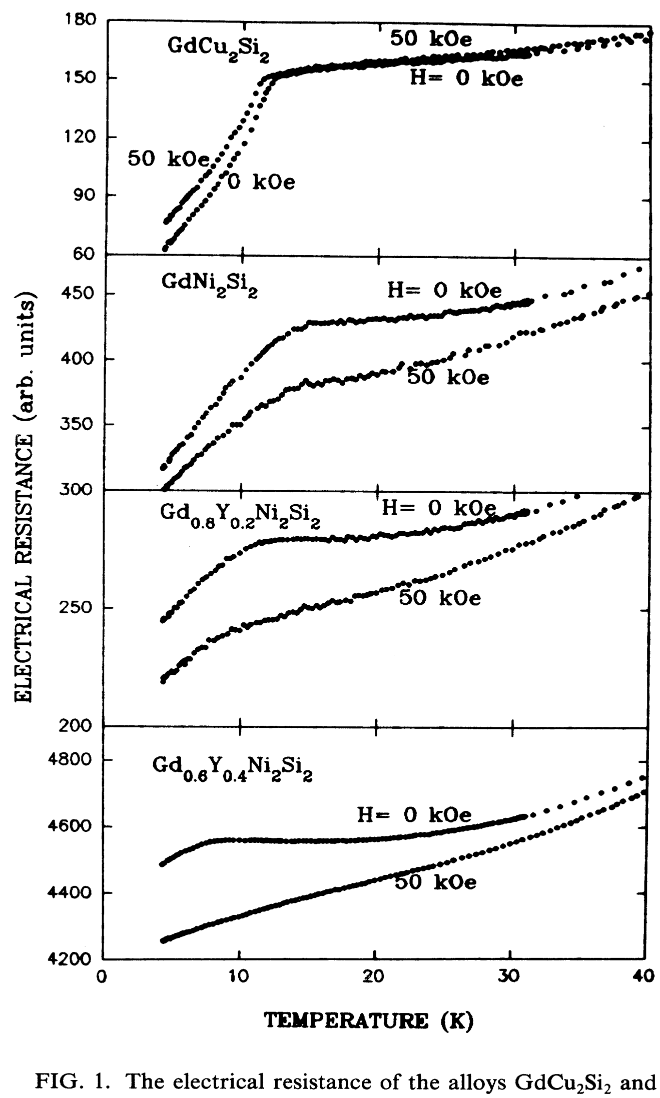

FIG. 1. The electrical resistance of the alloys $GdCu_2Si_2$ and $Gd_{1-x}Y_xNi_2Si_2$ ($x=0.0$, 0.2, and 0.4) in the temperature interval 5–40 K in zero field and in the presence of a field of 50 kOe.

the freshly prepared specimen (see Fig. 1). Distinct features at the magnetic ordering temperatures could be seen in all cases due to the loss of spin-disorder contribution in the plots of ρ versus T. In the case of $GdPt_2Si_2$, ρ exhibits a rise at the onset of antiferromagnetic ordering giving rise to a peak due to the formation of antiferromagnetic energy gap[8] and the application of a field of 50 kOe reduces the value of T_N from 10 to 8 K (Fig. 3). For $GdCu_2Si_2$ and $GdNi_2Si_2$, the application of H does not cause any apparent shift of T_N (Fig. 1). In the Y-substituted Ni alloys, ρ smoothly decreases as the temperature is lowered in the presence of $H=50$ kOe and hence it is not possible to infer the shift of T_N caused by this magnitude of field.

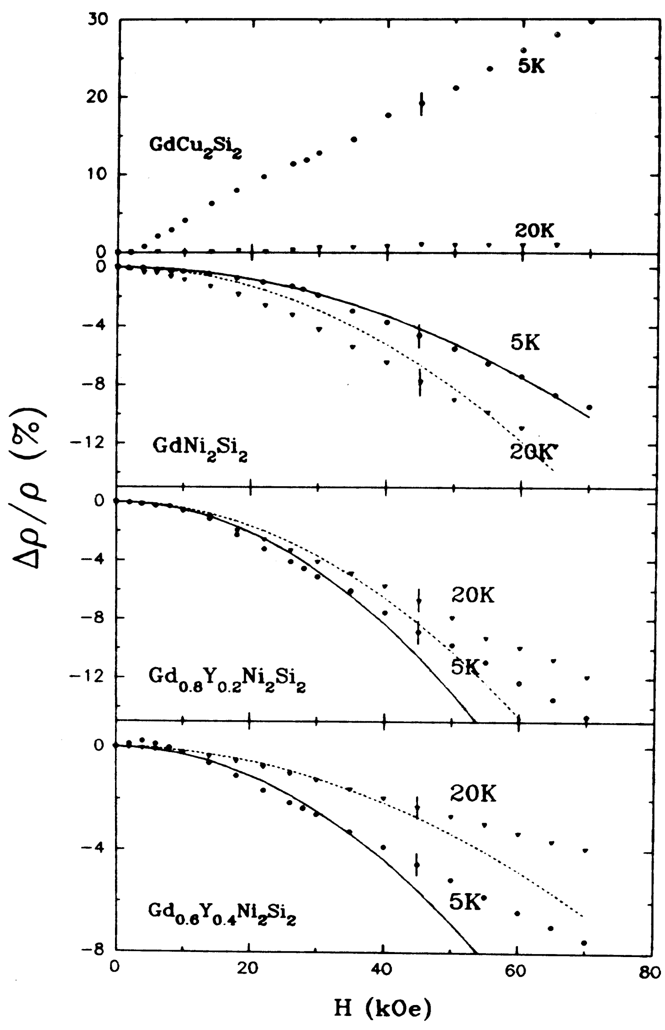

FIG. 2. The magnetoresistance of $GdCu_2Si_2$ and $Gd_{1-x}Y_xNi_2Si_2$ as a function of field at 5 K (circle) and 20 K (triangle). In the case of Ni-containing alloys, the fits to the function, $\Delta\rho/\rho=aH^2$ (where a is a constant) are shown by solid and broken lines for 5 and 20 K, respectively. For $x=0.2$ and 0.4, the fit is restricted to the data below 40 kOe so as to highlight the deviation from the quadratic dependence at higher fields. Vertical bars denote the error.

Now, we bring out the finding of central importance in our data. In the case of $GdCu_2Si_2$, $\Delta\rho/\rho$ is positive in the antiferromagnetically ordered state as expected.[9] It is not out of place to mention that the magnitude of $\Delta\rho/\rho$ is quite large at 5 K (about 30% in 70 kOe). The positive sign of $\Delta\rho/\rho$ persists in the paramagnetic state, thereby implying that the magnitude of the paramagnetic Gd-spin fluctuation contribution (which is negative) should be small enough compared to the positive contribution arising from the influence of H on the Fermi surface. On the other hand, in the case of $GdNi_2Si_2$, $\Delta\rho/\rho$ is negative in the paramagnetic state and its magnitude grows with decreasing temperature (Fig. 1); the magnitude of $\Delta\rho/\rho$ is large, for instance, about 15% at 20 K in the presence of $H=70$ kOe, varying almost quadratically with H (Fig. 2). This behavior, typical of Kondo spin fluctuation phenomena, cannot be attributed to the Gd-ion Kondo effect due to the well-localized character of Gd f electrons, but must be attributed to the Ni $3d$ band. It may be recalled that the transition-metal ions (except Mn) are known not to possess any magnetic moment in the compounds crys-

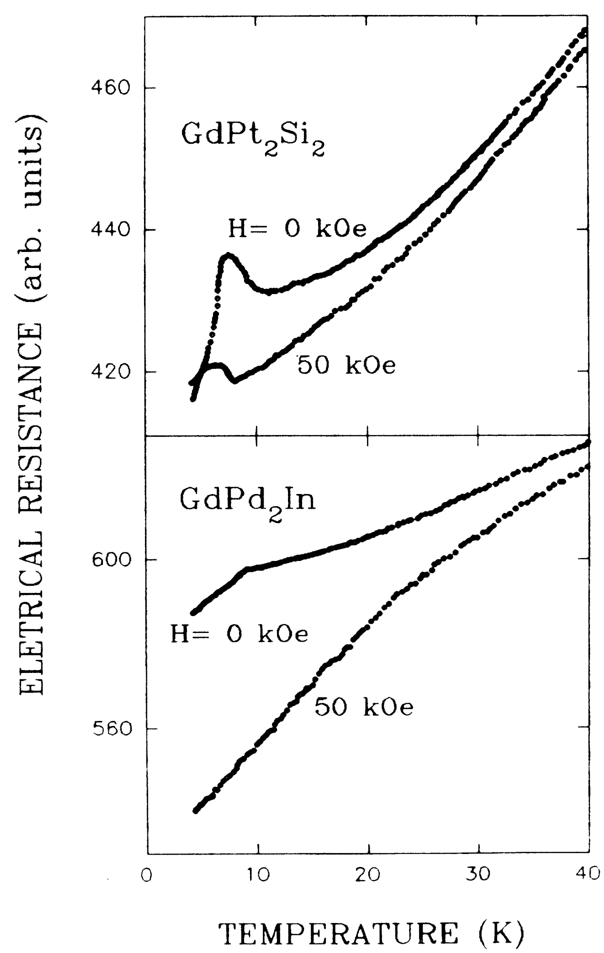

FIG. 3. The electrical resistance of $GdPt_2Si_2$ and $GdPd_2In$ in the temperature interval 5–40 K in zero field and in the presence of a field of 50 kOe.

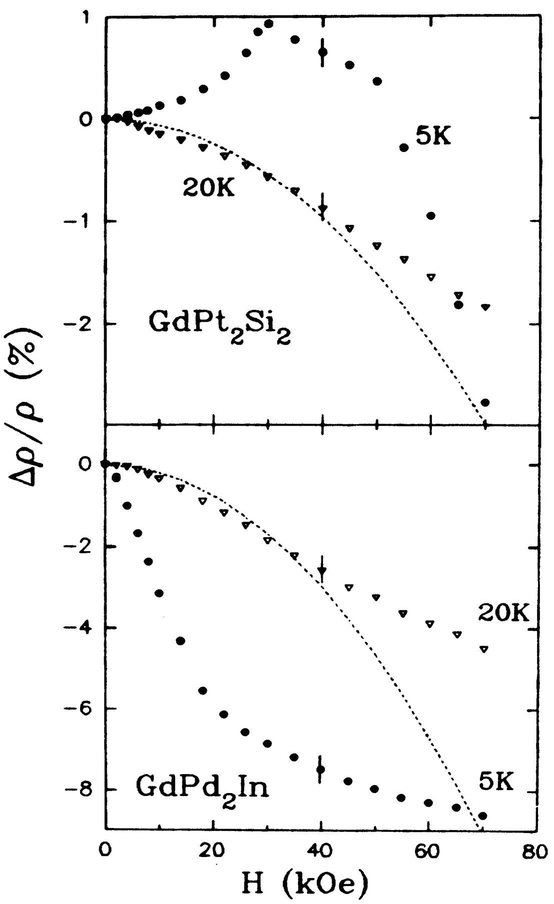

FIG. 4. The magnetoresistance of $GdPt_2Si_2$ and $GdPd_2In$ as a function of field at 5 K (circle) and 20 K (triangle). The fits to the function, $\Delta\rho/\rho = aH^2$ (where a is a constant) for the data below 40 kOe at 20 K, are shown by broken lines. Vertical bars denote the error.

tallizing in this type of structure.[5] Therefore, we propose that Gd moment polarizes the Ni 3d band thereby causing spin fluctuations. A point to note is that $\Delta\rho/\rho$ varies quadratically even at 5 K, as if the Ni 3d band spin fluctuation persists so as to dominate $\Delta\rho/\rho$ even in the magnetically ordered state (Fig. 2). In order to find out the sensitivity of these features to Gd concentration, the experiments were performed on the Y-substituted alloys, $Gd_{1-x}Y_xNi_2Si_2$ ($x = 0.2$ and 0.4). It is apparent that the features noted for $x = 0.0$ persist even when the Gd sublattice is diluted by Y, though the field dependence of $\Delta\rho/\rho$ tends to deviate from quadratic form for $H > 40$ kOe.

With respect to $GdPt_2Si_2$ and $GdPd_2In$, the response of ρ to the application of H in the paramagnetic state is qualitatively the same as that noted for $GdNi_2Si_2$ (Figs. 3 and 4). The $\Delta\rho/\rho$ is negative varying quadratically with H (below 40 kOe) as if Pt and Pd bands exhibit spin fluctuations. The magnitude however is relatively small compared to that of $GdNi_2Si_2$. With respect to the magnetically ordered state, the $\Delta\rho/\rho$ behavior as a function of H at 5 K is different from that of $GdNi_2Si_2$. For the Pt sample, the values are positive up to 30 kOe typical of antiferromagnets; for higher values of H, it appears that the Gd sublattice tends to become ferromagnetic as indicated by negative $\Delta\rho/\rho$. In the case of $GdPd_2In$, the nature of the spin alignment is believed to be very complex[6] and the plot of $\Delta\rho/\rho$ versus H at 5 K mimics that of weak ferromagnetic materials of itinerant type.[10] Since Gd is not expected to exhibit itinerant magnetism, this observation may have to be attributed to Pd d-band itinerant magnetism. In short, our observations render strong evidence for an earlier proposal[6] that Pd d band undergoes spin fluctuation above 7 K and the magnetism may be of complex type due to the coexistence of itinerant magnetism from Pd d band and localized magnetism from Gd f electrons.

To conclude, we have made interesting observations on the magnetoresistance behavior of a number of Gd alloys. Particularly, the results indicate the existence of spin fluctuations from the Ni, Pd, and Pt bands. If this additional set of competing interaction arising from transition metal bands persists in Ce and U systems, the understanding of the physics of these systems would be more complex.

[1]U. Rauchschwalbe, Physica B **147B**, 1 (1987).

[2]W. Rieger and E. Parthe, Monatsch. Chem. **100**, 444 (1969).

[3]K. Hiebl and P. Rogl, J. Magn. Magn. Mater. **50**, 39 (1985).

[4]M. Ishikawa, J. L. Jorda, and A. Junod, in *Proceedings of the International Conference on Superconductivity in d- and f-based Metals,* edited by W. Buckel and W. Weber (KFZ, Karlsruhe, 1982), p. 141.

[5]A. Szytula, in *Handbook of Magnetic Materials,* edited by K. H. J. Buschow (Elsevier Science, New York, 1991), Vol. 6, p. 85.

[6]K. U. Neumann, J. Crangle, R. T. Giles, D. Visser, N. K. Zayer, and K. R. A. Ziebeck, Solid State Commun. **84**, 577 (1992).

[7]I. Das and E. V. Sampathkumaran, Phys. Rev. B **49**, 3972 (1994).

[8]I. Das, E. V. Sampathkumaran, and R. Vijayaraghavan, Phys. Rev. B **44**, 159 (1991), and references therein.

[9]K. A. McEwen, in *Handbook on the Physics and Chemistry of Rare-Earths,* edited by K. A. Gschneidner, Jr. and L. Eyring (North-Holland, Amsterdam, 1978), Vol. 1, p. 411.

[10]T. Moriya, *Spin Fluctuations in Itinerant Electron Magnetism* (Springer-Verlag, Berlin, 1985).