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# *Lu*Gd: A positive–exchange-constant Kondo system

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We report transport and magnetic measurements in dilute alloys of *Lu*Gd which provide the first unambiguous observation of a ferromagnetic coupling between conduction electrons and isolated local moments.

Antiferromagnetic exchange coupling between localized magnetic moments and conduction electrons is well established in a wide variety of dilute magnetic alloys, principally from the Kondo effect in which the resistivity at low temperatures shows a minimum, and a variation below the minimum of the form $-\ln T$, the sign being directly controlled by the sign of the exchange constant $J$. By contrast, transport data have not yet provided any such clear observation of *ferromagnetic* coupling in such systems, which is surprising since there is no *a priori* reason why $J$ should not be positive. In the few well-defined local-moment systems in which there are indications of positive exchange, the evidence is clouded by impurity-impurity interactions, superconductivity, or Matthiessen's-rule breakdown.[1]

In this paper we wish to report the first unambiguous observation of a positive $\ln T$ variation of the resistivity which scales with the concentration of magnetic impurity in a series of alloys of Lu containing up to 1.4-at.% Gd.

The alloys were made by Rare Earth Products (UK) by arc melting sublimed grades (99.99% purity) of both metals. Resistivity samples of approximate dimensions $3 \times 0.05 \times 0.05$ cm were cut by diamond saw from the as-cast buttons, etched and then annealed for 6 h at 650 °C under a pressure of $10^{-6}$ Torr. Resistivity ratios $[\rho(T) - \rho(4.2)]/\rho(4.2)$ were measured to better than 1 part in $10^5$ using an ac technique.[2] Absolute resistivities were then determined after measuring $\rho(4.2)$ with a conventional four terminal dc method, with a shape uncertainty of $\pm 1\%$. Temperature was found from $^4$He vapor pressure.

The incremental resistivity $\Delta\rho(T) = \rho_{\text{alloy}}(T) - \rho_{\text{Lu}}(T)$ between 1.07 and 4.2 °K is illustrated in Fig. 1 for all the alloys. For samples with less than 1-at.% Gd, the variation of $\Delta\rho(T)$ is strictly logarithmic, i.e.,

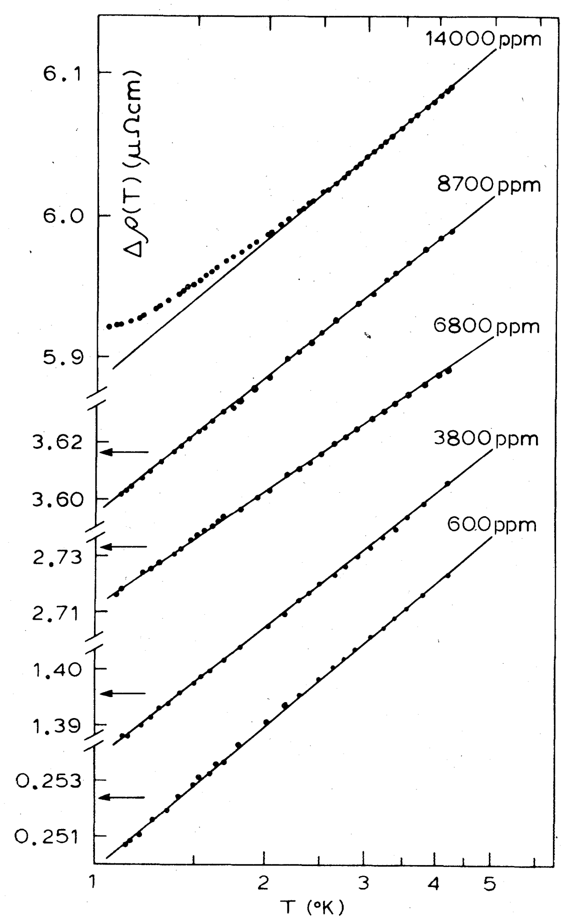


FIG. 1. Incremental resistivity $\Delta\rho(T) = \rho_{\text{alloy}}(T) - \rho_{\text{Lu}}(T)$ as a function of $\log_{10} T$ for all the alloys studied.

$$\Delta\rho(T) = A + D \ln T \quad , \tag{1}$$

where $D$, and hence the appropriate exchange constant $J$, is positive.

To ensure that the above behavior is characteristic of single Gd ions we have measured the low-field (<5 kOe) magnetic susceptibility of these same samples between 1.3 and 80 °K using a vibrating-sample magnetometer.[3] The excess Gd susceptibility of all samples with less than 1-at.% Gd obeys a Curie-Weiss law with a Curie temperature of $0.0 \pm 0.5$ °K. This simple behavior is shown in Fig. 2 and has been used to determine the Gd concentration, using $g = 2$ and $S = \frac{7}{2}$.

In Fig. 3 we combine the magnetic and transport data, showing that the resistivity behavior[1] is indeed a single impurity effect, since $A$ and $D$ vary linearly with concentration.[4]

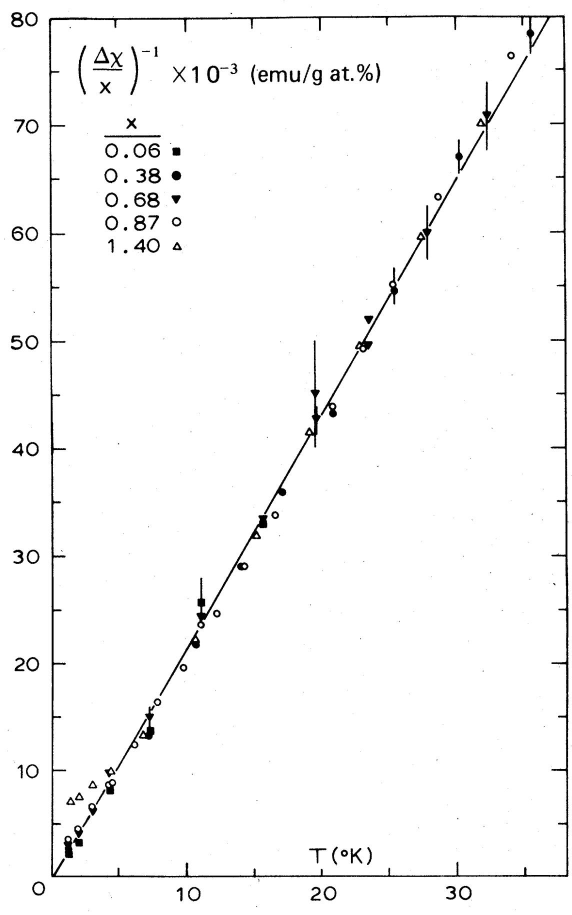


FIG. 2. Inverse incremental susceptibility per at.% Gd plotted against the absolute temperature for the same alloys shown in Fig. 1.

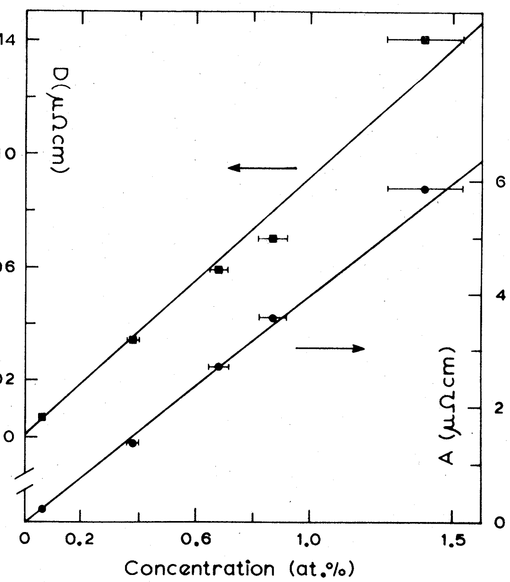


FIG. 3. Resistivity coefficients $A$ and $D$, determined from Fig. 1 plotted as a function of the Gd concentration $X$.

Quantitative analysis of $\Delta\rho(T)$ starts with the usual expression[5] derived from taking the Coulomb and exchange scattering to third order

$$\Delta\rho(T) = \alpha x \{ V^2 + J^2 S(S+1) \times [1 + (3JZ/E_F) \ln T] \} \quad , \tag{2}$$

where $\alpha = 3\pi m\,\Omega / 2\hbar e^2 E_F$. $\Omega$ is the atomic volume, $Z$ is the number of valence electrons per atom, $V$ is the direct Coulomb interaction, and the remaining parameters assume their usual meaning. Using $Z = 2$,[6] a density of states at the Fermi level of 4.6 states/eV atom[7] and the free-electron mass, the measured values of $A$ and $D$ yield $J = 0.057$ eV, $V = 0.56$ eV. The value of $J$ is in excellent agreement with the values deduced by Baberschke and Nagel[8] from EPR measurements and, furthermore, the comparatively small value of $V$ is reassuring since one would expect Gd to act as a nearly isoelectronic impurity in Lu.

A positive value for $J$ is exactly what we would expect for Gd. Theoretically one finds two contributions to $J$: a direct atomic ferromagnetic term $J_a$, and an antiferromagnetic admixture term controlled by some matrix element $V_m$. Thus we write[9]

$$J = J_a - |V_m|^2 / \Delta E \quad , \tag{3}$$

where $\Delta E$ is, in essence, the energy required to add or remove an electron from the magnetic ion. Now the $4f^7$ configuration of Gd is known to be extremely stable, so that $\Delta E$ is large. Thus $J_a$ dominates and $J$ is positive. By contrast, the stability of $3d$ magnetic ions is much weaker, so that the admixture term may dominate, as it apparently usually does.

Finally, it should be noted that the sign of $J$ and the smallness of $V$ have important consequences for magnetoresistance. With positive $J$, a field changes the second and third-order terms [the terms in $J^2$ and $J^3$, respectively, in Eq. (2)] in opposite directions. Thus the magnetoresistance should be much weaker than when $J$ is negative. We have in fact measured the magnetoresistance and its magnitude is indeed understandable only if we take $J$ positive. Full details of this will appear later.

In conclusion transport and magnetic measurements in *Lu*Gd have given the first clear example of a positive Kondo effect of single impurity origin. In addition, the close agreement between these results and EPR measurements establish the first unambiguous instance of local-moment conduction-electron scattering with a positive exchange coupling.

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[1]S. Das and A. Gerritsen, J. Phys. Chem Solids 27, 1167 (1966); T. Sugarawa, J. Phys. Soc. Jpn. 20, 2252 (1965).

[2]W. B. Muir and J. Ström-Olsen, J. Phys. E 9, 163 (1976).

[3]R. W. Cochrane, M. Plischke, and J. O. Ström-Olsen, Phys. Rev. B 9, 3013 (1974).

[4]The samples containing 1.40-at.% Gd shows signs of interaction effects both in resistivity and susceptibility, as may be seen in Figs. 1 and 2. The resistivity $\Delta\rho(T)$ is well fitted by the expression $\Delta\rho = A + D\ \ln(T^2+\Delta^2)^{1/2}$ with $\Delta$, a measure of the interaction strength, being 0.8 °K. The latter is comparable with the Curie temperature $\theta = 1 \pm 1$ °K, deduced from susceptibility.

[5]J. Kondo, Prog. Theor. Phys 32, 37 (1964); K. Yoshida, Phys. Rev. 107, 396 (1957).

[6]We assume that the two 6*s* electrons dominate the conductivity.

[7]K. A. Gschneidner, Solid State Phys. 16, 275 (1964).

[8]K. Baberschke and J. Nagel, Phys. Rev. B 13, 2793 (1976). EPR measurements give two estimates for $J$, which may be different since $J$ is wave-vector ($q$) dependent. The $g$ shift gives $J$ at $q = 0$, while the linewidth gives $J(q)$ averaged over the Fermi surface. Strictly speaking it is this last which should be comparable with our $J$. Baberschke and Nagel in fact give almost identical values from both: 0.055 eV from $g$ shift and 0.047 eV from linewidth.

[9]J. R. Schrieffer and P. A. Wolff, Phys. Rev. 194, 491 (1966); L. L. Hirst, AIP Conf. Proc. 24, 11 (1975). Equation (3) is taken from Hirst's paper since we believe that the configuration-based approach is applicable to Gd.