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ALLOYS WITH DILUTE Gd IMPURITIES

S. J. Poon and J. Durand

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CALIFORNIA INSTITUTE OF TECHNOLOGY
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California Institute of Technology
W. M. Keck Laboratory of Engineering Materials

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by

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Professor Pol Duwez, principal investigator.

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ABSTRACT

From magnetization measurements on some amorphous dilute $\text{La}_{79-x}\text{Gd}_x\text{Au}_{20}$ alloys with $x \leq 1$ we have shown that the magnetic behavior follows the scaling laws of a spin-glass system, characteristic of the $1/r^3$ dependence of the pairwise interaction. We have also determined the strength of the Ruderman-Kittel-Kasuya-Yosida interaction $V(r) = (V_0 \cos 2k_F r)/r^3$, to be $V_0 = 0.20 \times 10^{-37} \text{ erg cm}^3$. The corresponding value of the s-f exchange integral is $|J_{sf}| = 0.14 \text{ eV}$, which is compared with values determined from other experiments.

Recently, the spin-glass behavior in dilute magnetic alloys containing 3d-magnetic solutes has been the subject of experimental and theoretical work.¹ The indirect Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction² between the pairs of magnetic impurities in host metals via the host conduction electrons is believed to be responsible for the observed magnetic behavior in the spin glasses. From the invariance of the product xr^3 (where x is the concentration of magnetic impurities) scaling prediction³ for the various thermodynamical parameters was made, which constitutes an additional manifestation of the $1/r^3$ dependence of the s-d exchange interaction. The values of the s-d exchange integral J_{sd} were determined from measurements of the host NMR line broadening, EPR line width, and recently by Smith et al⁴ from magnetization measurements. Relatively fewer studies have been made on the spin-glass behavior in both crystalline and amorphous alloys containing rare-earth 4f-impurities.^{5,6} Especially in amorphous alloys, the spin-glass behavior can be extended to higher concentrations of magnetic solutes, as shown in the La-Gd-Au system. In general, the localized nature of the 4f wave functions is expected to give a smaller value of the s-f exchange integral J_{sf} and thus a weaker RKKY interaction. Various NMR, EPR, and resistivity measurements⁷⁻¹⁰ gave $|J_{sf}|$ varying from 0.03 to 0.31 eV.

We report in this letter measurements of the magnetic properties of a series of amorphous dilute $\text{La}_{79-x}\text{Gd}_x\text{Au}_{20}$ alloys with $x \lesssim 1.0$. The magnetic behavior follows the scaling prediction up to ~ 1 at. % Gd. We have also determined the value of the s-f exchange integral to be $|J_{sf}| = 0.14 \text{ eV}$ using the method first employed by Smith et al.⁴ The result is compared with values from other experiments. We believe

that this technique can be generalized to determine J_{sf} in other alloys doped with rare-earth impurities. Knowing the variation of J_{sf} in the rare-earth series, it might help to understand the recently observed abnormal depression of the superconducting transition temperature of LaSn_3 doped with rare-earth impurities.¹¹

The sample foils were prepared in the usual way as discussed elsewhere.⁶ The glassy nature of the foils was checked by x-ray scanning. Magnetization measurements were made between 1.8°K and 290°K in fields up to 70 kOe using the Faraday method.⁶ The Gd concentrations in the samples are nominal. Measurements were made on four $\text{La}_{79-x}\text{Gd}_x\text{Au}_{20}$ alloys ($x = 0, 0.24, 0.5, \text{ and } 1.0$). The magnetization curves for Gd impurities were obtained by subtracting from the data the magnetization contributions from the $\text{La}_{80}\text{Au}_{20}$ matrix at corresponding temperatures. The latter was found to contain ~ 200 ppm rare-earth impurities. The initial susceptibility χ_0 is determined from the slope dM/dH as $H \rightarrow 0$.

In Figure 1, the initial susceptibility χ_0 versus T/x for the three alloys is plotted for $1.8^\circ\text{K} \leq T < 100^\circ\text{K}$. At low temperatures ($T < 20^\circ\text{K}$), χ_0 can be fitted for each alloy to a Curie-Weiss law $\chi_0 = C_1/(T + \theta)$. For $T > 30^\circ\text{K}$, $\chi_0 = C_2/T$. The resulting values of C_1 , C_2 , and θ are listed in Table I. The values of the spin S per Gd atom determined from C_1 , C_2 and the saturation magnetization M_∞ are also included. The implication of these values will be given in a subsequent publication.⁶ To within experimental error, a universal curve characteristic of a scaling law is obtained in Figure 1. In Figure 2, M/x versus H/x at three different values of T/x are plotted. The values of T/x for each curve varies within $\sim 5\%$. Again, three

TABLE I. Magnetic properties of $(\text{La}_{100-x}\text{Gd}_x)\text{Au}_{20}$ alloys studied.

x (at. % Gd)	θ (°K)	C_1 ($10^{-6}\text{emu}^\circ\text{K/g}$)	C_2 ($10^{-6}\text{emu}^\circ\text{K/g}$)	M_∞ (10^{-2}emu/g)	$S^{(a)}$	$S^{(b)}$	$S^{(c)}$
0.24	0.6	168	138	70	4.00	3.70	3.79
0.5	0.9	329	305	145	3.89	3.82	3.77
1.0	1.5	794	617	290	4.41	3.85	3.81

(a) Spin per Gd atom determined from low temperature Curie constant C_1 .

(b) Spin per Gd atom determined from high temperature Curie constant C_2 .

(c) Spin per Gd atom determined from extrapolated magnetization M_∞ .

Fig. 1. Initial susceptibility χ_0 for three $(\text{La}_{100-x}\text{Gd}_x)_{80}\text{Au}_{20}$ alloys as a function of reduced temperature T/x .

Fig. 2. Reduced magnetization M/x as a function of reduced magnetic field H/x at three reduced temperatures T/x .

universal curves are obtained to within experimental error. The scaling behavior so observed originates from the invariance of xr^3 , which manifests the $1/r^3$ dependence of the pairwise interaction between Gd impurities. Preliminary magnetization results on samples with higher Gd concentrations (e.g. $x \sim 4$) indicate that the magnetic properties no longer observe the scaling laws. The interactions become more complicated which marks the emergence of a micromagnetic regime.⁶

To obtain the strength V_o of the RKKY interaction, we follow Smith's approach⁴ which is based on the asymptotic expression derived by Larkin et al¹² for the approach to saturation of the magnetization.

$$M = xg\mu_B S [1 - 4(2S + 1) xV_o / 3g\mu_B H] \quad (1)$$

valid for $g\mu_B H \gg k_B T$. In a maximum field of 70 kOe and $g = 2$, $g\mu_B H / k_B \approx 9.4^\circ K$. As the temperature is comparable to this critical value, a thermal factor should be included in (1). Since M/x is a universal function of T/x and H/x for the concentration range investigated, to lowest order, the temperature and field should appear in the combination T/H for the approach to saturation of the magnetization. Thus for $g\mu_B H \approx k_B T$, (1) can be modified to read

$$M/x = g\mu_B S [1 - H_o(x, T)/H]$$

$$\text{with} \quad H_o(x, T) = Ak_B T + 4(2S + 1) x V_o / 3g\mu_B \quad (2)$$

where A is a constant.

From the plots of M versus H^{-1} at various temperatures one obtains $M_{\infty} = xg\mu_B S$ in the limit $H^{-1} \rightarrow 0$ and also the slope $H_0(x, T)$. The approach to saturation for two samples is demonstrated in Figure 3. In Figure 4, we plot $H_0(x, T)/x$ as a function of T/x for the three alloys studied. The linear dependence observed illustrates the validity of expression (2) which yields $4(2S + 1)V_0/3g\mu_B$ as $T \rightarrow 0$. From the intercept we obtain $V_0 = 0.20 \times 10^{-37}$ erg cm³, using $g = 2$ and an averaged $S^{(c)} = 3.79$ from Table I. The small variation in S is ignored and thus no estimate is made for the error in V_0 . We also find from the slope that $Ak_B = 1.4 \times 10^3$ Oe/°K. Using $J^2 = 16\pi n E_F V_0 / 3z^2$ where the parameters n , E_F , and z refer to those of the host metal. For the La₈₀Au₂₀ matrix, we use¹³ $n = 0.87 \times 10^{23}$ cm⁻³, $E_F = 7$ eV, $z = 2.6$, and $V_0 = 0.20 \times 10^{-37}$ erg cm³ to obtain $|J_{sf}| = 0.14$ eV. By comparison, NMR⁷ and resistivity⁸ measurements gave $|J_{sf}| = 0.03$ and 0.31 eV respectively for Gd in the intermetallic GdAl₂. EPR measurements on La_{1-x}Gd_xAl₂⁹ and ThGd¹⁰ systems yielded $|J_{sf}| = 0.1$ and 0.042 eV respectively. The averaged saturation moment $S = 3.79$ for Gd in La₈₀Au₂₀ is comparable to that of crystalline Gd. The s-f exchange integral is found to be an order of magnitude smaller than the s-d exchange integral. Further experiments are needed to study the variation of J_{sf} in the rare-earth series based on this convenient technique. However, except in the case of Gd which is an S-state ion, the crystal field effects on the approach to saturation of magnetization must be taken into account for other rare-earth elements.

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Fig. 3. Magnetization M as a function of reciprocal magnetic field $1/H$ for two alloys.

Fig. 4. $H_0(x, T)/x$ as a function of reduced temperature T/x for three alloys illustrating the validity of equation (2).

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† On leave from Laboratoire de Structure Electronique des Solides,
E.R.A. 100 4, rue Blaise Pascal, 67000 Strasbourg, France.

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