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FROM SUPERCONDUCTIVITY TO FERROMAGNETISM IN AMORPHOUS Gd-La-Au ALLOYS

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by

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NOTICE

ABSTRACT

Splat cooled alloys of composition $(La_{100-x}Gd_x)_{80}Au_{20}$ have been obtained by complete substitution of gadolinium for lanthanum in amorphous $La_{80}Au_{20}$ matrix. Results of high field magnetization (up to 70 kOe), ac and dc low field susceptibility, and resistivity measurements over temperature range of 1.7 to 300°K for these alloys are reported. The $La_{80}Au_{20}$ alloys are superconducting at 3.5°K. For $x \lesssim 1$, a suppression of T_c described by the relation $dT_c/dx \approx -4.0$°K per atomic per cent gadolinium is observed. For alloys within the concentration range $1 \lesssim x \lesssim 70$, maxima in low field susceptibility measurements are observed. The 'ordering' temperatures T_M are proportional to x for $1 \leq x \leq 16$, similar to those observed in crystalline spin-glass alloys. For $16 \lesssim x \lesssim 70$, T_M is increasing at a faster rate than in the low concentration region, and this intermediate type of ordering corresponds to a mictomagnetic regime. As $x \gtrsim 70$, a ferromagnetic regime emerges. The maximum Curie temperature is observed for $Gd_{80}Au_{20}$ at ~ 150°K. The moment per gadolinium atom is found to be constant and close to that of the crystalline value throughout the concentration range investigated. Results of resistivity measurements are correlated with the magnetic properties of different regimes in the magnetic phase diagram.

EXPERIMENTAL PROCEDURES

The sample foils were prepared in the usual way as discussed elsewhere.[1] X-ray scanning of the foils indicated patterns with broad maxima centered at $\sim 31.5^{\circ}$ with full widths at half maxima of $\sim 4.5^{\circ}$, which were typical of a glassy metal. Magnetic ordering temperatures were observed using a standard ac inductance bridge technique. Magnetization measurements were made between $1.7^{\circ}K$ and $290^{\circ}K$ in fields up to 70 kOe using the Faraday method.[1] Electrical resistivity as a function of temperature was measured using a standard four-probe technique.

RESULTS AND DISCUSSION FOR $(La_{100-x}Gd_x)_{80}Au_{20}$ ALLOYS

A. $La_{80}Au_{20}$ Superconductors.

The critical behavior and transport properties of amorphous superconducting $La_{80}Au_{20}$ alloys has been discussed previously.[2,3] The alloys are ideal type II superconductors characterized by $T_c \simeq 3.5^{\circ}K$, $H_{c2}(0) \simeq 60$ kOe, $J_c(0) \simeq 10^4$ A/cm^2, and a Ginzburg-Landau parameter κ of ~ 70. Spin-orbit scattering effects are found to be stronger in the amorphous samples than in disordered crystalline samples. Fluctuation conductivity in three dimensional amorphous superconductors has been investgated by Johnson and Tsuei.[4] We obtained magnetization results between $1.7^{\circ}K$ and $290^{\circ}K$. The susceptibility is found to be temperature independent with a value of $\sim 0.5 \times 10^{-6}$ emu/g.

Table 1. Parameters derived from magnetization measurements for amorphous $Gd_{80}Au_{20}$ alloys.

T_c (°K)	β	γ	δ	$\mu_{Gd}(\mu_B)$	$\mu_{eff}^{(a)}(\mu_B)$	θ_p (°K)	J_n (°K) $(T > T_c)$	J_n (°K) $(T < T_c)$
149.45	0.439	1.294	3.948	7.00	9.37	165	2.28	1.34

(a) $\mu_{eff} = g\,[J(J+1)]^{\frac{1}{2}}$, some authors use p_{eff}.

B. $Gd_{80}Au_{20}$ Ferromagnets.

The magnetic properties of this amorphous ferromagnet[1] are summarized in Table 1. The normalized M(H, T) data are fitted to an equation of state derived for second order phase transition in fluid systems. The equation takes the form $h/m = f_{\pm}(m)$ where the $\pm$ signs stand for temperatures above and below the Curie temperature (T_c) respectively. This is illustrated graphically in figure 1. Together with the equality relation observed by the critical exponents, the results indicate clearly a second order phase transition in the amorphous state. The small deviations of the exponents from the Heisenberg values are probably due to the crystal-field anisotropy. The exchange integrals J_n for temperatures above and below T_c are found to be lower than those in crystalline Gd. The values equal 2.28 and 1.34^{o}K as determined from the Rushbrooke-Wood formula and spin-wave theory respectively. The low temperature saturation magnetization follows the $T^{3/2}$ law from 0.13 to 0.80 T_c. Amorphousness is found to be more detrimental for T_c than for μ_{Gd}. Fluctuations in J_n due to structural randomness are under investigation.[1] The differences in the exchange integrals J_n, and that between the effective moment μ_{eff} and saturation moment μ_{Gd} at different temperature regimes can be attributed to the nearest-neighbors antiferromagnetic couplings in the presence of Au below T_c. This is supported by case studies in stochiometric Gd-Au compound.[5]

C. $x < 1$, Coexistence of Magnetic Short-Range Ordering and Superconductivity Regime.

In figure 2, the suppression of superconducting transition tem-

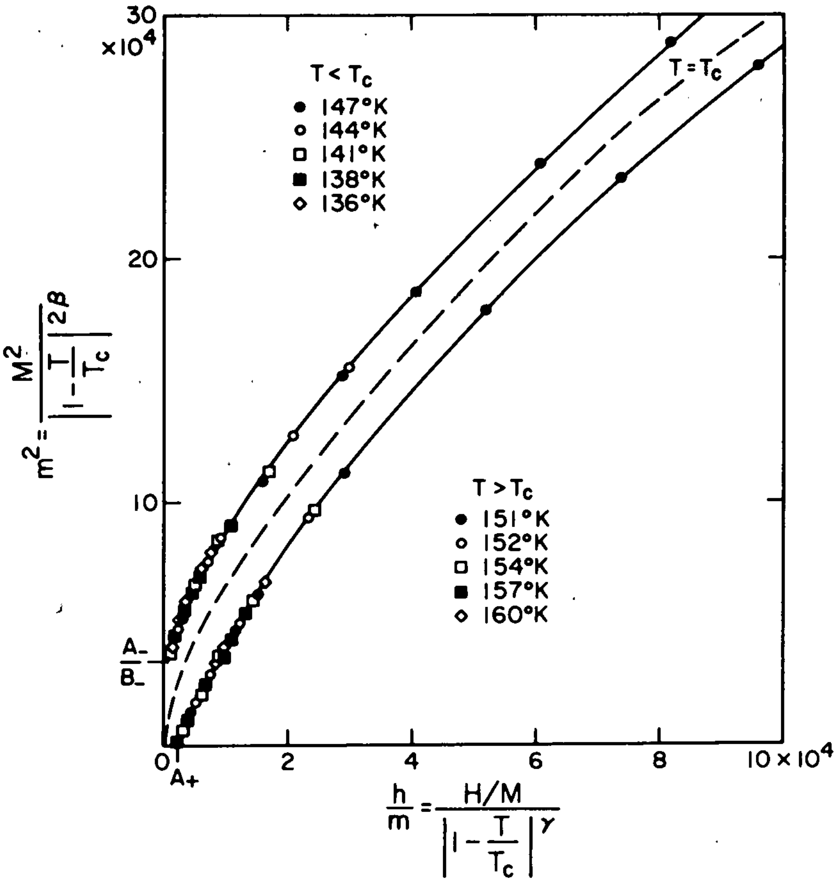


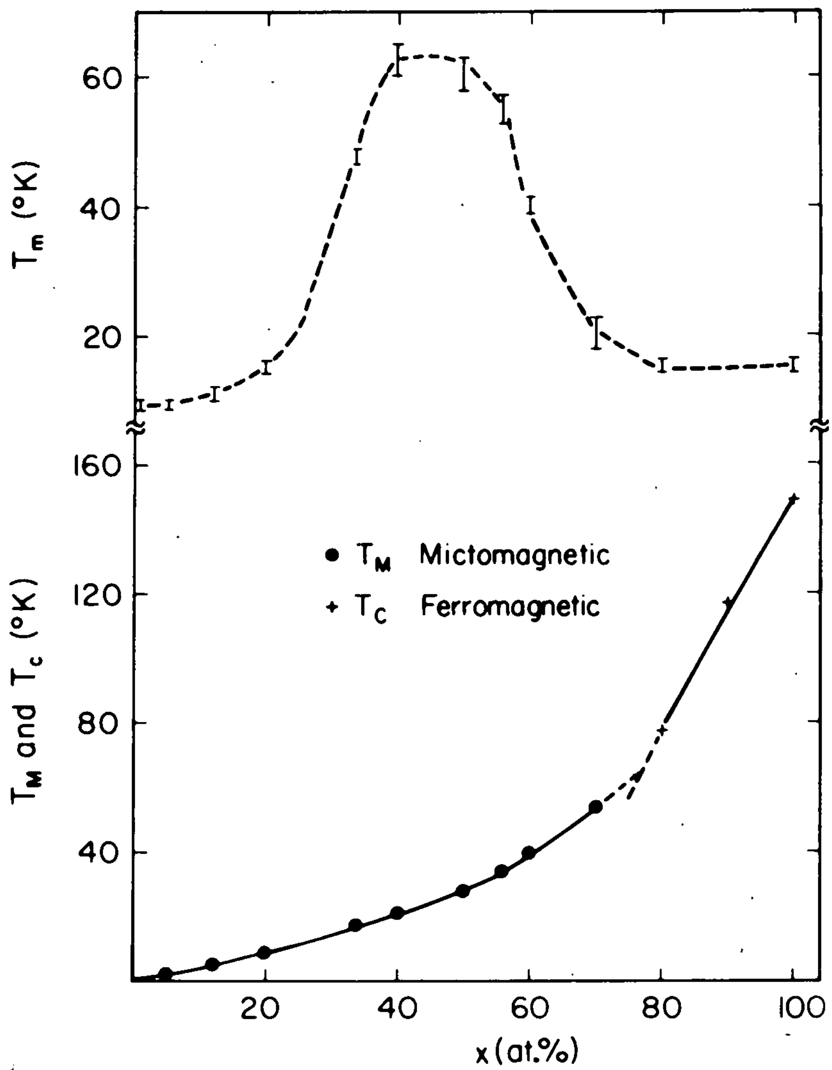
Fig. 1. The normalized magnetization m^2 versus normalized inverse susceptibility χ_o^{-1} for temperatures around T_c. The dashed line indicates asymptotic behavior of the two curves for large m above and below T_c.

perature in the presence of magnetic impurity Gd follows closely the Abrikosov-Gor'kov theory[6] below the critical concentration x. The gradient dT_c/dx gives a value of - $4^{\circ}K$ per atomic per cent Gd. Superconductivity in La-Gd dilute solid solutions has been investigated thoroughly by Matthias et al.[7,8] by employing specific heat results, Finnemore et al.[9] demonstrated the coexistence of antiferromagnetic coupling and superconductivity in the dilute Gd limit. It might be more interesting to investigate the coexistence phenomena directly by performing magnetization measurements. Such results have been obtained recently.[1] The magnetizations, after correcting for the $La_{80}Au_{20}$ matrix contributions to the susceptibility, are found to satisfy a Brillouin equation of the form $M(H,T) = M(\infty, 0)B_s(H/T + \theta)$. The parameters S and θ (> 0) give the spin of the moments and the antiferromagnetic characteristic temperature respectively. For the samples with 0.24 and 0.5 per cent Gd investigated, it is found that $\theta \lesssim 1^{\circ}K < T_c$ as shown in figure 2. The Gd atoms carry a moment of about 7 Bohr magnetons per atom which are close to the $^8S_{7/2}$ ionic value.

D. Mictomagnetic Regime (1 < x < 70).

Alloys in this regime are characterized by susceptibility maxima in low field measurements and thermomagnetic effects[10, 11](isothermal remanent magnetization and thermal remanent magnetization at least for high concentrations). The dependence of the 'ordering' temperature T_M on Gd concentration can be divided into two regimes

$$T_M(\text{in}^{\circ}K) \simeq \begin{cases} 0.38\,x & 1 < x < 16 \\ 6 + f(x) & 16 < x < 70 \end{cases}$$



(Fig. 2 is on page 9)

Fig. 3. Magnetic phase diagram of $(La_{100-x}Gd_x)_{80}Au_{20}$ indicating the transition temperatures T_M and T_c in the mictomagnetic and ferromagnetic regimes respectively as function of x. The resistivity minima T_m are also included for comparison.

where $0 < f(x) < 48$ is a monotonic increasing function of x. The distribution of the regimes are evident from the magnetic phase diagram of figure 3. The values of the initial susceptibility $\chi_o(T)$ for x = 20 and 60 in "zero" field and in fields up to 500 Oe on zero-field cooled samples are shown in figure 4. It is clear that the peaks in $\chi_o(T)$ are reduced and rounded off in small applied fields as observed in the crystalline case. They disappear in samples cooled in fields greater than $\sim$ 1 kOe. The paramagnetic Curie temperature θ_p ($\simeq 3\ T_M$) is found to increase with x indicating a trend towards stronger ferromagnetic couplings above the 'ordering' temperature. The large value of $\theta_p - T_M$ also measures the temperature range of inhomogeneous ferromagnetic interactions. Using the classical molecular field approach, the effective number of Bohr magnetons per Gd atom p_{eff} is found to remain constant at the value of 8 which corresponds to the ionic value of 7.94.

The linear dependence of T_M on concentration in the first regime resembles those observed in typical spin glass systems.[12] However, unlike the well studied spin glass systems, the present magnetization study does not indicate any scaling law relations and the concentration of magnetic impurities is far beyond those observed in the spin glass alloys in the sense of reference 12. Therefore, it might be more appropriate to call this region a mictomagnetic[13] regime without loss of generality.

The classical Arrott plots (M^2 versus H/M) in both regimes exhibit strong departures from linearity at small and high fields for all temperatures below θ_p, so that any spontaneous magnetization and Curie temperature cannot be defined by this method. However, the $M^2(H/M)$

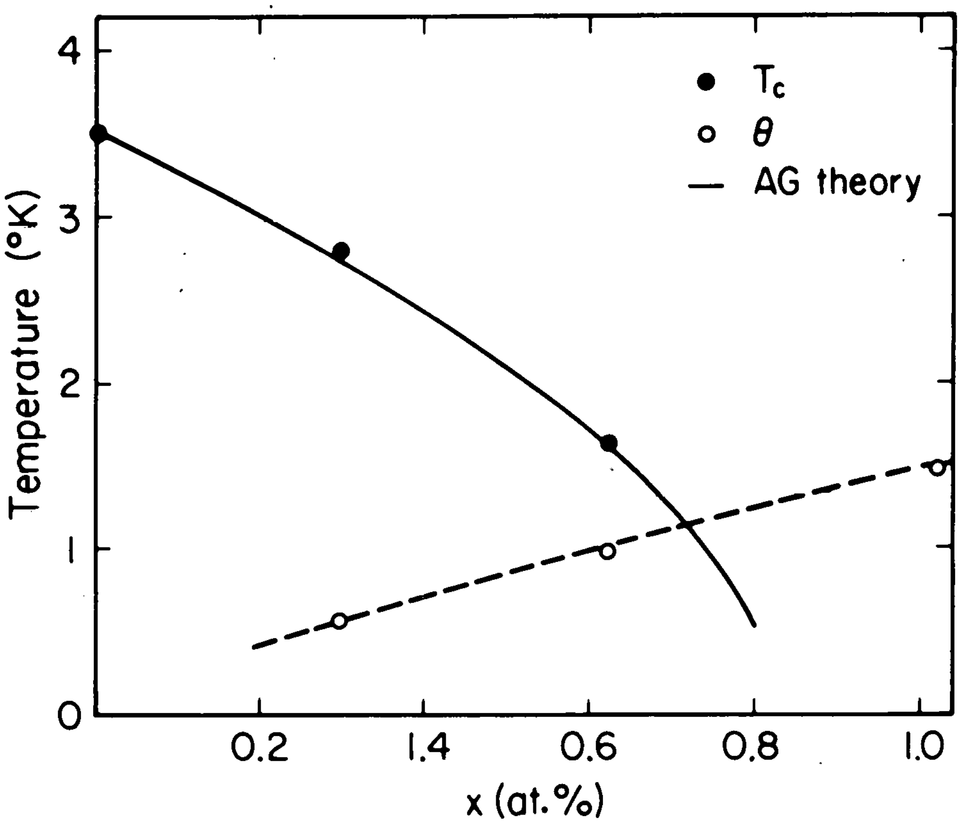


Fig. 2. The dependence of T_c and θ on Gd concentration in $La_{80}Au_{20}$ matrix.

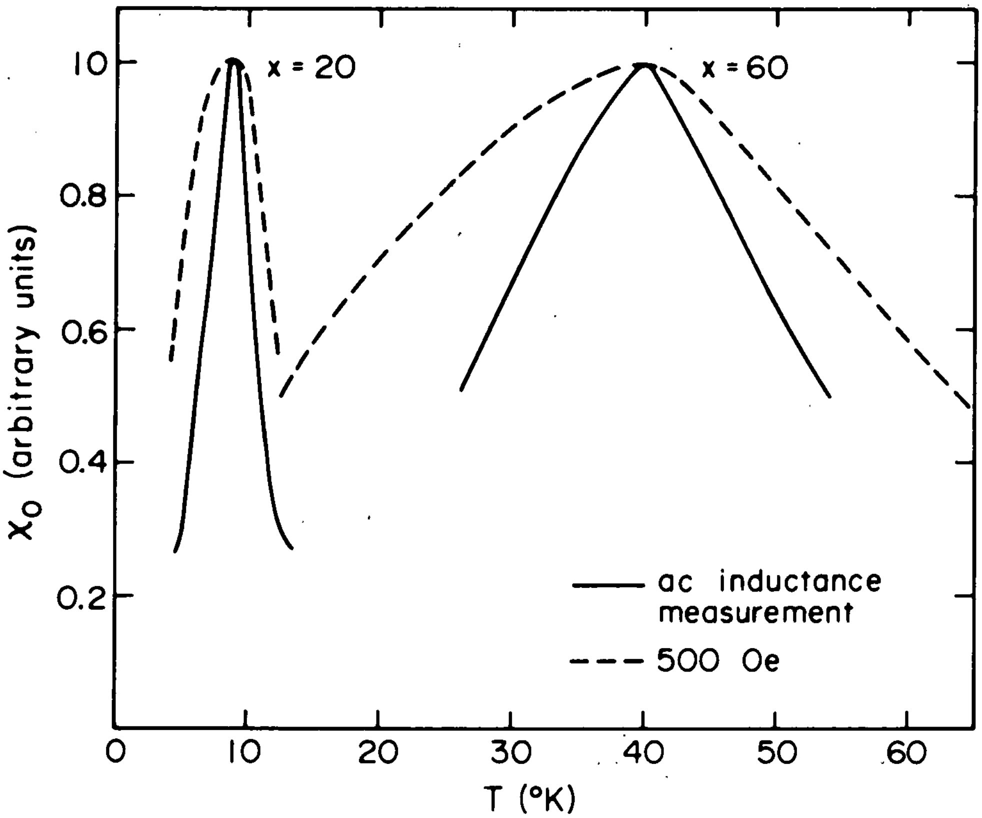


Fig. 4. Susceptibility (in arbitrary unit) as a function of temperature measured in "zero" field and in 500 Oe for x = 20 and 60 samples.

isotherms are observed to approach closer to the M^2-axis for higher Gd concentrations indicating a gradual onset of spontaneous magnetization for $x \gtrsim 70$. The absence of spontaneous magnetization from the Arrott plots for all $T < \theta_p$ also points towards the possibility of weak and inhomogeneous ferromagnetic interactions. For temperatures between T_M and θ_p, the superparamagnetic clusters break up gradually at increasing temperature to yield single magnetic atom moments above θ_p. The persistence of mictomagnetic regime at high Gd concentrations (up to ~ 56 atomic per cent) is probably favored by two conditions. First, the structural randomness in the amorphous state introduces inhomogeneities in ferromagnetic couplings which depend strongly on local structural environment. Second, it has been mentioned beforehand[1] that Gd atoms tend to couple antiferromagnetically in the presence of Au and La. Even in crystalline Gd, there is already a trend towards this type of couplings, as the difference in the exchange interactions J_n at different temperatures can be accounted for by the RKKY interaction.[14]

E. Ferromagnetic Regime ($70 < x < 100$).

Alloys in this regime are characterized by a well defined Curie temperature. The magnetic phase transition determined from ac inductance bridge measurement gives a transition width of ~ 10°K. The Curie temperature is defined by the inflection point on the signal intensity versus temperature curve.[1] The spontaneous magnetization can be determined from the Arrott plots. Nonlinearity in M^2 versus H/M is observed even for $x > 70$ indicating inhomogeneities in ferromagnetic couplings. This is also supported by the fact that the inflection point on the M(H) versus T plots disappears in fields greater than ~ 2 kOe.[1] The temperature domain over which ferromagnetic inhomogeneities dominate narrows as x increases until $Gd_{80}Au_{20}$, $(\theta_p - T_c)/T_c \simeq 0.1$. The variation of the mean magnetic moment per atom when La substitutes for Gd obeys fairly well a dilution law, which means the possible polarization of La (or Au) atoms has to be rather small.[15] The effective moment p_{eff} determined at $T > \theta_p$ gives ~ 8 Bohr magnetons for Gd atom if the effects due to La and Au are ignored. The saturation moment μ_{Gd} gives approximately the same value. However, the detail trends in μ_{eff} and μ_{Gd} can be explained in terms of antiferromagnetic couplings, conduction electron polarizations, and crystal-field effects.[1] The suppression of T_c defined by $(1/T_c)(dT_c/dx)$ when La is substituted for Gd in crystalline Gd[15] and amorphous $Gd_{80}Au_{20}$ alloys are found to be 1.82×10^{-2} and 2.32×10^{-2} per La atom respectively.

F. Resistivity Results.

Resistivity minima have been observed over the whole concentration range for $x \gtrsim 0.6$. The variation of the resistivity minima (T_m) follows a bell shaped curve as shown in figure 3. The invariance in T_m (10-15°K) at both the low concentration range and ferromagnetic regime suggests a structural rather than a magnetic origin for this phenomena[16] based on the following reasons. For x = 0.6 dilute alloys, it is unlikely that the resistivity minimum results from the Kondo effect.[17] Recent magnetoresistivity measurements[18] indicate that the shape of the resistivity curve is unaltered except for the sign of magnetoresistance in fields up to 40 kOe. For the ferromagnetic regime, conjectures had been made to explain the occurrence of resistivity minima in terms of conduction electrons being scattered off low energy magnons.[19,20] It is not clear in which way the magnon dispersion spectrum is changed in a field of 40 kOe ($\sim 5^{\circ}K$). Present experiments give the same results for the ferromagnetic samples as in the low concentration limit in an applied field. However, the strong variation of T_m in the mictomagnetic regime might point towards a magnetic origin.[21] Thus, the resistivity minimum phenomena might be caused by either structural or magnetic mechanism, and the interplay of these two mechanisms on the shape of the resistivity curves is under investigation.

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