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CALIFORNIA INSTITUTE OF TECHNOLOGY

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ABSTRACT

Magnetization and resistivity measurements are performed on amorphous $La_{80}Au_{20}$ alloys doped with Gd impurities. The anomalous behavior observed in the dependence of the superconducting transition temperature on Gd concentration is related to the onset of spin-spin correlations among the Gd impurities. We have identified the coexistence of superconductivity and magnetic ordering as a spin glass.

It was reported[1-5] that for several alloys systems the superconducting transition temperature T_c instead of decreasing monotonically with increasing impurity concentration, exhibits an anomalous peak close to the Abrikosov-Gor'kov critical concentration x_{AG}. This behavior was attributed to a magnetic ordering of the impurity spins in the vicinity of the peak.[6-8] Low field magnetization was used to probe the type of magnetism involved.[2,4,9] However, the data obtained so far only imply the occurrence of antiferromagnetism of some sort: either a Kondo effect or an antiferromagnetic interaction among impurities, which has significantly different influence on the superconductivity around x_{AG}.[10] In amorphous alloys, the interaction between magnetic impurities via conduction electrons is attenuated while the spin-orbit coupling is significantly enhanced.[11] The former effect which tends to enhance the spin-flip scattering[6] can be isolated as the latter suppresses the effect of the local exchange field on Cooper pairs.[12] By using both magnetization and resistivity measurements on amorphous superconductors doped with magnetic impurities, we can clarify the type of magnetic ordering coexisting with a superconducting phase. Comparison with existing theories on the effects of spin-spin correlations on superconductivity is thus possible.

Sample foils of nominal composition $La_{80-x}Gd_xAu_{20}$ (x = 0, 0.24, 0.42, 0.5, 0.55, 0.59, 0.63 and 0.67) were prepared in the usual way as discussed elsewhere.[13] The amorphousness of each foil was checked by x-ray diffraction. Low field susceptibility measurements were made by the ac inductance method down to 1.3°K. Detailed magnetization measurements on four alloys (x = 0, 0.24, 0.5 and 1.0) were made between 1.8°K and 290°K in fields up to 70 kOe using the Faraday method.[13]

Resistivity measurements were performed between 1.3°K and 20°K by the standard four probe technique. The $La_{80}Au_{20}$ matrix was found to contain $\sim$ 0.02 at.% Gd. The superconducting transition temperature T_c (transition width $\sim 0.2°K$) was determined consistently for each sample by adopting the mean-field T_c extrapolated from the onset of superconductivity.

In figure 1, T_c values of at least two samples for each nominal composition (within ± 0.01 at.% uncertainty) is represented by a two dimensional uncertainty bar. Comparison with the Abrikosov-Gor'kov curve is made by fitting the low Gd concentration data to theory. Thus, the T_c of pure amorphous $La_{80}Au_{20}$ alloy is estimated to be 3.6°K, and the critical concentration x_{AG} extrapolated to $\sim$ 0.7 at.% Gd. For $x = 0.59$, T_c is observed to exceed the AG value T_{AG} by $\sim 0.16°K$. Unfortunately, we were not able to measure T_c below 1.3°K for higher Gd concentrations. However, the T_c of $x = 0.63$ ($\sim 1.15°K > T_{AG} = 0.94°K$) can be estimated from the onset of superconductivity transition around 1.35°K which is compared with other samples with measurable T_c. The latter demonstrate similar resistivity behavior around T_c. Resistivity minima are observed at $T \sim 8°K$ for the $x \gtrsim 0.42$ alloys.

At low temperature ($T < 20°K$), the initial susceptibility can be fitted to a Curie-Weiss law $\chi_o = C/(T + \theta)$ with the values of θ included in figure 1. It can be seen that the values of T_c are enhanced around the region where the AG curve intersects the θ line. We fitted the magnetization data to a Brillouin function of the form $M(H, T) = M(\infty, 0)\ B_J\ (\mu H/(T + \theta))$. The value of J determines the average spin value of the polarization 'clouds'.[14] Such plot for the 0.24 sample ($g = 2$, $J = 4.35$) is shown in figure 2. Within experimental

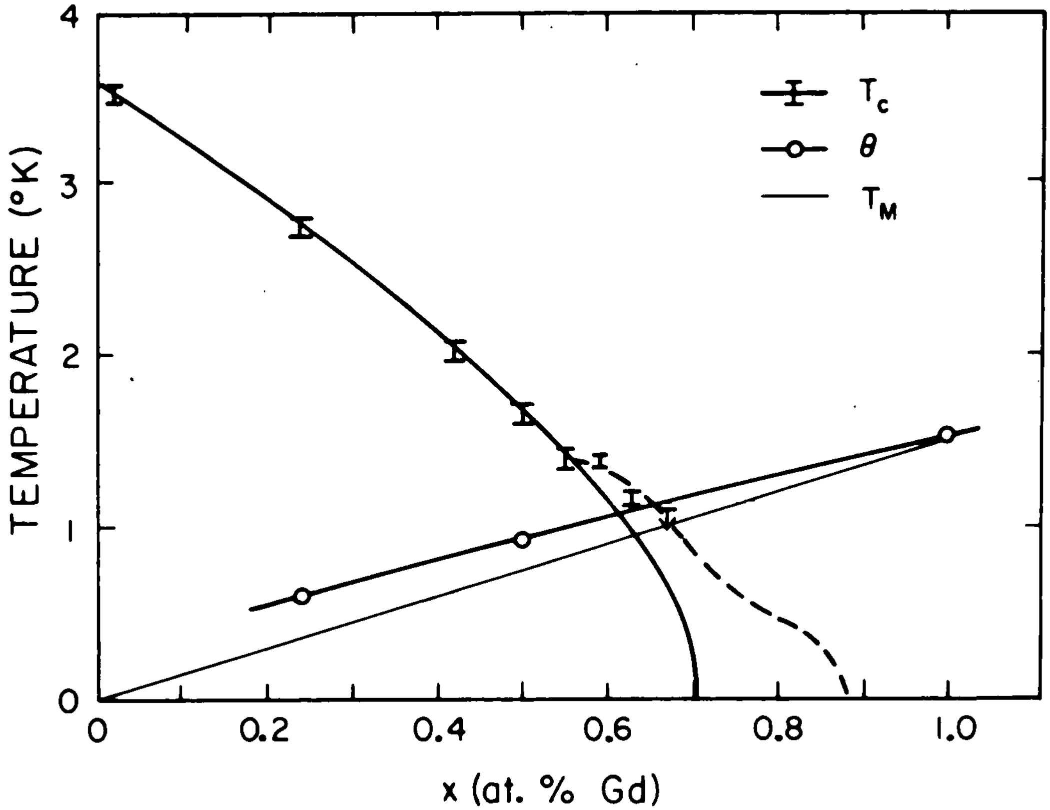

Fig. 1. T_c, T_M, and θ vs Gd composition. The solid line —I— is obtained from the AG theory, dashed line is derived from the Bennemann theory.

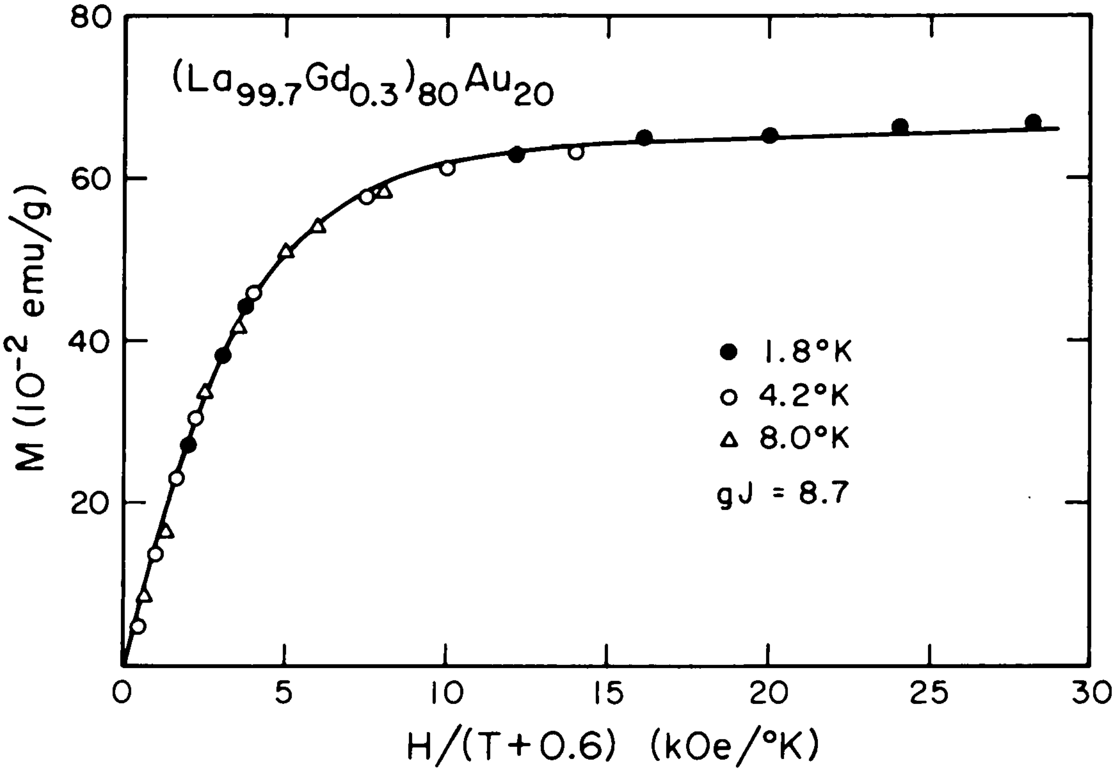

Fig. 2. Variation of the magnetic impurities magnetization vs H/(T + 0.6) for a 0.24% sample. Full line represents a Brillouin function (g = 2, J = 4.35).

error, the θ values so determined agree well with those obtained from low temperature χ_o. Such positive values of θ indicate an antiferromagnetic coupling of some sort among the Gd atoms carrying a moment close to the ionic value in the low concentration limit.

A more accurate insight on the nature of the magnetic ordering may be obtained from two supplementary experimental facts. First, for a 4% sample, we observe a susceptibility maximum at $T_M = 1.9^oK$ while low temperature χ_o also gives $\theta = 1.9^oK$. Similar result is observed for a 1% sample. The linearity of T_M as a function of x at low concentration gives values reasonably close to those obtained for θ, as shown in figure 1. Second, we obtain scaling laws in the magnetization and susceptibility of the form $M/x = f(H/x, T/x)$ and $\chi_o = g(T/x)$.[13] The scaling behavior[15] so observed originates from the $1/r^3$ dependence of the long-range pairwise interaction between Gd impurities. The latter is made possible in an amorphous matrix probably due to the polarization of La atoms with an empty f-band.[16] Thus, the values of θ are characteristic of a spin-glass transition temperature in the restricted sense.[15]

The pair breaking parameter in the AG theory was modified by Fulde and Maki[12] as $\alpha = (\hbar/\tau_s + I^2\tau_{so}/2\hbar)$, where the mean exchange field $I \sim \langle J_z \rangle$, τ_s and τ_{so} are the spin-flip and spin-orbit scattering time respectively. It was shown from upper critical field measurements[11] that τ_{so} is rather small in amorphous $La_{80}Au_{20}$ alloys, and that $I \sim \langle J_z \rangle = 0$ when the spins start to order antiferromagnetically. The latter is valid since the mean-free-path limited coherence length $\xi \approx 100Å$[11] is significantly greater than the spin-spin correlation distance $\ell_s \approx 20Å$ for the 0.6% sample. The pair breaking parameter then reduces to $\alpha = \hbar/\tau_s$. Bennemann[6] pointed out that the spin-flip scattering time τ_s is reduced by a factor $J/(J+1)$ between the randomly oriented spins and the ordered spins in the limit of very high local exchange fields acting between the ions. In the present case, one would expect an enhanced critical concentration of $\sim$ 0.88 at.% Gd. From high field magnetization data on the 0.5% sample, we deduced[13] an average local field of $\sim$ 4kOe (or 0.5°K) which obviously violates the high field assumption at $\sim$ 1°K. Instead, smaller enhanced T_c values are expected around $x \sim 0.6$ at.% Gd. The theoretical T_c enhancement based on the Bennemann theory[6] is plotted in figure 1. Without crystal-field splitting, the initial depression $-(dT_c/dx)_{x=0} \simeq 3.6$°K per at.% Gd yields a value of the exchange interaction parameter $|J| \approx 0.16$ eV. Similar value of $|J_{sf}| \simeq 0.14$ eV due to indirect exchange interaction was obtained from high field measurements.[13] The presence of a resistivity minimum at $T \sim 8$°K for $x \gtrsim 0.42$ implies that J_{sf} is negative in sign. Beyond the first Born approximation, a more rapid depression of superconductivity around $x \gtrsim x_{AG}$ is thus expected. In fact, such effect was observed in crystalline LaGd alloys.[1]

From magnetization measurements, we have demonstrated unambiguously the existence of a spin-glass regime within the superconducting phase in amorphous La-Gd-Au alloys. This allows us to clarify the type of magnetic ordering which is favorable for the enhancement of superconductivity.

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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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