Universal phase diagram for superconducting spin glasses

Thomas R. Lemberger
Physics Department, Ohio State University, Columbus, Ohio 43210
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Published measurements of the superconducting and spin-glass transition temperatures, T_c and T_g, respectively, as functions of the magnetic atom concentration, x, suggest the universal phase diagram for superconducting spin glasses presented in this paper. The universality occurs because, as x increases from zero, both the increase in T_g and the decrease in T_c are proportional to the exchange scattering rate of the conduction electrons.

In this paper, a universal phase diagram is presented for superconducting spin glasses. The phase diagram is based primarily on published measurements of the superconducting and spin-glass transition temperatures, $T_c(x)$ and $T_g(x)$, respectively, as functions of the magnetic atom concentration x for a wide variety of alloy combinations.[1] It is consistent with recent theoretical predictions that the mutual effects of spin-glass and superconducting order are relatively small,[2,3] and it can be understood physically with the rough theoretical arguments presented below. This is the first quantitative prediction of a relationship between T_c and T_g, and it allows one to estimate T_g from measurements of T_c.

The main goals of the paper are to motivate the universal phase diagram through a discussion of the relevant data, and then to provide a physical basis for the phase diagram by appealing to theories for T_c and T_g. It should be noted that Ginsberg[4] introduced the concept of universal behavior when he considered the relationship between T_c and the temperature at which correlations between the magnetic atoms become important.

The proposed phase diagram is shown in Fig. 1. The curve representing $T_c(x)$ is that of Abrikosov and Gorkov (AG)[5] and others.[6,7] The $T_c(x)$ curve is dashed for values of x where interactions between magnetic atoms often result in deviations from theory. The line representing $T_g(x)$ assumes that $T_g \propto x$ for all x, in agreement with the data described below. It also assumes that superconducting order does not inhibit spin-glass order, in agreement with measurements on *La*Eu described below, and with calculations.[2] The two most important features of the phase diagram in Fig. 1 are that the ratio $(dT_g/dx)/(-dT_c/dx)_{x\to 0}$ is about 0.13, regardless of the values of T_{c0} and x_{cr}, and that spin-glass ordering always occurs in the superconducting state, with $T_g \ll T_c$ for $x \lesssim 0.8x_{cr}$.

Representative data for three alloys are shown in Fig. 2. The solid curves represent the AG theory with values of T_{c0} and x_{cr} from Table I. The dashed lines show that T_g is approximately proportional to x, even when x is larger than a few mole percent. Note that $T_g(x) = T_c(x)$ at $T_c \approx 0.1T_{c0}$, in agreement with Fig. 1.

Data on nine alloys are summarized in Table I, where $T_{c0} \equiv T_c(x=0)$; $-(dT_c/dx)_0$ is the initial rate of depression of T_c with increasing x; $x_{cr} = 0.691T_{c0}/-(dT_c/dx)_0$ is the

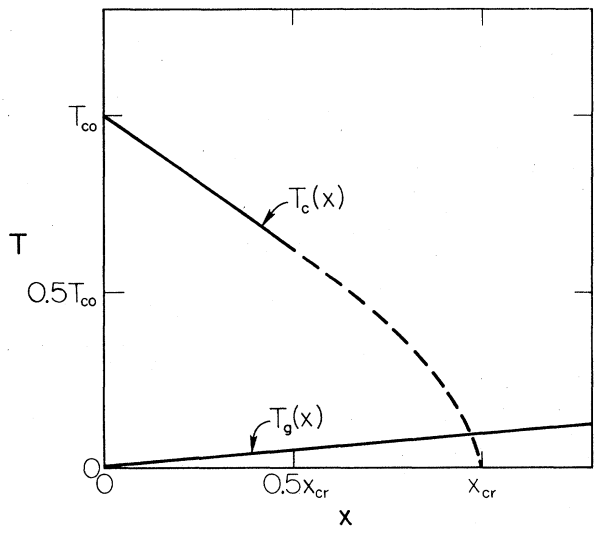

FIG. 1. Universal phase diagram for superconducting spin glasses. The ratio of dT_g/dx to $-(dT_c/dx)_0$ is about 0.13, so that $T_g = T_c$ at $T_c = T_{c0}/10$.

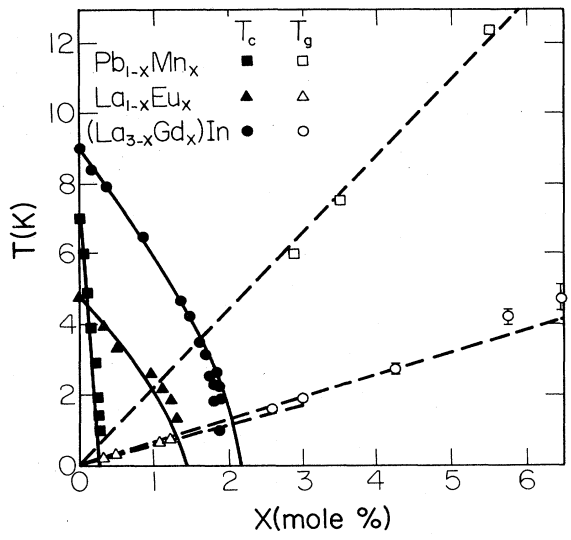

FIG. 2. Graph of $T_c(x)$ and $T_g(x)$ for three alloys, for comparison with Fig. 1. The solid curves represent the AG theory, with T_{c0} and x_{cr} from Table I. The dashed lines show that T_g is approximately proportional to x.

TABLE I. Superconducting and spin-glass properties of nine alloys that show both phenomena.

System	T_{c0} (K)	$-(dT_c/dx)_0$ (K/mole%)	x_{cr} (mole%)	dT_g/dx (K/mole%)	x range (mole%)	$\frac{dT_g}{dx} / -\left(\frac{dT_c}{dx}\right)_0$
$Pb_{1-x}Mn_x$ (film)	7.20[a]	16[a]	0.31[a]	2.2[b]	3–10[b]	0.138
(film)	7.00[c]	20[c]	0.24[c]			0.110
(film)	7.18[d]	21[d]	0.24[d]			0.105
$Zn_{1-x}Mn_x$ (bulk)	0.857[e]	305[e]	0.0019[e]	22[f]	< 0.01[f]	0.072
(film)	1.58[e]	285[e]	0.0038[e]			0.077
$La_{1-x}Gd_x$	5.7[g]	4.8[g]	0.82[g]	0.56[h]	5[h]	0.117
$La_{1-x}Eu_x$	4.8[i]	2.4[i]	1.4[i]	0.57[i]	0.3–1.2[i]	0.24
$(La_{1-x}Gd_x)Al_2$	3.24[j]	3.8[j]	0.59[j]	0.30[k]	0.6–1[k]	0.079
$(La_{3-x}Gd_x)In$	9.0[l]	2.9[l]	2.15[l]	0.63[h]	2.5–6[h]	0.22
$(La_{0.8-x}Gd_x)Au_{0.2}$	3.55[m]	3.5[m]	0.70[m]	0.54[m]	4–20[m]	0.15
$(La_{0.8-x}Sm_x)Au_{0.2}$	4.5[n]	2.1[n]	1.5[n]	0.34[n]	10–80[n]	0.16
$(Pd_{1-x}Mn_x)H$	9.3[o]	21[o]	0.33[o]	0.71[p]	3–10[p]	0.034

[a]Reference 8. [b]Reference 11. [c]Reference 9. [d]Reference 10. [e]Reference 12. [f]Reference 13 [g]Reference 14. [h]Reference 15. [i]Reference 16. [j]Reference 17. [k]Reference 18. [l]Reference 19. [m]Reference 20. [n]Reference 21. [o]Reference 22. [p]Reference 23.

critical concentration for superconductivity calculated from the data by using the AG theory; dT_g/dx is the slope of $T_g(x)$; and x range is the concentration range over which T_g was measured. Note that in *La*Eu T_c and T_g were measured for the same values of x.

T_g was determined from the maximum in the magnetic susceptibility χ, except in *La*Eu where it was determined from Mössbauer measurements. In (*La*Gd)Al_2, T_g was frequency dependent, and the value of dT_g/dx in Table I is from data taken at 117 Hz; it would be only 5% higher if the data at 10 Hz were used. The uncertainties in the experimental values of dT_g/dx and $(dT_c/dx)_0$ may be as large as 20%–30%, but since they are not large enough to affect the major features of the phase diagram, they will not be discussed in detail.

Several general features of the data in Table I must be emphasized. First, Table I includes all alloys for which T_c and T_g have been measured, and for which $(-dT_c/dx)_0 \gtrsim 2$ K/mole%. The data on each of these alloys are consistent with the proposed phase diagram, with the exception of (*Pd*Mn)H. All other alloys that are not consistent with the proposed phase diagram, e.g., the Ru-based Laves-phase compounds,[1] have $|(dT_c/dx)_0| \lesssim 0.4$ K/mole%. The compound (*Th*Nd)Ru_2 shows an initial increase, not a decrease, in T_c of 0.1 K/mole%,[24] suggesting that alloying effects on T_c may be as important as pair-breaking effects in these compounds. Second, $T_g \propto x$ is observed for each alloy (in *La*Gd only one data point is available). Third, the alloys cover a sufficiently wide variety of host metals and magnetic dopants, including simple metal/transition metal (*Pb*Mn), transition metal/transition metal (ZnMn), rare earth/rare earth (*La*Gd, *La*Eu), and intermetallics $(La_{1-x}Gd_x)Al_2$, $(La_{3-x}Gd_x)In$, $(La_{0.8-x}Gd_x)Au_{0.2}$, and $(La_{0.8-x},Sm_x)Au_{0.2}$, to justify a universal phase diagram.

The key point to be derived from Table I is that $(dT_g/dx)/(-dT_c/dx)_0 = 0.13$, within a factor of 2, for these diverse systems, even though the values of x_{cr} vary by a factor of 1000 and values of T_{c0} vary by a factor of 10. This remarkable result is the basis for the phase diagram in Fig. 1. The following analysis shows that the physical reason for this constant ratio is that both $(dT_c/dx)_0$ and dT_g/dx are determined entirely by the exchange scattering rate of conduction electrons from the magnetic atoms.

AG first showed that in the Born approximation T_c vs x has the form shown in Fig. 1, provided that the magnetic atoms are isolated from each other. A key result of their theory is that the initial decrease in T_c is proportional to the exhange scattering rate, so that

$$-(dT_c/dx)_0 = \pi\hbar/4k_B x\tau_{ex0} \quad (x \ll x_{cr}) \ , \tag{1}$$

where $1/\tau_{ex0}$ is the value of the exchange-scattering rate in the absence of impurity-impurity interactions, and it is proportional to x. Corrections due to spin-glass correlations were considered by Soukoulis and Grest,[3] and are expected to be small, at least for $x \lesssim 0.8x_{cr}$. Corrections to the AG theory from higher-order terms can be important when the magnetic dopant is a transition element.[6,7] [These corrections may explain the anomalous behavior of (*Pd*Mn)H.] The present theory of spin glasses is not sufficiently sophisticated to include them in a calculation of T_g, so they are omitted here in the calculation of T_c as well.

The Edwards-Anderson[25] (EA) theory of spin glasses can be used to calculate T_g. In the EA theory, the combination of a spatially oscillating Ruderman-Kittel-Kasuya-Yosida (RKKY)[26] interaction and a random magnetic dopant distribution is approximated by a regular lattice of dopant atoms whose interaction strength is random, with a Gaussian distribution. One result of the theory is

$$k_B T_g = \sqrt{2/9} J_{rms} \ , \tag{2}$$

where J_{rms} is the rms strength of the RKKY interaction between spin i and all other spins $[j]$, averaged over all configurations of $[j]$. The value of J_{rms} can be estimated by combining some physically reasonable approximations with what is known about the RKKY interaction.

The RKKY interaction occurs because the dopant spins $\mathbf{S}_i$ polarize nearby conduction electron spins $\mathbf{s}_j$ via the exchange interaction

$$H_{\rm ex} = -J\mathbf{S}_i \cdot \mathbf{s}_j \ , \tag{3}$$

where J is the exchange constant. The exchange interaction between the polarized electrons and nearby solute spins gives rise to the indirect RKKY interaction. This interaction can be written

$$H_{\rm RKKY} = J_{ij}\mathbf{S}_i \cdot \mathbf{S}_j / S^2 \ , \tag{4}$$

where

$$J_{ij} = A\left[-\sin(2k_F R_{ij})/2k_F R_{ij} + \cos(2k_F R_{ij})\right] \ , \tag{5}$$

and[27]

$$A = 3\pi z J^2 S^2 N(0)\Omega/(2k_F R_{ij})^3 \ . \tag{6}$$

In these relations, Ω is an atomic volume, S the magnitude of the dopant spins, $N(0)$ the volume density of states per spin, k_F the Fermi wave vector, z the valence of the host metal, and R_{ij} the separation between spins i and j.

To estimate the rms value of J_{ij}, assume that the magnetic atoms are arranged approximately on a simple cubic lattice with an atomic concentration x. Then each magnetic atom has six nearest neighbors, and is separated from them by an average distance given by

$$R_{ij}^3 \sim \Omega/x \ . \tag{7}$$

It is reasonable to neglect the sine term in J_{ij}, at least for concentrations less than ~ 1 at.%. Also, because J_{ij}^2 drops off as $1/R_{ij}^6$, the cumulative effect of magnetic atoms beyond nearest neighbor is negligible. From Eqs. (5)–(7)

$$J_{\rm rms} = \sqrt{6}6\pi z J^2 S^2 N(0) x/8k_F^3 \ . \tag{8}$$

The exchange-scattering rate of conduction electrons from uncorrelated spins, $1/\tau_{\rm ex0}$, is, in the Born approximation,

$$1/\tau_{\rm ex0} = x\pi N(0)\Omega J^2 S^2/2\hbar \ . \tag{9}$$

$1/\tau_{\rm ex0}$ is the high-temperature value of the exchange-scattering rate. Combining Eqs. (2), (8), and (9) with the free-electron relation, $k_F^3 = 3\pi^2 z/\Omega$, yields the final result

$$k_B T_g \approx 0.03\hbar/\tau_{\rm ex0} \ . \tag{10}$$

The ratio,

$$(dT_g/dx)/(-dT_c/dx)_0 = 0.04 \ , \tag{11}$$

follows from Eqs. (1) and (10). Equation (11) demonstrates that the universal phase diagram should describe any alloy in which the exchange interaction is the dominant interaction between the magnetic atoms and the conduction electrons. It is reassuring that the quantitative result of the calculation, 0.04, is so close to the average experimental value, 0.13, despite the crudeness of the approximations used in the estimates of T_c and T_g.

In summary, in this paper a universal phase diagram was presented for superconducting spin glasses. Published data on eight alloys with $-(dT_c/dx)_0 \gtrsim 2$ K/mole% were shown to be consistent with the phase diagram; the only exception was (*Pd*Mn)H. A simplified analysis showed that the physical basis for this diagram was the proportionality of both the depression in T_c and the increase in T_g to the exchange-scattering rate of the conduction electrons. These results enable one to make a reasonable estimate for T_g from measurements of T_c, and they reinforce the view that spin-glass ordering results primarily from the RKKY interaction.

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