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SPIN GLASS - PARAMAGNETIC PHASE BOUNDARY IN AMORPHOUS MAGNETIC ALLOYS


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Abstract.- A compositionally disordered magnetic alloy $(A_xB_1-x)_cD_{1-c}$ where $A$ and $B$ are magnetic species while $D$ represents the nonmagnetic atoms is considered. The competition among the exchange interactions $J_{AA}$, $J_{AB}$ and $J_{BB}$ in these alloys can lead to the disappearance of the ferromagnetism (for ferromagnetic $J_{MM}$) below a critical concentration $x_c$. For $x < x_c$, we show that while the appearance of the spin glass state at low temperatures is dependent on the relative signs of the exchange interaction, the details of the phase boundaries are controlled by the relative strength of the exchange interactions. These conclusions are borne out by our experimental results on the amorphous metal-metalloid alloys $(Fe_xNi_{1-x})_cG_{25}$, $(Fe_xMn_{1-x})_cG_{25}$ and $(Co_xMn_{1-x})_cG_{25}$ where $G_{25}$ represents $FeAl_{13}$. The magnetic phase diagrams for the three alloy systems were obtained from the ac susceptibility measurements and while the slope of the spin glass-paramagnetic phase boundary in the vicinity of the tricritical point is negative for the Fe-Ni alloy, it is almost zero for the Fe-Mn alloy and is positive for the Co based alloy.

Introduction. Spin glass behavior has been observed in a large number of systems including dilute alloys, where magnetic impurities are distributed at random on a crystalline lattice; or concentrated alloys, which are compositionally and/or topologically disordered [1]. Both metallic and insulating solids have been shown to exhibit the spin glass state and it is well established that competing exchange interactions, when present in adequate proportion, are the essential element for the existence of such a state.

In a previous paper [2], we have obtained the magnetic phase diagram for a bond disordered alloy in which ferromagnetic and antiferromagnetic bonds are randomly distributed on a lattice with a probability distribution $P(J) = x\delta(J-J_1) + (1-x)\delta(J-J_2)$, where $J_1 > 0$, $J_2 < 0$ and $x$ is the concentration of $J_1$ bonds while $(1-x)$ that of $J_2$ bonds. For such a distribution, it was shown [2] that below a critical concentration $x_c$, the spin glass state results with a transition temperature, $T_{sg}$, proportional to $[xJ_1^2 + (1-x)J_2^2]^{1/2}$. Thus the spin glass-paramagnetic phase boundary, defined by $dT_{sg}/dx$, exhibits an initial slope which is positive, zero or negative depending on whether the magnitude of $J_1$ is greater than, equal to or less than the magnitude of $J_2$. The zero slope case results from the fact that for $J_1 = |J_2|$, $T_{sg}$ is independent of $x$. The situation is equivalent to the case of symmetric continuous distribution of $J$ (e.g. Gaussian distribution) which also leads to a phase boundary with zero slope [3].

In the present investigation we analyze the random site disordered model and show the validity of the general conclusions regarding the phase boundary briefly described above for the bond case. Certain modifications that result for the site case and the relevance of the conclusions to our experimental data on amorphous metal-metalloid alloys is described below.

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Model. A compositionally disordered random magnetic alloy \((A_xB_{1-x})_cG_{1-c}\) is considered where A and B are magnetic species while G represents non-magnetic atoms. The exchange interaction \(J_{AA}\) is taken to be positive so that the pure A system \((x=1, c=1)\) is ferromagnetic below a certain critical temperature \(T_c\). The system remains ferromagnetic with reduced values of \(T_c\), for values of \(c\) in the range \(c_0 < c < 1\) where \(c_0\) is the critical concentration for the onset of ferromagnetism in the alloy \(A_cG_{1-c}\). For \(c \geq c_0\), we investigate the disappearance of ferromagnetism in the alloy \((A_xB_{1-x})_cG_{1-c}\), as interactions \(J_{AB}\) and \(J_{BB}\) are introduced where at least one of them is antiferromagnetic. It is shown that for \(x < x_0\), depending on the temperature, ferromagnetism disappears into a paramagnetic or a spin glass state. Of particular interest in the present study is the phase boundary between these two states which is found to be determined by the relative strength of \(J_{AB}\) and \(J_{BB}\) with respect to \(J_{AA}\). In particular, \(T_{BG}\) is found to increase, decrease or stay constant with decreasing \(x\) depending on the relative strengths of the exchange constants. The confirmation of these trends is found in our experimental results on amorphous metal-metalloid alloys with \(c = 0.75\) and \(A = \text{Fe or Co}\) while \(B = \text{Ni or Mn}\), and \(G,_{25} = \text{P,}_{18}\text{B,}_{04}\text{Al,}_{25}\). The magnetic phase diagrams for these alloys are obtained from ac susceptibility measurements and it is found that in vicinity of the tricritical point, \(T_{BG}\) increases with decreasing \(x\) for \((\text{Co}_x\text{Mn}_{1-x}),_{75}\text{G,}_{25}\) while it decreases in \((\text{Fe}_x\text{Ni}_{1-x}),_{75}\text{G,}_{25}\) and is essentially constant for \((\text{Fe}_x\text{Mn}_{1-x}),_{75}\text{G,}_{25}\). The random alloy \((A_xB_{1-x})_cG_{1-c}\) is represented by a Heisenberg Hamiltonian which is analyzed within the Bethe-Peierls-Weiss approximation. The details of the methodology have been previously described for amorphous binary alloys [4], spin glasses with Gaussian distribution of exchange interactions [5] and bond disordered random magnet [2], and will not be repeated here. It is sufficient to state that in BPU approximation, one considers a cluster of a central spin \(S_o\) and its 2 nearest neighbors \(S_A\)'s embedded in an effective medium defined by an internal field \((H_1-H_2)\) which acts on the atoms on the surface of the cluster; \(H_2\) being the externally applied field. The Hamiltonian is then written as

\[
3c = -\frac{1}{z} J_{AA} S_o \cdot S_A - H_z \cdot S_A - H_1 \cdot S_A
\]  

where \(J_{AA} = J\) is a variable which for our random alloy takes on values according to the probability distribution

\[
P(J) = x^2 J^2 \delta(J-J_{AA}) + (1-x)^2 J^2 (J-J_{BB}) + 2x(1-x)J^2 \delta(J)
\]  

The partition function \(Z\) for the above Hamiltonian is most easily calculated in the limit of large \(z\), where it can be obtained in a form that facilitates [2, 4] taking the configurational average of the free energy (i.e., \(\ln Z\)). Taking the Edwards-Anderson order parameter for the spin glass state [6], we find that the phase boundaries are defined by the equations [2],

\[
2\langle J^2 \rangle + 2zQ\langle J \rangle + 3 = 0 \text{ Ferro-Para}
\]

and

\[
\langle J^2 \rangle = \frac{1}{2} (2zQ^2 - 3) \text{ Para-Spin glass}
\]

where,

\[
J = \frac{3S}{2KQ} = aJ \text{ and } Q = \left( \frac{2}{3} zS^2 \right)^{1/2}
\]

The angular brackets in Eqs. (3) and (4) represent the configurational average over the probability distribution [Eq. (2)], which is readily performed to obtain,

\[
\langle J \rangle = ac^2 J_{AA} [x^2 + (1-x)^2 \delta + 2x(1-x)\alpha]
\]
\[ \langle J^2 \rangle = a^2 c^2 J_{AA}^2 [x^2 + (1-x)^2 \beta^2 + 2x(1-x)\alpha^2] \]  

where \( \alpha = J_{AB}/J_{AA} \) and \( \beta = J_{BB}/J_{AA} \). These equations are used to obtain complete phase diagrams and details of the calculation, without the large \( z \) restriction, will be published elsewhere [7]. Here our main concern is the spin glass-paramagnetic phase boundary and this is obtained by substituting Eq. (7) in Eq. (4) to obtain \((S=1)\)

\[ \frac{[kT_{SG}]}{[\alpha_{AA}]} = \frac{z}{2} \frac{z-1}{c^2} [x^2 + 2x(1-x)\alpha^2 + (1-x)^2 \beta^2]. \]  

Therefore the sign of \( dT_{SG}/dx \) is determined by the expression \([x + (1-2x)\alpha^2 - (1-x)\beta^2]\). Thus, in the vicinity of the tricritical point \( x_{st} \), \( dT_{SG}/dx \) is positive, zero or negative depending on whether

\[ |\beta| > \left[ \frac{x_{st} + (1-2x_{st})\alpha^2}{(1-x_{st})} \right] \]  

Schematic phase diagram based on above considerations is shown in Figure 1.

Experiment. All the alloys in the three systems, Co-Mn, Fe-Mn and Fe-Ni, were prepared in the form of ribbons by the melt spinning technique on the outer surface of a rotating copper-bronze disc [8]. X-ray techniques were used to ascertain the amorphous nature of the alloys. Low field ac-susceptibility techniques were used to determine the magnetic ordering temperatures in both the ferromagnetic and spin glass regimes of the alloys.

About 3 mm lengths were cut from ribbons of amorphous alloys of approximately 1.55 mm(25-40) \( \mu \)m cross-section and packed to form a cylinder embedded in candle wax. This was then packed in the coil of an ac-susceptibility bridge [9] with the longer dimension along the coil axis. The ac-susceptibility was measured in a field of \( \sim 3 \) Oe (rms) at 300 Hz. A check at several frequencies between 100 Hz and 1 KHz showed no significant differences in the data obtained. Data taken both on warming and cooling the sample were completely reproducible. The absolute temperature of the sample, in good thermal contact through an He exchange gas with the cooling/warming chamber, was determined using a calibrated platinum thermometer. In the ferromagnetic regime, the ac-susceptibility as a function of temperature exhibited a sharp rise near \( T_C \), and achieved a constant value, determined by the appropriate demagnetization factor, below it. The temperature corresponding to the 'kink-point' was then taken as the Curie temperature \( T_C \). The values of \( T_C \) thus obtained agree to within \( \pm 2K \) with those determined by other standard techniques like, for example, VSM measurements. At concentrations corresponding to the spin glass regime, a sharp characteristic ac-susceptibility cusp was observed for all the alloys and the spin glass temperature, \( T_{SG} \), was thus determined to better than \( \pm 1K \).

Results. In the Fe-Mn alloys, Fe and Mn possess comparable moments and while nearest neighbor Fe-Fe pairs are ferromagnetically coupled, the nearest
neighbor Mn-Mn and Fe-Mn pairs are antiferromagnetically coupled. Figure 2 depicts the observed

![Figure 2](image)

Phase diagrams for the amorphous alloy series \((\text{Fe}_x \text{Mn}_{1-x})_{0.75} \text{Ge}_{0.25}\) of the transition temperatures as a function of the alloy composition for the amorphous \((\text{Fe}_x \text{Mn}_{1-x})_{0.75} \text{Ge}_{0.25}\) series. Ferromagnetism prevails for \(x > 0.6\) and increasing concentration of Mn introduces enough competition among exchange interactions (frustration) to lead to the spin glass phase. It has been shown that the magnetic susceptibility along the spin glass-paramagnetic phase boundary as well as the ferromagnetic-paramagnetic boundary of this series of alloys satisfies a scaling hypothesis appropriate to a multicritical point common to both boundaries [10]. The value of \(T_{sg}\) and the magnitude of the susceptibility at \(T_{sg}\) both decrease with decreasing value of \(x\) from 0.6 to 0.4. The initial drop in \(T_{sg}\) near the multicritical point is however very slow and the slope of the paramagnetic-spin glass phase boundary is almost zero corresponding to the equality sign in Eq. (9).

Figure 3 shows the concentration dependence of the transition temperatures \((T_C, T_{sg})\) for the alloy series \((\text{Co}_x \text{Mn}_{1-x})_{0.75} \text{Ge}_{0.25}\). Again, while this alloy is a ferromagnet for \(x > 0.75\), a spin glass type behavior is observed for \(x < 0.6\). However, in contrast to the behavior observed in the case of Fe-Mn alloys, \(T_{sg}\) increases with higher concentrations of Mn for this system; a behavior predicted by the upper sign in Eq. (9). In addition, it is found that the maximum value of the susceptibility of \(T_{sg}\) decreases rather rapidly with further addition of Mn in this alloy system; so much so that it was not possible to obtain reliable \(T_{sg}\) values for \(x < 0.4\) by this method. It is useful to point out that such an unusual behavior of the concentration dependence of \(T_{sg}\) has also been reported recently for crystalline Co-Mn alloys [11]. A further evidence for the observed slope of the spin glass-paramagnetic boundary is obtained from the ac-susceptibility data for alloy compositions close to the multicritical point. For example, it is found that for \(x = 0.7\) alloy the ac-susceptibility exhibits a sharp characteristic kink-point correspond-
ing to the ferromagnetic transition, $T_c = 110K$, which is then followed by a sharp drop in $x_{ac}$ characteristic of a spin glass phase at a lower temperature $T_{sg} \approx 60K$. Further details of these will be published elsewhere [12]. These lower transitions observed for alloy concentrations close to the multicritical point extrapolates well to the spin glass line as shown in Figure 3.

While the two amorphous alloy series described above belong to the concentrated spin glass systems, the series $(Fe_{x}Ni_{1-x})_{0.75}S_{0.25}$ belongs to a dilute spin glass system. For the particular composition of the glass former chosen, Ni has essentially zero moment and simply acts as a diluant. The behavior in the spin glass region is indeed found to be similar to the dilute crystalline Au-Fe series [13]. As shown in Figure 4, the spin glass state sets in only for $x < 0.18$ and corresponding to the lower sign in Eq. (9), the value of $T_{sg}$ decreases with decreasing $x$.

![Phase diagram for the amorphous alloy series $(Fe_{x}Ni_{1-x})_{0.75}S_{0.25}$.](image)

References


