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# Spin-Glass–Ferromagnetic Multicritical Point in Amorphous Fe-Mn Alloys

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The ac susceptibility of a sequence of amorphous Fe-Mn alloys shows lines of both spin-glass and ferromagnetic transitions. A detailed scaling-law analysis, treating the common point on the two magnetic phase boundaries as a multicritical point, verifies the scaling hypothesis and yields multicritical, spin-glass, and crossover exponents. The observed behavior compares quite will with position-space renormalization results.

It is now generally accepted that materials with competing exchange interactions may become spin-glasses at low temperatures, rather than acquiring long-range order in the usual sense.[1-3] There is far from general agreement, however, as to whether the spin-glass state is achieved through a phase transition or a more gradual freezing process. In this Letter, we report the results of a detailed study of the transition from a ferromagnet to a spin-glass as a function of material composition. We show, for the first time, that the magnetic susceptibility along both the spin-glass and ferromagnetic lines satisfies a scaling hypothesis appropriate to a multicritical point common to both lines. This strongly implies that there is a competition between the length scales for ferromagnetic and spin-glass order, with both simultaneously divergent at the special point, which we will refer to as "bicritical." In the course of the analysis, the critical, bicritical, and crossover exponents are also determined. We assert that the applicability of scaling laws along the spin-glass line indicates that this is a line of phase transitions.

The materials chosen for this study are well-characterized amorphous alloys with the composition $(Fe_{1-x}Mn_x)_{75}P_{16}B_6Al_3$.[4,5] Alloys were prepared with $0 \leq x \leq 1$ by centrifugal spin quenching.[6] We report here results for alloys in the range $0.3 \leq x \leq 0.6$, in steps of 0.05. There are several reasons for this choice of material:

(i) The alloys can be prepared in a single phase for all values of $x$.[6]

(ii) Rapid quenching preserves the liquid state of the melt, thus avoiding possible chemical clustering effects.

(iii) Crystal-field, magnetocrystalline-anisotropy, and grain-boundary effects are minimized in these amorphous materials.

(iv) Fe and Mn atoms have local moments with nearly equal spin, so that the material approximates a bond-random model for the ferromagnet–spin-glass phase diagram.

Ribbons of the amorphous alloys were cut to approximately 3 mm× 1 mm× 25 μm and packed in the coil of an ac susceptibility bridge, with the longer dimension along the coil axis. The ac susceptibility was measured in a field of ≈ 3 Oe (rms) at 400 Hz, but a check at several frequencies between 100 Hz and 1 kHz showed no significant differences. Data taken both during warming and cooling were completely reproducible. The susceptibility at larger values of $x$ shows the characteristic spin-glass cusp as may be seen in Fig. 1. As $x$ is reduced, the peak temperature increases, as does the peak amplitude. At $x = 0.4$, the ac susceptibility signal becomes equal to the inverse of the demagnetizing factor $N$. For smaller $x$, the ac susceptibility shows a flattened top, indicating that the susceptibility is much larger than $N^{-1}$. To proceed with an analysis of the susceptibility, we first calculate the actual suscepti-

 

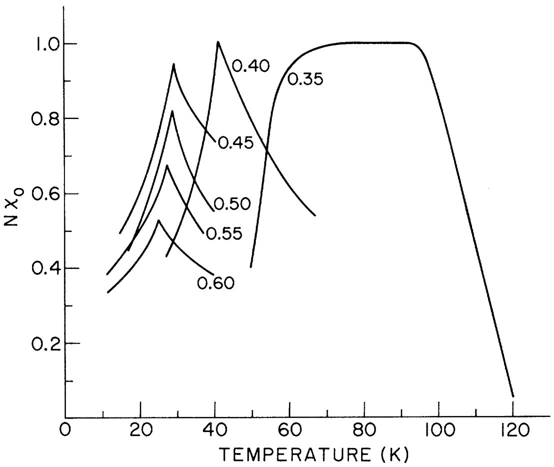


FIG. 1. Measured ac susceptibility scaled by $N^{-1}$, the inverse of the demagnetizing factor, for various Mn concentrations.

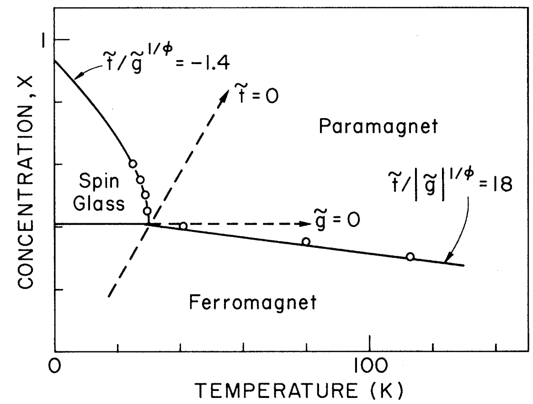


FIG. 2. Experimentally determined transition temperatures (circles) and the parametrized phase boundaries (solid lines). The dashed lines are the axes defined by the scaling fields $\tilde{g}$ and $\tilde{t}$, as described in the text.

bility from

$$\chi^{-1}=\chi_{\rm obs}^{-1}-N. \tag{1}$$

This leads to considerable error when $\chi_{\rm obs}$ is nearly equal to $N^{-1}$. We are therefore not able to obtain the susceptibility with any reliability for $x<0.4$. Because of the composite nature of the sample, $N$ is determined empirically.

As is evident from Fig. 1, there is no sharp distinction between a random ferromagnet and a spin-glass. In cases such as this, the crossover between one type of order and another is precisely described by scaling with respect to the multicritical point common to both the phases.[7] It is all the more important to use the full power of scaling theory here, since it is not really known that the spin-glass transitions form a line of critical points. We proceed by hypothesizing the existence of a multicritical point and then collapsing the data along the spin-glass line according to the dictates of scaling. Since the multicritical point defines for itself the directions of relevant fields, we can distinguish between spin-glass and a ferromagnet according to the scaling properties of the susceptibility and the peak position relative to the scaling-determined multicritical point. As we show, this leads to an internally consistent analysis, and gives the first detailed information about this important crossover effect.

From Fig. 1, we construct the bicritical phase diagram[7] in Fig. 2. The spin-glass points are taken as the cusp temperatures, while the ferromagnetic transition is estimated from the onset of the flattened portion, and is consequently less precise. Unlike the corresponding spin-flop bicritical point,[7] the line of ferromagnetic transitions curves upward, indicating that the crossover exponent is less than unity, as we shall see. It may be that the concentration axis is not an appropriate scaling field for an exact theory, but we shall proceed, assuming that concentration measures the random field which competes with ferromagnetic order.

To construct a scaling hypothesis for the random bicritical point, we require two scaling fields $\tilde{g}$ and $\tilde{t}$. These are functions of the reduced temperature and concentration, $t=T/T_b-1$ and $g=x/x_b-1$. The line $\tilde{g}=0$, by analogy to the spin-flop case, should continue the first-order line separating the spin-glass and ferromagnetic phases.[7] We do not observe any feature in our data indicating such a transition, and so assume, arbitrarily, that this line is parallel to the temperature axis, and, therefore, not observable. This means that $\tilde{g}=g$, which simplifies the analysis. We next seek a temperature scaling field of the form

$$\tilde{t}=t+q\tilde{g}, \tag{2}$$

such that the two phase boundaries may be parametrized by an exponent $\varphi$ and a constant according to

$$\tilde{t}/|\tilde{g}|^{1/\varphi}=\begin{cases}K_{\rm FM}, & \text{along the ferromagnetic line}\\ -K_{\rm SG}, & \text{along the spin-glass line.}\end{cases} \tag{3}$$

We find that, with $q=-0.71$ and $\varphi=0.77\pm0.05$, the two lines may be fitted with $K_{FM}=18$, and $K_{SG}=1.4$. Note that at $g=-1$ (pure Fe glass) the ferromagnetic transition is predicted to occur at $(19\pm 1)T_b = 574\pm 30$ K in good agreement with the $T_C = 620$ K observed (as quenched samples have $T_C \simeq 580$ K).[4] The bicritical point was taken to be at $x_b = 0.41$ and $T_b = 30.2$ K.

Having chosen a pair of scaling fields, we may now make a scaling hypothesis for the susceptibility. By the usual multicritical scaling arguments[8] the scaling form will be

$$\chi(T,x)=|\tilde{g}|^{-\gamma/\varphi}\chi^*(\tilde{t}/|\tilde{g}|^{1/\varphi}). \qquad (4)$$

The crossover exponent $\varphi$ is the same as determined above, and $\gamma$ is the susceptibility exponent for the bicritical point. For positive values of $\tilde{g}$ the scaling function $\chi^*(y_2)$, where $y_2=\tilde{t}/|\tilde{g}|^{1/\varphi}$, describes the crossover from bicritical to spin-glass behavior, with values along the spin-glass line corresponding to $y_2=-K_{SG}$. Equation (4) then reflects the divergence of the peak value of the spin-glass susceptibility cusp as $\tilde{g}\to 0$ (approaches the bicritical point). For negative $\tilde{g}$, the crossover is to the ferromagnetic critical line.

The two branches of the scaling function $\chi^*(y_2)$ can be plotted together by defining a new variable[8]

$$y_2' = \begin{cases} y_2+K_{SG}, & \text{for } \tilde{g}>0 \\ y_2-K_{FM}, & \text{for } \tilde{g}<0. \end{cases} \qquad (5)$$

Because $\tilde{g}$ is parallel to the temperature axis, $y_2'$ is particularly easy to define experimentally, being just $y_2'=[T-T_{f,C}(\tilde{g})]/T_b|\tilde{g}|^{1/\varphi}$, where $T_{f,C}(\tilde{g})$ is the freezing or Curie temperature appropriate for the concentration being studied. Clearly, far from the critical lines the distinction between the two branches should be unimportant and bicritical behavior will dominate.

In Fig. 3, we have plotted the susceptibility data derived from Fig. 1 after scaling according to (4) and (5). Only one ferromagnetic concentration is included ($x=0.4$), and it is seen to scale with the spin-glass data for large values of $|y_2'|$. The bicritical exponent used in this plot was $\gamma = 1.18 \pm 0.08$. Since the susceptibility at $\tilde{g}=0$ should diverge as $|t|^{-\gamma}$, the scaling function must have the asymptotic limit $\chi^*(y_2')\sim|y_2'|^{-\gamma}$. The solid lines for large $|y_2'|$ in Fig. 3 are plots of this asymptotic behavior. The amplitudes are in the ratio $A^+/A^- \simeq 3$. The data for $x=0.4$ follow this behavior out to $|y_2'|\sim 70$. In the inset to Fig. 1 are plotted the data close to the spin-glass line. The data scale very well, and approach the line in a sharp cusp. The solid lines are of the form $a-b|y_2'|^{1/2}$; the values of $a$ are the same for positive and negative $y_2'$, with the amplitudes in the ratio $b^+/b^- \simeq 1.2$.

The data of Fig. 3 indicate that the bicritical point dominates the susceptibility for $y_2' \lesssim -2$ and for $y_2' \gtrsim 6$ although the data for the $x=0.4$ alloy follow the bicritical behavior to smaller

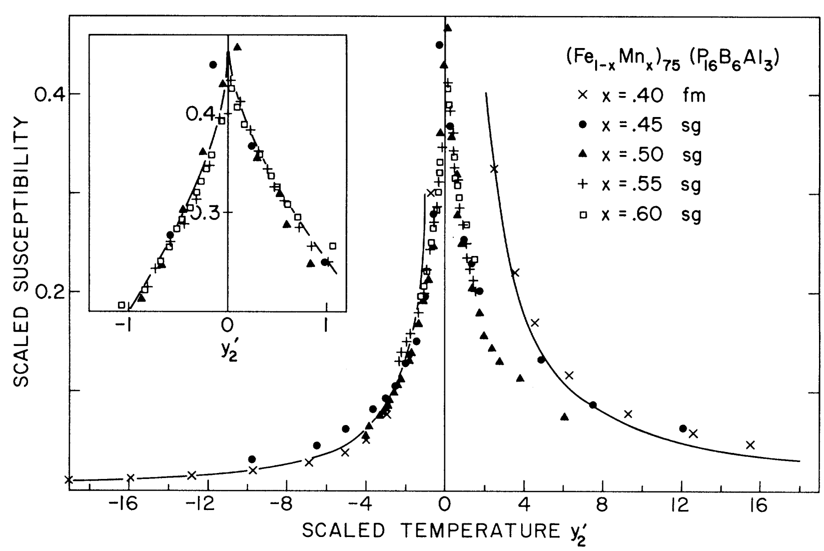


FIG. 3. Scaled susceptibility $\chi^*$ vs scaled temperature, $y_2'$, for five compositions. The solid lines for large $|y_2'|$ are proportional to $|y_2'|^{-\gamma}$. The inset shows the region close to the spin-glass phase boundary. The solid lines there are of the form $a-b|y_2'|^{1/2}$. The scaling exponents for this plot are $\varphi = 0.77$ and $\gamma = 1.18$.

$y_2'$. This crossover to bicritical behavior should be especially important in the ferromagnetic regime, since it implies a change in the development of a spontaneous magnetization and a noninfinite initial susceptibility. Indeed, for $x=0.35$, for which $g=-0.15$, crossover at $y_2'=-2$ corresponds to a temperature of 67 K, which agrees well with the observed decrease in the susceptibility for this concentration in Fig. 1. Our assertion, then, is that the drop in the ac susceptibility in the ferromagnetic phase is due not to a second-phase transition, but rather to a crossover to a bicritical behavior within that phase.

A phase diagram very similar to the one we find here has been determined by Wortis, Jayaprakash, and Riedel[3,9] from a real-space renormalization calculation of the two-δ-function model. Other models have predicted a multicritical point between spin-glass and ferromagnetic phases,[10,11] but could make no specific predictions for the exponents; here exponents are predicted for dimensions $d=2$ and $d=3$. As in our phase diagram (Fig. 2), the spin-glass/ferromagnetic phase boundary is found to be parallel to the temperature axis. The crossover exponent is predicted to be $\varphi=0.72$ for $d=2$, and $\varphi=0.91$ for $d=3$, bracketing our value for $\varphi=0.77\pm 0.05$. The exponent for the spin-glass order parameter, which is the same as that of the susceptibility at the cusp, was determined only for $d=2$, and is $\beta_{SG}=0.8$; our results give $\beta_{SG}=0.50\pm 0.08$, considerably smaller. Unfortunately, no calculation of the multicritical $\gamma$ was given for any dimensionality.

The close correspondence between the phase diagram and critical exponents obtained in the position-space renormalization calculation and that of the Fe-Mn amorphous spin-glass represents the first detailed verification of any spin-glass model. We have not, of course, established the nature of the spin-glass critical line, nor determined the order parameter directly. However, the ability to perform a scaling analyses with a single crossover exponent goes far toward establishing the general picture of a multicritical point on the ferromagnetic line and a true spin-glass phase boundary for this three-dimensional random-alloy system.

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## Exciton Dynamics within an Inhomogeneously Broadened Line

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Exciton dynamics in $Tb(OH)_3$ is shown to depend upon the excitation location within the $k\simeq 0$ inhomoheneously broadened absorption profile. The dynamics is observed by monitoring the populations of the different $k$ states from time-resolved band-to-band luminescence. The results indicate the presence of experimentally separable microscopic regions within which delocalized exciton states occur with well-defined $k$ values.

Much progress in our understanding of the broadening of optical transitions in solids has been made in recent years with the advent of new laser spectroscopy techniques working in the time[1-3] and frequency domains.[4-5] For instance, fluorescence line-narrowing techniques have