

Spin-Glass-Ferromagnetic Critical Line in Amorphous Fe-Mn Alloys

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The critical properties of a line of transitions between a spin-glass and a ferromagnet have been determined for a series of amorphous Fe-Mn alloys. Detailed scaling analysis of the magnetization leads to the conclusion that the transition is continuous and similar to the Curie transition. The values of the critical exponents are given for the first time.

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Magnetic materials with a random mixture of ferromagnetic (FM) and antiferromagnetic (AFM) bonds may have a spin-glass (SG) phase over some range of composition.¹⁻⁵ In the vicinity of the multicritical point where FM and SG transition lines intersect, there is a subtle interplay between long-range magnetic order and the randomness of the SG state. Some indications for a transition from FM to SG at fixed composition have been reported,⁶⁻⁹ but these have not been pursued adequately to establish this as a *critical* line separating the two phases nor to determine its properties. An experimental examination of such a line is the subject of this Letter.

The distinction between a spin-glass and a random ferromagnet is not sharp. Thus, a search for the FM-SG transition through the disappearance of the FM order parameter requires careful measurement of the magnetization at low fields coupled with full use of the scaling properties of thermodynamic quantities near the proposed transition. We chose for this study the series of amorphous alloys $(\text{Fe}_{1-x}\text{Mn}_x)_{75}\text{P}_{16}\text{B}_6\text{Al}_3$, previously¹⁰ shown to have a FM-SG multicritical point. Our scaling analysis of the magnetization leads to the following results:

(1) The low-field magnetization $M(H, T)$ has a maximum at a temperature T_m ($T_m < T_c$, the FM Curie temperature), and tends to zero in zero applied field as $M(0, T) \sim (T/T_{fg} - 1)^{\beta}$; T_{fg} decreases with x .

(ii) At $T = T_{fg}$, the magnetization increases with applied field as $M(H, T_{fg}) \sim H^{1-\delta}$.

(iii) The susceptibility tends to diverge as T_{fg} is approached from below as $\chi(T) \sim (1 - T/T_{fg})^{-\gamma}$.

(iv) Hysteresis and time-dependent effects set in for $T \approx T_{fg}$.

The critical behavior described by properties

(i)-(iii) is entirely analogous to an ordinary Curie transition, except that the direction along the temperature axis is reversed. The existence of a continuous transition between FM and SG phases is a feature of most mean-field models for random systems, and it is somewhat remarkable how closely the observed transition resembles the model results. A number of other features which occur in the ferromagnetic phases—the maximum in $M(H, T)$, at T_m ; the drop of the ac susceptibility⁸⁻¹⁰ at T_d ; and the characteristic temperature governing the exponential rise in the coercive field—are all considerably (25–30 K) above T_{fg} . These appear to be associated with “crossover” phenomena; that is, where random order *begins* to dominate the FM order, rather than with the FM-SG transition itself.

To carry out the magnetic measurements, ribbons of the alloys were cut to approximate dimensions $3 \text{ mm} \times 1 \text{ mm} \times 25 \text{ } \mu\text{m}$. Stacks of approximately twenty such ribbons were mounted in the sample holder of a vibrating-sample magnetometer with the longest dimension parallel to the dc field to minimize demagnetizing effects. The samples were cooled in zero field and the magnetization measured with increasing field. The sample was then heated, until the remnant magnetization completely disappeared, and then cooled again in zero field. Failure to carry out this procedure results in irreproducible results at low temperatures. At temperatures above T_m , no such difficulties were encountered.

The results of this measurement for¹¹ $x = 0.32$ are given in Fig. 1. The most striking feature of these data is the presence of a maximum in the magnetization curves. The temperature of the maximum is a decreasing function of the magnetic field. Comparable data for $x = 0.3$ and $x = 0.2$ re-

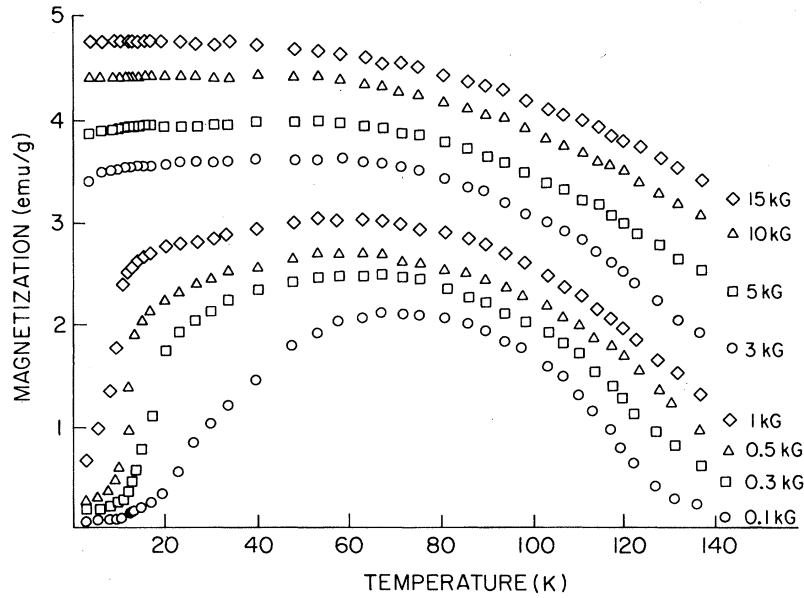


FIG. 1. Temperature dependence of the magnetization for $(\text{Fe}_{0.68}\text{Mn}_{0.32})_{75}\text{P}_{16}\text{B}_6\text{Al}_3$ for different applied fields.

veal a concentration dependence of T_m . For low external fields, T_m coincides approximately with T_d , the temperature where the ac susceptibility starts dropping from the demagnetization limit.¹⁰ Moreover, we find experimentally that the coercive field is of the same order of magnitude as the applied field at this temperature. Thus T_m , T_d , and the opening of the hysteresis loops are closely related and seem to represent the temperature at which random effects begin to dominate the behavior.

Although the decrease in M at lower temperatures is suggestive of a phase transition, detailed analysis is required to establish its existence. One possibility is to make modified Arrott plots; i.e., by plotting isotherms of $M^{1/\beta}$ vs $(H/M)^{1/\gamma}$ with the proper exponent values. However, as there is no *a priori* reason to assign particular values to them, exponents should be independently determined. A more general method employs a scaling hypothesis¹² to obtain a simple form of the magnetic equation of state in the critical region,

$$M/t^{\tilde{\beta}} = m^*(H/|t|^{\tilde{\beta}\tilde{\delta}}), \tag{1}$$

where $t = (T - T_{fg})/T_{fg}$ and we take $y = H/|t|^{\tilde{\beta}\tilde{\delta}}$. The scaling function m^* has two branches, m_+^* for $t > 0$ and m_-^* for $t < 0$. The limiting behaviors

for a critical point are

$$\lim_{y \rightarrow 0} m_+^* = \text{const}, \tag{2a}$$

$$\lim_{y \rightarrow \infty} m_{\pm}^* = y^{1/\tilde{\delta}}, \tag{2b}$$

$$\lim_{y \rightarrow 0} m_-^* = y. \tag{2c}$$

These limiting values reflect properties (i)–(iii) listed above.

In Fig. 2, we show results of the scaling procedure for the $x = 0.32$ alloy. The collapsing of the

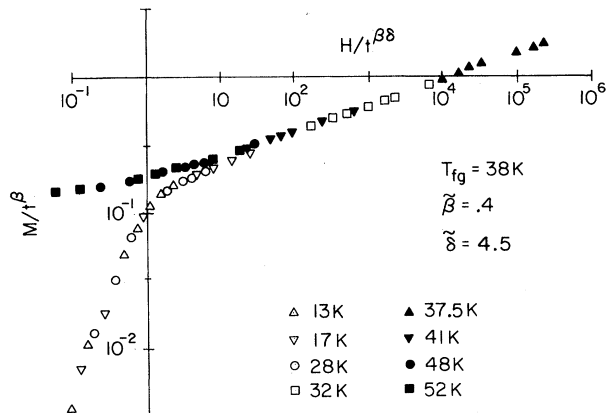


FIG. 2. Scales magnetization m/t^{β} vs scaled field $H/t^{\beta\tilde{\delta}}$ (in arbitrary units), for isotherms around T_{fg} .

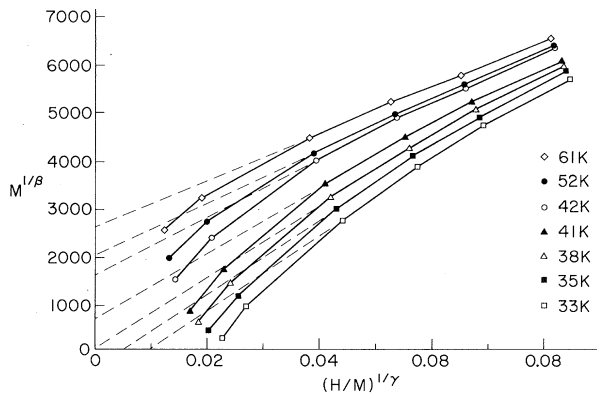


FIG. 3. Modified Arrott plot near the spin-glass-ferromagnetic transition temperature.

isotherms to two branches m_{\pm}^* exhibits a critical behavior in concordance with (2a)–2(c) with a transition temperature $T_{fg} = 38 \pm 1$ K and critical exponents $\tilde{\beta} = 0.4 \pm 0.03$ and $\tilde{\delta} = 4.5 \pm 0.3$. Data for $x = 0.3$ and $x = 0.2$ alloys were analyzed by the same procedure. We find the same critical behavior and exponents as above with transition temperatures of 31 ± 1 and 14 ± 2 K for the $x = 0.3$ and $x = 0.2$ alloys, respectively.

In Fig. 3, we have made a modified Arrott plot of the data for the $x = 0.32$ alloy using $\tilde{\beta}$ and $\tilde{\delta}$ as determined above and $\tilde{\gamma} = \tilde{\beta}(\tilde{\delta} - 1)$. The isotherm which extrapolates linearly to the origin is, as usual, the critical isotherm. For $T > T_{fg}$, there is a tendency for the curves to intersect the vertical axis, reflecting the appearance of a spontaneous moment. Lower temperature curves tend to intersect the horizontal axis and indicate the divergence of the susceptibility (actually $\chi^{1/\gamma}$) at the critical temperature. However, such a plot is clearly inferior to the scaling analysis in determining the critical behavior near T_{fg} . The general appearance of the curves differs from an ordinary ferromagnet and for fields below 100 G (the two lowest fields used in Fig. 3) demagnetization corrections become important, but cannot be treated easily because of the composite nature of the sample. We also cannot rule out the possible presence of weak (≈ 50 G) anisotropy fields at this transition.

To complete the analysis we used the same data processing for the PM-FM transition. The critical exponents are found to be $\beta_c = 0.4(\pm 0.03)$ and $\delta_c = 5(\pm 0.3)$. The Curie temperatures are 100, 107, and 295 K for $x = 0.32$, 0.30, and 0.20, respectively. We summarize the experimental results with a phase diagram (Fig. 4) for the Fe-

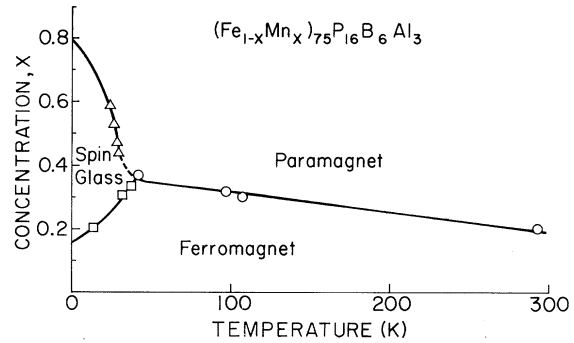


FIG. 4. Magnetic phase diagram for $(\text{Fe}_{1-x}\text{Mn}_x)_{75}\text{P}_{16}\text{B}_6\text{Al}_3$.

Mn system.¹³

The critical exponents we find, both for the FM-PM and SG-FM transitions, are quite large compared with those from the 3D Heisenberg model. Very similar values for β and δ have been previously reported for Metglas^{14,15} and other amorphous ferromagnetic alloys¹⁶ and seem to be characteristic of such random ferromagnets.

Nothing is known, however, about the SG-FM critical exponents beyond the mean-field model. Assuming scaling to hold, we find our results for $\tilde{\beta}$ and $\tilde{\delta}$ to lead to the estimate $\tilde{\gamma} = \tilde{\beta}(\tilde{\delta} - 1) = 1.4 \pm 0.2$, which is similar to the multicritical point exponent $\tilde{\gamma} = 1.2 \pm 0.1$ found in Ref. 10. The values here are closer to 3D-Heisenberg-model values of the critical exponents, a result we find somewhat surprising. Interestingly, the specific-heat exponent at the SG-FM transitions is expected from scaling to be on the order of $\tilde{\alpha} = -0.2$, considerably larger than the $\alpha_c = -0.5$ expected at the FM-PM transition. Indeed, no evidence for a specific-heat peak has been detected at the latter transition. Measurements of the specific heat at the FM-SG transition are in progress.

Phase diagrams similar to Fig. 4 have been predicted by several models. In the Kirkpatrick-Sherrington model,² the phase diagram is calculated as a function of the ratio \tilde{J}_0/\tilde{J} , where \tilde{J}_0 and \tilde{J} are, respectively, the first and second cumulant averages of the random exchange interaction. From our ratios of T_c to T_{fg} we estimate that \tilde{J}_0/\tilde{J} ranges from 1.1 ($x = 0.32$) to 1.2 ($x = 0.2$). A similar phase diagram has been presented in the (equivalent) mean-field model of Chen and Lubensky⁴ and in the real-space renormalization calculation of Wortis, Jayaprakash, and Riedel.³ None of these models, however, gives us an estimate of the critical exponents, beyond mean-field theory, to be found in a 3D spin-glass-to-ferromagnetic

transition.

Although careful analysis of the Sherrington-Kirkpatrick model and others based on the Edwards-Anderson spin-glass order parameter seems to rule out their applicability in fewer than four dimensions,⁴ the 3D metallic glasses we have studied conform quite closely to the general behavior expected from these simple mean-field models. As we have demonstrated here, the ferromagnetic phase of these materials is unstable with respect to a new phase with all of the attributes of a spin-glass, and the transition between the two phases is continuous. This presents a challenge of understanding this spin-glass-like phase which may have little resemblance to the Edwards-Anderson model. We hope this additional evidence for the vanishing of the spontaneous moment will spur the effort.

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¹¹The alloy concentrations were checked by electron microprobe technique.

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¹³This differs from the phase diagram in Ref. 10 where the FM-SG line was arbitrarily chosen parallel to the temperature axis. This choice reflected our inability to accurately locate the transition from the ac data alone.

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