Anomalous critical behavior in amorphous Fe-Mo alloys: Evidence for random anisotropy effects

L. S. Barton and M. B. Salamon
Department of Physics and Materials Research Laboratory, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801
(Received 30 November 1981)

Anomalous low-field critical behavior was found in amorphous Fe-Mo alloys. The deviations from normal ferromagnetic behavior are exactly those recently predicted to arise from random anisotropy in an otherwise isotropic material. In disagreement with that model, however, the low-temperature susceptibility is finite. The difference in the spin-orbit coupling constants of Fe and Mo is suggested as a source of the anisotropy.

The properties of many random ferromagnetic materials have been closely examined in recent years and found to display critical behavior very similar to corresponding ordered samples.[1] This was not surprising, since calculations on the $d=3$ Heisenberg model suggested that randomness does not significantly change critical behavior.[2] However, in the presence of random anisotropy or random exchange that is off diagonal in the spin components, theoretical arguments indicate that the ferromagnetic state is not stable for a rotationally invariant system of $d<4$ dimensions.[3] The addition of nonrandom anisotropy can restore the ferromagnetic ground state, but modifies the nature of the phase transition.[4] Recently, Aharony and Pytte[5] (AP) calculated the equation of state for a ferromagnet (uniform exchange model) in the presence of random uniaxial anisotropy. They found an additional term in the equation of state which drives the spontaneous magnetizaton to zero, but leaves the susceptibility infinite for all $T \leqslant T_c$.

In this Communication, we report the first observation of AP-like critical behavior, in the amorphous alloys $(Fe_{1-c}Mo_c)_{75}P_{16}B_6Al_3$. The addition of Mo to amorphous Fe alloys strongly reduces the transition temperature and greatly modifies the critical behavior.[6] The features we report are as follows: (i) Breakdown of ferromagnetic critical behavior: curvature in modified Arrott plots, low-field deviations from $M \propto H^{1/\delta}$ along the critical isotherm, and deviations from scaling; (ii) correction of behavior described in (i) by the addition of the AP term in the equation of state; (iii) absence of kink-point phenomena; and (iv) demagnetization-limited susceptibility at, and somewhat below, T_c. We will argue that strong, random off-diagonal exchange interactions could arise from the difference in spin-orbit coupling constants of Fe and Mo, as calculated recently by Fert and Levy.[7]

Two samples of $(Fe_{1-c}Mo_c)_{75}P_{16}B_6Al_3$ were used,[8] having $c=0.15$ and 0.20. Ribbons of the alloys, prepared by centrifugal spin quenching, were cut to the approximate dimensions $3 \times 0.5 \times 0.025$ mm^3. Approximately 10 such ribbons were stacked in the sample holder of a vibrating sample magnetometer. Sufficient rotational data were taken to permit an accurate empirical determination of the demagnetizing factor of the composite sample, which was oriented to minimize demagnetizing effects. The magnetization $M(H,T)$ was measured in applied fields ranging from 20 Oe to 5 kOe ($c=0.15$) or to 600 Oe ($c=0.20$). As seen in Fig. 1, data for applied fields greater than 200 Oe appear ferromagneticlike. Isochamps at low field, however, show a maxima in $M(H,T)$ just below the transition temperature. Such maxima could be caused by Hopkinson effect[9]: the pinning of normal ferromagnetic domain walls. Very little difference was found between zero-field-cooled and field-cooled magnetization, especially in the region of the maxima, which rules out this explanation. A second transition to a spin-glass state, such as observed in the reentrant regime of the Fe-Mn or Fe-

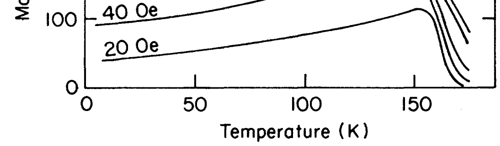

FIG. 1. Temperature dependence of the magnetization of $(Fe_{0.80}Mo_{0.20})_{75}P_{16}B_6Al_3$ at different applied fields. For $T>130$ K data are in 0.3-K increments for each of the nine applied fields. Individual data points not shown, for clarity.

Cr amorphous alloys,[10] would also yield maxima in the low-field $M(T)$ data. However, neither evidence for a second, low-temperature transition, nor the strongly time-dependent effects typical of spin-glasses were found.

To analyze the data, the usual scaling form[11] of the ferromagnetic equation of state was employed; that is, $y = m/h^{1/\delta}$ was plotted versus $x = t/h^{1/\beta\delta}$, for $t = (T - T_c)/T_c$, h being the dimensionless internal field, and m the dimensionless magnetization. For the correct choice of T_c, the demagnetizing factor, and the exponents β and δ, all of the data should collapse onto a single function $y = f(x)$. We are unable to find a set of parameter for which this holds. The "best" data collapsing for the $c = 0.20$ alloys was as found for $\beta = 0.41$ and $\delta = 4.8$ and is shown in Fig. 2. Also, regardless of the choice of T_c or the demagnetizing factor, h/m^δ on the critical isotherm depends on field.

Low-field deviations from ferromagnetic critical behavior have been calculated by Aharony and Pytte[5] for a model containing weak random uniaxial anisotropy of strength D. They predict that the equation of state for a $d = 3$ system will take the form

$$(h/m)^{1/\gamma} = t + m^{1/\beta} + Bm^2(m/h)^{1/2} \ , \tag{1}$$

where the AP parameter $B = 2a_A(D/J)^2/15$ for a Heisenberg system, J is the exchange energy, and a_A is a constant of order unity. We have extended, arbitrarily, the AP equation of state to include non-mean-field exponents, equivalent to the spherical model when $B = 0$.[11]

Along the critical isotherm ($t = 0$), the scaling law $\gamma = \beta(\delta - 1)$ was used to rewrite (1) as

$$(h/m^\delta)^{1/\gamma} = 1 + Bm^{2-1/\beta}(m/h)^{1/2} \ . \tag{2}$$

In Fig. 3, we test this prediction by plotting the left-hand side of (2) versus the term proportional to B. The low-field data (larger abscissa) satisfy (2), and yield the values for B given in Table I.

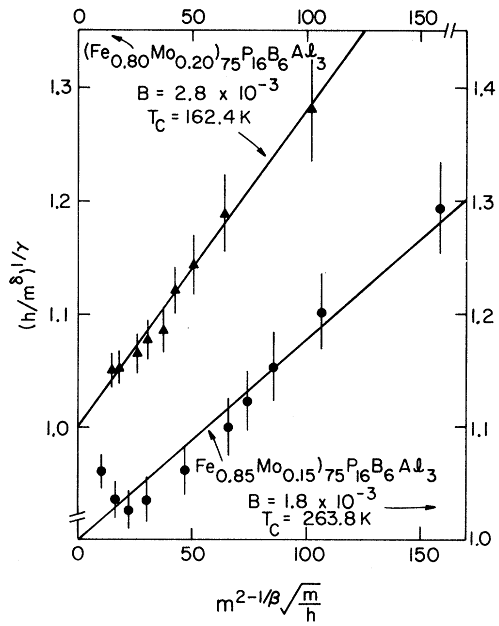

FIG. 3. Aharony-Pytte-like behavior on the critical isotherm. The slope equals the parameter B, in dimensionless units.

A more stringent test of AP-like critical behavior requires that (1) be cast into a scaling form. We find that the added term can be included in the scaled temperature, giving a new variable $\tilde{x} = x + By^{5/2}h^{-\phi/\beta\delta}$. The crossover exponent ϕ is given by $\phi = (2 + \beta\delta - 5\beta)/2$. Thus, y is plotted versus $\tilde{x}$, which reproduces Fig. 1 for $B = 0$. In Fig. 3, over 400 data points in the critical regime ($|t| < 0.05$) for each of the two samples are plotted in this way.

The exponents β and δ were varied in increments of $\pm 4\%$ over a range of 25%, T_c was varied in 0.1-K increments over a ± 1-K range, and for each set of parameters the AP parameter B was increased from zero until the best scaling behavior was observed. The best scaling fit was then chosen from the matrix of possible choices, giving $\beta = 0.41 \pm 0.02$ and

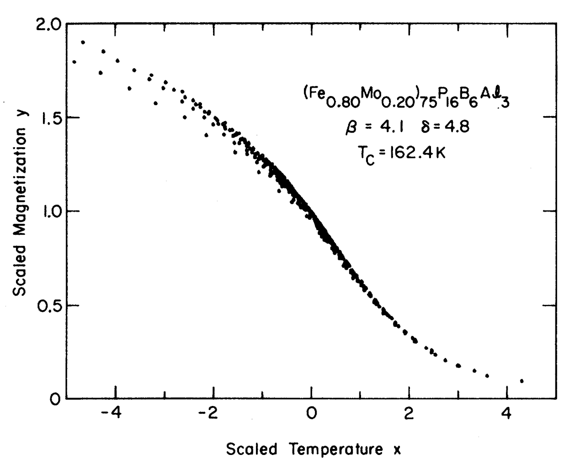

FIG. 2. Scaled magnetization $y = m/h^{1/\delta}$ vs scaled temperature $x = t/h^{1/\beta\delta}$ in dimensionless units.

TABLE I. Aharony-Pytte parameter $B = 2a_A(D/J)^2/15$.

c (Conc.)	B Critical isotherm	B Scaling	B/a_A Calc.
0.15	1.8×10^{-3}	1.6×10^{-3}	0.8×10^{-3}
0.20	2.8×10^{-3}	2.8×10^{-3}	1.2×10^{-3}

$\delta = 4.8 \pm 0.1$ (see Fig. 4). The critical temperatures are 162.4 ± 0.1 K ($c = 0.20$) and 263.8 ± 0.2 K ($c = 0.15$). The AP parameter B is accurate to $\sim 10\%$ for the exponents used; but is extremely sensitive to small changes in the exponents.

We did not observe a kink point in low-field $M(T)$ data, indicating the absence of spontaneous magnetization.[12] Experimental sensitivity limited our search to applied fields $\geqslant 5$ Oe. A search for kink-point effects at still lower fields is planned.

The dc susceptibility, determined from the intercept of modified Arrott plots [$m^{1/\beta}$ vs $(h/m)^{1/\gamma}$], revealed that the susceptibility χ saturates at the inverse demagnetizing factor for $T_c > T \gtrsim T_c - 10$ K, and decreases at lower temperatures. The susceptibility reaches a finite, nonzero value at 4.2 K. Although AP predict $\chi = \infty$ for $T \leqslant T_c$, convincing counterarguments exist[3,4] which require χ to remain finite at low temperatures, due to the spontaneous formation of domainlike clusters driven by statistical variations of the random anisotropy fields.

The origin of sufficiently large random anisotropies, D, cannot be explained by dipolar or pseudodipolar interactions alone. Recently, Fert and Levy[7] have extended the Ruderman-Kittel-Kasuya-Yosida (RKKY) calculation to include spin-orbit scattering of conduction electrons by nonmagnetic transition-metal ions and found this to induce a surprisingly large off-diagonal random exchange interaction of the Dzyaloshinsky-Moriya type. Although their calculation is not intended for concentrated magnetic systems, we have used its formalism to estimate the magnitude of spin-orbit-induced anisotropy. Within this framework, the ratio of the anisotropic to isotropic exchange energy is given by

$$\frac{D}{J_{\mathrm{RKKY}}} = \frac{15\varphi_d}{2E_F}\sin^2\left(\frac{\pi Z_d}{10}\right) \tag{3}$$

in which φ_d and Z_d are the spin-orbit coupling constant and number of d electrons on the atom providing the additional spin-orbit scattering (Mo). We assumed a simple cluster of atoms, using nearest neighbors only, typical Fermi energies, φ_d's appropriate to singly ionized atoms, and Z_d typical of the bcc metals. With these parameters, values of D/J_{RKKY} calculated from (3) are within a factor of 2 of the D/J observed experimentally, and an order of magnitude larger than pseudodipolar interactions could provide (see Table I).

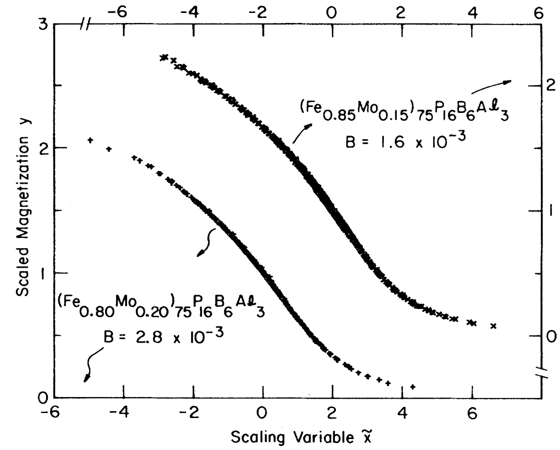

FIG. 4. Modified scaling: $y = m/h^{1/\delta}$ vs $\tilde{x} = t/h^{1/\beta\delta} + By^{5/2}h^{-\phi/\beta\delta}$, using $\beta = 0.41$ and $\delta = 4.8$.

We have demonstrated that the anomalous low-field critical behavior of amorphous Fe-Mo can be explained in the context of Aharony and Pytte's theory of a nonferromagnetic transition in materials containing random anisotropy. The amorphous nature of these samples greatly reduces the nonrandom anisotropy which would, otherwise, stabilize the ferromagnetic state. Inclusion of the AP correction to the ferromagnetic equation of state greatly enhances the scaling behavior of data in the critical regime. The origin of the anisotropy is consistent with calculated local anisotropies induced by nonmagnetic impurities with large spin-orbit coupling. Future work on amorphous Fe alloys with other nonmagnetic heavy-metal diluents is planned.

This work was supported in part by the National Science Foundation under Grant No. DMR-81-06143 with facility support of the Illinois Materials Research Laboratory under Grant No. DMR-80-20250.

[1]T. Mizoguchi, N. Ueda, K. Yamauchi, and H. Miyajima, J. Phys. Soc. Jpn. 34, 1691 (1973); K. Yamada, Y. Ishikawa, Y. Endoh, and T. Masumoto, Solid State Commun. 16, 1335 (1975); R. Malmhall, G. Backstrom, K. V. Rao, S. M. Bhagat, M. Meichle, and M. B. Salamon, J. Appl. Phys. 49, 1727 (1978).

[2]A. B. Harris, J. Phys. C 7, 1671 (1974); G. Grinstein and A. Luther, Phys. Rev. B 13, 1329 (1976).

[3]Y. Imry and S.-K. Ma, Phys. Rev. Lett. 35, 1399 (1975).

[4]A. Aharony, Phys. Rev. B 12, 1038 (1975); R. A. Pelcovits, E. Pytte, and J. Rudnick, Phys. Rev. Lett. 40, 476 (1978).

[5]A. Aharony and E. Pytte, Phys. Rev. Lett. 45, 1583 (1980).

[6]C. L. Chien and H. S. Chen, J. Appl. Phys. 50, 1574 (1979).

[7]P. M. Levy and A. Fert, Phys. Rev. B 23, 4667 (1981); A. Fert and P. M. Levy, Phys. Rev. Lett. 44, 1538 (1980).

[8]We are grateful to H. S. Chen, Bell Telephone Labora-

tories, for providing the samples.

[9]S. Chikazumi, *Physics of Magnetism* (Wiley, New York, 1964), p. 247.
[10]Y. Yeshurun, M. B. Salamon, K. V. Rao, and H. S. Chen, Phys. Rev. Lett. 45, 1366 (1980); Y. Yeshurun, K. V. Rao, M. B. Salamon, and H. S. Chen, Solid State Commun. 38, 371 (1981).
[11]P. Pfeuty and G. Toulouse, *Introduction to the Renormalization Group and Critical Phenomena* (Wiley, New York, 1977), p. 53.
[12]M. Rayl and P. J. Wojtowicz, Phys. Lett. 28A, 142 (1968).