Critical behavior of an amorphous ferromagnet

P. Gaunt, S. C. Ho, and Gwyn Williams
Department of Physics, University of Manitoba, Winnipeg, Canada R3T 2N2

R. W. Cochrane
Department of Physics, University of Montreal, Montreal, Canada H3C 3J7

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In two separate experiments, the static magnetization and ac susceptibility of METGLAS® 2826A have been measured in the vicinity of the critical region, $2\times10^{-3}\leq t\leq 2\times10^{-1}$, in applied fields up to 1 kOe. In the latter experiment, the critical behavior gives rise to peaks whose variation with magnetic field is shown to be governed uniquely by the index δ. These two experiments, yield respectively, the following values for the critical indices: $\beta=0.42\pm0.04$, $\gamma=1.35\pm0.03$ with $\delta=4.3\pm0.2$; and $\gamma=1.49\pm0.06$ with $\delta=4.3\pm0.1$. Within experimental error they satisfy the scaling law relationship $\gamma=\beta(\delta-1)$ and are in good agreement with values deduced for other amorphous ferromagnets. They are also remarkably close to the values for the crystalline ferromagnetic elements Ni and Fe. The one unusual feature of these data concerns the effective value for γ beyond the critical regime; in METGLAS® 2826A, and other amorphous ferromagnets, $\gamma^*(T)$ initially *increases* with increasing temperature above the transition, whereas in Ni it decreases continuously towards the high-temperature mean-field value of 1.

INTRODUCTION

Amorphous metallic alloys exhibit a wide spectrum of magnetic behavior ranging from diamagnetism through ferromagnetism including random or spin-glass freezing into noncoherent magnetic phases. It is worth remarking, however, that many noncrystalline alloys (particularly $3d$ metal-metalloid systems) are magnetically very soft, indicating that their magnetic moments are at most only weakly constrained by the local environment. While the effects of random atomic packing on the transition to ferromagnetism has yet to be examined theoretically, experimentally many amorphous alloys are observed to undergo sharp transitions into their ordered state. Consequently, there has been considerable recent effort directed at describing the critical behavior of such amorphous ferromagnets[1–3] in terms of the usual critical indices β, γ, and δ, defined by[4]

$$M(0,T)\propto t^{\beta},\quad T<T_c\ , \tag{1}$$

$$\chi(0,T)\propto t^{-\gamma},\quad T>T_c\ , \tag{2}$$

$$M(H,T_c)\propto H^{1/\delta},\quad T=T_c\ , \tag{3}$$

where M, H, and χ represent the magnetization, internal field, and susceptibility, respectively, while $t=|T-T_c|/T_c$. In the three systems $Co_{70}B_{20}P_{10}$,[1] $Fe_{80}P_{13}C_7$,[2] and $Gd_{80}Au_{20}$,[3] for which extensive analyses have been carried out in the critical region ($t\ll 1$), the critical indices in each case have been found to be similar to those determined for crystalline Ni (Refs. 5–7) and Fe (Ref. 8).

In the course of examining the magnetic and transport properties of several amorphous alloys,[9] we have followed the onset of ferromagnetism in the alloy $Fe_{32}Ni_{36}Cr_{14}P_{12}B_6$ (METGLAS® 2826A, manufactured by the Allied Chemical Corporation, New Jersey). In this paper, we report both dc magnetization and ac susceptibility measurements on this system in the region $2\times10^{-3}\leq t\leq 2\times10^{-1}$ for $H\leq$ kOe. In addition, we show that the field dependence of the critical susceptibility peak is determined uniquely by the index δ. The two sets of experimental data were acquired and analyzed separately, but within the experimental uncertainties they yield identical values for the critical indices which correspond to those of crystalline Ni and the amorphous alloys listed above. One unexpected feature of our data is the unusual manner in which γ approaches the critical region. Indeed, the significantly higher values of γ and δ reported by Figueroa *et al.*,[10] which were derived from data acquired well away from the transition region, correctly indicate the variation in χ—and hence in γ—as T increases above T_c.

EXPERIMENTAL DETAILS

Magnetization

The field and temperature dependence of the static magnetization was measured by means of a vibrating sample magnetometer. The latter operates near 37 Hz with the sample situated in the axial field of a solenoid which was swept through zero in the range $|H| \leq 700$ Oe using a Hewlett-Packard 6824A bipolar power supply–amplifier fed from a slow ramp generator. Temperatures were monitored with a calibrated GaAs diode mounted directly onto the sample holder; the estimated temperature uncertainty is $\pm 1\%$. The sample used in the magnetization study weighed approximately 10 mg, with dimensions $50 \times 2 \times 0.05$ mm^3.

ac susceptibility

The real part of the ac susceptibility was monitored in a modified linear phase-lock magnetometer[11] operating at 1.8 kHz with a driving field amplitude of 0.4 Oe rms. The detection coils consisted of two liquid-nitrogen-cooled, balanced coils connected in series opposition. dc biasing fields up to 1 kOe were produced by a liquid-nitrogen-cooled copper solenoid wound coaxially with the detecting system. The susceptibility sample consisted of three strips of METGLAS® 2826A, each $25 \times 3.5 \times 0.05$ mm^3, secured in a sandwich form with adjacent faces electrically insulated from each other with masking tape. This sample was suspended in a bundle of fine copper wires in the tail section of a glass Dewar system; the copper wires supporting the sample were about 15 cm long and were soldered at their upper ends into a cone-shaped copper block. A heater wound onto this block was used to provide warming rates of about 0.2 K/min between refrigerant fixed points. Sample temperatures were measured using a Au-0.03 at.% Fe vs chromel thermocouple; while the absolute accuracy of this thermocouple is $\pm 1\%$, relative temperatures can be determined with far greater precision.

RESULTS AND ANALYSIS

Magnetization data

Figure 1 shows the low-field-magnetization curves for METGLAS® 2826A in the critical region: Above the ordering temperature they are linear, become more concave as T_c is approached, and show a sharply stepped behavior below the transition temperature. On the scale of these measurements there is practically no hysteresis ($H_c \leq 1.0$ Oe) and only a slight

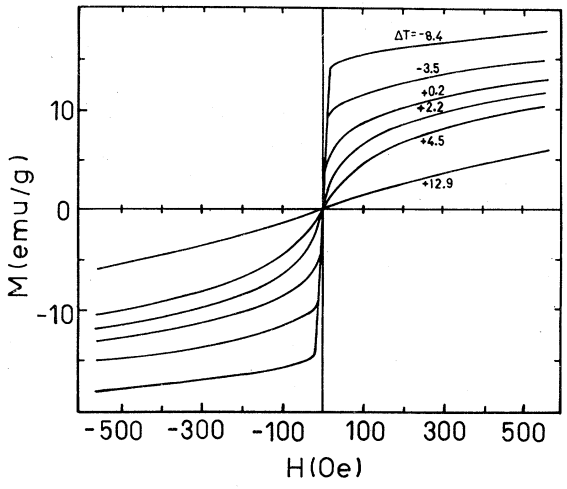

FIG. 1. Magnetization curves at various temperatures, $\Delta T = T - T_c$, throughout the critical region.

skewing of the magnetization curves due to the small demagnetizing field within the sample. Even without detailed analysis, the ordering temperature can be located within two degrees via the onset of remanence in the ordered regime.

The detailed analysis of these data follows the procedure outlined by Kouvel and Fisher,[6] and requires the processing of these data in two steps. The first is to obtain zero field values for the susceptibility above T_c and the spontaneous magnetization below T_c by extrapolating the thermodynamically based Arrott plot of $M^2(H,T)$ vs $H/M(H,T)$ to its intercept on the appropriate axis. H, as previously defined, refers to the internal field, viz., $H = H_{\text{ext}} - DM$, where D is the demagnetizing factor. From these quantities, we then calculate the logarithmic derivatives:

$$X(T) = \left| \frac{1}{\chi^{-1}} \frac{d\chi^{-1}}{dT} \right|^{-1} \tag{4}$$

and

$$Y(T) = \left| \frac{1}{M} \frac{dM}{dT} \right|^{-1} \tag{5}$$

which are displayed as a function of temperature in a combined plot [Fig. 2(a)]. The smooth extrapolation in these figures defines the ordering temperature T_c as $T_c = 249.0 \pm 0.1$ K and in the neighborhood of this temperature the critical indices are estimated as $\gamma = 1.35 \pm 0.03$, $\beta = 0.42 \pm 0.04$; the concave curvature of $X(T)$ towards the temperature axis results from a somewhat unexpected dependence of the susceptibility on temperature, which is presented by de-

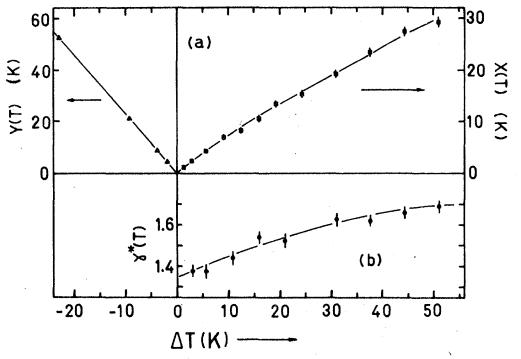

FIG. 2. (a) The parameters $X(T)$(K) and $Y(T)$(K) plotted against temperature T(K) in the vicinity of the ordering temperature. (b) The estimated value for the critical index $\gamma^*(T)$ as a function of temperature in the vicinity of the critical region.

fining the effective exponent:

$$\gamma^*(T) = \frac{T - T_c}{X(T)} \quad . \tag{6}$$

As is evident from Fig. 2(b), and in contrast to the situation existing in itinerant crystalline ferromagnets,[6] $\gamma^*(T)$ *increases* with increasing temperature in the region immediately above T_c, reaching a value of 1.7 near room temperature. This surprising variation accounts for the value of $\gamma = 1.7$ deduced by Figueroa *et al.*[10] using only data in the region $T > 290$ K. These latter data indicate, in addition, that beyond 400 K $\gamma^*(T)$ decreases towards the high-temperature value of 1 predicted by theory.

The third parameter, δ, has been estimated from the magnetization curve close to T_c (i.e., 249.0 K); curve a in Fig. 3 shows this isotherm on a log-log graph and yields a value of $\delta = 4.3 \pm 0.3$. However, in view of the sensitivity of this parameter to small temperature variations, as well as the uncertainties associated with the rounding of the transition due to sample inhomogeneities, this number must be viewed as an estimate only.

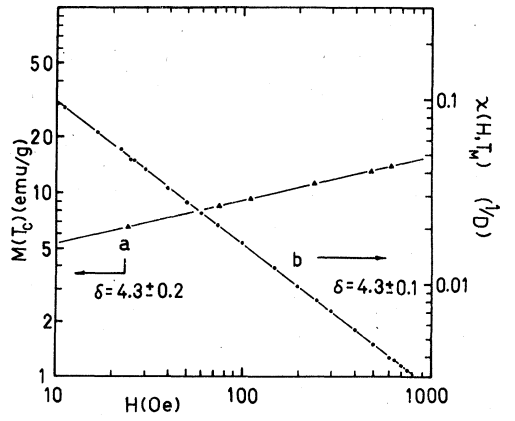

FIG. 3. Curve a the magnetization $M(H,T_c)$(emu/g) plotted against the internal field H(Oe) on a double logarithmic plot. Curve b the ac susceptibility $\chi(H,T_m)$ at the maxima (in units of the demagnetizing factor) plotted against the internal field H(Oe) on a double logarithmic plot.

ac susceptibility data

Above T_c, the ac susceptibility in zero field increases rapidly with decreasing temperature and below T_c saturates at a value determined by the demagnetizing factor.[12,13] Upon application of a small dc field the curve splits into two parts; the principle contribution is broadened and shifted rapidly towards lower temperatures, leaving behind a smaller but much sharper peak. Increasing the field displaces this secondary peak to *higher* temperatures and gradually smears it out, as indicated in Fig. 4. Similar behavior has been reported in other ferromagnetic alloy systems[14] by one of the present authors. Evidently, the principal peaks result from the details of the magnetization process (domains; hysteresis; nonuniform magnetostatic modes, etc.), whereas only the secondary maxima are controlled by the critical fluctuations. From these observations it is also concluded that the principal contributions to the zero-field curve close to the transition render it unreliable as a measure of the critical behavior and consequently it is necessary to focus on the secondary peaks to find the critical indices.

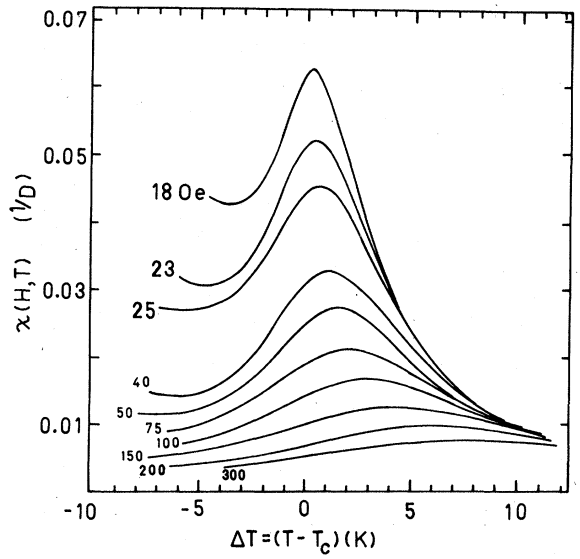

FIG. 4. The measured ac susceptibility $\chi(H,T)$ (in units of the demagnetizing factor) plotted against temperature (K) in the critical region. The number marked against each curve is the external field (Oe).

The outstanding characteristic of these secondary critical contributions is their sharp maximum moving to higher temperatures with increasing magnetic fields. In fact, the peak height is uniquely determined by the critical index, δ.[15] This follows directly from the scaling equation of state:

$$M = t^{\beta} F(H/t^{\beta\delta})$$

from which

$$\chi(H,t) = t^{\beta(1-\delta)} F'(H/t^{\beta\delta}) = H^{1/\delta-1} G(H/t^{\beta\delta}) \ .$$

The condition for maximum susceptibility as a function of temperature is given immediately by $G'(H/t_p^{\beta\delta}) = 0$, that is to say, that at the peak temperature, t_p, the function $G(H/t_p^{\beta\delta})$ is a constant independent of H. Hence $\chi(H,t_p) \propto H^{1/\delta-1}$. A log-log plot of $\chi(H,t_p)$ against H (after corrections for demagnetizing effects) is shown in Fig. 3(b) and gives $\delta = 4.3 \pm 0.1$. It should be emphasized that this value is *independent of the choice* T_c, in sharp contrast with the analysis of the magnetization data. That the two experiments gave the same value is an indication that T_c has been correctly estimated in the magnetization experiment.

An immediate corollary of the above theorem requires the peak temperature, T_p, to scale as $H^{1/\delta\beta}$. Indeed, when T_p is plotted against H, a continuous curvature is noted. As shown in Fig. 5, this curvature is removed on plotting T_p against $H^{1/\beta\delta}$ (using the value of β found from the magnetization analysis). (We have recently been able to demonstrate a similar dependence of T_p on H crystalline ferromagnets, viz., magnetically soft *Pd*Mn alloys.[16]) Extrapolating to the $H = 0$ axis gives $T_c = 256.9$ K. This value is somewhat higher than the value quoted earlier, a consequence, we believe of small concentration variations since this sample came from a different alloy batch than the magnetization sample. As

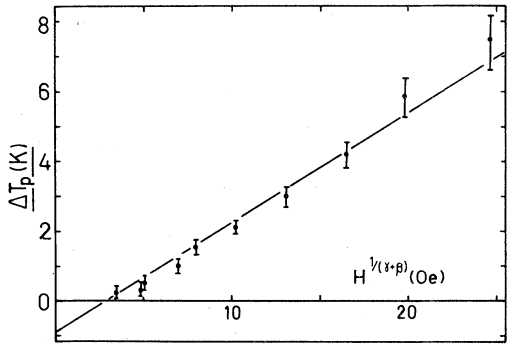

FIG. 5. Peak temperatures, T_p(K), as a function of $H^{1/\beta\delta}$. The zero on the temperature axis was arbitrarily taken at 257.7 K.

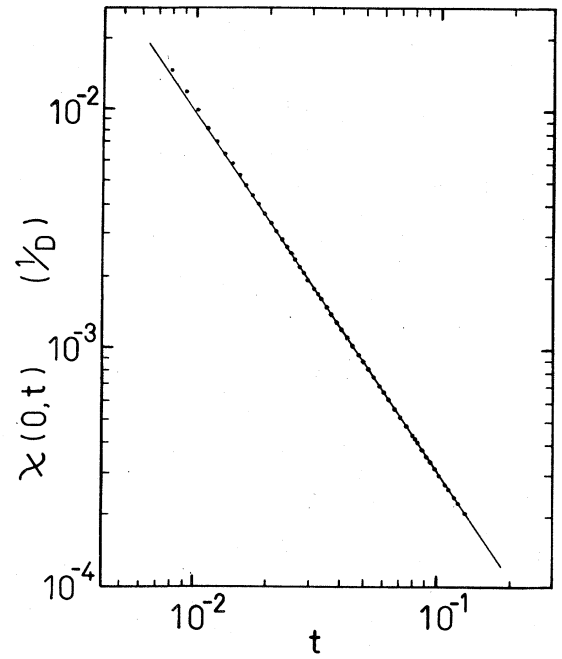

FIG. 6. The susceptibility $\chi(0,T)$ (in units of the demagnetizing factor) plotted against the reduced temperature t in the vicinity of the critical region, using $T = 256.9$ K.

for trying to determine β directly from such a plot, it is apparent that the peak positions cannot be defined well enough to produce an accurate measure, although an estimate would be possible. Since a value of β is readily available, it seems preferable to use it to fix T_c uniquely.

With T_c determined, it is now possible to extract the remaining index, γ, from the temperature variation of the zero-field susceptibility above T_c. Figure 6 shows a plot of $\chi(0,t)$ against t on a double logarithmic scale, and yields a well-defined value of $\gamma = 1.49 \pm 0.06$. This latter value falls within the range found in the magnetization experiment over the same temperature interval $2 < T - T_c < 25$ K.

CONCLUSION

In summary, we believe that the double approach to ferromagnetic critical behavior via the analysis of magnetization and low-field-susceptibility experiments provides a unique and unambiguous determination of the critical indices. These two experiments are, in fact, complementary, the first being best suited for values of γ, β, and T_c, whereas the second gives direct information about δ, T_c, and γ. We note that the secondary susceptibility peaks will only be observable in soft magnetic materials, but when

detected, the result is categoric—the peak temperatures give the index δ independently of T_c and the other critical parameters.

In particular, for the $Fe_{32}Ni_{36}Cr_{14}P_{12}B_6$ (METGLAS® 2826A) amorphous alloy, we find $\gamma = 1.35 \pm 0.05$, $\beta = 0.42 \pm 0.04$, and $\delta = 4.3 \pm 0.1$, which satisfy the scaling relation[4] $\gamma = \beta(\delta - 1)$. These values for the critical indices are close to those found in other amorphous ferromagnets, $Co_{70}B_{20}P_{10}$,[1] $Fe_{80}P_{13}C_7$,[2] and $Gd_{80}Au_{20}$,[3] as well as the crystalline ferromagnetic elements Ni (Refs. 5–7) and Fe (Ref. 8). In this respect, the present results provide another example of the universality of the scaling hypothesis.

One prominent difference between the amorphous and crystalline metals concerns the temperature dependence of the susceptibility beyond the critical region. Specifically, for crystalline Ni (Ref. 6) there is a continuous decrease in $\gamma^*(T)$ from 1.34 towards a high-temperature value of 1; for METGLAS® 2826A $\gamma^*(T)$ initially increases with temperature. Indeed, other recent data[10] on this system as well as all the other amorphous ferromagnets mentioned above exhibit this same feature. It is thus tempting to ascribe this observation as a characteristic of highly disordered systems. Physically, this dependence indicates that the susceptibility is decreasing with temperature at a faster rate than does a uniform magnet with moments of fixed magnitude. The observed variation in $\gamma^*(T)$ could result from a moment which decreased with increasing temperature, originating possibly from the thermally induced breakup of magnetic clusters. A temperature-dependence exchange interaction between fixed moments would also yield a similar behavior. In the absence of concrete theoretical guidance these suggestions remain speculations which we offer as incentives for detailed theoretical and model calculations.

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