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Anomalous magnetic behavior in the critical region of an amorphous magnetic alloy containing chromium

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A close scrutiny of the Mössbauer data previously taken on amorphous $Fe_{32}Ni_{36}Cr_{14}P_{12}B_6$ (Metglas 2826A) alloy and on the other Fe-Ni-P-B alloys containing similar transition-metal concentrations by different workers reveals that Fe atoms in Metglas 2826A have two types of environment; one in which Fe atoms have primarily Fe, Ni, and a very small amount of Cr as their neighbors and the other in which Fe atoms are surrounded by Cr atoms. In the present investigation, Curie temperatures for these so-called "FeNi" and "FeCr" magnetic phases are found to be 249 and 320± 10 K, respectively. The magnetic study performed on Metglas 2826A in the temperature range 220 to 300 K in fields up to 10 kOe gives the Curie temperature T_C and critical exponents β, γ, and δ as 250 ± 1 K, 0.43 ± 0.02, 1.33 ± 0.05, and 4.43 ± 0.17, respectively, for the "FeNi" phase. The significantly large error limits in the determination of these exponent values result from the presence of the "FeCr" phase. The above-mentioned values for the critical exponents are seen to follow very closely the predictions of a three-dimensional Heisenberg model and thereby suggest dominance of the short-range forces in the critical region (long-range forces, e.g., dipolar forces are shown to have negligible influence on the critical fluctuations of magnetization). The data satisfy the magnetic equation of state characteristic of a second-order phase transition over the entire temperature range. Furthermore, the observation that the magnetic order for the "FeCr" phase persists well above the Curie temperature of the "FeNi" phase is shown to provide a straightforward explanation for the anomalies observed in various properties of Metglas 2826A and permits us to conclude that it is not necessary to invoke superparamagnetism in this system.

I. INTRODUCTION

Considerable effort has been directed over the years to studying ferromagnetic alloys in the structurally amorphous state both from technological as well as fundamental points of view. Critical phenomena in such materials have received increased experimental[1–7] and theoretical[8–13] attention particularly during the last decade. Such an intense activity has been motivated primarily by basic questions such as: Do the amorphous magnetic systems have a well-defined critical point and if so, is it the manifestation of a second-order magnetic phase transition as evidenced in crystalline systems? What is the range of the exchange forces in such systems in the critical region? A large number of amorphous alloys have been prepared using various techniques.[14] Among the amorphous systems studied so far, $Co_{70}B_{20}P_{10}$, $Fe_{80}P_{13}C_7$, $Fe_{32}Ni_{36}Cr_{14}P_{12}B_6$ (Metglas 2826A), $Fe_{29}Ni_{49}P_{14}B_6Si_2$ (Metglas 2826B), and $Gd_{80}Au_{20}$, in particular, exhibit a sharp, well-defined magnetic phase transition with the reduced magnetization and field following an equation of state derived for second-order phase transition in fluid systems[12] and with the critical exponents satisfying an equality relation resulting from the static scaling law.[12, 15–17] The above mentioned alloys, with the exception of Metglas 2826A and 2826B, give values for the critical exponents in striking agreement with those derived for a three-dimensional Heisenberg ferromagnet.[18] Such a close agreement, suggestive of dominant short-range forces, between Heisenberg values and the experimental critical exponent values has been observed previously for ferromagnets in the crystalline state.[19–22] For Metglas 2826A and 2826B the observed exponent values are larger in magnitude than Heisenberg values. This discrepancy in the values has been attributed to superparamagnetism[4, 7] in these systems. Though the tendency in the disordered materials towards formation of localized clusters of strongly interacting moments is generally recognized,[14, 23] it is not clear as to why only Metglas 2826A and 2826B of all the amorphous systems should exhibit superparamagnetic behavior above Curie temperature.

Among the amorphous alloys stated above, only Metglas 2826A stands out as a unique system in itself because magnetic,[4] electrical-resistivity,[24, 25] specific-heat,[6] and Mössbauer[26] investigations on this alloy reveal anomalous behavior not observed in any other amorphous system. While magnetic and specific-heat results support the existence of short-range magnetic ordering or superparamagnetic behavior for temperatures above the Curie temperature in this system, Mössbauer data rule out both these possibilities. The above findings, therefore, point to the fact that no clear-cut explanation exists in the literature for the anomalies observed in various properties of Metglas 2826A and the contradiction between the conclusions drawn from different observations remains. An obvious way out of this situation is to examine the magnetic behavior of this alloy in the critical region much more thoroughly than done previously[4] and to scrutinize Mössbauer data more closely.

We report here the results of magnetization measurements performed on Metglas 2826A in the temperature range 220 to 300 K in fields up to 10 kOe. From the magnetization data we evaluate the spontaneous magnetization and initial susceptibility values at different temperatures in the critical region. This permits determination of the critical exponents and the Curie temperature. Contrary to the observations made in the previous work,[4] the present investigation yields critical exponent values which are in close agreement with the corresponding theoretical values obtained for a three-dimensional Heisenberg ferromagnet. Such a striking agreement between the experimental and theoretical values obviously suggests that the short-range forces dominate in the critical region. However, in conformity with the earlier data,[4] spontaneous magnetization is found to have a sizable value at the Curie temperature and the magnetization versus field curve taken at 295 K in fields up to 16 kOe exhibits a finite curvature.

A comparison of Chien's Mössbauer data[26] on Metglas 2826A with the Mössbauer data obtained for amorphous Fe-Ni-P-B alloys, containing similar transition-metal concentrations,[27–30] reveals that in this alloy Fe atoms have two types of environment; one in which Fe atoms have primarily Fe, Ni, and a very small amount of Cr as their neighbors and the other in which Fe atoms are surrounded mainly by Cr atoms. Chien's data, when scrutinized closely, further brings out the fact that the experimental values[26] of the most probable hyperfine field $(H_{\text{peak}})_1$ due to Fe atoms having the latter type of environment as a function of temperature do not follow the power law $\sim(1-T/T_C)^{1/3}$ with $T_C=249$ K given by Chien but instead their temperature dependence is closely described by a power law $\sim(1-T/T_C)^{1/2}$ with $T_C=320\pm10$ K. The observation that the magnetic order for this so-called "FeCr" phase extends about 70 K beyond the Curie temperature 250 ± 1 K of the "FeNi" phase is shown to provide adequate explanation for the anomalies found in various properties of this alloy. The possibility for this amorphous system to exhibit superparamagnetic behavior is ruled out on the basis of deductions made from the available Mössbauer data and the present magnetization results.

II. EXPERIMENTAL DETAILS

Two batches of the amorphous $Fe_{32}Ni_{36}Cr_{14}P_{12}B_6$ alloy (prepared by the splat-cooling technique and commerically known as Metglas 2826A) were procured from the Allied Chemical Company, New Jersey, in ribbons having 0.05 mm thickness and 1.5 mm and 1.8 mm widths. The magnetization measurements were performed on the samples taken from both the batches using a Faraday balance. In order to achieve sufficient accuracy even in the low-field (15 Oe) data (taken over the temperature range 220 to 300 K), three pieces belonging to the same batch of the alloy ribbon (each 10 mm in length) with a total weight of 11.4 mg were placed side by side in a vertical slot cut in a 5-mm-diameter Teflon rod which, in turn, was contained in a cylindrical copper sample holder. The magnetic field was applied in the plane of the ribbon pieces parallel to their breadth. The demagnetizing factor was estimated to be 0.012. Magnetization as a function of temperature was measured at constant external fields $\geqslant 50$ Oe in the temperature interval $223 \leqslant T \leqslant 283$ K with temperature increasing from 220 K at a rate of 0.5 K/min. The sample temperature was controlled by means of a resistance heater and a small pressure of helium exchange gas in a continuous-flow cryostat. The voltage across a standard platinum resistance (carrying a constant current of 100 μA), which monitors the sample temperature and the Faraday balance output, which gives the corresponding force change proportional to magnetization at a particular fixed external field, were continuously recorded on a XY recorder. With the help of a digital readout of the platinum resistance thermometer output, the force change for every 0.5 K was also independently noted. In addition, the magnetization was measured as a function of the external magnetic field at 4.2 and 295 K in fields up to 16 kOe. The Curie temperature determined from the low-field magnetization data was found to be different for the samples taken from two different batches, e.g., 250 K for 1.5-mm-wide ribbons and 254 K for 1.8-mm-wide ribbons. These Curie temperature values are in excellent agreement with those observed by other workers.[4,24–26] Except for the difference in the T_C values the samples taken from both the batches yield the same values for the critical exponents within the error limits given later in the text. In the following section we, therefore, present only the results obtained on the samples with $T_C=250$ K.

III. EXPERIMENTAL RESULTS AND DISCUSSION

The experimental results of the temperature dependence of magnetization for about 25 K on either side of Curie temperature T_C in various constant external

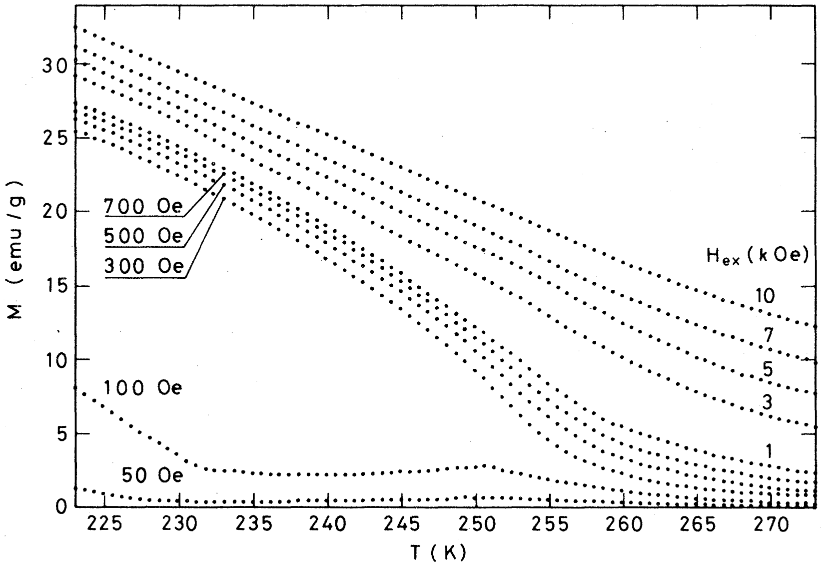

FIG. 1. The temperature dependence of magnetization in constant external magnetic fields (50 Oe $\leq H \leq$ 10 kOe).

magnetic fields in excess of 50 Oe are shown in Fig. 1. The T_C was found to be 250 K from the sharp kink observed in the magnetization versus temperature curve taken at 15 Oe (not shown in Fig. 1) in the temperature range 220 to 300 K. The kink point shifts to lower temperatures for external fields greater than 50 Oe, as is usually observed in ferromagnetics either in crystalline or in amorphous state.

A. Critical exponents and the magnetic equation of state

It is well known[12] that the second-order phase transition around T_C is characterized by a set of critical exponents β, γ, and δ, defined as

$$\lim_{H\to 0} M \equiv M_s = A(T_C - T)^\beta, \quad T < T_C \ , \tag{1}$$

$$\lim_{H\to 0} (H/M) \equiv \chi_0^{-1} = B(T - T_C)^\gamma, \quad T > T_C \ , \tag{2}$$

and

$$M = CH^{1/\delta}, \quad T = T_C \ , \tag{3}$$

where M_s and χ_0 are spontaneous magnetization and initial susceptibility, respectively, and A, B, C are proportionality constants, with the static scaling law[12,15–17] giving the following relation between them

$$\delta = 1 + (\gamma/\beta) \tag{4}$$

and a magnetic equation of state given by

$$M_\pm/|T - T_C|^\beta = f_\pm(H/|T - T_C|^{\beta\delta}) \ , \tag{5}$$

where the plus and minus signs denote temperatures above and below T_C, respectively. Equation (5) implies that the reduced magnetization $m \equiv M/|T - T_C|^\beta$ as a function of the reduced field $h \equiv H/|T - T_C|^{\beta\delta}$ falls on two different universal curves: $f_-(h)$ for temperatures below T_C and $f_+(h)$ for temperatures above T_C.

Equations (1) and (2) demonstrate that the computation of the values for the critical exponents β and γ needs evaluation of M_s and χ_0^{-1} as functions of temperature, respectively. The values of M_s and χ_0 at every 0.5 K interval below and above Curie temperature were obtained by extrapolating the high-field portion of the isothermal magnetization curves taken at various temperatures below T_C to $H = 0$ and by using the low-field (15 Oe) magnetization data, respectively. At this stage it should be mentioned that all the above values were deduced only after making proper corrections for the demagnetizing field and the alternative method using Arrott plots suggested by Poon and Durand[5] to obtain M_s and χ_0 values could not be used for the present alloy since M_s remains finite even for temperatures far above T_C and consequently, large errors result not only in the determination of T_C but also in that of M_s and χ_0. The values for M_s and χ_0^{-1} computed at different temperatures using the above-mentioned method are shown in Figs. 2 and 3, respectively. Equations (1) and (2) can be rewritten in the form

$$\left[\frac{d(\ln M_s)}{dT}\right]^{-1} = M_s \left[\frac{dM_s}{dT}\right]^{-1} = (T - T_C)/\beta \tag{6}$$

and

$$\left[\frac{d \ln \chi_0^{-1}}{dT}\right]^{-1} = \chi_0^{-1} \left[\frac{d\chi_0^{-1}}{dT}\right]^{-1} = (T - T_C)/\gamma \ . \tag{7}$$

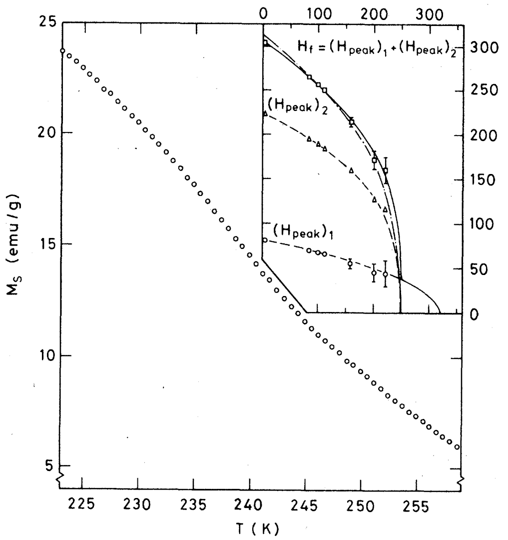

FIG. 2. Spontaneous magnetization M_s as a function of temperature. Insert shows the experimental data of Chien for the temperature dependence of the most probable hyperfine fields $(H_{\text{peak}})_1$ and $(H_{\text{peak}})_2$ and their sum H_f; the dashed curves through the data points are obtained from the theoretical values computed using Eqs. (9) and (10) and the solid curve represents the sum of the corresponding theoretical $(H_{\text{peak}})_1$ and $(H_{\text{peak}})_2$ values at a particular temperature. The dash-dot curve represents the power law $\sim(1-T/T_C)^{0.4}$ (see text).

Equations (6) and (7) evidently show that the plots of $[d(\ln M_s)/dT]^{-1}$ versus T and $[d(\ln\chi_0^{-1})/dT]^{-1}$ versus T should be straight lines with slopes $(1/\beta)$ and $(1/\gamma)$, respectively, and such straight lines when extrapolated to ordinate equal to zero should yield intercepts on their T axes equal to the Curie temperature. According to Kouvel and Fisher,[21] such plots, shown in Figs. 4 and 5, give precise values of T_C, β, and γ. However, certain sources of error deserve proper attention. First, small variations are possible while drawing a smooth curve through M_s or χ_0^{-1} data points. Second, the temperature T at which the local derivative dM_s/dT or $d\chi_0^{-1}/dT$ is defined is uncertain to within ±0.05 K. Also, due consideration should be given to the accuracy limit ±0.05 K of the temperature measurement. All these combined together give two-dimensional uncertainity limits which are included in the size of the closed circled points in Figs. 4 and 5. The straight lines obtained from a least-squares fit through the centers of the data points in Figs. 4 and 5 give values of $\beta=0.43$, $T_C=249$ K and $\gamma=1.33$, $T_C=252$ K, respectively. In order to obtain the limits of uncertainity in these values, the least-squares fits through the edges of the data points were made to yield $\beta=0.43\pm0.02$, $T_C=249\pm0.3$ K and $\gamma=1.33\pm0.05$, $T_C=252\pm0.3$ K. Large deviations

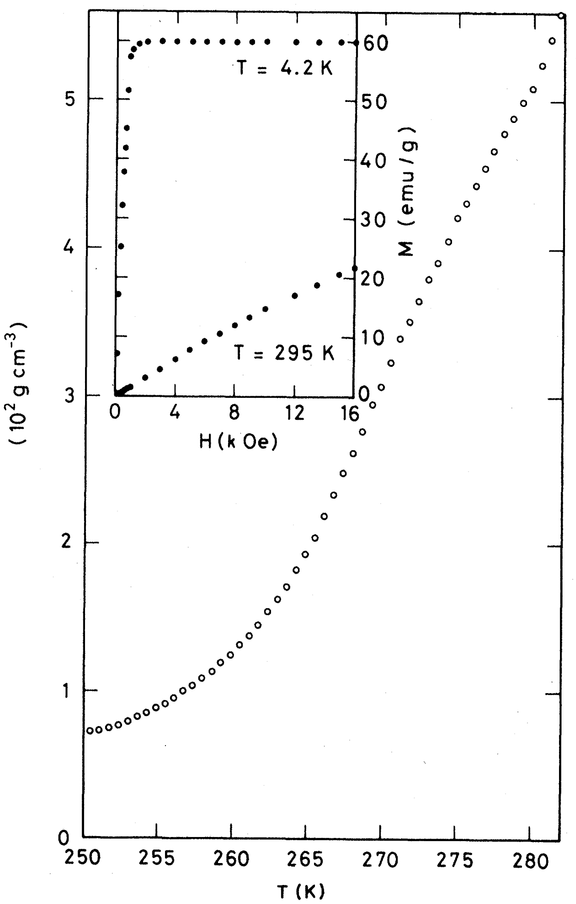

FIG. 3. Temperature dependence of the inverse initial susceptibility χ_0^{-1}. Insert shows the magnetization versus field curves taken at 4.2 and 295 K in fields up to 16 kOe.

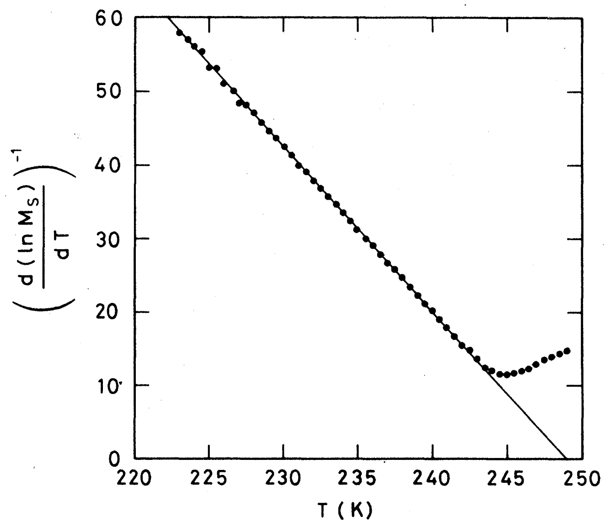

FIG. 4. Inverse of $d(\ln M_s)/dT$ versus temperature plot gives the critical exponent β and Curie temperature (see text).

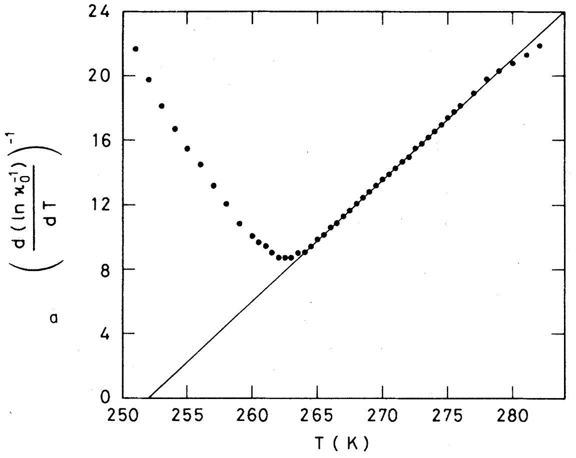

FIG. 5. Inverse of $d(\ln\chi_0^{-1})/dT$ versus temperature plot gives the critical exponent γ and Curie temperature (see text).

of the data points from the linear fits apparent in Fig. 4 above 244 K and in Fig. 5 below 263 K result from the fact that above 244 K M_s varies with temperature much slower than $(T_C - T)^{0.43}$ power law and tails off slowly beyond T_C (Fig. 2) to a finite value even at 295 K as evidenced by a slight curvature in M vs H curve at 295 K (insert in Fig. 3) and also from the fact that a straight-line fit to χ_0^{-1} vs T curve (Fig. 3) can be obtained only in the temperature range 263 to 280 K. Peculiarities such as the persistence of spontaneous magnetization for temperatures well above 250 K and the absence of a proper divergence in the susceptibility versus temperature curve at T_C are shown in the later text to be caused by the presence of the so-called "FeCr" phase for which the magnetic order extends up to a temperature (Curie temperature) as high as ~ 320 K. The M_s value at 4.2 K, evaluated from the magnetization versus field plot (insert in Fig. 3), is found to deviate only by about 2% from the $(T_C - T)^{0.43}$ power law. An abrupt change in the slope of χ_0^{-1} vs T curve at ~ 280 K clearly seen in Fig. 3 is also manifested in Fig. 5. In the temperature range 285 to 300 K, a double-logarithmic plot of χ_0^{-1} vs T gives the slope value as 1.68, which is in close agreement with the corresponding value obtained by Figueroa *et al.*[4] in the temperature range 290 to 350 K.

In Fig. 6, ln M is plotted against ln H for the T_C values obtained from Figs. 4 and 5, i.e., 249 and 252 K and also for their mean temperature value 250.5 K. It is evident from Fig. 6 that Eq. (3) is valid for fields in excess of 300 Oe (lower the field value than 300 Oe, more the magnetization values deviate from these straight-line plots) with $\delta = 4.6$, 4.35, and 4.1 for 249, 250.5, and 252 K, respectively. Validity of Eq. (3) in a relatively large temperature range suggests the following temperature dependence for

$$\delta = 4.35 + 0.17(250.5 - T) \quad . \tag{8}$$

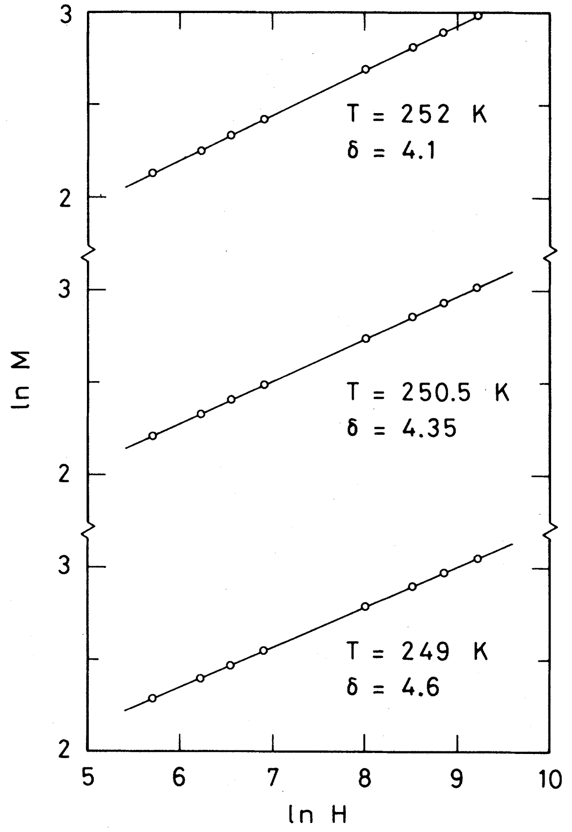

FIG. 6. ln M versus ln H curves for 249, 250.5, and 252 K.

Similar relation between δ and T was also observed by Figueroa *et al.*[4] However, in view of the fact that the value of T_C determined from the low-field magnetization data is found to be 250 K with a maximum possible error of ± 1 K, the value of δ without any loss of accuracy comes out to be 4.43 ± 0.17. Substitution of the values $\beta = 0.43 \pm 0.02$ and $\gamma = 1.33 \pm 0.05$ in Eq. (4) yields $\delta = 4.10 \pm 0.28$, which is in reasonable agreement with its presently determined value. The value of the specific-heat exponent α computed using the relation $\alpha = 2(1-\beta) - \gamma$ is -0.19 ± 0.09. At this stage it should be emphasized that the values for the critical exponents given above pertain only to the so-called "FeNi" phase (see next section) and the large error limits in their determination arise from the presence of the "FeCr" phase.

The present values for the critical exponents are listed in Table I together with the exponent values observed for crystalline elements[19–22] and amorphous alloys[1–7] and those deduced from a three-dimensional Heisenberg model.[18] Table I presents the following salient features: (i) the present values for the critical

TABLE I. Experimental values of the critical exponents β, γ, δ, and Curie temperature T_C for crystalline elements and amorphous alloys. The values of the specific-heat exponent α listed in this table are computed from the corresponding β and γ values. The theoretical values for these exponents deduced from a three-dimensional Heisenberg model are also included.

Compound	β	γ	δ	α	T_C(K)
Three-dimensional Heisenberg ferromagnet (Ref. 18)	≈ 0.37	≈ 1.38	≈ 5.0 4.73^a	-0.12	
Iron[b]	0.37 ± 0.03	1.30 ± 0.06		-0.04 ± 0.12	1043
Iron[c]	0.389	1.33	4.35	-0.10	1044
Cobalt[d] (Ref. 5)	0.36 ± 0.08	1.23 ± 0.05 $(1.21 \pm 0.04)^e$		0.05 ± 0.21	1388 ± 2^e
Nickel[f]	0.378 ± 0.004	1.34 ± 0.01	4.58 ± 0.02	-0.10 ± 0.01	627.4
Gadolinium (Ref. 22)	0.38	1.19	3.61	0.06 $(-0.20 \pm 0.02)^g$	292.85
Amorphous $Fe_{80}P_{13}C_7$ (Ref. 2)	0.38 ± 0.02	1.30 ± 0.05	4.47 ± 0.05	-0.06 ± 0.09 $(-0.18 \pm 0.04)^h$	586.55 ± 0.1
Amorphous $Co_{70}B_{20}P_{10}$ (Ref. 1)	0.402 ± 0.007	1.342 ± 0.025	4.39 ± 0.05	-0.15 ± 0.01	452.30 ± 0.07
Amorphous (Ref. 4)	0.41 ± 0.02	1.67 ± 0.08	5.07 ± 0.20	-0.49 ± 0.12	254.50 ± 1.0
$Fe_{32}Ni_{36}Cr_{14}P_{12}B_6$[i]	0.43 ± 0.02	1.33 ± 0.05	4.43 ± 0.17	-0.19 ± 0.09	250.0 ± 1.0
Amorphous (Ref. 7) $Fe_{29}Ni_{49}P_{14}B_6Si_2$	0.40 ± 0.01	1.70 ± 0.10	5.25 ± 0.10	-0.50 ± 0.08	384.5
Amorphous $Gd_{80}Au_{20}$ (Ref. 5)	0.44 ± 0.02	1.29 ± 0.05	3.96 ± 0.02	-0.17 ± 0.09	149.45 ± 0.2

[a] Value of δ obtained from β and γ.

[b] M. F. Collins, V. J. Minkiewicz, R. Nathans, L. Passell, and G. Shirane, Phys. Rev. 179, 417 (1969).

[c] S. Arajs, B. L. Tehan, E. E. Anderson, and A. A. Stelmach, Int. J. Magn. 1, 41 (1970).

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[f] J. S. Kouvel and J. B. Comly, Phys. Rev. Lett. 20, 1237 (1968).

[g] M. B. Salamon, D. S. Simmons, and C. C. Huang, Physica (Utrecht) B86–88, 583 (1977), experimental value of α for crystalline Gd.

[h] L. J. Schowalter, M. B. Salamon, C. C. Tsuei, and R. A. Craven, Solid State Commun. 24, 525 (1977), experimental value of α for amorphous $Fe_{75}P_{15}C_{10}$ alloy.

[i] Present results.

exponents, though in obvious disagreement particularly for γ and δ, with the previously[4] determined values on the same system, are in close agreement not only with those obtained for other amorphous systems (except for Metglas 2826B) but also with three-dimensional Heisenberg values; such a close agreement between experimental and theoretical values being also observed for crystalline elements. (ii) Compared with the values of β observed for crystalline elements, β values for amorphous systems are consistently larger in magnitude (≥ 0.4). The higher values of γ observed by Figueroa *et al.*[4] can be justified on the grounds that they could not have fit a straight line with only two points on their χ_0^{-1} vs T plot in the temperature range 260 to 280 K and had, therefore, to evaluate γ in the region 290 to 350 K (i.e., about 35 K above T_C) where χ_0^{-1} vs T curve has larger slope than that within the previous temperature range (we have also observed the same γ value in the temperature interval 285–300 K). An explanation for observing the value of γ as high as 1.68 in the above-said temperature range has been provided in the next section, where this γ value is shown to characterize the critical region of the so-called "FeCr" phase. That our value for γ should be characteristic of the magnetic phase transition observed at 250 ± 1 K (i.e., of the "FeNi" phase), is also justified from the argument that the most appropriate value of γ is the one obtained in the temperature region as close to T_C as possible. The difference in the δ values observed for Metglas 2826A, though small within the specified error limits, defies any obvious explanation at present. The observation (i) that the present exponent values follow closely Heisenberg values, sug-

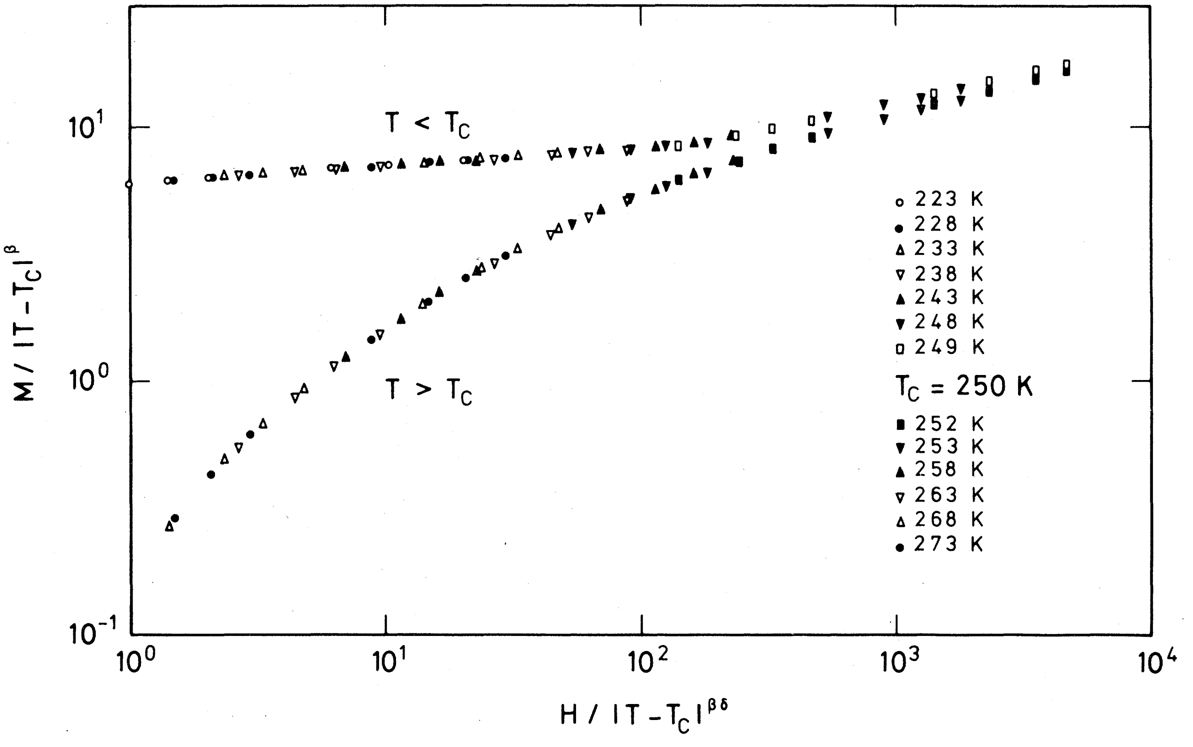

FIG. 7. Reduced magnetization $m \equiv M/|T - T_C|^{\beta}$ versus reduced field $h \equiv H/|T - T_C|^{\beta\delta}$ for temperatures below and above Curie temperature T_C.

gests that the short-range forces dominate the critical region. In order to rule out any influence from the long-range forces (e.g., dipolar forces), we use the criterion of Kadanoff *et al.*[13] that the effect of long-range forces on the critical fluctuations of the magnetization can be neglected in case

$$\epsilon = (1 - T/T_C) >> [\mu M_s(0)/kT_C]^{1/\beta(\delta-1)} \equiv T_r \ ,$$

where $\mu = g\mu_B S$ is the moment per spin and $M_s(0)$ the saturation magnetization at 0 K. The value of T_r for Fe and Ni calculated using their S and $M_s(0)$ values comes out to be 0.002 and 0.003, respectively. Experimentally, we have determined the critical exponents for our system in the region $\epsilon = 0.108$ for $T < T_C$ and $\epsilon = 0.128$ for $T > T_C$. Considering the fact that the value of ϵ for the present system is about 50 times larger than T_r values computed for Fe and Ni, the agreement between the present and Heisenberg values is not surprising at all. Also, long-range forces are expected to be significantly reduced in amorphous materials with a very short mean free path.[31] A qualitative explanation for the enhanced values of β observed in amorphous systems [observation (ii)] can be given within the framework of a dilution model proposed by Müller-Krumbhaar[11] (for details see Ref. 5).

The reduced magnetization $M/|T - T_C|^{\beta}$ plotted against the reduced field $H/|T - T_C|^{\beta\delta}$ on a log-log plot with the best choice of the exponent values $\beta = 0.43$ and $\delta = 4.43$ is shown in Fig. 7. This figure clearly demonstrates the validity of Eq. (5) over a wide temperature range. Plots given in Fig. 7 are not, however, very much sensitive to the choice of δ because of the large range of δ values observed.

B. Other observations

Preliminary Mössbauer data,[32] taken on Metglas 2826A in the amorphous state in the temperature range 4.2 to 295 K at close temperature intervals especially around T_C, besides confirming the essential observations made by Chien[26] on the basis of his Mössbauer data on the same system, provide the additional information that no observable change in the linewidth occurs in the temperature range 240 to 295 K and a small contribution due to the magnetic hyperfine interaction persisting even up to 295 K, over and above a well-defined quadrupole splitting, cannot be ruled out completely. The latter statement is justified from the observation that the data taken at 240 K (i.e., $T < T_C$) showed asymmetry in the quadrupole splitting similar to that observed at 270 and 295

K (i.e., $T >> T_C$) and also from the recognition of the fact that at present it is not possible to estimate quantitatively the contributions to asymmetric quadrupole splitting arising from hyperfine-field distribution and from the distribution of the electric field gradients as well as isomer shifts in amorphous materials. A much weaker dependence of the quadrupole splitting on temperature observed by Chien[26] for temperatures below ~ 320 K than that above this temperature may be a direct consequence of the above conclusion.

Chien's observations[26] and the deductions from them can be summarized as follows: (i) The hyperfine-field distribution $P(H)$ as a function of H exhibits two maxima, $(H_{\text{peak}})_1$ corresponding to a low-field component and $(H_{\text{peak}})_2$ corresponding to a high-field component whereas for Fe-Ni-P-B systems[27–30] only a single peak (corresponding to the high-field component) is observed in $P(H)$, suggesting thereby that Cr plays an important role in this alloy. (ii) $(H_{\text{peak}})_1$ and $(H_{\text{peak}})_2$ are temperature dependent. (iii) While the half width ΔH_2 of the high-field component varies with temperature as expected, the half width ΔH_1 of the low-field component does not show any significant dependence on temperature within the error limits. (iv) A comparison of the hyperfine-field distribution observed in Metglas 2826A with that observed for $Fe_{45}Ni_{35}P_{14}B_6$,[26] $Fe_{40}Ni_{40}P_{14}B_6$,[27] and Metglas 2826B[30] (i.e., alloys containing comparable transition-metal concentration) demonstrates that the high-field component in the present alloy has smaller peak value but comparable half width. Considering the fact that the metalloid atoms only contribute to the width[28,29] of $P(H)$ and the presence of Cr in the neighborhood can drastically reduce the peak value of $P(H)$, the observation (iv) suggests that in Metglas 2826A the high-field component should be due to those Fe atoms that have primarily Fe, Ni, and some Cr atoms sitting next to them whereas the low-field component (not observed in alloys without Cr) should be associated with those Fe atoms that have Cr atoms as their neighbors. The values of $(H_{\text{peak}})_1$, open circles, and $(H_{\text{peak}})_2$, open triangles, as a function of temperature evaluated by Chien[26] are reproduced in the insert of Fig. 2, where their temperature dependences have been empirically fitted by the power laws (dashed curves) given by

$$(H_{\text{peak}})_1 = 81.164(1 - T/T_{C_1})^{1/2}\ \text{kOe}\ ,\quad T_{C_1} = 320\ \text{K}\ , \tag{9}$$

$$(H_{\text{peak}})_2 = 224.0(1 - T/T_{C_2})^{1/3}\ \text{kOe}\ ,\quad T_{C_2} = 249\ \text{K}\ . \tag{10}$$

While Chien has given a correct power law [a power law of the type $(1 - T/T_C)^{0.37}$ also fits the data equally well] for describing the temperature dependence of $(H_{\text{peak}})_2$, surprisingly enough, he has failed to arrive at the best-fitting empirical relation (9) for $(H_{\text{peak}})_1$. Graphical as well as experimental errors in $(H_{\text{peak}})_1$ values can lead to a maximum error limit of ± 10 K in the T_{C_1} determination. The empirical relation (9) was arrived at by taking T_{C_1} and n as parameters in the general expression $(1 - T/T_{C_1})^n$ and finding out the combination of these parameters which gives the best fit to the experimental $(H_{\text{peak}})_1$ values when the theoretical value obtained at 4.2 K was normalized to the corresponding experimental value. It is interesting to note that the recent Mössbauer studies on crystalline FeCr alloys[33] have revealed that both hyperfine field and bulk magnetization as functions of temperature follow the same power law as given in Eq. (9) from 4.2 K to their Curie temperatures. The value of T_{C_1} mentioned above can also be justified otherwise.[34] Assuming hyperfine field due to Ni atoms to be negligibly small,[28,29] the total hyperfine field H_f for Metglas 2826A should be the sum of $(H_{\text{peak}})_1$ and $(H_{\text{peak}})_2$. The sum of the experimentally determined values of $(H_{\text{peak}})_1$ and $(H_{\text{peak}})_2$ gives the open square data points (insert of Fig. 2) whereas the sum of the theoretically determined values from Eqs. (9) and (10) gives the solid curve through the data points. Also shown in this insert is an empirical fit, the dash-dot curve, $H_f = 314(1 - T/\,T_C)^{0.4}$ kOe with $T_C = 249$ K to the H_f data points. Evidently, the data point taken at 4.2 K deviates by about 2% from this fit whereas large deviations are apparent for data points very close to T_C. An immediate consequence of the above observation is that spontaneous magnetization also varies as $(T_C - T)^{0.43}$ over the entire temperature range 4.2 K ($0.017T_C$) to 245 K ($0.98\ T_C$), $T_C = 250$ K.

Based on the above findings, the present alloy can be considered to comprise two magnetic phases[35]; one essentially a "FeCr" phase with $T_C = 320 \pm 10$ K and the other a "FeNi" phase (containing also a very small amount of Cr) with $T_C = 249$ K. The conclusion that the magnetic order for the so-called "FeCr" phase extends about 70 K beyond the Curie temperature 250 ± 1 K of the "FeNi" phase now provides a straightforward explanation for the anomalies observed for Metglas 2826A in (a) magnetic properties, e.g., (i) unusually high value of M_s at 250 K (also the bulk Curie temperature) and the finite curvature in the magnetization versus field curve taken at 295 K, (ii) large value[36] of γ observed[4] in the temperature range 290 to 350 K, (iii) Curie-Weiss law being obeyed only in the temperature interval 350 to 450 K (Ref. 4) (i.e., well above the Curie temperature of the "FeCr" phase), and (iv) the observation of a strong spin resonance signal for temperatures as high as 350 K (Ref. 37); (b) specific heat, the failure[6] to detect a sharp specific-heat peak, normally observed for all other amorphous materials (implying that the spin entropy in this system is not released abruptly but instead continuously over a broad temperature range and suggesting thereby a magnetic order extending well above T_C); and (c) resistivity, (i) no clear indication[24,25] of a sharp magnetic phase transition at 250 K in resistivity and (ii) the Kondo-like resistivity minimum observed at $T_{\min} = 270$ K[24,25] should be attributed to the scattering of the conduction electrons by weakly coupled Fe moments as proposed by Tsuei and Hasegawa[38] and not to the scattering from the disordered structure as proposed by Cochrane *et al.*[39] in view of the following arguments. Fe moments in this so-called "FeCr" phase around 270 K are expected to be weakly coupled for two reasons. First, Cr atoms not only have their moments antiparallel to those of Fe in crystalline FeCr alloys[40] but also drastically reduce the moment at the Fe sites[41] even though they may be at a large (several angstroms) distance from them. Second, 270 K is a temperature fairly close to the Curie point 320 ± 10 K for this phase (an abrupt change of slope observed in the present χ_0^{-1} vs T plot at ~ 280 K may have a direct bearing on this fact).

Finally, the possibility for Metglas 2826A to exhibit superparamagnetic behavior is ruled out on the following grounds: First, practically no change in the Mössbauer linewidth is observed across T_C or for temperatures well above T_C. Second, no anomaly is found in the temperature dependence of the quadrupole splitting. Third, magnetization versus H/T curves do not fall on a universal curve. Fourth, the so-called "FeCr" phase with $T_C = 320$ K [this value of T_C in crystalline FeCr alloys corresponds to 33 at.% Fe (Ref. 34)] present in our alloy cannot give rise to superparamagnetism because normal long-range ferromagnetic ordering is known[33,34] to exist beyond 29 at.% Fe for the crystalline FeCr alloys and the situation in the amorphous state cannot be expected to be different.

IV. CONCLUSIONS

Our exhaustive magnetization data taken in the critical region of Metglas 2826A when analyzed with caution permit us to arrive at reliable values for the critical exponents characterizing the so-called "FeNi" phase. The exponent values follow closely the predictions of a three-dimensional Heisenberg model and this observation brings to the limelight the dominance of the short-range forces in the critical region. A close scrutiny of the Mössbauer data taken on the present alloy and comparison of these data with similar data taken on other amorphous alloys enables us not only to rule out the possibility of superparamagnetism in this system but also to provide a clear-cut explanation for the anomalies observed in the various properties of Metglas 2826A.

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[35] Since these magnetic phases are presumably microscopic regions in intimate contact with each other, it is rather difficult to conceive such phases to behave as independent thermodynamic entities. A conceptual difficulty of this nature could, in our opinion, be resolved in case the following possibility is considered: Fe atoms in the neighborhood of Ni atoms retain their full moment value whereas those Fe atoms sitting next to Cr atoms possess a diminished moment value. These two types of Fe moments are ferromagnetically coupled with each other, form the so-called "two sublattices" (in a sense analogous to the two-sublattice model for ferromagnetism in crystalline systems) and exhibit different functional dependence on temperature. Though a phase separation of the type mentioned in the text could be a gross oversimplification of the actual situation, such an approximation nevertheless places all the anomalous experimental results obtained for Metglas 2826A within a fairly consistent framework.

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