Quenched Disorder and the Critical Behavior of a Partially Frustrated System

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We report the direct observation of the effects of quenched disorder on the critical behavior of partially frustrated amorphous FeMnZr alloys by the systematic analysis of high-precision ac susceptibility data and dc magnetization data. Interestingly, the analysis reveals that the presence of short-range quenched disorder does not alter the actual critical behavior. However, it does affect quantities such as the Curie temperature, the peak value of effective exponent γ, width of the peak, and crossover temperatures. The observed temperature dependence of the effective critical exponent can be understood in terms of the field-theoretical renormalization group approach. Also, the present results would help in identifying the main source of the spread in the exponent values reported in the literature.

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The effects of disorder on the critical properties of systems that undergo a second order phase transition is an important problem in the fields of condensed matter and statistical physics. An understanding of experimental results in this field in the context of existing theoretical models has significant implications and allows for a determination of the importance of the universal characteristics of the system such as lattice dimensionality and order parameter symmetry. The investigation of critical properties of disordered magnetic systems has provided substantial information over the years and continues to be an active area of research that contributes to the understanding of critical phenomena [1]. Existing results suggest that the phase transition in systems with quenched disorder may be qualitatively different from that in pure systems. However, one consequence of this behavior is that the effective critical exponent for the susceptibility, $\gamma_{\rm eff}$, in systems with quenched disorder has approximately the same value [2] as it does in the asymptotic critical region (ACR) of a pure system. In crystalline materials, the susceptibility exponent decreases monotonically [3] from its asymptotic value towards the mean-field value with increasing reduced temperature [$\eta = (T - T_c)/T_c$, where T_c is Curie temperature]. In amorphous ferromagnets, however, the exponent value goes through a peak [4] before decreasing towards the mean-field value. The peak value of $\gamma_{\rm eff}$ and its dependence on the concentration [5] of magnetic atoms in a given amorphous alloy have found possible interpretation in terms of a number of theoretical models. One of the central problems here is to answer the following basic questions: (i) Do the critical exponents of a homogeneous magnet change under the presence of quenched disorder? If so, (ii) how do they change? Since the functional dependence of $\gamma_{\rm eff}$ on η is different in amorphous and crystalline ferromagnets, a systematic investigation of the influence of structural relaxation [caused by isothermal annealing (IA)] in glassy ferromagnets is expected to provide a precise assessment for the existing theories. On the other hand, close scrutiny of the results published in the literature thus far reveals that the exponents deviate from the expectated values due to various possible problems in the analysis [6]. Within this context, in this Letter we explain (i) how the critical behavior of a partially frustrated system is influenced by the presence of quenched disorder and (ii) the reasons for the spread in the previously reported critical exponent values. For this purpose, values of the asymptotic [i.e., leading correction to scaling (LCTS)] critical exponent γ have been accurately determined from high-precision low-field ac susceptibility (ACS) and high-field dc magnetization (DCM) measurements made on a series of amorphous ferromagnets through an elaborate data analysis. Results of various methods of analysis have been compared so as to clearly distinguish between the effective and asymptotic critical exponent values.

The system studied in the present work is amorphous (a-) $Fe_{90-x}Mn_xZr_{10}$ alloys, where significant changes in magnetism are observed over a composition range $0 < x < 16$. Figure 1 summarizes the magnetic properties of the system [7]. This figure resembles both the phase diagram of binary FeZr alloys [8] in the Fe rich region and part of the mean-field theory diagram [9] for Heisenberg spin glasses. Basically, magnetic transitions are seen at two temperatures; T_c and spin-glass-like transition temperature (T_{sg}). Two features have been observed in this system that deserve particular note: (i) Below T_c, the alloys order ferromagnetically with a finite magnetic

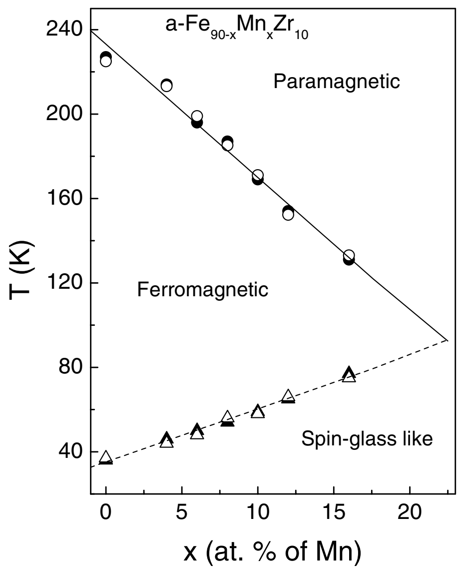

FIG. 1. The magnetic phase diagram of amorphous $Fe_{90-x}Mn_xZr_{10}$ ($0 < x < 16$) alloys. The solid and open symbols are the data obtained from magnetization and ac susceptibility measurements, respectively. The solid and dashed lines are shown for clear view.

correlation length of about 25 Å. Although the susceptibility behaves quite like that of a soft ferromagnet, the value of magnetization is much less than (about 7% at 75 kOe field) that expected for a collinear ferromagnet. There is a large high-field slope in the magnetization curve [7] and the slope increases with increasing Mn concentration; (ii) T_{sg} marks the temperature at which a partially frustrated three-dimensional (3D) Heisenberg magnet develops static spin components perpendicular to the ferromagnetic (FM) order established at T_c. Between T_c and T_{sg}, the system is FM, while below T_{sg} the magnetic structure is characterized by coexisting, and mutually perpendicular, FM and xy-spin-glass ordering [10].

Although the extrapolation procedure from the nonsaturated magnetization curves even up to 75 kOe applied field in the partially frustrated system yields considerable error in obtaining the zero-field quantities [6], by focusing on both high-precision ACS and DCM measurements it is possible to probe the effect of quenched disorder on the critical behavior of the present system by systematic analysis on the data obtained for various samples that have undergone for systematic IA treatment. ACS measurements were performed at 20 and 50 mK intervals in rms ac driving fields of $H_{ac} = 8$ and 12 μT at a frequency of 81 Hz using the mutual inductance method [11], while DCM measurements were done in external fields up to 75 kOe. Microstructural details have been investigated through standard x-ray diffractometry and electron microscopy techniques.

Amorphous samples have been prepared by melt-spinning [11] techniques in an argon atmosphere. Prior to heat treatment, the crystallization temperatures of the samples were determined by differential scanning calorimetry and found to be about 750 K. Subsequently, the samples were annealed at 600 K with annealing times of 1, 2, 4, 6, 8, and 10 h. For this purpose, the samples consisting of several lengths of ribbon were sealed in quartz tubes after evacuating to 10^{-7} Torr. The furnace temperature was raised to a particular annealing temperature and maintained within ± 2 K throughout the annealing time. After the first heat treatment, XRD, ACS, DCM measurements, and SEM studies were carried out. Subsequently, the samples were annealed for longer periods and the measurements were repeated.

In order to understand the effects of quenched disorder on the critical behavior, the high-precision ACS and DCM measurements were performed in the temperature range close to the magnetic order-disorder phase transition temperatures. As shown in Figs. 2(a) and 2(b), increased accuracy and substantially reduced scattering in the ACS and DCM data make them amenable to more rigorous analysis. Inverse susceptibility data, obtained from both ACS and DCM [12], were fitted to various models {Eq. (1a) simple power law, (1b) Kouvel-Fisher method [13], (1c) [(1d)] LCTS with linear [nonlinear

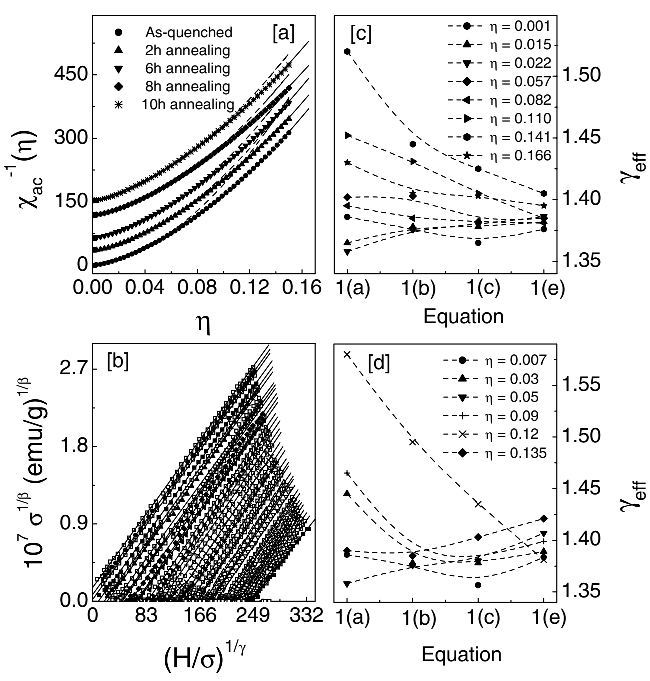

FIG. 2. (a) The inverse ACS data plotted against reduced temperature for a-$Fe_{86}Mn_4Zr_{10}$ alloy annealed at 600 K for different annealing times. Note that the y-axis value is shifted by 35 (2 h), 65 (6 h), 118 (8 h), and 153 (10 h) with respect to the as-quenched sample value. (b) A modified-Arrott plot for a-$Fe_{86}Mn_4Zr_{10}$ alloy annealed at 600 K for 8 h. (c) The effective critical exponents for the sample annealed at 2 h obtained from ACS data. (d) The effective critical exponents for the sample annealed at 8 h obtained from DCM data.

(NL)] variables, and (1e) NL relevant scaling models [14]} as a function of η, as shown below:

$$\chi^{-1}(\eta) = L^{\text{eff}}\eta^{\gamma_{\text{eff}}}, \qquad \eta > 0, \tag{1a}$$

$$\left[\frac{d\{\ln[\chi^{-1}(\eta)]\}}{d\eta}\right]^{-1} = \left[\frac{\eta T_c}{\gamma}\right], \qquad \eta > 0, \tag{1b}$$

$$\chi^{-1}(\eta) = L\eta^{\gamma}[1 + u^+_{\chi_1}(\eta)^{\Delta_1} + u^+_{\chi_2}(\eta)^{\Delta_2}]^{-1}, \quad \eta > 0, \tag{1c}$$

$$\chi^{-1}(\eta) = L' t\tilde{\eta}^{\gamma}[1 + \tilde{u}^+_{\chi_1}(\tilde{\eta})^{\Delta_1} + \tilde{u}^+_{\chi_2}(\tilde{\eta})^{\Delta_2}]^{-1}, \quad \tilde{\eta} > 0, \tag{1d}$$

$$\chi^{-1}(\eta) = L'' t\tilde{\eta}^{\gamma}[1 + \tilde{u}^+_{\chi}(\tilde{\eta})]^{-1}, \qquad \tilde{\eta} > 0, \tag{1e}$$

where γ is the asymptotic critical exponent, Δ_1, Δ_2 are the LCTS exponents, $u^+_{\chi_1}$, $u^+_{\chi_2}$, $\tilde{u}^+_{\chi_1}$, $\tilde{u}^+_{\chi_2}$, $\tilde{u}^+_{\chi}$, and $L = (h_0/m_0)$ are the critical amplitudes, and $\tilde{\eta} = (t-1)/t$, $t = T/T_c$ are the NL scaling variables. A detailed analysis of $\chi^{-1}(T)$ data reveals that, in the ACR, the LCTS analysis based on expressions that include nonanalytic correction terms alone yields the same result regardless of whether these correction terms are expressed in linear variables [Eq. (1c)] or in nonlinear variables [Eq. (1d)]. On the other hand, Eq. (1e) provides a very good overall fit over temperature ranges as wide as $T_c \le T \le 1.4T_c$ for $\chi^{-1}(T)$. With reference to the effect of IA on γ_{eff} and other relevant magnetic parameters, the important findings are (i) the asymptotic critical exponent γ remains unaffected in the ACR [Figs. 2(c) and 2(d)] even though thermal relaxation leads to a substantial increase in T_c, and (ii) the IA influences quantities such as the peak value of the exponent γ ($\gamma^{\text{peak}}_{\text{eff}}$), width of the peak, and crossover temperature. The effective critical exponent, γ_{eff}, was calculated [13] from both ACS and DCM data in different temperature ranges and is illustrated in Fig. 3(a). A comparison between effective exponents and Eq. (1c) reveals that in the ACR ($|\eta| < |\eta_{\text{cross}}|$) [16], γ_{eff} and γ are related as

$$\gamma_{\text{eff}}(\eta) = \gamma - u^+_{\chi_1}\Delta_1(\eta)^{\Delta_1} - u^+_{\chi_2}\Delta_2(\eta)^{\Delta_2}, \tag{2}$$

and in the limit $\eta \to 0^+$, $\gamma_{\text{eff}}(\eta)$ coincides with γ.

Interestingly, the critical exponent is found to change with distance from T_c [Fig. 3(a)] beyond η_{cross}. We consider the observed behavior of the critical exponent values in terms of the following two methods: (i) the renormalization group (RG) approach of Sobotta *et al.* [17] used to describe the critical properties of site and bond frustrated systems and (ii) field-theoretical RG (FTRG) approach used by Dudka *et al.* [15] to describe the critical properties of random quenched magnets.

According to Sobotta *et al.*, the quenched disordered magnet may be described as an equilibrium system with additional forces of constraints [18] to keep the atoms at their randomly distributed positions. This assumption results in an effective Hamiltonian that contains both the spin Hamiltonian and an additional term that involves the chemical potential and depends on the random variables. With this simplified calculation, the most important result of the investigation was connected with the fixed-point structure which showed that different concentrations lead to different crossover temperatures. Particularly, it predicts that there exists a limiting value of the concentration, where the critical behavior is completely governed by the Fisher-renormalized critical exponents ($\alpha = -1$, $\beta = \frac{1}{2}$, $\gamma = 2$, $\delta = 5$). Although the concentration dependence of the crossover temperature has been observed in the present system [5], the critical exponent values are not in agreement with the values predicted for the above-mentioned model.

An alternative approach we have followed uses the FTRG technique with the two-loop approximation calculations refined by the resummation of the perturbation theory series for dilute Heisenberg-like magnets. In the work of Dudka *et al.*, the effective Hamiltonian is obtained by the RG approach using replica tricks [19]. In the RG scheme, the effective critical exponents are calculated in the region where the couplings $u(l)$, $v(l)$ depend on the flow parameter, l, and have not reached their fixed-point values. Here l is related to the distance η

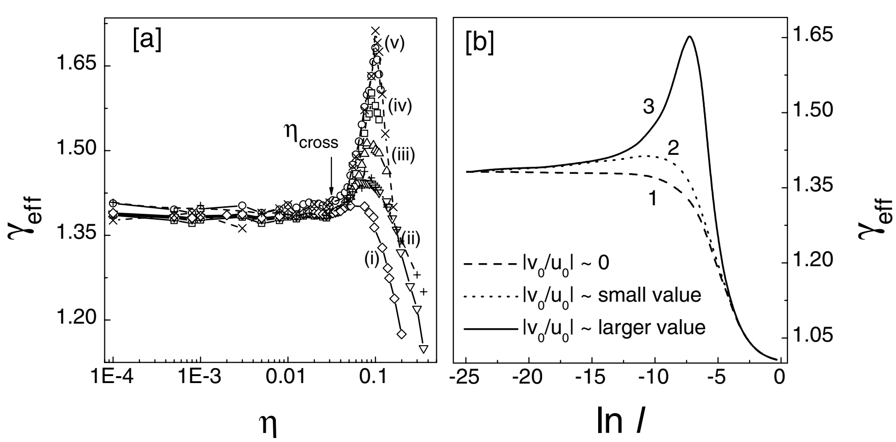

FIG. 3. (a) The effective critical exponent obtained from ACS data [(i) 10 h, (ii) 8 h, (iii) 6 h, (iv) 2 h annealing, and (v) as-quenched state] and DCM data (-+- 8 h and -×- as-quenched state) as a function of reduced temperature range for the a-$Fe_{86}Mn_4Zr_{10}$ alloy annealed at 600 K. (b) The effective critical exponent versus the logarithm of the flow parameter. This figure is taken from Ref. [15] and shown for comparison.

to Curie point. Particularly, for the critical exponent γ, they obtained

$$\gamma_{\mathrm{eff}}^{-1}(\eta) = 1 - \frac{\bar{\gamma}_{\phi^2}[u\{l(\eta)\}, v\{l(\eta)\}]}{2 - \gamma_{\phi}[u\{l(\eta)\}, v\{l(\eta)\}]} + \cdots. \quad (3)$$

For the quenched random magnets model, the coupling v is proportional to the variance of disorder and the ratios $|v_0/u_0|$ define the degree of disorder. The effective exponent values obtained for different ratios $|v_0/u_0|$ are shown in Fig. 3(b) (Fig. 4 of Ref. [15]). The curves in Figs. 3(a) and 3(b) reflect some similarities between the present experimental observations and the theoretical approach. Curve 1 of Fig. 3(b) corresponds to the pure Heisenberg model, whereas curves 2 (weak disorder) and 3 (strong disorder) provide two possible scenarios for the effective exponents of the disordered Heisenberg model. To our best knowledge, the results shown in Fig. 3(a) are the first experimental report of this type of γ_{eff} behavior for the disordered Heisenberg-like magnet. Curves (ii)–(v) of Fig. 3(a) show that, before reaching the ACR, the exponent exhibits a distinct peak and depends strongly on the amount of disorder in the system. On the other hand, curve (i) of Fig. 3(a) is comparable to curve 2 of Fig. 3(b) suggests the presence of weak disorder in the system, which is further confirmed through XRD and SEM studies.

The important finding is that the values of asymptotic critical exponent for the present series of alloys are the same as those of an isotropic 3D Heisenberg ferromagnet. This refutes the earlier claim based on logarithmic analysis [20] that the critical exponent γ for the same alloys has anomalously large values. The results of the present investigation make it clear that such unphysically large exponent values as well as the spread in the reported exponent values are an artifact of the analysis carried out on the magnetization data taken either partly or completely outside the ACR. In fact, even in the ACR, γ_{eff} weakly depends on temperature and possess anomalously large values for $|\eta| > |\eta_{\mathrm{cross}}|$. Also, it is seen that the exponent values in the ACR do not depend on the presence of quenched disorder in the system.

In summary, we have shown experimentally that the asymptotic critical exponent values do not depend on the presence of amount of quenched disorder in the system. Interestingly, it is found to have substantial effects on other quantities such as Curie temperature, peak values of exponent, crossover temperature, and width of the peak outside the critical region. We believe that the extension of this work to various published data [21] on different samples can help further in identifying the main source of the spread in the exponent values.

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