Effect of ferromagnetic critical fluctuations on the phase transition to a spin-glass state

V. G. Bar'yakhtar, A. B. Surzhenko, and G. A. Takzeĭ

Institute of Metal Physics, Academy of Sciences of the Ukraine
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Measurements of the dynamic magnetic susceptibility and its nonlinear components are used to study the "paramagnet to spin glass" phase transition in the amorphous alloy $(Fe_{0.65}Mn_{0.35})_{75}P_{16}B_6Al_3$ and in the fcc alloy $Fe_{56}Ni_{24}Cr_{20}$. These compounds are characterized by a substantial ferromagnetic exchange. The critical exponents describing the phase transition in these systems are different from those in classical spin glasses.

1. INTRODUCTION

The huge amount of experimental evidence which has now been accumulated confirms the conclusion of spin-glass theory[1] that a cooperative paramagnet to spin glass (PM-SG) phase transition occurs at a critical temperature $T_c > 0$. This conclusion is based primarily on results on nonlinear magnetic susceptibilities of dilute alloys of transition metals with noble metals[2,3] and also of concentrated alloys of $3d$ transition metals.[4,5]

For these reasons, the PM-SG phase transition can be described by critical exponents,[6] among which we focus on three here: β, γ, and δ. Here β and γ are the critical exponents of the order parameter and of the generalized (nonlinear) susceptibility of the SG, respectively, while δ characterizes the effect of a static magnetic field on the order parameter of the SG. The selection of these exponents, which are related by the scaling relation[7]

$$\beta\delta=\beta+\gamma, \tag{1}$$

makes it possible to experimentally test similarity theory for describing the PM-SG phase transition in specific systems.

For SGs with compositions far from the critical concentration ($X^{(0)}$) for the onset of a long-range ferromagnetic order on the magnetic phase diagram, the contributions of the ferromagnetic and antiferromagnetic interactions to the overall exchange energy are approximately equal. For such systems, regardless of the type of competing exchange, the corresponding critical exponents are approximately the same[4] ($\beta\approx 1$, $\gamma\approx 2$, $\delta\approx 3$). These SGs can thus be assigned to a common universality class.

For SGs in the immediate vicinity of $X^{(0)}$, whose description requires consideration of the critical ferromagnetic fluctuations, much less is known. In particular, for the PM-SG phase transition in the case $X\to X^{(0)}$ we do not have a theory of critical phenomena like that derived in Ref. 8 for the paramagnet-ferromagnet phase transition. We should also point out that the wide scatter in the values of the critical exponents for SGs of various types with $X\to X^{(0)}$ means that the experimental situation regarding this question is also extremely uncertain.

We have accordingly analyzed the changes which occur in the characteristics of the PM-SG phase transition as the composition of the alloys is varied.

2. EXPERIMENTAL PROCEDURE

According to Refs. 6, 9, and 10, the phase transition can be described by a scaling equation. If the magnetic field is weak, this equation is

$$\chi_s=\tau^{\beta}F(h^2/\tau^{\varphi})=a_2\tau^{\beta-\varphi}+a_4\tau^{\beta-2\varphi}+\ldots, \tag{2}$$

where $\chi_s \equiv M/H-\chi_0$ is the singular part of the susceptibility, M is the magnetization, $\chi_0=(dM/dH)_{H=0}$ is the initial susceptibility, $\tau=(T-T_c)/T_c$ is the reduced temperature, $h=\mu gH/(k_BT_c)$ is the reduced magnetic field, $F(X)$ is a scaling function, and the coefficients a_{2n} are the coefficients of a series expansion of χ_s in powers of $h^2\tau^{-\varphi}$, where $\varphi=\beta+\gamma$, and β and γ are the Edwards–Anderson critical exponents of, respectively, the order parameter,

$$q_{EA}=\langle\langle s_i\rangle_T^2\rangle_J$$

and the generalized (nonlinear) susceptibility,

$$\chi_{EA}=N^{-1}\sum_{ij}\langle(s_is_j)\rangle_T-\langle s_i\rangle_T\langle s_j\rangle_T)^2\rangle_J$$

of a spin glass. Here $\langle\ldots\rangle_T$ means a thermodynamic average, and $\langle\ldots\rangle_J$ means a configurational average. The terms of series (2) are obviously proportional to nonlinear components of the dynamic magnetic susceptibility, which can be measured at odd harmonics of the frequency of the magnetization-reversal field.

The linear component χ_0 and the nonlinear components of the dynamic magnetic susceptibility were studied with the help of a mutual-induction bridge. The vertical component of the geomagnetic field was canceled with the help of a copper solenoid in order to suppress the signal from parasitic even harmonics of the dynamic magnetic susceptibility induced by the geomagnetic field.

The critical exponent δ was estimated from the relation

$$1-\chi_0'(H,\ T_m)/\chi_0'(0,\ T_f)\propto q_{EA}\propto H^{2/\delta}, \tag{3}$$

which was proposed in Ref. 11. Here T_f and T_m are the temperatures at which the real component χ_0' of the dynamic magnetic susceptibility reaches its maxima in a zero magnetic field and in a static field $H\neq 0$.

For this study we selected the amorphous alloy

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$(Fe_{0.65}Mn_{0.35})_{75}P_{16}B_6Al_3$ ($X_{Fe}^{(0)} = 0.68$; Ref. 12) and the crystalline (fcc) alloy $Fe_{56}Ni_{24}Cr_{20}$ ($X_{Fe}^{(0)} = 55$ at. %; Ref. 13). The amorphous sample was a parallelepiped ($3\times3\times15$ μm) formed from a tape 20 μm thick. The crystalline sample was a cylinder 3 mm in diameter and 15 mm long. In the former case, the magnetization-reversal field was applied along the plane of the tape; in the latter case, it was applied along the axis of the cylinder.

3. EXPERIMENTAL RESULTS

We first note that in molecular field theory it is customary to plot the magnetic phase diagrams of the SG systems in the coordinates $T/\bar{J} - \bar{J}_0/\bar{J}$, where T is the temperature, $\bar{J}_0$ is the mean exchange, and $\bar{J}$ is the standard deviation.[14,15] A long-range magnetic order exists at values $\bar{J}_0/\bar{J} \geqslant 1$. Experimentally, in contrast, the phase diagrams are usually plotted in the coordinates $T - X$ (see, for example, Refs. 12 and 13). In this case, as was mentioned above, a ferromagnetic order arises at $X = X^{(0)}$. This representation, however, is not always convenient, since it often fails to reflect the one-to-one correspondence between the parameter $\bar{J}_0/\bar{J}$ and the concentration of the magnetic element, X. For this reason, the parameter[16]

$$\eta = \theta/T_f = \bar{J}_0/\bar{J}$$

which can easily be determined experimentally, is introduced in molecular field theory in order to evaluate $\bar{J}_0/\bar{J}$; here θ is the paramagnetic Curie temperature, and T_f is the temperature at which the SG freezes. As we will show below, the classification of real SGs on the basis of this parameter frequently turns out to be extremely convenient.

3.1. The amorphous alloy $(Fe_{0.65}Mn_{0.35})_{75}P_{16}B_6Al_3$

Figure 1 shows the temperature dependence of the real part χ_0' and the imaginary part χ_0'' of the dynamic magnetic susceptibility, along with that of the nonlinear susceptibilities χ_2 and χ_4. There are anomalies in these components of the susceptibility near $T_f \approx 42.5$ K, which indicate a transition of this alloy into a SG state. The value found for the paramagnetic Curie temperature from the Curie–Weiss law in the temperature interval $T_f < T < 2T_f$ is $\theta = 20.9 \pm 0.2$ K. For this SG, the value $\eta = \theta/T_f = 0.49$ thus indicates a fairly large ferromagnetic-exchange component in the overall exchange energy, while for classical SGs we would have $\eta \approx 0$.

Working from the temperature dependence of the nonlinear susceptibilities shown here, we can draw conclusions about the nature of the PM-SG transition in this alloy. Specifically, from expansion (2) we find

$$|\chi_2| \propto \tau^{-\gamma} \tag{4}$$

and

$$\chi_4 \propto \tau^{-(\beta+2\gamma)}, \tag{5}$$

so for temperatures $T > T_f$ we easily find

$$|\chi_4/\chi_2| \propto |\chi_2|^{(1+\beta/\gamma)}. \tag{6}$$

It follows that expansion (2) is valid if relation (6) holds. In other words, in this case the PM-SG phase transition described by a scaling equation of state occurs.

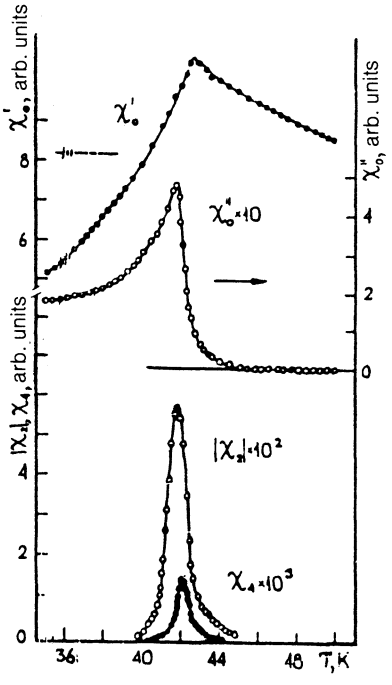

FIG. 1. Temperature dependence of the real component χ_0' and the imaginary component χ_0'' of the dynamic magnetic susceptibility and of the nonlinear susceptibility components χ_2 and χ_4 for the alloy $(Fe_{0.65}Mn_{0.35})_{75}P_{16}B_6Al_3$. The amplitude of the magnetization-reversal field is 1 Oe, and its frequency is 83.7 Hz.

Figure 2 shows relation (6) in full logarithmic scale. We see that this relation holds over the fairly broad temperature interval with $1 + \beta/\gamma = 1.37 \pm 0.02$, so the PM-SG transition is of a cooperative nature in this alloy. It follows that the nonlinear susceptibility χ_2 must be of a power-law nature as in (4); this conclusion is supported by the data in Fig. 3. A least-squares fit of (4) to the experimental data yields a critical temperature $T_c = 42.2 \pm 0.1$ K for the PM-SG phase transition and the corresponding value $\gamma = 2.05 \pm 0.05$. Knowing the values of γ and $1 + \beta/\gamma$, we then easily find $\beta = 0.77 \pm 0.05$.

The critical exponent β describing the temperature dependence[1]

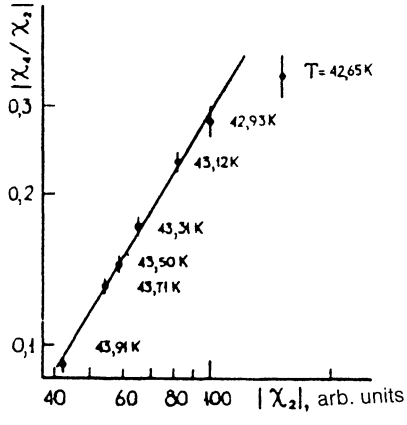

FIG. 2. Plot of $|\chi_4/\chi_2|$ versus $|\chi_2|$ for the amorphous alloy $(Fe_{0.65}Mn_{0.35})_{75}P_{16}B_6Al_3$.

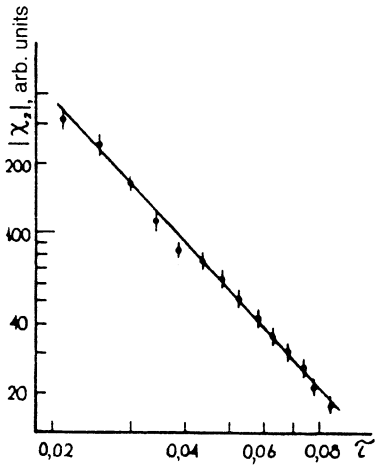

FIG. 3. Nonlinear magnetic susceptibility χ_2 versus the reduced temperature τ for the alloy $(Fe_{0.65}Mn_{0.35})_{75}P_{16}B_6Al_3$.

$$q_{EA} \propto \tau^{\beta} \tag{7}$$

can be determined independently. To do this, we use the relation

$$q_{EA}=1-T\chi_0'(T)/[C+\theta\chi_0'(T)], \tag{8}$$

which was found in Ref. 17; C is the Curie constant.

A plot of $q_{EA}(\tau)$ in full logarithmic scale for the value $T_c = 42.2$ K found above can be approximated well by a straight line (Fig.4). From the slope of this line at small τ we find $\beta = 0.76 \pm 0.04$. This value agrees completely with the value found for β from the nonlinear susceptibilities in the discussion above. With β and γ known, it is now a simple matter to use scaling relation (1) to find the critical exponent $\delta = 3.7 \pm 0.2$. For an independent test of the validity of (1), we then examine the effect of a magnetic field on this transition.

It can be seen from the field dependence $1-\chi_0'(H,T_m)/\chi_0'(0,T_f)$ in Fig. 5 that Eq. (3) holds over a broad range of magnetic fields. The value $\delta = 3.8 \pm 0.3$ found in this manner agrees well with the value found above from (1).

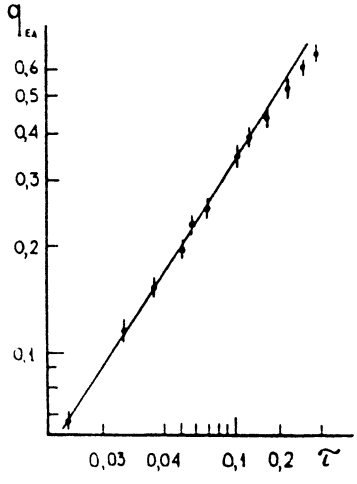

FIG. 4. The Edwards–Anderson order parameter q_{EA} versus the reduced temperature τ for the spin glass $(Fe_{0.65}Mn_{0.35})_{75}P_{16}B_6Al_3$.

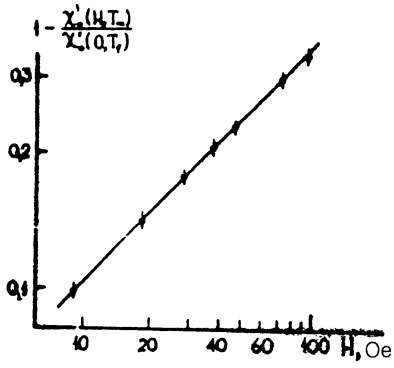

FIG. 5. Field dependence of the parameter $1-\chi_0'(H,T_m)/\chi_0'(0,T_f)$ for the spin glass $(Fe_{0.65}Mn_{0.35})_{75}P_{16}B_6Al_3$.

On the basis of these results, we conclude that a PM-SG phase transition which can be described by a scaling state function and by corresponding critical exponents occurs at the critical temperature $T_c = 42.2$ K in the amorphous alloy $(Fe_{0.65}Mn_{0.35})_{75}P_{16}B_6Al_3$. This alloy lies near the critical concentration $X_{Fe}^{(0)}$ ($\eta = 0.49$) for the onset of a long-range ferromagnetic order on the magnetic phase diagram.[12] The critical exponents β and γ for this alloy, however, differ from the corresponding values ($\beta \approx 1$, $\delta \approx 3$; Ref. 4) found for classical SGs with $\eta \approx 0$.

3.2. The fcc alloy $Fe_{56}Ni_{24}Cr_{20}$

The other alloy which we selected for study was the fcc alloy $Fe_{56}Ni_{24}Cr_{20}$. Several circumstances led us to this alloy. First, its composition is in the immediate vicinity of the critical concentration $X_{Fe}^{(0)} = 55$ at. % for the onset of a long-range ferromagnetic order, according to Ref. 13. Second, this alloy has been studied in detail by a variety of methods.[13,18–20] In particular, studies of the static susceptibility (of thermomagnetic-history effects) have made it possible to construct a magnetic H–T phase diagram[18] and to show that this alloy exhibits the properties of an Ising SG. The anomalously low de Almeida–Thouless field ($H_{AT} \approx 40$ Oe) confirms the conclusion[13] that the composition of this alloy is close to the critical concentration $X_{Fe}^{(0)}$.

On the other hand, studies of the cross section for small-angle neutron scattering[19] indicate that a long-range ferromagnetic order prevails in this alloy at temperatures $T > 4.2$ K. Measurements of the real component χ_0' of the dynamic magnetic susceptibility in static magnetic fields indicate significant critical ferromagnetic fluctuations[20] at temperatures $T \geqslant T_f$.

It can thus be concluded that both SG and ferromagnetic critical fluctuations occur in the fcc alloy $Fe_{56}Ni_{24}Cr_{20}$ in its paramagnetic state near the SG freezing temperature T_f. It is this circumstance that makes this alloy particularly interesting for a study of the PM-SG phase transition. To illustrate the situation, we show the temperature dependence of χ_0', χ_0'', χ_2, and χ_4 of the alloy $Fe_{56}Ni_{24}Cr_{20}$ in Fig. 6. These curves are qualitatively similar to the corresponding curves in Fig. 1 for the SG $(Fe_{0.65}Mn_{0.35})_{75}P_{16}B_6Al_3$. In this case, however, a Curie–Weiss law holds only at temperatures

$T > 200\ \text{K} \gg T_f \simeq 21$ K. Experimentally, a value $\theta = 80 \pm 5$ K, i.e., $\eta \simeq 4$, is found. This anomalously large value of η for an alloy in which a long-range ferromagnetic order does not arise is evidence that the result

$$\bar{J}_0/\bar{J} = \theta/T_f$$

of molecular field theory[14–16] does not apply to SG systems in which critical ferromagnetic fluctuations are large. The value found for η for the alloy $Fe_{54}Ni_{26}Cr_{20}$ also indicates that, on the magnetic X–T phase diagram, the composition of this alloy is much closer than the composition of the amorphous alloy discussed above to the corresponding critical concentration $X^{(0)}$.

Let us see how these distinguishing features of the alloy $Fe_{54}Ni_{26}Cr_{20}$ affect the picture of magnetic transitions which occur in it. We will analyze the experimental data (Fig. 6) by fitting (6) to them. The results of this fit, shown in Fig. 7, lead to the paradoxical conclusion that the ratio β/γ is negative. Here we wish to stress that neither β nor γ could be negative if a PM-SG phase transition occurred. On the other hand, an examination of this transition in the alloy $Fe_{56}Ni_{24}Cr_{20}$ on the basis of the fluctuation theory[7] of paramagnet-ferromagnet phase transitions also leads to the physically meaningless result $\beta_{FM} < 0$, where β_{FM} characterizes the temperature dependence of the ferromagnetic order parameter (the spontaneous magnetic moment) near its Curie temperature.

It follows that the transition observed in $Fe_{56}Ni_{24}Cr_{20}$ at $T \approx 21$ K cannot be described satisfactorily by the critical exponents corresponding to PM-SG and paramagnet-ferromagnet phase transitions. The probable reason for this result is a coexistence of SG and ferromagnetic critical fluctuations in this alloy at temperatures $T \gtrsim T_f$.

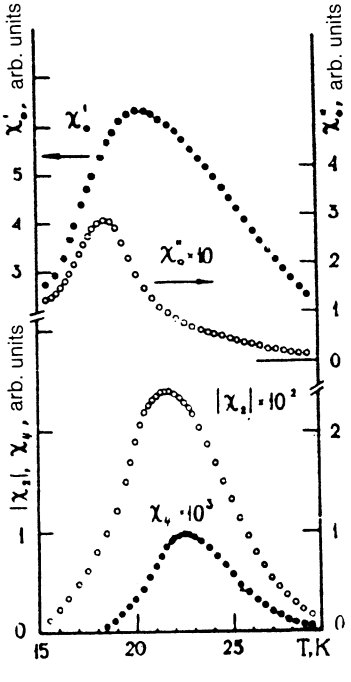

FIG. 6. Temperature dependence of the real component χ_0' and the imaginary component χ_0'' of the dynamic magnetic susceptibility and of the nonlinear susceptibility components χ_2 and χ_4 for the crystalline alloy $Fe_{56}Ni_{24}Cr_{20}$. The amplitude of the magnetization-reversal field is 1 Oe, and its frequency is 83.7 Hz.

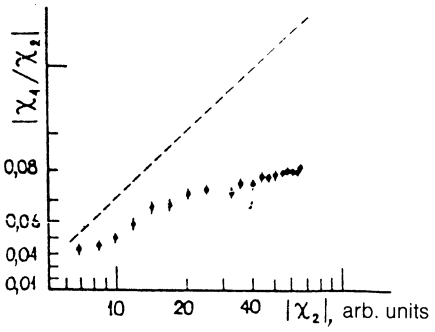

FIG. 7. Plot of $|\chi_4/\chi_2|$ versus $|\chi_2|$ for the crystalline alloy $Fe_{56}Ni_{24}Cr_{20}$. The dashed line corresponds to $\beta/\gamma = 0$.

4. DISCUSSION OF EXPERIMENTAL RESULTS

To put the results of research on the PM-SG phase transition in real systems into systematic form, we summarize the values of the critical exponents describing this transition in Table I. The particular alloys shown here were chosen because they differ in the relative weight of the ferromagnetic-exchange component in the overall exchange energy. It can be seen from these results that with increasing $\eta = \theta/T_f$, i.e., as the composition of the alloy approaches the critical value $X^{(0)}$, the critical exponent β decreases, while δ increases, for both the crystalline and amorphous systems.

Let us examine this tendency from the standpoint of the effect of a ferromagnetic exchange, which strengthens as $\eta \to 1$ ($X \to X^{(0)}$), on the nature of the critical phenomena near the temperature of the PM-SG phase transition. As we mentioned above, in the limit $n \approx 0$ most SGs can be assigned to a single universality class, regardless of the type of competing exchange interaction.[4] This circumstance indicates similarities in the nature of the critical phenomena which developed near the phase-transition temperature T_c. An increase in the ferromagnetic-exchange component of the overall exchange energy ($\eta \to 1, X \to X^{(0)}$) leads to significant ferromagnetic critical fluctuations, in addition to the SG critical fluctuations, in the paramagnetic region[20] ($T \gtrsim T_c$). Clearly, even in the simplest case, in which the SG and ferromagnetic critical fluctuations do not interact with each other (strictly speaking, it is incorrect to assume that this is the case), the presence of these fluctuations should lead to an additive contribution to the linear and nonlinear responses of the system to a magnetization-reversal field. In other words, because of these factors, the increase in ferromagnetic exchange in the alloys should cause the critical exponents to deviate from the values characteristic of classical SGs; this is just what we see in practice (Table I). For alloys which are in the immediate vicinity of $X^{(0)}$, in contrast, e.g., the alloy $Fe_{56}Ni_{24}Cr_{20}$, the distortion of the PM-SG phase transition by critical ferromagnetic fluctuations is so pronounced that the conventional macroscopic methods used in this study to determine the critical exponents lead to physically meaningless results, although these methods are valid for SGs with $\eta \approx 0$.

TABLE I. Critical parameters of certain spin glasses.

Alloy	T_c, T_f, K	$\eta = 0/T_f$	β	ν	δ	$\delta = 1+\gamma/\beta$	Reference
Crystalline							
$Fe_{58}Ni_{22}Cr_{20}$	15.6	0.25	0.9	2.1	–	3.1	[4]
$Pd_{94}Mn_6$	3.5	0.90	0.9	2.0	3.0	–	[21]
$Pd_{95}Mn_5$	2.84	0.97	$\leqslant 0.3$	3.2	–	$\geqslant 11$	[22]
$Fe_{56}Ni_{24}Cr_{20}$	~ 21	~ 4	–	–	–	–	Present study
Amorphous							
$(Fe_{0,65}Mn_{0,35})_{75}P_{16}B_6Al_3$	42.2	0.49	0.76	2.05	3.8	3.7	Present study
$(Fe_{0,07}Ni_{0,93})_{77}B_{13}Si_{10}$	15.0	$\leqslant 1$	0.38	2.1	–	6.6	[4]
$(Fe_{0,15}Ni_{0,85})_{75}P_{16}B_6Al_3$	22.1	~ 4.8	0.38	3.4	11	–	[5]

In conclusion, we wish to stress that the approximation of noninteracting SG and ferromagnetic critical fluctuations is apparently not correct for an analysis of the real physical process which occur in alloys with $X \to X^{(0)}$ near the temperature of the PM-SG phase transition. This conclusion is supported by numerical calculations carried out in Ref. 23 for Ising SGs. Clearly, the interaction between the critical fluctuations in two (or, in general, several) order parameters must be taken into account in this problem.

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