Nonlinear ac response of spin glasses in a magnetic field

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The scaling hypothesis has been used to predict nonlinear ac properties of spin glasses in the external magnetic field. The experimental data prompt a method which allows us to determine lines where a parameter of the scaling function remains unaltered, and to estimate the critical temperature and three critical exponents, needed for a test of the scaling relation. The method gives the same results for both low and high applied magnetic-field values, even if the field is strong enough to suppress the spin-glass transition. The procedure is applied for the case of the amorphous $(Fe_{0.65}Mn_{0.35})_{75}P_{16}B_6Al_3$ alloy, which is a spin glass with the transition temperature $T_F \approx 42$ K. [S0163-1829(99)00618-9]

I. INTRODUCTION

Magnetic phase transitions in systems of spins mutually interacting through an Ising coupling with the coupling constants varying both in sign and amplitude have been investigated extensively. It was shown that if statistical distribution of the coupling constants J_{ij} can be approximated by Gaussian function with a maximum at J_0 and width J,[1] two low-temperature magnetic states are realized: the ferromagnet (FM) and the spin glass (SG). The SG state, in contrast to the FM state, has no spontaneous magnetic moment, $M_S = \langle\langle S\rangle_T\rangle_J = 0$, but nevertheless the Edwards and Anderson parameter $q = \langle\langle S\rangle_T^2\rangle_J$ is nonzero.

The FM state evolves from the paramagnetic (PM) state ($M_S = q = 0$) when the ratio $\eta = J_0/J$ exceeds unity. This phenomenon, which is well known as the PM-FM second-order[2–5] phase transition, may be qualitatively analyzed using the following static scaling law:

$$M_{FM}(H,\tau) = |\tau|^{\beta}\mathbf{F}_{\pm}(H/|\tau|^{\phi}), \tag{1}$$

where $F_{\pm}(x)$ is the scaling function [suffix $+(-)$ refers to the temperatures above (below) the critical temperature $T_C \sim J_0$], H is the applied magnetic field value, and $\tau = T/T_C - 1$ is the reduced temperature.

For a particular case $\eta < 1$, it is not yet clear whether the SG state is a result of the gradual freezing of spins[6] or a genuine phase transition at the nonzero freezing temperature $T_F \sim J$.[1,7–17] Numerous theoretical speculations[1,6–9] did not give an unambiguous answer, while some experimental data,[10–17] supporting the phase-transition concept, have been obtained. Also, it was found that scaling methods, developed earlier for empirical description of PM-FM (Ref. 2) phase transitions, gave consistent results when applied to the PM-SG transformation. Specifically, the scaling analysis confirmed that the scaling relation[7]

$$\chi_{SG}(H,\tau) = \chi(0,\tau) - |\tau|^{\beta'}\mathbf{F}_{\pm}(H^2/|\tau|^{\phi'}) \tag{2}$$

is valid in the case of a PM-SG transition and demonstrated a certain similarity between critical phenomena which accompanied both PM-SG and PM-FM transitions. For example, the scaling function $F_{\pm}(x)$ of the SG state[12,13] was restored in good agreement with the following approximations:

$$\lim_{x\to\infty} \mathbf{F}_{\pm}(x) \sim x^{1/\delta'},$$

$$\lim_{x\to 0} \mathbf{F}_{+}(x) \sim x,$$

$$\lim_{x\to 0} \mathbf{F}_{-}(x) = \text{const}, \tag{3}$$

which coincided with those in the FM's,[2,3] provided the scaling parameters were, respectively,

$$x = \begin{cases} H/|\tau|^{\phi}, & \text{(FM)} \\ H^2/|\tau|^{\phi'}, & \text{(SG)}. \end{cases} \tag{4}$$

However, a direct technique, which is supposed to be applied to derive $F_{\pm}(x)$, was often reduced to a separate estimation of critical exponents with the following test of their consistency. The common method, used to avoid determination of the unknown function $F_{\pm}(x)$, was usually based on assumption parameters (4) to be constants, each of them corresponded to a certain line on the experimental surfaces, $M = M(H,T)$, $\chi = \chi(H,T), \ldots$. Until recently it was known how to select only two of these lines, $x_0 = 0$ and x_0. Zero-field ($H_0 = 0$) behavior was studied for $x_0 = 0$ and critical isotherm ($\tau_0 = 0$) for $x_0 = \infty$. Now, a method to determine another line $0 < x_0 = \text{const} < \infty$ was proposed by Williams *et al.*[4] They studied linear FM susceptibility, which was assumed to obey the following scaling relation, provided $\phi = \beta\delta = \beta + \gamma^2$:

$$\frac{\partial M_{\rm FM}}{\partial H}(H,\tau) \equiv \chi_{\rm FM}(H,\tau) = |\tau|^{-\gamma}\mathbf{F}'_{\pm}(x), \tag{5a}$$

and demonstrated that the extrema of its temperature dependences at constant magnetic field $H_0 \neq 0$,

$$\left.\frac{\partial \chi_{\rm FM}}{\partial \tau}(H_0,\tau)\right|_{\tau=\tau_m} = -2\phi|\tau_m|^{-\gamma-1}\left[\frac{\gamma}{\phi}\mathbf{F}'_{\pm}(x_{0\tau}) + x_{0\tau}\mathbf{F}''_{\pm}(x_{0\tau})\right] = 0$$

corresponded to the same $x_{0\tau}$'s value. Proceeding from evident similarity (3) between the FM and SG scaling functions, we suggested that similar extrema should also be expected for the nonlinear SG response

$$\frac{\partial \chi_{\rm SG}}{\partial H}(H,\tau) \equiv \chi_2(H,\tau) = -2H|\tau|^{-\gamma'}\mathbf{F}'_{\pm}(x) \tag{5b}$$

Here $\chi_k(H,\tau)$ and $F^{(k)}_{\pm}(x)$, respectively, stand for $\partial^{(k)}M_{\rm SG}(H,\tau)/\partial H^{(k)}$ and $d^{(k)}F_{\pm}(x)/dx^{(k)}$. Although the experimental evidence for this effect could shed some light onto the problem of the PM to SG transition, until now no thorough study was performed. The previous $\chi_2(H,\tau)$'s data[14] were too incomplete for the predicted extrema to be revealed.

Assuming the scaling hypothesis (2) to be valid, we carried out an experimental research of nonlinear phenomena in SG's in magnetic field. Analysis of the experimental data resulted in a method for separate estimation of three critical exponents and the freezing temperature. The method was applied to the case of $(Fe_{0.65}Mn_{0.35})_{75}P_{16}B_6Al_3$ amorphous alloy, which is a SG with $T_F \approx 42$ K.[18]

II. EXPERIMENTAL DETAILS

Linear and nonlinear susceptibilities were measured by the standard mutual-inductance technique. The coil setup included an exciting coil, used to induce the ac component ($h=6$ Oe, $f=\omega/2\pi=75$ Hz), and a Helholtz pair, which supplied the dc component $H_{\rm dc}\leqslant 200$ Oe of the total magnetic field $H=H_{\rm dc}+h\sin(\omega t)$, as well as an astatic pair of equivalent secondary pickup coils. The coils were wound around the same axis and registered the *longitudinal* (parallel to the magnetic-field direction) response of the studied sample. Since the magnetization near an arbitrary point $(H_{\rm dc},\tau)$ may be presented as a Taylor's series:

$$M(H,\tau) = M(H_{\rm dc},\tau) + \sum_{k=1}^{\infty}\chi_k(H_{\rm dc},\tau)(H-H_{\rm dc})^k, \tag{6}$$

the differential output voltage, induced in the secondary coils after the sample was inserted, equals

$$E = -A\frac{d\Phi}{dt} \sim A\frac{dM}{dt}(H,\tau,t) \sim \omega A\sum_{k=1}^{\infty}\Theta_k(H_{\rm dc},\tau)\sin[k\omega t+\varphi_k], \tag{7}$$

where A is a numerical factor, which depends on the dimension of the coils and the filling factor, Φ is the magnetic flux, φ_k are the phase shifts due to the magnetic losses. If the exciting amplitude h is small enough, the kth harmonic signals

$$\Theta_1 = h^1\times\left[\chi_1+\frac{3}{4}\chi_3h^2+\frac{5}{8}\chi_5h^4+\cdots\right],$$
$$\Theta_2 = h^2\times\left[\frac{1}{2}\chi_2+\frac{1}{2}\chi_4h^2+\frac{15}{32}\chi_6h^4+\cdots\right],$$
$$\Theta_3 = h^3\times\left[\frac{1}{4}\chi_3+\frac{5}{16}\chi_5h^2+\cdots\right],$$
$$\Theta_4 = h^4\times\left[\frac{1}{8}\chi_4+\frac{3}{16}\chi_6h^2+\cdots\right],\ \text{etc.} \tag{8}$$

are equal (within the constant factor of $\sim h^k$) to the derivatives $\chi_k(H_{\rm dc},\tau)$. In our experiment the minimum amplitude of the exciting field h was limited by unharmonicity of our sine-wave generator. We had to increase the exciting field up to $h=6$ Oe, until the relative contribution of the high-order harmonics in the exciting ac field (i.e., the linear susceptibility $\Theta_1\sim h$, multiplied by the high-order harmonic content in the exciting signal $\sim$0.1%) to the nonlinear signals $\Theta_k \sim h^k$, $k=2,3,\ldots$ was reduced to 2%. On the other hand, nonzero amplitude of h results in an error, which is caused by high-order terms in brackets in Eqs. (8). We will demonstrate that this error disappears, as the dc magnetic field H and/or the reduced temperature τ (see the Appendix) become large enough.

Amorphous $(Fe_{0.65}Mn_{0.35})_{75}P_{16}B_6Al_3$ alloy was prepared by the melt-spinning technique and was shaped as a long thin ($\sim$ 20 μm) flexible ribbon. Its amorphous nature was confirmed by x-ray diffraction. The ribbon was cut and packed into a sample with a mass of 0.215 g.

III. RESULTS

Since both PM's and SG's have no spontaneous magnetic moment, their magnetization (6) must exhibit an inverse symmetry $M(H,\tau)=-M(-H,\tau)$. Thus, the first $M(H,\tau)$'s derivative with respect to H, i.e., linear susceptibility $\chi_1(H,\tau)$, is independent on inversion $H\Leftrightarrow -H$. Each further differentiation alternates these types of symmetry:

$$\chi_k(-H,\tau) = (-1)^{k+1}\chi_k(H,\tau). \tag{9}$$

It is worth to note that the odd harmonics $\Theta_{2m-1}(H,\tau)$, $m=1,2\ldots$ contain only the terms with odd $\chi_{2n-1}(H,\tau)$, $n\geqslant m$ derivatives [see Eq. (8)], whereas the even harmonics $\Theta_{2m}(H,\tau)$ consist only of even $\chi_{2n}(H,\tau)$, $n\geqslant m$ ones.[19] So, Eq. (9) is also valid for the experimental signals $\Theta_k(H,\tau)$, and we will, hereafter, refer to the absolute values of the dc magnetic field H. It is also obvious why even $\Theta_{2k}(H=0,\tau)$ harmonics must be absent in the SG state.[20] When $H\neq 0$,[21] analysis of nonlinear properties of SG's requires in this work that we use the scaling hypothesis (2) for this purpose. In the simplest case of the second harmonics, χ_2, we take Eq. (5b) as a starting point.

A. Scaling analysis (H_0=const)

Except for the singular point (0,0), Eq. (5b) confirms the previous result $\chi_2(0,\tau)=0$. To estimate the $\chi_2(0,0)$ value one needs:

(i) to dissect the $\chi_2(H,\tau)$ surface on isofield ($H_0 = \text{const}$) curves;

(ii) to find whether extrema, similar to those in FM's,[4] do exist;

(iii) if the latter is valid, to establish the relation between the applied magnetic field H_0 and the extremum value $\chi_2(H_0,\tau_m)$;

(iv) to determine the $\chi_2(0,0)$'s value by extrapolating $H_0 \to 0$.

Since only experimental study may answer (ii) (while the other issues can be resolved mathematically), let us suggest *a priori* that the extrema mentioned in (ii) exist. Proceeding from this assumption, namely

$$\frac{\partial\chi_2}{\partial\tau}(H_0,\tau)\bigg|_{\tau=\tau_m} = 2\phi' H_0|\tau_m|^{-\gamma'-1}\left[\frac{\gamma'}{\phi'}F'_\pm(x_{0\tau}) + x_{0\tau}F''_\pm(x_{0\tau})\right] = 0, \tag{10}$$

one has the crossover line(s) of constant scaling parameter(s)

$$\chi_{0\tau} = H_0^2/|\tau_m|^{\phi'} = \text{const}. \tag{11}$$

Then, the extremum amplitudes are described by the usual power dependences

$$\chi_2(H_0,\tau_m) = -2H_0|\tau_m|^{-\gamma'}F'_\pm(x_{0\tau}) \sim H_0^{2/\delta'-1} \sim |\tau_m|^{\beta'-\phi'/2}. \tag{12}$$

Therefore, to find the $\chi_2(0,0)$ quantity correctly, it is important to consider all possible cases

$$\delta' < 2, \tag{13a}$$

$$\delta' = 2, \tag{13b}$$

$$\delta' > 2, \tag{13c}$$

and select the suitable one(s). This may, for example, be performed with a reasonable condition

$$\chi_2(H_0,\tau_m)|_{H_0\to\infty} \to 0, \tag{14}$$

that, at once, disagrees with the assumption (13a). The mean-field exponent $\delta' = 2$ (Ref. 10) also looks unreasonable. In this case, $\chi_2(H_0,\tau_m)$ becomes independent of H_0, which contradicts either the restriction (14) or the experimental evidence for the nonzero $\chi_2(H_0 = 10\,\text{Oe},\tau)$ signal.[14] As a result, the inequality (13c) remains the only suitable approximation that, by the way, was found to be valid in several real SG's.[10–17] Hence, as the magnetic field decreases, the $\chi_2(H_0,\tau_m)$ amplitude approaches infinity and so does the second-harmonic value $\chi_2(H\to 0,\tau\to 0)$.

B. Scaling analysis ($\tau_0 = \text{const}$)

Criterion (10), introduced in earlier works of Williams *et al.*,[4] seems to be the only one of a few methods to register the lines of constant scaling parameters. But the larger the number of these lines, the more accurate the future experimental values of critical exponents and the freezing temperature T_F. So, we propose an alternative method. Let us consider the isotherms $\chi_2(H,\tau_0)$. Following the procedure (i)–(iv)

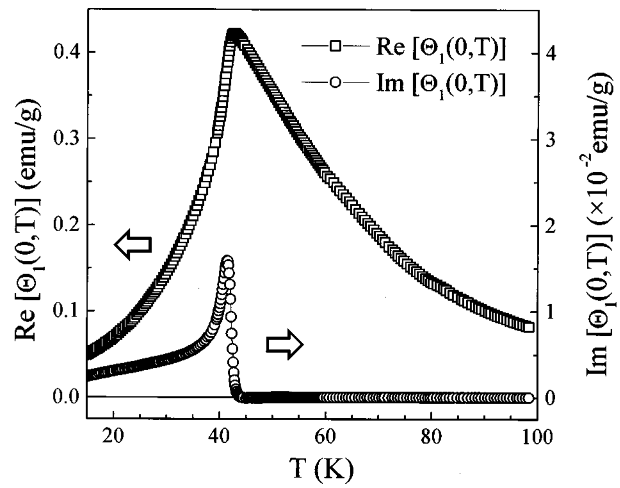

FIG. 1. The in-phase $\text{Re}[\Theta_1(H,T)]$ and out-of-phase $\text{Im}[\Theta_1(H,T)]$ components of the linear ac susceptibility measured in the amorphous $(Fe_{0.65}Mn_{0.35})_{75}P_{16}B_6Al_3$ alloy without an external magnetic field. The amplitude of the exciting field is $h = 2$ Oe.

$$\frac{\partial\chi_2}{\partial H}(H,\tau_0)\bigg|_{H=H_m} = -4|\tau_0|^{-\gamma'}\left[\frac{1}{2}F'_\pm(x_{0H}) + x_{0H}F''_\pm(x_{0H})\right] = 0, \tag{15}$$

we also obtain the crossover line(s)

$$x_{0H} = H_m^2/|\tau_0|^{\phi'} = \text{const}, \tag{16}$$

different from the previous one(s) Eq. (11) [compare the factors of $F'_\pm(x)$ in the brackets of Eqs. (10) and (15)], but certainly described by the same asymptotic dependences

$$\chi_2(H_m,\tau_0) \sim H_m^{2/\delta'-1} \sim |\tau_0|^{\beta'-\phi'/2}. \tag{17}$$

At last, noticing that $\partial\chi_2/\partial H$ definitely equals χ_3 and, therefore, may be measured by directly detecting the total ac response (7) at the triple exciting frequency, one can formulate more general criterion. Among an infinite number of lines $0 < x_0 = \text{const} < \infty$, which constitutes the experimentally accessible surface $\chi_k(H,T)$, $k = 2,3,\ldots$ this criterion prompts us to select those, if any, where either $\chi_k(H,T)$ itself or any of its partial derivatives equals zero.

C. Experimental results

To confirm our concept, the amorphous $(Fe_{0.65}Mn_{0.35})_{75}P_{16}B_6Al_3$ alloy has been studied as an example of a typical SG. The in-phase $\text{Re}[\Theta_1(0,T)]$ and out-of-phase $\text{Im}[\Theta''_1(0,T)]$ components of its linear ac susceptibility (Fig. 1) agree well with ones in materials undergoing the PM to SG phase transition.[10,11] The freezing point T_F can be approximately taken as a temperature (in our case ≈42.5 K), at which the susceptibility real part $\text{Re}[\Theta_1(0,T)]$ temperature dependence exhibits a cusp.

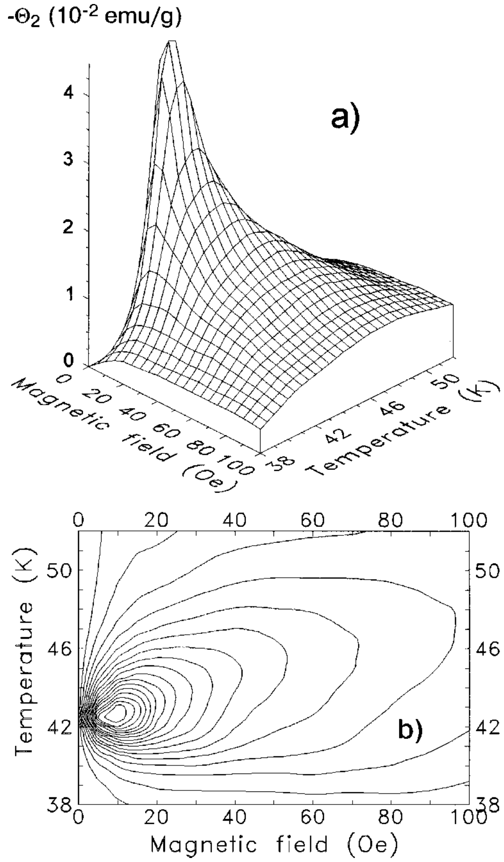

FIG. 2. The nonlinear $\Theta_2(H,T)$ susceptibility represented as (a) perspective $3d$ plot and (b) contour curves. The amplitude of the exciting field is $h=6$ Oe.

The nonlinear susceptibility $\Theta_2(H_0\neq 0,T)$ vs T dependences have been measured after the sample was field cooled (FC) from a temperature well above T_F. These data are summarized in Fig. 2 as (a) $3d$ plot and (b) contour curves. Then, following the proposed criterion, let us dissect the presented surface onto a certain number of isofields $\Theta_2(H_0,T)$, selected with the step $\Delta H=5$ Oe, and isotherms $\Theta_2(H,T_0)$ ($\Delta T=0.5$ K) and consider the curves connecting their extrema. Since $\Theta_2(H,T_0)$ dependence defines isotherms both above ($\tau>0$) and below ($\tau<0$) the critical temperature, two solutions are possible Eq. (16). Each of them satisfies Eq. (15), provided the proper branch of the scaling function, $F_+(x)$ or $F_-(x)$, is used. The curve $\Theta_2(H_0,T)$ may be connected with the parameter (11). Since the extremal temperature T is increased with H_0 (see Fig. 2), this solution belongs to the upper branch $F_+(x)$. Projecting these crossover lines onto each of three mutually perpendicular planes, where H, τ or Θ_2 is of constant value, one can estimate three critical exponents and check their relationship.[22] Since for this procedure a method used to select these lines has no effect on the result, the subsymbols will be hereafter omitted.

At first, let us consider projections onto $\tau=$const plane and find δ'. When $H\gg h=6$ Oe, these projections are parallel (in log-log scale) lines with the slope $2/\delta'-1=-0.67\pm 0.01$, which corresponds to $\delta'=6.0\pm 0.3$ (Fig. 3). An evident departure from this behavior ($H\leqslant h$) may be caused by high-order terms in Eqs. (8). Fortunately, their relative con-

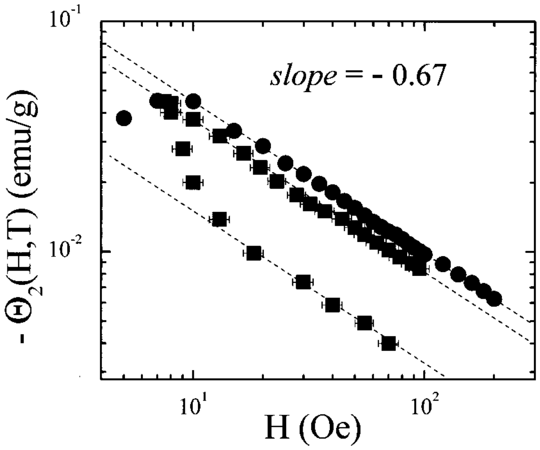

FIG. 3. Projections of crossover lines onto the plane $\tau=$const represented in the log-log scale. The $\Theta_2(H_0,T)$ vs T dependences' extrema are marked by circles. Squares denote the extrema of the $\Theta_2(H,T_0)$ vs H curves. The same slope$=2/\delta'-1\approx-0.67$ of the dotted lines corresponds to the critical exponent $\delta'=6.0$.

tribution diminishes as the magnetic field H is increased. At least, it noticeably improves ($\sim H^2$) the ratio of the useful signal

$$\chi_k(H,\tau)\sim H^{2/\delta'-(k-1)}\sim|\tau|^{\beta'-(k-1)-\phi'/2} \tag{18}$$

to the first term of the experimental error $\chi_{k+2}(H,\tau)$ (see the Appendix). Since along the studied lines $H^2\sim\tau^{\phi'}$, the same advantage is gained as the reduced temperature τ is increased. Therefore, critical indices, that describe temperature divergences, also have to be obtained from experimental points, which are far enough from the singularity (0,0). Besides, as compared with the previous log-log plot (see Fig. 3), the estimation of these indices sensitively depends on the critical temperature choice and usually requires predefined T_F. The exception is the Kouvel-Fisher technique,[2] which allows estimating both the exponent and T_F values simultaneously. In SG's, where the nonlinear response is too small for this approach to give reliable results, we propose another method that has the same advantage. Using the idea by Geschwind *et al.*,[16] let us rewrite Eqs. (11) and (17) to linearize τ

$$\tau=1-T/T_F\sim|\chi_2(H,\tau)|^{2/(2\beta'-\phi')}\sim H^{2/\phi'}. \tag{19}$$

If chosen properly, each of both exponents in Eq. (19), missing at this point to test the scaling relationship $\phi'=\beta\cdot\delta'$, should satisfy the following requirements:

(i) a valid exponent rescales three corresponding projections into straight lines.

(ii) three projections intersect the T axis at the same point $T=T_F$.

(iii) this point $T=T_F$ must surely coincide for both optimization procedures.

Because the mentioned error [see Eq. (18)] reaches its maximum near the singular point ($H=0,T_F$), the experimental data $\Theta_2(H,T)$ in the vicinity of the point were ignored. So, when separating exponents, we were guided by the requirements (ii),(iii). The best exponents and the freezing temperature were found to be $2/(2\beta'-\phi')=-0.95\pm 0.05$,

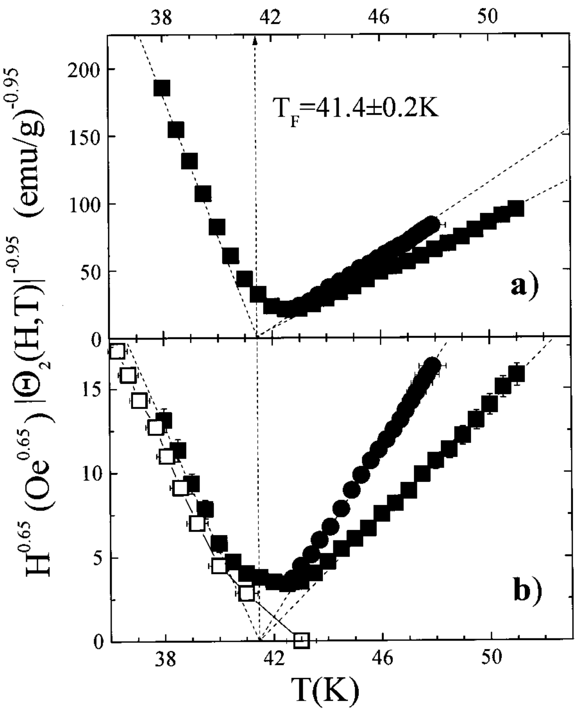

FIG. 4. Linearized projections of crossover lines onto the planes of equal (a) H's and (b) Θ_2's values. The critical freezing temperature and exponents are found to be $T_F = 41.4 \pm 0.2$ K, $2/(2\beta' - \phi') = -0.95 \pm 0.05$ and $2/\phi' = 0.65 \pm 0.05$, respectively. The symbols denoting the crossover lines are the same as in Fig. 3.

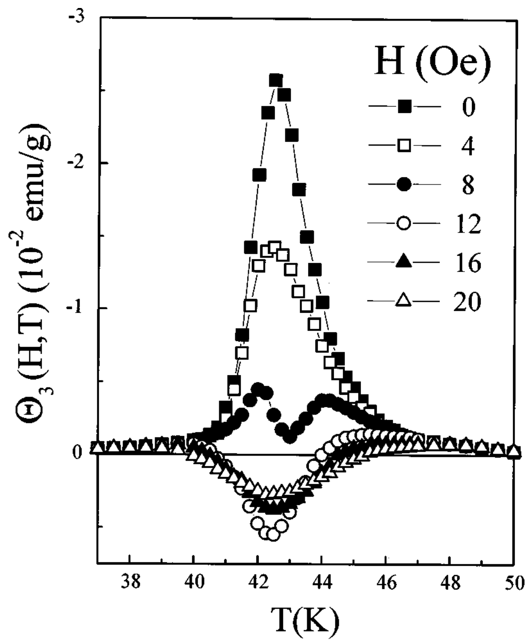

FIG. 5. Nonlinear $\Theta_3(H,T)$ susceptibility vs T dependences measured in various magnetic fields H. The amplitude of the exciting field is $h = 6$ Oe.

$2/\phi' = 0.65 \pm 0.05$, and $T_F = 41.4 \pm 0.2$ K (see Fig. 4). Since the obtained exponents are bound to each other by the relation $(2/\delta' - 1) = -0.67$, it is a convincing proof of the scaling assumption (2). Hence, the other scaling relations are acceptable to estimate the unknown indices: $\phi' = 3.08$, $\beta' = \phi'/\delta' = 0.49$, $\gamma' = \phi' - \beta' = 2.59$, etc.

We also consider another scaling prediction that can be easily checked by direct detecting the nonlinear SG response at triple (3ω) exciting frequency. As the squares in Fig. 4(b) restrict the sector where $\partial\chi_2/\partial H$ has an positive sign, negative $\chi_3(H,T)$ values, that are usually registered in zero-field measurements,[10–17] should be expected only outside this sector. Shown in Fig. 5, the experimental $\Theta_3(H,T)$ data strengthen this conclusion. Similar phenomena were observed in the dilute alloy Au 1.5 at.% Fe.[15]

IV. DISCUSSION

When the final results are obtained, it is worth answering the question: To what extent may they be relied on? One can name, at least, three reasons that cause some doubts. The first of them: the exciting field amplitude h in Eqs. (8) is presumed to be small enough, as it was already discussed above. This requirement sets a low-field boundary of (H,τ) range for reliable measurements. The other two are related to the scaling hypothesis (2) and exclude usage of too large τ and H values, respectively.

For instance, the static scaling law (2) is valid while the sample is in equilibrium. But it is well known that the SG magnetic state possesses a wide spectrum of relaxation times. At temperature low enough the relaxation time may exceed reasonable experiment time,[10] no matter whether one uses dc methods with the observation time $t_{obs} \sim 10^2$ s or ac techniques ($t_{obs} \sim \omega^{-1}$). In the former case the boundary (H,τ) line between equilibrium and nonequilibrium states is usually estimated from field-cooling–zero-field cooling (FC–ZFC) curves that deviate one from another just below this line.[11] For our case ($t_{obs} \sim 10^{-2}$ s) this approach seems inapplicable. We use an alternative method. To make sure that our observation time is more than SG relaxation time, we monitored magnetic losses, i.e., the phase angle φ_1 in Eq. (7), provided the exciting frequency ω remained the same and so did the t_{obs} value. Indeed, if the largest relaxation time is exceeded, SG's are assumed to reach the equilibrium state and $\varphi_1(H,\tau) = 0$. Otherwise, e.g., at low temperatures, large relaxation times create a nonzero phase delay between the exciting field and the sample's response. In the reported case, for example, at the liquid helium point $T = 4.2$ K the signal-response lag attained abnormally large values of $\varphi_1 \sim 4$ angle degrees. Results of our $\varphi_1(H,T)$ measurements are presented in Fig. 6. We have chosen the threshold criteria $\varphi_1 = 0.1$ [to avoid the overestimation of the equilibrium temperature $\varphi_1(H,T) = 0$ induced by the experimental error] and found that the equilibrium boundary depends on the applied dc magnetic field as the de Almeida–Thouless line[23]

$$\tau(H) = \frac{T_F(0) - T_F(H)}{T_F(0)} = \left[\frac{H}{H_{AT}}\right]^{2/\phi'}, \qquad (20)$$

where the *longitudinal* spin components are presumed frozen. It allows us to include these data in Fig. 4(b) (open squares) and to show directly that all crossover lines are in an equilibrium region.

Equation (20) demonstrates the requirement for the applied dc magnetic field to be small enough. It is resulted from the scaling assumption (2), which states that the temperature $T_F(H)$ remains constant. Fortunately, the deviation

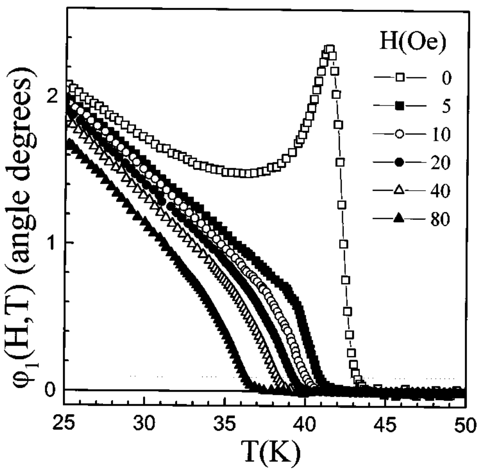

FIG. 6. Temperature dependences of the phase angle φ_1 measured in various magnetic fields H.

$\tau(H)$, if any, reveals the same asymptotic behavior as the crossover lines (11),(16). In other words, within the proposed in Fig. 4(b) approach it changes the crossover line slopes only, but has no influence on the exponent $2/\phi'=0.65\pm0.05$ as well as the temperature $T_F(H=0)=41.4\pm0.2$ K. Since the method to extract another exponent, $\delta'=6.0\pm0.3$, is also independent of whether $T_F(H)$ is constant or not, the obtained indices are more reliable than their values estimated by the best-fit procedure for the scaling function $F(x)$,[24,25] when T_F is defined as constant. Moreover, to show this difference, we *a priori* selected the sample $(Fe_{0.65}Mn_{0.35})_{75}P_{16}B_6Al_3$ that is close ($\eta\approx0.5$) to the percolation threshold $\eta=1$,[18] and is more sensitive, $H_{AT}\sim(1-\eta)$,[23] to an external magnetic field H. But the exponents appear nearly the same—$\delta'=6.4\pm0.4, \phi'=3.0\pm0.1$ (see Fig. 7). This means that $\tau(H=100\,\text{Oe})\times T_F$ barely exceeds the double experimental error for T_F, i.e., $2\times0.2\,\text{K}=0.4\,\text{K}$, and this fact allows us to estimate the lowest limit for the denominator $H_{AT}\geqslant10$ T. In contrast to the experimental value $H_{AT}=0.19\pm0.02$ T, calculated from the magnetic losses data (see Fig. 6), this limit well agrees with the theoretical prediction $H_{AT}=2k_BT_F(1-\eta)/(\mu_B\sqrt{5})\approx20$ T.[23]

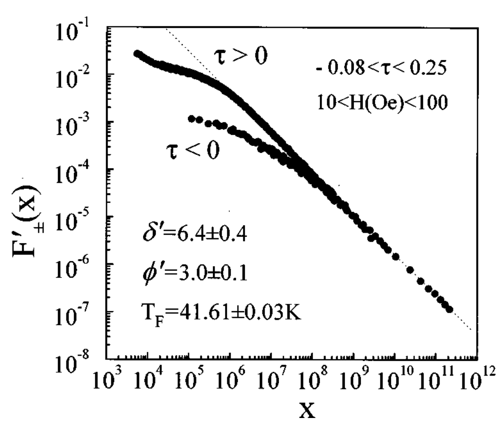

FIG. 7. Scaling function $F'_{\pm}(x)$. The slope$=1/\delta'-1\approx-0.84$ of the dotted line is consistent with the critical exponent $\delta'=6.4$.

V. CONCLUDING REMARKS

The experimental methods, used to check the scaling assumption, may be roughly discerned by techniques the authors use to resolve the problem of unknown scaling function $F_{\pm}(x)$. Compared with direct calculations of $F_{\pm}(x)$, the test of the scaling relationships seems somewhat indirect method, but no less convincing. Moreover, the exponents, which yield an optimum data collapse, strongly depend upon the (H,τ) range where the fit is performed.[24] Since in magnetic materials near the percolation threshold $\eta=1$ this range shrinks dramatically,[25] our approach looks preferable for such cases.

Until a recent work by Williams *et al.*[4] the indices were usually extracted from the critical isotherm ($\tau=0$) and the zero-field ($H=0$) behavior. So, it was known how to select only two, $x_0=0$ and $x_0=\infty$, of an infinite number of crossover lines forming the surface. Unfortunately, each of these lines was described by a single exponent, i.e., none of them included information about all three critical indices, which are needed to check the scaling relationship. The lines with $0<x_0<\infty$ are free of this fault. Although these lines was discovered and studied in FM's, we expanded this concept for SG's (and, in general terms, for any system where the scaling hypothesis can be applied).

APPENDIX

Here we roughly estimate the dc biasing magnetic field H that is needed to suppress the high-order terms in Eqs. (8). Let us consider the equation that defines the second-harmonic amplitude Θ_2, and require for the first term of the total experimental error $[\chi_4h^2/2+15\chi_6h^4/32+\cdots]$, normalized to the useful signal $\chi_2/2$, to be less than ε, where $\varepsilon\ll1$. In this case, the second derivative of Eq. (5b)

$$\chi_4=\frac{d^2\chi_2}{dH^2}=-\frac{4H}{\tau^{\phi'+\gamma'}}[3F''(x)+2xF'''(x)]$$

should satisfy the condition

$$\frac{\chi_4}{\chi_2}h^2=\frac{2x[3F''(x)+2xF'''(x)]}{F'(x)}\left(\frac{h}{H}\right)^2<\varepsilon.$$

Along the line $x\to\infty$ (i.e., the critical isotherm $\tau=0$), where the scaling function has the well-known form $F(x)\sim x^{1/\delta'}$, this condition may be noticeably simplified

$$\frac{H}{h}>\sqrt{\frac{2}{\varepsilon}\frac{(\delta'-1)(\delta'-2)}{(\delta')^2}}.$$

Considering our case ($h=6$ Oe, $\delta'=6.0$) and assuming an acceptable error to be $\varepsilon=5\%$, one can easily obtain: $H>30$ Oe. With H's amplitude constant, the experimental accuracy along the other lines (including the ones in Fig. 3) turns out to be even better, since the ratio χ_4h^2/χ_2 is partly suppressed ($\sim\tau^{\phi'}$) by the reduced temperature τ. So, the smaller the scaling parameter x_0, responsible for a crossover line, the larger the distance $\tau=[H^2/x_0]^{1/\phi'}$ between this line and the critical isotherm [see Fig. 4(b)], the lower seems to be the admissible magnetic field H (Fig. 3).

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[19] When Levy (Ref. 14) expanded $M(H,\tau)$ around the zero magnetic field as the powers of $[H_{\mathrm{dc}}+h\sin(\omega t)]^k$, the experimental amplitudes $\Theta_k(H,\tau)$ included all the terms with $\chi_n(H,\tau), n \geqslant k$. However, this result looks questionable. As soon as H_{dc} becomes different from zero, his expansion loses an accuracy that strongly depends on whether the parameter $[H_{\mathrm{dc}}+h\sin(\omega t)]^k$ is small or not.
[20] Since FM's possess the spontaneous magnetic moment, neither their linear susceptibility $\chi_{\mathrm{FM}}(H,\tau)$ nor nonlinear responses have any symmetry with respect to an inversion $H \Leftrightarrow -H$. Registered without a magnetic field (Ref. 5) the nonzero signal of the second harmonic confirms this assumption.
[21] Levy ungroundly suggested that the even derivatives $\chi_{2m}(H=0,\tau), m=1,2,\ldots$ must be absent in an arbitrary magnetic field $\chi_{2m}(H,\tau)\equiv 0$ (Ref. 14). Despite its complete agreement with Eq. (9) this requirement is excessive. In fact, since χ_{k+1} definitely equals $\partial\chi_k/\partial H$, his assumption also proscribes the odd nonlinear responses, i.e., $\chi_k(H,\tau)\equiv 0$ $(k \geqslant 2)$.
[22] Since any projection onto the last of three mutually perpendicular planes is completely determined with help of the previous two and thereby so is the third exponent, confirming the scaling assumption (2) does not require us to estimate all three critical indices as reported in this paper.
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