Critical dynamics of metallic spin glasses

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We have measured the ac nonlinear susceptibilities χ_3', χ_5', and χ_7' of very dilute *Ag*Mn spin glasses above and below the transition temperature T_g as a function of frequency, temperature, and magnetic field. In the static limit, these quantities display well-defined critical singularities at T_g and imply a divergence of the spin-glass correlation length. The nonlinear susceptibilities can be fitted above T_g to powers of frequency with weakly temperature-dependent exponents. While the effective exponents satisfy the relations imposed by static and dynamic scaling, the observed temperature and frequency roundings suggest that the spin-glass correlation length does not actually become infinite and is cut off at a length scale of 2000 Å. Activated behavior in the vicinity of T_g with a logarithmic dynamic scaling also accounts for the measured dynamical nonlinear susceptibilities. The measured critical exponents ($\beta=0.9$, $\gamma=2.3$, $\delta=3.3$, $\nu=1.3$, $z=5.4$) are not mean field, and differ from the exponents obtained by Monte Carlo simulation of short-range Ising systems.

I. INTRODUCTION

In metallic spin glasses, transition-metal impurities such as Mn are diluted in a noble metal such as Cu or Ag. Their magnetic interaction which is mediated by conduction electrons oscillates rapidly [$\cos(2k_FR)$] as a function of their separation R. Since the impurities are randomly distributed through the host lattice, their effective exchange interaction is random. At the simplest level this interaction is described by the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction[1] which falls off as $1/R^3$. The effective interaction between the Mn ($S=\frac{5}{2}$) impurities is then described by a Heisenberg Hamiltonian with random exchange.

The dependence of the susceptibility of these alloys with temperature exhibits a sharp cusp (Fig. 1) at a temperature T_g, which has been associated in previous studies, either with a gradual freezing of the spins or a genuine phase transition. This question cannot easily be resolved by theoretical considerations[2] since Heisenberg spin glasses are believed to be either below or at their lower critical dimension in $d=3$ (d is the space dimension).[3] This has been confirmed by Monte Carlo studies of isotropic RKKY spin glasses in $d=3$, which have failed, in the absence of anisotropic interactions, to observe any phase transition at a nonzero temperature.[4] These arguments suggest that the existence of a phase transition in Heisenberg systems may be very sensitive to the range of interaction or to the presence of weaker anisotropic forces. In $d=3$, the $1/R^3$ decay of the RKKY law produces a "volume invariant" interaction, which has been argued to lead to a marginal critical behavior.[5] Also, Monte Carlo simulations of Heisenberg systems have shown that the addition of dipolar (anisotropic) interactions was sufficient to give a phase transition at a nonzero temperature.[4] In analogy with ferromagnets, it has been argued that anisotropic forces can lead to an Ising-like behavior "close enough" to T_g.[6] At present, Monte Carlo simulations[7] and mean-fields theories[8] can definitively conclude that short- and long-range Ising systems have a thermodynamic phase transition. They are not sufficient to say with certainty whether (i) Heisenberg spin glasses have a phase transition, (ii) what amount of anisotropy is necessary to produce an Ising behavior, (iii) how can critical universality classes of spin glasses be defined.

In light of this discussion, it is important to characterize as accurately as possible the interaction between impurities in metallic spin glasses. First, we note that the spin coupling between the local Mn moments and the conduction-electron spins occur by s-d exchange with the d electrons of the local moments. The $1/R^3$ decay of the

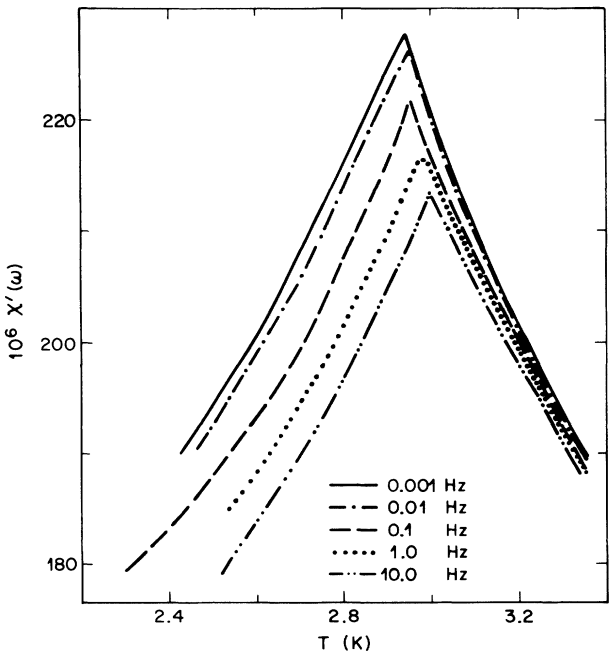

FIG. 1. Frequency dependence of χ_1' above and below T_g. Below T_g, the decrease of χ_1' is nearly logarithmic at frequencies above 10^{-2} Hz.

RKKY interaction is obtained within the free-electron model by taking the s-d mixing as a zero-range contact interaction. In reality, this coupling is extended over the ionic potential. This introduces a preasymptotic deviation from the $1/R^3$ law,[9] which is anisotropic and reflects the shape of the Fermi surface. According to a recent calculation the asymptotic regime is reached at distances of the order of 60 Å in *Cu*Mn and 35 Å in *Ag*Mn.[10] In alloys with concentration of magnetic impurities larger than 10^{-4}, the average distance between local moments always falls within this range and the deviations from the simple RKKY behavior are relevant.[11] In addition, manganese impurities are known to "anticluster" (i.e., to preferentially locate as second neighbors where they interact ferromagnetically rather than first where they are antiferromagnetic).[12] This short-range atomic order can bias the distribution of exchange as can be inferred from the ratio of the Curie-Weiss temperature Θ to T_g. As shown in Fig. 2,[13] this ratio becomes small and slightly negative for Mn concentrations below 1 at.%, indicating a good balance between ferromagnetic and antiferromagnetic interactions. A short-range ($\sim$4 lattice constants) magnetic oscillation in the spin density with a wave vector $\sim 2k_F$ has also been observed in neutron scattering experiments.[14] Since there is local atomic order and since the RKKY interaction introduces an anisotropic modulation of the electronic spin density at wave vector $2k_F$, this short-range magnetic order may be incidental to the atomic short-range order, but other views exist in the literature.[15] In any event, the existence of short-range order is not expected to affect significantly the spin-glass state as long as a spin-glass correlation can develop on much longer length scales. The main thrust of this paper is to demonstrate that this is indeed the case. On general grounds, the asymptotic $1/R^3$ behavior of the RKKY interaction will determine the critical regime when the correlation length becomes sufficiently large.

Experimental evidences for anisotropies in metallic spin glasses have been observed in torque (Ref. 16), NMR (Ref. 17), and ESR (Ref. 18) experiments. They arise from the spin-orbit coupling of the conduction electrons with the magnetic impurities, giving rise to a random anisotropy (no preferred axis) well described by the Dzyaloshinskii-Moriya Hamiltonian[19] $D_{ij}\cdot S_i \times S_j$, where the D_{ij} are random variables with variance D. The magnitude of D is known from ESR measurements to be 3.6×10^{-2} (*Cu*Mn) and 9.5×10^{-2} (*Ag*Mn) times smaller than the exchange. Some experimental studies of the nonlinear susceptibilities of metallic spin glass with gold doping (which alters the anisotropy constant D and to a lesser extent the exchange J) have made the relevance of anisotropy forces to the critical behavior plausible.[20] Since these studies were carried out far from T_g and with rather large fields, their conclusions remain uncertain.

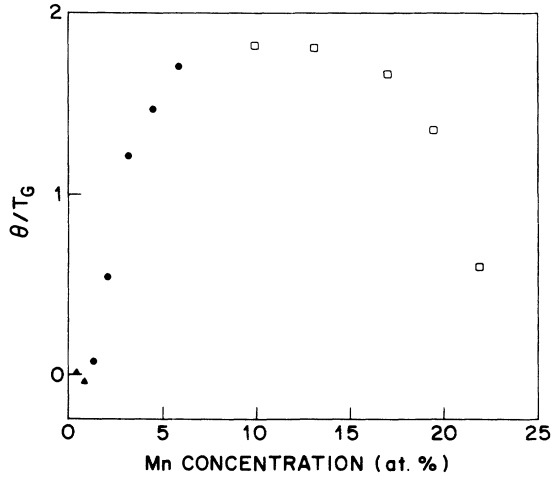

FIG. 2. Plot of the ratio of the Curie-Weiss temperature Θ to T_g as a function of Mn concentration. The data are taken from Ref. 13.

The signature of a phase transition is a singularity of the free energy. For spin glasses, this singularity should be manifest in the divergence of the nonlinear susceptibility $\chi_{SG}=\partial^3 M/\partial H^3$.[21] The nonlinear susceptibility χ_{SG} is, in fact, related to $\partial^2 q/\partial H^2$ where $q=[\langle S_i\rangle^2]_{av}$ is the spin-glass order parameter (q is the variance of the local magnetization). We note the analogy with ferromagnets (where the order parameter is the magnetization), for which the susceptibility $\chi=\partial M/\partial H$ diverges at the Curie temperature.

Since the experimental evidence for a phase transition is based on the observation of this divergence, we question to what extent it is unique to spin glasses. In an antiferromagnet, there is an exact cancellation of the molecular field at each site, and consequently an external magnetic field cannot couple to the magnetization. However, if diluted, this argument does not hold and a nonlinear magnetization appears in an external field. It is known that this nonlinear susceptibility also diverges at the Néel temperature. Since antiferromagnetic tendencies are known to exist below 1 at.% Mn concentration, the interpretation of a divergent nonlinear susceptibility at T_g in terms of genuine spin-glass transition requires more care. We can first rely on Monte Carlo simulation of dilute antiferromagnets to argue that the critical exponent associated with this divergence must be quite small, since the increase of χ_3 appears to be weak.[22] In fact, we argue[23] in Sec. IV that the critical exponent for the divergence of χ_3 should be $\gamma_{AF}-2\beta_{AF}$ in a dilute antiferromagnet, where β_{AF} and γ_{AF} are, respectively, the critical exponents of the staggered magnetization and susceptibility of the pure system. In the Ising case, it is approximately 0.6.

To explore the behavior of spin glasses in the vicinity of T_g, experiments have to be able to isolate any static critical properties from dynamical effects. Since there is now good evidence that the relaxation times become very long in the spin-glass phase, the only way to know how far the system is from static equilibrium is to perform a dynamical study at ultralow frequencies and see whether the system reaches a static limit as the frequency is lowered.

We have carried out very systematically such a study on the linear and the nonlinear susceptibilities.[24] This study shows unambiguously the following. (i) A divergence of the spin-glass correlation length as $T\rightarrow T_g$, which is characteristic of a phase transition. (ii) Power-law divergences of all the nonlinear susceptibilities with reduced temperature $[\tau=(T-T_g)/T_g]$ consistent with the static

scaling hypothesis; however, some temperature and frequency rounding of the divergences suggest that the spin-glass correlation length does not actually become infinite but is cutoff at a length scale of order 0.2 μm. (iii) The dynamics in the vicinity of the transition can be as well described within the dynamic scaling hypothesis, which implies critical slowing down as with a logarithmic scaling that implies activated dynamics close to T_g. (iv) Values of the critical exponents that differ from the value obtained in mean field and in the short-range Ising model.

This discrepancy between the critical exponents measured in metallic spin glasses and in numerical simulations of short-range Ising systems[7] can be interpreted in several ways. The most obvious interpretation is that RKKY metallic spin glasses do not belong to the same critical universality class as short-range Ising systems. The range of interaction or the weakly Heisenberg character can give rise to a crossover region that would account for the data. Another alternative is that the concept of critical universality classes developed for pure and dilute systems are not applicable to random magnets. It has been conjuctured[25] that the entire distribution of random exchange constants (which is altered by dilution) should be used to define universality instead of the variance alone. This issue has been addressed in numerical simulation.[26] The results are roughly consistent with the Harris criterion (i.e., the distribution of exchange constants is not relevant as long as α, the critical exponent for the specific heat, is negative) although the statistics remain poor. The identification of the universality class of the spin glasses remains therefore largely unresolved both experimentally and theoretically.

The paper is organized as follows. In Sec. II we show that some knowledge of the higher-order correlation functions can be extracted from nonlinear susceptibilities. This result is relevant to spin glasses, since there are no direct experimental methods to measure the spin-glass order parameter and its associated correlation functions. The experimental techniques used to extract the various nonlinear susceptibilities are described in Sec. III. The static limit is analyzed in Sec. IV, testing the scaling hypothesis. The data obtained in a magnetic field are also presented. The dynamical measurements are analyzed in Sec. V for linear and nonlinear processes in the framework of dynamical scaling as well as logarithmic scaling. Section VI deals with some of the observation below T_g, and we close the paper by discussing the implications of this study.

II. SUSCEPTIBILITIES AND CORRELATION FUNCTIONS

The presence of 11 harmonics in the Fourier transform of the magnetization response (Fig. 3) indicates the growth of multispin spin-glass correlation functions. It is known that the correlation function $C_q(r,t) = \langle q(r,t)q(0)\rangle$ of the spin-glass order parameter can be related to four-spin correlation functions. For most issues associated with the spin-glass state, a knowledge of the correlation length ξ on which $C_q(r,t)$ decays is essential. Since there are no direct ways to measure ξ, valuable information can be gained from the experimentally accessible nonlinear susceptibilities. Indeed χ_3 is also related to four-spin correlation functions and is expected to be closely related to C_q. There are two ways to infer the value of ξ from measurements of χ_3. The first one relies on the experimental validity of scaling (see Sec. IV) for singular thermodynamic quantities such as χ_3, in the vicinity of the transition temperature T_g. With this hypothesis, ξ scale close to T_g as

$$\frac{\xi}{l_0} \approx x \left(\frac{\chi_3}{\chi_1}\right)^{\gamma/\nu} \left(\frac{H}{T_g}\right)^2 , \tag{1}$$

where γ and ν are, respectively, the critical exponent for the nonlinear susceptibility and the correlation length. In Eq. (1), l_0 is the average distance between spins while H is the strength of the external field in temperature units. These factors are introduced so the critical amplitude x is close to unity.

Another way to relate C_q to χ_3 is based on the extension of the fluctuation-dissipation theorem to nonlinear response and does not rely on scaling. It has been shown recently[27] that a lower bound on the spectrum of higher-order equilibrium fluctuation can be set with the imaginary part of χ_3. Specifically,

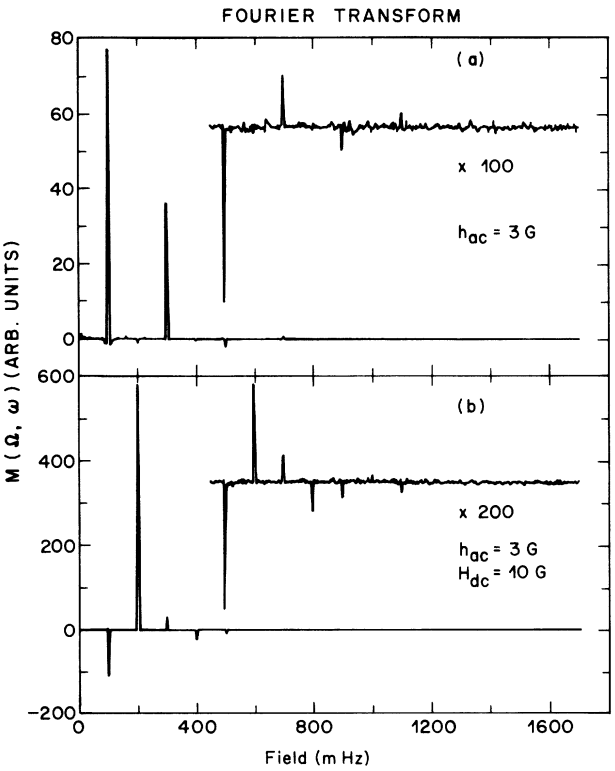

FIG. 3. (a) Fourier-transform spectrum of the magnetization response to a 0.1-Hz field at T_g. The intensity of the fundamental measures the linear susceptibility minus the fixed signal subtracted via the center tap of the modulation coil. The resultant amplitude is about -5×10^{-3} of the linear susceptibility. The magnified signal in the upper trace shows the fifth, seventh, ninth, and eleventh harmonics. (b) With a static field of 10 G, even harmonics are also observed.

$$\mathrm{Im}[\chi_3(\omega,0,\omega)+\chi_3(\omega,0,-\omega)] \leq (\beta\omega)^3/8\langle S(\omega)S(-\omega)S(\omega)S(-\omega)\rangle_c , \tag{2}$$

where $\langle\cdot\rangle_c$ is the connected part of the correlation function. The implication of this result to our work is that it is possible to interpret a divergence of χ_3'' directly in terms of a divergence of the spin-glass correlation length ξ, without relying on any scaling relations. This may prove to be essential for studies of the spin-glass phase.

III. EXPERIMENTAL PROCEDURES

All measurement were made by superconducting quantum interference device (SQUID) magnetometry. The gradiometer developed for this experiment allows the sweeping of large fields (up to 100 G) at fast rates (≥ 1 Kg/sec), making this instrument ideal for nonlinear dynamical studies. The flux transformer (see Fig. 4) consists of an astatic pair (2×7 turns) 9 mm in diameter wound on a thin quartz tube. Its total inductance ($L \sim 1.6$ μH) matches the inductance (~ 2 μH) of the input coil of the SHE model No. 330X SQUID probe giving a flux transformation ratio of $\sim \frac{1}{60}$ between the sample coil and the SQUID. The sample is mounted on a sapphire rod 8 mm in diameter and 1.5 cm long, mounted on a copper slug providing the thermal anchoring to a ^{3}He cryostat and a top loading system to interchange samples rapidly. The uniqueness of this instrument comes from the modulation coil producing the swept magnetic field. It is made of 90 turns of Nb-Ti superconducting wire 0.125 mm in diameter wound on a quartz tube 1.2 cm in diameter. It is accurately centered around the astatic pair and has in the middle a center tap through which a small current can be added to the main driving current. Its purpose is twofold: it enables compensation for the imperfect balance of the astatic pair (about one part in 200) and subtraction of a constant term from the signal. In this way, the contribution coming from the linear susceptibility of the sample can be eliminated. However, contributions arising from eddy currents in the sample which are at low frequencies in quadrature with the driving magnetic field cannot be compensated for. Since the instrument is phase sensitive, the eddy currents only distort the measurement of χ'' and χ_3''. We found it necessary to have very-low-resistance (<0.3 Ω) copper and superconducting leads to drive the modulation coils to eliminate spurious nonlinearities associated with nonlinear resistance. For fields below 5 G, we used as a current source an Hewlett-Packard (HP) model No. 3325 synthesizer with a very stable 50 Ω resistance in series. Later on, the synthesizer output was amplified with a high-current operational amplifier (supplied with large Ni-Cd batteries) to produce larger fields. The superconducting shields used in this experiment were either open cylinders (Nb or Sn were used) to trap a static magnetic field (up to 1.2 kG) or closed cylinders to expel the earth's magnetic field. The output of the SQUID is synchronously digitized by an HP voltmeter during an integer number of cycles (typically 20), and the Fourier transform with typically 1024 or 4086 points is computed by a micro-Vax minicomputer. The real part of the transform gives the in-phase response ($\propto\chi'$, χ_3', ...) of the magnetization (Fig. 3), while the imaginary part gives the out-of-phase response ($\propto\chi''$, χ_3''). Above T_g and in absence of magnetic field, this nonlinear response

$$M(\Omega,t)=\sum \theta_k' \cos(k\Omega t)+\theta_k'' \sin(k\Omega t)$$

contains only odd harmonics. The amplitudes θ_k' contain contributions from all susceptibilities χ_p with $p \geq k$. At the spin-glass temperature, all the nonlinear susceptibilities are divergent and χ_5 diverges faster than χ_3. It is therefore important to account for the reaction of all the measurable higher-order processes on the lower-order ones. Specifically, it is straightforward for the general theory of nonlinear processes to evaluate

$$\theta_1' = \chi_1'(\Omega)h + [\chi_3'(\Omega,2\Omega,\Omega)+2\chi_3'(\Omega,0,\Omega)]\frac{h^3}{4} + [4\chi_5'(\Omega,0,\Omega,0,\Omega)+2\chi_5'(\Omega,0,\Omega,2\Omega,\Omega)+2\chi_5'(\Omega,2\Omega,\Omega,0,\Omega) + \chi_5'(\Omega,2\Omega,\Omega,2\Omega,\Omega)+\chi_5'(\Omega,2\Omega,3\Omega,2\Omega,\Omega)]\frac{h^5}{16}+\cdots , \tag{3a}$$

$$\theta_3' = \chi_3'(3\Omega,2\Omega,\Omega)\frac{h^3}{4}+[\chi_5'(3\Omega,2\Omega,3\Omega,2\Omega,\Omega)+\chi_5'(3\Omega,4\Omega,3\Omega,2\Omega,\Omega) + 2\chi_5'(3\Omega,2\Omega,\Omega,0,\Omega)+\chi_5'(3\Omega,2\Omega,\Omega,2\Omega,\Omega)]\frac{h^5}{16}+\cdots , \tag{3b}$$

$$\theta_5' = \chi_5'(5\Omega,4\Omega,3\Omega,2\Omega,\Omega)\frac{h^5}{16}+\cdots . \tag{3c}$$

The measurement of all the harmonic amplitudes θ_k gives a measurement of the susceptibilities in two limits: (a) if $\chi_1' h \gg \chi_3' h^3 \gg \chi_5' h^5$ the back reaction is negligible and each harmonics measures the susceptibility of the same order; (b) in the static ($\Omega \to 0$) limit, where the linear system (3) simplifies to

$$\theta_1' = \chi_1' h + \tfrac{3}{4}\chi_3' h^3 + \tfrac{5}{8}\chi_5' h^5 + \tfrac{34}{64}\chi_7 h^7 + \cdots ,$$
$$\theta_3' = \tfrac{1}{4}\chi_3' h^3 + \tfrac{5}{16}\chi_5' h^5 + \tfrac{21}{64}\chi_7' h^7 + \cdots , \tag{4}$$
$$\theta_5' = \tfrac{1}{16}\chi_5' h^5 + \tfrac{7}{64}\chi_7' h^7 + \cdots , \quad \theta_7' = \tfrac{1}{64}\chi_7' h^7 + \cdots$$

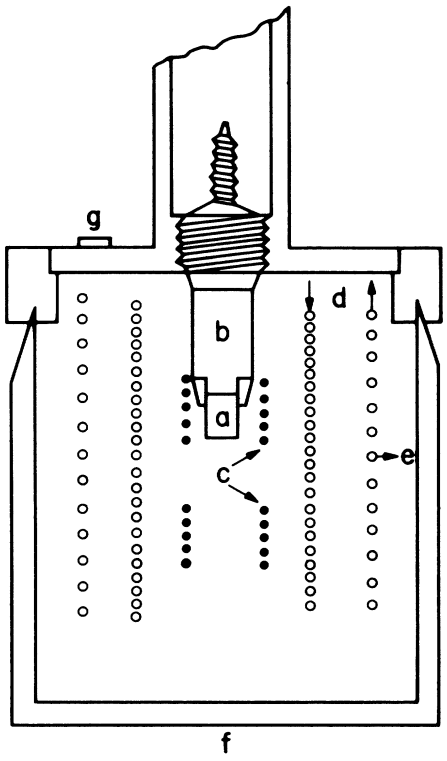

FIG. 4. Schematic diagram of the susceptometer: (a) *Ag*Mn sample; (b) sapphire rod; (c) astatic pair mounted on a thin quartz tube (not shown); (d) Nb-Ti modulation coil producing the ac magnetic fields; (e) center tap used to balance the susceptometer and to cancel part of the linear susceptibility; (f) superconducting shield; (g) low-temperature Hall probe used to monitor dc magnetic fields.

and its solution fully accounts for the back reaction. In absence of dc magnetic field, it is not difficult to keep the ac field sufficiently small so that the back reaction is negligible, mainly because the divergence of the nonlinear susceptibilities is cut off at $\tau < 10^{-2}$ (Sec. IV). In the presence of a dc magnetic field the magnetization response contains even and odd terms as a result of interference between the dc and ac components of the magnetic field, and the nonlinear susceptibilities can be reliably measured only in the static limit, since the back reaction is important. In the limit where $H \gg h$, the harmonic amplitudes θ_k are approximately given by

$$\begin{aligned}
\theta_1 &= (\chi_1' + 3\chi_3' H^2 + 5\chi_5' H^4 + 7\chi_7' H^6 + \cdots)h\,, \\
\theta_2 &(\tfrac{3}{2}\chi_3' + 5\chi_5' H^2 + \tfrac{21}{2}\chi_7' H^4 + \cdots)Hh^2\,, \\
\theta_3 &= (\tfrac{1}{4}\chi_3' + \tfrac{5}{2}\chi_5' H^2 + \tfrac{35}{4}\chi_7 H^4 + \cdots)h^3\,, \qquad (5) \\
\theta_4 &= \tfrac{5}{8}(\chi_5' + 7\chi_7' H^2 + \cdots)Hh^4\,, \\
\theta_5 &= \tfrac{1}{16}(\chi_5' + 21\chi_7' H^2 + \cdots)h^5\,.
\end{aligned}$$

To summarize, the Fourier transform of the magnetization response gives a measurement of the harmonic amplitudes θ_k', with the exception of θ_1' from which a constant has been subtracted by the gradiometer compensation. In the absence of dc field, the back reaction can be made sufficiently small so that the dynamic susceptibilities can be obtained with Eq. (3). In a dc magnetic field, the back reaction requires the solution of the linear system Eq. (5) truncated to the highest measurable harmonic.

IV. STATIC LIMIT

A. Critical behavior in zero field

To analyze a critical behavior requires a precise determination of a transition temperature. T_g has been determined in two ways: (a) as the peak of the linear susceptibility as a function of temperature (Fig. 1) at the lowest frequency (10^{-3} Hz) giving values of $T_g = 2.945$ K for the 0.5 at.% *Ag*Mn sample and $T_g = 1.225$ K for the 0.2 at.% sample; and (b) as the peak of the nonlinear susceptibility with temperature shown in Fig. 5 at a frequency of 3×10^{-3} Hz leading to values of T_g of 2.940 and 1.220 for the 0.5 and 0.2 at.% samples, respectively. These numbers are essentially identical and close to the value obtained in the scaling analysis described below, where T_g is a fitting parameter. It remains true that all the methods used can only overestimate T_g, leading to an underestimate of the exponent γ. From Fig. 5, the nonlinear susceptibility increases by 2 orders of magnitude close to T_g, and its asymmetry with respect to T_g is presumably associated with the strong dynamical effects in the spin-glass phase. The frequency dependence of the nonlinear susceptibility χ_3 above T_g is shown in Fig. 6. It is, at all temperatures, constant below 10^{-2} Hz. We therefore take these values as the static limit of the nonlinear susceptibility.

We first analyze this data in terms of the scaling hy-

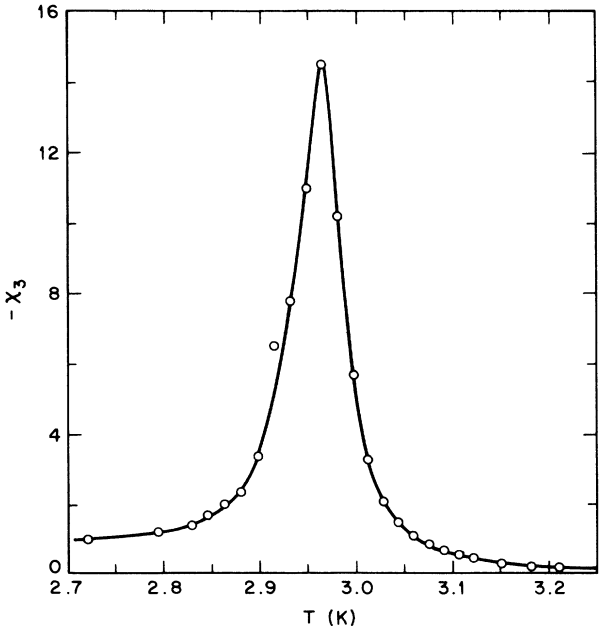

FIG. 5. Temperature dependence of the nonlinear susceptibility χ_3 across the spin-glass transition at a frequency of 10^{-2} Hz. Note the asymmetry above and below T_g.

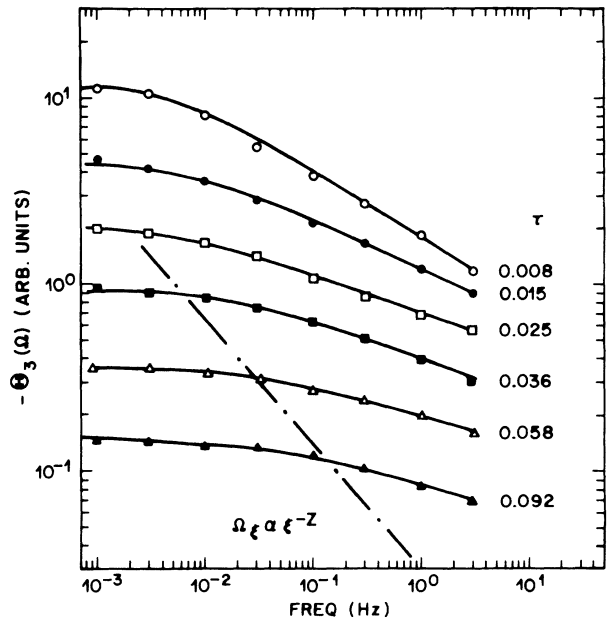

FIG. 6. Frequency dependence of $\theta_3 \sim -\chi_3' h^3/4$ above T_g for several reduced temperatures. The slope close to T_g and above 10^{-2} Hz is $\sim \gamma/(z\nu)$. The dashed-dotted line qualitatively shows the shift of Ω_ξ as a function of τ expected from dynamic scaling.

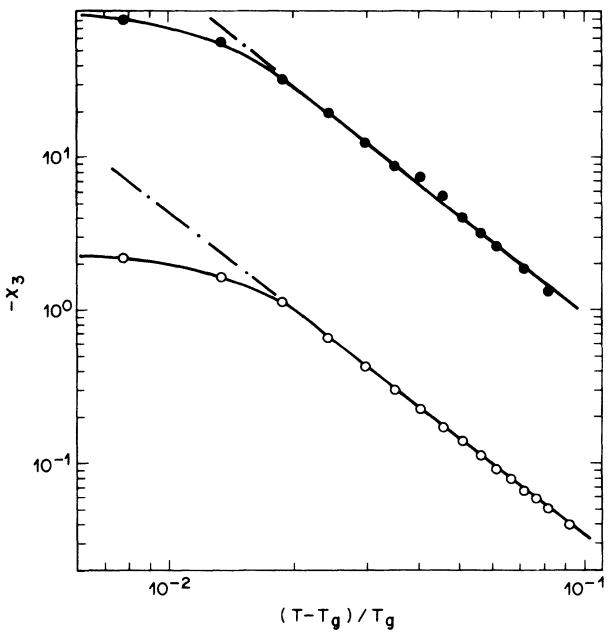

FIG. 7. Temperature dependence of $-\chi_3$ above T_g measured at 10^{-2} Hz in static fields of 0 (open circles) and 90 G (solid circles) as a function of reduced temperature τ. The same slope $(-\gamma)$ indicates that the divergence of χ_3 in dc fields less than 100 G remains well described by the same exponent $\gamma = 2.3$ as in zero field.

pothesis which has been successfully applied to the Monte Carlo studies of Ising spin glasses.[7] It states that any field or temperature deviations away from the critical point should have the same effect on a singular thermodynamic quantity when measured on comparable scales. This assumption implies that the nonlinear susceptibility $M/H - \chi_1$, which is expected to be singular at a spin-glass transition, should be expressible in terms of a function of a single variable,

$$\frac{M}{H} - \chi_1 = \tau^\beta f(h^2/\tau^\phi) \tag{6}$$

where $h = H/T_g$ is the scaled magnetic field, β is the exponent governing the growth of the order parameter q below T_g, while ϕ is the scaling exponent. To relate this quantity to the susceptibilities χ_3, χ_5, ... experimentally accessible, we perform a high-temperature expansion of f and identify the nonlinear susceptibilities χ_3, χ_5, and χ_7 as the coefficient of h^2, h^4, and h^6 in the series expansion. Defining γ as the power governing the divergence of χ_3 with reduced temperature, we have $\phi = \beta + \gamma$ and

$$\chi_3' = \frac{\chi_3}{\tau^\gamma}, \quad \chi_5' = \frac{\chi_5}{\tau^{2\gamma+\beta}}, \quad \chi_7' = \frac{\chi_7}{\tau^{3\gamma+2\beta}}. \tag{7}$$

The critical amplitudes χ_3, χ_5, ... are nonuniversal constants expected to be of the order of χ_1 as in ferromagnets. The data shown in Fig. 7 show a power-law divergence of χ_3 for values of reduced temperature between 10^{-2} and 10^{-1}. For values of τ below 10^{-2}, we observe a rounding of the transition which will be discussed below. The determination of γ has been made with a one- (γ) and a two- (γ and T_g) parameter fit of the data shown in Fig. 7, with values of τ in the range $[10^{-2}, 10^{-1}]$ to avoid underestimates of γ associated with the rounding of the transition. We find, respectively, $\gamma = 2.1 \pm 0.1$ with a one-parameter fit and $\gamma = 2.3 \pm 0.15$ with $T_g = 2.935$ K with a two-parameter fit. The exponent γ for the 0.2 at.% sample is found with the same procedure to be $\gamma = 2.3 \pm 0.2$. Since this method provides absolute measurements, we are also able to estimate the critical amplitude to be $\chi_3 \sim 0.4\chi_1$, suggesting that all the spins contribute to the critical fluctuations. The most stringent test of the scaling assumption (6) can be made without the precise knowledge of T_g: from Eq. (7), χ_5/χ_3 and χ_7/χ_5 scale as $\tau^{-(\gamma+\beta)}$ or alternatively as $\chi_3^{\{1+\beta/\gamma\}}$. The plots of χ_5/χ_3 and χ_7/χ_5 vs χ_3 shown in Fig. 8 are two parallel lines with a slope of 1.45 ± 0.1, as predicted by the scaling assumption. From this slope, we infer the value of $\beta = 0.9 \pm 0.2$. This test of scaling is satisfied at all temperatures, even where the rounding effects take over. This observation suggests that all quantities are related according to the scaling relations, although they do not actually become infinite. This is the first indication that the correlation length ξ does not become infinite, but saturates at a finite value ξ_r. An estimate of ξ_r can be made, assuming that at the temperature τ_r where rounding starts to take place, ξ still scales as $\xi = x l_0 \tau^{-\nu}$, where l_0 is the average distance between impurities and x is the critical amplitude. For the 0.5 at.% sample, $l_0 = 5.8$ Å,[28] while x is of order unity in Monte Carlo simulation of Ising spin glasses.[7] From the divergence of the nonlinear susceptibilities, we conclude that the spin-glass correlation length ξ is the only relevant length scale until $\xi = \xi_r$ is of the order of 0.2 μm, where it presumably levels off. At this point, the correlation

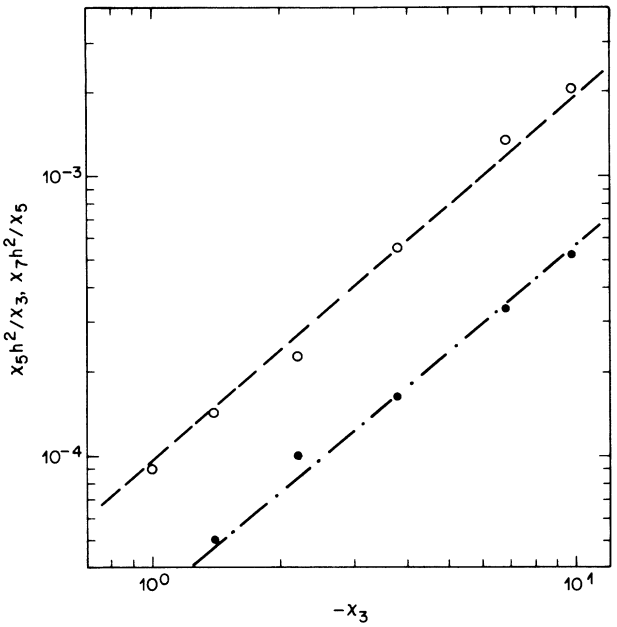

FIG. 8. Plot of the susceptibility ratios $-\chi_5/\chi_3$ (open circles) and $-\chi_7/\chi_5$ (solid circles) as a function of $-\chi_3$. The slope is $1+\beta/\gamma$. The rounding of the transition is not observed, indicating that the scaling relations are satisfied even when rounding takes place.

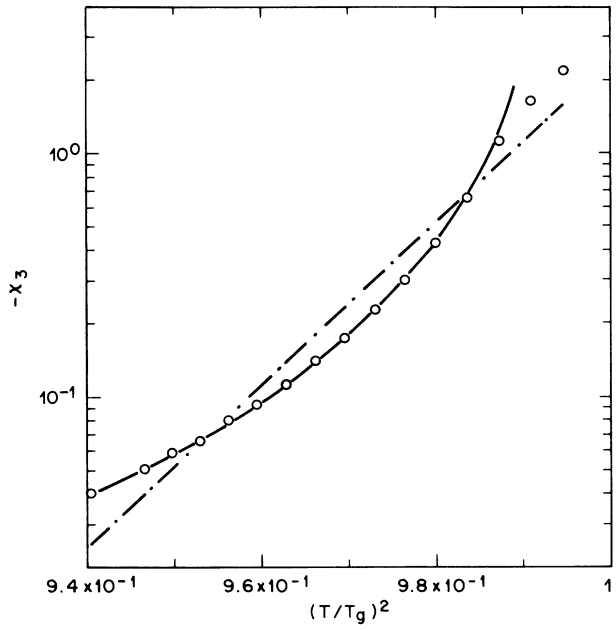

FIG. 9. Plot of $-\chi_3$ as a function of $(T_g/T)^2$. The dashed-dotted line is the result expected for a zero-temperature transition.

volume contains 10^8 spins. The exponent δ may be obtained from the scaling relation $\delta=1+\gamma/\beta=3.3\pm0.2$. Similarly, the hyperscaling relation $d\nu=\gamma+2\beta$ leads to a value $\nu=1.3\pm0.2$ for the exponent governing the divergence of the correlation length. It appears unlikely that the precision in the determination of the exponents can be improved in the future: scaling holds only in the vicinity of T_g (say $\tau<0.15$), while rounding effects limit in a fundamental way how close to T_g one can get. Thus, spin-glass critical behavior is only observable over one decade of reduced temperature.

The values of the exponents measured here are in good agreement with two other experiments done in similar conditions.[29] It differs from earlier measurements made far from T_g and with larger fields.[30]

Some comments on these rounding effects are in order: dynamical effects will round up the transition as the systems falls out of equilibrium. As will be discussed in the next section, the observed rounding is consistent with this picture. Inhomogeneities in the sample will also broaden the transition. Simple models assuming smooth variation of the impurity concentration are not consistent with the transition width as determined by the linear susceptibility and the observed critical amplitude. (A critical amplitude of order unity implies that all the spins contribute to the divergent quantities). In ferromagnets, such simple models also fail to account properly for the transition width which arises from inhomogeneities and defects, so their applicability to disordered systems is questionable.

We now examine the possibility of a zero-temperature transition. At the lower critical dimension the nonlinear susceptibility should scale as $\exp(J^2/T^2)$.[31] The plot of χ_3 shown in Fig. 9 as a function of $(T_g/T)^2$ shows some significant deviation from this scaling law and the best fit corresponds to $J\sim8T_g$, a value unphysically high. With this fit, the nonlinear magnetization in a field of 3 G would become equal to the linear magnetization 5% below T_g and would reach the value of the saturated magnetization 13% below T_g. Thus this exponential scaling transition can safely be ruled out. We have also considered the scaling function $\chi_3\propto T^{-\gamma}$.[32] We find, as did earlier authors, that our data are inconsistent with this scaling law and can be ruled out. In conclusion, a zero-temperature transition cannot adequately account for our data.

The existence of a phase transition appears to be the only consistent picture. As noted earlier, the dilute antiferromagnet also has a singular nonlinear susceptibility. We can estimate the critical exponent from the scaling function for the susceptibility,

$$\chi=\tau^{-\gamma}f(h^2/\tau^{\phi}). \tag{8}$$

The scaling exponent $\phi=2\beta$, since χ must scale as $(h/h_l)^2$ below T_g, where the local field $h_l\propto\tau^{\beta}$.[23] The scaling function f goes to zero linearly for small arguments, so there is no singular contribution to the linear susceptibility. On the other hand, the nonlinear susceptibility diverges as $\gamma-2\beta$, which for the three-dimensional Ising model is of the order of 0.6, an exponent four times smaller than the one obtained for spin glasses. This leads to a nonlinear susceptibility about 3 orders of magnitude smaller at $\tau=10^{-2}T_N$ than for spin glasses, which is consistent with Monte Carlo simulations.[22] Such a small exponent is not consistent with this measurement, and the interpretation of the divergent nonlinear susceptibility in terms of antiferromagnetic ordering appears problematic. Measurements of the nonlinear susceptibility of dilute antiferromagnets would be helpful here.

B. Behavior in a static field

As noted earlier, there is even and odd harmonic responses in a static field (Fig. 4). Also, the back reaction of the higher-order nonlinear susceptibilities is so large that the harmonic amplitude θ_2 changes sign as shown in Fig. 10. The nonlinear susceptibilities can nevertheless be measured with the procedure given in Sec. III. We find that the maximum of χ_3 occurs at the same temperature as in zero field. We stress here that the spin-glass susceptibility χ_{SG} does not coincide with the nonlinear susceptibility $\chi_3=\partial^4 F/\partial H^4$ (F is the free energy) in a field.[33] There are therefore no *a priori* reasons to identify the maximum of χ_3 with the transition temperature T_g in a magnetic field. In particular, the absence of a shift with field does not rule out the existence of a de Almeida-Thouless transition line, since in mean-field theory the maximum of χ_3 occurs always at the same temperature, while the transition temperature shifts down. The measurements of χ_3 (Fig. 7) also display a divergence with the same power law ($\gamma=2.2\pm0.2$) as in zero field. This indicates that dc fields below 100 G do not significantly alter the critical behavior which remains well described by the exponents measured in zero field.

V. CRITICAL DYNAMICS

A. Linear susceptibility

The frequency dependence of the linear susceptibility was carried out at ultralow frequencies (Fig. 1). The shift in T_g, plotted in Fig. 11, appears to saturate below 10^{-2} Hz as expected for a finite-temperature phase transition. If the depression of T_g is interpreted within dynamic scaling ($\delta T_g/T_g \propto \Omega^{1/z\nu}$),[34] we find an effective exponent $z\nu\sim5.4$ with considerable uncertainties. Such determination of the dynamical exponent is very unreliable compared to the proper dynamic scaling analysis of singular thermodynamic quantities given below. The uncertainties in absolute calibration of χ_1 make it difficult to perform a precise scaling analysis of $\delta\chi_1/\chi_1$ in the vicinity of T_g. It is on the other hand clear from the data below T_g that χ_1' scale with frequency as

$$\chi'(\omega)-\chi'(\omega')=-[\log_{10}(\omega/\omega')]^{\mu} \tag{9}$$

with $-1.5<\mu<1.5$. A similar functional dependence was predicted by Fisher and Huse using a description of the low-temperature excitations of a spin glass with a two-level droplet model.[35] Only negative values of the exponent μ are meaningful in this picture.

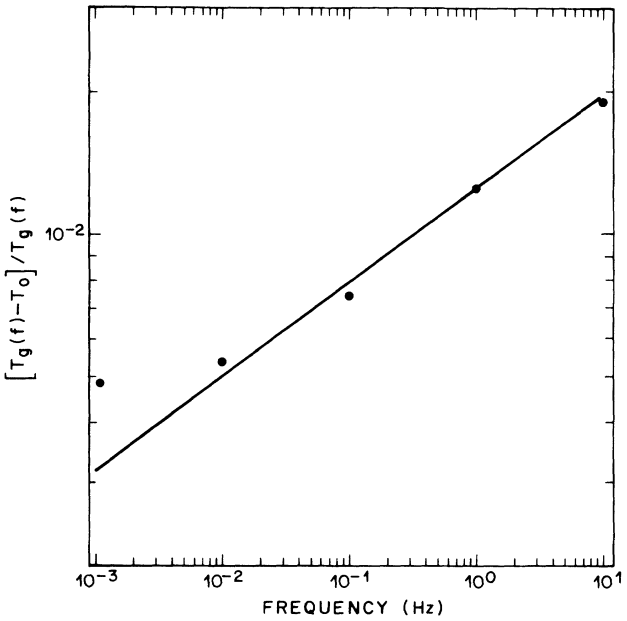

FIG. 11. Frequency dependence of the transition temperature T_g. Within dynamic scaling, the slope approaches $1/(z\nu)$ at high frequency.

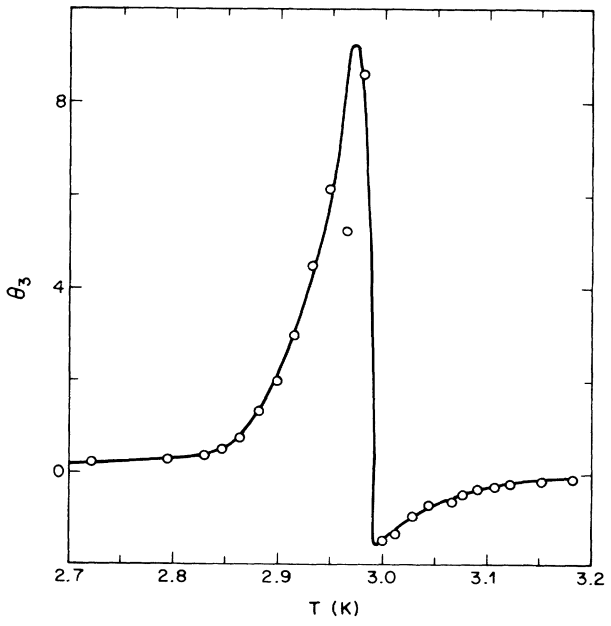

FIG. 10. Temperature dependence of θ_2 through the transition. The rapid sign change close to T_g is caused by the back reaction of χ_7 and χ_5 onto θ_2.

B. Nonlinear susceptibility

We have seen that in sufficiently low ac field, the harmonic amplitude θ_3 measures directly the nonlinear susceptibility $\chi_3(3\Omega,2\Omega,\Omega)$, since the back reaction is negligible. We found the shift in the maximum of $\chi_3(T)$ with frequency to be small, and the value of T_g determined from the static limit (Sec. IV) will be assumed in our analysis. As shown in Fig. 6, we find that close to T_g, this quantity diverges as a power law of frequency, above 10^{-2} Hz. This suggests an analysis within the dynamic scaling hypothesis, which was successfully applied to Monte Carlo simulation of Ising systems. In this picture, the relaxation time of the order parameter $t^*\propto\tau^{-z\nu}$ is the only relevant time scale. It is then straightforward to derive the scaling laws for the nonlinear susceptibilities χ_3

and χ_5 as a function of temperature and frequency,[36]

$$\chi_3 = \Omega^{-\gamma/z\nu} g_3(\Omega t^*) = \tau^{-\gamma} f_3(\Omega t^*), \quad (10a)$$

$$\chi_5 = \Omega^{-(2\gamma+\beta)/z\nu} g_5(\Omega t^*) = \tau^{-(2\gamma+\beta)} f_5(\Omega t^*). \quad (10b)$$

As $T \to T_g$, the slopes $\log_{10}(\chi_3')/\log_{10}(\Omega)$ and $\log_{10}(\chi_5')/\log_{10}(\Omega)$ should therefore approach $-\gamma/z\nu$ and $-(2\gamma+\beta)/z\nu$, respectively. At frequencies greater than 10^{-2} Hz and close to T_g, their measured values (Fig. 6) 0.30 ± 0.02 and 0.69 ± 0.07 give the same value of the effective exponent $z\nu = 7 \pm 0.6$. This is consistent with a unique relaxation time, in agreement with dynamic scaling. As long as $\xi < \xi_r$, the relaxation time of a correlated region of size ξ is of order $\Omega_\xi^{-1} \sim t_0(\xi/l_0)^z$, where t_0 is a microscopic relaxation time of the order of $10^{-11 \pm 1}$ sec. Since frequencies below Ω_ξ probe length scales greater than ξ, the nonlinear susceptibilities should level off when $\Omega < \Omega_\xi$. This behavior is qualitatively observed in Fig. 6 at reduced temperature greater than 0.04. Although this value is greater than τ_r, the reduced temperature at which the static nonlinear susceptibility stops growing, it is consistent with a large correlation volume (10^6–10^9 spins). For $\tau < 0.04$, there are real deviations from scaling which cannot be absorbed in the dynamic scaling function g shown in Fig. 12, unlike the change in slope of the frequency-dependent nonlinear susceptibilities shown in Fig. 6. The significance of these deviations will have to be assessed in further work.

We note here that the reduced temperature at which we expect the system to fall out of equilibrium when probed with a frequency of 3×10^{-3} Hz is $\tau = (\Omega t_0)^{1/z\nu} \sim 1.2 \times 10^{-2}$. This is remarkably close for the temperature at which the transition is rounded. Nonequilibrium effects provide therefore a consistent explanation of the observed rounding of the phase transition.

We now consider the logarithmic scaling introduced by Malozemoff and Pytte as an alternative to dynamic scaling.[37] Logarithmic scaling laws imply activated dynamics in the vicinity of the transition temperature T_g.

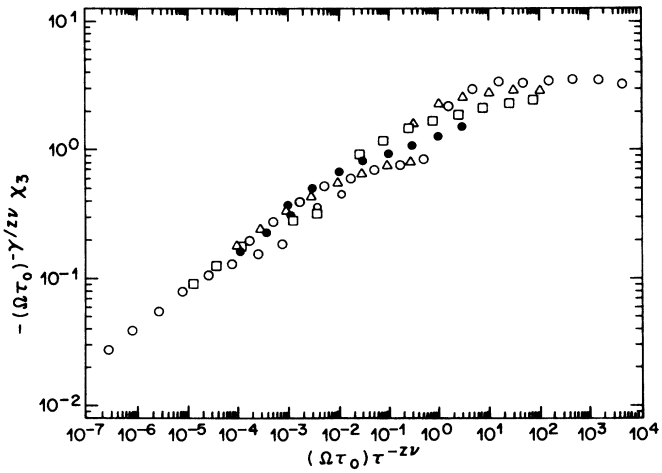

FIG. 12. All the measurements of χ_3 as a function of temperature and frequency are gathered on this scaling plot of $-(\Omega t_0)^{-\gamma/z\nu}\chi_3$ as a function of Ωt^*. This curve is a measurement of the scaling function g [Eq. (8)].

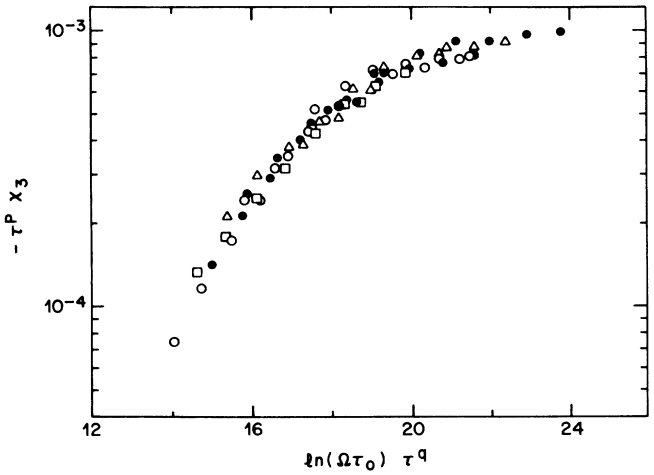

FIG. 13. Logarithmic scaling plot of $\tau^\gamma\chi_3$ as a function of $\ln(\Omega t_0)\tau^q$. The good collapse of the data on the single function f shows that this form of activated dynamics fits the data well with a small exponent $q \sim 0.1$.

Specifically, the relaxation time τ follows a generalized Volger-Fulcher law ($\tau = \tau_0 \exp[a/(T-T_g)^q]$). This dynamical behavior is expected for random-field systems,[38] but its applicability to spin glasses remains at present unclear. In this picture, the scaling ansatz

$$\chi_3 = \xi^{-p/\nu} f(-\ln(\omega t_0)/\xi^{q/\nu}) \quad (11)$$

describes the dynamic nonlinear susceptibility in the vicinity of T_g. In Eq. (11), p and q are scaling exponents, while t_0 is a microscopic relaxation time. Here, there is also a thermodynamic phase transition at T_g with a divergent correlation length $\xi \propto \tau^{-\nu}$. Because of the large number of adjustable parameters in Eq. (11) (p, q, and τ_0), we have obtained the scaling function by biasing the fit with $p = \gamma$ measured in the static limit and $t_0 = 10^{-11}$ sec. This procedure makes the quality of the scaling achieved in Fig. 13 more significant, since there is now only one scaling exponent q adjusted to be ~ 0.1. Clearly, this experiment can very well be interpreted with a finite-temperature transition at which the time scales diverge according to a generalized Volger-Fulcher law. There are, however, two difficulties. First, the small exponent q suggests that the activation energy scales are very small. Second, we expect that in the paramagnetic regime $\chi_3'' \to \Omega^3$, implying that the scaling function shown in Fig. 13 should become exponential at large arguments. This unphysical behavior suggests that logarithmic scaling can only apply in the vicinity of the transition.

To conclude, it is almost impossible to distinguish dynamic scaling with an exponent z as large as 5.4 from an activated behavior with an exponent q as small as 0.1. It is again difficult to imagine that experiments will ever be able to distinguish between the two scaling laws.

VI. BELOW T_g

In the spin-glass phase, the time scales are long and it is difficult to measure equilibrium quantities. In zero field,

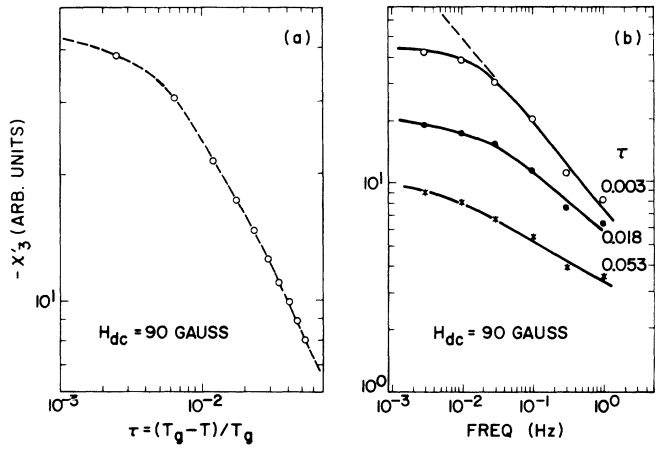

FIG. 14. (a) Temperature dependence of χ_3' measured at 3×10^{-3} Hz below T_g. (b) Frequency dependence of χ_3' just below T_g for several reduced temperatures.

the dynamics is so slow that we could not identify a static limit at any frequency. In a static field of 90 G and close to T_g, time scales are significantly shorter and we identified a quasistatic limit below 3×10^{-3} Hz (Fig. 14). In this regime, we have attempted to determine the exponent γ' $\{\chi_3\propto[(T_g-T)/T_g]^{-\gamma'}\}$. From the data shown in Fig. 14(a), we estimate that the effective exponent γ' is close to 2, which is consistent with scaling ($\gamma=\gamma'$). The slight curvature at larger values of τ is presumably due to dynamical effects.

The dynamics is summarized in Fig. 14(b). The presence of severe aging effects and drifts in the dynamics make a quantitative analysis of this data more speculative. Close to T_g, the fit to a power law $\Omega^{-\lambda}$ gives the same value $\lambda\sim0.30\pm0.14$ as above T_g. We have also considered logarithmic scaling $[-\ln(\Omega t_0)]^{-\mu}$. With the assumption that t_0 is a microscopic time of order 10^{-11} sec, the exponent μ is $\sim0.65\pm0.25$.

At a temperature of $0.3\times T_g$, we find that χ_3 is only a few percent of its critical value, while the higher-order susceptibilities χ_5 and χ_7 are unmeasurable, indicating a weak dynamic nonlinear susceptibility in the spin-glass phase. This gives little support to the idea that the entire spin-glass phase is critical as it has been argued in the literature.[39]

VII. DISCUSSION

We have described static and dynamic measurements of the nonlinear susceptibilities in the vicinity of the spin-glass transition T_g of AgMn. They clearly demonstrate the existence of a spin-glass-like phase transition at T_g and rule out a homogeneous freezing of the spins. Throughout the paper, we have interpreted the observed divergence of the nonlinear susceptibilities as a divergence of the spin-glass correlation length. We justify this interpretation with (i) the existence of a generalized fluctuation-dissipation theorem relating four-spin correlation functions to the nonlinear susceptibility χ_3'' (Sec. II), and (ii) the scaling relation (7) for χ_3' in the vicinity of the transition. The observed divergence of all the nonlinear susceptibilities χ_3, χ_5, and χ_7 check the relations imposed by static scaling. Since Eq. (6) is verified experimentally, the only consistent picture is a divergent spin-glass correlation length. A recent study of finite-size effect in spin glasses[40] has largely confirmed this interpretation with a direct measurement of the spin-glass correlation length of the order of 0.1 μm, compared to the value of 0.2 μm obtained in this work.

The observed rounding of the phase transition can be interpreted as the nonequilibrium response of the system as the relaxation times exceed the period of the ac field. Other interpretations (inhomogeneities) are difficult to rule out, although we have indicated some inconsistencies (critical amplitudes).

We have found the dynamics to be consistent with critical slowing down or with a weakly activated dynamic describe by a generalized Volger-Fulcher law.

The value of the critical exponent obtained in this study and in Monte Carlo simulation of Ising spin glasses are summarized in Table I. The value of β is close to its mean-field value indicating a stiff ordering, which contrasts very much with the soft ordering ($\beta=0.5$) observed in simulation of short-range Ising systems. The difference in the values of γ (2.3 vs 2.9) is also significant, since they are well measured by simulations and experiments. A first interpretation of the discrepancy is that the RKKY spin glasses do not belong to the same universality class as short-range Ising systems. An alternative interpretation is that the experiments on RKKY systems have been carried out in a crossover region associated either with the range of their interactions or their weakly Heisenberg character. In favor of a crossover, we note that the measured exponents fall systematically between the mean-field value and the Ising short-range value. Finally, a more radical interpretation would dispute that universality classes in random magnets can be identified with the value of the exponents, but should rather be defined with the probability distribution of exchange constants between spins. Clearly, it will take more simulations and more experimental studies to sort out interpretations as radically different as the one we have outlined.

TABLE I. Value of the critical exponent obtained in this study and in Monte Carlo simulations of Ising spin glasses.

	α	β	γ	δ	ν	z
This work	1.9 ± 0.3	0.9 ± 0.2	2.3 ± 0.2	3.3 ± 0.3	1.3 ± 0.15	5.3 ± 0.8
Simulations	1.9 ± 0.3	0.5 ± 0.1	2.9 ± 0.1	6.8 ± 1.2	1.3 ± 0.1	6.0 ± 0.5

ACKNOWLEDGMENTS

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$$\chi_{SG} = \left[\sum(\langle\sigma_i\sigma_j\rangle - \langle\sigma_i\rangle\langle\sigma_j\rangle)^2\right]$$

and

$$\chi_3 = \left[\sum(\langle\sigma_i\sigma_j\sigma_k\sigma_l\rangle - 4\langle\sigma_i\sigma_j\sigma_k\rangle\langle\sigma_l\rangle - 3\langle\sigma_i\sigma_j\rangle\langle\sigma_k\sigma_l\rangle + 6\langle\sigma_i\sigma_j\rangle\langle\sigma_k\rangle\langle\sigma_l\rangle - 6\langle\sigma_i\rangle\langle\sigma_j\rangle\langle\sigma_k\rangle\langle\sigma_l\rangle)\right].$$

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