Critical scaling of the EPR linewidth in the Ag-Mn spin glass

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We have measured the EPR linewidth and shift between 1 and 10 GHz in the paramagnetic regime of the Ag-Mn(2.6 at. %) spin glass. The critical part of the linewidth can be described equally well within experimental error either by dynamic (frequency) scaling, $W^{ex} \propto t^{-g} G(\omega/t^{\nu z})$, or by field scaling, $W^{ex} \propto t^{-g} G(H/t^{(\beta+\gamma_s)/2})$. The experiments give $g=1.5$ and either $\nu z=2.5$ or $(\beta+\gamma_s)/2=2.5$. The adequacy of the scaling form is taken as evidence favoring a thermodynamic phase transition of the usual type at T_g. Within experimental error, T_g is independent of applied magnetic field, and within the Sherrington-Kirkpatrick model, T_g may be identified with the Gabay-Toulouse line for transverse freezing.

I. INTRODUCTION

The spin-glass state is signaled by a maximum in the temperature dependence of the zero-field magnetic susceptibility χ, defining the temperature T_g.[1] Despite intense theoretical and experimental effort,[2] it remains controversial whether there exists a thermodynamic phase transition in the usual sense at T_g. Besides theoretical arguments, the lack of structure in the specific heat[3] is largely responsible for this situation, although some data are consistent with a specific-heat critical exponent[4] $\alpha < -1$. Arguments against a phase transition are also found in the strongly frequency-dependent maximum of χ in some materials,[5] in the scaling laws for time-dependent response,[6] and in broad, nonexponential tails found in neutron scattering.[7]

Among the evidence in favor of a phase transition at finite temperature is the electron paramagnetic resonance (EPR) linewidth in the paramagnetic regime, which shows a strong increase just above T_g.[8] Over a broad range of T, the width behaves as a power law[9] in $t=(T-T_g)/T_g$, suggesting that some effective correlation time would become infinite at T_g. Yet, as T_g is approached, the measured EPR width ceases to increase,[10,11] attaining instead a finite value near and at T_g. This "saturation" of width may be interpreted in several ways. On the one hand, the most skeptical view is that there is no phase transition at T_g, with the "power-law linewidth divergence" over some temperature range being only accidental. Growth of static clusters could provide a model for such behavior. On the other hand, accepting the reality of the phase transition, the failure of the power law admits two explanations.[11] Either the lengthening of the correlation time is suppressed by some intrinsic behavior of spin glasses in applied magnetic fields, or continued lengthening of the correlation time is not reflected in larger linewidths because of the nature of the EPR probe.

Here we present EPR data for $T > T_g$ in the Ag-Mn(2.6 at. %) spin-glass at several frequencies between 1 and 10 GHz. The full temperature and frequency dependence of the excess linewidth is analyzed from the standpoint of critical scaling. The experimental results can be described equally well by field scaling[12] and by dynamic (frequency) scaling. Thus we cannot distinguish whether the "saturation" of the width near T_g is due to the nonzero measuring field or to the finite timescale of the experimental probe. Nevertheless, either analysis supports the case for a phase transition of the usual kind at T_g, complementing the conclusions of earlier work on scaling of the dc magnetization.[13]

II. EXPERIMENTAL METHOD

All measurements were made on the same Ag-Mn(2.6 at. %) sample used in Ref. 11, for which the low-field static cusp temperature was measured as $T_g=10.24$ K. The X-band and (1—2)-GHz data were taken as described in Ref. 11. For all measurements reported here, temperature control and measurement, data reduction from the raw spectra, and demagnetization correction of the resonance shift were performed as described therein.

The sample and flow system were inserted into a rectangular cavity operated in the TE_{102} and TE_{104} modes for 4.6 and 7.2 GHz, respectively. For 3.3 GHz, the resonance frequency was depressed by filling most of the volume of the cavity with a Teflon insert. Energy was coupled to the cavity through a loop at the end of a semirigid coaxial line. An "elementary" spectrometer was constructed from a Hewlett-Packard 8620C/86240A microwave sweeper, followed by an isolator, circulator, and diode detector. The source was locked to the cavity resonance by means of a simple frequency-modulation phase-locked loop.

III. RESULTS

The temperature dependence of the EPR linewidth in the paramagnetic phase of a spin glass can be expressed in the form[11] $W=a+bT+W^{ex}$, where a is the residual width, b the thermal broadening coefficient, and W^{ex} an "excess" contribution which is manifest as $T \to T_g$ from above. We take W^{ex} to arise from exchange narrowing of an anisotropic spin-spin interaction, which was argued previously[11] to consist principally of the Dzaloshinski-

Moriya form proposed by Fert and Levy.[14] We present here a more thorough analysis than in Ref. 11 of the temperature dependence of W^{ex} at several frequencies between 1 and 10 GHz. $T_g = 10.24$ K was used for all data at all magnetic fields when expressing the temperature in reduced units. The relevance of this choice will be discussed in Sec. IV.

Figure 1 exhibits W^{ex} versus reduced temperature $t = (T - T_g)/T_g$ at five frequencies, illustrating the following points.

(1) The higher temperature data are the same for all frequencies, within experimental error, and the common data obey the power-law behavior,

$$W^{ex} = ct^{-g}, \tag{1}$$

with $g = 1.5 \pm 0.1$ (This g was called γ in Ref. 11. We have changed notation to avoid confusion with the exponent γ of the magnetic susceptibility.)

(2) The data depart from the power-law behavior below a frequency-dependent temperature t^*, which is larger for higher frequency.

(3) W^{ex} approaches a constant as $t \to 0$. This frequency-dependent constant falls off with frequency somewhat more rapidly than $\omega^{-1/2}$. The frequency dependence of W^{ex} is exhibited in Fig. 2 at several fixed temperatures. The magnetic field shift from the high-temperature resonance position, ΔH^{res}, is also shown. While ΔH^{res} falls off rapidly at high frequencies, at low frequency $\Delta H^{res} \propto \omega$, whence the shift below 2 GHz can be characterized as a temperature-dependent g shift.

Saslow[15] has presented a phenomenological theory for EPR above T_g. He finds

$$\Delta H^{res} = \frac{g\mu_\beta}{\hbar} \frac{(K/\chi)\omega}{\omega^2 + U^{-2}}, \tag{2a}$$

$$W^{ex} = \frac{g\mu_\beta}{\hbar} \frac{(K/\chi)U^{-1}}{\omega^2 + U^{-2}}, \tag{2b}$$

where K is an anisotropy constant, χ is the static magnetic susceptibility, and U^{-1} is the rate at which the "spin triad" relaxes toward equilibrium. According to Saslow's picture, $K = 0$ for $T > T_g$ in zero magnetic field, but a finite field induces a finite anisotropy: $K \propto H^2$. U^{-1} is also strongly field (or frequency) dependent. Equations of these forms were analyzed in Ref. 11 under the assumptions that K/χ and U^{-1} were independent of ω or H, and that $\omega U \ll 1$ at 1 GHz. It was found there that the temperature dependence of the low-frequency width and shift

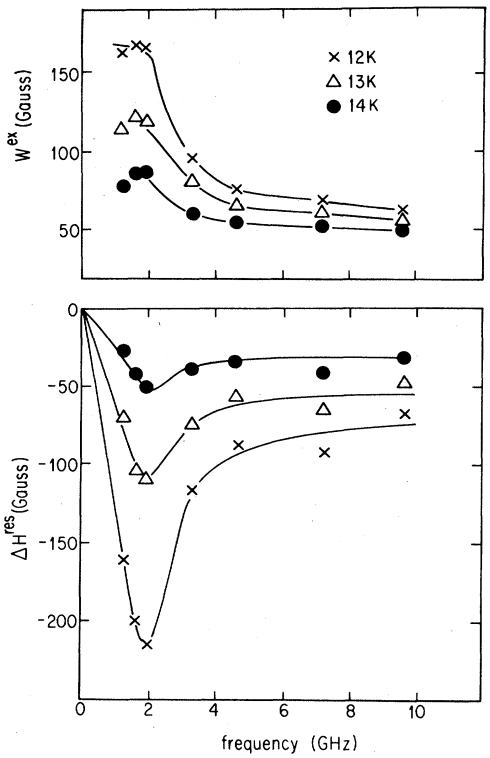

FIG. 2. Frequency dependence of W^{ex} and of the line-shift ΔH^{res} at three fixed temperatures. Lines are guides to the eye.

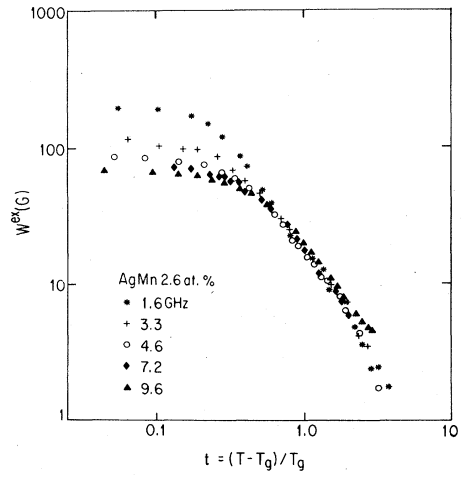

FIG. 1. Excess linewidth $W^{ex} = W - a - bT$ vs reduced temperature t. This log-log plot shows a common power-law behavior $W^{ex} \propto t^{-1.5 \pm 0.1}$ for large t.

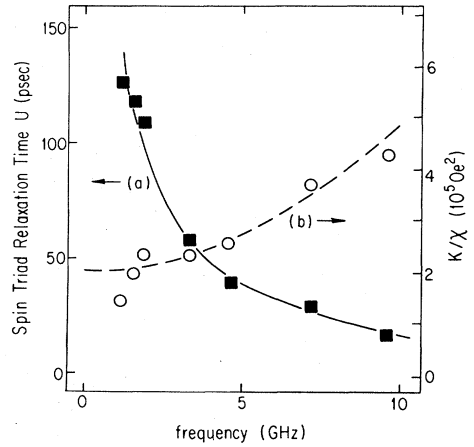

FIG. 3. Frequency dependence of the parameters K/χ and U required to fit the forms of Eq. (2), $T = 12$ K. In the spirit of Ref. 15, these dependences should be regarded as field (not frequency) effects. The dashed line has the form $A + BH^2$, while the solid line is a guide for the eye.

could be fit with $U^{-1} \propto t^g$ and K/χ independent of temperature, but the predicted decreases of W^{ex} and H^{res} with ω were much stronger than observed.

If the forms of Eq. (2) are assumed correct, then U can be deduced unambiguously from the ratio $\Delta H^{res}/W^{ex} = \omega U$. This procedure is useful only very near T_g, where both W^{ex} and ΔH^{res} are large. Figure 3 presents the frequency dependence of U and K/χ at 12 K, deduced in this manner. However even at the lowest frequency, $\omega U \gg 1$, invalidating the previous analysis.[11] Furthermore, K/χ is indeed consistent with a quadratic dependence on H, but only for K/χ nonzero in zero field. Some investigations[16] point to finite K/χ for $T > T_g$, while others[17] show that the torque associated with K/χ vanishes above T_g. In any case, the validity of Eqs. (2) is an *assumption* for this approach to the analysis of the data. In the next section we analyze W^{ex} in the alternative form suggested by critical scaling.

IV. SCALING ANALYSIS

The critical part of the spin-relaxation rate is determined from the correlation function,[18]

$$C_Q(\omega,\mathbf{k}) = \int_{-\infty}^{\infty} dt\, e^{i\omega t} \langle [\mathbf{S}_0^+(\mathbf{k},t), \mathscr{H}'] \times [\mathbf{S}_0^-(\mathbf{k},0), \mathscr{H}'] \rangle ,$$

where $\mathbf{S}_0$ is the total spin. For EPR in dirty metals, one takes $k = \lambda^{-1} \approx 0$, where λ is the skin depth. The perturbation $\mathscr{H}'$ contains an anisotropic part, commonly dipolar in origin, although in metallic spin glasses a Dzaloshinski-Moriya term is probably dominant.[19,11] In either case $C_Q(\omega)$ becomes a linear combination of four-spin correlation functions whose values depend on temperature, measuring frequency, and any applied fields. According to the critical scaling hypothesis C_Q is a homogeneous function of its parameters:

$$C_Q(t,\omega,H) \propto t^{-g} G(\omega/t^{\nu z}, H/t^{(\beta+\gamma_s)/2}) .$$

The forms for the frequency and field arguments are obtained from Refs. 20 and 12, respectively. The exponents are ν for the correlation length ($\xi \propto t^{-\nu}$), z for dynamic scaling ($\tau \propto \xi^z$), β for the order parameter, and γ_s for the nonlinear susceptibility. It is not known a priori for a spin glass whether the effect of frequency or of field is more important. Therefore, assuming that the effects are not of equal importance, we will treat them separately and in turn, beginning with the frequency dependence.

For dynamic scaling the correlation function obeys[20] $C_Q(\omega) \propto t^{-g} G(\omega/t^{\nu z})$. The dynamic scaling function $G(\omega\tau)$ goes to unity for $\omega\tau \ll 1$ and falls off as a power law, $G(\omega\tau) \propto (\omega\tau)^{-\kappa}$, for $\omega\tau \gg 1$. Thus the linewidth can be written[20]

$$W^{ex} = c t^{-g} G(\omega\tau) , \tag{3}$$

where

$$\tau = \tau_0 t^{-\nu z} . \tag{4}$$

Now, as $T \to T_g$, $\tau \to \infty$, and any measurement performed in a finite time cannot diverge. Therefore in the limit $\omega\tau \gg 1$ the linewidth must approach a constant; i.e.,

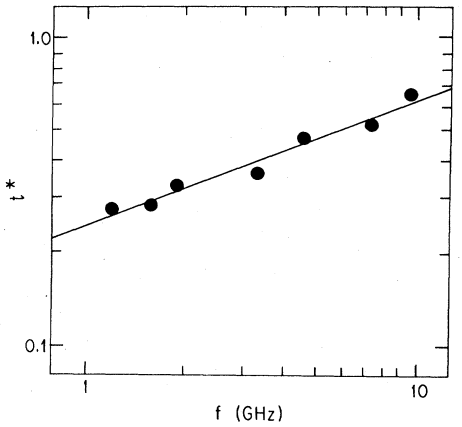

FIG. 4. Frequency dependence of t^* (frequency-dependent) temperature below which the power law of Fig. 1 fails. The slope is $(\nu z)^{-1} = 0.40$.

$t^0 \propto t^{-g} t^{\kappa\nu z}$ or

$$\kappa = g/\nu z . \tag{5}$$

The temperature t_c which is intermediate between the two limits is defined by $\omega\tau = 1$, whence

$$t_c \propto \omega^{1/\nu z} .$$

Now t^* is not quite the same as t_c, being the highest temperature at which a departure from the t^{-g} behavior is detectable. Yet t^* should have the same dependence on ω, allowing the extraction of νz. Figure 4 exhibits the frequency dependence of t^*, from which we deduce $\nu z = 2.5 \pm 0.3$. The large uncertainty is due to the imprecise definition of t^*.

The dynamic scaling hypothesis, Eq. (3), can now be tested by seeing whether the scaled width, W^{ex}/ct^{-g}, is a unique function of $\omega\tau$. The scaling plot is shown in Fig. 5. Data from all temperatures and all frequencies do, in fact, fall on a single curve. The data attains a constant

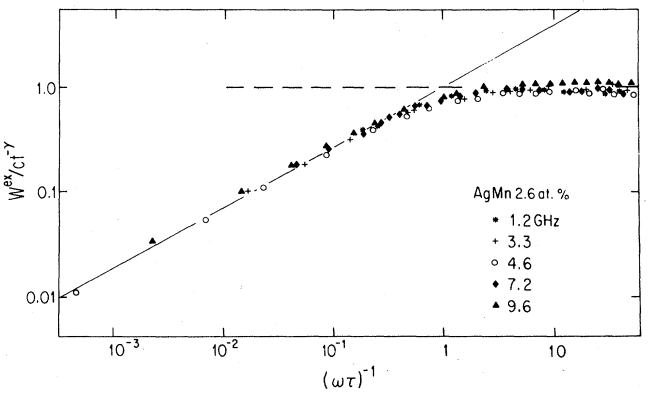

FIG. 5. Scaled width W^{ex}/ct^{-g} vs scaling parameter $(\omega\tau)^{-1}$. $c = 19$ G, $g = 1.5$. The solid line has slope $K = 0.58$, and the horizontal scale has been adjusted so that $\omega\tau = 1$ where the reduced width equals 0.8. Data from all temperatures and all frequencies fall on the same curve, $G(\omega\tau)$, demonstrating the validity of the scaling hypothesis.

for large $(\omega\tau)^{-1}$, by construction. For small $(\omega\tau)^{-1}$, the slope is $\kappa=0.58\pm0.06$, which agrees with Eq. (5): $g/\nu z=0.60\pm0.07$. The abscissa has been adjusted so that $\omega\tau=1$ when $W^{\rm ex}/ct^{-g}=0.8$, as should be expected if we assume $G(\omega\tau)=[1+(\omega\tau)^2]^{-\kappa/2}$. This sets the scale of the characteristic time to be $\tau_0=3\times10^{-12}$ sec. This τ_0 falls within an order of magnitude of the timescale deduced from T_g, $\hbar/k_BT_g=0.8\times10^{-12}$ sec. The exact value of τ_0 depends of course on the assumed form for $G(\omega\tau)$.

We now consider the case in which magnetic field scaling[12] dominates the behavior of the correlation function: $C_Q\propto t^{-g}G(H/t^{(\beta+\gamma_s)/2})$. Within experimental error the analysis goes through as before except for writing H in place of ω and $(\beta+\gamma_s)/2$ in place of νz. From Fig. 4 we find $(\beta+\gamma_s)/2=2.5\pm0.3$, yielding a "crossover" exponent[12,13] $\phi=\beta+\gamma_s=5.0\pm0.6$. This result compares favorably with values extracted from magnetization data.[13] For example, for CuMn (4.6 at. %) Malozemoff *et al.*[13] obtain $\phi=5.0\pm0.5$ from static scaling analysis. Thus field scaling also appears to be compatible with the EPR data.

Our principal result is that scaling is obeyed for all data to the smallest measured t. Previous analyses of EPR data,[9–11] while strongly *suggesting* that something special happens at T_g, have been unable to interpret the behavior below about 1.5 T_g, leaving the extrapolation to T_g open to criticism. The present scaling analysis erases such objections. Moreover, this analysis singles out a unique T_g. While $T_g=10.24$ K has been used throughout the preceding analysis, we can ask how sensitive the scaling results are to variation of T_g. Because of scatter in the data, the transition temperature T_g may be adjusted by about $\pm1°$ without significantly altering the quality of the scaling. However, variation of T_g by $\pm2°$ seriously degrades the quality of the fit. Thus a unique T_g can be identified by the EPR measurements to within about 10%, on the assumption of scaling behavior.

The nature of the transition at T_g is of course not specified by a scaling analysis. Within the Sherrington-Kirkpatrick framework, one may try to equate T_g with the de Almeida–Thouless (dAT) longitudinal instability line:[21]

$$\frac{T_{\rm dAT}(H)}{T_g(0)}=1-\left[\frac{m+2}{4}\right]^{1/3}\left[\frac{g\mu_BH}{k_BT_g(0)}\right]^{2/3},$$

with $m=3$ for Heisenberg spins. $T_{\rm dAT}(H)$ has been shown to be relevant to the static longitudinal magnetization properties of spin-glass systems.[22] However, using the field dependence of T_{dAT} (amounting to $\sim1°$ at 2000 G) introduces substantial spread into the scaling plot. The EPR data are consistent only with a field-independent, or much more weakly field-dependent transition, such as the Gabay-Toulouse line for *transverse* freezing,[23] $T_{\rm GT}(H)$. The $T_{\rm GT}(H)$ line is essentially independent of H for the fields at which we worked. This identification is physically reasonable: The narrowing process requires spin-flip events, and is therefore sensitive to fluctuations in the *transverse components* of the magnetization.[19,24,25]

Chen and Slichter[26] have measured the correlation time of Fe spins in the spin glass 2H-$NbSe_2Fe_x$ using nuclear quadrupole resonance in zero magnetic field. They found $\tau\propto t^{-2.5\pm0.5}$ for fluctuations of the z component of spin, where z is determined by crystal symmetry. Since $T_g=T_{\rm dAT}=T_{\rm GT}$ for $H=0$, their result for a very different class of material is consistent with the exponent νz reported here.

The significance of the exponent g is unclear for either type of scaling. Considering the ω case, Bruinsma[27] obtained $g=\nu z$ by straightforward averaging of the Dyaloshinski-Moriya interaction, while Henley[28] deduced $g=\nu z-2\beta$ by decoupling the four-spin correlation functions. Neither is consistent with our measurements ($g=1.5$, $\nu z=2.5$) unless β does not take its expected value[13] of 1. The interpretation of g in the field scaling case is problematic, because a recent theoretical analysis[27] of that case expresses g in terms of a new, unknown exponent associated with the anisotropy energy.

V. SUMMARY

This investigation of the Ag-Mn spin glass has shown that critical scaling can explain the frequency and temture dependence of the EPR linewidth in the paramagnetic phase. The scaling holds for all frequencies from 1 to 10 GHz and for all temperatures from within one degree of the low field magnetization cusp temperature to $>4T_g$.

The scaling analysis constitutes strong evidence in favor of a thermodynamic phase transition at T_g.

$T_g(H)$ is experimentally indistinguishable from $T_g(0)$ for all accessible magnetic fields. Within the Sherrington-Kirkpatrick framework, T_g cannot therefore be identified with the de Almeida–Thouless longitudinal instability, but is consistent with the Gabay-Toulouse prediction for transverse freezing.

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