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Dynamical susceptibility of spin glasses in the fractal cluster model

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We study the dynamical susceptibility χ of spin glasses above the critical temperature using a fractal cluster model. We derive scaling relations for the zero-field limit of the real and imaginary parts of χ which are general since they do not depend on a particular relaxation model. Comparison to data on $Eu_{0.4}Sr_{0.6}S$ yields critical exponents in good agreement with independent determinations. We discuss the different criteria which have been used to extract the critical relaxation time τ_ξ from experimental χ's. The smallness of the ratio $\beta/\nu z$ between critical exponents in the spin-glass problem justifies the approximations used to interpret and relate experimental data, for example, with the equation $\chi''=-\pi d\chi'/2d\ln\omega$.

The dynamical susceptibility has been one of the most widely studied physical properties of spin glasses.[1] It was, in fact, the very discovery of a cusp in the ac susceptibility of Au-Fe alloys as a function of temperature that suggested the existence of a phase transition in these systems.[2] Ironically, it was also the observation of a frequency dependence of the cusp temperature that started a long-standing controversy about the nature of the freezing process.[3]

In the last few years, however, scaling analysis of the field- and temperature-dependent magnetization of different spin glasses made possible the calculation of critical exponents which exhibit an impressive degree of universality.[4] On the other hand, the success of dynamic scaling in describing the critical slowing down of the fluctuations as one approaches the freezing temperature T_g, provides almost unambiguous support for the existence of a phase transition[5,6] and a critical line in the H-T plane separating spin-glass and paramagnetic phases. The agreement between the experimentally observed exponents and those obtained in sophisticated computer simulations[7] lends further support to this point of view.

The mean-field theory is capable of describing many of the features of spin glasses, including the existence of a field-dependent critical line.[8] However, the critical exponents arising from this theory are quite different from the observed ones.[4] Also, certain scaling relations, which are found to be valid experimentally,[5,6] cannot be obtained within a mean-field theory;[9] one example is $\phi=2\psi$, relating the crossover exponent ϕ to the exponent ψ which characterizes the shift ($\Delta T\propto H^{1/\psi}$) of the critical temperature with applied magnetic field.

An alternative model of the spin-glass phase transition has recently been proposed and is able to give a unified description of static and dynamic phenomena in spin glasses.[10,11] This "critical fractal cluster" model also makes clear the physical significance of scaling, which has been so successful in describing this transition.[4,10,12]

In this paper we extend further this model to treat the dynamical susceptibility $\chi(\omega)$ of a spin glass, assuming power-law dynamics. We derive novel scaling relations for χ as a function of temperature, frequency, and field in the ergodic region above the transition temperature T_g. We compare our results with experiment and obtain critical exponents in agreement with those found earlier by other methods. In the context of this model, we can also test a variety of criteria which have been used in treating experimental data in the past: for example, in extracting lines of constant relaxation time. We derive an essential requirement on scaling exponents for these criteria to work.

The basic assumption of the critical fractal cluster model is the existence of a characteristic field- and temperature-dependent cluster size (or number of spins) s_ξ, on which all physical quantities depend.[10,11] Assuming power-law dynamics, the characteristic relaxation time of the system τ_ξ is related to s_ξ by the equation $\tau_\xi=\tau_0 s_\xi^x$, where $x=z/D$, with D the fractal dimension of the typical cluster and z the conventional dynamical exponent.[11]

The typical cluster size s_ξ is given in terms of the correlation length ξ by $s_\xi=\xi^D$. Since ξ diverges at the transition temperature like $|\varepsilon|^{-\nu}$, where ν is a standard critical exponent and $\varepsilon=(T-T_g)/T_g$ is the reduced temperature, we get $s_\xi\propto|\varepsilon|^{-\nu D}=|\varepsilon|^{-\phi}$ as the transition is approached. The exponent $\phi=\nu D$ is the crossover exponent in the field-temperature plane.[10]

We should keep in mind that, in spite of the existence of a characteristic relaxation time in the system which diverges at T_g, there is, in fact, a whole spectrum of relaxation times, associated with a distribution of cluster sizes, through the relation $\tau=\tau_0 s^x$. A complete characterization of the system near the phase transition requires a knowledge of this distribution. Within the percolation model of a phase transition, the distribution of cluster sizes is given by[13]

$$n_s=s^{-\tau}f(s/s_\xi)\ , \tag{1}$$

where $\tau=2+\delta^{-1}$ is another critical exponent, not to be confused with the relaxation time. The scaling function $f(x)$ approaches a constant as $x\to 0$ and decays as $\exp(-s/s_\xi)$ for large clusters above the transition temperature.[11,13]

In order to derive the scaling relations for the dynamical susceptibility $\chi(\omega)$, we shall start by considering the simple Debye relaxation form $\chi=\chi_0/(1+i\omega\tau)$ for this quantity. Later on we shall discuss to what extent our results depend on the choice of a particular relaxation model. Integrating over cluster sizes at temperatures above T_g, we find[10]

$$\chi(\omega) \propto H^{-1} \int_0^\infty ds\, n_s s^{1/2} \tanh(s^{1/2}H/kT)/(1+i\omega\tau) \ , \quad (2)$$

where we use the tanh function rather than the Langevin function because we assume an Ising, rather than Heisenberg, spin-glass system. This is also appropriate for Heisenberg systems with local anisotropy, provided the applied field is small enough.

Let us first consider the imaginary part of the dynamical susceptibility $\chi''(\omega)$ in the limit of low fields. Using $\tau = \tau_0 s^x$ for the relaxation time of a cluster, we obtain from Eq. (2)

$$\chi''(\omega) = \frac{1}{T} \int_0^\infty \frac{ds\, s^{1-\tau} f(s/s_\xi) \omega \tau_0 s^x}{1+\omega^2 \tau_0^2 s^{2x}} \ . \quad (3)$$

Making the change of variables $y = \omega\tau_0 s^x$ and noticing that $f(s/s_\xi) = f_1(y/\omega\tau_\xi)$, where $\tau_\xi = \tau_0 s_\xi^x$, we derive the following scaling relation:

$$\chi''(\omega) \propto \frac{1}{T} (\omega\tau_0)^{\beta/\nu z} g_2(\omega\tau_\xi) \ . \quad (4)$$

Here, $g_2(x)$ is a scaling function, and we have used the scaling relations $2-\tau = -1/\delta$, $\nu D = \phi$, and $\beta = \phi/\delta$, where β, τ, δ, ϕ, ν, and D are standard critical exponents.[10,13]

For the real part of the dynamical susceptibility $\chi'(\omega)$, including the regular term, we get a similar type of equation

$$\chi'(\omega) \propto \frac{1}{T} [1 - (\omega\tau_0)^{\beta/\nu z} g_1(\omega\tau_\xi)] \ , \quad (5)$$

where we have ignored multiplicative constants, and g_1 is another scaling function.

It can be easily verified that the scaling relations given by Eqs. (4) and (5) are completely general and do not depend on the particular model susceptibility describing the relaxation dynamics of a given cluster, as long as ω and $\tau(s)$ appear in these susceptibilities multiplying each other. This holds for all well-known relaxation models, such as the Cole-Cole, Cole-Davidson, and Havriliak-Negami equations[14] and also obviously for the Debye model.

The form of the scaling functions $g_1(x)$ and $g_2(x)$ will depend, in general, on the particular relaxation model, except at the critical temperature where both $g_1(x)$ and $g_2(x)$ tend to constants, due to the fact that the scaling function $f(x)$ in Eq. (1) also approaches a constant as $T \rightarrow T_g$. As a consequence, we can write very generally

$$\chi''(\omega) \propto (\omega\tau_0)^{\beta/\nu z} \ ,$$
$$\chi'(\omega) \propto 1 - (\omega\tau_0)^{\beta/\nu z} \quad (6)$$

for $T = T_g$ independent of the model susceptibility.

Next we obtain the form of the scaling functions $g_1(x)$ and $g_2(x)$ in the $T > T_g$, low-frequency limit $\omega\tau \ll 1$. For the Debye model, we get for $\chi''(\omega)$ from Eq. (3)

$$\chi''(\omega) \propto (\omega\tau_0)^{\beta/\nu z} (\omega\tau_\xi)^{1-\beta/\nu z}/T$$
$$\propto (\omega\tau_0) \varepsilon^{-\nu z + \beta}/T \ , \quad (7)$$

which predicts a linear frequency dependence for $\chi''(\omega)$ as ω goes to zero.

Let us verify the generality of this result by considering as our model susceptibility the phenomenological Cole-Cole equation[14]

$$\chi(\omega) = \chi_0/[1 + (i\omega\tau)^{1-\alpha}] \ . \quad (8)$$

Since χ must be analytic in ω above T_g, it should be recognized that results based on this *ad hoc* nonanalytic form will be invalid at sufficiently low $\omega\tau$. Assuming some intermediate range of validity $0 < \omega\tau \ll 1$ and using Eq. (8) we obtain for the imaginary part of the dynamic susceptibility

$$\chi''(\omega) \propto \frac{1}{T} (\omega\tau_0)^{\beta/\nu z} (\omega\tau_\xi)^{1-\alpha-\beta/\nu z}$$
$$\propto \frac{1}{T} (\omega\tau_0)^{1-\alpha} \varepsilon^{-\nu z(1-\alpha)+\beta} \ . \quad (9)$$

These expressions relate the frequency dependence of $\chi''(\omega)$ to the exponent α of Eq. (8). One may also easily derive a similar equation for the real part of the dynamical susceptibility in the same limit ($\omega\tau \ll 1$) from Eq. (8)

$$\chi'(\omega) \propto [1 - (\omega\tau_0)^{1-\alpha} \varepsilon^{-\nu z(1-\alpha)+\beta}]/T \ , \quad (10)$$

which, as for $\chi''(\omega)$, turns out to be different from the Debye prediction in the same limit. In principle, relations like Eqs. (9) and (10) should allow one to distinguish between different model susceptibilities.

In Fig. 1 we present a scaling plot of the data on $\chi''(\omega,T)$ obtained by Paulsen, Williamson, and Maletta[15] on the $Eu_{0.4}Sr_{0.6}S$ spin glass according to Eq. (4) for $T \geq T_g$. The exponent $\nu z = 9 \pm 1$ obtained from the scaling plot is in good agreement with that obtained through a scaling of the phase of the dynamical susceptibility on the same system[5] in the low-frequency limit. The exponent $\beta = 1.17 \pm 0.1$ is rather large compared with what would be expected from the computer simulations on an Ising system,[7] although it turns out to be similar to the value obtained through a static scaling analysis of a Cu-Mn spin glass.[4] The error bars on the exponents are rough indications of the range of values within which the quality of the scaling does not noticeably degrade.

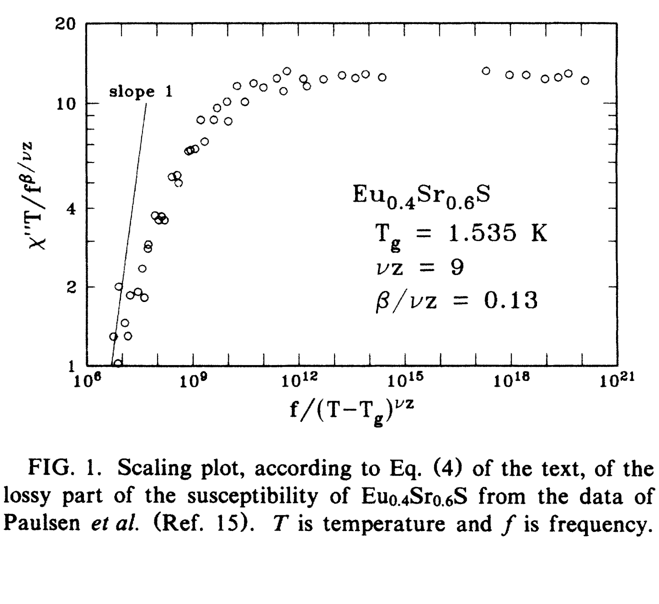

FIG. 1. Scaling plot, according to Eq. (4) of the text, of the lossy part of the susceptibility of $Eu_{0.4}Sr_{0.6}S$ from the data of Paulsen *et al.* (Ref. 15). T is temperature and f is frequency.

From Eqs. (7) and (9) we see that the inclination of the scaling curves for small values of $\omega\tau_\xi$ allows one to probe the frequency dependence of $\chi''(\omega)$ for $T > T_g$. The results are consistent with $\chi''(\omega)\propto\omega^{1/2}$, which on the basis of Eqs. (7) and (9) would rule out a Debye susceptibility in favor, for example, of a Cole-Cole equation with $\alpha=\frac{1}{2}$. Nevertheless, we believe that the data do not probe sufficiently small values of $\omega\tau_\xi$, so as to convincingly establish the limiting inclination. We point out that a linear frequency dependence of $\chi''(\omega)$ above T_g, consistent with the Debye model, has been observed[16] in the insulating spin glass $(Ti_{0.9}V_{0.1})_2O_3$.

Recent dynamical studies of spin glasses have determined the lines of constant relaxation time in the magnetic field versus temperature plane.[5,6] These lines, which in our model represent contours of constant s_ξ, are important since they allow for the determination of the critical line in the H-T plane.[5,6] Essentially two different experimental criteria have been used to determine these lines, and we shall discuss them based on our fractal cluster model.

In the first method, which can be called the constant phase method, one defines an average relaxation time τ_{av}, for $\omega\tau\ll 1$, through the phase of the dynamical susceptibility

$$\omega\tau_{av}\equiv\chi''(\omega)/\chi'(\omega) \ . \tag{11}$$

It can be easily seen using the Debye model susceptibility and the normalization condition[10] for the number of clusters $\int sn_s(s)ds=1$ for $T > T_g$, that the average relaxation time in the limit $\omega\tau\ll 1$ is given by

$$\tau_{av}\propto\tau_\xi^{1-\beta/vz}\propto\varepsilon^{-(vz)_{\rm eff}} \ . \tag{12}$$

As a consequence, the effective dynamical exponent which is obtained from this analysis is $(vz)_{\rm eff}=vz(1-\beta/vz)$. This was, in fact, already noticed by Ogielski,[7] and if we assume the validity of hyperscaling in spin glasses, one can easily check that his exponent $x=(d-2+\eta)/2z$ is identical to β/vz. Since β/vz is of order 0.1 in most cases studied experimentally or theoretically so far,[5-7] this correction is a small one in practice. The most serious problem with this criterion is that it relies on the validity of the Debye model susceptibility to describe the data as can be seen, for example, if one uses the Cole-Cole equations (9) and (10) in Eq. (11).

In the second method, lines of constant relaxation time are obtained from the inflection points of $\chi''(\omega)$ as a function of temperature.[6] Starting with the general scaling form of χ'' in Eq. (4), we find it advantageous to consider the inflection point of $T\chi''$. After some algebra, we find that this inflection point is determined by the condition

$$\omega\tau_\xi=\text{const} \ . \tag{13}$$

This simple but general result, valid for the limit of low field ($\varepsilon^\phi/H^2\rightarrow\infty$), allows a straightforward determination of the critical exponents independent of any particular model for the dynamical relaxation. For the Debye model, the constant in Eq. (13) can be evaluated exactly by making a simple approximation for the cluster distribution $f(x=s/s_\xi)$ in Eq. (1), namely, by taking it to be a step function which is $f(x)=1$ below $x=1$, and $f(x)=0$ above $x=1$. Differentiating for the inflection point, one finds

$$\omega\tau_\xi=[(vz+1-\beta)/(vz-1+\beta)]^{1/2} \ . \tag{14}$$

For typical parameters with β close to 1 and vz large, the constant in Eq. (14) turns out to be close to 1. This means that, in fact, the inflection point of $T\chi''$ corresponds quite closely to the sample intuitive condition $\omega\tau_\xi=1$, and so gives directly the relaxation time of the characteristic cutoff cluster size as the inverse of the known experimental angular frequency ω.

In practice, this criterion has usually been used without taking into account the regular term of the susceptibility, that is, by using the inflection point of χ'' rather than of $T\chi''$. We can test the validity of this approximation in the context of the Debye model and the step function approximation for $f(s/s_\xi)$. Differentiating χ'' of Eq. (2), we obtain two terms: The first term gives $-\chi''/T$ and represents the effect of the regular term in the susceptibility. The second term comes from differentiating the integral in Eq. (2). Evaluating these expressions in the limit $\delta x\gg 1$ or equivalently $\beta/vz\ll 1$, we find the first term proportional to $\pi/4$ and the second to $\phi x=vz$. Thus, provided β/vz is small (as is usually the case in simulation and experiment) it is reasonable to ignore the T factor and determine the inflection point simply from χ''.

The above results can be extended to finite fields. For example, in the limit $H^2/\varepsilon^\phi\rightarrow\infty$, that is, in the field-dominated critical region of the H-T plane, we can again evaluate the constant of Eq. (13) exactly by using the Debye relaxation and the step function approximation for $f(s/s_\xi)$. We must now start with Eq. (2) and consider the behavior of the tanh function. This depends on the parameter[10] $p=s_\xi^{1/2}H/kT$, which is a constant independent of field in the field-dominated critical region since $s_\xi\propto 1/H^2$. A value greater or less than 1 reflects a greater or lesser tendency of the largest clusters to saturate. In particular, if p is much less than 1 the tanh function never saturates over the range of integration of cluster sizes from 0 to s_ξ. Then the inflection point calculation gives the same result as in Eq. (14). On the other hand, if p is much larger than 1 most clusters which contribute to the relaxation are saturated, and one can approximate the tanh function by 1. The calculation then gives a result identical to Eq. (14), but with β replaced everywhere by $\beta+\phi/2$. Intermediate values of the parameter p will give intermediate results. Since $\phi/2$ is also small compared to vz in typical cases, these results lead to the general conclusion that in this limit, the inflection point criterion is a good approximation for determining $\tau_\xi\simeq 1/\omega$.

The same limit of $\beta/vz\ll 1$ also justifies a widely used approximate relation between χ'' and the logarithmic frequency derivative $d\chi'/d\ln\omega$ of the in-phase susceptibility χ'. Taking χ' from Eq. (2) in the limit of zero field, differentiating with respect to $\ln\omega$, and integrating over s, we find for $\omega\tau_\xi\gg 1$

$$-\frac{\chi''}{d\chi'/d\ln\omega}=\pi/2\sin\left[\pi\left(1-\frac{\beta}{vz}\right)/2\right]\Gamma\left(1-\frac{\beta}{2vz}\right)\Gamma\left(1+\frac{\beta}{2vz}\right) . \tag{15}$$

In the limit $vz \gg \beta$, the right-hand side reduces to $\pi/2$, which is precisely the original result of Lundgren, Svendlindh, and Beckman,[17] and which has at least been confirmed approximately in experiment.[15,17]

The time dependence of the magnetization is, of course, closely related to the dynamic susceptibility. Treating a generalized cluster distribution model with Debye relaxation, Lundgren *et al.* derived the relation[17]

$$\frac{dM}{d\ln t} \propto \frac{H}{T} \int_{\tau_{\min}}^{\tau_{\max}} d\ln\tau\, m_0(\tau) g(\tau) e^{-t/\tau} t/\tau \ , \quad (16)$$

where $m_0(\tau)$ is a cluster magnetization, and $g(\tau)$ is a weighting factor for clusters with relaxation time τ. Assuming the distribution function $m_0(\tau)g(\tau)$ was slowly varying with τ, they took it out of the integral and so argued that one could determine the distribution function directly from the experimentally measured $dM/d\ln t$. We can now test this approximation in our model. In our case $m_0(\tau)$ and $g(\tau)$ can be shown to be $\tau^{2/x}$ and $\tau^{-(2/x)-(1/\delta x)}$, respectively; so indeed the product, going as $\tau^{-1/\delta x}$, is weakly varying in the limit $1/\delta x = \beta/vz \ll 1$.

Explicitly, the result for the limit of experimental times t, short compared to the longest relaxation time, is

$$dM/d\ln t \propto \frac{H}{T} \Gamma\left(1+\frac{\beta}{vz}\right) / t^{\beta/vz} \ . \quad (17)$$

For increasing temperatures corresponding to decreasing maximum relaxation times, the integral in Eq. (16) gives a curve which drops off from the value of Eq. (17) towards zero, with an inflection point corresponding to $t \simeq \tau_\xi$. Thus such a curve can be described as a smeared-out step function, or more crudely as a wave, which advances to the right as the measurement time is shortened. In view of the measurement-time dependence in Eq. (17), the crest of the wave is very weakly dependent on this time. These features describe the experimental observations quite well.[17] At longer times, a stretched exponential behavior is expected to develop, as discussed elsewhere.[11]

We conclude that our fractal cluster model is quite successful in describing many, if not all, aspects of the dynamical spin-glass susceptibility and its counterpart, the time-dependent magnetization relaxation. Typically, these properties have been previously interpreted in terms of generalized cluster distributions. The language of those earlier cluster models often pointed towards the early Néel cluster-blocking picture, which differs in a fundamental way from our model by having no phase transition. Our model shows how the phase-transition picture can, in fact, incorporate many of the properties previously taken as evidence against a phase transition. In this sense it bridges between the two early perspectives in the study of spin glasses to achieve a new synthesis.

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