## **VOLUME 33, NUMBER 5**

1 MARCH 1986

## Dynamic scaling and the field-dependent critical line in a fractal cluster model of spin glasses

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(Received 20 December 1985)

We extend an earlier fractal cluster model of spin glasses to study (1) the magnetization relaxation M(t) induced by a magnetic field, (2) magnetic noise, and (3) dynamics near a critical line  $T_g(H)$ . Above the zero-field transition temperature  $T_g$ , M(t) follows a stretched exponential form  $\exp(-t^{1-n})$  with  $n = vz/(\phi + vz)$ , where  $\phi$ , v, and z are standard static and dynamical critical exponents. A dynamic scaling relation is derived for the entire region  $T_g(H) < T < T_g$  in agreement with experiments. The equations associated with lines of constant relaxation time are obtained, and it is shown how nonuniversality of exponents along the critical line can be tested.

Recently, there has been a lot of interest in the study of spin-glass dynamics both theoretically<sup>1-4</sup> and experimentally.<sup>5-16</sup> At the same time a critical fractal cluster model of spin glasses has been proposed which is able to describe the essential features of the phenomena occurring near a spinglass phase transition and to account for the static critical exponents which experimentally turn out to be different from those predicted by mean-field theory.<sup>17</sup> The basic assumption of this fractal model is the existence of a temperature- and magnetic-field-dependent characteristic cluster size  $s_{\xi}$  on which all relevant physical quantities depend and which diverges at the transition temperature  $T_g$ . It is related to the correlation length  $\xi$  and the cluster fractal dimension D by  $s_{\xi} \propto \xi^{D}$ .

In this Rapid Communication we extend this critical fractal cluster model to describe several aspects of spin-glass dynamics which are of current interest: (1) stretched exponential relaxation,  $^{3,4,9,10,12,15,16}$  (2) magnetic noise,  $^{12,13}$ and (3) scaling<sup>8,11,14</sup> above the field-dependent critical line  $T_g(H)$ , where conventional equilibrium dynamics can be assumed to apply. Although the critical cluster model predicts complementary behavior from finite-size clusters below the critical line, this dynamic behavior is in most cases masked by the slow dynamics of the infinite cluster which we do not consider here.

We start by introducing the relaxation time of a cluster of size s which we assume to be given by  $\tau = \tau_0 s^x$ , where  $\tau_0$  is a constant and x a critical exponent related, as shown below, to the exponent z introduced earlier in a generalization of the standard dynamical scaling hypothesis<sup>8</sup> to spin glasses. In the fractal model then, the slowing down of the dynamical processes, as the critical temperature is approached, is due to the growth of the characteristic cluster size  $s_{f}$ .

Since  $s_{\ell} \propto \xi^D$  and  $\xi \propto |\epsilon|^{-\nu}$ , where  $\epsilon = (T - T_g)/T_g$  is the reduced temperature and  $\nu = \phi/D$  is the usual critical exponent associated with the correlation length, we find  $s_{\ell} \propto |\epsilon|^{-\nu D}$ . The relaxation time of the characteristic cluster is  $\tau_{\xi} = \tau_0 s_{\xi}^{*}$ , and consequently,  $\tau_{\xi} = \tau_0 |\epsilon|^{-\nu Dx}$ . Comparing this result with the standard dynamical scaling hypothesis which assumes<sup>8</sup>  $\tau_{\xi} \propto |\epsilon|^{-\nu z}$ , we obtain the desired relation z = Dx between the dynamical exponents.

In an Ising fractal cluster model, the magnetization M at a temperature T induced by an external magnetic field H is an average over all cluster sizes:

$$M \propto \int_0^\infty ds \ n_s s^y \tanh(\mu_0 s^y h) \ . \tag{1}$$

In this equation h = H/kT is the reduced field and the exponent y = 1 and  $\frac{1}{2}$  for the ferromagnet and spin glass, respectively.<sup>17</sup> According to scaling theory, the cluster size distribution  $n_s$  (number of clusters with s spins per magnetic site) is given by<sup>18</sup>

$$n_{\mathbf{s}} = \mathbf{s}^{-\tau} f(\mathbf{s}/\mathbf{s}_{\boldsymbol{\xi}}) \quad , \tag{2}$$

where  $\tau = 2 + \delta^{-1}$  is another critical exponent<sup>18</sup> (not to be confused with the relaxation time  $\tau$ ), and f(x) is a scaling function which approaches a constant as  $x \to 0$ , but drops off towards zero above x = 1. Clusters are assumed to be independent, interactions being taken into account by the growth with decreasing temperature of the clusters themselves.<sup>17</sup> We also ignore here transverse ordering and Heisenberg behavior, assuming that local anisotropy effects predominate at low fields.<sup>1</sup>

Let us now calculate the time decay of the magnetization induced in a spin glass by a weak magnetic field which is turned off at t=0. We get

$$\frac{M(t)}{M(0)} = \frac{\int_0^\infty s^{2y-\tau} f(s/s_{\xi}) e^{-\omega_0 t s^{-x}} ds}{\int_0^\infty s^{2y-\tau} f(s/s_{\xi}) ds} \quad , \tag{3}$$

where  $\omega_0 = \tau_0^{-1}$ , and we assumed that the relaxation of a given cluster is exponential. From the equation above we can derive the following scaling expression for the time dependence of the magnetization:

$$\frac{M(t)}{M(0)} = g(\omega_0 t/s_{\xi}^{x}) = g(\omega_0 t/|\epsilon|^{-\nu z}) \quad . \tag{4}$$

The last equality was obtained using z = Dx and  $s_{\xi} \propto |\epsilon|^{-\nu D}$ . This is an important equation since it shows that although the system has a distribution of relaxation times, there is a single characteristic time, which is the relaxation time of the typical cluster, that determines the overall time scale of the dynamical processes. This makes explicit the underlying assumption of earlier scaling treatments.<sup>8, 11, 14</sup>

We now calculate the specific form of the relaxation of the magnetization in the long-time limit. For this purpose we consider that as in the percolation model of a phase transition,<sup>18</sup>  $f(s/s_{\ell}) \propto \exp[-(s/s_{\ell})^{\nu}]$  for sufficiently large clusters, close to  $T_g$  and ignoring algebraic prefactors. The exponent is  $\nu = 1$  and (d-1)/d, where d is the dimension of the system, at temperatures above and below the transition, respectively.<sup>18</sup> The relaxation of M(t), after the small ap3592

plied field is turned off at t = 0, is then given by

$$M(t) \propto \int_0^\infty ds \exp[-(s/s_{\xi})^{\nu} - \omega_0 t s^{-x}] \quad . \tag{5}$$

An expression similar to the equation above has been previously<sup>19</sup> obtained in a study of the relaxation function of conventional glasses. It can be evaluated in the limit of long times to yield

$$M(t) \propto e^{-(t/\tau_1)^{1-n}}, \quad n = x/(x+v) = vz/(vz + \phi v) \quad , \qquad (6)$$

with  $\tau_1 \propto s_{\tau}^{x}$  as expected from Eq. (4). Since v decreases from 1 to  $\frac{2}{3}$  in three dimensions (3D) as the system is cooled from above to below the transition, the exponent *n* increases with decreasing temperature.

Equation (6) is of stretched exponential or Kohlrausch form, which has recently been found to describe the relaxation of the magnetization in spin glasses, both experimentally<sup>9,10,12,15,16</sup> and theoretically.<sup>3,4</sup> Initial attention focused on the nonergodic region below  $T_g$  and a derivation of this relaxation form was made in the context of the nonergodic Parisi solution for the mean-field spin glass.<sup>14</sup> However, our result highlights the fact that the same kind of relaxation can be obtained above  $T_g$  in a region described by equilibrium dynamics. Recently, the first observation<sup>16</sup> of stretched exponential relaxation in a spin glass above  $T_g$  has been made on Eu<sub>0.4</sub>Sr<sub>0.6</sub>S, where n = 0.9 below  $T_g$  and decreases slowly above  $T_g$ . Using dynamic scaling results on the same system,<sup>8</sup> which give  $\phi = 4$  and  $\nu z = 7.2$ , we predict n = 0.64 above  $T_g$  and n = 0.73 below  $T_g$  in rough agreement with experiment. Numerical calculations<sup>3</sup> of a 3D Ising model have given n = 0.65 at  $T_g$  (see Fig. 11 of Ref. 3), decreasing linearly above  $T_g$ . In the simulation  $\nu z = 7.9$ , and if we assume  $\phi = 4$ , we obtain the same predictions for *n*.

Equation (6) gives for the first time the stretched exponential power in terms of critical exponents in spin glasses. While the order of magnitude and trend are correct, the observed continuous change in n as a function of temperature needs to be understood, which probably requires taking into account crossovers to power-law relaxation (see below) and finite-field effects. Another interesting point is that the derivation of Eq. (6) is valid both for ferromagnets and for spin glasses in the cluster model. Usually, relaxation behavior of the type predicted by Eq. (6) is difficult to see in ferromagnets because of the exceedingly small temperature range near  $T_g$ , where the time constants are experimentally accessible. However, recently such relaxation has been seen<sup>15</sup> in the reentrant ferromagnet Eu<sub>0.54</sub>Sr<sub>0.46</sub>S, giving a value n = 0.85.

In the limit of linear response and equilibrium thermodynamics, Ocio, Bouchiat, and Monod<sup>13</sup> have shown that from a knowledge of the stretched exponential power 1 - n, a magnetic fluctuation noise-power spectrum going as  $\omega^{n-2}$ can be derived for frequencies greater than the characteristic frequency  $\omega = 1/\tau_{\xi}$ . Thus, we predict a noise spectrum going as

$$S(\omega) \propto \omega^{-(\nu z + 2\phi \nu)/(\nu z + \phi \nu)}$$
(7)

which gives  $\omega^{-1.36}$  for  $T > T_g$  in Eu<sub>0.4</sub>Sr<sub>0.6</sub>S. No experimental results have yet been reported above  $T_g$ . This is the first prediction relating the noise spectrum to the critical exponents, although 1/f noise has previously been derived<sup>20</sup> using distributed activation barriers as appropriate for  $T < T_{\mathbf{g}}$ .

Let us now study the relaxation of the magnetization of a spin glass at the transition temperature. At  $T_g$  the cluster size distribution is given by  $n_s = s^{-r}$ , since the characteristic cluster size is infinite. The equation describing the relaxation of the magnetization after a small field has been turned off at t=0 is

$$M(t) \propto \int_0^\infty ds \ s^{1-\tau} e^{-\omega_0 t s^{-x}} \propto \left(\frac{1}{\omega_0 t}\right)^{D/\delta z} = \left(\frac{1}{\omega_0 t}\right)^{\beta/\nu z}, \quad (8)$$

where we used the standard scaling relations  $\beta \delta = \phi = \nu D$ for the critical exponents,<sup>18</sup> and z = Dx as obtained before. The divergence of M(t) at t = 0 is unphysical and can be traced back to the assumption of s going continuously to zero in the integral. In the frequency domain, Eq. (8) gives rise to a noise spectrum<sup>13</sup> varying as  $\omega^{\beta/\nu z - 1}$ .

Notice that the exponent which appears in this equation is that of the order parameter  $(\beta)$  and not that of the susceptibility  $(\gamma)$ . This is characteristic of spin-glass response and arises because  $y = \frac{1}{2}$  instead of 1 as in ferromagnets.<sup>17</sup> This algebraic type of relaxation at the transition temperature is a standard feature of critical phenomena and in our model it is a direct consequence of the form of the cluster size distribution at  $T_g$  and of our assumption of a cluster relaxation time which is a function of its size. Equations (7), (8), and (2) also illustrate the algebraic nature of the relaxation associated with small clusters ( $s \ll s_{\xi}$ ). The exponent  $\beta/\nu z$ can be easily estimated. If we take  $\beta = 0.5$  as in 3D Ising simulations,<sup>3</sup> we predict  $M(t) \propto t^{-0.063}$ , in agreement with the numerically observed exponent of -0.06 at  $T_g$ . The noise spectrum would be close to 1/f. On the other hand, most experiments seem to show stretched exponential relaxation at  $T_{\mathbf{g}}$ . Here we believe the effects of finite field, ignored in the simulations and in our derivation of Eq. (8), will be important. Where power-law relaxation has been observed,  $^{13}$  as in CsNiFeF<sub>6</sub>, there is good agreement between the M(t) exponent and the noise spectrum prediction, but so far these observations are all below  $T_g$ . We should also point out that the small exponent  $\beta/\nu z$  makes it difficult to distinguish the algebraic decay from other forms of relaxation.

The form of Eqs. (6) and (8) for M(t) leads us to suggest the following interpolation expression to describe the relaxation of the magnetization in a spin glass:

$$M(t) \propto t^{-\beta/\nu z} \exp[(-at|T - T_g|^{\nu z})^{1-n}] , \qquad (9)$$

where a is a constant. In this case, the relaxation crosses over from exponential to algebraic as the transition is approached. An expression like the one suggested above has been found to describe the time-dependent magnetic correlation function of a spin glass simulated in a special purpose computer.<sup>3</sup> The temperature dependence of the exponents which was found there may be a consequence of the interpolation character of Eq. (9) which gives rise to effective exponents.

In order to get a deeper understanding of the dynamics of a spin glass and its response to an external magnetic field, it is crucial to have a detailed knowledge of the behavior of this system in the H-T plane and in particular the location of any phase transition line, such as the de Almeida-Thouless line<sup>21</sup> of the infinite-range Ising model, separating the spin-glass and paramagnetic phases. We shall now extend

$$T_{g}(H) = T_{g}(1 - gH^{1/\psi})$$
, (10)

or alternatively  $\epsilon_{gH} = gH^{1/\psi}$ , where  $\epsilon_{gH} = (T_g - T_g(H))/T_g$ ; g is a constant, and  $\psi$  an exponent characterizing the shift in the critical temperature due to the magnetic field.<sup>22</sup>

The scaling expression for the characteristic cluster size can be generalized for the case of finite fields in the standard way:<sup>17</sup>

$$s_{\ell} \propto |\epsilon|^{-\phi} f_{\ell}(H^2/|\epsilon|^{\phi}) \quad , \tag{11}$$

where  $\phi = \nu D$  is the crossover exponent, and  $f_{\xi}(x)$  is a scaling function. In the conventional "analytic" regime for  $T > T_g$  and  $y = H^2/\epsilon^{\phi} \rightarrow 0$ , the function  $f_{\xi}(y)$  goes as 1-y. In the field-dominated regime, where  $|y| = H^2/|\epsilon|^{\phi} \rightarrow \infty$ ,

$$f_{\xi}(|y|) \to |y|^{-1}(1 \mp |y|^{-1/\phi})$$
, (12)

with  $\mp$  referring to  $T \gtrless T_g$ , respectively. These forms insure the proper behavior of the relaxation time, as will be seen below.

It remains to define the form of  $f_{\xi}(y)$  as the critical line is approached from above. Since  $s_{\xi}$  diverges on this line, one can write in general<sup>22</sup>

$$s_{\boldsymbol{\xi}} \propto A(H) |\boldsymbol{\epsilon}_{H}|^{-\phi}, \quad \boldsymbol{\epsilon}_{H} = [T_{\boldsymbol{g}}(H) - T]/T_{\boldsymbol{g}} \quad .$$
 (13)

Although there is some evidence for nonuniversality of dynamic exponents,1 we will assume that the static exponents do not depend on field. In this case  $\phi = \phi$ , and A becomes independent of H (see below). The generalized scaling assumption of Eq. (11), together with Eq. (10), implies the important relation  $\phi = 2\psi$ , relating the crossover exponent and the exponent  $\psi$  characterizing the shift in the critical temperature by the magnetic field, independently of the universality assumption.<sup>22</sup> This relation can be directly verified substituting Eq. (10) for the critical line in Eq. (13) and requiring that  $s_{\ell}$  be independent of temperature to lowest order in the field-dominated regime. Conversely, it can be shown that the validity of  $\phi = 2\psi$  guarantees that  $s_{\xi}$ is independent of temperature in this regime, even without universality. The relationship  $\phi = 2\psi$  has recently been confirmed in  $Eu_xSu_{1-x}S$  (Refs. 8 and 17) and in an amorphous spin glass.<sup>11</sup>

A convenient expression for the typical cluster size near the critical line is  $s_{\xi \alpha} |H - H_c|^{-\phi}$ , where the critical field  $H_c(T_0)$  at a given temperature  $T_0$ , is defined by Eq. (10) with  $T_g(H) = T_0$ . This is a useful equation when approaching the critical line at constant temperature.

From Eqs. (11) and (13) we can now derive the form of the scaling function  $f_{\xi}$  near the critical line:

$$f_{\xi}(|y|) = |y|^{-\dot{\phi}/\phi} (g - |y|^{-1/\phi})^{-\dot{\phi}}, \quad |y| \to g^{-\phi} \quad . \tag{14}$$

Clearly this diverges when  $|y| = H^2/|\epsilon|^{\phi} = g^{-\phi}$ , which is equivalent to the critical line of Eq. (10). The amplitude relation can also be explicitly obtained and is given by  $A(H) \propto H^{-2(1-\phi/\phi)}$ . Let us compare this result to that of Eq. (12) in the field-dominated region. Since  $|y| \rightarrow \infty$  in this region, Eq. (12) can be rewritten, to first order in  $|y|^{-1/\phi}$  as

$$f_{\xi}(|y|) \to |y|^{-1}(1 \pm \phi^{-1}|y|^{-1/\phi})^{-\phi}, |y| \to \infty$$
, (15)

where  $\pm$  refer to  $T \gtrless T_g$ . Here we have kept the factor  $\phi^{-1}$  even though this form and that in Eq. (12) ignore additional constant factors. Comparing Eq. (14) (with  $\phi = \dot{\phi}$ ) and Eq. (15), we now see that aside from these constant factors, the form of  $f_{\xi}(y)$  as given by Eq. (14) is unchanged throughout the range  $T < T_g$ , and in the simplest case, the constants are the same in both regions. As we shall see below, this simple case appears to be observed in experiments.

Let us now relate these results to experiment by using the basic dynamic scaling relationship  $\tau_{\xi} = \tau_0 s \xi$ . Obviously, this relationship implies that critical slowing down occurs as the typical cluster size diverges approaching the critical line. It also implies that lines of constant characteristic relaxation time  $\tau_{\xi}$ , which are accessible experimentally (see below), are also lines of constant  $s_{\xi}$ . From Eqs. (11) and (13), these lines are given by

$$(\tau_{\boldsymbol{\xi}}/\tau_0)^{1/\boldsymbol{x}} = |\boldsymbol{\epsilon}|^{-\boldsymbol{\phi}} f_{\boldsymbol{\xi}}(H^2/|\boldsymbol{\epsilon}|^{\boldsymbol{\phi}}) \quad , \tag{16}$$

or alternatively, near the critical line,

$$(\tau_{\xi}/\tau_0)^{1/x} = \{[(T - T_g(H)]/T_g\}^{-\phi} .$$
(17)

Equation (16) implies the well-known result that, in the analytic regime  $H^2/\epsilon^{\phi} \ll 1$ , lines of constant  $s_{\xi}$  or  $\tau_{\xi}$  leave the temperature axis perpendicularly and then bend over as  $H^2$ :

$$T_f(H) = T_f(0) - \{C / [T_f(0) - T_g]^{\phi - 1} \} H^2 \quad , \tag{18}$$

where C is a constant and  $T_f(H)$  is the characteristic temperature for a given field and frequency (or relaxation time). At H=0,  $T_f(0) - T_g \propto \tau_{\xi}^{-1/\nu z}$ , so as might be expected, the curvature increases as  $\tau_{\xi}$  increases and  $T_f(0)$  approaches  $T_g$ . Equation (16), coupled with Eq. (12), also implies that at  $T_g$  ( $\epsilon=0$ ),  $s_{\xi} \propto H^{-2}$ , and on a line of constant  $\tau$ ,  $H \propto \tau^{-1/2x}$  and  $dH/dT \propto H$ . This means that the lines of constant  $\tau$  are linear in the H-T plane as they cross the vertical axis at  $T_g$ . As  $\tau$  increases they cross at lower fields and with lower slope. These features are well known in experimental studies of several spin-glass systems, <sup>6,8,11,14</sup> where  $\tau_{\xi}$  is taken as  $1/\omega$  ( $\omega$  is the frequency of the ac susceptibility measurement), and where  $T_f(H)$  is determined, for example, from the inflection point of the out-of-phase susceptibility  $\chi''$  at a fixed frequency.

Equation (11) with  $s_{\xi} = \text{const}$  relates the magnetic field and temperature along the lines of constant relaxation time. Implicit differentation of this equation shows that in the case where these lines have inflection points these points should fall on a curve where  $H \propto |\epsilon|^{\phi/2}$ , which is consequently governed by the same exponent  $\psi = \phi/2$  of the critical line. This curve joining the inflection points of the lines of constant relaxation time constitutes an alternative, precise definition of a crossover line.

Of special interest is the determination of the critical line  $T_g(H)$ , which has been attempted in a number of studies recently. Normalizing to the zero-field limit<sup>8,11,14</sup> one can transform Eq. (11) with Eqs. (14) and (15) into

$$|\epsilon_{f0}/\epsilon_{fH}| = (H^{2/\phi}/|\epsilon_{fH}|) - 1 \quad . \tag{19}$$

Here  $\epsilon_{f0}$  and  $\epsilon_{fH} = [T_f(H) - T_g]/T_g$  are the reduced temperatures at zero and finite fields, respectively, deduced from a criterion like the inflection point of  $\chi''$  at a frequency f. In Eq. (19) we have neglected constant factors. By contrast, if we use the nonuniversal form of Eq. (13) for  $s_{\epsilon}$ 

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near the critical line, we obtain in the same way

$$|\epsilon_{f0}/\epsilon_{fH}|^{\phi/\dot{\phi}} = (H^2/|\epsilon_{fH}|^{\phi})^{1/\dot{\phi}} - (H^2/|\epsilon_{fH}|^{\phi})^{1/\dot{\phi}-1/\phi} .$$
(20)

It is obvious that Eq. (20) reduces to Eq. (19) as  $\dot{\phi} \rightarrow \phi$ .

Comparing Eqs. (19) and (20), it is apparent that the linearity of scaling plots of  $|\epsilon_{f0}/\epsilon_{fH}|$  as a function of  $(H^{2/\phi}/|\epsilon_{fH}|)$  can test the universality hypothesis (see Fig. 1). However, caution should be exercised because in the high-field limit, an equation similar to Eq. (19) [but with +1 instead of -1, see Eq. (12)] applies for  $T > T_g$ . It is tempting to use such an equation<sup>14</sup> to extrapolate to  $|\epsilon_{f0}/\epsilon_{fH}|=0$ , where  $\tau \to \infty$  and  $\epsilon_{gH} = gH^{2/\phi}$ ; however, such a formal determination of the critical line does not, in fact, test the critical behavior at all except at H=0. Two studies<sup>8,11</sup> have explored the  $T < T_g$  regime and have indeed found linear scaling behavior as predicted by Eq. (19), with no change in slope over the entire range of data for  $T < T_g$ .

Our analysis, coupled with these results, has thus brought out the following new points: (1) While the linear scaling behavior of Eq. (19) was originally found empirically, we have derived it theoretically from the assumption of a critical line  $\epsilon_{gH} \propto H^{2/\phi}$  and of universality in the exponents along the critical line. (2) Our derivation does not rule out the possibility of a change of slope in Eq. (19) as the critical line is approached, but such a change of slope can only affect data at times longer than conveniently accessible in present experiments. (3) The fact that the scaling behavior follows Eq. (19) rather than (20) provides evidence for universality



FIG. 1. Dynamic scaling plot of  $Eu_{0.4}Sr_{0.6}S$  data by Bontemps et al. (Ref. 8) (circles), and of Eq. (20) for different values of  $\dot{\phi}/\phi$  (same units as Fig. 3 of Ref. 8). Deviations from a straight line indicate nonuniversality of exponents.

## of exponents.

In summary, the critical cluster model of spin glasses provides a simple framework for understanding many static and dynamic properties of spin glasses.

This work was begun in close collaboration with B. Barbara and S. Barnes.

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