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Relaxation behavior of fractal-cluster spin glasses

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The relaxation behavior of spin glasses has been analyzed in the framework of the critical fractal-cluster model. It is found that the *relaxation rate* of the magnetization can be expressed as $\partial m/\partial \ln t \propto t^{-\beta/z\nu}\exp[-(t/\tau_\xi)^{\beta\delta/z\nu}]$, where β,δ,z,ν are standard static and dynamic critical exponents and τ_ξ is the relaxation time of a characteristic cluster which diverges at the spin-glass freezing temperature T_g. The expression is valid through T_g and is found to describe excellently the fundamental features of the relaxation in real spin glasses.

A characteristic feature of spin glasses is the very wide time range for the relaxation process.[1] Relaxation occurs continuously from atomic time scales to a characteristic time, which in the vicinity of the spin-glass freezing temperature goes beyond laboratory time scales. In the linear-response regime the relaxation of the order parameter $q(t)$ may be probed by ac susceptibility, zero-field-cooled, and thermoremanent magnetization measurements. Unfortunately, not only are experiments on spin glasses hampered by the restriction to just a small time segment of the total relaxation with one experimental probe, but also they are generally far too ill defined regarding the influence of the aging process[2,3] and magnitude of the external magnetic field.[4] Relaxation in spin glasses is likely to be more universal than implied by the profound dissimilarities reported in the literature. A general understanding of the static and dynamic properties of spin glasses is given by the critical fractal-cluster model, introduced by Malozemoff and co-workers.[5] Within this unusually descriptive model, Continentino and Malozemoff[6] (CM) derived the relaxation behavior of the magnetization in spin glasses in the limit of zero field. It was particularly emphasized that the relaxation above the spin-glass freezing temperature should follow a stretched-exponential form. In this Rapid Communication we argue that the stretched exponential form for the relaxation only refers to a minute fraction of the total relaxation and that the main conclusion by CM does not give full credit to the critical fractal-cluster model. Instead, within the same model, we propose a relaxation function which accounts for the total relaxation including the profound nature of the equilibrium approach. Our relaxation function is valid through the spin-glass freezing temperature and is found to describe excellently the general features of the relaxation in real spin-glass systems.

In the critical fractal-cluster model the relevant physical quantities are governed by a characteristic cluster size s_ξ which is related to the correlation length ξ through $s_\xi \propto \xi^D$, where D is the fractal dimensionality of the cluster. The cluster-size distribution n_s (number of clusters with s spins) is given by

$$n_s = s^{-2-1/\delta} f(s/s_\xi) \ , \tag{1}$$

where δ is a standard static critical exponent and $f(s/s_\xi)$ is a distribution function, which close to T_g may be expressed as $f(s/s_\xi)\propto\exp(-s/s_\xi)$. Assuming an exponential decay for the moment of each cluster, CM derive the time decay of the remanent magnetization $M(t)$ in the limit of zero field:

$$M(t) = C\int n_s s \exp(-t/\tau) ds \ . \tag{2}$$

C is a constant and τ is the relaxation time of a cluster, related to its size through $\tau=\tau_0 s^x$, where τ_0 is a constant and x a critical exponent. Following CM, x is related to the dynamic exponent z through $z=Dx$ and can also be expressed in standard critical exponents as $x=z\nu/\beta\delta$. The normalizing criterion is

$$M(0) = C\int n_s s \, ds \ . \tag{3}$$

We reformulate the equations above in terms of relaxation times τ. The general equation for $M(t)$ can then be written as

$$M(t) = C\int_{\tau_0}^{\infty} (1/x)(\tau/\tau_0)^{-1/\delta x} \times \exp(-(\tau/\tau_\xi)^{1/x})\exp(-t/\tau) d\ln\tau \ . \tag{4}$$

In the normalization we put $f(\tau/\tau_\xi)=1$ for $\tau<\tau_\xi$ and $f(\tau/\tau_\xi)=0$ for $\tau>\tau_\xi$. Integration from the minimum relaxation time τ_0 (which we interpret to be the single spin-flip time, 10^{-13} sec) and the maximum relaxation time τ_ξ gives

$$M(0) = C\int_{\tau_0}^{\tau_\xi} (1/x)(\tau/\tau_0)^{-1/\delta x} d\ln\tau \ , \tag{5}$$

which becomes

$$M(0) = C\delta(1-(\tau_\xi/\tau_0)^{-1/\delta x}) \ . \tag{6}$$

Equation (4) can be written in the abbreviated form

$$M(t) = \int_{\tau_0}^{\infty} g(\tau)\exp(-t/\tau) d\ln\tau \ , \tag{7}$$

which is analogous to the expression found[7] from the phenomenological picture[8,9] prescribing a wide distribution of relaxation times. The fractal-cluster model gives an analytic expression for the distribution of relaxation times $g(\tau)$, which varies slowly with $\ln\tau$ (since the exponent $1/\delta x\sim 0.1$) and exhibits a well-defined maximum relaxation time τ_ξ. From the basic assumption of a wide distribution of relaxation times, it has previously been shown that the relaxation rate $[(1/H)\partial M/\partial\ln t]$ in zero-field-cooled (ZFC) and thermoremanent magnetization

(TRM) measurements,[7] and the imaginary part $\chi''(\omega)$ of the dynamic susceptibility[8] directly reflect the density of relaxation times $g(\tau)$ at the experimental observation time. In ZFC and TRM measurements the observation time equals t and in dynamic susceptibility measurements $1/\omega$. By taking the partial derivative of $M(t)$ with respect to $\ln t$ in Eq. (7) one obtains

$$\partial M(t)/\partial \ln t = -\int_{\tau_0}^{\infty} g(\tau)(t/\tau)\exp(-t/\tau)d\ln\tau \ . \quad (8)$$

Since the integrand is highly peaked around $t=\tau$ we bring out $g(\tau)$ from the integral with the value $g(t)$. After integration we obtain the approximate result

$$\partial M(t)/\partial \ln t \simeq -g(t) \ . \quad (9)$$

Using the analytic expression for $g(t)$, the normalization criterion [Eq. (6)], and the relation $x=z\nu/\beta\delta$, the expression for the relaxation rate, valid through T_g, becomes

$$[1/M(0)]\partial M/\partial \ln t = -(\beta/z\nu)(1-(\tau_\xi/\tau_0)^{-\beta/z\nu})^{-1}(t/\tau_0)^{-\beta/z\nu}\exp(-(t/\tau_\xi)^{-\beta\delta/z\nu}), \quad \tau_\xi=\tau_0(T/T_g-1)^{-z\nu} \ . \quad (10)$$

The equation yields a slow power-law decay followed by a sudden disappearence of the relaxation rate at $t\sim\tau_\xi$ characterized by a stretched-exponential decay. Since τ_ξ diverges at T_g, only a power-law decay is found below T_g. As shown below, these are the fundamental characteristics of the zero-field equilibrium relaxation in real spin glasses. At temperatures below T_g, CM find the power-law behavior given by Eq. (10), but derive the following expression for the relaxation at long times above T_g:

$$M(t)\propto\exp(-(t/\tau_\xi)^{1-n}), \quad n=z\nu/(\beta\delta+z\nu) \ . \quad (11)$$

In Figs. 1(a) and 1(b) we have visualized the general appearence of the relaxation rate as given by Eqs. (10) and (11), respectively. The values of the critical exponents are $z\nu=8.2$, $\beta=0.38$, $\beta\delta=3.8$, and $\tau_0=2\times10^{-13}$ s as obtained from static[10] and dynamic[11] scaling on the amorphous metallic spin glass $(Fe_{0.15}Ni_{0.85})_{75}P_{16}B_6Al_3$. At temperatures above T_g the two curves refer to $T/T_g=1.05$ and 1.01. In Fig. 1(a) no extra parameter has been introduced to change the magnitude or shape of the relaxation rate curves. The magnitude of the curves in Fig. 1(b) is arbitrarily scaled. Figure 2 shows the relaxation rate (S) of the amorphous metallic spin glass $(Fe_{0.15}Ni_{0.85})_{75}P_{16}B_6Al_3$ at three temperatures around T_g, as obtained from ZFC and ac susceptibility measurements.[12] The data points in the observation time range from 3×10^{-6} to 3×10^{-1} s are found from ac susceptibility measurements, and the quantity $-(2/\pi)\chi''(\omega)$ is plotted. From experiments on real spin glasses it has been shown[8] that this quantity equals $-\partial\chi'(\omega)/\partial\ln\omega$. This relation applies to systems where there exists a wide distribution of relaxation times, which consequently include fractal-cluster spin glasses.[13] The solid curves in the time range from 3 to 10^4

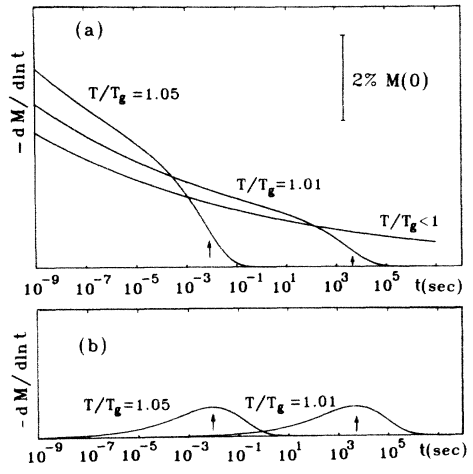

FIG. 1. (a) Relaxation rate obtained from Eq. (10) for $T=1.05T_g$, $T=1.01T_g$, and $T<T_g$. Parameters are $\beta=0.38$, $\beta\delta=3.8$, $z\nu=8.2$, and $\tau_0=2\times10^{-13}$ s. The arrows indicate the location of τ_ξ as claculated from $\tau_\xi=\tau_0(T/T_g-1)^{-z\nu}$. 2% of $M(0)$ is indicated. (b) Corresponding relaxation rate curves above T_g obtained from Eq. (11). The magnitude of the curve is arbitrarily scaled.

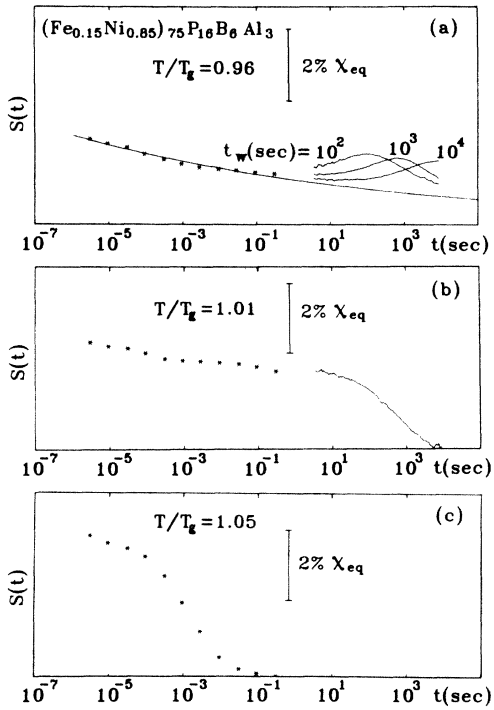

FIG. 2. Relaxation rate (S) curves for $(Fe_{0.15}Ni_{0.85})_{75}P_{16}B_6Al_3$ obtained from ac susceptibility [$S=-(2/\pi)\chi''(\omega)=-\partial\chi'(\omega)/\partial\ln\omega$, $3\times10^{-6}<1/\omega<3\times10^{-1}$ s] and zero-field-cooled magnetization [$S=(1/H)\partial M/\partial\ln t$, $3<t<10^4$ s] measurements. 2% of the equilibrium susceptibility is indicated. $T_g=22.6$ K. (a) $T=0.96T_g$. The various zero-field-cooled curves refer to different wait times ($t_w=10^2$, 10^3, and 10^4 s) before the field ($H=0.1$ G) is applied. The solid line represents the best fit ($t^{-0.05}$) to the ac susceptibility data. (b) $T=1.01T_g$. (c) $T=1.05T_g$.

s are obtained from measurements of the relaxation rate $(1/H)\partial M/\partial \ln t$ in ZFC experiments. At temperatures below T_g the aging process markedly influences the experimental ZFC curves. The various ZFC curves at $T=0.96T_g$ [Fig. 2(a)] refer to different wait times ($t_w=10^2$, 10^3, and 10^4 s) before applying the external field ($H=0.1$ G). The characteristic feature of these curves is a maximum of the relaxation rate at an observation time equal to the wait time. From the evolution of the relaxation rate curves with wait time it is not hard to imagine that they, at equilibrium (i.e., $t_w=\infty$), level down on the $t^{-0.05}$ curve [solid line in Fig. 2(a)], which represents the best fit to the ac susceptibility data. At $T=1.01T_g$ it is possible to wait out the effect of aging, and the measured ZFC curve in Fig. 2(b) reflects the equilibrium relaxation. As T increases [Fig. 2(c)] the edge of the relaxation rate curve drastically moves towards shorter times.

A direct comparison between the calculated curves of Fig. 1 and the experimental curves of Fig. 2 clearly demonstrates that a pure stretched exponential form does not signify the relaxation behavior and emphasizes the importance of a wide time scale perspective. Equation (11) ony applies in the asymptotic limit of long times (i.e., $t \gg \tau_\xi$) and represents only the minute fraction of the total relaxation left well beyond τ_ξ. The very final approach towards equilibrium is by no means unimportant, but facing the facts that the equilibrium approach is exceptionally sensitive to the magnitude of the external field and sample constitution, it is exceedingly difficult to experimentally determine the relevant functional form for the asymptotic behavior in the limit of zero field. Instead, including the general results from neutron scattering,[14] Eq. (10) gives a good qualitative description of both the magnitude[7,8,12,15] and functional form of the total relaxation, including the profound nature of the equilibrium approach. Apparently strongly influenced by the frequent reports of pure stretched exponential relaxation in spin glasses, both above and below T_g, and less confident with the predictions of the fractal-cluster model, CM virtually squeeze out the stretched exponential term from the model. The proposal[16] that the time decay of the TRM can be described by a pure stretched exponential below T_g is due to a fundamental misinterpretation of the influence of the aging process on the experimental curves. A stretched exponential only appears to characterize the *dynamics of the aging process.* This has been amply demonstrated by Nordblad, Svedlindh, Lundgren, and Sandlund[2] and Alba, Ocio, and Hammann.[3] At equilibrium and in the limit of zero field there are no experimental indications whatsoever of a stretched-exponential relaxation below T_g. Close to T_g, measurements of TRM generally refer to the time decay of the saturated remanent magnetization,[17,18] which by nature is different from the zero-field equilibrium relaxation.[4]

In the regime of linear response ac susceptibility, ZFC, and TRM experiments mirror the time variation of the order parameter $q(t)$. From computer simulation on a short-range 3D-Ising spin glass by Ogielski,[19] it was found that the time variation of the q parameter at thermal equilibrium follows the empirical law

$$q(t)=At^{-a}\exp(-(t/t_p)^b) \ . \tag{12}$$

This relation has a great similarity to Eq. (10), but in order to account for the fundamental experimental observation on real spin glasses of a monotonic decrease[8,12,15] of the relaxation rate with time, there are constraints on the ratio between the exponents b and a. It is readily shown that a monotonic decrease requires $b/a<4$. This inherent ambiguity of the form of the relaxation function does not exist in Eq. (10). Figure 3 shows the ratio b/a between the exponents of Eq. (12) using data from Ogielski.[19] As is seen from the figure, the ratio between the exponents becomes larger than 4 at some temperature above T_g, and a simple extrapolation gives $b/a=5.5$ close to T_g. In Fig. 4 the relaxation rate $\partial q/\partial \ln t$ obtained from Eq. (12) is plotted with $a=0.06$ (from the simulations), $b/a=5.5$, and assuming $t_p=10^3$ s for comparisons with the results of Figs. 1 and 2. Figure 4 shows a pronounced maximum of the relaxation rate (at $t\sim t_p$), a behavior which is not found in experiments on real spin glasses. It should be noticed that a maximum in the relaxation rate was barely seen by Ogielski, and the parameter fittings were made from data at shorter times. On the other hand, the data from the computer simulations can also be fitted to the functional form of Eq. (10). Oddly enough, in spite of a

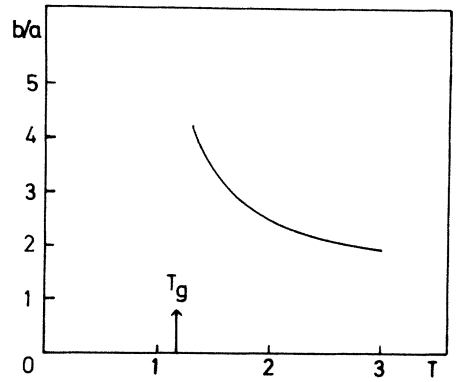

FIG. 3. Ratio b/a of the exponents in Eq. (12). Data from computer simulations by Ogielski (Ref. 19).

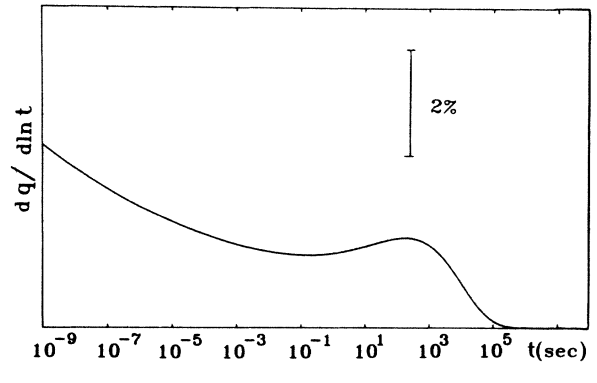

FIG. 4. Relaxation rate $\partial q/\partial \ln t$ obtained from Eq. (12) with $a=0.06$, $b=0.33$, and $t_p=10^3$ s. The curve has been normalized to $q(t)=1$ at $t=10^{-13}$ s. A relative change of 2% is indicated.

short maximal time in the computer simulations by Ogielski, it appears that these simulations give the best summarizing description of the equilibrium relaxation of spin glasses. This is primarily due to a good definition of the experimental conditions as well as the possibility of covering an unusaully wide time scale with only one experimental probe. Simulations of the q parameter at nonequilibrium are called for in order to simulate the aging phenomenon, which is the main cause for the current confusion as to the behavior of the spin-glass relaxation.

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