Temperature dependence of aging in spin glasses

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Experimental data for $Cu_{0.9}Mn_{0.1}$ are presented for the change with temperature of: the dependence of the initial magnetization on waiting time before applying a magnetic field to a zero-field-cooled sample, the maximum value of $S=\partial M/\partial \ln t$, and the time at which the maximum occurs. The results as well as published data [Granberg *et al.*, J. Magn. Magn. Mater. **92**, 228 (1990)] on the dependence of S on $\ln t$ and waiting time are found to be in good agreement with a domain model.[S0163-1829(99)06837-X]

INTRODUCTION

In this paper we wish to present experimental data on the temperature dependence of aging phenomena in spin glasses, and examine the agreement between a domain model for spin glasses and experiment.

An important difference between glasses and crystalline solids lies in their relaxation: structural glasses below the glass transition temperature exhibit structural relaxation phenomena on a wide range of time scales that slow dramatically as the temperature is lowered. Spin glasses are similar, showing magnetic relaxation phenomena with a wide range of relaxation times that slow down rapidly as the temperature decreases below the spin-glass transition, T_g.

A characteristic feature of the relaxation of spin glasses is the aging phenomenon: the magnetization of a spin glass that has been cooled in zero field below T_g, depends on the time elapsed t_w before application of the magnetic field. The larger t_w, the lower the magnetization. These effects have been studied extensively (Ref. 1, and references therein), but only at temperatures close to T_g. Although it may be anticipated that aging phenomena in spin glasses are strongly temperature dependent because relaxation effects can be expected to disappear at low temperatures where the relaxation times become very long, and at high temperatures where these times become very short, this aspect of their behavior has not been studied in any detail heretofore, to our knowledge.

Although spin glasses have been the subject of considerable attention, both experimental and theoretical, only recently has any quantitative comparison between theory and experiment been attempted.[2–4] The time dependence of the magnetization M is relatively featureless and is incapable of providing a useful testing ground for different theories. It is faintly sigmoidal, however, and the derivative $S=\partial M/\partial \ln t$ displays a maximum when plotted against $\ln t$. Thus the dependence of S on $\ln t$ and t_w, the temperature dependence of the ac susceptibility, and the strong temperature dependence of aging phenomena, that will be reported in this paper, provide experimental features which can test theoretical models.

The principal objective of this paper is to attempt to provide an adequate model for the data (in the sense that calculated curves agree with experimental points). Four sets of data will be considered: the dependence of S on t_w and $\ln t$ published by Granberg *et al.*[1] (this data has already been analyzed by Joh *et al.*[3]), and the temperature dependence of three other aging parameters reported in this paper: the difference between the initial moment for two values of t_w, the maximum value of S, and the time at which the maximum in S occurs.

At present, two seemingly quite different explanations for aging are advanced: the Parisi[5] solution of the Sherrington-Kirkpatrick Hamiltonian[6] has inspired a model in which metastable states are hierarchically organized in phase space. Aging is the result of the system overcoming barriers and populating additional states, and the $\ln(t)$ dependence results from a wide spectrum of energy barriers. The other considers aging to result from the growth of droplets[7] or domains of correlated spins.[1,8] A number of different theoretical approaches[1,7,9,10] yield the result that the domain size R is proportional to $\ln(t)$ in disordered systems (this contrasts with the well-known $t^{1/2}$ behavior for nondisordered materials).

It is sometimes claimed that domains cannot be present in spin glasses because $\langle \mathrm{Si}\rangle = 0$. $\langle \mathrm{Si}\rangle$, however, is only 0 in the limit as the number of spins approaches infinity, and fluctuations ensure that local departures from the average must occur. Therefore small volumes will have a net ferromagnetic moment. When a sample is cooled from its paramagnetic state into the spin glass phase it will contain a number of small regions, or domains, for which $\langle \mathrm{Si}\rangle \neq 0$. The system can decrease its energy by decreasing the total boundary area between different domains by domain growth, and this results in aging phenomena.[4] For this reason, to quote Bouchaud and Dean[11] a spin glass can be considered a ''disguised ferromagnet.''

It is clear that it is the net magnetic moment that is ferromagnetic in a spin glass. Competing interactions can result in a significant number of spins in a domain having orientations different from that of the domain itself. In the Ising model the ferromagnetic moment is a result of the majority of spins being oriented parallel to each other, while the rest are antiparallel. Since the average fraction of antiparallel spins is independent of domain size, the average ferromagnetic moment is proportional to the total number of spins. In this connection it is worth remembering that whereas the interaction between the Mn at high concentrations is antiferromagnetic, in the low-concentration spin-glass phases, the interaction is ferromagnetic.[12]

The states of the Ising model are twofold degenerate in the absence of an external field. The presence of a magnetic

field removes this degeneracy, and the problem to be addressed here is how a domain changes its state in response to the magnetic field. The hierarchical model concentrates on the transitions of individual spins without any reference to possible movements of domain walls. On the other hand, the spins on the domain walls are the most easily reversed in the system, so that the domain walls must move in the presence of a magnetic field. It also appears that the higher barriers to spin reversal in the hierarchical model require an ergodic contribution. If this is the case it is impossible to ignore cooperative effects.

The positions of the boundary of a domain that has been growing for a time t are continually fluctuating on a broad range of time scales between some microscopic relaxation time τ_0 and t, resulting in a highly irregular structure: if the scale of the domain is R, the scale of the fluctuations in boundary position are on the order of R. This makes a description in terms of domains somewhat imprecise because the highly irregular nature of the boundaries will lead to considerable interpenetration of the network of domains. Nevertheless, it is possible to define a domain size R in the sense that the number of spins in a domain will be R^D, where D is the fractal dimension. In three dimensions $D \sim 2.5$.[13]

A major objective of this paper is to account quantitatively for both the experimental data in the literature and the new results on the temperature dependence presented here. For the reasons outlined above, a domain model will be used to analyze the experimental results. The domain model has already been shown to account for the temperature and frequency dependence of the ac susceptibility.[2] It will be found that a domain model is also successful in reproducing the data on aging, with one exception: the change in the peak position of $S = \partial M / \partial \ln t$ with temperature. The failure appears to be related to the replacement of the spectrum of energy barriers to spin reversal by a single average barrier.

The remainder of this paper will be organized as follows: first the experimental techniques and the results will be presented, then the theory used to analyze that data, and finally, a discussion of the implications of these results.

EXPERIMENTAL DETAILS AND RESULTS

A polycrystalline sample of $Cu_{0.9}Mn_{0.1}$ was prepared by quenching from the melt (in order to minimize segregation of the manganese). The moment as a function of time was measured with a superconducting quantum interference device (SQUID) magnetometer in a field provided by a superconducting magnet. Temperature control was better than 0.01 K. T_g depends on the waiting time, and for a waiting time of 300 s was about 42 K.

In order to measure the effect of waiting time on the initial moment the sample was cooled in zero field from a temperature of 100 K to a temperature below T_g. After reaching thermal equilibrium a field of 5 Oe was switched on after waiting for 30 s, and the evolution of the moment with time was measured, the experiment was then repeated except that the sample was held in zero field for 300 s before the field was applied. The difference in the magnetization after the field had been applied for 30 s for the two waiting times is shown in Fig. 1.

The detailed dependence of $S = \partial M / \partial \ln t$ on $\ln t$ and waiting time has been exhaustively reported in the literature, and will not be reported here. Instead we will focus on the change in the magnitude of the maximum value of S, S_{max}, and the change in its position with temperature.

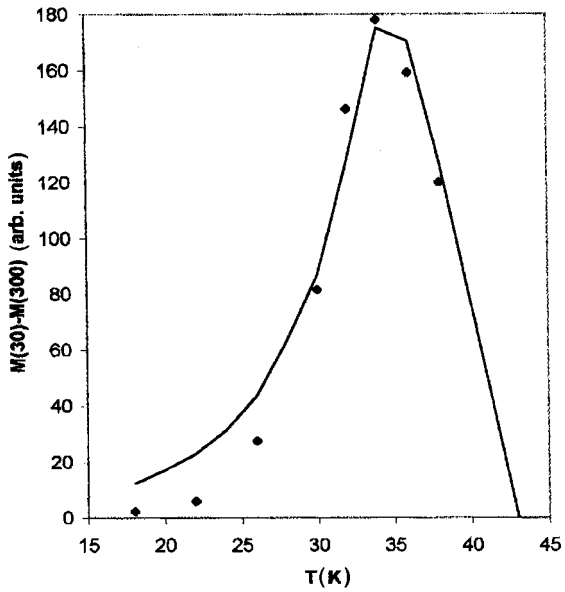

FIG. 1. The change in ΔM with temperature. ΔM is the difference between the initial values of the magnetization of a zero-field-cooled sample upon waiting for 30 and 300 s before turning on a 5-Oe field. The points are the experimental data, the solid line was calculated using a domain model.

The change in the magnitude of S_{max} with temperature is shown in Fig. 2. The behavior is similar to that of another spin glass that has already been reported.[14] It is interesting that virtually all the data in the literature are obtained at a temperature $\sim 0.9 T_g$ but the effect is strongest at a much lower temperature.

The change in position of the peak is shown in Figs. 3 and 4. While it is claimed in the literature that this peak occurs at a time equal to the waiting time, it is clear from Fig. 4 that this is only the case at one temperature, about $0.9 T_g$. There-

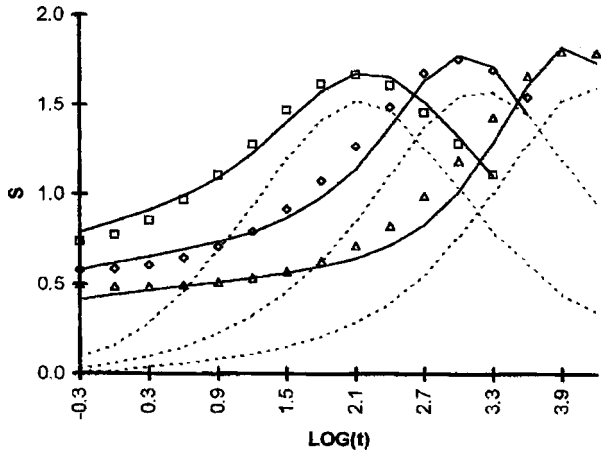

FIG. 2. The change in $S = \partial M / \partial \ln t$ with $\ln t$ for three waiting times. The points are data from Ref. 4 for waiting times of 100 s, squares; 1000 s, diamonds, and 10 000 s, triangles. The solid lines are a fit using the domain model, and the dashed lines are one using the hierarchical model from Joh *et al.* (Ref. 3). Note the lack of agreement of the hierarchical model for small t.

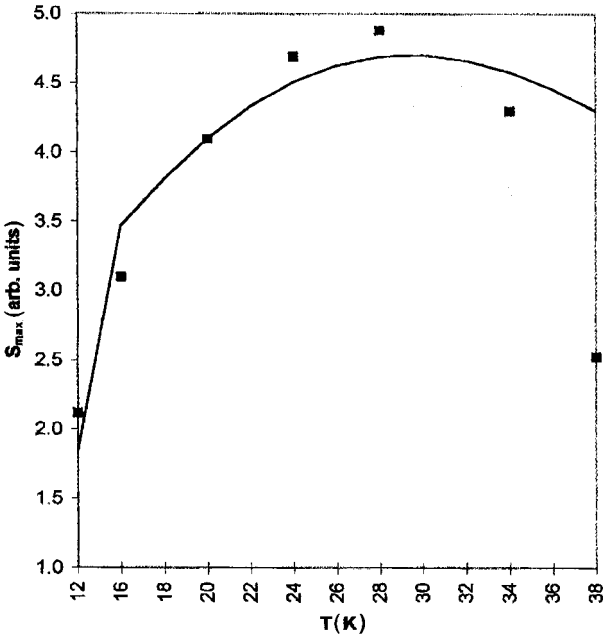

FIG. 3. The temperature dependence of the maximum value of S for a waiting time of 300 s. The points are the data, the line was calculated using a domain model.

fore the identification of the peak position with waiting time is clearly wrong. This simple idea may possibly be rescued if the effect of a finite cooling time is included but this cannot be simply a question of measuring the waiting time from the time when the sample began to cool.

The lines in the figure were calculated, and the theory for those calculations will now be outlined.

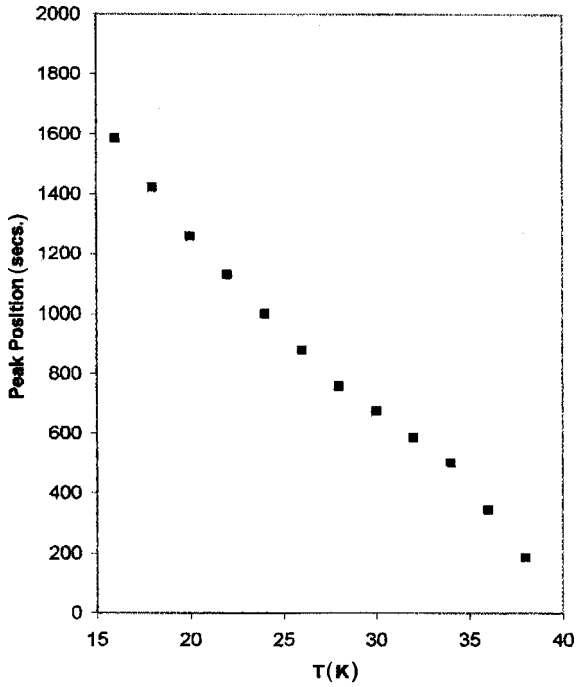

FIG. 4. The temperature dependence of the time at which the maximum value of S, for a waiting time of 300 s, occurs.

THEORETICAL BACKGROUND

A domain model has been shown[4] to yield quantitative agreement with experimental results published by Svedlidh *et al.*[15] for the change in T_g with frequency, and S with $\ln t$ for $Fe_{0.5}Mn_{0.5}TiO_2$. The frequency dependence of S for $Cu_{0.9}Mn_{0.1}$ published by Granberg *et al.*,[1] and the temperature dependence of the effect of waiting time on aging in this material obtained as outlined above will be addressed here.

There are three contributions to the moment produced by the applied field: the first is due to domain boundary motion, the second arises from the nucleation of new domains, and the third from the percolation structure.[16] Nucleation and growth of new domains limits the effects of aging: eventually waiting before applying a magnetic field will cease to have any effect because the system will simply reach equilibrium. It is evident from the experimental data that this is only important at temperatures close to T_g.

An Ising spin glass with only nearest-neighbor interactions will be considered. The Hamiltonian is

$$\mathcal{H}=-J\sum_{\langle i,j\rangle} S_iS_j-H\sum_i S_i,$$

where the spins, $S_i=\pm 1$, J is the nearest-neighbor exchange, and $\mathcal{H}$ is a uniform magnetic field.

A number of different theoretical approaches[2,7,9,10] yield the result that the domain size R is a function of $\ln t$ in disordered systems. Since it has also been well-established experimentally that domains in disordered systems scale with $\ln t$,[1] the following expression should be able to account for the experimental data:

$$R\sim\left(A\ln\frac{t}{\tau}\right)^a, \tag{1}$$

where R is the domain size, $\tau=\omega_0^{-1}e^{-E/T}$ is a microscopic relaxation time with ω_0 a fundamental attempt frequency, and E a barrier height to spin reversal. A range of barrier heights are present; so E should be interpreted as the result of an effective medium approximation. E will be treated as an adjustable parameter. It is not always satisfactory to replace a spectrum of relaxation barriers by an average, and this approximation may be the least satisfactory aspect of the whole model.

A is a temperature dependent constant. Fisher and Huse[7] have $A=T/\Delta(T)$, where Δ is an energy on the order of J. The calculation in the appendix yields a similar result: $A=T/(T+Q)$.

t/τ can be identified as the number of spin reversals in a time t along a line of spins. In that case a should relate to the ''tortuosity'' of the lattice, and should be the ''spreading'' or ''chemical'' dimension that computer simulations find to be 0.88 in three dimensions.[17]

It is clear that if t_w is measured from the end of a waiting time t_w,

$$R\propto\left[A\ln\left(\frac{t_w+t}{\tau}\right)\right]^a.$$

The waiting time plays a crucial role because while waiting in zero field domains grow. However domain growth

also takes place while the sample is cooling to the measurement temperature; so an effective cooling time t_c should be added to t_w:

$$R \propto A^a \ln^a\left(\frac{t_c+t_w+t}{\tau}\right) \equiv A^a \ln^a(g_c+g_w+k). \tag{2}$$

g_c, g_w, and k are the number of spin reversals after cooling, waiting, and a time t, respectively. As the sample is cooled, eventually a temperature will be reached where domain growth ceases and g_c becomes constant. The time at which the peak in S will occur is $t_p=t_w+g_c\omega_0^{-1}e^{E/T_f}$, and, if the effective medium approach is valid, should increase exponentially as the temperature decreases after g_c becomes constant. The results shown in Fig. 4, however, show no evidence of such an exponential increase at low temperatures.

AGING

Contribution from domain-wall motion

Equation (2) implies that domains much larger than R are improbable. Due to the fluctuations in boundary position domains disappear, thus domains of scale much smaller than $\sim R$ are also improbable. This suggests that the size distribution is relatively narrow. The domain size distribution can be obtained by solving the stochastic equation involved, however that is beyond this work, and the distribution of domain sizes will simply be replaced by the average size R.

Fluctuations in the position of the boundary of a domain that has been growing for a time t are occurring on all time scales less than t. Therefore the domains will be irregular on all scales $r<R$: a domain that has grown for a time t_w+t, and for which $R=\{A\ln[(t_w+t)/\tau]\}^a$ will have structure on all scales $R'=[A\ln(t'/\tau)]^a<R$, which has developed in times $t'<t_w+t$.

In a magnetic field H the relaxation time $\tau=\omega_0^{-1}e^{E/T}$ becomes

$$\tau^{\pm}=\omega_0^{-1}e^{(E\pm\mu H)/T}\equiv \tau e^{\pm h}. \tag{3}$$

When a magnetic field is applied the fluctuations in boundary position will no longer be equal. If the field has been applied for a time t, the boundaries will have moved a distance $\sim[A\ln(t/\tau^{\pm})]^a$, involving $[A\ln(t/\tau^{\pm})]^{aD}$ spins (a possible effect of the field on A has been neglected). Thus the number of favorably oriented spins will exceed the others, and a net moment will result: $m\sim\{[A\ln(t/\tau^{+})]^{aD}-[A\ln(t/\tau^{-})]^{aD}\}$.

Thus, at the end of t_w, this model envisions domains of average scale $R\sim[A\ln(t_c+t_w)/\tau]^a$, with very irregular boundaries whose moments average to zero. After the field has been applied for a time t the average domain size becomes $\sim[A\ln(t_c+t_w+t)/\tau]^a$, but the fluctuations in boundary position occurring during t no longer average to zero

The net moment will be proportional to the total boundary area. The boundary is fractal, and the area of a domain of size R will be $\sim R^{d_s}$ where $d_s>d-1$, the total number of domains is N/R^D, where N is the total number of spins; so the total boundary area is $\sim NR^{d_s-D}=N/[A\ln(g+k)]^{a(D-d_s)}$. The moment is $\sim mN/R^{D-d_s}$,

$$M_h \propto \frac{A[\ln^b(t/\tau^+)-\ln^b(t/\tau^-)]}{[A\ln(g+k)]^{b-c}} \simeq \frac{2b\mu H}{T}\left[\frac{A^c\ln^{b-1}k}{\ln^{(b-c)}(g+k)}\right], \tag{4}$$

where $b=aD, c=ad_s$, and it has been assumed that H is small.

The spins can be expected to lie on a percolation structure that can be viewed as a collection of clusters.[18] Below the percolation limit the clusters are isolated from each other. Above it an infinite cluster appears. The infinite cluster can be viewed as an assembly of clusters, some of which are connected, this is the ''blobs'' and ''links'' model.[19]

Below the percolation threshold the cluster size distribution can be approximated by $w_s=s^{-\tau+1}e^{-Cs}$[18] where τ (not to be confused with the relaxation time) is ~ 2.18,[18] s is the number of spins in the cluster , and $C\sim|p-p_c|^{1/\sigma}$.[18] In three dimensions $\sigma=0.45$,[18] and since the threshold for site percolation on a fcc lattice is $p_c=0.198$, for $p=0.1, C\sim 0.0057$.

If nucleation of new domains is neglected, only those clusters large enough to contain at least one domain boundary, i.e., for which $R>A\ln^a(g+k)$, can contribute to M_h. Thus Eq. (4) must be multiplied by the fraction of the total number of spins in multidomain clusters, which is

$$\frac{\int_{R^D}^{\infty} sw_s}{\int_1^{\infty} sw_s} \approx e^{-CR^D}.$$

And the moment produced by domain-wall motion becomes

$$M_h \propto \frac{2b\mu H}{T}\left[\frac{A^c\ln^{b-1}k}{\ln^{(b-c)}(g+k)}\right]e^{-C[A\ln(g+k)]^b}. \tag{5}$$

Contribution from percolation clusters

Clusters smaller than $(A\ln g)^a$ are smaller than the domain size, and will be single domain. If the nucleation of new domains is neglected they cannot contribute to the moment. After the field is applied, domain growth will take place in the larger, multidomain, clusters at different rates for the two spin orientations. Some clusters, those for which $A\ln^a g<R^{\pm}\leqslant[A\ln(g+ke^{\pm h})]^a$, will become single domain, and, because their average volumes are different, will contribute to the moment:

$$M_{cl} \propto \int_{A^b\ln^b(g+ke^{-h})}^{A^b\ln^b(g+ke^{h})} e^{-Cs},$$

which, for small H becomes

$$\simeq 2aD\frac{\mu H}{T}\left(\frac{k}{g+k}\right)[A\ln(g+k)]^{b-1}e^{-CR^D}. \tag{6}$$

The total is $M=M_h+M_{cluster}$, and

$$M \propto \frac{1}{T}\left[\left(\frac{A^D\ln^{aD-1}k}{R^{D-d_s}}\right)+\left(\frac{k}{g+k}\right)\frac{R^D}{\ln(g+k)}\right]e^{-CR^D}, \tag{7}$$

$S \propto dM/d\ln t$, and for $CR^D < 1$ and after some algebra

$$S \propto \frac{1}{T}\left\{\left(\frac{A^D \ln^{aD-1} k}{R^{D-d_s}}\right)\left[\frac{aD-1}{\ln k} - \frac{ak}{(g+k)\ln(g+k)}\right] \times (D - d_s + CDR^D)\right\} + \frac{1}{T}\left[\frac{gk}{(g+k)^2}\right]\left[\frac{R^D}{\ln(g+k)}\right] \times \left\{1 + \frac{(b-1)k}{g\ln(g+k)} - \frac{k}{g\ln(g+k)} CR^D\right\} e^{-CR^D}. \quad (8)$$

The second term in Eq. (8) is multiplied by $gk/(k+g)^2$ which has a maximum when $k=g$. It is responsible for the peak in S, and it is easy to see why the maximum occurs when $k \sim g$. It should be noted that this contribution depends on the existence of percolation clusters. The existence of percolation clusters in turn depends on site disorder. Therefore a material in which this disorder is absent, and which, consequently, does not display percolation clusters, should not display the peak in S at $k=g$. This conclusion is supported by results for the nondisordered pyrochlore $Y_2Mo_2O_7$.[16]

DATA ANALYSIS AND DISCUSSION

The experimental data used provided a stringent test of the model: the fits were very sensitive to the values of the parameters employed (with the exception of d_s). While fitting any one set of data was relatively easy, fitting all three was not.

In fitting the data, $A = T/(Q+T)$ was used, and Q was calculated using $J = 50$ K, the value obtained by Morgownyk and Mydosh,[20] which led to $Q = 4504$ K. The remaining parameters are the values of E and ω_0, and d_s, since no data is available that would yield values for them these were adjusted to yield the best fit, which resulted in $E = 220$ K, $\omega_0 = 10^{13}$. The fits were quite insensitive to the value of d_s, which must be greater than $d-1=2$, and obviously less than $D = 2.5$. A value of 2.2 was used for d_s.

Thus, with two adjustable parameters, the domain model provides good agreement with all the experimental data with one exception, the change in t_p with temperature. As discussed previously, this probably illustrates the inadequacy of replacing a spectrum of relaxation barriers with a single average barrier.

Another limitation of this simple theory results from neglect of nucleation and growth of new domains. This is clearly responsible for the shift in the peak position of S to times shorter than the waiting time as T_g is approached from below. It presumably is also responsible for the decrease in the peak value of S close to T_g.

It is likely that the hierarchical model would be able to reproduce the data in Fig. 4. This model also avoids the messy problem of accounting for domain nucleation and growth. Unfortunately, quoting Joh *et al.*: ''The peak of $S(t)$ at $t=t_w$ in the hierarchical model arises from the massing of occupied states at $\Delta(t_w)$ at $t=t_w$.'' If that is the case, S should display a maximum at $t=t_w$ in $Y_2Mo_2O_7$, which is contrary to observation. Another, less serious difficulty, is the fact that the hierarchical model does not account at all well for the initial value of S that depends quite strongly on t_w.

It would appear that neither model is entirely satisfactory in its present form. The domain model can be improved by including a spectrum of relaxation times. A possible improvement to the the hierarchical model would be to include correlations between relaxing spins, which might then lead to a sensitivity to the percolation structure. Possibly the time dependence of such correlations might be viewed as domain growth by another name.

APPENDIX

At the concentrations of interest here the clusters have a highly ramified character, thus the spin correlations must propagate along one-dimensional paths. It will be the thermal fluctuations along these paths that will control the evolution of the magnetization. The spins in these sequences must have a minimum of two nearest neighbors (NN). In what follows it will be assumed that the average time for a spin with two NN to reverse is much longer than any laboratory time, and this direct relaxation will be neglected. This approximation will clearly break down close to T_g.

Consider the microscopic nature of the spin relaxation: it is clear that the relaxation of a spin reduces its constraint on its neighbors, one of which can then relax, thereby reducing the constraint on *its* neighbors, and so on, along a sequence of spins. Consider a sequence of spins, the relaxation of a spin at position n in the sequence (the sequence starts at $n=0$) is much shorter if the spin at $n-1$ has relaxed. The relaxation time of the spin at n is reduced by a factor $\sim e^{-2J/T}$ by relaxation of the spin at $n-1$. If the spin at $n-1$ only has two neighbors, the spins at n and $n-2$, and the spin at $n-2$ is reversed, then both orientations for the spin at $n-1$ are equally probable. However, the relaxation of the spin at n is more probable if $n-1$ is in one of its orientations, thus $\tau_n \sim 2\tau_{n-1}$. If the spins in the line of n spins all have two NN, then $\tau_n \sim 2^n \tau_0$.

If the spin at $n-1$ has three neighbors, one of which has relaxed, the probability that the spin at $n-1$ has reversed is $e^{-J/T}/(e^{J/T}+e^{-J/T})$, and $\tau_n \sim (1+e^{2J/T})\tau_{n-1}$. With four NN, $\tau_n \sim (1+e^{4J/T})\tau_{n-1}$, and so on.

If p_q is the probability that a spin has q NN, and z is the maximum number of NN,

$$\tau_n \sim \tau_0 2^{np_2}(1+e^{2J/T})^{np_3}(1+e^{4J/T})^{np_4}\cdots(1+e^{(2z-4)J/T})^{np_z} \approx \tau_0 2^{np_2}(e^{2J/T})^{np_3}(e^{4J/T})^{np_4}\cdots(e^{(2z-4)J/T})^{np_z},$$

which may be written

$$\tau_n \sim \exp[p_2 \ln 2(1+Q/T)n]\tau_0,$$

where, $Q \approx 2J[p_3 + 2p_4 + \cdots + (z-2)p_z]/p_2 \ln 2$.

Finally, identifying t with τ_n,

$$n \sim \frac{1}{p_2 \ln 2}\left(\frac{T}{T+Q}\right)\ln\frac{t}{\tau_0}.$$

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