Relaxation and aging in Ising systems

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A sequential relaxation model for the susceptibility of site diluted Ising systems is introduced. Most of the necessary parameters are known for the two-dimensional Ising spin glass $Fe_{0.4}\ Mg_{0.6}\ Cl_2$ and a good fit to experimental results for the ac susceptibility is obtained. Predictions of the model yield good agreement with aging experiments on $Rb_2Cu_{0.89}Co_{0.11}F_4$ and $Fe_{0.5}Mn_{0.5}\ TiO_3$. [S0163-1829(96)06321-7]

I. INTRODUCTION

It has been known for some time that at low temperatures the spectrum of relaxation times in disordered Ising magnets becomes very broad. More recently,[1–3] interesting aging effects, in which the susceptibility is time dependent, have been reported. A very elementary microscopic model will be introduced below that can, in large measure, account for these experimental results.

In an important series of papers, Fisher and Huse[4] have developed a droplet model of relaxation in disordered Ising systems in which the coherent relaxation of droplets of oppositely ordered spins controls the dynamics. A droplet is defined as a coherent group of reversed spins embedded in a matrix of oppositely oriented spins. The dynamics of the system is determined by coherent reversals of the droplet spins. As the temperature is lowered the important droplet relaxations, i.e., those that can take place in an experimental time, are confined to smaller and smaller droplets. The droplet relaxations occur in a system of domains, and movement of the domain boundaries and the consequent decrease in domain boundary area is responsible for aging. The only difference between droplets and domains is one of size, and hence the time scale of fluctuations. The domains are taken to be stable for a time on the order of the experimental time, whereas the droplets are not. The distinction is, of course, inexact because there must be a continuous distribution of sizes, and a complete theory would avoid the distinction. In fact the process by which a cluster of reversed spins (a droplet) becomes a domain is important for the effect of cooling rate on the thermal remanent moment (TRM), and aging.

The model to be presented here is similar in that the system is supposed to consist of domains of oppositely oriented spins. In the low temperature limit, when all relaxation times are much longer than the experimental time, all domains are stable. As the temperature increases fluctuations become important, leading to changes in the domain structure responsible for aging effects. However, coherent fluctuations, and hence droplets, are specifically discarded in favor of a sequential model to be described below in which spins are assumed to relax sequentially with the relaxation of one reducing the constraint on a neighbor, thereby allowing it to relax, which in turn reduces the constraint on another spin in the sequence and so on. Thus the focus is on the individual relaxation of the spins in the domains, rather than their collective relaxation. Nevertheless, clusters of flipped spins are the result, common to both theories.

Both models predict that a two-dimensional (2D) random ferromagnet ages as $(A+B\ \ln t)^{\alpha}$. The droplet model predicts that $\alpha \approx 4$, whereas the sequential requires α to be 1/2. The droplet model has been applied to the aging of the ac susceptibility by Schins *et al.*,[3] however, it will be shown below that the sequential approach yields better agreement with the data.

The sequential is a constrained dynamical model for relaxation. Frederickson and Andersen[5,6] have described a constrained dynamical model for relaxation in Ising systems which yields the susceptibility, and similar models have been applied to other relaxation phenomena in disordered systems.[7–10] In a similar vein, a hierarchical model was quite successful in accounting for relaxation in orientational glasses[11–13] accounting for the frequency and temperature dependence of not only the structural but also the dielectric relaxation. The model was based on the hierarchical relaxation model of Palmer *et al.*,[14] in which Ising spins are assumed to occupy a hierarchy of levels such that the spin in one level, say n, cannot relax unless spins in the adjacent lower level, $n-1$, has relaxed. No microscopic model for the levels was provided by the authors; so, in order to apply it and compare it with experiment the levels were identified with the number of nearest neighbors, so that relaxation of a spin with a given number of neighbors required the relaxation of spins with fewer neighbors. In other words the fluctuations in concentration were modeled to consist of clusters of spins with the highest concentration at the center of the cluster.

A hierarchical model for the organization of the energy barriers in the rough free-energy landscape of spin glasses has been proposed by Lederman *et al.*[15] which has been applied to the decay of a TRM.

Sibani,[25] using a model based on motion involving thermally activated hopping in an utrametric space has obtained results which qualitatively reproduce experimental susceptibilities. No comparison with experiment is attempted, however, possibly because the change in the temperature of the susceptibility peak with frequency is much larger than is observed in most experiments.

While both the hierarchical and the droplet model can account for aging effects, neither does so for the temperature and frequency dependence of the ac susceptibility. In fact, despite more than two decades of intense theoretical and experimental interest, quantitative agreement with the ac sus-

ceptibility does not exist for any theory at present. This will be provided by the sequential model which will also be shown to account for aging phenomena.

The sequential model generates a distribution of relaxation times which diverge exponentially with the number of spins in the sequence. There are other constrained dynamical models which could, in principle, be expected to achieve the same result, and, as outlined above some display attractive features. The detailed comparison with experimental susceptibilities is lacking, however.

This paper is organized as follows: in Sec. II the model is described and compared with experimental data for the ac susceptibility of the two-dimensional Ising spin glass, $Fe_{0.4}Mg_{0.6}Cl_2$ obtained by Wong *et al.*[16] Next the change in ac susceptibility with time after a quench is treated in Sec. III. The time dependent acquisition of a moment in a magnetic field is considered in Sec. III, and the results are discussed in Sec. V.

The model is a nearest neighbor model so only results for insulating Ising systems with short range interactions are considered. Unfortunately aging data are not available at present for $Fe_{0.4}Mg_{0.6}Cl_2$ and the analysis of the aging data requires parameters which have not been measured in the systems for which aging data is available. Decay of a TRM is not considered, because cooling in a field results in a complex structure for which there is no obvious approximation: since the TRM decay is sensitive to the waiting time before the field is turned off,[15] the starting distribution of clusters must depend on cooling rate. Calculation of the cooling rate dependence of a TRM is not difficult but is beyond the purpose of this communication.

II. THE SPECTRUM OF RELAXATION TIMES

The model is the following: a site-diluted 2D Ising random magnet with clusters of spins which are unfrustrated is considered. The major interaction is with the nearest neighbors (NN's), and is calculated explicitly. A single ion anisotropy energy Q must be overcome for a spin to flip (of course, a substantial anisotropy must exist for the Ising model to provide a realistic approximation).

The following is the basic assumption of the model: at low temperatures the field due to its nearest neighbors will prevent a spin in a cluster from relaxing. But, if at least half its neighbors have flipped, the field of the rest will be canceled, and the spin will no longer be constrained. The only remaining barrier to its relaxation will be that due to anisotropy. The simplest such situation is one where the spin has only two NN's. In that case the relaxation time of a spin n sites from where the process was initiated is determined by the probability that, since these are Ising spins, the neighbor at $(n-1)$ will occupy one of its two possible states, i.e.,[14]

$$\tau_n' = 2\tau_{n-1}.$$

The spin at n can, of course relax thermally, and the relaxation time for this process is

$$\omega_0^{-1} e^{(Q+4J)/T},$$

where ω_0 is the attempt frequency, J is the exchange interaction with NN, and Q is the anisotropy energy. The total relaxation rate will be

$$\frac{1}{\tau_n} = \frac{1}{2\tau_{n-1}} + \omega_0 e^{-(Q+4J)/T}. \tag{1}$$

If the spin at n has three NN's relaxation of one of them will still leave it constrained by the other. The other NN forms part of another sequence and when both have relaxed the sequence can continue. It is unlikely that both spins have the same relaxation time; so, while the spin with the longer time is favorably oriented, the other, with the shorter time will flip more than once, and it is the longest relaxation time of the two that will control the process; so the faster relaxation of the second spin will be neglected.

If the spin has four NN's, relaxation of two neighbors removes the constraint imposed by the other two, and the same remarks apply, except that with three NN's only one sequence can continue, whereas with four NN's both can. If five and six NN's are possible it is necessary for three spins to relax. In order to simplify the calculation it will be assumed that the relaxation of only one NN controls the relaxation of a spin in a sequence. In effect correlations between the sequences are being neglected. This is a poor assumption if the number of NN's is 5 or 6, but at the concentrations of interest the concentration of spins with more than four NN's is very small.

With these assumptions Eq. (1) can be written as

$$\frac{1}{\tau_n} = \frac{1}{2\tau_{n-1}} + \omega_0 e^{-(Q+2z_nJ)/T}, \tag{2}$$

where z_n is the number of NN's of n. Equation (1) also holds for the nearest neighbors of n

$$\frac{1}{\tau_{n-1}} = \frac{1}{2\tau_{n-2}} + \omega_0 e^{-(Q+2z_{n-1}J)/T}, \tag{3}$$

where z_{n-1} is the number of nearest neighbors of the spin at $n-1$. z_{n-1} must be greater than 1 for a sequence to be possible. Substitution of (3) in (2) yields

$$\frac{1}{\tau_n} = \frac{1}{2}\left(\frac{1}{2\tau_{n-2}} + \omega_0 e^{-(Q+2z_{n-1}J)/T}\right) + \omega_0 e^{-(Q+2z_nJ)/T}.$$

An expression similar to (3) can be derived for τ_{n-2}, and if the sequence is retraced to the spin which originated it, τ_i, we obtain

$$\tau_{i,n}^{-1} = \frac{1}{2^n\tau_i} + \omega_0 e^{-\frac{Q}{T}} \sum_{k=0}^{n-1} 2^{-k} e^{-(2z_{n-k}J)/T}. \tag{4}$$

When the first term in (3) is larger than the second, the relaxation is a sequential process with the relaxation of each spin leading to the relaxation of a neighbor along a line of spins. Eventually the second term will dominate, the sequence stops, and the relaxation times become those for thermal relaxation of the individual spins.

The contribution of a sequence to the suceptibility is

$$\chi'_{i,n}=\frac{1}{T}\frac{P(z_i)P(z_j)P(z_k)\cdots P(z_n)}{1+(\omega\tau_{i,j,\ldots,n})^2}, \tag{5}$$

where $\tau_{i,j,k,\ldots,n}$ is the relaxation time of a sequence that begins at a spin with i NN's, and continues with spins with successively j, k, and so on to n NN's. ω is the measurement frequency, and $P(z_k)$ is the probability that a spin at k has z_k nearest neighbors.

Spins in the sequences must have at least two neighbors. The spin which starts the sequence must have at least one neighbor. If it is on a domain boundary the effective number of NN can be as small as zero because the presence of oppositely oriented spins on the other side of the boundary can cancel the effect of some of its NN's on its side.

In order to compute the contribution of the relaxing spins to the real part of the susceptibility it is necessary to sum the contributions of all the sequences:

$$\chi'_{\rm seq}=\frac{1}{T}\sum_{i=0}^{m}P(z_i)\sum_{j,k,\ldots,n=2}^{m}\frac{P(z_j)P(z_k)\cdots P(z_n)}{1+(\omega\tau_{i,j,k,\ldots,n})^2}. \tag{6}$$

In this expression i refers to the spin that starts the sequence. It can effectively have zero NN's if it is on a domain boundary; m is the maximum number of neighbors a spin can have.

The number of possible combinations contributing to Eq. (5) can be very large: If there are m possible values of z it is $m(m-2)^{n-1}$. Fortunately, at the concentrations of interest here the probabilities $P(z)$ are small, and only the contribution of the first few terms is necessary.

There are two additional contributions to the susceptibility which are significant: the first is the coherent relaxation of small independent clusters of spins, i.e., where the cluster reverses its total moment. This is

$$\chi'_{\rm cl}=\frac{1}{T}\sum_{p=2}^{p_{\max}}\frac{P(p)}{1+[\omega\tau_{\rm cl}(p)]^2}, \tag{7}$$

where the probabilities $P(p)$ have been calculated by Sykes and Glen,[17] and the relaxation time of a cluster of p spins is

$$\tau_{\rm cl}(p)=\omega_{\rm cl}^{-1}e^{pQ/T}, \tag{8}$$

where Q is just the anisotropy energy.

The other contribution is a frequency independent static susceptibility, $\chi'_{\rm st}$, from spins which cannot relax at the measurement frequency. If spin wave data are available this can be estimated using elementary spin wave theory.[18] The problem with this approach in disordered systems is first that the lack of translational invariance makes it impossible to define a consistent set of spin wave vectors. Another difficulty is that as some phase transition temperature is approached the contribution becomes smaller, and eventually disppears. That temperature is difficult to identify, and must be specified in some *ad hoc* fashion. An alternative is to use the classical theory for antiferromagnets.[19] If this is done the magnetization must be calculated. If a mean field theory is used the magnetization will be overestimated, and in any case mean field theory ignores the reaction field which is important in disordered systems.[20] If the reaction field is included it is necessary to solve the TAP (Ref. 21) equations. However, the contribution of the static susceptibility is small, and the labor involved in attempting even an approximate solution to these equations is difficult to justify. Therefore the simple expedient of adjusting the magnitude of the exchange field will be used instead:

Since this is a two level system, the magnetization in a small field h is given by[19]

$$M^{\pm}=\frac{N}{2}\tanh\left(\frac{\gamma J\pm h}{T}\right), \tag{9}$$

where γ is an adjustable parameter as discussed above, and N is the number of spins per unit volume. The susceptibility will be

$$\chi'_{\rm st}=\left[\frac{\partial(M^+-M^-)}{\partial h}\right]_{h\to 0}=\frac{N}{T}\mathrm{sech}^2\left(\frac{\gamma J}{T}\right). \tag{10}$$

The total susceptibility is $\chi'=\chi'_{\rm seq}+\chi'_{\rm cl}+\chi'_{\rm st}$.

In order to test the model a system was required with as many measured parameters as possible. The two-dimensional spin glass $Fe_{0.4}Mg_{0.6}Cl_2$ is unmatched in this respect: $FeCl_2$ is an antiferromagnet consisting of Fe^{++} spins aligned ferromagnetically along the hexagonal axis within the layer, and antiparallel in neighboring layers.[22] The spins in the layers are located on a triangular lattice. The nearest neighbor exchange interaction is ferromagnetic, and about an order of magnitude greater than the next nearest neighbor, and the antiferromagnetic interplanar interactions. When diluted with Mg, $FeCl_2$ reportedly[16,23] forms a two-dimensional spin glass at Fe concentrations below 0.5, the two-dimensional nearest neighbour percolation limit.

If a nearest neighbor model is used for $Fe_{0.4}Mg_{0.6}Cl_2$ frustration vanishes in the model, and the system becomes a two-dimensional diluted ferromagnet. For such a system no spin glass transition would be expected from the model: a Griffiths phase would be expected to exist at all temperatures below some magnetic transition temperature.[24]

The ac susceptibility of $Fe_xMg_{1-x}Cl_2$ has been measured by Wong *et al.*[16] for $x=0.4$, and Bertrand *et al.*[26] for $x=0.3$. Wong *et al.* have also made a rather complete inelastic neutron scattering study which provides most of the required parameters for a fit. These results are not available for Bertrand *et al.*'s material, so a detailed comparison will only be made with Wong *et al.*'s data.

From the neutron results the average sizes of the clusters was calculated by Wong *et al.* They point out that they are substantially smaller than the percolation correlation length for this concentration. This in turn implies that domain boundaries are present, whose average length is easily calculated from the measured domain size.

It was assumed that the sequences could start at any spin. The most likely starting points are on the boundary, where the smallest barriers to thermal activation are encountered. The fraction of spins on the boundaries was calculated from the average boundary length.

The contribution to the susceptibility of the sequences was computed using the model outlined above. The value of J used was that determined by Birgeneau *et al.*[22] The value of the anisotropy energy was obtained from the neutron data.

The contribution of the sequences was computed numerically. In doing so it was found that sequences longer than

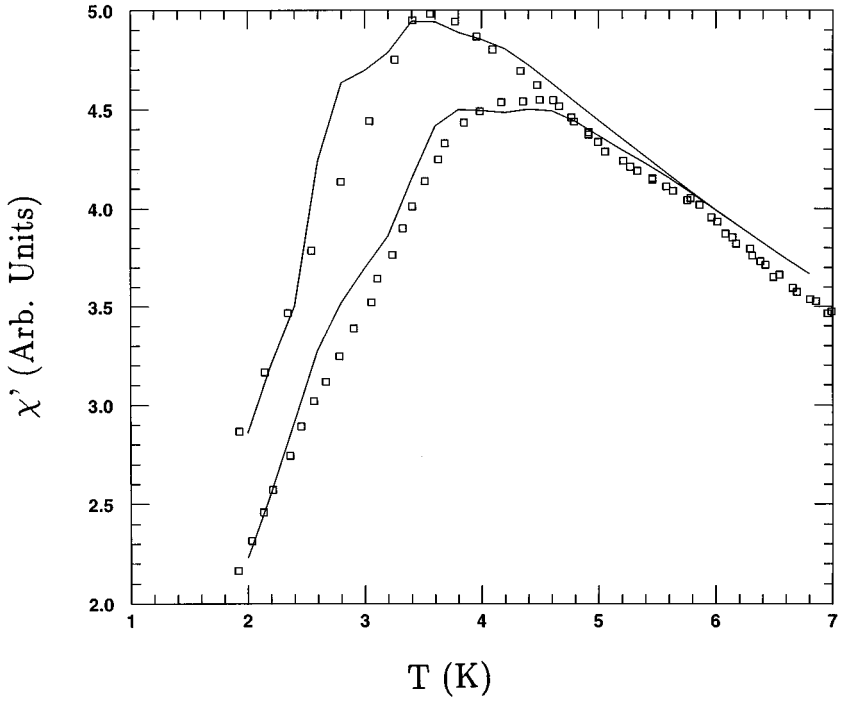

FIG. 1. The real part of the susceptibility of $Fe_{0.4}Mg_{0.6}Cl_2$. The points are taken from Wong *et al.* (Ref. 16), at 11 Hz, upper curve, and 2784 Hz, lower curve. The lines were calculated.

five spins made negligible contribution. Apart from the first spin to relax, no distinction was made between sequences which are confined to the domain boundaries and those in the interior of the domain.

The contribution of coherently relaxing clusters $\chi'_{\rm cl}$ was calculated as outlined above. These are small clusters, and they can also relax via the sequential mechanism. Thus only those clusters were included whose relaxation time was shorter than the sequential relaxation time of a sequence of half the spins in the cluster. $\chi'_{\rm cl}$ amounted to about 10% of the total.

The contribution of the static susceptibility $\chi'_{\rm st}$ was only about 20% of the total in the neighborhood of the susceptibility peak, and about 5 to 10% elsewhere. It was obtained using Eq. (9), with $\gamma = 0.5$.

Because the susceptibility data is quoted in ''arbitrary units'' $\chi' = A(\chi'_{\rm seq} + \chi'_{\rm cl} + \chi'_{\rm st})$ was calculated leaving ω_0 and A as adjustable parameters. The results, shown in Fig. 1, were obtained with $\omega_0 = 10^{11.5}$ rad/sec.

III. AGING

Summarizing the preceding analysis, relaxation occurs by a sequential process whereby flipping a spin relieves the constraint on a neighbor which then can flip thereby allowing its neighbor to flip, and so on. The sequences can start either on domain boundaries or in the interior of the clusters. The relaxation time of a spin n spins from where the sequence began is

$$\tau_{i,n}^{-1} = 2^{-n}\tau_i^{-1} + \tau_0^{-1}\sum_{k=0}^{n-1} 2^{-k} e^{-(2z_{n-k}J)/T}, \qquad (11)$$

where τ_i is the relaxation time of the spin that started the sequence, Q is an anisotropy barrier, and ω_0 is an attempt frequency.

The relaxation time of the spin which starts the sequence, τ_i, is, of course much shorter than the thermally activated relaxation times of the rest of the spins in the sequence, and the first term in Eq. (3) will dominate at temperature at and below the peak in the susceptibility. Consider temperatures low enough for this to be the case, neglecting the second, thermally activated term. If the system is in the field for a time t, spins will have relaxed for which $\tau_{i,n} \sim t$, and $2^n \tau_i \sim t$, or

$$n \sim \frac{1}{\ln 2} \ln(t/\tau_i). \qquad (12)$$

If the system has been rapidly quenched to a temperature where it is no longer paramagnetic, it can be expected to consist of a large number of domains. The faster the quench, the smaller the domains can be expected to be, and a large fraction of the spins will be on the boundaries. In that event sequences which start on the boundaries can make a major contribution to the total susceptibility.

If the temperature is high enough for relaxation to proceed, it can be expected that the domain size will increase with time, and the total length of domain boundaries will decrease. The sequences which can start on the boundary will be fewer, and the susceptibility will decrease.

In order to estimate the change in susceptibility with waiting time, consider that the material has been cooled to a temperature where the average domain of size R contains N spins, if the dimension is d,

$$N \sim R^d. \qquad (13)$$

In general, if the system has been quenched it can be expected that relaxation of spins on the boundary will increase the size of favorably oriented domains. Thus if n spins

in the unfavorably oriented domain relax, the cluster will now contain $N+n$ spins, and its size will increase to

$$R+\delta R\sim(N+n)^{1/d}. \tag{14}$$

The number of clusters in the sample is $\sim 1/R^d$ and each cluster has boundary $\sim R^{d-1}$, so the total area of domain boundary in the sample is $\sim R^{-1}$. If the fractal nature of the domain boundaries is considered, and the dimension of the boundary is d_s where $d-1<d_s<d$, the total boundary area becomes[4] $R^{-(d-d_s)}$. The contribution to the susceptibility from spins on the domain boundaries is proportional to the total area of domain boundary, and therefore is

$$\chi' \propto 1/(R+\delta R)^{d-d_s}=A/(N+n)^{(d-d_s)/d} \tag{15}$$

and, with Eq. (10),

$$\chi'=\frac{A}{\left[N+\dfrac{1}{\ln 2}\ln(t/\tau_i)\right]^{(d-d_s)/d}}. \tag{16}$$

An ideal way to monitor the change in susceptibility is with an ac measurement, and a beautiful series of experiments of this kind have been performed by Schins *et al.*[3] on the 2D Ising random-exchange ferromagnet $Rb_2Cu_{0.89}Co_{0.11}F_4$. The experiment consists of quenching the sample, and then monitoring the change in the ac susceptibility in *zero* field as a function of time.

Let the size of a 2D average cluster in the quenched material be R, and contain N spins; $N\sim R^2$. With Eq. (14), and neglecting the fractal nature of the boundary

$$\chi'=\frac{A}{(N\ \ln 2+\ln\omega_f+\ln t)^{1/2}} \tag{17}$$

or

$$(\chi')^{-2}=a+b\ln t, \tag{18}$$

N can be obtained from the average domain size. The constant A is not easy to calculate analytically. Using the theory above it can be evaluated numerically for $Fe_{0.4}Mg_{0.6}Cl_2$, but unfortunately aging data are not available for this material. On the other hand, aging data are available for $Rb_2Cu_{0.89}Co_{0.11}F_4$.[3] Although this material being a random-exchange system with a high degree of frustration is significantly different from that on which the analysis is based, as shown in Fig. 2, the time dependence of its susceptibility agrees very well with that predicted by Eq. (13).

IV. MAGNETIZATION IN CONSTANT FIELD

A sample cooled in zero field will initially consist of domains of up and down spins of equal volume. The size of the domains will depend on the cooling rate, and the waiting time. When a magnetic field is applied spins in the unfavorably oriented domains will begin to flip, and the sample will begin to develop a moment. If the number of spins in such a domain initially is N_0^-, N_0^- will depend on the waiting time before the magnetic field is turned on. The longer the time, the larger N_0^-, but the smaller the total boundary area, and the smaller the susceptibility. Using Eq. (12) the rate of spin reversal is initially

$$\left(\frac{dn}{d\ \ln t}\right)_{t\to 0}=\sum_i N_0^-\frac{p_i^{\rm cl}}{\ln 2}+(N_0^-)^{d_s/d}\frac{p_i^{\rm bd}}{\ln 2}=A+B, \tag{19}$$

where $p_i^{\rm cl}$ ($p_i^{\rm bd}$) is the probability of initiating a sequence in the cluster (on the boundary).

After the field has been on for a time t, N_0^- will have decreased. Ignoring correlations between the sequences, the number of spins in the unfavorably oriented domains is $N^-=N_0^--n$ spins on the average. Taking this into account by simply scaling Eq. (13)

$$\frac{dn}{d\ \ln t}=(A+B)\frac{N^-}{N_0^-}=C(N_0^--n), \tag{20}$$

where $C=(A+B)/N_0^-$.

The solution is

$$\ln(N_0^--n)=-C\ \ln t+\text{const.}$$

Since $n=0$ when $t<\tau_i$, letting $n=0$ at $t=\tau_i$ the const $=N_0^-e^{C\ \ln\tau_i}$, and

$$n=N_0^-(1-e^{-C\ \ln(t/\tau_i)}). \tag{21}$$

The magnetization is simply proportional to n, and the ''relaxation rate,'' S, of Svedlindh *et al.*[2] is

$$S=\frac{\partial M/H}{\partial\ \ln t}\propto N_0^-Ce^{-C\ \ln(t/\tau_i)}. \tag{22}$$

This expression provides the correct behavior initially, and at long times. However, it is observed that at temperatures near and below the peak in the susceptibility, S goes through a distinct maximum at times on the order of the waiting time before the field is turned on. As it stands this theory is inca-

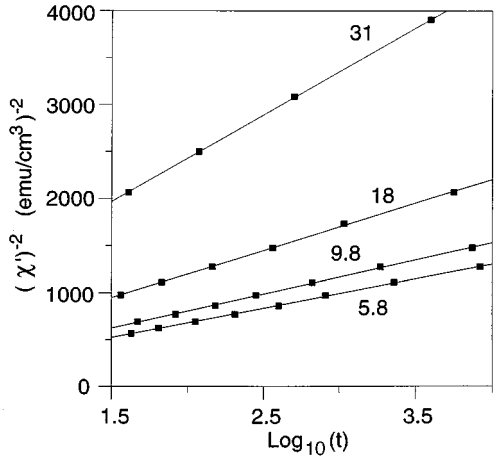

FIG. 2. Aging in the two-dimensional random exchange ferromagnet $Rb_2Cu_{0.89}Co_{0.11}F_4$. The reciprocal of the square of the susceptibility has been plotted against $\log_{10}(t)$ yielding a straight line in agreement with the predicted behavior of the sequential model. The data was taken from Schins *et al.* (Ref. 3). The numbers on the lines refer to the measurement frequency.

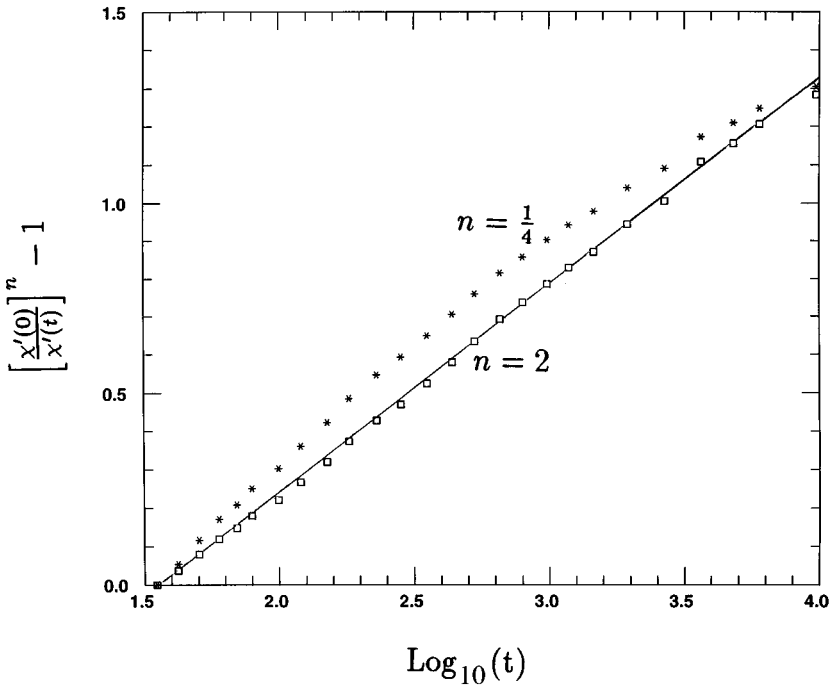

FIG. 3. Comparison of the predicted behavior of the droplet model (Ref. 4) and the sequential model with aging in the two-dimensional random exchange ferromagnet $Rb_2Cu_{0.89}Co_{0.11}F_4$. The reciprocal of the square of the normalized susceptibility has been plotted against $\log_{10}(t)$ yielding a straight line in agreement with the predicted behavior of the sequential model. On the other hand, if n is taken to be $\frac{1}{4}$, which is the prediction of the droplet model (Ref. 3), the line is clearly not straight. The two lines were scaled in order to facilitate the comparison. The data was taken from Schins *et al.* (Ref. 3).

pable of providing an explanation for this behavior. However, if correlations between the sequences are taken into account it may be possible for S to increase initially because the sequences have the effect of increasing the number of sites at which relaxation can start, since each flipped spin along the sequence can initiate another sequence.

V. DISCUSSION

A simple model for relaxation has been described which provides reasonable agreement between theory and experiment for the susceptibility. It predicts that the time dependent susceptibility during aging should be proportional to $(a+b\ln t)^{-d-d_s/d}$, which leads to $(a+b\ln t)^{1/2}$ for $d=2$ if the fractal nature of the domain boundary is neglected. The droplet model, on the other hand, predicts $(T\ \ln t/t_0)$,[4] and Schins *et al.*[3] have analyzed their data according to that law. However, as shown in Fig. 3, the agreement is clearly inferior to that obtained from the sequential model, in that a plot of $\chi'^{-1/4}$ against $\ln t$ does not yield a straight line.

By an obvious extension of the arguments presented, in three dimensions the susceptibility should age according to $(a+b\ln t)^{2/3}$. Suitable data for a 3D Ising system could not be found, but data for the relaxation of strain (i.e., creep) of the orientational glass $KBr_{0.5}KCN_{0.5}$ (Ref. 26) obeys this relation.[27]

The model describes a random magnet rather than a spin glass. However, Schins *et al.*[3] have also obtained data for aging in $Rb_2Cu_{0.67}Co_{0.33}F_2$. This composition yields a spin glass, and it is interesting that the magnitudes of the susceptibilities are about a factor of 5 smaller, and the shift in temperature of the susceptibility maximum is also much less.

Many materials classed as spin glasses show aging phenomena and a shift in the temperature of the susceptibility peak with frequency. The model described above would predict no time dependent phenomena for frustrated spins. Therefore it is tempting to ascribe these phenomena in spin glasses to constrained relaxational behavior of the spins that are not frustrated. It is interesting to speculate that as the number of unfrustrated spins decreases aging phenomena, the susceptibility, and the change in temperature of the susceptibility maximum with frequency also decreases. This could easily be verified.

VI. CONCLUSIONS

Constrained dynamical models in general, and the sequential model in particular, can account in large measure for relaxation in disordered Ising systems.

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