

Evidence for differing short- and long-time decay behavior in the dynamic response of the insulating spin-glass $\text{Eu}_{0.4}\text{Sr}_{0.6}\text{S}$

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The time decay of the thermoremanent magnetization of the insulating spin-glass $\text{Eu}_{0.4}\text{Sr}_{0.6}\text{S}$ has been measured with use of Faraday rotation to probe short- and long-time regimes (3 ms–3000 s) between $0.86T_g$ and $1.04T_g$. Aging effects are negligible in this compound to within our experimental accuracy, so that equilibrium response is accessible. We find evidence for two time regimes. The decay is of a power-law form at short times. Beyond a well-defined crossover time, the decay is of a stretched exponential form. The crossover time decreases as the temperature increases from below T_g , and drops abruptly at T_g . No field dependence of the crossover time is observed for the field range investigated (5–40 Oe). Connection is made with computer simulations of Ogielski, and with the existence of a finite length scale in the equilibrium spin-glass state.

I. INTRODUCTION

The controversy raised by the characterization of the decay of the thermoremanent magnetization (TRM) in spin-glasses began with the work of Chamberlin *et al.*¹ They identified the decay of the TRM over three decades of time with a “stretched exponential” form:

$$\sigma = \sigma_0 \exp[-(t/\tau_p)^{1-n}] \quad (1)$$

τ_p is the apparent response time.

The relevance of “aging” was stressed afterwards:^{2,3} the response of the system depends upon the amount of time, t_w , one waits at an assigned temperature (below T_g , the spin-glass transition temperature) before switching off the field. This problem has been examined quantitatively for various spin-glasses.^{3–8} One suggestion has been that one should define an effective time, λ , given by

$$\lambda = [t_w / (1 - \mu)] [(1 - t/t_w)^{1-\mu} - 1] \quad (2)$$

(where μ is an “aging” parameter,^{6–8} if the system is aging while the TRM is decaying. In this picture, the decay is represented by the product of a power law by a stretched exponential

$$\sigma = \sigma_0 \lambda^{-\alpha} \exp[-(\lambda/\lambda_0)^{1-n}] \quad (3)$$

In the most general case, λ , λ_0 , and σ_0 depend on the waiting time t_w . The effective time λ becomes relevant when the measurement or laboratory time t approaches the waiting time t_w . As long as $t \ll t_w$, $\lambda \simeq t$. However, this analysis exhibits an inconsistency when confronted with susceptibility measurements. Though the fluctuation-dissipation theorem (FDT) is shown to be valid,⁸ there remains a specific discrepancy between the exponent α as defined by Eq. (3) and the value which is extracted from χ'' measurements in the proper frequency range. This point will be discussed later.

For our part, we have been unable to find any evidence for aging in $\text{Eu}_{0.4}\text{Sr}_{0.6}\text{S}$, to within our experimental accu-

racy, over the range of temperature ($0.86T_g$ – $0.97T_g$) or field⁹ (5–16 Oe) where such effects are expected to be observable. We therefore claim that the response of this compound is not affected by aging phenomena.

The purpose, therefore, of our experiments is to examine the time dependence of the TRM in this spin-glass compound over a wide range of times. The absence of aging effects is evidence of the field-cooled state being at equilibrium. Hence, our study of the time dependence of the TRM enables us to examine spin-glass dynamics *starting* from an equilibrium condition. Our most important result is our observation of two distinct time regimes for the decay of the TRM. The decay at short times is found to obey a power law (for times less than 3 s in our accessible temperature range). The form of the decay of the TRM at longer times can be satisfactorily characterized by a stretched exponential. The power-law regime is present for times as long as 1 s even at $0.97T_g$, but then drops sharply to below a millisecond time scale at T_g . Slightly above T_g (up to $1.04T_g$), a simple stretched exponential describes the decay over our full time window.

We shall suggest that the occurrence of the two time regimes implies the existence of a characteristic length, L^* , relevant to the spin-glass phase, which could conceivably be related to a typical domain size. The early time period then refers to diffusion over length scales $L < L^*$, while the longer time domain to length scales $L > L^*$. The crossover time t_{co} specifies the time for diffusion to the length L^* . At the present time, the only microscopic theory which has introduced an explicit length scale is that of Fisher and Huse,¹⁰ and Bray and Moore.¹¹

II. EXPERIMENTAL PROCEDURE

We have performed our measurements on the same sample for which we carried out our dynamic scaling analysis.¹² The freezing temperature was determined by the divergence of a characteristic relaxation time, when

approaching T_g from above, and was found to be $T_g = 1.50 \pm 0.02$ K.

Our experimental approach uses Faraday rotation as a probe of the TRM. The shortest time (3 ms) is set by the experimental time constant of our apparatus. The longest time (3000 s) is chosen so that the drifts of our setup are less than, or at worst comparable to, the variation of the TRM on that time scale. The signal is acquired one decade after another, with sufficient overlap in order to accurately link the regimes together. This procedure allows us to use a longer integration time when recording on longer time scales, and therefore to achieve a better signal-to-noise ratio. The accuracy achieved under such conditions is, over the entire time range, 8×10^{-4} degrees for the Faraday rotation angle; which in turn represents approximately 3×10^{-4} of the field-cooled magnetization at our smallest field (5 Oe).

III. DESCRIPTION AND ANALYSIS OF THE RESULTS

A. Aging

The search for aging has been performed over the last three decades of our time scale. The effect is known to be more pronounced when the time scale is comparable to the waiting time. As our shortest available waiting time is 300 s, we have compared the decay of the TRM recorded between 3 s and 3000 s for $t_w = 300$ s and $t_w = 8000$ s. We have found no significant difference between the two measurements for various fields (5–16 Oe) or temperatures (1.290–1.450 K). Comparing $\text{Eu}_{0.4}\text{Sr}_{0.6}\text{S}$ to Ag:Mn (Ref. 7) or $\text{CdIn}_{0.3}\text{Cr}_{1.7}\text{S}_4$,⁸ the difference expected between the two relaxation curves for two such waiting times would be quite measurable in our setup. We are therefore led to the conclusion that there is much less aging, if any, in the $\text{Eu}_{0.4}\text{Sr}_{0.6}\text{S}$ system.

An alternate reason for the lack of observation of aging effects could also be that the system ages very slowly. However, the system would then be expected to thermalize nearly instantaneously for temperatures above T_g . There would presumably then have to be a temperature range in the immediate vicinity of T_g where aging would become observable. This is not the case. We conclude that, in contrast with other spin-glasses, $\text{Eu}_{0.4}\text{Sr}_{0.6}\text{S}$ achieves thermal equilibrium on a time scale short compared to our experimental one.¹³ Therefore we claim that we measure the decay of the TRM from an equilibrium state (the *equilibrium* response).

B. Characterization of the decay of the TRM

We exhibit in Fig. 1 the time dependence of the TRM measured at 1.290 K ($0.86T_g$) for two different fields (5 and 25 Oe). We reproduce in Fig. 2 three representative TRM decay curves at 1.290, 1.450, and 1.510 K (the last approximately equal to T_g to within ± 0.01 K). The two curves in Fig. 1 have been arbitrarily shifted in order to make more obvious the fact that the short-time behaviors are very much the same, whereas the long-time behaviors differ. These figures demonstrate two clearcut time decay

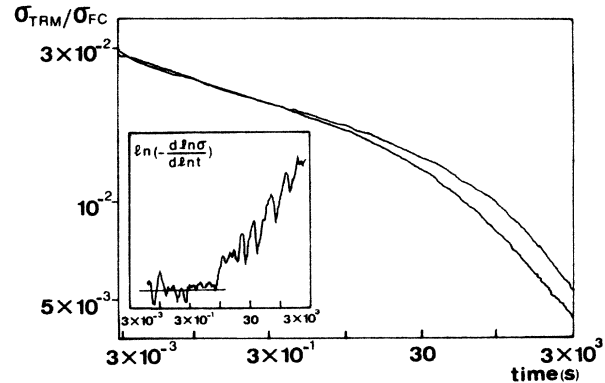


FIG. 1. Log-log plot of the time decay of the TRM at $T = 1.290$ K for $H = 5$ Oe and $H = 25$ Oe. Numerical values on the ordinate axis are referred to the field-cooled magnetization σ_{FC} . Inset: plot of $\ln[-d(\ln\sigma_{TRM})/d(\ln t)]$ vs $\ln t$ for $T = 1.290$ K and $H = 16$ Oe.

regimes for the TRM. Plotting $\ln[-d(\ln\sigma_{TRM})/d(\ln t)]$ versus $\ln t$ (inset of Fig. 1) shows rather convincingly a break in the time variation of the decay of the TRM. At short times, the plot is constant, signifying a power-law time decay. The long-time regime fits a straight line, representing a stretched exponential form on this plot.

Our data are therefore highly suggestive of two different time decay regimes. A power law $t^{-\beta}$ describes the short-time regime. We define a crossover time t_{co} as being the time where the experimental points depart from a straight line on a log-log plot by more than twice the rms deviation. We then fit the remaining longer-time regime to the stretched exponential form, Eq. (1). As can be seen from Fig. 2, the fits are excellent. They show that

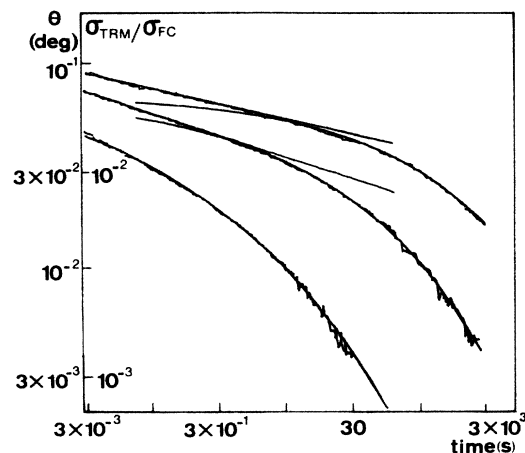


FIG. 2. Log-log plot of the time decay of the TRM at three different temperatures (1.290, 1.450, and 1.510 K) for $H = 5$ Oe. The left-hand side of the ordinate axis gives the Faraday rotation angle in degrees. The right-hand side gives the TRM referred to the field-cooled magnetization. The power law is represented by the solid straight line, the stretched exponential by the solid curve. Nothing but the noise differentiates the experimental curves from the fits.

TABLE I. Variation of the exponent β vs temperature when fitting the short-time decay to a simple power law $t^{-\beta}$. The values given are within ± 0.003 . $T_g = 1.50 \pm 0.02$ K.

T (K)	1.290	1.320	1.380	1.420	1.450	1.480	1.500
β	0.071	0.074	0.080	0.093	0.100	0.130	0.164

the crossover time is experimentally well defined, and that the decay of the TRM does actually "switch" from one behavior to the other.

We report the values for the power-law (short-time) decay exponent β at various temperatures in Table I. Table II contains the values of the parameters for the stretched exponential (long-time) decay derived as described above. Below T_g ($T \leq 1.50 \pm 0.02$ K) the temperature variation of τ_p is similar to what has been observed for stretched exponential decay in metallic spin glasses.⁴ That is, τ_p becomes shorter rapidly as T_g is approached from below. For our system, however, we are unable to obtain a precise variation of τ_p with temperature because of the limited temperature range over which we can make measurements. Above T_g ($T > 1.50$ K), a power-law regime is not present in the time window in which we can make measurements. Instead, we find that a stretched exponential form for the time decay of the TRM fits our measurements over our time scale of measurement. That time scale shrinks with increasing temperature (3 ms \rightarrow 3 s at $T = 1.56$ K; 3 ms \rightarrow 300 ms at 1.58 K). In order to compare these results to those of, for example, Ref. 12, one must be able to work at a well-determined reduced temperature $(T - T_g)/T_g$. This quantity is reasonably well determined at $T = 1.56$ or 1.58 K, but at 1.51 K possesses a very large uncertainty because of the uncertainty in T_g and the closeness of T to T_g . Moreover, across T_g , the decay undergoes a dramatic change because τ_p is expected to jump from a very large value above T_g (critical slowing down), to a very small value below T_g . Such a large variation in τ_p over such a narrow temperature range for which the reduced temperature is not well determined results in very large uncertainties in the experimental data, and makes analytic comparisons difficult in the immediate vicinity of T_g . These considerations explain why the expected variation of τ_p through T_g is smeared out, but the trends are clear. In fact, in the temperature range where the reduced temperature is well defined ($T > 1.56$ K), the temperature variation of τ_p is consistent with the $z\nu$ exponent already determined for this system.¹²

We now discuss the form for the decay of the TRM described by Eq. (3) from Ref. 8, in terms of its relevance to

our results. It is stated in there that, at short times ($t \ll t_w$), the *equilibrium* decay of the TRM is a power law. The stretched exponential form for the decay is said to be caused by the aging of the system. Our present experimental results argue against the latter point.¹³ As described above, we believe that we measure the equilibrium response. We find that a power law does not describe the decay over the full time range. Therefore, the *stretched exponential* form for the TRM decay can also be an *equilibrium response*.¹⁴

We should note that, nevertheless, it is possible to use Eq. (3) to fit our data if we set $\lambda = t$ in order to represent the equilibrium limit. One can obtain a reasonably satisfactory fit for the values of α reported in Table III. However, there are some rather fundamental problems with the form of Eq. (3) for the time decay of the TRM which mitigate against its use. Presumably, according to linear response, whether one extracts α from the time decay of the TRM, or from the harmonic response χ'' (derived from a power-law decay), the two values should be the same. Close to T_g , it has been already noted⁸ that they are not. This is consistent with our interpretation of our measurements where we find the *equilibrium* response not to be a simple power law. This difference may account for the observed discrepancies in α , but it also raises doubts about the adequacy for Eq. (3) being a proper description of the TRM decay.

For $T \geq T_g$, our data show that a simple stretched exponential accounts satisfactorily for the decay of the TRM over the full time range of our measurements (see Fig. 2). This appears to be consistent with what one should presumably find above T_g , namely, eventually a simple exponential decay. However, the measured stretched exponential form appears to be heading towards a simple exponential form as the temperature is increased. Table II shows that n is decreasing above T_g as T increases ($n = 0$ is equivalent to simple exponential decay). Our interpretation for this continuity across T_g is that the power-law regime occurs at shorter and shorter times as the temperature increases from below towards T_g . Above T_g , this short-time regime is inaccessible on the time scale of our experiments, allowing for observation of only the stretched exponential decay.

TABLE II. Typical values for the stretched exponential parameters, n and τ_p , at nine temperatures. The uncertainties in n and τ_p are listed below the relevant entries.

T (K)	1.290	1.320	1.360	1.450	1.480	1.500	1.510	1.560	1.580
n	0.70	0.84	0.83	0.80	0.78	0.84	0.82	0.66	0.65
Error	± 0.02	± 0.02	± 0.02	± 0.02	± 0.03	± 0.03	± 0.04	± 0.05	± 0.05
τ_p (s)	750	180	180	21	1.3	0.04	0.03	0.015	0.003
Error	$\pm 20\%$	$\pm 20\%$	$\pm 20\%$	$\pm 20\%$	$\pm 20\%$	1 order of magnitude		$\pm 100\%$	$\pm 100\%$

Thus there appear to be a number of reasons to believe that there are two distinct time decay regimes for the TRM. We have tried to rule out one of the two possible decay characterizations. We have compared both exponents β and α extracted from the time decay of the TRM (Tables I–III) to the values obtained from our susceptibility data in the corresponding frequency range ($\omega \geq 1$ Hz). One must be careful, however, because of the large demagnetizing fields occurring in $\text{Eu}_{0.4}\text{Sr}_{0.6}\text{S}$.¹² It is easy to show that, if the actual χ'' is proportional to ω^α , this would remain true for the measured χ'' in the presence of demagnetizing fields with a slightly different exponent $\alpha + \delta\alpha$. Also, the FDT should hold for the measured values as well as for the actual ones as already discussed.¹⁵ We are able to show that, for our sample, $\delta\alpha$ is negligible compared to α . The measurement of α through the frequency variation of the susceptibility is unfortunately rather inaccurate. We find $\alpha = 0.06 \pm 0.03$ at 1.3 K and $\alpha = 0.085 \pm 0.03$ at 1.45 K. These values are consistent with fits to both forms for the decay of the TRM (Tables I–III). This check is therefore incapable of distinguishing between the two.

The results in Tables I–III exhibit, however, a few interesting features. The values for β that we find, and the α values found from the χ'' measurements in $\text{CdIn}_{0.3}\text{Cr}_{1.7}\text{S}_4$,⁸ are in remarkable agreement even in the temperature range where the authors of Ref. 8 observed discrepancies with their TRM data using Eq. (3) to determine α . Also note that the difference between our β and α values, very close to T_g , exhibits a similar discrepancy. This suggests that our fit to a simple power law at short times, and a stretched exponential at longer times, provides a more consistent characterization of the decay.

IV. PHYSICAL INTERPRETATION

We wish now to discuss the physics which may underly these two time regimes. It is tempting to think of the crossover time t_{co} as suggesting two different diffusion processes, one at short-, the other at large-length scales. That is, the crossover time implies a crossover length. After switching off the field, relaxation may begin by nucleation at a site, then spreading over a spatial region which grows in time. At the crossover time, the character of the diffusion would change, leading to a change in the time decay of the TRM.

It is interesting to note that the picture introduced by Fisher and Huse,¹⁰ and developed by Bray and Moore,¹¹ is not inconsistent with this concept. These models have been developed for Ising spin-glasses. We believe that such a model could be relevant to $\text{Eu}_{0.4}\text{Sr}_{0.6}\text{S}$ because of

TABLE III. Temperature dependence of the α exponent from Eq. (3). The values contain a ± 0.003 uncertainty.

T (K)	1.290	1.320	1.380	1.450	1.480	1.500
α	0.067	0.066	0.074	0.087	0.094	0.150

TABLE IV. Values for the exponent β , and the stretched exponential parameters, n and τ_p , at two temperatures, $T = 1.290$ and 1.380 K ($T_g = 1.500 \pm 0.02$ K), as a function of cooling field H . The errors for β are ± 0.003 ; for n are ± 0.03 ; and for τ_p are $\Delta\tau_p/\tau_p = \pm 0.2$.

H (Oe)	β	n	τ_p (s)
$T = 1.290$ K			
2.5	0.069	0.75	800
5.0	0.070	0.76	750
10.0	0.070	0.79	340
16.0	0.070	0.86	80
25.0	0.077	0.83	70
40.0	0.083	0.88	3
$T = 1.380$ K			
5.0	0.082	0.82	200
16.0	0.090	0.82	40
40.0	0.12	0.88	0.03

the large random dipole anisotropy present in this system. Such anisotropy can lead to effective Ising-like behavior as previously discussed in Ref. 12.

According to Ref. 11, “. . . as the temperature is increased from zero, $L^* \sim T^{-2/\xi}$ is the length scale at which the entropy first plays an important role, i.e., the length scale at which the ordering pattern $\{\langle S_i \rangle_T\}$ (where $\langle \rangle_T$ indicates a thermal average) loses coherence with the ground state.” The numerical factor in the exponent depends upon the particular bond distribution, but the trend of L^* with increasing temperature is clear. It is tempting to interpret the power-law decay of the TRM at short times to diffusion within the strongly correlated spatial region $L < L^*$. According to Fisher and Huse,¹⁰ the equilibrium autocorrelation function, hence the TRM according to linear response, should decay as $(\ln t)^{-\theta/\psi}$. Our data do not appear to support this explicit time dependence. For longer times, the diffusion would take place over a length scale where “. . . the ground state is unstable against arbitrarily weak perturbations to the bonds” and could therefore generate a different time decay for the TRM.

In order to more fully characterize the two time regimes, we have examined the temperature and the field dependence of the exponent β , and of the crossover time t_{co} .

The temperature dependence of the power-law exponent β is reported in Table I, while the temperature dependences of the stretched exponential parameters are given in Table II. The β values are systematically higher (by a factor of the order of 2) than those found in numerical simulations (see below).

We exhibit the temperature variation of the crossover time t_{co} in Fig. 3. We find a very small decrease, if any, as T increases, then a sharp drop at T_g . No power-law region for the decay of the TRM at 1.51 K ($T_g = 1.50 \pm 0.02$ K) is found in Fig. 2. This means that, in a very narrow range of temperature near T_g , t_{co} shifts to below our available experimental time scale. Because of such behavior, we are led to infer that the characteristic length is shortening rapidly in the vicinity of T_g . We

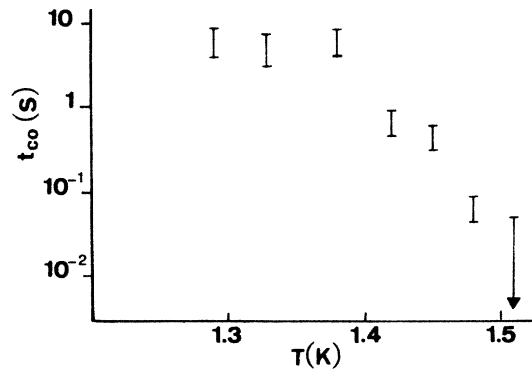


FIG. 3. Plot of the crossover time t_{co} vs temperature for a fixed field $H = 5$ Oe. The arrow at 1.510 K is a reminder of the fact that at this temperature, t_{co} might be well below the millisecond time range as well.

cannot determine at which precise temperature because the very short measuring times required are not available in our apparatus.

Because of the very limited temperature range below T_g where we can determine t_{co} (we cannot measure below $0.86T_g$), we cannot make a direct comparison between the temperature dependence for the crossover length inferred from our data, and the predictions of Bray and Moore for the temperature dependence of L^* , their calculation being limited to the regime near $T = 0$.

We now turn to the magnetic field dependence of our parameters. Our data are presented in Table IV. Note that, at $0.86T_g$ (1.290 K), the response does not depend upon the value of the field (linear-response regime) for fields up to 5 Oe. Thus, for this field regime, the response is comparable to the lower field experiments exhibited in Refs. 4–8. Checking the linear-response field range with the same accuracy close to T_g was not possible, but we note that the interpretation which leads to Eq. (3) has also been used for much higher fields, out of the linear-response regime.¹⁶ At $0.86T_g$, we do not find any significant variation of β up to 16 Oe. In contrast, the parameters in the stretched exponential vary quite dramatically with field at and above 10 Oe. This is reminiscent of what has been found near T_g for the Ag:Mn spin-glass.¹⁷

However, we do not find a measurable field dependence of the crossover time t_{co} for fields varying from 5 to 40 Oe, either at 1.290 K ($0.86T_g$) or 1.400 K ($0.92T_g$). At the latter temperature, a 40-Oe field takes the system close to the “De Almeida–Thouless line” as determined from dynamical scaling in this system from previous measurements¹⁸ (but not so close that one would expect for t_{co} a comparable change to that found near T_g).

Numerical simulations are instructive to examine in order to see how a characteristic length may give rise to a crossover decay time. Ogielski¹⁹ has calculated the time decay of the dynamic autocorrelation function over eight decades of time [10^8 Monte Carlo steps (MCS)]. In the spin-glass phase, only algebraic decay could be observed.

Above T_g , he found that the decay could be empirically described by the product of a power law times a stretched exponential. We argue that his simulations are consistent with what we observe. The samples are very small in numerical simulations. Therefore a physical length scale would be expected to be much larger than the size of the simulation sample. At and below T_g , one would expect a single domain throughout the sample, and therefore a single relaxation regime, namely a power law.

Above T_g , the relevant length scale, namely the correlation length, would start from an atomic scale at high enough temperatures, and become very large at T_g . A crossover between two different relaxation regimes associated with a crossover between short- and long-length scales could show up in a limited temperature range near to T_g . We argue that it does through Ogielski’s observation of the product of a power law by a stretched exponential form for the decay of the dynamic autocorrelation function. As we have shown, this can very likely also be analyzed in terms of the two time regimes.

It is also interesting to examine the effect of finite size for various lattice sizes. Ogielski finds that “. . . for a 16^3 lattice, the relaxation is not affected by finite size for times $t < 10^6$ MCS.” Figure 9 of Ref. 18 shows conclusively that the power law is indeed unchanged up to 10^6 MCS, and that for longer times, the decay departs from a power law. This suggests (not surprisingly) that the occurrence of a length scale (in this case, the finite size of the sample) sets a time scale beyond which the decay is modified. Such similarities between our data and the simulations are striking and are in agreement with an interpretation involving the existence of a crossover length scale.

V. SUMMARY

In conclusion, we have argued that the equilibrium decay of the thermoremanent magnetization of the $\text{Eu}_{0.4}\text{Sr}_{0.6}\text{S}$ spin-glass exhibits two distinct time regimes, separated by a crossover time t_{co} . The temperature variation of t_{co} displays the temperature variation of a characteristic length scale. A power-law form is found at short times, and can be ascribed to relaxation in a highly correlated spatial region of the spin-glass phase in analogy with the explicit picture of Bray and Moore.¹⁰ For times longer than t_{co} , the decay takes the form of a stretched exponential, and one can ascribe relaxation to take place in the uncorrelated spatial regime.

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