

## Extraction of the Spin Glass Correlation Length

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The peak of the spin glass relaxation rate,  $S(t) = d[-M_{\text{TRM}}(t, t_w)/H]/d \ln t$ , is directly related to the typical value of the free energy barrier which can be explored over experimental time scales. A change in magnetic field  $H$  generates an energy  $E_z = N_s \chi_{\text{fc}} H^2$  by which the barrier heights are reduced, where  $\chi_{\text{fc}}$  is the field cooled susceptibility *per spin*, and  $N_s$  is the number of correlated spins. The shift of the peak of  $S(t)$  gives  $E_z$ , generating the correlation length,  $\xi(t, T)$  for Cu:Mn 6 at. % and CdCr<sub>1.7</sub>In<sub>0.3</sub>S<sub>4</sub>. Fits to power law dynamics,  $\xi(t, T) \propto t^{\alpha(T)}$  and activated dynamics  $\xi(t, T) \propto (\ln t)^{1/\psi}$  compare well with simulation fits but possess too small a prefactor for activated dynamics. [S0031-9007(98)08108-3]

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The study of the irreversible behavior of the spin glass magnetization under a change of magnetic field allows exploration of the available states of a random frustrated system [1,2] There are various representations for the long time evolution and the dynamics of spin glasses [3–5], but a coherent, overall accepted real space description remains lacking [6]. The purpose of this paper is to extract a time and temperature dependent spin glass correlation length from a specially structured set of experiments and to compare our results with available theoretical predictions.

The definition of a correlation length for a spin glass is difficult to express in measurable terms. Marinari *et al.* [7] and Kisker *et al.* [8] introduced the time dependent equal time correlation function at time  $t$ . In the notation of Ref. [7],

$$G(x, t) = V^{-1} \overline{\sum_i \langle \sigma_{i+x} \tau_{i+x} \sigma_i \tau_i \rangle_t}, \quad (1)$$

where the average is done at time  $t$ , and  $\sigma_i$  ( $\sigma_{i+x}$ ) and  $\tau_i$  ( $\tau_{i+x}$ ) represent the  $z$  component of Ising spins at sites  $i$  ( $i+x$ ) in two thermalized configurations in a box of volume  $V$ . To avoid accidental contributions to  $G(x, t)$ , the two configurations are chosen to have zero overlap,  $q = V^{-1} \sum_i \sigma_i \tau_i$ . References [7] and [8] both observed, through their simulation studies, that for large times  $t$  the correlation function  $G(x, t)$  differs from zero for distances not too much larger than a dynamic correlation length  $\xi(t, T)$ . Simulations of Marinari *et al.* [7] obtain satisfactory fits for  $\xi(t, T) \propto (t/\tau_0)^{\alpha T/T_g}$ , appropriate to power law dynamics [4], while Kisker *et al.* [8] fit satisfactorily both this proportionality and equally well  $\xi(t, T) \propto [(T/T_g) \ln(t/\tau_0)]^{1/\psi}$ , appropriate to activated dynamics [5].

Our measurements consist of cooling a sample in a magnetic field through the glass temperature  $T_g$  to the measuring temperature  $T$ , waiting a time  $t_w$ , then cutting

the field to zero and measuring the decay of the magnetization. This generates the response function

$$S(t) = d \left[ \frac{-M_{\text{TRM}}(t, t_w)}{H} \right] / d \ln t, \quad (2)$$

where  $M_{\text{TRM}}(t, t_w)$  is the thermoremanent magnetization at time  $t$  after cutting the magnetic field to zero.

Our approach, justified previously through magnetic field cycling [9] and used to determine the Parisi physical order parameter  $P(q)$  [1], makes use of the scaling relationship introduced by Vincent *et al.* [2]. They show that barrier heights surmounted during aging are reduced upon a change in magnetic field by  $E_z$ , a quantity related to the change in Zeeman energy. Our model [1] assumes that barriers  $\Delta < E_z$  in the initial state (before the magnetic field is changed) are quenched, with the population of occupied states transitioning essentially instantaneously to those states of lowest energy corresponding to the new value of the magnetic field. The associated change in magnetization is referred to as the reversible part of the magnetization. The states “left behind” comprise the irreversible component and decay by diffusion to the sink created by the quenching of barriers  $\Delta < E_z$ .

At small magnetic field changes, states at the barrier height corresponding to the waiting time  $t_w$ ,

$$\Delta(t_w, T) = k_B T (\ln t_w - \ln \tau_0), \quad (3)$$

where  $1/\tau_0$  is an attempt frequency  $\approx k_B T_g / \hbar$ , diffuse towards the sink, resulting in a local equilibration of state occupancies on a time scale of the order of  $t_w$ . This causes [10] a peak in the response function  $S(t)$ , Eq. (2), for measurement times  $t$  close to the waiting time  $t_w$ . When the magnetic field change is increased,  $E_z$  increases, and the *effective* barrier height from which diffusion takes place to the sink is  $\Delta(t_w, T) - E_z$ . This

results in a *reduction* of the measuring time at which  $S(t)$  peaks and was first noted for experiments upon the insulating thiospinel  $\text{CdCr}_{1.7}\text{In}_{0.3}\text{S}_4$  by Vincent *et al.* [2] and the amorphous system  $(\text{Fe}_x\text{Ni}_{1-x})_{75}\text{P}_{16}\text{B}_6\text{Al}_3$  by Djurberg *et al.* [11]. We follow their analysis and associate the peak in  $S(t)$  with an effective waiting time,  $t_w^{\text{eff}}$ , so that as a function of magnetic field change,

$$\Delta(t_w, T) - E_z = k_B T (\ln t_w^{\text{eff}} - \ln \tau_0). \quad (4)$$

Systematic experiments were carried out by us for the metallic spin glass Cu:Mn 6 at. % and for the thiospinel [12]. Representative curves for  $-M_{\text{TRM}}/H$ ,  $S(t)$  are exhibited in Fig. 1.

Systematic studies have been performed for a variety of waiting times,  $t_w$ , magnetic field changes,  $H$ , and temperatures,  $T$ , enabling us to extract values of  $E_z$  over a wide range of parameter space.

We interpret  $E_z$  to be the magnetic energy associated with a change in magnetic field. In the field cooled state, we associate each spin to possess a magnetic susceptibility  $\chi_{\text{fc}}$  *per spin*, which we can calculate from the total value of the field cooled magnetic susceptibility  $M_{\text{fc}}/H$  by dividing by the total number of spins  $N$ . Then the number of spins participating in barrier quenching (and barrier hopping) can be derived from setting

$$E_z = N_s \chi_{\text{fc}} H^2. \quad (5)$$

The quantity  $N_s$  defines a volume over which the spins are effectively locked together for barrier hopping [2,13] the radius of which we define as the spin glass correlation length  $\xi$ . Were  $N_s$  to be the sum of smaller independent clusters, the activation energy would relate to smaller

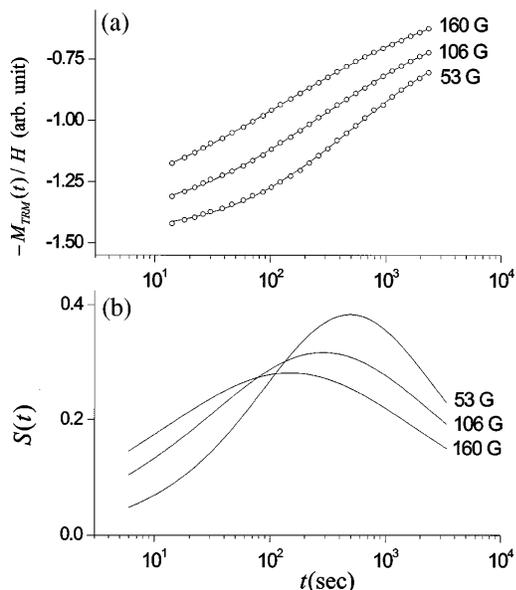


FIG. 1. (a) Plots of the data points for  $-M_{\text{TRM}}/H$  vs measurement time  $t$ , with the solid lines the analytic fit; and (b)  $S(t)$  defined in Eq. (2) vs  $t$  for Cu:Mn 6 at. % at various changes in magnetic field.  $S(t)$  was calculated by taking the time derivative of the analytic fit (solid line) in (a).

barriers, and  $S(t)$  would not shift its peak from the vicinity of measurement times  $\approx t_w$ .

Extensive studies were made of both Cu:Mn 6 at. % and  $\text{CdCr}_{1.7}\text{In}_{0.3}\text{S}_4$  to examine the  $H^2$  dependence predicted in Eq. (5). Experimental results for both systems, with  $H$  scales adjusted to their respective de Almeida–Thouless lines [14], are exhibited in Fig. 2.

Experimentally, the measured quantity  $N_s$  depends both upon waiting time  $t_w$  and temperature  $T$ . It represents the number of correlated spin units which flip together within a volume  $\xi^3$  growing as a function of  $t_w$  and  $T$ .

We summarize our results for  $N_s$  in Cu:Mn 6 at. % as a function of waiting time  $t_w$  for fixed temperature  $T = 0.89T_g = 28$  K in Fig. 3. The solid curve in Fig. 3 is a fit to the form derived from power law dynamics [4],

$$\xi(t_w, T) = 0.653(t_w/\tau_0)^{0.169T/T_g}, \quad (6)$$

with  $1/\tau_0 = 4.1 \times 10^{12} \text{ sec}^{-1}$ . A similar analysis for the thiospinel at  $T = 0.72T_g = 12$  K, but for only two waiting times, 1100 and 3410 sec, gives  $\xi(t_w, T) = 0.53(t_w/\tau_0)^{0.132T/T_g}$ . The two expressions are remarkably close and suggest that Eq. (6) may be universal (see Fig. 4 below).

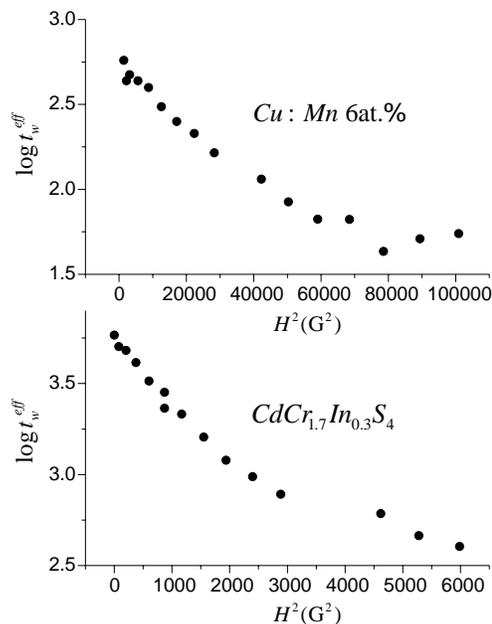


FIG. 2. A plot of  $\log t_w^{\text{eff}}$  [equivalently  $E_z$  from Eq. (4)] vs  $H^2$  for Cu:Mn 6 at. % ( $T/T_g = 0.83$ ,  $t_w = 480$  sec) and  $\text{CdCr}_{1.7}\text{In}_{0.3}\text{S}_4$  ( $T/T_g = 0.72$ ,  $t_w = 3410$  sec) at fixed  $t_w$  and  $T$ . The dependence is linear for magnetic fields less than 250 and 45 G, respectively, then breaks away to a slower dependence. The ratio of these “break” fields is in agreement with the respective de Almeida–Thouless lines. We do not have a satisfactory explanation for this change in slope. A different description, Ref. [2], predicts a linear dependence of  $E_z$  upon  $H$ , which can be made to fit the data over the entire range of  $H$  for the thiospinel, but with a significant deviation at small field changes. We have chosen to focus in the text on the fit at small field changes because we want to be certain to be in the linear regime for our analysis.

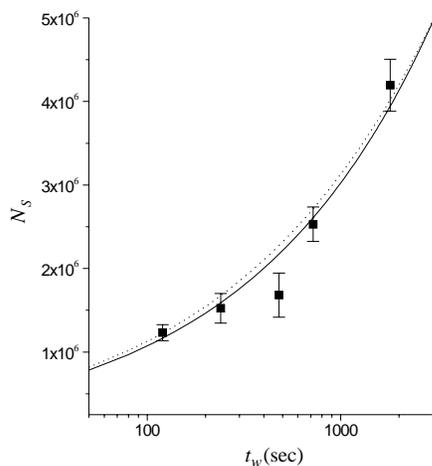


FIG. 3. A plot of  $N_s$ , the number of spins participating in barrier quenching (and barrier hopping) from Eq. (4) for Cu:Mn 6 at.%, vs  $t_w$  on a log scale at fixed temperature  $T = 0.89T_g = 28$  K. The solid curve drawn through the points is the prediction for power law dynamics, Ref. [4], while the dashed curve is the prediction for activated dynamics, Ref. [5], with their exchange factor set equal to  $T_g$  (i.e., independent of  $T$  and  $t$ ). As can be seen from the two curves, the two fits are equally good.

The dashed curve in Fig. 3 is a fit to the predictions of activated dynamics [5], leading to

$$\xi(t_w, T) = 10^{-5}[(T/T_g) \ln(t_w/\tau_0)]^{1/0.21}, \quad (7)$$

where we have taken the nominally  $t, T$  dependent exchange factor of Ref. [5] to be a constant equal to  $T_g$ . As seen from Fig. 3, both Eqs. (6) and (7) fit remarkably well. However, the factors in Eq. (7) deserve comment. The prefactor is very troublesome, for it would give  $\xi \approx O(1)$  for  $t_w/\tau_0 = 10^5$ , while simulations [7,8,15] clearly see an increasing dynamic correlation length in the regime  $t/\tau_0 < 10^5$ . The exponent in Eq. (7), equal to  $1/\psi$ , is also at the lower allowable value [5],  $\psi = 0.2$ .

A comparison of Eq. (6) with the numerical results of the simulations in Refs. [7], [8], and [15] is remarkable. The prefactor in the first is of order unity, with an exponent equal to  $0.16T/T_g$  (compare with our exponent  $0.169T/T_g$ ), very close to our fitted results. The second finds a prefactor of order unity and an exponent between  $0.12T/T_g$  and  $0.13T/T_g$ , and the third a prefactor close to unity, with an exponent  $0.13T/T_g$ . All three are in substantial numerical agreement with the experimental result, Eq. (6).

In addition, Kisker *et al.* [8] find a proportionality of the same form as Eq. (7) for activated dynamics [5], but with an exponent  $1/\psi = 1/0.71 \pm 0.02$ , somewhat different from our fitted value  $1/\psi = 1/0.21$ . However, we have set the exchange factor of Ref. [5] equal to a constant,  $T_g$ . This quantity may, however, possess a  $t_w$  and  $T$  dependence over the temperature region of our measurements [16].

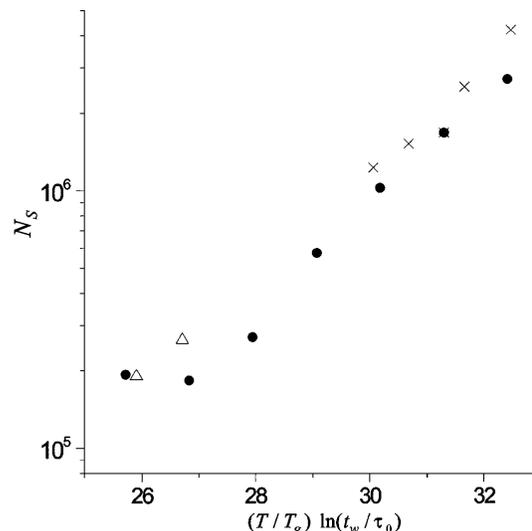


FIG. 4. A plot of  $N_s$  on a log scale vs  $(T/T_g) \ln(t_w/\tau_0)$  for Cu:Mn 6 at.% (solid circles and crosses) and the thiospinel (open triangles). The solid circles are for variable  $T$  and fixed  $t_w = 8$  min, and the crosses for variable  $t_w$  and fixed  $T = 0.89T_g = 28$  K for Cu:Mn 6 at.%. The open triangles are for variable  $t_w$  and fixed  $T = 0.72T_g = 12$  K for the thiospinel.

In order to test the universality of our fit to the power law form for  $N_s$  as given by Eq. (6), we plot in Fig. 4,  $N_s$  on a log scale against  $(T/T_g) \ln(t_w/\tau_0)$ , for both Cu:Mn 6 at.% and the thiospinel. It is seen that all of our data falls roughly along a universal line. It should be noted that two data points for the thiospinel, taken at low temperatures ( $0.36$  and  $0.48 \times T/T_g$ , respectively), would not fall on the data line in Fig. 4, and are not exhibited. Both points correspond to data taken by a different method from those exhibited in Fig. 4, in that the peak in  $S(t)$  could not be observed, and an estimate of the effective age of the curves was obtained by a scaling procedure (see Fig. 5 of Ref. [2]). The precise fitting could be in error, but we cannot rule out that the “line” in Fig. 4 may cross over to a weaker slope at low temperatures.

The values of  $N_s$  extracted by us from Eq. (4) are in the range of  $10^5$ , varying with  $t_w$  and  $T$  according to Eqs. (6) and (7). This suggests that there is a mesoscopic number of spins participating as a unit in the aging process, as argued initially in Ref. [2]. Lederman *et al.* and Hammann *et al.* [17] suggest a large highly correlated spin system is involved in the dynamical processes of aging and irreversibility.

The nature of  $G(x, t)$  in Eq. (1) lends itself to such an interpretation. The spin realizations  $\sigma_i$  and  $\tau_i$  have zero overlap. Yet their correlation is finite so long as  $x < \xi(t, T)$ . These realizations can be interpreted as rotations one from another, consistent with a change in Zeeman energy, relating the theoretical quantity  $G(x, t)$  to  $N_s$ .

The power law dynamics form for  $\xi(t_w, T)$ , exhibited in Eq. (6), and the maximum occupied barrier  $\Delta(t_w, T)$ ,

stipulated by Eq. (3), generate an interesting relationship. Substituting  $t_w$  from Eq. (6) into Eq. (3) leads to

$$\Delta(t_w, T)/k_B T_g = 6[\ln \xi(t_w, T) + 0.44]. \quad (8)$$

This result, supported by our experimental data, was anticipated by Koper and Hilhorst [4] as well as by Rieger [13] and Sibani *et al.* [18]. The form of Eq. (8) was also exhibited qualitatively in an analysis similar to our own by Granberg *et al.* on Cu:Mn 10 at. % [19]. Activated dynamics would yield a power law relationship between  $\Delta(t_w, T)$  and  $\xi(t_w, T)$ .

Equation (8) shows that it would be wrong for a finite time scale to think of a rigid set of barriers extending to infinite height (the so-called “pure states”). Barriers are created as the correlation length increases, with detailed balance obeyed throughout the hierarchy.

In summary, we have interpreted the magnetic field dependence of the response function to generate the *number* of spins,  $N_s(t_w, T)$ , locked together in the barrier quenching (hopping) process. We identify  $[N_s(t_w, T)]^{1/3} \approx \xi(t_w, T)$  with the spin glass correlation length. The fitted expressions are nearly identical for measurements on Cu:Mn 6 at. %, and for the thiospinel. This suggests that power law dynamics [4] may well be universal. Finally, our fitted expressions for  $\xi(t_w, T)$  were very close to the numerical simulations of Refs. [7], [8], and [15].

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