Spin Glass Dynamics under a Change in Magnetic Field

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A barrier model is developed for spin glass dynamics. An ultrametric arrangement for spin glass states at magnetization M, with barrier heights increasing linearly with Hamming distance, is used to represent aging. Upon a change in magnetic field, with an associated change in Zeeman energy E_z , states with barrier heights $\Delta < E_z$ rapidly transition to the new ground state magnetization. Calculation of the time rate of change of the magnetization for finite hierarchies yields results in agreement with experiment. Forms for the most probable values of the Parisi physical order parameter for infinite hierarchies, P(q), are extracted from experiment at representative temperatures. [S0031-9007(96)01764-4]

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Aging is one of the characteristic features of spin glasses [1]. The model of barrier heights [2] monotonically increasing with metastable state Hamming distance on a hierarchical tree can account [1] for the aging phenomenon (but see Ref. [3]). What remains is to relate this structure to spin glass dynamics, e.g., the time dependence of the thermoremanent magnetization, $M_{\text{TRM}}(t, t_w)$ or of the zero field magnetization, $M_{\text{ZFC}}(t, t_w)$, where t is the time of measurement after waiting a time t_w below the spin glass transition temperature T_g .

Steps in this direction have been taken by Chu *et al.* [4] and Kenning *et al.* [4], and within a trap model by Bouchaud and Dean [5] and Vincent *et al.* [5].

The pure states introduced by Parisi [6] are thought to originate from metastable states when the largest barrier diverges as the temperature is lowered [1]. These infinite barriers encompass metastable states separated by a self-similar distribution of barrier heights regardless of temperature. When the spin glass is cooled through T_g in a magnetic field (which could be zero) and held at the measuring temperature T_m for t_w , the system ages by overcoming barriers and populating additional metastable states. The *ultrametric* symmetry of states [7] ensures that this diffusion is effectively one dimensional, with barriers of increasing height being surmounted as time progresses.

The model for extraction of spin glass dynamics of which we shall make use [4] relies on the assumption that the states separated by barrier heights $0 \le \Delta \le E_z$ empty instantaneously into the new ground states upon a corresponding change in magnetic field H. This assumption can be related to the work of Narayan and Fisher [8]. Magnetic field cycling experiments [4] have shown that the exchange of occupations "respects" the barrier heights: the new ground state occupation is bounded by the same barrier heights, $0 \le \Delta \le E_z$, as the original state occupation. Subsequent diffusion within the initial manifold of states takes place over barriers larger than those encoun-

tered in the time t_w , $\Delta(t_w)$, and to the *sink* arising from the change in Zeeman energy overcoming the barriers between phase space states, $\Delta \leq E_z$. This model can account for $M_{\text{TRM}}(t, t_w)$ if the initial manifold is identified with the field cooled states (with magnetization M_{FC}), and the ground state upon $H \rightarrow 0$ is identified with the zero magnetization state. The difficulty lies with the solution of the diffusion equation on the hierarchical tree appropriate to $M = M_{\text{FC}}$ in the presence of a sink at $\Delta \leq E_z$.

The solution to the problem of a random walk in the absence of a sink on a finite *bifurcating* ultrametric space was presented by Ogielski and Stein [9]. We have extended their calculation to include a sink, and solved for the time dependence of $M_{ZFC}(t, t_w)$. In this manner, we are able to compare directly with the classic experiments of the Uppsala group [10].

We consider a hierarchical structure with branching ratio *r*. Starting at the order parameter q_0 at the top of the tree, we assign an increasing value of *q* at each subsequent branch down to the maximum value q_{EA} (the Edwards-Anderson order parameter [11]). The Hamming distance *D* is defined by $D = \frac{1}{2}(q_{\text{EA}} - q)$. Note that in the Ogielski and Stein treatment [9] *D* is given by the number of branches that one must meet before two branches merge. It is more natural for our purposes to define the Hamming distance *D* in terms of *q* because *q* does not necessarily increase linearly with an increasing number of branches. This relationship will not enter into the calculation of $M_{\text{ZFC}}(t, t_w)$ at given magnetic field, but will be required when we compare $M_{\text{ZFC}}(t, t_w)$ at different magnetic fields where it will be introduced through the Parisi physical order parameter P(q) [12].

At $t = -t_w$ (the time of quenching the system below T_g), the states of magnetization of $M_{\rm FC}$ are occupied initially at Hamming distance D = 0, and aging begins. Let the number of states of given magnetization at the site of distance k be given by $N_k(t)$, the barrier height

at D = k be Δ_k , and the probability per unit time of jumping from *any* one state at D = i to *any* of r^{j-1} states at D = j be ϵ_{ij} . Then, $\mathbf{N}(t)$ satisfies the differential equation

$$\frac{\partial \mathbf{N}(t)}{\partial t} = \boldsymbol{\epsilon} \mathbf{N}(t). \tag{1}$$

By this definition, the diffusion matrix ϵ results in an $n \times n$ eigenvalue problem in contrast to that of Ogielski and Stein which would be $2^n \times 2^n$. With n = 80 and r = 2, we would have $2^{80}-10^{23}$ metastable states. An additional benefit of our definition is that we can solve the diffusion problem even for noninteger r.

For convenience we shall choose r = 2 and n = 20. With this choice of n, all of the states occupied for waiting times and measurement times up to 10^4 are available. Note that the matrix ϵ is not symmetric. Because there are 2^{k-1} degenerate states at the Hamming distance k, we have $\epsilon_{ij} = 2^{j-i}\epsilon_{ji}$. For i < j, we have $\epsilon_{ij} = \sum_{i=0}^{j-1} \epsilon_{ji}$ which means the probabilities per unit time to overcome the barrier height Δ_k from below and above are the same. If the hopping is thermally activated, $\epsilon_{ij} = e^{-\Delta_j/T}$ for i < j. We note from Ref. [2] that, for small D, energy barriers grow linearly with Hamming distance, i.e., $\Delta_j = j\Delta$. By solving the $n \times n$ eigenvalue problem with the initial condition $N_0(-t_w) = \mathcal{N}$ where \mathcal{N} is the total number of states of given magnetization and $N_{i\neq 0}(-t_w) = 0$, we obtain $N_i(t < 0)$ during the waiting time t_w .

At t = 0, the system has aged for t_w , and there is a field change δH . With the field change $H \to 0$, we would be calculating $M_{\text{TRM}}(t, t_w)$, while $0 \to H$ would give $M_{\text{ZFC}}(t, t_w)$. We focus on $M_{\text{ZFC}}(t, t_w)$ in order to compare with the experiments of the Uppsala group [10] though the behavior of $M_{\text{TRM}}(t, t_w)$ is the same [and thus will give the same absolute value for S(t) as calculated below].

Upon the field change $0 \rightarrow H$, the rapid increase of magnetization $M_{\rm ZFC}(t=0)$ is caused by the instantaneous transfer of states within the sink of the M = 0manifold to states with comparable barrier heights within the $M = M_{\rm FC}$ manifold. The subsequent increase of $M_{\rm ZFC}(t, t_w)$ with time is associated with the diffusion of states within the M = 0 manifold to the sink, with the concomitant instantaneous transfer to the $M = M_{\rm FC}$ manifold. The Zeeman energy E_z is so small compared to the barrier height $\Delta(t_w)$ explored during the experimental waiting time t_w that for purposes of calculation only the D = 0 site is included in the sink. In case the field dependence of the magnetization were to be important [see below where the nonlinear field dependence of $M_{\rm ZFC}(t, t_w)$ generates Parisi's P(q), one must cut off the region of phase space up to $\Delta \simeq E_z$. This defines a new diffusion matrix modified to $\epsilon_{0i} = 0$; i.e., there is no transition from the D = 0 site to D = i sites. The new initial conditions at t = 0 would be $N_0(0^+) = 0$ and $N_{i\neq 0}(0^+) = N_{i\neq 0}(0^-).$

The solution $N_0(t > 0)/\mathcal{N}$ added to the initial decay $N_0(0^-)/\mathcal{N}$ corresponds to the magnetization $M_{\rm ZFC}(t, t_w)/M_{\rm FC}$. We plot our calculation of the quantity used by the Uppsala group [10] $S(t) = dM_{\rm ZFC}(t, t_w)/d \ln t$ versus t with n = 20 in Fig. 1(a).

The comparison with the experiments of the Uppsala group [10], Fig. 1(b), is remarkable. We find that S(t)given by the hierarchical model is peaked at the measurement time $t \approx t_w$. This feature was pointed to by Grandlund et al. [10] as a signature of the droplet model [13] arising when nucleated spin domains grow to reach the equilibrium domain size. 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 $t = t_w$. While this does not distinguish between the two models, it at least demonstrates that *each* is capable of explaining the structure of S(t). In addition, the current calculation generates a quantitative fit, while the droplet model so far has only suggested qualitative features. It should be noted that Hoffmann and Sibani [14] have generated qualitatively similar properties for S(t) using a parametrized master equation on a set of states which have the topology of a tree.

Reference [1] extended temperature cycling methods [15] to extract the temperature dependence of a particular barrier of height Δ . Hammann *et al.* [1] were able to fit $-d\Delta/dT_r$ to both a power law and an exponential in Δ , where T_r is the reduced temperature T/T_g . We have used



FIG. 1. (a) Using the finite bifurcating ultrametric tree for n = 20, we plot S(t) vs t for a three different waiting times. (b) Experimental values for S(t) from Ref. [10].

the latter [Eq. (5) of Ref. [1]],

$$-\frac{d\Delta}{dT_r} = \alpha \, \exp(\beta \Delta), \qquad (2)$$

with their values of $\alpha = 0.5$ and $\beta = 0.2$ to fit to the temperature cycling results of Granberg *et al.* [15] using the hierarchical structure of finite extent (n = 20) outlined above. The results are exhibited in Fig. 2. Again, the agreement is remarkable, now especially so because of the use of the temperature dependence of the barrier heights as extracted in Ref. [1] from a completely different experiment. The need to account for the temperature dependence of the barrier heights, and the use of values for the parameters α and β from a different experiment, strongly suggests that the results of Ref. [1] are universal.

We are also able to formulate a continuum expression for $M_{ZFC}(t, t_w)/M_{FC}$ applicable to physical systems in terms of the most probable value of the Parisi physical order parameter P(q), the probability to find two *pure states* with an overlap q. Our model, using the most probable order parameter, contrasts with the use of free energy fluctuations by Bouchaud and Dean [5]. Our observations [1] demonstrate that a large number of metastable states separated by *finite* barriers form an ultrametric space within a pure state. In our treatment of the finite system, the assumption of linear increase in Δ as a function of D, defined by the number of branches



FIG. 2. (a) Plots of S(t) vs *t* for temperature cycling changes $\Delta T(K) = 0, 0.15, 0.3, 0.45$, and 0.6 according to the protocol of Ref. [13]. (b) Experimental values for S(t) for the same $\Delta T(K)$ from Ref. [13].

one must meet before the two sites merge, automatically connotes a linear dependence of Δ (or *D*) on the number of branches occurred. In real systems, q_k need not linearly increase with *k*. Let $P_1(D)$ be the most probable probability density for branching at Hamming distance *D*. Then the most probable value of the Parisi physical order parameter P(D) [6], the probability to find two states with overlap $q = q_{EA} - 2D$, is related to $P_1(D)$ by

$$P(D) = P_1(D)r^{\int_0^D P_1(D) \, dD},\tag{3}$$

where r is the average branching ratio. A similar conclusion has been reached by Sibani and Hertz [16], and by Weissman *et al.* [17].

For an ultrametric tree with r branches at each vertex, in a manner entirely analogous to the finite structure treated previously, the continuum approximation defined above generates

$$r \int_{0}^{D(E_{z})} P_{1}(D) dD = \frac{M_{ZFC}(t, t_{w})}{M_{FC}} w(t_{w}).$$
(4)

The factor $w(t_w)$ is the number of explored states during t_w . The denominator $M_{\rm FC}$ is used to account for the magnetization of each occupied state. It is seen that the derivative of the left-hand side of Eq. (4) with respect to the Hamming distance $D(E_z)$ gives P(D) as defined by Eq. (3). By virtue of the sum rule [10] relating $M_{\rm TRM}$ and $M_{\rm ZFC}$, this means, from Eq. (4), that P(D) follows immediately from

$$P(D) = \omega(t_w) \frac{d}{dD(E_z)} \left[\frac{M_{\text{TRM}}(t, t_w)}{M_{\text{FC}}} \right].$$
(5)

Calculations of the dependence of the barrier height with D show a linear relationship for small D [2]. Noting $E_z \propto H^2$, we see that P(D), [P(q)], can be obtained, to within a multiplicative constant, by taking the derivative of $M_{\text{TRM}}(t, t_w)/M_{\text{FC}}$ with respect to H^2 , $[-H^2]$, respectively.

We have measured $M_{\text{TRM}}(t, t_w)/M_{\text{FC}}$ over a wide range of temperatures and a very fine mesh of magnetic fields. These measurements, and their derivative with respect to $-H^2$, are plotted in Fig. 3 for three representative temperatures ($T_g = 31.5$ K): (a) $T_r = 0.13$; (b) $T_r = 0.47$; and (c) $T_r = 0.88$.

At low temperatures, Mézard *et al.* [18] find $P(q) \propto (1-q)^{-3/2}$ away from the delta function at $q = q_{\rm EA} \approx 1-aT^2$, where *a* is a constant. Our result [Fig. 3(a')] fits that form exceedingly well. The extracted shape for P(q) remains similar for reduced temperatures up to about $T_r \approx 0.5$.

Near T_g , Mézard *et al.* [18] show that P(q) flattens out to a constant. Our measurements from about $T_r \approx 0.5$ to our highest temperature of measurement ($T_r = 0.88$) exhibit a gradual evolution away from the power law at lower temperatures to a pronounced plateau [Fig. 3(c')] at $T_r = 0.88$, though with a sharp but small area divergence close to q_{EA} . The height and width of the plateau region are observed to increase as T_g is approached from below [compare Figs. 3(b') and 3(c')].

These measurements and analysis are the first to generate explicit forms for the most probable value of the Parisi physical order parameter, P(q). They appear to be consistent with the predictions of mean field theory, and represent a quantitative approach to examination of the experimental consequences of the hierarchical model for spin glass dynamics. Comparison with the Monte Carlo calculations of Young [19] displays consistency between our data and numerical methods.

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FIG. 3. Plots of $M_{\text{TRM}}(t, t_w)/M_{\text{FC}}$ vs $-H^2$ for Cu: Mn 6 at. % $(T_g = 31.5 \text{ K})$ at t = 10 sec and $t_w = 5$ min for (a) $T_r = 0.13$; (b) $T_r = 0.47$; and (c) $T_r = 0.88$. The solid lines are *best fits* to the data, the derivative of which with respect to $-H^2$ is plotted to the right, giving P(q) from Eq. (5) in the text, with $D = \frac{1}{2}(q_{\text{EA}} - q)$.

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- M. Lederman, R. Orbach, J. Hammann, M. Ocio, and E. Vincent, Phys. Rev. B 44, 7403 (1991); J. Hammann, M. Ocio, R. Orbach, and E. Vincent, Physica (Amsterdam) 185A, 278 (1992).
- [2] N. Nemoto, J. Phys. A 21, L287 (1988); D. Vertechi and M. A. Virasoro, J. Phys. (Paris) 50, 2325 (1989).
- [3] J.-P. Bouchaud [J. Phys. I (France) **2**, 1705 (1992)] has shown that it would be possible to have a distribution of barriers up to infinite values without having aging. If one instead introduces the characteristic time of crossing barriers, the condition for aging is then that there is no finite mean value for these times.
- [4] D. Chu, G.G. Kenning, and R. Orbach, Philos. Mag. B
 71, 479 (1995); G.G. Kenning, Y.G. Joh, D. Chu, and R. Orbach, Phys. Rev. B 52, 3479 (1995).
- [5] J.-P. Bouchaud and D.S. Dean, J. Phys. I (France) **5**, 265 (1995), and E. Vincent, J.-P. Bouchaud, D.S. Dean, and J. Hammann, Phys. Rev. B **52**, 1050 (1995) have shown that the trap model with hierarchical steps exhibits the essential behavior of $M_{\text{TRM}}(t, t_w)$, including $\frac{t}{t_w}$ scaling.
- [6] G. Parisi, Phys. Lett. **73A**, 203 (1979); Phys. Rev. Lett. **43**, 1574 (1979); J. Phys. A **13**, L115 (1980).
- [7] M. Mézard, G. Parisi, N. Sourlas, G. Toulouse, and M. A. Virasoro, J. Phys. (Paris) 45, 843 (1984).
- [8] O. Narayan and D.S. Fisher, Phys. Rev. B 49, 9469 (1994).
- [9] A. T. Ogielski and D. L. Stein, Phys. Rev. Lett. 55, 1634 (1985).
- [10] L. Granlund, P. Svedlindh, P. Granberg, P. Nordblad, and L. Lundgren, J. Appl. Phys. 64, 5616 (1988).
- [11] S.F. Edwards and P.W. Anderson, J. Phys. F 5, 965 (1975).
- [12] V.S. Dotsenko, Usp. Fiz. Nauk **163**, 1 (1993) [Sov. Phys. Usp. **36**, 455 (1993)], writes " $\dots P(q) \dots$ could be considered as the physical order parameter, and it is in terms of the function P(q) that the spin-glass phase looks essentially different from any other phase."
- [13] D. S. Fisher and D. A. Huse, Phys. Rev. B 38, 373 (1988);
 38, 386 (1988).
- [14] K. H. Hoffmann and P. Sibani, Z. Phys. B 80, 429 (1990).
- [15] Ph. Refregier, E. Vincent, J. Hammann, and M. Ocio, J. Phys. (Paris) 48, 1533 (1987); P. Granberg, L. Sandlund, P. Nordblad, P. Svedlindh, and L. Lundgren, Phys. Rev. B 38, 7097 (1988).
- [16] P. Sibani and J. A. Hertz, J. Phys. A 18, 1255 (1985).
- [17] M. B. Weissman, N. E. Israeloff, and G. B. Alers, J. Magn. Magn. Mater. **114**, 87 (1992).
- [18] M. Mézard, G. Parisi, and M. A. Virasoro, *Spin Glass Theory and Beyond* (World Scientific, Singapore, 1987), pp. 42–43.
- [19] A.P. Young, Phys. Rev. Lett. 51, 1206 (1983).