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Ordering and relaxation in spin glasses

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Ordering and relaxation in spin glasses are discussed in terms of the diffusion of the point representing the system in eigenstate space. On this approach, the spin-glass transition T_g appears as a percolation threshold; stretched exponential relaxation behavior extends from T_g to an upper characteristic temperature T_c. Predictions are consistent with three-dimensional Ising simulations by Ogielski. Experimental data suggest that for real metallic spin glasses $T_c \simeq 1.5T_g$. This approach may be relevant for other broad-spectrum relaxation phenomena.

I. INTRODUCTION

The question of the existence of an ordering temperature for spin glasses has been hotly debated for a long time; it is generally assumed, at least implicitly, that if there is an ordering temperature it can be identified with the ac susceptibility cusp temperature T_g. However, it has been found that a precise definition of the ordering temperature is made difficult by relaxation effects, or in other words, the effective T_g is dependent on the time scale of the measurement. Because of this, the existence of a real transition has remained in doubt.

In this article I will discuss relaxation and ordering in spin glasses and conventional magnetic systems in general terms, concentrating on the Ising case for simplicity, and following along the lines of a brief analysis published elsewhere.[1] The spin-glass transition appears as a percolation threshold in eigenstate space and is preceded at higher temperature by a crossover from paramagnetic behavior to nonparamagnetic stretched exponential relaxation. I will quote extensively from simulation results[2] plus a selection of experimental data which can be interpreted in this way. Finally, and more speculatively, I will propose that this type of description of relaxation in a disordered system may be sufficiently general to be applicable to a wide range of complex systems which show characteristic broad-time-spectrum phenomena: magnetic lag, mechanical creep, $1/f$ noise, and so on.

II. THERMODYNAMICS OF TRANSITIONS

In order to underline the essential difference between a conventional transition and a spin-glass transition, I will describe things on a very elementary level using a slightly unconventional viewpoint of standard thermodynamics.

Suppose we have a Hamiltonian for our system which can be diagonalized to give ν eigenstates of energy E_i, $i=1$ to ν. In addition, we have a very weak time-dependent interaction which can induce transitions between the levels without changing the E_i. This interaction puts the system in contact with a bath at temperature T. The populations of the levels will be given by the Boltzmann factor

$$p(i)=e^{-E_i/k_BT}\Big/\sum_i e^{-E_i/k_BT} .$$

Now if ν is large, we can make a plot of the log of the density of states per unit energy $\ln\rho(E)$ against E. We can identify $\ln\rho(E)$ with the entropy S and E with the internal energy U. We know that $dS/dU=1/T$. This means [see Fig. 1(a)] that once we have the curve $S(U)$ and we know the temperature T, the only eigenstates that contribute to the physical properties of the system at that particular temperature are those with $E_i \simeq U(T)$, where $U(T)$ is determined by the condition that the line with slope $1/T$ is tangent to the curve $S(U)$ for $U=U(T)$. This is because higher-energy states are too high in energy to be populated, and states of lower energy are populated but are so relatively few in number that their contribution to any thermodynamic average is negligible.

One further thermodynamic relation that will be useful is for the specific heat:

$$C=-(T^2\,d^2S/dU^2)^{-1} . \qquad (1)$$

This follows immediately from the standard definitions. We can now describe different sorts of transitions. If $S(U)$ is concave upwards over some range of U, the system will have a first-order transition, going from point A to point B in Fig. 1(b). There need be no discontinuity in $S(U)$ or any of its derivatives, so a first-order transition need not be strictly a "phase" transition, as is well known.

As an example of a second-order transition, let us take an Ising ferromagnet with N spins so $\nu=2^N$. All eigenstates have energy E_i and moment m_i as good quantum numbers. Schematically, $\rho(m)$ is shown as a function of m for dif-

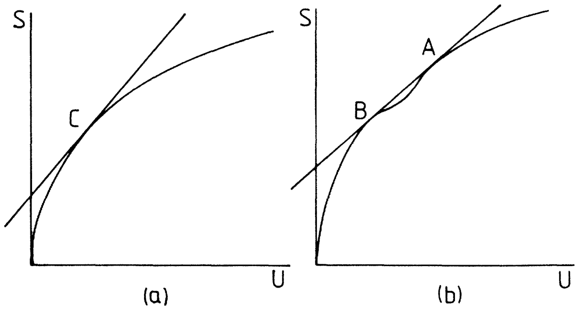

FIG. 1. (a) Schematic plot of entropy S as a function of internal energy U. The straight line is a tangent with slope $1/T$. Only states corresponding to point C, where $dS/dU=1/T$, contribute effectively to the physical properties of the system at temperature T. (b) As (a) for a system with a first-order transition which takes it from A to B at temperature T_c. The slope of the common tangent is $1/T_c$.

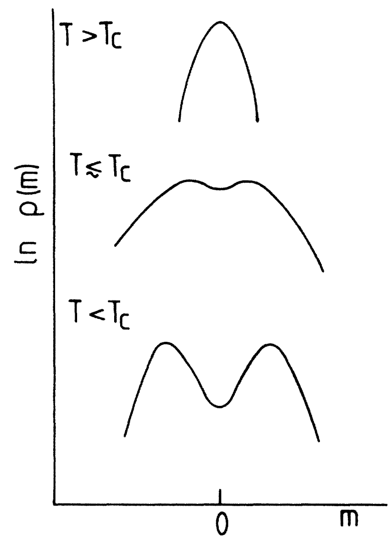

FIG. 2. The density of states as function of magnetization m for an Ising ferromagnet at certain fixed values of U (or T).

ferent E_l (i.e., different temperatures) in Fig. 2. Below T_c, ρ_m has two maxima at $m = \pm m_T$, and the number of states with $m = 0$ is infinitesimally small compared with the number in one or other of the maxima in the thermodynamic limit $N \rightarrow \infty$. Then for $T < T_c$ once the system is in one of the maxima by a random walk among states of the same energy, it will (thermodynamically) never end up in the other maximum because the probability of it finding itself in a bridge state with $m = 0$ is just too small. The system has become nonergodic, i.e., states with the same energy are no longer mutually accessible by the relaxation process. However, below T_c all states of given E having the same sign of m are mutually accessible. At T_c there is an incident on the specific-heat curve, as above T_c the system follows the curve $S = \ln\rho$ $(\nu, m = 0)$ and below T_c the curve $S = \ln\rho$ $(\nu, m = m_T)$. d^2S/dU^2 or d^3S/dU^3 will be discontinuous, leading to a step or change of slope in $C(T)$, from Eq. (1).

We can also represent the eigenstates of any N-spin Ising system by the vertices of an N-dimensional hypercube.[2] Suppose we relax by single spin flips; then the system will diffuse from vertex to near-neighbor vertex. For the ferromagnet, the states of different energies are so arranged on the hypercube that they are either all mutually accessible $(E > U_{T_c})$ or split into two families, each internally mutually accessible $(E < U_{T_c})$.

III. ISING SPIN GLASS

Now consider an Ising spin glass. Because of the random nature of the interactions between the spins, the eigenstates of each given energy E will be distributed in a much more irregular manner on the hypercube than in the case of the ferromagnet. At low temperature there will be a thermodynamically significant number $(\sim e^{\alpha N})$ of alternative ground states,[3] isolated states on the hypercube surrounded by states of higher energies (these ground states have a distribution of energies).[3]

Suppose we start at high temperature. The entropy is high, so the hypercube is densly covered in available states which will all be mutually accessible by relaxation. The system is paramagnetic. As the temperature is lowered, the entropy drops, and the available states thin out. At some temperature there will be a crossover to a situation where the available states form a ramified structure resembling a fractal. The system will percolate among these available states; they will all be mutually accessible on a finite time scale. The system is still ergodic, but the way it relaxes has changed qualitatively. As I have pointed out,[1] because of the ramified nature of the available diffusion path, long-time relaxation will be of the stretched exponential form $\exp(-t^{\beta})$ with $\beta < 1$ in this range of temperature. Alternatively, for an experiment done on a short-time scale the system will be effectively trapped in one part of phase space in this temperature range. As temperature is lowered further at a well defined temperature, the density of available states on the hypercube will attain a value corresponding to the percolation threshold. At this temperature the relaxation exponent β should be equal to $\frac{1}{3}$ (Ref. 1), from the known properties of the percolation threshold fractal in infinite dimension space.[4] Below this temperature the eigenstate space will split up into nonpercolating clusters, i.e., the system will be nonergodic as states on different clusters will not be mutually accessible. We thus have a strictly defined temperature at which ergodicity breakdown sets in. At lower temperatures still, clusters will break up further into smaller noncommunicating clusters.

Because of the irregular structure of the eigenstate distribution, there is no reason to expect an accident in the thermodynamic specific-heat curve at the ergodicity breakdown temperature. The number of available states will evolve smoothly as a function of temperature through this temperature, so d^2S/dU^2 will evolve smoothly. The contrast with the Ising ferromagnet case outlined above appears to be related to the absence of a conventional ordering parameter in the spin glass. There may be practical problems in some systems to measure the specific heat slowly enough to reach the true thermodynamic value.

The difference between the standard second-order transition and the spin-glass transition is that in the former, the system is either ergodic $(T > T_c)$ or nonergodic $(T < T_c)$ on any time scale, while for the latter strict ergodicity breakdown only occurs at T_g, but apparent ergodicity breakdown starts at T_c on short-time scales.

IV. COMPARISON WITH SIMULATION RESULTS

By far the most information available on the $d = 3$ near-neighbor Ising spin glass is from large-scale simulations by Ogielski.[2] The long-time relaxation of the correlation function $q(t) = \langle S(t)S(0)\rangle$ shows qualitative changes of regime. Above a characteristic temperature $T = 4.5$ J, relaxation is exponential $(\beta = 1)$; from $T = 4.5$ J down to $T = 1.2$ J, relaxation is stretched exponential $\exp[-(t/\tau)^{\beta}]$ with τ tending to ∞ and β to $\frac{1}{3}$ at $T = 1.2$ J, which Ogielski identifies as the glass temperature. The scenario for spin-glass ordering which I have outlined above is consistent with these results; we can associate the upper characteristic temperature with the "dense to nondense" crossover, and the lower spin-glass ordering temperature with the percolation threshold. The fact that in the simulation the exponent β tends to $\frac{1}{3}$ at the ordering temperature is particularly en-

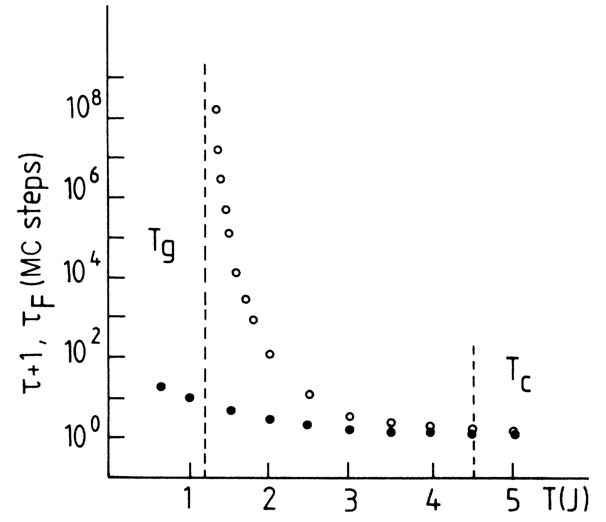

FIG. 3. The relaxation times $(\tau+1)$, ○; τ_F, ● plotted against temperature from simulation results on a (three-dimensional, $\pm J$) Ising spin glass, Ogielski (Refs. 2 and 5). τ is defined from $q(t)=t^{-x}\exp[-(t/\tau)^{\beta}]$ and $\tau_F=1/[1-q(1)]$. $(\tau+1)$ is plotted, rather than τ for consistency of definition in the high-temperature limit.

couraging.

In addition to the long-term relaxation behavior, it is possible to identify two other relaxation regimes. The first is on very short-time scales, one Monte Carlo step per spin. From the results for q (Ref. 1), which Ogielski does not discuss explicitly, we can directly obtain the rate at which individual spins are flipping at any given temperature. This characteristic time given by the inverse flip rate $\tau_f=1/[1-q(1)]$ can be very much faster than the relaxation time τ, as each given spin can flip backwards and forwards many times while its overall time-average correlation on an intermediate time scale remains almost constant. I have plotted (Fig. 3) τ_f and τ both taken from Ogielski's results.[5] The important point is that τ_f becomes essentially equal to τ at the upper characteristic temperature T_c, which one could expect as above this temperature relaxation is exponential, so there is only one characteristic time scale. τ_F varies smoothly through T_g. I will compare with the experiment in Sec. V. Secondly, on an intermediate-time scale or below T_g [and always for $q(t)\gtrsim 0.5$] Ogielski finds $g(t)\sim t^{-x}$ with $x<0.5$. On the diffusion approach this regime would correspond to diffusion within a nonpercolating cluster for $T<T_g$, or within an analogous short-range structure for $T_c>T>T_g$. This corresponds to the time scale within which the system is diffusing among states "within one well." Because of the wide range of distance scales needed to describe the nonpercolating cluster, it is obvious that the relaxation will be characterized by a wide range of relaxation times, but it is not yet clear to me why the result should be algebraic relaxation.

V. COMPARISON WITH EXPERIMENT

A number of experiments have shown that short-time-scale relaxation properties of metallic spin glasses are quite different from those of standard ferromagnets. Ogielski's simulation results show that for the three-dimensional Ising spin glass, relaxation is nonexponential up to a limiting temperature T_c which is about $3.75T_g$. The arguments I have given suggest that the existence of T_c should be a general property; however, one can expect the ratio of T_c/T_g to be system dependent. I will now show that there is experimental evidence in metallic spin glasses that a temperature T_c can be defined with T_c/T_g about 1.5.

It is difficult to measure $q(t)$ directly at different temperatures and to pin down a temperature at which relaxation becomes strictly exponential. Neutron-spin-echo (NSE) measurements get close to this,[6] but existing data well above T_g are rather scanty. The results, as they stand, show that relaxation is definitely nonexponential for $T\lesssim 1.3T_g$.[6] As an alternative, we can use the technique suggested by Fig. 3. As I have noted[7] the temperature-dependent part of the resistivity of a spin glass is due to the inelastic local-moment-conduction electron-spin-flip scattering, and so can be used to estimate the local-moment spin-flip rate (defined in just the same way as τ_F^{-1} in Sec. IV) in absolute terms. This can be compared with experimental values of the local-moment correlation times in the same alloy, when these are available. Figures 4(a) and 4(b) are comparisons

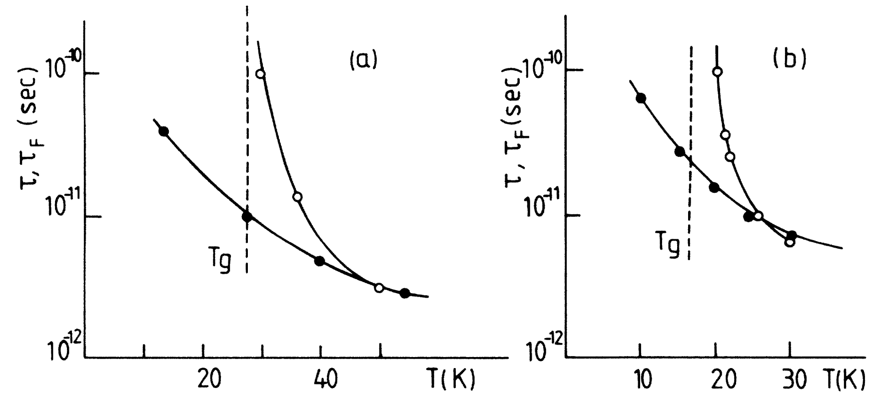

FIG. 4. (a) Estimates of the local-spin-correlation time τ, ○ from NSE (Ref. 6) and of the spin-flip scattering time (Ref. 7) τ_F, ● for Cu-5 at.% Mn. (b) Estimates of the local-spin-correlation time τ, ○ from Mössbauer linewidth (Ref. 8) and of the spin-flip scattering time (Ref. 7) τ_F, ● for Au-3 at.% Fe.

of this sort. In the first case, τ is estimated by fitting with a stretched exponential to the NSE data for *Cu*Mn 5 at.%.[6] τ_F is estimated from data in Ref. 7. In the second case, for *Au*Fe 3-at.% effective τ values from Mössbauer linewidth data[8] are compared with τ_F values from resistivity data.[7] In both cases, the τ extracted from the data is an "effective τ" obtained by fitting experimental results to a model of relaxation which is incomplete. Also, the τ_F values have a systematic uncertitude because a free-electron model was used to pass from $\rho(T)$ to τ_F. Nevertheless, even taking the figures with a pinch of salt, there is a clear resemblance to Fig. 3, with $\tau_F \simeq \tau$ for T above about $1.5T_g$. This is evidence that for T above this temperature there is only a single characteristic relaxation time for the spin glass, or, in other words, $T_c \simeq 1.5T_g$ in real metallic spin glasses.

This result throws some light on ESR measurements in spin glasses. It is well established that for $T << T_g$ there is a resonance line shift which is due to Dzyaloshinsky-Moriya interaction anisotropy.[9] This anisotropy is nonzero because the system of spins is blocked "within one well" at low temperatures. However, when T is increased, the resonance shift only tends to zero at around $1.5T_g$.[10–12] Following the arguments given above, for temperatures up to T_c, the system is blocked "within one well" on short-time scales (i.e., it has not had time to percolate completely). The ESR measurement is on a short-time scale of about 10^{-10} sec; we would then expect a quasistatic resonance field shift up to almost T_c. The fact that a shift appears near $1.5T_g$ is consistent with the estimate for T_c from the other experiments.

The observed frequency dependence of the effective T_g in susceptibility measurements[13] could clearly be interpreted in terms of the temperature variation of the relaxation.

VI. DISCUSSION

Spin glasses are not only of interest in themselves, but also as test-bed systems in which to develop concepts which one can hope will apply to even tougher disorder problems. Relaxation with a very broad spectrum of characteristic times appears to be a hallmark of disordered systems. Lag in ferromagnets has been analyzed in terms of logarithmic[14] or stretched exponential[15] decay. The susceptibility of concentrated dielectrics shows behavior which is equivalent to a stretched exponential time dependence.[16,17] Creep in polymers and glasses also follows the stretched exponential form (with an exponent $\beta = \frac{1}{3}$) and is accompanied by aging effects which appear to vary logarithmically with time after quench.[18]

It has been suggested a number of times[15–17,19] that a single physical mechanism should be at the base of the explanations for all these phenomena in apparently diverse materials. I have argued above that the successive broad-spectrum relaxation regimes observed in the Ising spin-glass simulations[2] can be understood in terms of general properties of diffusion of the point representing the system in the eigenstate space of the complicated spin-glass Hamiltonian, and that it is possible to interpret results on real spin glasses along the same lines. The physical approach in terms of diffusion in an eigenstate space where the energies of the states have an irregular structure would seem to be applicable, *mutatis mutandis,* to any strongly disordered system and not to be restricted to spin glasses. It remains to be seen whether the similarities between these different phenomena are only superficial, or whether the systematics for the behavior of the Ising spin glass carry over in detail to the other materials. A rapid survey of some of the vast set of existing experimental results seems to be encouraging.

In conclusion, I have discussed relaxation and onset of nonergodicity in spin glasses in terms of diffusion of the system point in eigenstate space. The spin-glass transition appears as a percolation threshold; between T_g and an upper characteristic temperature T_c the long-term relaxation should be stretched exponential with an exponent β going from $\frac{1}{3}$ at T_g to 1 at T_c. These predictions are consistent with results of large-scale simulations on a three-dimensional Ising spin glass.[2] An examination of experimental data on real metallic spin glasses suggests that for these Heisenberg materials $T_c \simeq 1.5T_g$. This approach may be relevant in other disorder problems, where there are broad-spectrum relaxation processes.

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