Critical Line for Strong Irreversibility in Spin-Glass and Ferro–Spin-Glass Alloys

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Torque measurements have been made as a function of temperature on a Cu–2-at.%-Mn and a Au–18-at.%-Fe alloy. At a field-dependent temperature $T^*(H)$ torque effects go to zero indicating the frontier for the low-temperature strong-irreversibility regime. For the *Cu*Mn sample the present experimental $T^*(H)$ curve coincides with the calculated mean-field–model de Almeida–Thouless line. For the *Au*Fe, $T^*(H)$ also follows the behavior predicted for the strong-irreversibility crossover on the mean-field model.

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The study of spin-glasses has been given considerable impetus over the last few years by the careful theoretical analysis of the Sherrington-Kirkpatrick (SK) mean-field model,[1] first for Ising[2,3] and then for Heisenberg[4-6] spin systems. One of the main conclusions which has been drawn is that for a simple mean-field Ising spin-glass in an applied field there is a line of transitions from paramagnetic to replica-symmetry-broken order, the de Almeida–Thouless (AT) line.[2] For reduced field h and temperature t, the critical field-temperature relation is

$$h^2 = [4/(m+2)](1-t)^3, \qquad (1)$$

where m is the spin dimensionality. In a Heisenberg model system this transition is demoted to a crossover from weak to strong irreversibility, and another transition line appears, the Gabay-Toulouse (GT) line[4] where canting and some form of transverse irreversibility set in. By continuity,[2,3] for a system where the interaction distribution is biased ferromagnetically, the zero-field-model phase diagram shows a paramagnetic to ferromagnetic transition at T_c, then a canting transition at T_{GT}, and finally a crossover to strong irreversibility at T_{AT}.

It is hoped that these model phase diagrams bear some relation to real systems. The AT and GT transitions have been sought with care experimentally, but have proved elusive. Detailed studies of the temperature dependence of the magnetization of *Ag*Mn and *Cu*Mn samples in different fields[7,8] have shown features at temperatures T_X such that $(T_g - T_X) \propto H^{2/3}$ as suggested by Eq. (1), but it is difficult to decide unequivocally which feature to identify with the model transition. We have made torque measurements as a function of temperature in two canonical systems, *Cu*(2 at.% Mn) and *Au*(18 at.% Fe). The data show rather well defined field-dependent temperatures $T^*(H)$ at which the irreversibility disappears; we suggest that $T^*(H)$ can be identified with the model de Almeida–Thouless line $T_{AT}(H)$.

The idea behind the present experiment is simple: It is now well established that a field-cooled spin-glass shows strong anisotropy effects at low temperature, leading to torque when the direction of the applied field is turned,[9] whereas in the paramagnetic state we can expect the magnetization to follow strictly the direction of the applied field so that the torque will be zero. The onset of torque can be used as a criterion for the onset of irreversibility or memory effects intrinsic to the spin-glass state.

We cool the sample from high temperature in an applied field H to a temperature T; we then turn the field through a small angle (usually 5°) and observe the torque acting on the sample. In this way we can estimate the temperature $T^*(H)$ at which the torque disappears for each value of H.

We will first discuss the simpler case, *Cu*(2 at.% Mn), which is a standard spin-glass. The T_g of our sample as estimated in a separate magnetization measurement with a field of 500 G is 13.5 ± 0.5 K.[10] Part of a set of experimental data is shown in Fig. 1. Our torque results show an initial torque (1 sec after the field is turned) which for fixed values of H decreased with increasing temperature as shown in Fig. 2. For present purposes we will not discuss the form of the temperature dependence of the torque (although this is certainly an interesting subject) but merely note that for low values of torque, Fig. 2 shows that for each field the torque varies as $T^*(H) - T$. We use this to define a series of transition temperatures $T^*(H)$. We cannot rule out the possibility that if our experimental sensitivity was much higher we would observe a high-temperature tail of weak but nonzero torque at each field. However, even if this was so the curves as they stand allow us to define $T^*(H)$ from the extrapolation to zero of the linear part

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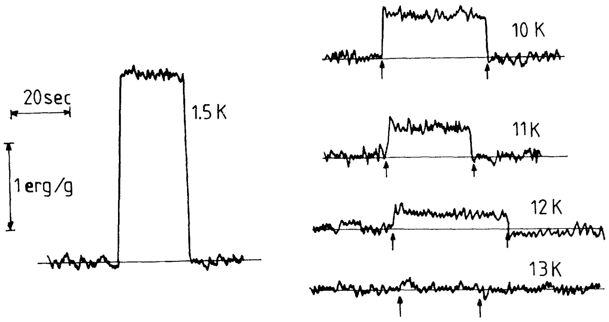

FIG. 1. Experimental torque curves at different temperatures for Cu(2 at.% Mn), as a function of time. The field-angle sequence is 0°-5°-0° with the turning points indicated by arrows. The applied field is 3 kG. The scales are the same at all temperatures.

of the torque versus temperature curve; $T^*(H)$ is where "strong" torque effects begin. With improved sensitivity (which poses no fundamental technical problem) the form of the $T^*(H)$ curve could be established much more precisely.

As we have stated above, $T^*(H)$ defines experimentally the onset of a certain type of memory effect. It is obviously associated with the anisotropy—if there were no ansiotropy terms linking the magnetization to the lattice this effect would not exist. However, we suggest that the torque is a good probe of the general irreversibility (or nonergodic) properties of the system; the presence of torque after the field is turned shows that the system has a memory of its earlier state, or in other words it cannot choose freely to go to the lowest-energy configuration where M would be parallel to H. From this argument, we would expect the torque at low fields to go to zero at T_g; indeed we find that within the experimental error,[11] $T^*(H)$ at 500 G from the torque measurements coincides with T_g estimated independently from magnetization measurements. It therefore seems justified to take $T^*(H)$ as the temperature for the onset of strong irreversibility and to compare the experimental $T^*(H)$ with the model curve $T_{AT}(H)$. We have calculated the model curve[4] for vector spins ($m=3$); this curve has no fitting parameters once T_g is fixed, defining[4,8] $t=T/T_g$ and $h=g\mu_B H/kT_g$. As can be seen in the inset in Fig. 2, the model curve is almost identical to the experimental curve. Despite the fact that for the Heisenberg model system the only true transition is at T_{GT}[5,6] and T_{AT} is just a crossover temperature to more marked irreversibility, we find the strong-irreversibility onset temperature to be rather well defined experimentally and to follow the field dependence

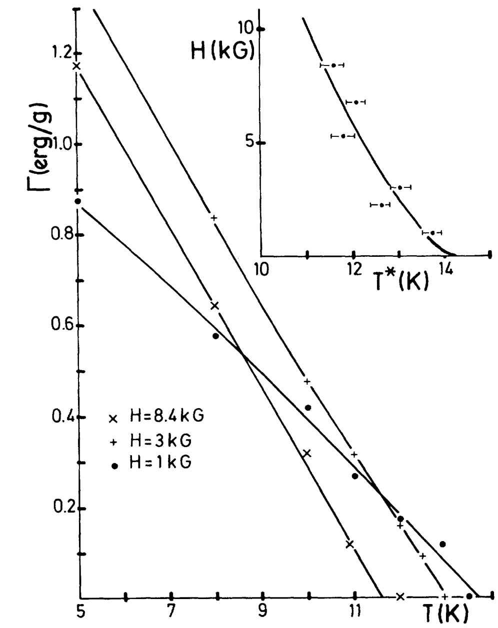

FIG. 2. Cu(2 at.% Mn). Measured torque Γ after a 5° turn of the field H. Inset: The critical field-temperature relation; circles, experimental values of $T^*(H)$; full curve, the de Almeida–Thouless line for $m=3$ (Ref. 4), $H^2=\frac{4}{5}(1-T)^3$ with $T_g=14.25$ K.

expected for it.

Dynamic experiments on CuMn and other spinglasses[12,13] have given $T_{AT}(H)$ curves of the same functional form as the model curve, but with a very different scaling factor for H which appears to depend strongly on the time scale of the measurement. The model curve is approached from below only at high frequencies. Our CuMn torque results, Fig. 1, show a $T^*(H)$ which is rather insensitive to weak relaxation effects over the time scale range 1 to 20 sec but we should point out that we have discussed only the small-turn-angle behavior—for large turn angles we have observed strong torque relaxation effects which accelerate as $T^*(H)$ is approached. We explain this as follows: Experiments which involve large changes in the strength of the field (as in remanence measurements or in Ref. 13) or in its direction drive the system into an energetic metastable state from which it relaxes, at a rate which accelerates as $T^*(H)$ is approached. Small-angle field rotation on the other hand is a gentle perturbation, putting the system into a state only slightly more energetic than the ground state and for which relaxation towards the ground state is slow even very near $T^*(H)$. The fast relaxation observed in certain experiments would then be characteristic of the system evolving from a highly perturbed state while the small-angle torque results are closer to representing observations on the unperturbed spin-glass at fixed H and T. Relaxation effects in other systems may not be the same. Our *Cu*Mn results show that irreversibility on a "long" (~20-sec) time scale persists up to at least the model $T_{AT}(H)$.

We now turn to the more complicated case of *Au*(18 at.% Fe), which shows mixed ferro–spin-glass character. For this concentration, $T_c = 170$ K and Mössbauer data[14,15] have shown that there is canting-transition behavior near 60 K, while magnetic measurements[15–17] indicate slight irreversibility at higher temperatures which becomes rapidly stronger below 15 K. The torque measurements show a strong spin-glass type of torque at 1.5 K. As the temperature is increased the torque drops and, in contrast to *Cu*Mn, relaxation first appears and then accelerates. In the region below 15 K the torque becomes weak and relaxes towards zero with a time constant of seconds, and at slightly higher temperature the torque disappears; once again the temperature for this disappearance is field dependent as shown in Fig. 3. Defining an exact onset temperature $T^*(H)$ is not quite as easy as for *Cu*Mn since the torque-against-temperature plots curve at the higher temperatures. The $T^*(H)$ values in the inset in Fig. 3 have been derived with the assumption that the plots approach the axis parabolically; alternatively we could for instance define $T^*(H)$ as the point where the torque is 10^{-3} of its value at 1.5 K. This would change the absolute values of the temperatures $T^*(H)$ by a fraction of one degree, but not their relative positions since all curves for the different fields are parallel. As for *Cu*Mn we have the caveat that higher-precision measurements could reveal a high-temperature tail, but any effects below the range of our present sensitivity would be extremely weak. Thus whatever criterion we choose, we find that strong irreversibility sets in only below 15 K, and the field dependence of the onset temperature $T^*(H)$ is as given in Fig. 3.

The $T^*(H)$ obtained from these torque data are in good agreement with strong-irreversibility temperatures estimated earlier from the region of rapid increase of hysteresis[15]; however, the torque data give a more clear-cut transition temperature and show the variation of this temperature with the applied field.

For fields below about 200 G the torque behavior changes drastically. For temperatures

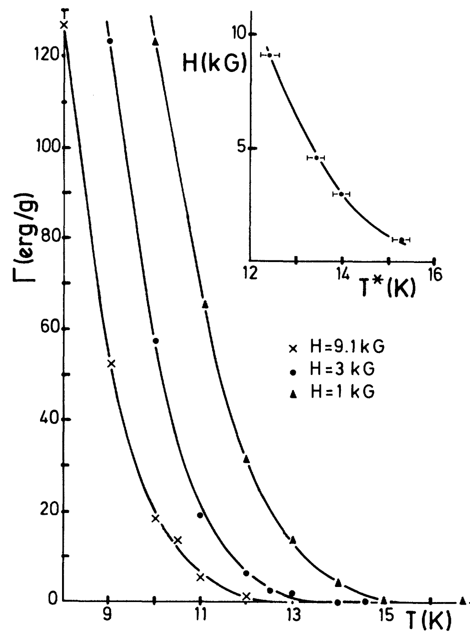

FIG. 3. *Au*(18 at.% Fe). Measured torque Γ after a 5° turn of field H. The saturation low-temperature Γ is about 1000 ergs/g. Inset: The experimental $T^*(H)$ curve.

of 16 K or above where the torque has gone to zero for the higher fields, there is still a strong torque in these lower fields, with visible relaxation effects. This low-field torque drops off gradually as the temperature is increased; we have studied the temperature dependence in detail for an arbitrarily chosen field value of 45 G. Here the low-field torque drops continuously until $T \cong 60$ K, above which it remains constant at a low level up to 90 K (the highest temperature to which we have measured). We ascribe the radical difference between the high-field and the low-field results to the presence of domains and domain walls in the latter situation where the demagnetizing field begins to play a role. From magnetization data[18] it is not possible to identify precisely a technical saturation field for this system at any temperature, but the magnetization tends to approximate saturation above about 200 G for $T > 15$ K. The sample in a high field is monodomain, and in a low field is multidomain.

We can again compare the results with the mean-field model. First we will discuss the high-field data. As in *Cu*Mn and for the same reasons we identify $T^*(H)$ as a transition to strong irreversibility, and so we compare it to T_{AT}. We can see that this transition temperature is very much distinct from the canting temperature of about 60 K.[14,15] As far as the field dependence is concerned, no explicit expression has been published for the field dependence of T_{AT} in the mixed ferro–spin-glass regime, but an estimate can be given quite easily in the Ising version of the model.[2] With both ferromagnetic interactions and an applied field, $|(J_0 m+H)/kT|$ should be substituted for $J_0 m/kT$ in the self-consistent set of expressions for m, q, and $(kT/J)^2$ given by de Almeida and Thouless.[2,19] Putting on an applied field is essentially equivalent to an increase in J_0, so that T_{AT} should decrease linearly with H to first order. Substituting experimental values of T_c and T_{AT} into the model, we can estimate as an order of magnitude for our particular alloy

$$dT_{AT}/dH \cong -0.15 \text{ K/kG}.$$

This rough model value is in very reasonable agreement with the experimentally observed variation of $T^*(H)$, Fig. 3.

Finally, the low-field torque varies in a very similar way to the degree of canting as measured by the Mössbauer effect[14]; both show a "transition" in the region of 60 to 70 K, and for both there is a weak residual effect at higher temperatures. From this purely phenomenological parallel we can suggest heuristically that the local transverse irreversibility that should accompany the onset of canting at T_{GT}[5,6] actually manifests itself as a hindering of domain-wall movements in multidomain samples.

In conclusion, we have presented torque measurements on a simple spin-glass *Cu*(2 at.% Mn) and a mixed ferro–spin-glass *Au*(18 at.% Fe). We measure the temperature $T^*(H)$ at which the torque observed after turning the applied field goes to zero. We find that $T^*(H)$ is well defined for both alloys; $T^*(H)$ indicates the onset of strong irreversibility and in both cases follows closely the behavior expected for the de Almeida–Thouless temperature T_{AT} on the mean-field model for this type of system. For *Au*Fe under weak fields, the behavior of the torque can be linked with the canting rather than with the strong irreversibility.

Other problems such as the relaxation or the anisotropy properties per se which we have practically ignored in the present discussion are also important and will be more fully treated elsewhere. In any case, it is clear that torque measurements give new and essential information on the complexities of spin-glass behavior.

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