

Critical line for irreversibility in spin glasses

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ABSTRACT

An experimental study of irreversible phenomena in amorphous FeMn is reported. Irreversibility is characterized by the existence of both non-equilibrium susceptibility and magnetic viscosity. Both disappear above a critical line $T_c(H)$ which is identified as the de Almeida-Thouless line for spin glasses. $T_c(H)$ is, within experimental resolution, independent of time-scale in the range 30–700 s and has the form $\tau \propto h^\phi$, where τ and h are the reduced temperature and the reduced field respectively, $\phi = 0.75 \pm 0.15$ and the field prefactor is ~ 15 . These results are discussed within the framework of recent mean-field models for spin glasses.

§ 1. INTRODUCTION

The two main characteristic properties of spin glasses are the sharp cusp in the zero-field a.c. susceptibility at the freezing temperature T_f and the onset of irreversible phenomena below this temperature. The observation of the cusp (Cannella and Mydosh 1972) motivated Edwards and Anderson (1975, hereafter referred to as EA) to develop a theory which considers the spin-glass transition as a collective process. Models of the EA type, such as the Sherrington-Kirkpatrick (1975, hereafter referred to as SK) mean-field solution, indeed produce a cusp in the susceptibility. In the EA theory the spin-glass order parameter q_{EA} vanishes at T_f in zero magnetic field only. In the presence of a field, q_{EA} has a non-zero value at any finite temperature and a spin-glass transition cannot occur. Some experimental support for this 'polarization' of the spin-glass order parameter has been found in the strong smearing of the cusp in a rather small magnetic field. This has given rise to a general belief that the spin-glass transition is destroyed in the presence of a magnetic field. That this is *not* the case was first shown by de Almeida and Thouless (1978, hereafter referred to as AT) who calculated, in the framework of the SK model for Ising spin glasses, a line $T_c(H)$ which has been shown to be a line of spin-glass-paramagnetic transitions in the field-temperature (H - T) phase diagram (Parisi 1979, Gabay and Toulouse 1981, Sompolinsky 1981). The equation of this line in the vicinity of $T_f = T_c(0)$ is given by

$$\tau = Ah^{2/3}, \quad (1)$$

where $\tau \equiv 1 - T_c(H)/T_f$ and $h \equiv g\mu_B H/k_B T_f$ are the reduced temperature and the reduced field respectively, g being the gyromagnetic ratio, μ_B the Bohr magneton and k_B is the Boltzmann constant. The field coefficient A is of the order of 1.

Sompolinsky (1981), using a dynamic approach, has explicitly incorporated the slow irreversible relaxation processes as a central feature of the spin-glass

transition. The theory describes the spin-glass phase by order parameters which relax with a broad distribution of relaxation times. In the finite-time limit the non-equilibrium susceptibility obeys, in zero field, the Fischer relation

$$\chi_{ne} = (C/T)(1 - q_{EA}), \quad (2)$$

where C is the Curie constant. Another order parameter is the irreversible response Δ defined by the difference between χ_{ne} and the true equilibrium susceptibility χ_0 .

$$\chi_e = \chi_{ne} + (C\Delta/T). \quad (3)$$

The use of Δ as an order parameter is particularly useful if a finite field is present; q_{EA} is then non-zero, even at high temperatures, but Δ appears only below a field-dependent critical temperature $T_c(H)$. The loci of these critical temperatures define a line in the H - T phase diagram which coincides with the AT line (eqn. (1)).

The AT calculations for Ising spin glasses were extended to m -component spins by Gabay and Toulouse (1981, hereafter referred to as GT). For Heisenberg spin glasses the equation of the transition line, the GT line, is given by

$$\tau = A'h^2, \quad (4)$$

where $A' \simeq 7/20$. For this model system the transition at $T_c(H)$ is signalled by freezing of the spin components which are transverse with respect to the applied field. This in turn gives rise to an irreversible response in both the transverse and the longitudinal susceptibilities, the latter being relatively weak in the vicinity of T_c . The AT line defined by eqn. (1) is, in this model system, a cross-over line from weak to strong (longitudinal) irreversibility.

Thus, from the theoretical point of view, the existence of a spin-glass transition line in the H - T plane is well established and its physical meaning is well understood. The purpose in the present article is to examine the results of a systematic experimental study of the H - T phase diagram. We focus on the amorphous FeMn spin-glass system (Yeshurun, Ketelsen and Salamon 1982, Yeshurun and Sompolinsky 1982), but also discuss results for crystalline FeCr (Palumbo, Parks and Yeshurun 1982). The experimental criterion which is used here to identify the spin-glass transition is the disappearance of irreversible processes. We describe two different kinds of experiment based on this criterion:

- (1) branch point measurements for identifying the critical temperature $T_c(H)$ above which the two branches of the susceptibility, χ_e and χ_{ne} , coincide at a given field, and
- (2) magnetic viscosity measurements for identifying the highest field $H_c(T)$ below which relaxation phenomena are still observed at a given temperature.

In the following sections we describe in detail the two experimental procedures and compare the results. We conclude that both experiments yield one well-defined transition line, independent of the experimental time-scale (30–700 s). This line of transitions is identified as the AT line (eqn. (1)). Finally, we discuss the results and sketch the present experimental status of the H - T phase diagram for spin glasses.

§ 2. EXPERIMENTAL

This section is devoted mainly to a detailed description of the two experimental procedures, branch point and magnetic relaxation measurements. We exhibit results for amorphous $(\text{Fe}_{0.64}\text{Mn}_{0.36})_{75}\text{P}_{16}\text{B}_8\text{Al}_3$ which have been previously reported (Yeshurun and Sompolinsky 1982, Yeshurun *et al.* 1982). Our main interest here is to compare the results obtained using the two procedures.

2.1. Sample preparation

Ribbons of the amorphous samples were prepared by centrifugal spin quenching (Chen and Miller 1976). Small chips ($5 \times 1 \times 0.5$ mm) were cut from the ribbons, stacked in parallel and introduced into a vibrating sample magnetometer with the longest axis parallel to the applied magnetic field.

2.2. Susceptibility branch point measurements

Susceptibility was measured using a four-step procedure. First the sample was zero-field cooled (ZFC) to 4.2 K, a field H was applied and the ZFC magnetization curve M_1 was measured up to ~ 80 K. Then, without changing the field, the field-cooled (FC) magnetization M_2 was recorded down to 4.2 K. At this temperature the field was increased by ΔH and magnetization curves were recorded while sweeping the temperature up (M_3) and down (M_4). This procedure is demonstrated in fig. 1 for $H = 600$ Oe and $\Delta H = 100$ Oe.

From eqn. (3) the order parameter Δ is given by

$$\Delta = (T/C)(\chi_e - \chi_{ne}). \quad (5)$$

For a cooling field H and a step ΔH ,

$$\chi_e = (M_4 - M_2)/\Delta H, \quad (6a)$$

$$\chi_{ne} = (M_3 - M_2)/\Delta H. \quad (6b)$$

The order parameter Δ is therefore proportional to $\Delta M = M_4 - M_3$. The inset in fig. 1 exhibits the temperature dependence of ΔM . As can be seen from fig. 1, ΔM (and therefore Δ , too) increases continuously from zero. We show this by the solid line in the inset, which is the result of a non-linear least-squares fit of the difference,

$$\Delta(H) \propto M_4 - M_3 = A\bar{\tau} + B\bar{\tau}^2, \quad (7)$$

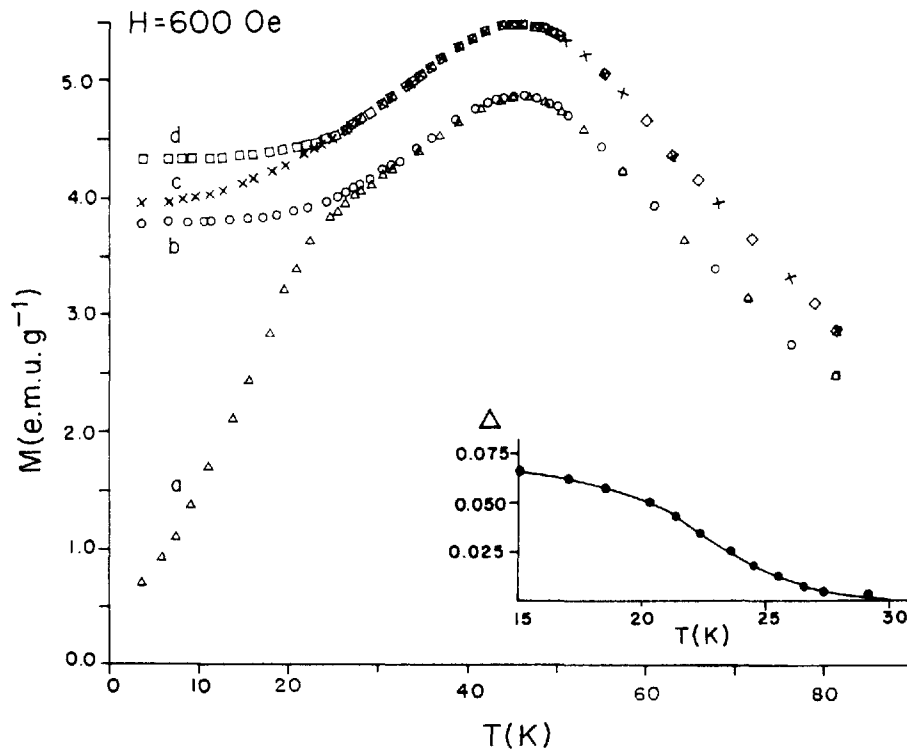
where $\bar{\tau} = 1 - T/T_c(H)$. The branch point $T_c(H)$ is thus one of the fitting parameters in eqn. (7).

The procedure for a zero-field run is a little simpler. For $H = 0$ we take $H = 8$ Oe (the lowest field in this experiment). Now χ_e and χ_{ne} are simply the FC and the ZFC low-field susceptibilities. The critical temperature $T_c(0) = T_f$ is identified as the location of the maximum of χ_{ne} (41.6 K). The order parameter Δ is extracted with the help of eqn. (5), while the EA order parameter is deduced from the Fischer relation, eqn. (2). The temperature dependence of q_{EA} and of Δ for $H = 0$ is shown in fig. 2; the inset shows a log-log plot of these order parameters in the vicinity of T_c . We find for $H = 0$ that

$$q_{EA} \propto [1 - (T/T_f)]^\beta, \quad \beta = 1.25 \pm 0.25, \quad (8a)$$

$$\Delta(H=0) \propto [1 - (T/T_f)]^{\beta'}, \quad \beta' = 2.0 \pm 0.2. \quad (8b)$$

Fig. 1



Magnetization field branches for amorphous $(\text{Fe}_{0.64}\text{Mn}_{0.36})_{75}\text{P}_{16}\text{B}_8\text{Al}_3$. Curve (a): zero-field-cooled magnetization, $H=600$ Oe; curve (b): field-cooled magnetization, $H=600$ Oe; curve (c) magnetization measured in field of $H=700$ Oe after cooling in field of 600 Oe; curve (d) magnetization measured while cooling in field of $H=700$ Oe. The inset shows the temperature dependence of ΔM .

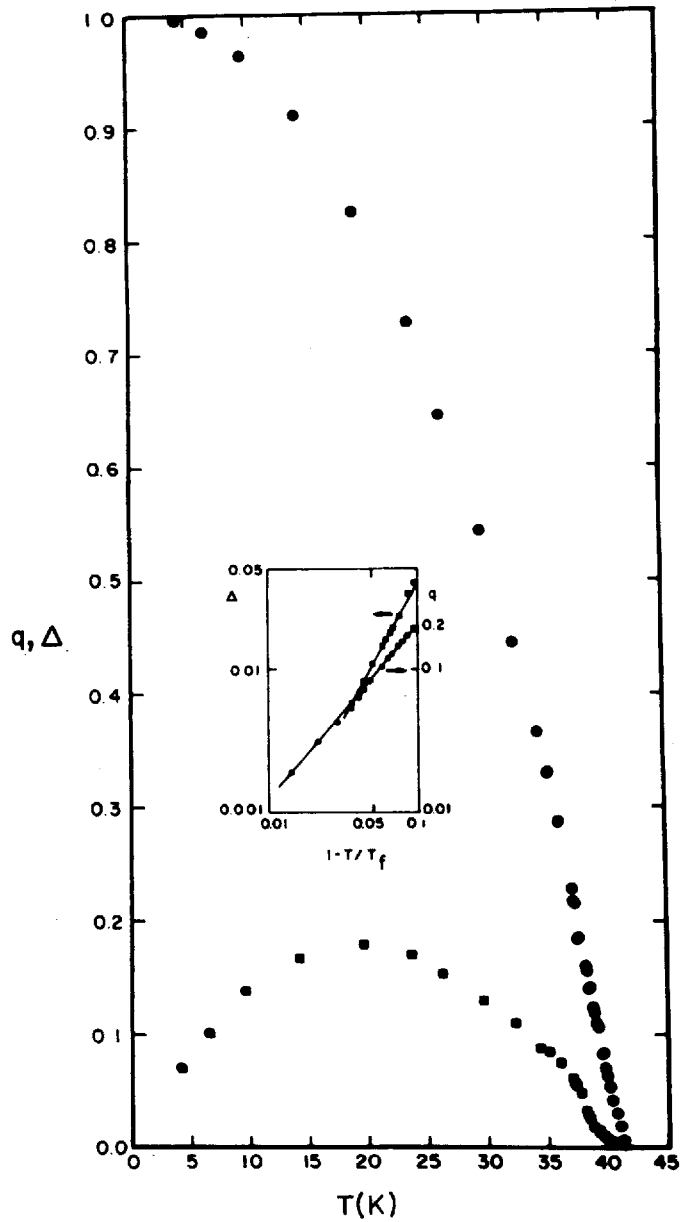
Note that the result (8b) is consistent with eqn. (7) provided $A(H=0)=0$. The exponents β and β' , as well as the trend of $A(H)$ in eqn. (7), are in good agreement with the predictions of the dynamic model (Sompolinsky 1981).

2.3. Magnetic viscosity measurements

Three different procedures were used.

- (1) The sample was cooled in zero field from $T \approx 60$ K $> T_f$ to the desired temperature T . The field was then increased abruptly to a value H , and changes in the magnetization recorded for approximately 12 min.
- (2) The field of step (1) was kept constant until quasi-equilibrium (defined by a constant reading of magnetization over a period of several minutes) was achieved. The field was then turned off and the relaxation recorded for 12 min.

Fig. 2



The spin-glass order parameters $q_{E\Delta}$ and Δ for $H=0$. The inset shows a log-log plot of these parameters in the vicinity of T_c .

- (3) After cooling in a field from $T \simeq 60$ K to $T < T_f$, the field H was turned off and the relaxation of the thermo-remanent magnetization (TRM) from equilibrium state was recorded for approximately 12 min.

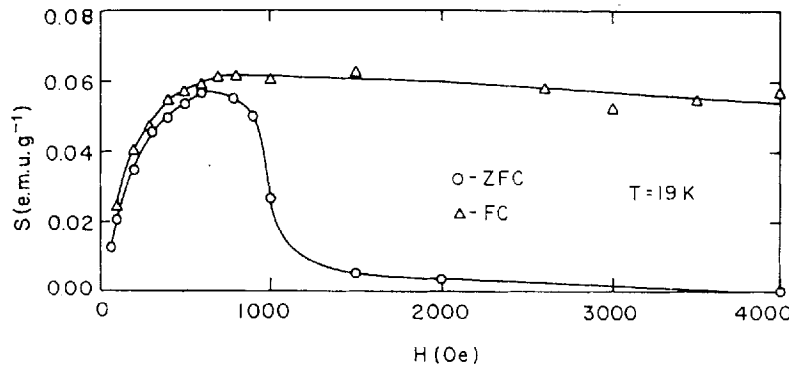
In all cases we find that the changes in the magnetization can be fitted to the equation

$$M(t) = M_1 + S \ln t, \quad (9)$$

in the range $t_1 = 1$ min to $t_2 = 12$ min.

The results obtained by following the first procedure (ZFC and application of a constant field) can be summarized as follows. First, for a constant temperature, S increases with field, peaks at $H_m(T)$ then decreases and vanishes at $H_c(T)$ (the circles in fig. 3 represent typical results). The amplitude of the peak S_m and its position $H_m(T)$ decrease with increasing temperature. We observe a parabolic dependence of S on H for most of the field interval, which enables extrapolation to $S = 0$ with quite reasonable accuracy. Note, however, that the parabolic dependence fails at high fields, above $H = H_p$, where the coefficient S levels off to a certain fraction of S_m and then decreases slowly to zero. The high-field part of $S(H)$ is extrapolated to $S = 0$ by fitting the data to a power law, $S \propto (1 - H/H_c)^\nu$. We find the exponent ν to vary between 1 and 1.5, whereas $H_c(T)$ is determined to within an accuracy of, typically, 20%. Second, for a constant field S increases with temperature, peaks at $T_m(H)$ and then decreases and vanishes at $T_c(H)$. The magnitude of the peak S_m increases and its location T_m decreases with increasing field. The lines described by $T_m(H)$ and $T_c(H)$ coincide, within experimental error, with the $H_m(T)$ and $H_c(T)$ lines defined above.

Fig. 3



The field dependence of the relaxation rate S (\circ) after a ZFC process to 19 K and application of field H , and (\triangle) for thermo-remanent magnetization. (From Yeshurun, Ketelsen and Salamon 1982.)

The results obtained via procedures (1) and (2) are quite different. The triangles in fig. 3 represent results for TRM decay at 19 K, in comparison with the in-field relaxation. $S(H)$ is now roughly constant at high fields. Similar

results are obtained when procedure (2) is used. Unfortunately, the temperature T_{p1} which defines the starting point of the plateau in $S(H)$ is experimentally ill-defined. Therefore, in the following discussions we restrict ourselves to results obtained using procedure (1). Qualitative interpretation of the plateau has been discussed previously (Yeshurun *et al.* 1982).

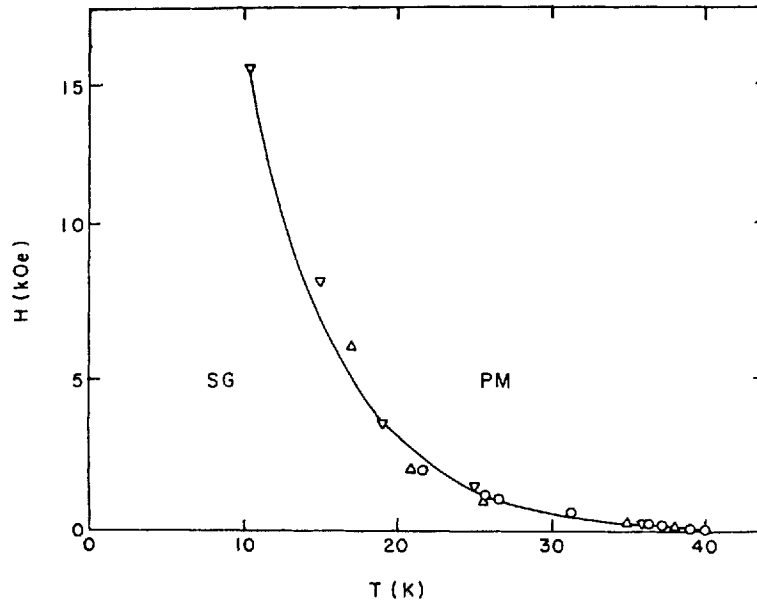
2.4. H - T phase diagram

The results for $T_c(H)$ obtained via eqn. (7) and the results for $H_c(T)$ obtained via extrapolation of $S(H)$ to zero are summarized in fig. 4 in a H - T phase diagram. The circles represent the results of branch point measurements and the triangles the results of viscosity measurements. The smooth solid line connecting the experimental points is the best fit to a power law $\tau = Fh^\phi$, where τ and h are reduced temperature and field respectively, and $T_c(0) = T_1 = 41.6$ K. We find that $\phi = 0.75 \pm 0.15$, in good agreement with the AT prediction for *Ising* spin glasses (eqn. (1)). The experimental field coefficient F is, however, bigger by a factor of ~ 15 than the predicted prefactor A (eqn. (1)). In other words, at a given low temperature, irreversibility is prevented by a field smaller than the one predicted.

§ 3. DISCUSSION

The H - T phase diagram for various spin-glass systems has been explored by several experimental groups by measuring d.c. susceptibility (Monod and Bouchiat 1982, Chamberlin, Hardiman, Turkevich and Orbach 1982, Yeshurun and Sompolinsky 1982, Malozemoff, Barnes and Barbara 1983), a.c. susceptibility (Salamon and Tholence 1983), Faraday rotation (Kett, Gebhardt, Krey and Furdyna 1981, Bontemps, Rajchenbach and Orbach 1983), magnetic relaxation (Yeshurun *et al.* 1982, Tholence and Salamon 1983, Palumbo *et al.* 1982), the magnetocaloric effect (Berton, Chaussy, Odin, Rammal and Tournier 1982) and magnetic torque (Campbell, Arvanitis and Fert 1983). This impressive list allows one to draw some 'universal' conclusions. There is fairly general agreement about the qualitative shape of the experimental AT line. In particular, the critical exponent in these experiments agrees well with the predicted value, $\frac{2}{3}$. The experimental field coefficient, however, ranges from the order of unity to ~ 25 . This range of values might be attributed either to different experimental criteria for the transition or to specific properties of the spin-glass system (clustering effects, spin of magnetic species, effective magnetic moment and Curie-Weiss temperature) which are not usually taken into account in calculating the field scale. Recently there have been some indications that the field scale depends, in the *same* sample, on the time-scale of the experiment (Salamon and Tholence 1983, Bontemps *et al.* 1983, Young 1983, Kinzel and Binder 1983). One expects, therefore, some deviation of the branch point line (represented by the circles in fig. 4; the time-scale is about 30 s) from the viscosity line (represented by the triangles; the time-scale is about 700 s). This is *not* observed in the present experiment. Both approaches yield one transition line, independently, within experimental resolution, of the time-scale. We note, however, that in order to determine total vanishing of irreversibility we have used a different extrapolation procedure in each experiment. This

Fig. 4



Field-temperature phase diagram for $(\text{Fe}_{0.64}\text{Mn}_{0.36})_{75}\text{P}_{16}\text{B}_6\text{Al}_3$, showing the results of (○) branch point measurements and (△) viscosity measurements.

might lead to a systematic error in the results and affect the field coefficient (but not the exponent).

A proper extrapolation procedure is indeed crucial for these kinds of experiment. Usually the extrapolation procedure introduces large errors in the experimental critical points. This problem can be drastically reduced by using the scaling properties of the experimental data (Salamon and Tholence 1983). We demonstrate this for magnetic relaxation data for a polycrystalline $\text{Fe}_{0.84}\text{Cr}_{0.16}$ spin-glass system (Palumbo *et al.* 1982). Relaxation data were recorded using procedure (1) (§ 2.3). The slow relaxation was fitted to a power law

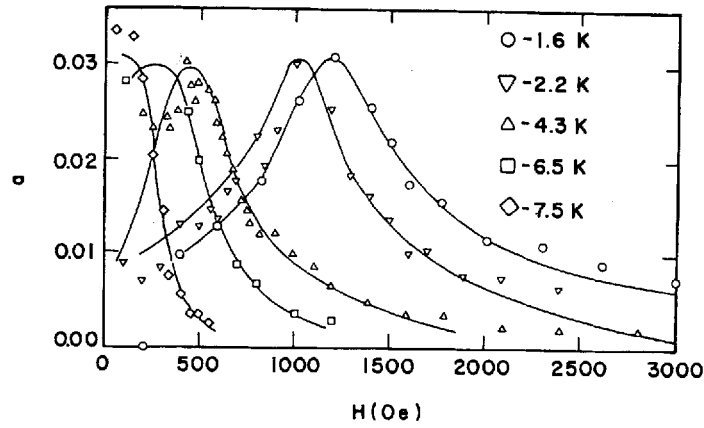
$$M = M_0 t^a \quad (10)$$

as well as to a logarithmic function (eqn. (9)). In all the relaxation experiments it was found that $a \ll 1$, and therefore it is difficult to distinguish experimentally between a power law and logarithmic behaviour. We note, however, that by taking derivatives with respect to $\ln t$ for both eqns. (9) and (10), and provided $a \ll 1$, one finds that

$$a \approx S/M_0. \quad (11)$$

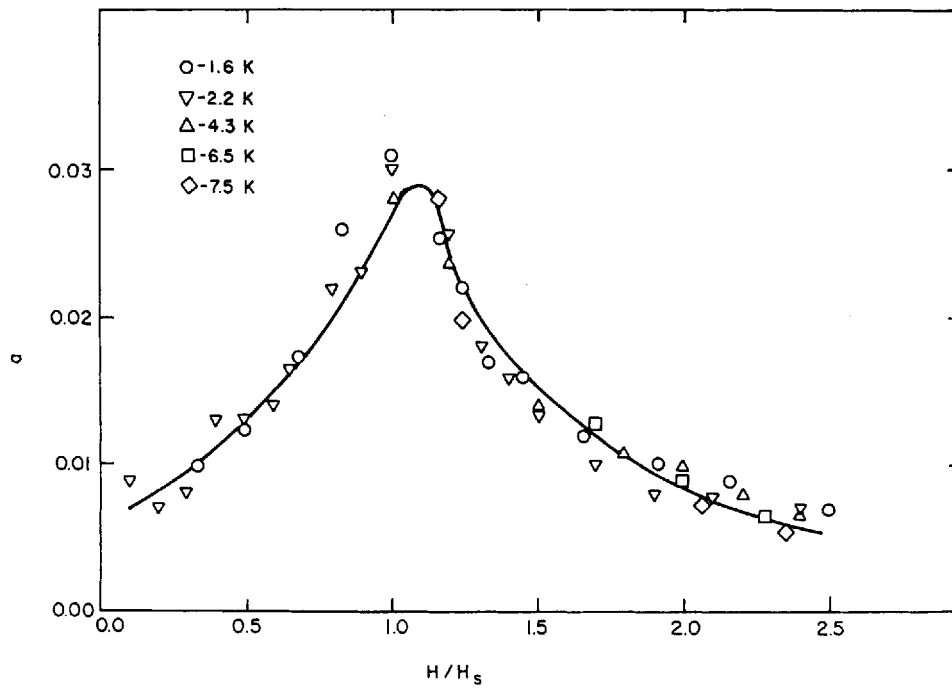
Figure 5 shows the exponent $a(H)$ of eqn. (10) for five isotherms. An interesting feature of the $a(H, T)$ data is the weak temperature dependence of the relaxation rate at the maximum. This simple behaviour enables all data points to be collapsed into a single curve with a scaling field parameter $H_m(T)$, where H_m is the field for which $a(H)$ has a maximum. This scaling procedure is shown in

Fig. 5



The exponent $a(H)$ in eqn. (10) for five isotherms. (From Palumbo, Parks and Yeshurun 1982.)

Fig. 6



The data from fig. 5 scaled to a single curve. The scaling parameter $H_s(T)$ is the field for which $a(H)$ is the maximum. (From Palumbo, Parks and Yeshurun 1982.)

fig. 6. Owing to the scaling property of the data, it is clear that $H_c(T)$, the critical field for the vanishing of viscosity data, is proportional to $H_m(T)$. The maxima $H_m(T)$ can, of course, be determined much more accurately than the critical fields $H_c(T)$. The procedure therefore yields better estimates of the critical exponent and the qualitative shape of the critical line, but with the penalty of an undetermined prefactor.

We turn now to one of the most intriguing questions in the present subject. This is the apparent absence of the GT line (eqn. (4)), in most experimental results. For isotropic samples this line marks the freezing of transverse spin components. Some evidence for such freezing is indeed found in Mössbauer experiments (see, for example, Campbell, Senoussi, Varret, Teillet and Hamzic 1983). This transverse freezing is expected to induce weak longitudinal irreversible response. At the AT line (eqn. (1)) a cross-over to strong irreversible response is expected (Cragg and Sherrington 1982, Kotliar and Sompolinsky 1984). The absence of the GT line here and for other isotropic spin-glass systems has been attributed to either insensitivity of the experimental apparatus to the weak longitudinal irreversibility (Cragg and Sherrington 1982) or to local random anisotropies which lead to an Ising behaviour (Kotliar and Sompolinsky 1984, Sompolinsky 1984). The first approach interprets the experimental AT line as a cross-over line, whereas the second approach views it as a true critical line in the presence of a field. In any case, there is general agreement that the AT prediction has found solid experimental evidence.

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