PHYSICAL REVIEW B VOLUME 26, NUMBER 3 1 AUGUST 1982

Irreversible response in spin-glasses: An experimental study in amorphous Fe-Mn

Y. Yeshurun*

Department of Physics and Materials Research Laboratory, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801 and Physics Department, Polytechnic Institute of New York, Brooklyn, New York 11201

H. Sompolinsky

Department of Physics, Harvard University, Cambridge, Massachusetts 02138

(Received 4 January 1982; revised manuscript received 20 May 1982)

The equilibrium and nonequilibrium susceptibilities χ_e and χ_{ne} of the spin-glass $(Fe_{0.64}Mn_{0.36})_{75}P_{16}B_6Al_3$ have been measured as functions of temperature ($4.2 \leq T \leq 80$ K) and a cooling field (8 Oe $\leq H \leq 2$ kOe). A phase transition occurs at a temperature $T_c(H)$ below which the irreversible response $\Delta(T) \propto T(\chi_e - \chi_{ne})$ appears. $T_c(H)$ decreases from the freezing temperature ($T_f = 41.6$ K) as $H^{0.75 \pm 0.1}$. For small $\tau \equiv 1 - T/T_c(H)$, Δ behaves as $A\tau + B\tau^2$ with field-dependent coefficients A and B and $A(H=0)=0$. The results are compared with the predictions of the mean-field theory of spin-glasses.

A well-known characteristic of spin-glasses is the appearance of slow irreversible phenomena at low temperatures.[1-3] A recent mean-field theory[4] of the Edwards-Anderson (EA)[5,6] spin-glass (SG) model has incorporated these relaxation processes as a central feature of the SG transition. The theory describes the SG phase by order parameters which relax with a broad distribution of relaxation times due to crossing of the energy barriers between highly degenerate ground states. These relaxation times diverge in the thermodynamic limit. In the *finite*-time limit denoted by t_1, the nonequilibrium susceptibility $\chi_{ne} = \int_0^{t_1} \chi(t)\, dt$ obeys, in zero field, the Fischer relation:

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

where q_{EA} is the nonequilibrium value of the EA order parameter, $q_{EA} = [\langle S_i(0) S_i(t_1) \rangle]_{av}$; C is the Curie constant, and we assume for simplicity that the Curie temperature Θ is zero. Another order parameter is the irreversible response Δ defined by the difference between χ_{ne} and the true equilibrium susceptibility χ_e,

$$\chi_e = \chi_{ne} + \frac{C}{T}\Delta \quad . \tag{2}$$

The use of Δ as an order parameter is particularly useful in the presence of a finite field H. In this case, q_{EA} is nonzero even at high temperature but Δ appears according to the mean-field theory, only below a field-dependent critical temperature $T_c(H)$.

In this work we present an experimental study of the irreversible response as a SG order parameter by measurements of χ_e and χ_{ne} of $(Fe_{0.64}Mn_{0.36})_{75}P_{16}B_6Al_3$ in the range of temperatures from 4.2 to 80 K and fields of 8 Oe up to 2 kOe. We assume that by *cooling* in a field, the system reaches quickly its true equilibrium state appropriate to that field.[2,7,8] Since we are interested in the effect of a true static field on the equilibrium properties of the SG, both χ_e and χ_{ne} have been measured as functions of the *cooling* field H. Thus, χ_e is defined here as

$$\chi_e(H) \equiv \frac{M(H+\delta H) - M(H)}{\delta H} \quad , \tag{3}$$

where $M(H)$ and $M(H+\delta H)$ are the magnetizations induced by *cooling* fields H and $H+\delta H$, respectively. On the other hand,

$$\chi_{ne}(H) \equiv \frac{M(H,\delta h) - M(H)}{\delta h} \quad , \tag{4}$$

where $M(H,\delta h)$ is the magnetization measured after *cooling* in a field H and *then increasing the field* to $H+\delta h$. From these measurements, we extracted Δ as a function of T and H and located its critical line $T_c(H)$. We compare the results with the predictions of the SG mean-field theory.

The sample chosen for the present work is the well-studied $(Fe_{0.64}Mn_{0.36})_{75}P_{16}B_6Al_3$, for which the low-field ac susceptibility measurement[9] shows a sharp cusp at $T_f \simeq 41$ K. Ribbons were prepared by centrifugal spin quenching[10] and small chips ($5 \times 1 \times 0.05$ mm^3) were stacked in parallel and introduced into a vibrating sample magnetometer. Magnetization data were recorded in the following procedure. The sample was cooled in a field H from 80 to 4.2 K where the field was increased by δh. Then, the temperature was increased in steps up to 80 K and at each temperature the magnetization

$M_1 = M(H, \delta h)$ was recorded. Next, temperature was reduced without changing the field, and the magnetization $M_2 = M(H + \delta H)$ was recorded. We refer to this procedure as a *differential branch-point* measurement. Since $\delta h = \delta H$ the order parameter Δ can be evaluated via Eqs. (2), (3), and (4)

$$\Delta = \frac{T}{C} \frac{M_2 - M_1}{\delta H} \quad . \tag{5}$$

The magnetization M_1 was recorded when the rate of change of M_1 was less than 0.5% over 30 sec. This is done in order to achieve stable temperature conditions on one hand and consistent measurement of the nonequilibirum state on the other hand.

The susceptibilities of $(Fe_{0.64}Mn_{0.36})_{75}P_{16}B_6Al_3$ were measured by the above procedure with $H = 0$ and $\delta h = \delta H = 8$ Oe. Note that for $H = 0$, χ_e and χ_{ne} are simply the field-cooled and zero-field-cooled low-field suscéptibilties. The critical temperature $T_c(0) = 41.6 \pm 0.1$ is identical to the freezing temperature identified by the maximum of χ_{ne}. In order to extract the order parameters q_{EA} and Δ we need to know the values of the Curie constant C and the Curie temperature Θ. The parameters, however, vary with temperature and in particular Θ decreases from about 10 K when extrapolated near 80 K to about zero near T_f. This phenomenon was observed in other SG systems,[11,12] and is presumably due to the gradual formation of short-range order above T_f. In Fig. 1 we present the values of q_{EA} and Δ as extracted from χ_e and χ_{ne} according to Eqs. (1) and (2) with the parameters $C(T_f) = 0.5$ K cm^3/g and $\Theta(T_f) = 0$ K, since they reflect better the behavior of the system at $T \lesssim T_f$. The qualitative features of the results are quite insensitive to variations in C and Θ. As can be seen from Fig. 1, $q_{EA}(H=0)$ and $\Delta(H=0)$ increase continuously from zero below T_f, as

$$q_{EA}(H=0) \propto (1 - T/T_f)^{\beta} \quad , \quad \beta = 1.25 \pm 0.25 \quad , \tag{6}$$

$$\Delta(H=0) \propto (1 - T/T_f)^{\beta'} \quad , \quad \beta' = 2.0 \pm 0.2 \quad . \tag{7}$$

The uncertainties in β and β' reflect error bars in χ and T_f and also possible positive values of Θ. The result (6) for β' is slightly higher than the mean-field value $\beta = 1$. A similar trend was found in other works.[13] The result (7) is consistent with the value $\beta' = 2$ of the mean-field theory.

We turn to the results for nonzero H. The values of M_1 and M_2 for the case $H = 200$ Oe, $\delta H = \delta H = 40$ Oe, are shown in Fig. 2. For comparison we also present the values of the *field-cooled* magnetization $M(H = 200$ Oe) and the *zero-field-cooled* nonequilibrium magnetization $M(h = 200$ Oe) measured after turning on the field h at low T. Since the experimental resolution of $M_2 - M_1$ puts a lower limit on the measured values of Δ, an accurate determination of $T_c(H)$ requires a procedure for extrapolating the measured $\Delta(H)$ to zero. In order to do this we note that at high fields, Δ has both linear and quadratic dependences on T as can be seen in the inset of Fig. 2. Hence we fitted the data for $\Delta(H,T)$ to the form

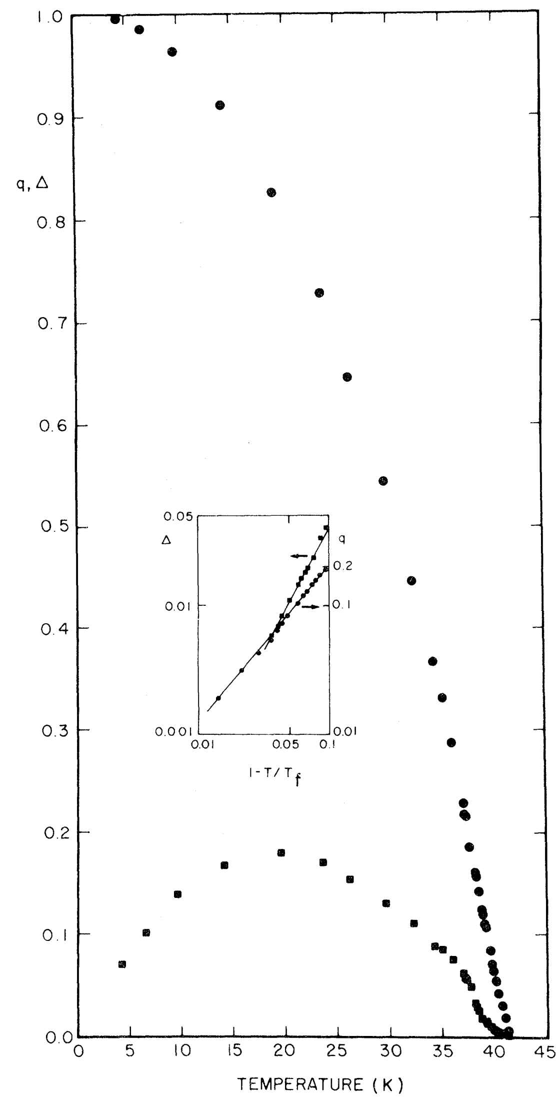

FIG. 1. Results for q_{EA} and Δ for $H = 0$ extracted from magnetic measurements at a field of 8 Oe. Inset shows ln-ln plot of these order parameters in the vicinity of T_f.

$$\Delta(H,T) = A(H)\tau + B(H)\tau^2 \quad , \quad \tau \equiv 1 - T/T_c(H) \tag{8}$$

in the range $0.03 \leq \tau \leq 0.1$, from which $T_c(H)$ was determined. The results, which are plotted in Fig. 3, show a power-law dependence of T_c on H,

$$\tau_c \equiv 1 - T_c/T_f \simeq a(g\mu_B H/k_B T_f)^{\delta} \quad , \quad \delta = 0.75 \pm 0.1 \tag{9}$$

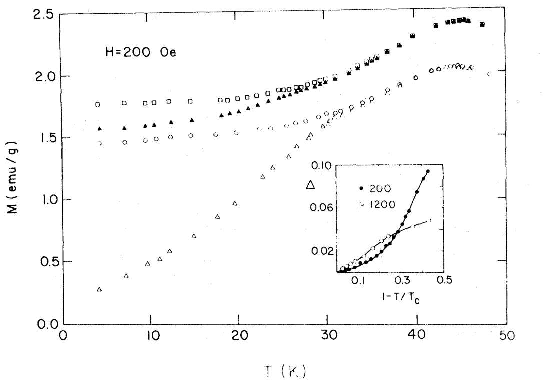

FIG. 2. *Differential branch-point* measurement at a field of 200 Oe. □ is the magnetization measured after *cooling* in a field 240 Oe; ▲ is the magnetization after *cooling* in a field 200 Oe and then *turning on* an additional field $\delta h = 40$ Oe; ○ refers to *cooling* in a field 200 Oe; and Δ to *zero-field cooling* and *turning on* a field of 200 Oe. Inset of the figure shows the values of the order parameter Δ is cooling fields 200 and 1200 Oe.

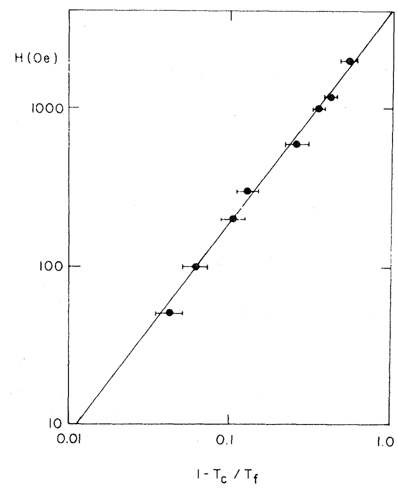

FIG. 3. Values of the reduced critical temperature $\tau_c \equiv 1 - T_c/T_f$ vs field H, as extracted from measurements of $\Delta(H,T)$. See text.

with $a \simeq 13 \pm 2$. This result is in agreement with the value of T_c extracted from measurements[3] of the onset of magnetic viscosity in this material. In addition, a recent study[14] of the SG transition in a finite field in Ag:Mn also yielded $\delta \sim 0.7$.

According to recent investigations[15,16] of the mean-field theory of Heisenberg SG, a phase transition occurs at a temperature

$$\tau_c \propto (g\mu_B H/k_B T_f)^2 \quad , \tag{10}$$

below which the transverse components of $q_{\rm EA}$ freeze. This in turn gives rise[16–18] to an irreversible response in both the transverse and the longitudinal susceptibilities. However, in the vicinity of τ_c of (10), the irreversible longitudinal response is expected to be proportional to τ^3 which may well be below the experimental resolution. This may explain the significant discrepancy between the results (9) and (10). It should also be noted that the result (10) holds only for a pure Heisenberg SG and inclusion of anisotropies leads to an Ising behavior in which[19]

$$\tau_c \simeq (3\mu_B g H/4k_B T_f)^{2/3} \quad . \tag{11}$$

In this case, $\Delta(H,T)$ is indeed of the form (8) with $A(H=0)=0$. Thus the experimental results may actually indicate that a significant amount of anisotropy is present in our system. This may also explain the observed T^2 behavior of $1-q_{\rm EA}(H=0)$ at $T \rightarrow 0$ K (see Fig. 1). Such a behavior is in accord with the mean-field theory of Ising SG whereas in the Heisenberg case $(1-q_{\rm EA})/T$ is expected to remain finite at $T \rightarrow 0$ K due to large fluctuations transverse to the *local* fields. It should also be noted that the coefficient in (9) is an order-of-magnitude larger than the result (10). This enhancement of the effect of the field is probably due to short-range ordering which gives rise to large effective magnetic moments.

In conclusion the experimental results are consistent with the prediction of a continuous SG transition in a finite field associated with the onset of irreversible magnetic response. The observed field dependence of the transition temperature indicates an Ising-type behavior probably due to the presence of relatively large anisotropy in the system.

ACKNOWLEDGMENTS

We are grateful to Professor R. Orbach for helpful discussions and for sending us a copy of his unpublished work, Ref. 14. We thank Professor B. I. Halperin, Professor M. B. Salamon, and Professor R. D. Parks for many useful discussions. The sample has been kindly prepared by Dr. H. S. Chen of Bell Laboratories. We also thank C. Henley for his help with the numerical analysis. Work done in Illinois was supported in part by National Science Foundation and Materials Research Laboratory Grant No. DMR-77 23999. Work in New York is supported by the Office of Naval Research and at Harvard by National Science Foundation Grant No. DMR-77 10210. One of us (H.S.) also acknowledges the support of a Weizmann Fellowship.

*Permanent and present address: Department of Physics, Bar-Ilan University, Ramat-Gan, Isreal.

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