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

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The equilibrium and nonequilibrium susceptibilities $\chi_e$ and $\chi_{ne}$ of the spin-glass
($Fe_{0.64}Mn_{0.36}$)$_3P_{16}B_6Al_1$ have been measured as functions of temperature ($4.2 \leq T \leq 80$ K)
and a cooling field ($8 \text{ Oe} \leq H \leq 2 \text{ kOe}$). A phase transition occurs at a temperature $T_c(H)$
below which the irreversible response $\Delta(T) = T(\chi_e - \chi_{ne})$ appears. $T_c(H)$ decreases from
the freezing temperature ($T_f = 41.6$ K) as $H^{0.75 \pm 0.1}$. For small $\tau = 1 - T/T_c(H)$, $\Delta$ 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. A recent mean-field theory of the Edwards-Anderson (EA) 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 $t_i$, the nonequilibrium susceptibility
$\chi_{ne} = \int_0^t \chi(t) \, dt$ obeys, in zero field, the Fischer relation:

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

where $q_{EA}$ is the nonequilibrium value of the EA order parameter, $q_{EA} = \{ \langle S_i(0) S_j(t_i) \rangle \}_{EA}$; $C$ is the Curie constant, and we assume for simplicity that the Curie temperature $\Theta$ is zero. Another order parameter is the irreversible response $\Delta$ defined by the difference between $\chi_{ne}$ and the true equilibrium susceptibility $\chi_e$:

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

The use of $\Delta$ 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 $\Delta$ 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 $\chi_e$ and $\chi_{ne}$ of ($Fe_{0.64}Mn_{0.36}$)$_3P_{16}B_6Al_1$ 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. Since we are interested in the effect of a
true static field on the equilibrium properties of the SG, both $\chi_e$ and $\chi_{ne}$ have been measured as functions
of the cooling field $H$. Thus, $\chi_e$ is defined here as

$$\chi_e(H) = \frac{M(H) + H - M(H + H)}{\delta H} ,$$

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

$$\chi_{ne}(H) = \frac{M(H, \delta h) - M(H)}{\delta h} ,$$

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 $\Delta$ 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 ($Fe_{0.64}Mn_{0.36}$)$_3P_{16}B_6Al_1$, for which the low-field ac susceptibility measurement shows a sharp cusp at $T_f = 41$ K. Ribbons were prepared by centrifugal spin quenching and small chips ($5 \times 1 \times 0.05 \text{ mm}^3$) 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 $\delta h$. Then, the temperature was increased in steps up to 80 K
and at each temperature the magnetization...
\( M_I = M(H, \delta h) \) 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 \( \Delta \) can be evaluated via Eqs. (2), (3), and (4)

\[
\Delta = \frac{T}{C} \frac{M_2 - M_I}{\delta H}.
\]

The magnetization \( M_I \) was recorded when the rate of change of \( M_I \) 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 nonequilibrium state on the other hand.

The susceptibilities of (Fe_{0.64}Mn_{0.36})_{7}P_{16}B_{8}Al_{3} were measured by the above procedure with \( H = 0 \) and \( \delta h = \delta H = 8 \) Oe. Note that for \( H = 0 \), \( x_e \) and \( x_{ne} \) are simply the field-cooled and zero-field-cooled low-field susceptibilities. The critical temperature \( T_c(0) = 41.6 \pm 0.1 \) is identical to the freezing temperature identified by the maximum of \( x_{ne} \). In order to extract the order parameters \( q_{EA} \) and \( \Delta \) we need to know the values of the Curie constant \( C \) and the Curie temperature \( \Theta \). The parameters, however, vary with temperature and in particular \( \Theta \) decreases from about 10 K when extrapolated near 80 K to about zero near \( T_f \). This phenomenon was observed in other SG systems, 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 \( \Delta \) as extracted from \( x_e \) and \( x_{ne} \) 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 \( T \leq T_f \). The qualitative features of the results are quite insensitive to variations in \( C \) and \( \Theta \). As can be seen from Fig. 1, \( q_{EA}(H = 0) \) and \( \Delta(H = 0) \) increase continuously from zero below \( T_f \), as

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

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

The uncertainties in \( \beta \) and \( \beta' \) reflect error bars in \( x_e \) and \( T_f \) and also possible positive values of \( \Theta \). The result (6) for \( \beta' \) is slightly higher than the mean-field value \( \beta = 1 \). A similar trend was found in other works. 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 \( M_I \) 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 \( M_2 - M_I \) puts a lower limit on the measured values of \( \Delta \), 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, \( \Delta \) 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

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

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 \( H \),

\[
\tau_c = 1 - T_c/T_f \approx a(g \mu_B H/k_B T_f)^\delta, \quad \delta = 0.75 \pm 0.1 \tag{9}
\]
with $a = 13 \pm 2$. This result is in agreement with the value of $T_c$ extracted from measurements of the onset of magnetic viscosity in this material. In addition, a recent study of the SG transition in a finite field in Ag:Mn also yielded $\delta \sim 0.7$.

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

$$\tau_c \propto (g \mu_B H/k_B T)^2,$$

below which the transverse components of $q_{EA}$ freeze. This in turn gives rise to an irreversible response in both the transverse and the longitudinal susceptibilities. However, in the vicinity of $\tau_c$ of (10), the irreversible longitudinal response is expected to be proportional to $\tau$ 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

$$\tau_c \propto (3 \mu_B g H/4k_B T)^{2/3}.$$

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 $T^n$ behavior of $1 - q_{EA}(H=0)$ at $T \to 0$ K (see Fig. 1). Such a behavior is in accord with the mean-field theory of Ising SG whereas in the Heisenberg case $(1 - q_{EA})/T$ is expected to remain finite at $T \to 0$ 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.

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