Low-temperature magnetic viscosity in the re-entrant ferromagnet

$(Fe_{0.68}Mn_{0.32})_{75}P_{16}B_6Al_3$

LETTER TO THE EDITOR

Low-temperature magnetic viscosity in the re-entrant ferromagnet $(Fe_{0.68}Mn_{0.32})_{75}P_{16}B_6Al_3$

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Abstract. Studies of magnetic viscosity in $(Fe_{0.68}Mn_{0.32})_{75}P_{16}B_6Al_3$ are presented. The magnetisation in a constant field H is described by $M = M_1 + S \ln t$. The coefficient $S(T)$ peaks at $T_m(H)$. We show that S and the susceptibility χ scale similarly and deduce the ferromagnetic/spin glass transition temperature $T_{fg} \simeq 35$ K, in agreement with our previous results.

Magnetic viscosity is a term used to describe the slow changes in magnetisation that lag behind the changes in the external magnetic field. This phenomenon, which has been known in ferromagnetic materials for almost a century (see Dunlop 1973), has attracted new interest in the study of spin glasses. The spin glass may be defined as a system of randomly oriented 'frozen' spins with a non-zero, time-average magnitude for each spin but with no long-range spin order. In the infinite time limit this definition corresponds to the Edwards–Anderson (EA) spin glass phase (Edwards and Anderson 1976). However, the existence of an EA-type spin glass phase transition is still controversial. One experimental approach to resolve this question is to examine the nature of time-dependent phenomena in spin glass systems. In particular, magnetic viscosity is one of the major characteristics of spin glasses, and has been studied in many systems (Guy 1978, Ferre *et al* 1980, 1981, von Löhneysen and Tholence 1979). The time dependence of the thermo-remanent, the isothermal remanent and the in-field magnetisation has been found to obey either a weak power law or a logarithmic law. (As a matter of fact, it is almost impossible from an experimental point of view to distinguish between the two). The exponent in the power law and the coefficient of the logarithmic term are found to be field and temperature dependent with a tendency—in some cases (Ferre *et al* 1981)— to diverge near the freezing temperature T_f. For $T \ll T_f$, the exponent has been predicted to be linear in T by Monte Carlo simulations of the EA spin glass (Binder and Schröder 1976, Kinzel 1979, Dasgupta *et al* 1979) and by several theoretical models based upon a simple physical picture of small-cluster, two-level systems (Dasgupta *et al* 1979, Ma 1980). The time dependence near T_f still lacks a theoretical explanation.

While the paramagnetic/spin glass (PM/SG) transition is still disputed, there seems to be more agreement about the existence of a ferromagnetic/spin glass (FM/SG) phase transition. This transition, which was first predicted by Sherrington and Kirkpatrick (SK) (1975), also occurs in other models (Klein *et al* 1979, Kinzel 1979, Fishman and Aharony 1980). Experimental evidence for such a spin glass phase at low temperature

may be found from data on several materials through susceptibility (Verbeek *et al* 1978, Rao *et al* 1980), DC magnetisation (Shull *et al* 1976, Dublon *et al* 1980) and neutron scattering measurements (Maletta and Convert 1979, Fincher *et al* 1980). Recently we demonstrated (Yeshurun *et al* 1980) that there is indeed a second-order FM/SG phase transition by analysing the critical behaviour of the magnetisation of amorphous $(Fe_{0.68}Mn_{0.32})_{75}P_{16}B_6Al_3$ which undergoes a PM/FM transition at $T_c \simeq 100$ K and a FM/SG transition at $T_{fg} \simeq 38$ K. In the course of studying this latter transition we observed considerable magnetic viscosity. We report here a set of in-field measurements on the same sample focusing on the time-dependent effects near T_{fg}. This is, to our knowledge, the first detailed report of time-dependent phenomena near a FM/SG transition temperature. The changes in the magnetisation in the present experiment can be described by a logarithmic law, as in other spin glasses. However, we find sharp peaks in the *coefficient* of the logarithmic term when plotted versus field at a constant temperature or versus temperature at a constant field. Later, we will argue that these are susceptibility-like peaks and we thus gain more, independent information about this transition.

The sample was prepared by H S Chen by the rapid quenching method in the form of long ribbons. Fifteen small pieces ($5 \times 0.1 \times 0.05$ mm) were stacked together in a vibrating-sample magnetometer (VSM) sample holder such that the external field H_{ext} is parallel to the longest axis, to minimise the demagnetisation factor. The sample was first cooled in zero field to a temperature $T < T_{fg}$, then the field was turned on and magnetisation data were recorded. Preliminary measurements had taught us that in most, but not all, cases $M(t)$ is linear in log t for 30s $\lesssim t \lesssim$ 300s. In the present report we limit ourselves to 4 K $\leqslant T \leqslant$ 40 K, 50 Oe $\leqslant H_{ext} \leqslant$ 4 kOe where a simple logarithmic law suffices to describe accurately the changes in the magnetisation. Within those limits we recorded $M(t)$ for $\Delta t \simeq$ 300s along different isotherms (temperature interval = 5 K) following step increases in H_{ext}. Between steps, $M(t)$ was recorded for time period $\Delta t \simeq$ 300s. Note that basically this is the way by which $M(H, T)$ data (Yeshurun *et al* 1980) were accumulated, with only the asymptotic value reported.
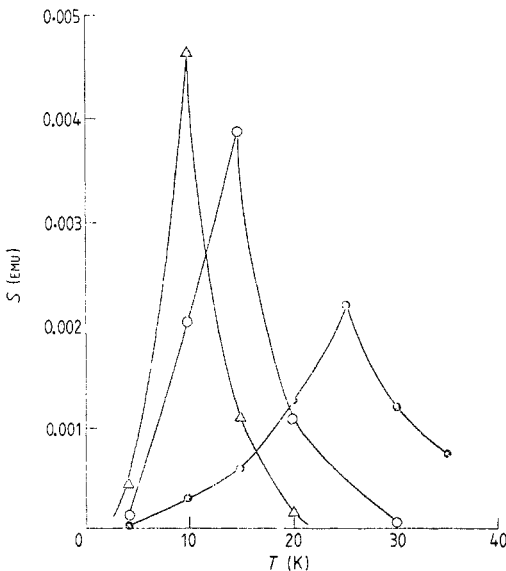

Figure 1. Slope of logarithmic tail versus temperature for three different fields: ●, 100 Oe; ○, 400 Oe; △, 800 Oe.

A change in H_{ext} causes a rapid change in the magnetisation followed by a logarithmic tail which was fitted to

$$M(t) = M_1 + S \ln t \tag{1}$$

where M, the magnetisation at $t = 1$ s found by least-squares analysis, is usually smaller than the actual value $M(1)$ due to deviations from (1) at short times.

The field and temperature dependence of the coefficient S is described as follows: (i) $S(T)$ exhibits a sharp cusp at $T_m(H)$ for each stepped external field H. (See figure 1 for some representative results.) The peak position $T_m(H)$ decreases with increasing final field while the amplitude of the peak increases with field. (ii) $S(H)$ peaks at $H_m(T)$ for a constant T. (See figure 2 for some representative results.) The field H_m and the amplitude of the peaks decrease with increasing temperature.
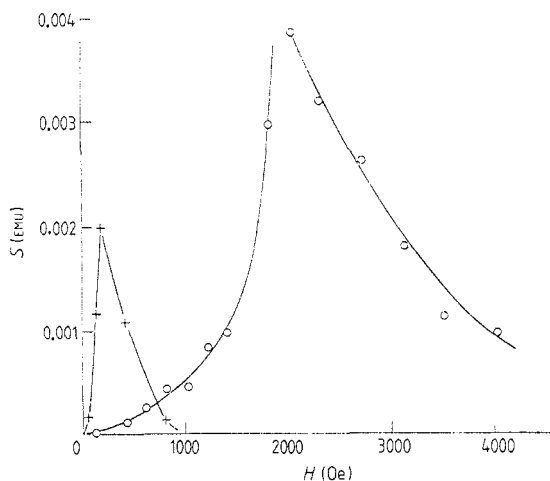

Figure 2. Slope of logarithmic tail versus external field at ($\bigcirc$) 4.2 K and (+) 20 K.

Richter (1937) suggested a phenomenological model to account for the logarithmic time dependence of the magnetic viscosity. The starting point of this model assumes exponential relaxation of the instantaneous magnetisation toward the equilibrium value. However, the relaxation times are assumed to be distributed according to

$$g(\tau) = g_0/\tau \qquad \text{for} \qquad \tau_1 \leq \tau \leq \tau_2 \tag{2}$$

$$= 0 \qquad \text{elsewhere.}$$

Integration of the relaxation function over all possible relaxation times gives a logarithmic decay for a restricted time interval.

Another phenomenological approach was proposed by Néel (1955) and by Street and Woolley (1956) who considered the thermal activation of domain walls and particle magnetisation over a distribution of energy barriers. An interesting aspect of this model is a semi-empirical relationship between S and the static susceptibility:

$$S(H, T) \sim kT\chi_0(H, T)/q, \tag{3}$$

where q is the change in activation energy with applied field; it reduces to the particle

moment for an ensemble of identical domains. The simplest linear response argument gives a similar result. The response of the system to a step change in magnetisation is governed by the frequency-dependent susceptibility which gives the magnetisation resulting from each Fourier component of the field step. For a step at $t = 0$, trivial arguments give

$$M(t) = H_a \int_{-\infty}^{t} \chi(t') \, dt', \tag{4}$$

where $\chi(t')$ is the Fourier transform of the frequency-dependent susceptibility. Because we expect the entire spectrum of $\chi(\omega)$ to scale with the static susceptibility, we predict
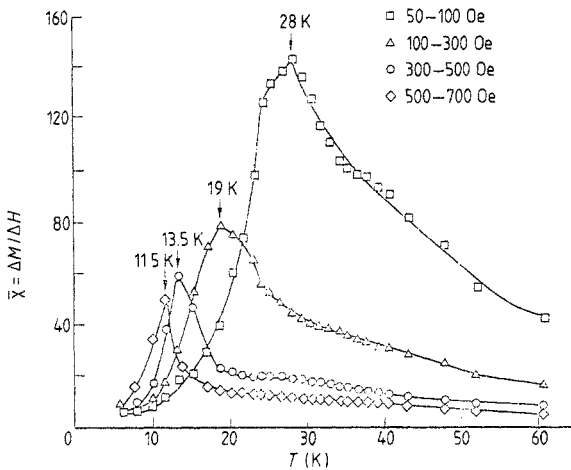
$$S(H, T) \sim H_a \chi_0(H, T). \tag{5}$$

Figure 3. Average susceptibility $\Delta M/\Delta H$ versus temperature for $(Fe_{0.68}Mn_{0.32})_{75}P_{16}B_6Al_3$.

In figure 3 we have plotted the ratio $\Delta M/\Delta H$ for this material, obtained by subtracting neighbouring isochamps reported earlier (Yeshurun *et al* 1980). The position of the peaks compares well with those of $S(H, T)$ at comparable applied fields, and the peak heights scale with H as in equation (5). The close relationship between S and χ has further implications. According to scaling laws, which we assert hold at this lower transition, the susceptibility scales as

$$\chi = t^{-\gamma} \chi^* (H/t^{\beta\delta}), \tag{6}$$

where here $t = 1 - T/T_{fg}$. Any feature, such as a peak, should scale with field so that

$$t_m \sim H^{1/\beta\delta}. \tag{7}$$

Our previous scaling results give $1/\beta\delta \simeq 0.5$. To test this, we plot T_m against $H^{1/2}$ in figure 4. The best-fitting line gives $T_{fg} \approx 35 \, K$ which agrees with our previous value of $38 \, K$ within experimental uncertainty.

In conclusion we point out two interesting observations. (i) Although theoretical models which treat magnetic viscosity assume a PM/SG transition, they agree with the
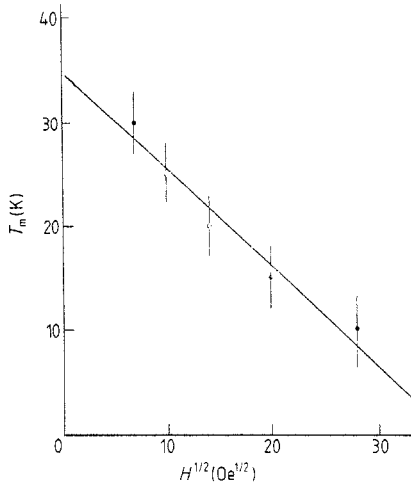
Figure 4. The peak temperature T_m versus square root of external field H. The straight line is the result of a least-squares analysis. Note that for $H = 0$, $T_m \approx 35\,\text{K}$.

present results, which deal with a FM/SG transition. This is seen clearly when we compared the peaks in $S(H)$ with Monte Carlo calculations (e.g. Kinzel 1979). Note that similar behaviour of $S(H)$ was reported by Guy (1978) for Au–Fe spin glasses. From this point of view the spin glass phase has the same properties, regardless of the type of transition. (ii) Scaling laws are used quite successfully here as well as in previous works. This, of course, favours the existence of an actual FM/SG phase transition. Note that the low-field relaxation peak agrees well with our previous determination of T_{fg}.

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