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LETTER TO THE EDITOR

Spin dynamics in the anisotropic spin glass Fe_2TiO_5

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Abstract. We have studied spin-freezing phenomena along the magnetic easy axis of the insulating spin glass Fe_2TiO_5 by magnetisation, AC susceptibility and neutron scattering experiments. The characteristic measurement time for these techniques varies over more than fourteen orders of magnitude. The results, which show a decrease of the freezing temperature with decreasing characteristic frequency, are analysed in terms of three models.

Measurements of magnetic relaxation phenomena in spin glasses are sensitive to the 'window' of the experimental measuring time. Thus, the increase in the relaxation time when the size of the correlations increases is manifested at a characteristic temperature T_g which depends on the experimental frequency f. Over the past few years much interest has been devoted to this frequency dependence (for a recent review see Fischer 1984), but most of the available data are limited to a small range of frequencies. In this Letter we investigate the functional form of $T_g(f)$ for the insulator spin glass Fe_2TiO_5 over more then fourteen orders of magnitude of measurement time scales, by magnetisation, AC susceptibility and neutron scattering measurements.

The system under study, a single crystal of Fe_2TiO_5, exhibits anisotropy in the magnetic susceptibility (Atzmony *et al* 1979, Yeshurun *et al* 1984, Yeshurun and Sompolinsky 1985). Here we limit ourselves to describing longitudinal (i.e. along the easy axis) relaxation only. A small sample ($2 \times 2.9 \times 5.5\ mm^3$) was used for DC and AC measurements. A bigger sample ($3 \times 5 \times 9\ mm^3$) of the same batch was used for the neutron-scattering experiments.

The zero-field-cooled (ZFC) and the field-cooled (FC) branches of the magnetisation were measured with a SQUID susceptometer. The experimental procedure is as follows. The sample is cooled in zero field to 2 K, a field H (10 Oe $\leqslant H \leqslant$ 1000 Oe) is applied, and the ZFC branch is measured up to $T \simeq 80$ K. Then, without changing the field, the sample is cooled slowly to 2 K and the FC magnetisation is measured while cooling. A typical time for scanning the whole temperature range is of the order of 10^4 s. The time associated within each temperature point in the ZFC run is estimated to be 10^2 s. The FC process is much closer to the equilibrium state (Malozemoff and Imry 1981, Lundgren *et al* 1983); we argue below that the time associated with the FC branch is 10^4–10^5 s. The ZFC branch increases with temperature and exhibits a cusp at $T_g = 48.8 \pm 0.1$ K

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independent of field up to 1 kOe. The FC branch depends weakly on temperature below T_{ZFC} but still exhibits a small and broad maximum at $T_g = 48.1 \pm 0.1$ K.

The AC susceptibility χ_{ac} was measured with a mutual inductance coil. For each measurement the sample was systematically displaced between the two pick-up coils to eliminate the empty coil contribution. Measurements were taken at 24.2, 242 and 2420 Hz with the same peak-to-peak value (~3 Oe) of the AC driving field. Both the in-phase part χ'_{ac} and the out-of-phase part χ''_{AC} of the susceptibility exhibit frequency and temperature dependence similar to the behaviour found in other spin glass systems (Lundgren *et al* 1981, Tholence 1981, Mulder *et al* 1982, Goldfarb *et al* 1982). More details of this frequency dependence will be presented elsewhere. We concentrate here on the frequency dependence of the cusp temperature $T_g(f)$, which we assume to coincide with the temperatures where the measuring frequencies match the sample's characteristic fluctuation frequencies. These temperature values are summarised in table 1 together with T_g deduced from the ZFC and FC experiments.

The neutron scattering experiments were carried out using the 1T1 triple-axis spectrometer at the Orphee reactor at Saclay. The spectrometer was equipped with a Ge(III) monochromator and a graphite (002) analyser; a graphite filter was used to eliminate the higher-order contaminants of the scattered beam. The horizontal collimators were between 45′ and 55′, giving an energy resolution with $k_f = 2.662$ Å^{-1} of 0.24 THz (FWHM) as determined by the width in energy of the incoherent scattering from the sample. The sample was mounted in a 'Displex' continuous cycle cryostat with the magnetic easy axis in the scattering plane. A full description of the results and a thorough discussion will be published shortly. Here we shall concentrate on the evolution of the energy width of the diffuse quasi-elastic scattering as a function of temperature.

Constant-q energy scans were performed at $(h, 4\frac{1}{2}, 0)$ for $h = 0.5$, 0.45 and 0.40 at selected temperatures in the range $T = 7.8$ to 200 K. The results were analysed using a standard procedure where the observed line profiles are fitted by least-squares refinement to the profiles obtained by a numerical folding of an assumed analytic cross section with the known resolution function. Good fits ($\chi^2 \leq 2$) were obtained by fitting the data to a broad and a narrow lorentzian component. The energy width of both lorentzians decreases with decreasing temperature until about $T = 120$ K, below which the widths

Table 1. Summary of characteristic frozen frequencies and the temperatures at which they have been measured by the various techniques. The error bars for the neutron scattering data are essentially those due to counting statistics and they do *not* include any estimates due to possible systematic errors in the analysis of the line shape. The two points denoted with asterisks are not taken into account in the fit to equations (1)–(3).

Experiment	T_g(K)	f(Hz)
FC	48.1	10^{-5}–10^{-4}
ZFC	48.8	10^{-2}
AC	50.6	24.2
AC	51.3	242
AC	52.4	2420
Neutron scattering*	78	$(1 \pm 2) \times 10^{10}$
Neutron scattering*	92	$(2.1 \pm 1.5) \times 10^{10}$
Neutron scattering	102	$(3 \pm 1) \times 10^{10}$
Neutron scattering	132	$(5.7 \pm 2) \times 10^{10}$
Neutron scattering	200	$(9.3 \pm 2) \times 10^{10}$

decrease rapidly with a dominating nearly resolution-limited quasi-elastic component below 90 K.

On the basis of the present experiments we cannot make a unique assignment of the different components. Most likely the sharp lorentzian corresponds to the S^{zz} fluctuations and the weaker and broader lorentzian to the transverse fluctuations. We restrict our attention to the longitudinal fluctuations and assume that $\langle S^z(0)S^z(t)\rangle$ gives a measure of a characteristic spin relaxation time, similar in its physical meaning to that found by the temperature maximum in AC measurements. We therefore add to table 1 these characteristic times and the temperatures at which they are measured.

We turn now to explore possible functional dependence for the experimental data of table 1. The relaxation fits most often tried are the Vogel–Fulcher law (e.g. Tholence 1984)

$$\tau = \tau_0 \exp(E_a/k(T_g - T_0)) \tag{1}$$

a power law (e.g. Bontemps and Rajchenbach 1984)

$$\tau = \tau_0(1 - T_0/T_g)^{-\nu z} \tag{2}$$

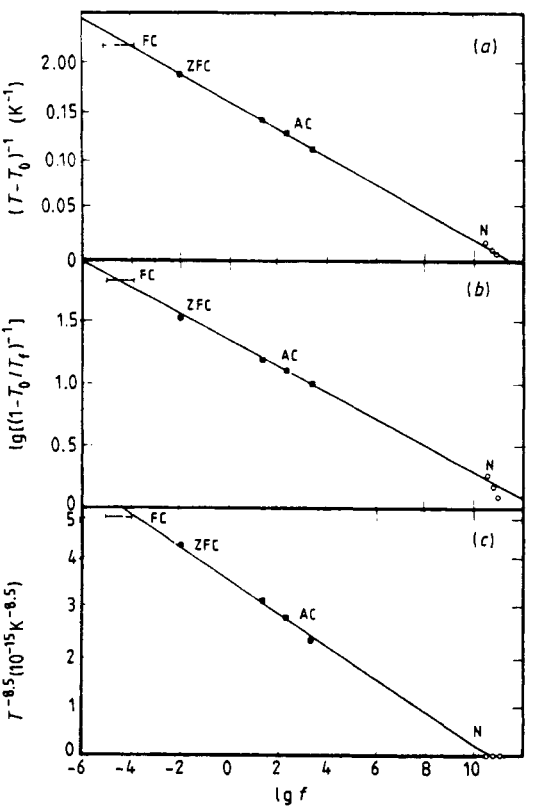

Figure 1. Characteristic frozen frequencies and the temperature at which they have been measured by magnetisation, AC susceptibility and neutron (N) scattering techniques. The Vogel–Fulcher law is fitted in figure 1(*a*), a power law in figure 1(*b*), and a generalised Arrhenius law in figure 1(*c*). The fitting parameters for which the straight line have been drawn are as follows. (*a*) $\tau_0 = 2.5 \times 10^{-12}$ s; $E_a/k = 170$ K; $T_0 = 43.5$ K. (*b*) $\tau_0 = 10^{-13}$ s; $\nu z = 9.5$; $T_0 = 47.4$ K. (*c*) $\tau_0 = 10^{-11}$ s; $\nu z = 8.5$; $T_0 = 0$.

and a 'generalised' Arrhenius law (e.g. Binder and Young 1984)

$$\tau = \tau_0 \exp(bT^{-\nu z}) \tag{3}$$

where ν is the critical exponent which describes the growth of the correlation length and z is the dynamic exponent which describes the slowing down of the relaxation. In figures 1(a), 1(b) and 1(c) we demonstrate the success of each model in describing the experimental data. The relevant fitting parameters are denoted in the figures. The range of time scales presented in our experiments is exceptional. (The only other example for which data are available for a similar range is CuMn (Tholence 1980, Murani *et al* 1981, Malozemoff and Imry 1981)). We therefore expect to eliminate one or more of the above laws. Quite surprisingly the fit to all three models is quite reasonable. It is true that the fit to equation (1) seems to be better than the other fits in the THz regime but, on the basis of the present data and due to possible systematic errors in the analysis of the neutron scattering line shape, we cannot exclude either the power law (which implies critical slowing down at a finite temperature) or the generalised Arrhenius law ($T = 0$ transition). More data points and a better energy resolution seem to be essential for experimental differentiation between existing models for the slowing-down process.

We compare our results with recent Monte Carlo (MC) experiments carried out by Ogielski and Morgenstern (1984) on a new, very fast, special-purpose computer at Bell Laboratories. They studied the relaxation times in a nearest-neighbour Ising system with random interactions distributed independently on each lattice site. Their relaxation times are fitted quite nicely with all the above three models and they reflect similar tendencies (unusually large values for z and a larger T_0 in the power law as compared to the Fulcher law).

We conclude with two comments. (i) The parameters obtained for the Vogel–Fulcher law, figure 1(a) yield $\alpha \equiv (T_g(24\text{ Hz}) - T_0)/T_g(24\text{ Hz}) \simeq 0.16$ which indicate, according to a criterion introduced by Tholence (1984), that Fe_2TiO_5 is comparable with other 'good' spin glasses (for CuMn, Mn aluminosilicate (Beauvillain *et al* 1984) and $Eu_{0.4}Sr_{0.6}S$ $\alpha \simeq 0.075$, 0.16 and 0.25 respectively). (ii) The FC maximum at 48.1 K can be incorporated in the fits assuming that it corresponds to a characteristic time of 10^4–10^5 s. This implies that the FC branch is *not* stable on this time scale in accordance with recent findings (Lundgren *et al* 1983, Bouchiat and Mailly 1984).

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