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Low-temperature study of the susceptibility in the anisotropic spin glass Fe_2TiO_5

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Abstract. We study the susceptibility of the anisotropic spin glass Fe_2TiO_5 below 1 K. An unusual contribution to the magnetic susceptibility is revealed by an increase in the susceptibility as temperature is decreased. This increase is attributed to 'loose' spins which, due to the frustration in this system, experience zero internal field and contribute a Curie–Weiss term to the susceptibility. This contribution is anisotropic in nature.

The insulator spin glass Fe_2TiO_5 has recently attracted experimental and theoretical interest due to its anisotropic magnetic properties (Atzmony *et al* 1979, Yeshurun *et al* 1984, Sherrington 1984, Yeshurun and Sompolinsky 1985). We report here on a low-temperature study of the magnetisation of Fe_2TiO_5 which reveals quite unusual magnetic behaviour. In particular, below 1 K we observe a paramagnetic-like contribution to the magnetic susceptibility. We attribute this behaviour to 'loose' or 'free' spins which experience a zero effective field even at very low temperatures. Similar phenomena have recently been observed in two other insulating spin glasses (Escorne and Mauger 1982, Fiorani *et al* 1984) and this might indicate that this behaviour is more general in spin glasses than was previously recognised. The phenomenon observed here is unique, however, in that the paramagnetic contribution exhibits anisotropic behaviour.

The system under study is a single crystal of the insulator Fe_2TiO_5 which exhibits a uniaxial anisotropy in its low-field AC susceptibility (Atzmony *et al* 1979). A cusp is observed at $T_g \simeq 50$ K in the 'longitudinal' susceptibility χ_c measured with the field along the crystal's $\boldsymbol{c}$ orthorhombic axis whereas χ_a and χ_b, measured with the field along the $\boldsymbol{a}$ and $\boldsymbol{b}$ axes respectively, exhibit smooth paramagnetic behaviour in the vicinity of T_g. Since no indication of long-range order was found in neutron diffraction experiments it was concluded (Atzmony *et al* 1979) that Fe_2TiO_5 is a spin glass with anisotropic characteristics. The spin glass features, which have been explored extensively by various techniques, resemble those of other 'good' spin glasses (Yeshurun *et al* 1985). The transverse susceptibilities deviate from a Curie–Weiss behaviour below 8 K where a small and broad maximum has been observed in the low-field magnetisation (Yeshurun and Sompolinsky 1985). It was suggested (Sherrington 1984, Yeshurun and Sompolinsky 1985) that this maximum might be an indication for a crossover to transverse freezing at $\simeq 8$ K, i.e. well below T_g where longitudinal freezing occurs. A further decrease of the

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transverse susceptibility is therefore expected at lower temperature. We demonstrate, however, that the enhanced contribution from the 'loose' spins dominates the low-temperature behaviour and masks any decrease of the susceptibility due to transverse freezing.

The magnetisation M was measured by the extraction technique using an apparatus provided with an adiabatic demagnetisation arrangement to cool the sample to 0.1 K. A copper coil in the nitrogen bath serves for the application of the measuring field. In the following we present magnetisation data as a function of field ($H \leqslant 2$ kOe) and temperature ($0.1\ \mathrm{K} \leqslant T \leqslant 10\ \mathrm{K}$). Data have been obtained after zero-field-cooling or field-cooling processes. The measurement process was started by zero-field cooling from room temperature but successive field-cooled runs were started at $10\ \mathrm{K} \simeq 0.2\ T_g$. The fact that we do not cool in field from above T_g affects, of course, the amount of induced irreversibility but does not prevent us from reaching quantitative conclusions, as discussed below.

Figure 1 exhibits values of $M(H)$ at $T = 0.15$ K with the field parallel to the ***a***, ***b*** and ***c*** directions. Data for this figure have been accumulated after a field-cooling process from 10 K and the initial values at $H = 0$ are the remanent magnetisations that have been picked up during this process. The most interesting aspect of figure 1 is the

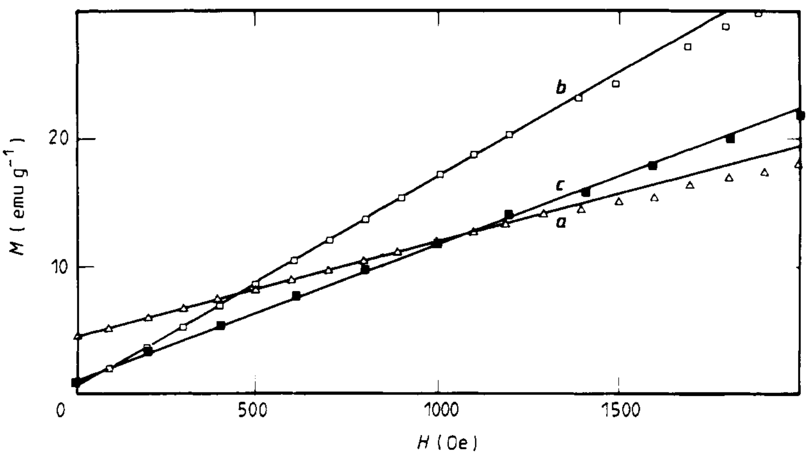

Figure 1. Magnetisation curves at 0.15 K measured with the field along the indicated (***a***, ***b*** or ***c***) directions.

difference in the initial slopes of the curves for $M(H)$ which demonstrate the anisotropy in the respective susceptibilities. Figure 1 also demonstrates that non-linear effects are negligible up to at least 800 Oe. This is the field we have chosen for the study of the temperature dependence of the susceptibility at low temperature.

The experimental procedure for obtaining data on $M(T)$ is the following. The sample is either field cooled or zero-field cooled from 10 to 0.1 K in small temperature intervals and at each temperature we measure the magnetisation $M(T)$ and the remanent magnetisation $M_R(T)$ 20 s after suppression of the field to zero. To avoid complications resulting from cooling being from 10 K we discuss in the following the temperature dependence of the *reversible* magnetisation $M_{rev}(T)$ defined as $M(T) - M_R(T)$. The temperature dependences of M_{rev} measured with the field along the ***a***, ***b*** or ***c*** direction

(M_a, M_b and M_c respectively) are exhibited in figure 2. Two features characterise the behaviour of $M_{rev}(T)$. First, the anisotropy ($M_b > M_a > M_c$) is preserved down to 0.1 K. Second, a remarkable *increase* of all components of the susceptibility is found when temperature is reduced below 1 K. This surprising increase seems to be superimposed on a susceptibility that varies linearly with temperature (the linear baselines are represented by the full lines in figure 2). This last observation brings us to suggest that in Fe_2TiO_5

$$\chi = \chi_0 + \alpha T + C/(T - \theta) \tag{1}$$

where $\chi_0 + \alpha T$ represents contributions from the non-magnetic matrix *and* from the frozen magnetic spins whereas $C/(T - \theta)$ is the contribution from 'paramagnetic' or 'loose' spins, i.e. weakly interacting spins for which the internal local fields cancel. According to this interpretation the differences between the raw data and the respective baselines of figure 2 are the contribution from the 'paramagnetic' spins. Indeed, the inverse of this difference plotted in figure 3 reveals a Curie–Weiss behaviour with θ = 0, 0.17 and 0.1 K and $C = 1.42 \times 10^{-4}$, 3.65×10^{-4} and 1.36×10^{-4} emu g^{-1} for the ***a***, ***b***

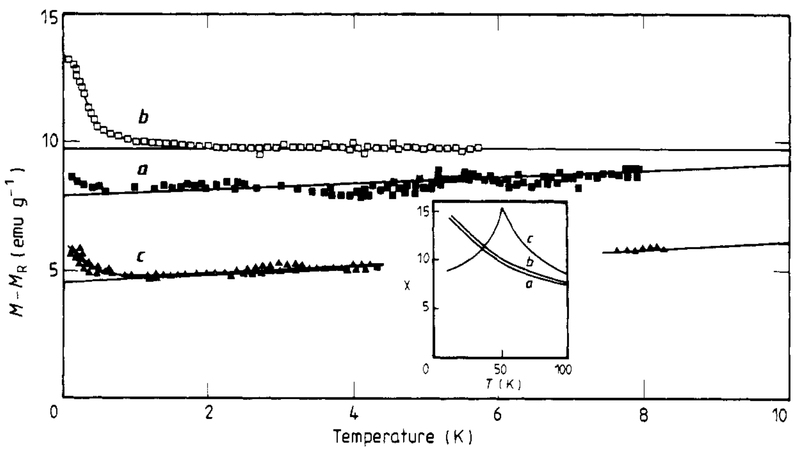

Figure 2. The reversible magnetisation as a function of temperature measured in 800 Oe with the field along the indicated directions. The full lines are the linear baselines, which denote contributions from the non-magnetic matrix and from interacting spins. The loose spins' contribution is the difference between the raw data and the full lines. Inset: AC susceptibility (in emu mol^{-1}) against temperature (in K) reproduced from Atzmony *et al* (1979).

and *c* directions respectively. In the high-temperature regime ($T \gg T_g$) the Curie constant, resulting from free Fe^{3+} ions, is found to be $C = 1.87 \times 10^{-2}$ emu g^{-1}. We therefore estimate the number of loose spins below 1 K to be $\simeq 10^{-2}$ of the total Fe^{3+} ions in the system. These spins experience a net zero internal field due to cancellation of conflicting information coming from their neighbours.

A similar paramagnetic-like increase in the spin glass susceptibility has already been observed in Monte Carlo experiments (Kirkpatrick 1977, Rapaport 1978, Bray *et al* 1978) and in two insulating spin glass systems: single crystals of $Cd_{1-x}Mn_xTe$ (Escorne *et al* 1981, Escorne and Mauger 1982) and a polycrystalline spinel $ZnCr_{1.6}Ga_{0.4}O_4$

(Fiorani *et al* 1984). In all of these cases 'loose' spins, which are easily flipped and therefore provide Curie behaviour, are invoked. Furthermore, it has been noted (Bray *et al* 1978) that flipping one of these spins may place another spin in zero field, so effectively the number of spins that may be readily aligned with an external field at low temperature is larger than the number counted at any instant of time. This, indeed, seems to be the case for Fe_2TiO_5 as well as for the other systems. The fact that this (predicted) phenomenon has been observed only in insulator spin glasses with dominating antiferromagnetic interactions is probably due to the frustration in these systems which makes cancellation of internal fields from neighbouring spins more likely to occur.

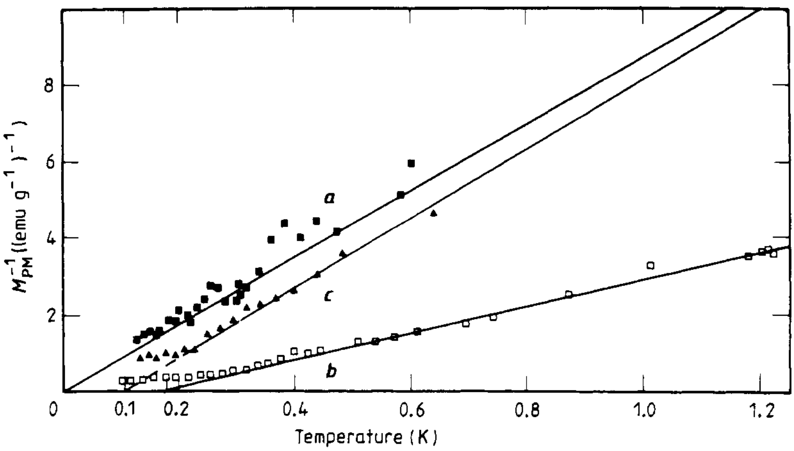

Figure 3. The inverse of the loose spins' contribution to the magnetisations as a function of temperature.

The anisotropy observed in this experiment (figure 3) in the loose spins' contribution to the susceptibility is the first observation of such a feature. At first glance it seems to be natural to this anisotropic system. However, the hierarchy of the 'paramagnetic' susceptibilities below 1 K ($\chi_b^* > \chi_c^* > \chi_a^*$ where χ^* is the loose spins' contribution deduced from figure 3) is different from the paramagnetic susceptibilities above T_g ($\chi_c > \chi_b > \chi_a$; Atzmony *et al* 1979). This might indicate either the failure of the phenomenological model (equation (1)) or a change in the anisotropic nature at low temperature. A further study of the low-temperature susceptibility will hopefully clarify this point.

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