Effect of Crystal-Field Anisotropy on Irreversible Phenomena in Spin-Glasses

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We study the angular dependence of the field-cooled susceptibility χ_{eq} of Fe_2TiO_5, a spin-glass with a strong uniaxial anisotropy, by rotating the measuring field $\vec{H}$ with respect to the cooling field $\vec{H}_c$. Unlike the behavior found in classical spin-glasses (i) the anisotropy in χ_{eq} *depends* on the direction of $\vec{H}_c$ and (ii) the irreversible magnetization does *not* rotate with $\vec{H}$. The de Almeida–Thouless line is identified via the field and temperature dependence of the anisotropy in χ_{eq}.

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Experimental study of anisotropy in spin-glasses has developed in two independent directions. In one, attention is focused on the magnetic properties of spin-glasses which, as a result of crystal-field anisotropy may exhibit susceptibility cusps only in certain lattice directions, thus giving rise to realization of Ising or *XY* spin-glass ordering.[1-6] The other direction deals with the macroscopic anisotropy which couples to the remanent magnetization and is independent of crystallographic orientation. The nature of this macroscopic anisotropy has been extensively studied in metallic spin-glasses[7-11] and it has been concluded that it is dominated by a directional (2π periodic) contribution arising from Dzyaloshinsky-Moryia (DM) interactions.[12] The effect of other anisotropic forces, such as crystal-field interactions, on the remanent susceptibility has not been explored. It is the purpose of the present work to report on a comprehensive experimental study of the anisotropic nature of irreversible phenomena in a spin-glass system with a strong crystalline anisotropy.

The system under study is a single crystal of the insulator Fe_2TiO_5 which exhibits a strong uniaxial anisotropy in its low-field ac susceptibility[1]: A sharp cusp is observed along the c orthorhombic axis at $T_g \simeq 53$ K whereas along the a and b axes a smooth paramagnetic behavior is found above and below T_g. Since no indication of long-range order was found in neutron diffraction and other measurements,[1] it was concluded that Fe_2TiO_5 is a spin-glass with anisotropic characteristics. The spin-glass nature has been attributed to frustration resulting from random distribution of Fe^{3+} and Ti^{4+} ions on the $(8f)$ and $(4c)$ sites; anisotropy has been attributed to crystal fields. In the present work we explore the anisotropy in the *field-cooled* susceptibility of Fe_2TiO_5 by rotating the magnetic field $\vec{H}$ with respect to the cooling field $\vec{H}_c$. Our study reveals several features which are very different from those observed in classical spin-glasses.[7-11] (i) The susceptibility exhibits a π-periodic anisotropy which *depends* strongly on the direction of the cooling field with respect to the c axis. (ii) The irreversible part of the magnetization is *fixed* in the c direction during rotation of $\vec{H}$ but π-periodic viscosity phenomena, resulting from the rotation, are observed. (iii) The anisotropy in the susceptibility persists well above T_g, but contributions from irreversible susceptibility, which depend strongly on field and temperature, vanish at the de Almeida–Thouless line[13] $T_c(H)$.

Anisotropic properties of Fe_2TiO_5 have been investigated via measurements of the magnetization on a vibrating-sample magnetometer (VSM) with a 2π-rotating sample holder. The sample is cooled in a field to the measuring temperature. With the field *on* and temperature stabilized to better than 0.1 K, the sample is rotated by ϕ relative to the magnetic field. (In the sample frame of reference, ϕ is the angle between the cooling field $\vec{H}_c$ and the measuring field $\vec{H}$.) We then measure the magnetization as a function of ϕ. The total sample magnetization is customarily written as

$$\vec{m} = \overleftrightarrow{\chi} \cdot \vec{H} + \vec{M}_{irr}. \quad (1)$$

where $\overleftrightarrow{\chi}\cdot\vec{H}$ and $\vec{M}_{irr}$ are the reversible and irreversible magnetization, respectively. We stress two features of Eq. (1) which are relevant to our study. (a) The susceptibility $\overleftrightarrow{\chi}$ is usually isotropic, i.e., $\chi_{\alpha\beta}=\chi\delta_{\alpha\beta}$, but for Fe_2TiO_5 it exhibits uniaxial anisotropy which dominates the anisotropic behavior. (b) The irreversible magnetization $\vec{M}_{irr}$ is *not* interchangeable with the remanent magnetization $\vec{M}_R$ obtained after switching off the field. In fact, we find (Fig. 1, inset) that for a constant temperature, $M_R(H)$ *saturates* whereas $M_{irr}(H)$ *vanishes* in strong enough fields.

Figure 1 exhibits the measured magnetization M as a function of ϕ for a cooling field $H_c=2$ kOe and temperature 4.2 K. In Fig. 1(a) the cooling field was applied along the c axis ($\phi=0$) and then ϕ was changed at a rate of 0.2 rpm. Several features characterize $M(\phi)$. First, the measured magnetization *increases* when the field is rotated from $\phi=0$ in either sense. Second, $M(\phi)$ reaches a pronounced minimum for $\phi=180°$. Finally, π-periodic, time-dependent phenomenon are observed. At lower rates of revolutions relaxational effects seem to be less and less important and the two curves tend to coincide.

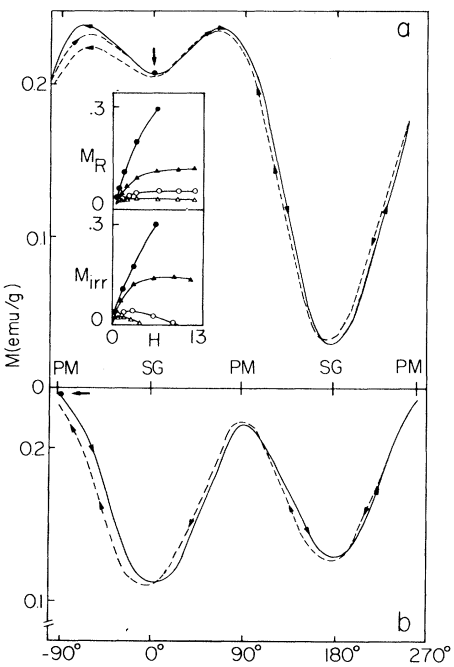

FIG. 1. Angular dependence of the magnetization measured after cooling the sample in 2 kOe. (a) $\vec{H}_c$ is along a spin-glass axis ($\phi=0°$). (b) $\vec{H}_c$ is along a paramagnetic axis ($\phi=-90°$). The solid arrows point to the value of the magnetization after the cooling process. The light arrows on the solid and broken lines indicate the order of measurements. Note the $\sim\pi/4$ segments for which the broken line (data taken later) is above and below the solid line. Inset: Remanent and irreversible magnetization as a function of field for 4.2 K (closed circles), 20 K (closed triangles), 32 K (open circles), and 40 K (open triangles).

The general shape of $M(\phi)$ is very different when the cooling field is applied perpendicular to the c axis ($\phi=-90°$) as in Fig. 1(b). Now $M(\phi)$ has a w shape with minima and maxima which correspond to states with $\vec{H}$ along the c and a (or b) axes, respectively. Note that $M(\phi)$ *decreases* when the field is rotated from $\phi=-90°$ in either sense. With a rate of 0.2 rpm we observe, as in the previous case, a π-periodic viscosity which disappears at slower revolution rates.

In Fig. 2 we exhibit $M(\phi)$ data for $H_c=5$ kOe parallel to the c axis ($\phi=0$) after a cooling process to temperature T (4.2 K $\leqslant T \leqslant$ 47 K). The effect of temperature on the general shape of $M(\phi)$ is twofold. First, the initial slope ($dM/d\phi$ at $\phi=0$) is positive at low temperature, decreases with temperature to zero at $T\simeq30$ K, and then turns to be negative at higher temperatures. Second, and the most important effect, the depth of the minimum around $\phi=180°$ decreases gradually with temperature. Around 30 K, the minimum is replaced by a maximum which increases gradually until $M(0)=M(180)$ and a $\cos^2\phi$ shape is observed. This fi-

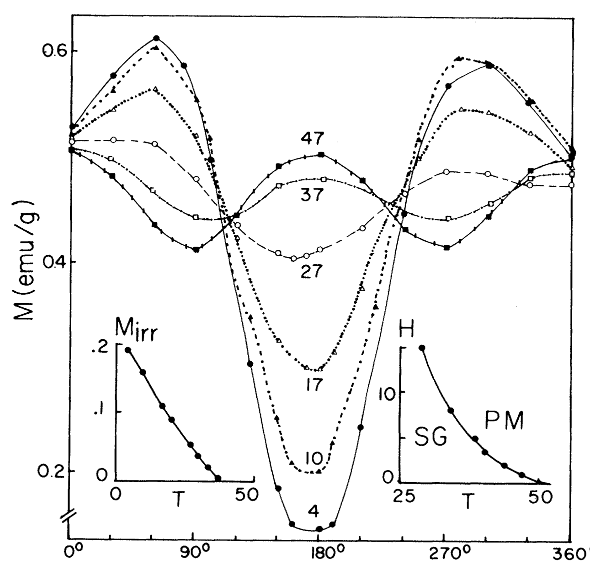

FIG. 2. Angular dependence of the magnetization measured after cooling the sample in a field $H_c=5$ kOe to temperature T (4 K $\leqslant T \leqslant$ 47 K). $\vec{H}_c$ is along a spin-glass axis ($\phi=0°$). Inset: $M_{irr}(T)$ for $H_c=5$ kOe (left) and the de Almeida–Thouless line for Fe_2TiO_5 (right).

nal shape persists up to at least $T = 2T_g$.

We turn now to discuss the gross features of Figs. 1(a) and 2, pointing out the main factors which affect $M(\phi)$ for $\vec{H}_c$ along the c axis. The reversible anisotropic susceptibility of Fe_2TiO_5 has two components, $\chi_\parallel$ and $\chi_\perp$, which are the susceptibilities measured parallel and perpendicular to the c axis, respectively. The magnetization, *measured along the applied field H,* is

$$M = \chi_\perp H + (\chi_\parallel - \chi_\perp) H \cos^2\phi + M_{\rm irr} \cos\phi, \quad (2)$$

where we take $\phi = 0$ to be along the c axis. The angular dependence of M is thus determined by two factors: the irreversible magnetization $M_{\rm irr}$ and the susceptibility *difference* $\chi_\parallel - \chi_\perp$. $\vec{M}_{\rm irr}$, resulting from the field-cooling process, is parallel and antiparallel to $\vec{H}$ for $\phi = 0°$ and $\phi = 180°$, respectively. It is therefore apparent that $M(0) - M(180) = 2M_{\rm irr}$. The changes in the depth of the minima therefore reflect the temperature dependence of $M_{\rm irr}$. On the other hand, the initial slope reflects the temperature dependence of the anisotropy in the reversible susceptibility. At low temperature $\chi_\parallel < \chi_\perp$, whereas above 30 K $\chi_\parallel > \chi_\perp$. Apparently, this affects the sign of the $\cos^2\phi$ term and therefore the initial slope of $M(\phi)$.

Equation (2) provides a basis for a quantitative analysis of the experimental data. The raw data were least-squares fitted by Eq. (2) with $\chi_\perp$, $\chi_\parallel$, and $M_{\rm irr}$ as parameters. The parameters $\chi_\perp(H,T)$ and $\chi_\parallel(H,T)$ obtained from this procedure are consistent with our measurements of the susceptibility along the a (or b) and c axes, respectively. Interesting results are obtained for $M_{\rm irr}(H,T)$. For a constant field $M_{\rm irr}(T)$ decreases with temperature (Fig. 2, inset) and *vanishes* at a temperature $T_c(H)$ above which $M(\phi)$ exhibits pure $\cos^2\phi$ behavior. Similarly, for constant temperature $M_{\rm irr}(H)$ increases, reaches a maximum, and then *decreases quite sharply to zero* (Fig. 1, inset). These results can be best understood in terms of the de Almeida–Thouless (AT) line[13] in the H-T phase diagram for spin-glasses. For Ising spin-glasses the AT line is a line of phase transitions from spin-glass to paramagnetism which are characterized by the vanishing of irreversible responses.[14] The irreversible characteristics of Fe_2TiO_5 are reflected in $M_{\rm irr}(H,T)$. We therefore take $M_{\rm irr} = 0$ as an experimental criterion for being above the AT line and by extrapolating $M_{\rm irr}(T)$ to zero (left-hand inset, Fig. 2), we are able to identify $T_c(H)$. The results of this extrapolation procedure are summarized in Fig. 2 (right-hand inset) with an AT-like line for Fe_2TiO_5 for which we find a critical exponent of 1.6 ± 0.1 in accordance with the theoretical predictions. We note that strikingly similar AT lines are found experimentally for *isotropic* systems.[15] For those systems theory[17] predicts a very different critical line; the observed AT line has been interpreted[5] as a *crossover* line from a weak (and undetectable) to a strong (and measureable) longitudinal irreversible response. Strong anisotropy, however, is expected to induce Ising-like *phase transitions,* the loci of which are described by the AT line.[4-6] The observed AT line in Fe_2TiO_5 is therefore consistent with theoretical predictions for Ising-like spin-glasses.

In the least-squares analysis of Eq. (2) we have also considered the possibility of a fourth parameter, namely, the angle θ which describes a rigid rotation of $\vec{M}_{\rm irr}$ with respect to $\vec{H}_c$. For such rigid rotations, which have been demonstrated experimentally in classical spin-glasses,[7-11,17] we should replace the last term in Eq. (2) by $M_{\rm irr}\cos(\phi - \theta)$, where θ is a function of ϕ and of the ratio $x = K/HM_{\rm irr}$, with K the anisotropic energy. Our least-squares analysis, however, yields $\theta = 0$, namely, that $\vec{M}_{\rm irr}$ is *fixed* in the c direction. The origin of this behavior lies in the behavior of the *reversible* susceptibility. For classical spin-glasses the reversible susceptibility is *isotropic,* i.e., spin-glass characteristics are not limited to certain lattice directions. Therefore, $\vec{M}_{\rm irr}$ can be (rigidly) rotated in any direction by overcoming the DM anisotropic energy. On the other hand, for Fe_2TiO_5, because of anisotropic crystal fields, reversible spin-glass characteristics and, as a result, irreversible characteristics are limited to the c direction. The term "rigid rotation" is therefore inapplicable for Fe_2TiO_5 and $M(\phi)$ is described as in Eq. (2).

The shape of $M(\phi)$ in a paramagnetic cooling process [Fig. 1(b)] might be interpreted by use of similar arguments as for the spin-glass cooling process. Since $\vec{H}_c$ is perpendicular to the c axis ($\phi = 90°$) no irreversible magnetization is induced along this axis. From the spin-glass point of view this is a *zero-field–cooled* (ZFC) process. Indeed, by rotating the field to $\phi = 0$ we obtain the spin-glass ZFC susceptibility value which, at 4.2 K, is smaller than the paramagnetic susceptibility. As a result of the ZFC process, irreversible contributions in Eq. (2) are negligible and therefore $M(\phi)$ in Fig. 1(b) is expected to exhibit $\cos^2\phi$ behavior. The small difference between the two minima which are observed at low temperature [Fig. 1(b)] suggests, in analogy with the behavior exhibited in Fig. 2, that transverse freezing occurs in Fe_2TiO_5 despite the absence of susceptibility anomaly (Fert *et al.*[4] have recently reached a similar conclusion via susceptibility measurements on rare-earth–based alloys). We cannot exclude, however, a different interpretation of the difference between the wells, namely, that small isothermal irreversible magnetization which is induced by the field as the field is rotated tends to slightly distort the $\cos^2\phi$ shape.

Finally, we turn to discuss the viscosity phenomena which are observed during rotation (Fig. 1). Time-dependent phenomena during rotations in classical spin-glasses have been attributed[11] to redefinition of the direction of the anisotropy axis with respect to $\vec{H}_c$. This is apparently not the case for Fe_2TiO_5. We suggest a simple mechanism which explains relaxation phenomena in the present experiment. As we have discussed above, the irreversible contribution to the magnetization in Fe_2TiO_5 remains fixed along the spin-glass axis, parallel to H_c. Rigid rotations[7–10, 14] of $\vec{M}_{irr}$ due to the rotation of the field $\vec{H}$ are therefore prevented. Instead, *the rotation of* $\vec{H}$ *with respect to* $\vec{M}_{irr}$ *is effectively equivalent to switching off and on the field along the spin-glass axis.* As a result, viscosity phenomena[18] are induced; magnetization slowly decreases during half the cycle and increases in the other half. By slowing down the rate of the change in the effective field (slower revolution rate) the viscosity can be practically eliminated.

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