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Transverse ordering in anisotropic spin glasses

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The low-field susceptibility along the easy axis of the spin glass Fe_2TiO_5 exhibits a sharp cusp at $T_g = 48$ K, whereas the susceptibility in transverse directions exhibits a broad maximum around 6 K. In all directions irreversibility appears already below T_g. Transverse irreversibility, however, is weaker by an order of magnitude than the longitudinal one. This behavior is discussed in terms of a model of a spin glass which has a weak random anisotropy in addition to a uniform easy axis.

Recent mean-field calculations have predicted a rather rich phase diagram for spin glasses (SG) with uniaxial anisotropy.[1-3] If the anisotropy field $D > 0$ is weak compared to the exchange field J, two successive transitions are expected. As the temperature is decreased, spin components along the easy ("longitudinal") axis freeze at T_g. Then, at a lower critical temperature T_2 the transverse components freeze as well. This second transition does not exist for sufficiently strong anisotropy. Indeed, extensive experimental work on various spin-glass systems[4,5] has demonstrated the absence of transverse freezing in spin glasses with strong uniaxial anisotropy. However, for spin glasses with moderate values of D it was found[5] that transverse freezing occurs at the *same* temperature as the longitudinal one, in clear disagreement with the theoretical predictions. In order to understand the origin of this behavior we have studied the characteristics of the transverse ordering in the uniaxially anisotropic spin glass Fe_2TiO_5.

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.[6,7] A sharp cusp is observed in the susceptibility χ_L measured with the field along the c orthorhombic axis at $T_g \simeq 50$ K, whereas along the a and b axes a smooth paramagnetic behavior is found in the susceptibility χ_T in the vicinity of T_g. Since no indication of long-range order was found in neutron diffraction and other measurements it was concluded[6] that Fe_2TiO_5 is a uniaxially anisotropic spin glass. The spin-glass nature is probably due to frustration resulting from random distribution of Fe^{3+} and Ti^{4+} ions on the $(8f)$ and $(4c)$ sites; anisotropy is due to crystal fields. In the present work we explore the zero-field-cooled (ZFC) and the field-cooled (FC) susceptibilities of Fe_2TiO_5 along the a,b, and c axes *down to 2* K. Our study reveals new features of the magnetic susceptibility of Fe_2TiO_5. (i) *At low temperature,* the *transverse* susceptibility χ_T deviates significantly from a paramagnetic behavior and exhibits a broad maximum around 6 K. (ii) Irreversible phenomena characteristic of a spin-glass order appear in *both* the longitudinal and transverse susceptibility *already in the vicinity of* T_g. (iii) Transverse irreversibility, however, is an order of magnitude weaker than the longitudinal one.

These results suggest that the appearance of transverse and longitudinal irreversibility at the same temperature is caused by a mechanism which *couples weakly* transverse and longitudinal order. Once transverse order is induced by this coupling at T_g, no further transition at a lower temperature is expected. However, if this perturbation is much weaker than the uniaxial anisotropy D and D *is much less than the exchange* J, one expects a crossover from a weak transverse order at $T \lesssim T_g$ to a strong one at $T < T_2$. The observed maximum of χ_T at $T = 6$ K might be an indication of such a crossover, although it might also be just the result of the gradual increase of the longitudinal freezing at low temperature. Two possible coupling mechanisms are random anisotropy and external magnetic field in a transverse direction. These mechanisms have been investigated theoretically using a model of a uniaxially anisotropic Heisenberg spin glass with the additional perturbation of random Dzyaloshinsky-Moriya-type anisotropy[8] as well as a transverse field. It is shown that either perturbation induces transverse order at T_g. However, the observed field dependence of the transverse irreversibility in Fe_2TiO_5 is more consistent with the predicted effect of a weak random anisotropy.

Two of the main features of spin glasses are the cusp in the nonequilibrium susceptibility and the appearance of two susceptibility branches which reflect reversible and irreversible magnetic responses.[9] These features have been investigated for the insulator Fe_2TiO_5 by using a SQUID susceptometer. The experimental procedure is the following. The sample is cooled in zero field to 2 K, a field H (10 Oe $\leq H \leq 700$ Oe) is applied and the ZFC susceptibility branch is measured up to $T \gtrsim 80$ K. Then, without changing the field, the sample is cooled to 2 K and the FC susceptibility branch is measured. Figure 1 exhibits ZFC and FC susceptibility branches (open and full symbols, respectively) measured with a field $H = 30$ Oe. The squares and circles represent data taken with the field H parallel to the c axis (χ_L) and to the b axis (χ_T), respectively. The ZFC branch of χ_L peaks at $T_g \simeq 48$ K. A broad maximum is observed in χ_T at $T \simeq 6$ K. Irreversibility, characterized by the difference between the FC and ZFC branches, is found, *for both* χ_L *and* χ_T, at $T < T_g$. We denote by Δ_L and Δ_T the differences between the FC and ZFC branches of χ_L and χ_T, respectively. From Fig. 1 it is clear that $\Delta_T << \Delta_L$. The ratio Δ_L/Δ_T is temperature dependent as shown in Fig. 2. Note that this ratio increases significantly as temperature is increased towards T_g. Close to T_g, however, where Δ_L and Δ_T reach the experimental resolution, the ratio cannot be determined unambiguously. Curves similar to those represented in Fig. 1 have been taken for fields up to 700 Oe. The effect of the field is twofold. First, it causes Δ_L and Δ_T to vanish at temperature $T_c(H) < T_g$. The line $T_c(H)$ resembles other experimental lines[9,10] which have been associated with the theoretical prediction of de Almeida and Thouless.[11] The second effect of the field is to

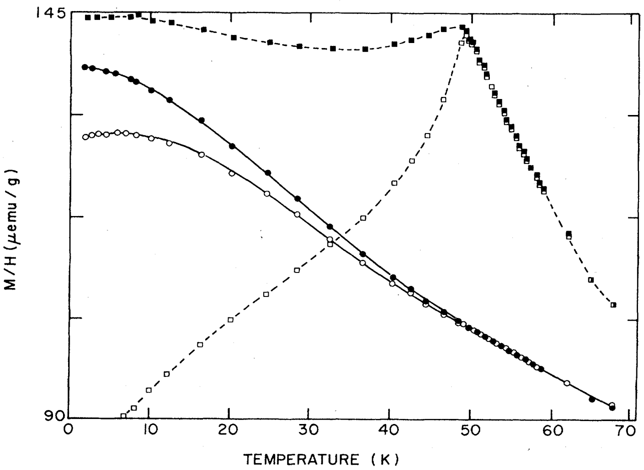

FIG. 1. Magnetization branches for Fe_2TiO_5 measured in a field $H=30$ Oe. The squares and the circles represent data taken with H along the c and the b axes, respectively. Open and full symbols represent ZFC and FC branches, respectively.

reduce the magnitude of the irreversibilities Δ_L and Δ_T, but we find that the ratio Δ_L/Δ_T does not vary significantly with the field.

In order to understand the origin of the weak transverse ordering near T_g we have studied the following spin-glass model

$$H=-\frac{1}{2}\sum_{\langle ij\rangle} J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j - D\sum_i (S_i^z)^2 - \frac{1}{2}\sum_{\langle ij\rangle}\mathbf{S}_i\cdot\overline{K}_{ij}\mathbf{S}_j \ . \tag{1}$$

The exchange interaction J_{ij} and the anisotropic interaction matrix $K^{\mu\nu}_{ij}$ are independent random variables with a Gaussian distribution around zero and variances $[J^2_{ij}]=J^2/z$, $[(K^{\mu\nu}_{ij})^2]=K^2/z$, z being the number of nearest neighbors. The spins $\mathbf{S}_i$ are m-component classical vectors satisfying $\sum(S_i^\mu)^2=m$, and $D>0$. We have assumed for simplicity a nonlocal anisotropy. However, our results apply also to local anisotropy which couples randomly the various spin components. Actually, in the insulator Fe_2TiO_5, it is likely that a random distribution of *local* crystal fields is responsible for this effect.

We assume in the following that the system undergoes a spin-glass phase transition at a finite temperature T_g (even with zero K) and furthermore that the spin-glass phase has the same qualitative structure as the mean-field transition. In such a case the ordered phase can be described[12] by the order parameter $Q^{\mu\nu}_{\alpha\beta}(i)$, the thermal average of which is

$$\langle Q^{\mu\nu}_{\alpha\beta}(i)\rangle = [\langle S^{\mu}_{\alpha,i}S^{\mu}_{\beta,i}\rangle] = q^{\mu}_{\alpha\beta}\delta^{\mu\nu} \ , \tag{2}$$

where α and β denote two copies of the system. The overlaps $q^{\mu}_{\alpha\beta}$ have a distribution of values which can be parametrized by monotonically increasing functions $q^{\mu}(x)$, $0\leq x\leq 1$. Physically, the cusp in the nonequilibrium magnetic susceptibility in the μ direction is related to $q^{\mu}(1)$. The difference Δ^{μ} between the equilibrium and nonequilibrium susceptibilities is proportional to $q^{\mu}(1)-\int_0^1 q^{\mu}(x)\,dx$. To describe the properties of the system near T_g, we use the replica formalism to study an effective Ginzburg-Landau-Wilson (GLW) free-energy functional, which is of the form[13]

$$F(Q_{\alpha\beta}(i))=\frac{1}{2}\sum_i\sum_{\substack{\alpha,\beta\\ \mu,\nu}}[r_0(Q^{\mu\nu}_{\alpha\beta})^2-\delta r^{\mu\nu}Q^{\mu\mu}_{\alpha\beta}Q^{\nu\nu}_{\alpha\beta}+|\nabla Q^{\mu\nu}_{\alpha\beta}|^2] \tag{3}$$

plus isotropic terms which are higher order in $Q_{\alpha\beta}$. The sum over α and β are from 1 to n, and the limit $n\rightarrow 0$ has to be taken. The reduced-temperature r_0 is proportional to T/T_0-1, where $T_0\sim J+O(D/J,K^2/J)$. The leading effect of the anisotropy is to induce the perturbation

$$\delta r^{\mu\nu}=D/J\delta^{\mu z}\delta^{\nu z}+K^2(m-1)/2J \ . \tag{4}$$

From Eq. (4) it is evident that the inclusion of K does not change the *critical* properties of the system. In the absence of anisotropy (i.e., $\delta r^{\mu\nu}=0$) all $m(m-1)$ components of $Q_{\alpha\beta}$ are simultaneously critical. Turning on the perturbation $\delta r^{\mu\nu}$ of (4) leaves *only one* critical field $q^0_{\alpha\beta}$, given by

$$q^0_{\alpha\beta}=\sum_\mu q^\mu_{\alpha\beta}\langle\lambda_0|\mu\rangle \ ,$$

where $\langle\lambda_0|\mu\rangle$ is the eigenvector of the matrix $\delta r^{\mu\nu}$ with the smallest eigenvalue δr_0. Associated with this critical field is a reduced-temperature $r_0-\delta r_0$. By integrating out the rest of the modes one is left with an Ising critical behavior (for all values of $D>0$ and K). The role of K is to change the "direction" of the critical field which is $q^0_{\alpha\beta}=q^z_{\alpha\beta}$ in zero K. Thus, by diagonalization of $\delta r^{\mu\nu}$ it is readily seen that in the limit of $T\rightarrow T_g^-$,

$$q^x_{\alpha\beta}/q^z_{\alpha\beta}=f(K^2/DJ) \ ,$$

where $f(x)\propto x$ as $x\rightarrow 0$ and $f(x)\rightarrow 1$ as $x\rightarrow\infty$.

Since $\chi_T\simeq\chi_T(T=T_g)-q^x/T$, it is clear that the random anisotropy has a minor effect on the total χ_T near T_g as long as $K^2/DJ \ll 1$. On the other hand, Δ_T/Δ_L will be proportional to $f(K^2/DJ)$, and this implies in particular that *near* T_g,

$$\frac{\Delta_T}{\Delta_L}\propto\frac{K^2}{DJ} \tag{5}$$

in the limit of $K^2/DJ\rightarrow 0$. This should be valid in any dimension at which a SG phase of the structure described above exists. The above analysis applies to the vicinity of T_g. As T decreases, the tendency to induce transverse order

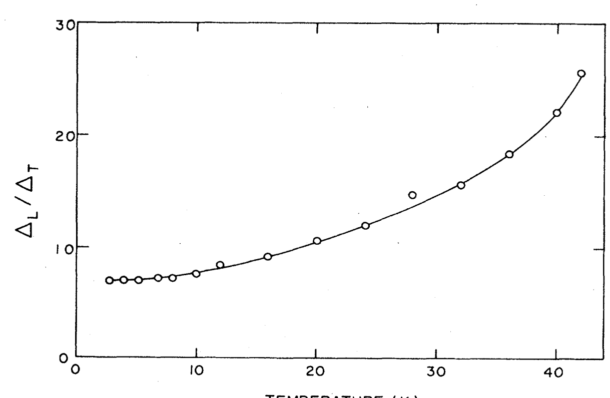

FIG. 2. The ratio Δ_L/Δ_T as a function of temperature for $H=30$ Oe. Δ_L and Δ_T are the difference between the FC and the ZFC branches in Fig. 1 for the longitudinal ($\mathbf{H}\parallel\hat{\mathbf{c}}$) and the transverse ($\mathbf{H}\parallel\hat{\mathbf{b}}$) measurements, respectively.

increases. Hence Δ_T/Δ_L is expected to increase; however, its value at low T depends on the nature of the low T phase. If, in the case $K=0$ the system has a transverse-ordering transition at $T_2 < T_g$, then as T decreases below T_2 the ratio Δ_T/Δ_L crosses over from Eq. (5) (in the case of $K^2/DJ \rightarrow 0$) to $(T_2-T)^{\beta_2}/(T_g-T)^{\beta_1}$, where β_1 and β_2 are the β exponents at the longitudinal and the transverse-ordering transitions, respectively, and eventually will be of order unity as $T \rightarrow 0$. These results are consistent with the calculation of the mean-field theory of the model (1). In particular, when $K^2/DJ << 1$ and $D/J << 1$, we find

$$\frac{\Delta_T}{\Delta_L} \sim \frac{K^2}{J(T_g-T+K^2/mJ-D/2J)} \tag{6}$$

valid for $T_2 < T < T_g$. Note that in this temperature regime T_g-T is of $O(D)$, which is consistent with Eq. (5). The result (6) applies only above the crossover regime which starts at $T \sim T_2+O(K^2/J)$. As mentioned in the beginning, qualitatively the experimental results displayed in Figs. 1 and 2 are consistent with the above predictions. However, a *microscopic* understanding of the source and magnitude of the random anisotropy in our system is still needed.

So far we have discussed the effect of a weak random anisotropy. We now discuss the affect of adding a term $-\mu H \sum_i S_i^x$ to Eq. (1) *with* $\mu H/J << 1$ *and* $K=0$. Qualitatively, the transverse internal fields induced by H at all temperatures couple to the frozen longitudinal moments below T_g and give rise to irreversibility in all directions. This is somewhat similar to the weak irreversibility induced below the Gabay-Toulouse[14] line in the direction of an external field in isotropic spin glasses. The important terms in the GLW replica Hamiltonian are

$$F=\frac{1}{2}\sum_{l}\sum_{\substack{\alpha\beta\\ \mu\nu}}\left(r_0(Q_{\alpha\beta}^{\mu\nu})^2-D(Q_{\alpha\beta}^{zz})^2+|\nabla Q_{\alpha\beta}^{\mu\nu}|^2-H^2Q_{\alpha\beta}^{xx}+w\sum_{\gamma}\mathrm{Tr}\,Q_{\alpha\beta}Q_{\beta\gamma}Q_{\gamma\alpha}+y_1\,\mathrm{Tr}\,Q_{\alpha\beta}^4+y_2(\mathrm{Tr}\,Q_{\alpha\beta}^2)^2\right) , \tag{7}$$

where the Tr refers to the spin indices. The quartic parts contain terms of the form $(Q_{\alpha\beta}^{xx})^2(Q_{\alpha\beta}^{zz})^2$. Since $q_{\alpha\beta}^x$ is nonzero, these terms give rise to a replica symmetry breaking in $q_{\alpha\beta}^x$, once it occurs in $q_{\alpha\beta}^z$. This is demonstrated by a mean-field solution of the model (7). Specializing to the regime $\mu H/J << 1-T/T_g << 1-T/T_2$ a straightforward solution of $\partial F/\partial Q_{\alpha\beta}=0$ yields $q_{\alpha\beta}^x \propto H^2/[r_0-y(q_{\alpha\beta}^z)^2]$. Note that in the mean-field approximation of (7) $T_g=T_0+D$, $T_2=T_0$, and hence in the above regime, $r_0 \sim D$ and $(q_{\alpha\beta}^z)^2 << r_0$. Thus, expanding in $(q_{\alpha\beta}^z)^2/r_0$ and using $\Delta_\mu \propto q^\mu(1)-\int_0^1 q^\mu(x)\,dx$ one obtains

$$\frac{\Delta_T}{\Delta_L} \propto (H/D)^2(1-T/T_g) \quad . \tag{8}$$

In short-range systems the dependence of Δ_T/Δ_L on H, T, and D may differ from Eq. (8); however, we expect a substantial variation of Δ_T/Δ_L as a function of H in all cases, since $\Delta_T \rightarrow 0$ as $H \rightarrow 0$. As mentioned above, no substantial dependence of Δ_T/Δ_L or H has been detected in our system implying that the measuring field is probably not the source for the appearance of transverse order in Fe_2TiO_5.

Discussions with P. Monod stimulated reexamination of the low-temperature transverse susceptibility. The possibility that random anisotropy or external field may be responsible for the observed single transition in the uniaxial spin glasses was raised in a discussion of one of us (H.S.) with A. J. Bray at the Orsay Spin Glass Meeting in January, 1984. This idea is also considered in a recent report by D. Sherrington. We thank D. Sherrington for helpful discussions and for sending us a report of his work prior to publication. The sample was kindly provided by B. Wanklyn, Oxford. This work is supported in part by the Fund for Basic Research administered by the Israel Academy of Sciences and Humanities.

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