Anisotropy of Spin-Glasses from Torque Measurements

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The anisotropy of *Cu*Mn spin-glasses was probed by torque measurements. In *Cu*Mn (20 at.%) the anisotropy is 2π periodic, increasing to its maximum when the spin system is rotated by π away from equilibrium. After a rotation by π around $\hat{x}$, the anisotropy energy remains at its maximum value during a subsequent rotation around $\hat{y}$ (perpendicular to $\hat{x}$), at least in a limited angle range. These properties are due to the existence of antisymmetric interactions.

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Magnetic,[1-3] NMR,[4] and ESR[5,6] measurements have revealed the existence of a macroscopic anisotropy energy in metallic spin-glasses. In most of the theoretical approaches[6-11] the spin system is supposed to rotate bodily (or quasi-bodily) and an anisotropy energy $E_A(\theta)$ is introduced in the macroscopic free energy. $E_A(\theta)$ is the energy needed to rotate the spin system (bodily) by an angle θ. Several specific features of $E_A(\theta)$ in *Cu*Mn and *Ag*Mn have been well established[2-6]:

(a) It has been known since 1961 that the anisotropy of *Cu*Mn and *Ag*Mn includes a directional (i.e., 2π periodic) contribution.[1] Most of the recent experiments have been interpreted by a combination of directional and axial terms, i.e.

$$E_A(\theta)=K_1(1-\cos\theta)+\tfrac{1}{2}K_2\sin^2\theta. \quad (1)$$

The axial term is introduced to explain why the equilibrium at π is less unstable than predicted for pure directional anisotropy, or even stable. However, recent susceptibility and torque measurements[12] have shown that this stabilization comes with time dependence and irreversibility effects and arises from rearrangements of the spins during large-angle rotations. For rigid rotations, at relatively small angles, the anisotropy appears to be purely directional.

(b) The anisotropy energy is independent of the magnitude of the remanent magnetization σ down to $\sigma=0$.[3,6] Consequently, it must cost the same energy to rotate around any axis.[7,10]

(c) The anisotropy of *Cu*Mn is enhanced by nonmagnetic impurities with strong spin-orbit coupling.[13] This led Fert and Levy[8,9] to ascribe the anisotropy to Dzyaloshinsky-Moriya (DM) type antisymmetric interactions due to spin-orbit scattering. In particular this explains the directional character.[9,10]

We now describe some remarkable properties of the anisotropy arising from DM interactions (some of these properties have already been described by Henley, Sompolinsky, and Halperin[10]). We consider a DM energy of the form

$$\sum_{ij}\vec{D}_{ij}\cdot(\vec{S}_i\times\vec{S}_j). \quad (2)$$

Since the vectors $\vec{D}_{ij}$ are randomly oriented,[8,9] the DM energy has a nonzero average only if correlations between the vectors $\vec{S}$ and $\vec{D}$ have been frozen in during the cooling. The assumption of rigid rotations of the spin system with respect to the $\vec{D}_{ij}$ lattice implies that there is no rearrangement of the spins to minimize the DM energy, as can be expected if the correlations arise from long-range arrangements involving a large number of spins. In practice, we know from our torque measurements that the rotations are nearly rigid in concentrated *Cu*Mn alloys at low temperature.

We first consider a rotation of the spins by θ out of the equilibrium state ($\theta=0$) around a given axis. This is equivalent to considering a rotation by $-\theta$ of the $\vec{D}$ vectors (lattice) with respect to the spins. The DM energy, Eq. (2), varies as

$$\sum_{ij}[R(-\theta)\vec{D}_{ij}]\cdot(\vec{S}_i\times\vec{S}_j). \quad (3)$$

The 2π periodicity of the rotation matrix $R(-\theta)$ leads to

$$E_A(\theta)=K(1-\cos\theta). \quad (4)$$

Since the system is isotropic K does not depend on the rotation axis.

We now consider, after a first rotation by θ around $\hat{x}$, a subsequent rotation by φ around $\hat{y}$ ($\hat{x}$ and $\hat{y}$ are perpendicular). By arguing, as above, from the 2π periodicity of the rotation matrix $R(\varphi)$, one finds

$$E_A(\theta,\varphi)=E_A(\theta,\varphi=0)+k(\theta)(1-\cos\varphi). \quad (5)$$

For $\varphi=\pi$, whatever θ, the product of the rotations is a rotation by π, which gives $E_A(\theta,\varphi=\pi)$

$= 2K$. We also know $E_A(\theta,\varphi=0)$ from Eq. (4), so that Eq. (5) can be written as

$$E_A(\theta,\varphi)=K(1-\cos\theta)+\tfrac{1}{2}K(1+\cos\theta)(1-\cos\varphi). \quad (6)$$

For a rotation around $\hat{x}$ by π (our experiments and Fig. 1) E_A increases as $(1-\cos\theta)$ from A to B and then is constant along BD.

The angular dependences predicted above are confirmed by torque measurements on 20-at.% and 4.7-at.% *Cu*Mn alloys (the capacitance torquemeter we used will be described elsewhere[14]).

After cooling the samples in a field along $\hat{z}$, we first rotate the remanent magnetization $\vec{\sigma}$ around $\hat{x}$ by rotating a field $\vec{H}$. θ_H and θ are the angles with $\hat{z}$ of $\vec{H}$ and $\vec{\sigma}$, respectively. We measure the x component of the torque acting on the sample:

$$\Gamma_x = dE_A/d\theta. \quad (7)$$

Moreover, the equilibrium condition for $\vec{\sigma}$ is written as

$$\Gamma_x = \sigma H \sin(\theta_H-\theta). \quad (8)$$

Since we know σ from independent measurements, Eq. (8) yields θ for a given experimental θ_H and so we know Γ_x and E_A as functions of θ. In an equivalent way we can also test a particular form of E_A by calculating the expected torque as a function of θ_H and comparing with the experimental curves, as we do in this Letter. The main features of our results (curve a in Fig. 2) agree with what is expected for directional anisotropy (dashed line), as already found in similar alloys.[12,15] To be more precise the torque curve is reversible and time independent for $\theta_H \lesssim 60°$ (i.e., $\theta \lesssim 47°$). Such a rigid-rotation range has also been found in other *Cu*Mn alloys and transverse susceptibility measurements have clearly shown that, in this range, the anisotropy is purely directional.[12] At larger angles there appear several effects which have already been seen in less concentrated alloys and ascribed to rearrangements of the spins.[12] However, these effects are much less pronounced in 20-at.% *Cu*Mn: The measured torque is only slightly time dependent, the torque curve is almost reversible, and it departs moderately from the calculated curve; the shifts of the equilibirum direction are very small. In view of the smallness of these effects we can reasonably consider that 20-at.% *Cu*Mn provides us with a good example of directional anisotropy with, even at large angles, quasirigid rotations.

We also probed the anisotropy during a rotation around $\hat{y}$ following a rotation by π around $\hat{x}$. Our aim is to drive the system along the constant-energy line BD in Fig. 1, which would give $\Gamma_y = 0$. However, from an experimental point of

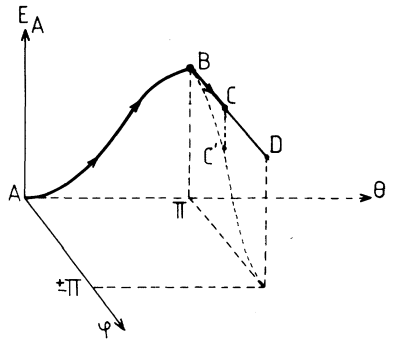

FIG. 1. Predicted variation of the anisotropy energy during successive rotations around $\hat{x}$ and $\hat{y}$ (see text).

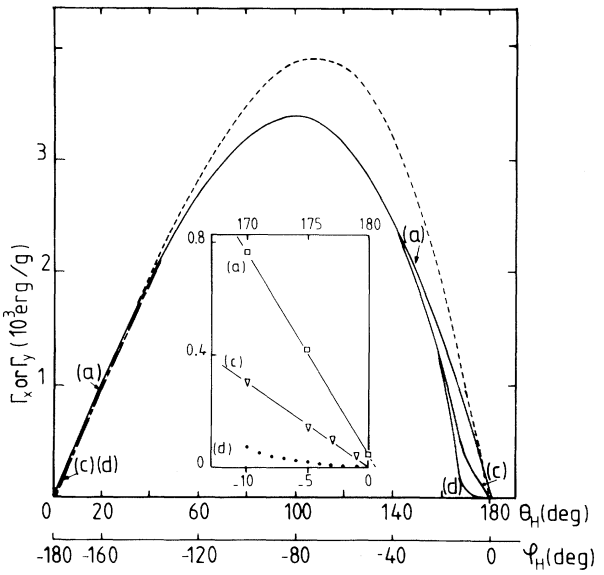

FIG. 2. Torque vs θ_H or φ_H for 20-at.% *Cu*Mn at 1.5 K in 6 kG (cooling field = 9.1 kG, $\sigma = 2.11$ emu/g). The inset is a blowup of the figure around $(\theta_H=\pi, \varphi_H=0)$. Curve a: Γ_x during a rotation around $\hat{x}$ from $\theta_H=0$ to $\theta_H \gtrsim \pi$. Curve d: Γ_y during a subsequent rotation around $\hat{y}$ from $\varphi_H=0$ to $\varphi_H \simeq -\pi$. Curve c: Γ_y during a rotation around $\hat{y}$ after an inversion. Dashed line: calculation from Eqs. (4), (7), and (8) with $K=3900$ erg/g. The values of K_θ', K_φ', and K_i' derived from the slopes of the curves a, d, and c near $\theta_H=\pi$ or $\varphi_H=0$ are $K_\theta' = -3100 \pm 50$ erg/g, $K_\varphi' \simeq 0$, $K_i' = -1510 \pm 50$ erg/g. Measurements in different fields give similar curves if the field is high enough to reverse $\vec{\sigma}$.

view, we drive the rotation of the remanent magnetization $\vec{\sigma}$ but we cannot control any rotation of the spin system around $\vec{\sigma}$. Such a rotation would lower the energy ($C \rightarrow C'$).

We find (curve *d* in Fig. 2) that for $|\varphi_H| \lesssim 10°$, Γ_y remains *extremely small*, as can be seen clearly in the inset. When $|\varphi_H|$ increases beyond 10°, Γ_y rises abruptly (with strong time-dependent effects) and, at about 30°, the curve *d* tends to the curve *a*. This abrupt increase of Γ_y is certainly due to a rotation of the spin system around $\vec{\sigma}$ (*CC'* line in Fig. 1). These results confirm the properties predicted by the theory (Fig. 1). However, because we have not been able to prevent a rotation around $\vec{\sigma}$, we could not probe the energy variation on the *BD* line very far from *B*.

We also reversed the magnetization by inverting the field and then came back to the initial direction by rotating around $\hat{y}$ (any axis of the x-y plane would be equivalent). This gives the curve *c* in Fig. 2, halfway between curves *a* and *d*. This suggests that, during the inversion, the spin system breaks into many blocks which rotate around different axes distributed in the x-y plane. During the subsequent rotation around $\hat{y}$ the torque is thus an average of the torques of curves *a* and *d*. Above about 10° the torque increases abruptly and the curve *c* tends to the curves *d* and *a*. This means that the blocks have rotated around their remanent magnetization to recompose a state simply rotated from the initial state.

The energy variation around $\theta = \pi$ or $\varphi = 0$ can be quantitatively characterized by the coefficients K_θ' and K_φ' defined as the second derivatives of E_A with respect to θ and φ, respectively (after rotation), and K_i' defined as the second derivative of E_A with respect to θ or φ after inversion. The values of K_θ', K_φ', and K_i' derived from the slopes of curves *a*, *d*, and *c* are given in the caption of Fig. 2 together with K. That $|K_\theta'|$ is slightly smaller than K is due to the stabilization by rearrangements already mentioned.

In 4.7-at.% *Cu*Mn at 1.5 K the spins rotate less rigidly than in 20-at.% *Cu*Mn, as indicated by the

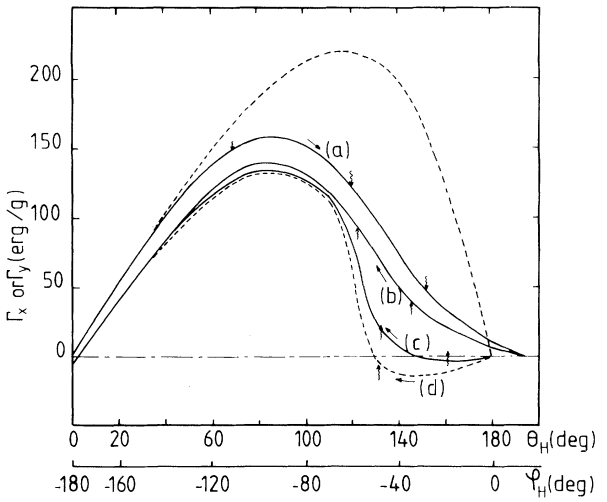

FIG. 3. As in Fig. 2 but for 4.7-at.% *Cu*Mn at 1.5 K in 1 kG (cooling field = 9.1 kG, $\sigma = 0.52$ emu/g). Curve *b*: Γ_x during a rotation around $\hat{x}$ from $\theta_H \gtrsim \pi$ to $\theta_H = 0$. Dashed line: calculation with $K = 220$ erg/g. Derived values of K_θ', K_φ', K_i': $K_\theta' = 34 \pm 5$ erg/g, $K_\varphi' = 40 \pm 5$ erg/g, $K_i' = 9 \pm 4$ erg/g. Wavy arrows indicate the direction of time-dependent drifts, when they exist.

definite irreversibility of the torque curves and the significant shifts of the equilibrium axes (Fig. 3, see also Ref. 12). However, the relative positions of the curves *a*, *c*, and *d* near π are the same as in 20-at.% *Cu*Mn. The only change is a general tendency to stabilization: After a rotation around $\hat{x}$, the equilibrium is still unstable with respect to a rotation around $\hat{x}$ (i.e., $K_\theta' < 0$) but it is quite stable with respect to a rotation around $\hat{y}$ (i.e., $K_\varphi' > 0$); after an inversion, the equilibrium is also stable (i.e., $K_i' > 0$). The stability of the reversed state increases with the temperature: at 4.2 K, K_θ', K_φ', and K_i' are positive.

The stabilization by nonrigid rotations can be qualitatively understood in the following picture. We suppose that the anisotropy triad[7] has rotated by θ_0 around $\hat{x}$ after a rotation of $\vec{\sigma}$ to $\theta \sim \pi$ (with some distribution of θ_0 in the sample). Thus the spins rotate with respect to the triad by only $\theta - \theta_0$, so that Eq. (6) is written as

$$E_A(\theta,\varphi) = K[1 - \langle\cos(\theta - \theta_0)\rangle] + \tfrac{1}{2}K[1 + \langle\cos(\theta - \theta_0)\rangle](1 - \cos\varphi). \quad (9)$$

This leads to

$$K_\theta' = -K\langle\cos\theta_0\rangle, \quad K_\varphi' = \tfrac{1}{2}K(1 - \langle\cos\theta_0\rangle) \quad (10)$$

which gives $K_i' = \frac{1}{2}(K_\theta' + K_\varphi') = \frac{1}{2}K(1 - 3\langle\cos\theta_0\rangle)$. Our experimental results are in qualitative agreement with Eq. (10) if one assumes that $\langle\theta_0\rangle$ progressively increases when the concentration decreases or the temperature increases.[16]

In conclusion, our torque experiments demonstrate some of the remarkable properties of the anisotropy in spin-glasses. The most striking

result is that, after a rotation by π around $\hat{x}$, the anisotropy energy remains at its maximum value during a subsequent rotation around $\hat{y}$, at least as long as an uncontrollable rotation around the axis of the remanent magnetization can be avoided. These properties (characteristic of tridirectional anisotropy) result from the existence of antisymmetric interactions as well as from the macroscopic isotropy of spin-glasses.[7,9,10] With respect to the problem of ESR in spin-glasses our results confirm the validity of the theoretical approach of Henley, Sompolinsky, and Halperin[10] and Legget.[11]

[1]J. S. Kouvel, J. Phys. Chem. Solids 21, 57 (1961).
[2]P. Monod, J. J. Prejean, and B. Tissier, J. Appl. Phys. 50, 7324 (1979).
[3]F. Hippert and H. Alloul, J. Phys. (Paris) 43, 691 (1982).
[4]H. Alloul, J. Appl. Phys. 50, 7330 (1979).
[5]P. Monod and Y. Berthier, J. Magn. Magn. Mater. 15–18, 149 (1980).
[6]S. Schultz, E. M. Gullikson, D. R. Fredkin, and M. Tovar, Phys. Rev. Lett. 45, 1508 (1980).
[7]W. M. Saslow, Phys. Rev. B 22, 1174 (1980), and Phys. Rev. Lett. 48, 505 (1982).
[8]A. Fert and P. M. Levy, Phys. Rev. Lett. 44, 1538 (1980).
[9]P. M. Levy, C. Morgan-Pond, and A. Fert, J. Appl. Phys. 53, 1168 (1982).
[10]C. L. Henley, H. Sompolinsky, and B. I. Halperin, Phys. Rev. B 25, 5849 (1982).
[11]A. J. Leggett, private communication.
[12]F. Hippert, H. Alloul, and A. Fert, in Proceedings of the Twenty-Eighth Conference on Magnetism and Magnetic Materials, Montréal, 1982 (to be published).
[13]J. J. Prejean, M. Joliclerc, and P. Monod, J. Phys. (Paris) 41, 427 (1980).
[14]A. Fert, to be published.
[15]T. Iwata, K. Kai, T. Nakamichi, and M. Yamamoto, J. Phys. Soc. Jpn. 28, 582 (1970).
[16]A. Fert and F. Hippert, to be published.

Many-Body Theory of Optical Bistability in Semiconductors

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A unified microscopic theory of the observed optical bistabilities in narrow-gap semiconductors, such as InSb and GaAs, is developed by calculating the complex, nonlinear dielectric function for arbitrary free-carrier concentrations from an integral equation for the polarization function.

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In 1979 dispersive optical bistability was observed in the narrow-gap semiconductors GaAs (Ref. 1) and InSb.[2] The driving optical nonlinearity in GaAs is connected with the ionization of the exciton, while the nonlinearity in InSb is mainly due to filling of the bands with free carriers. Recently, the observation of optical bistability has been reported also for Te.[3] In all these experiments a relatively small number of electron-hole (e-h) pairs is excited in the low-transmission state, while in the high-transmission state a relatively dense e-h plasma is generated. Thus, a microscopic theory of the optical bistability has to connect the low- and high-density regimes. The dominant correlations in the low-density regime are caused by the attractive e-h Coulomb interaction, which gives rise to the formation of excitons in GaAs. In this substance the exciton binding energy E_0 is—in bulk materials at least at low temperatures and in superlattice structures even at room temperature—larger than its broadening γ. In InSb the broadening always exceeds the binding energy E_0, so that the exciton resonance is not resolved. In the high-density regime the screening of the Coulomb interaction by intraband scattering is the most important process and is well described by the random-phase approximation (RPA). A consistent theory of the intermediate-density regime is rather difficult, especially for low temperatures and wide-gap semiconductors, where many-exciton effects become important.

In this Letter, we will neglect many-exciton effects and will combine consistently the processes

