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π-rotation minimum in spin-glasses

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For spin-glasses with large enough triad anisotropy of a uniaxial (as opposed to unidirectional) nature and an external field $\vec{H}$ not necessarily along the cooling field $\vec{H}_c$, we find a new energy minimum for the spin triad $(\hat{n},\hat{p},\hat{q})$, wherein it is rotated from the anisotropy triad $(\hat{N},\hat{P},\hat{Q})$ by 180°, about the axis midway between $\vec{H}$ and $\vec{H}_c$. Experimental realization of this state is discussed, as are ESR, torque, and transverse susceptibility measurements.

I. INTRODUCTION

When a spin-glass (SG) is cooled in a field $\vec{H}_c$, it goes into a complicated local equilibrium state.[1] Since the spins point in all three directions, one must specify any given spin in terms of an orthonormal reference triad, which we call the *spin triad* $(\hat{n},\hat{p},\hat{q})$.[2] We may take $\hat{n}$ to point along the remanence direction, but the orientation of $\hat{p}$ and $\hat{q}$ (about $\hat{n}$) is not directly observable. Because of interactions with the lattice, there is an anisotropy torque acting on the spin system if $(\hat{n},\hat{p},\hat{q})$ is rotated from its equilibrium orientation by an angle ψ, the torque being opposed to the axis of rotation $\hat{\psi}$.[3] This enables us to define an equilibrium orientation (with zero anisotropy torque) for $(\hat{n},\hat{p},\hat{q})$, which we call the *anisotropy triad* $(\hat{N},\hat{P},\hat{Q})$.[2] The anisotropy triad itself can rotate, but it does this only slowly, so that for many purposes it can be considered fixed.[2] If we assume this, then for the applied field $\vec{H}$ along $\vec{H}_c$, where $(\hat{n},\hat{p},\hat{q})$ coincides with $(\hat{N},\hat{P},\hat{Q})$, we see that $\hat{N}$ is along $\vec{H}_c$.

If $\vec{H}$ is not along $\vec{H}_c$, there will be a Zeeman torque tending to rotate $\hat{n}$ towards $\vec{H}$. It is thus natural to seek a ground-state solution wherein $\hat{n}$ lies in the $\vec{H}$-$\vec{H}_c$ plane. We refer to this as the *planar solution.* A ferromagnet with a single easy axis has the same equilibrium solution.[4]

Experimental studies, for a given transverse field, of how large a longitudinal field must be to cause a discontinuous flipping of the remanence, are in reasonable agreement with the predictions for a ferromagnet.[5] However, similar studies for the return flip of the remanence are not in such agreement.[5] This suggests that when the SG flips its remanence, it may go into a state which is not accessible to a ferromagnet, but is characteristic only of SG. We have found such a state theoretically, wherein $(\hat{n},\hat{p},\hat{q})$ is rotated from $(\hat{N},\hat{P},\hat{Q})$ by 180°, about the axis midway between $\vec{H}$ and $\vec{H}_c$. This state minimizes the Zeeman energy (i.e., the remanent magnetization points along $\vec{H}$) and locally minimizes the anisotropy energy. We have studied a number of its properties: the necessary material parameters for its realization, and how to produce the state when the material parameters are favorable; its ESR frequencies; and its expected behavior for torque and transverse susceptibility measurements. The return-flip data are somewhat clarified by the introduction of this new state (which we call the *π-rotation state).*

Before performing any detailed analysis, it would be worthwhile to discuss the physics behind the new solution. Although $\hat{n}$ is completely dependent upon the rotation $\vec{\psi}\equiv\psi\hat{\psi}$, one can alternately consider $\hat{n}$ and ψ as variables, subject to the constraint that $\psi\geq\cos^{-1}(\hat{n}\cdot\hat{N})$. If we choose the equal sign, then $\hat{n}$ and ψ are not independent, and we obtain the planar solution. However, if we choose the inequality, then we may vary $\hat{n}$ and ψ independently, subject only to the inequality. Variation of $\hat{n}$ minimizes the Zeeman energy (giving $\hat{n}$ along $\vec{H}$), and variation of ψ minimizes the anisotropy energy, giving $\psi=0$ and, for large enough uniaxial anisotropy, $\psi=\pi$. Since for $\vec{H}$ not along $\vec{H}_c$ we must have $\hat{n}\cdot\hat{N}<1$ if $\hat{n}$ is along $\vec{H}$, we see that the inequality eliminates the $\psi=0$ solution. All that remains, for large enough uniaxial anisotropy, is the $\psi=\pi$ solution.

II. STATICS

We now proceed to write down and minimize the energy density ϵ believed to be appropriate to SG's.[2,3,6] First, however, consider $(\hat{N},\hat{P},\hat{Q})$ fixed at a standard orientation, corresponding to a rotation $\vec{\Theta}=\vec{0}$. The rotation $\vec{\theta}\equiv\theta\hat{\theta}$ of $(\hat{n},\hat{p},\hat{q})$ from that standard orientation is then the same as the relative rotation $\vec{\psi}$ between $(\hat{n},\hat{p},\hat{q})$ and $(\hat{N},\hat{P},\hat{Q})$, and we may set $\vec{\psi}=\vec{\theta}$. Thus we write

$$\epsilon=\frac{m^2}{2\chi}-\vec{m}\cdot\left[\vec{H}+\frac{m_0}{\chi}\hat{n}\right]-K_1\cos\theta-\tfrac{1}{2}K_2\cos^2\theta \quad . \quad (1)$$

Here $\vec{m}$ is the magnetization, χ is the isotropic susceptibility, m_0 is the (zero-field) remanence, and K_1 and K_2 are the unidirectional and uniaxial triad anisotropy constants. Note that $\cos\theta=\frac{1}{2}(\hat{n}\cdot\hat{N}+\hat{p}\cdot\hat{P}+\hat{q}\cdot\hat{Q})-\frac{1}{2}$ for triad anisotropy, whereas $\cos\theta$ is replaced by $\hat{n}\cdot\hat{N}$ for single-axis anisotropy,[7] which is not considered in this paper. Minimizing ϵ with respect to $\vec{m}$, we obtain $\vec{m} = m_0\hat{n} + \chi\vec{H}$ in equilibrium. Placing this in (1), and eliminating constant terms which do not depend on the rotation, we obtain

$$\epsilon = -m_0\hat{n}\cdot\vec{H} - K_1\cos\theta - \tfrac{1}{2}K_2\cos^2\theta \ . \tag{2}$$

Expressing $\hat{n}$ in terms of $\hat{N}$, θ, and $\hat{\theta}$, we can rewrite Eq. (2):

$$\hat{n} = \hat{N}\cos\theta + \hat{\theta}(\hat{\theta}\cdot\hat{N})(1-\cos\theta) + (\hat{\theta}\times\hat{N})\sin\theta \ , \tag{3}$$

$$\epsilon = -m_0H[(\hat{H}\cdot\hat{N})\cos\theta + (\hat{H}\cdot\hat{\theta})(\hat{N}\cdot\hat{\theta})(1-\cos\theta) + \hat{H}\cdot(\hat{\theta}\times\hat{N})\sin\theta] - K_1\cos\theta - \tfrac{1}{2}K_2\cos^2\theta \ . \tag{4}$$

Minimization of ϵ with respect to θ gives, with $\tilde{K} \equiv K_1 + K_2\cos\theta$,

$$0 = \frac{\partial\epsilon}{\partial\theta}\bigg|_{\hat{\theta}} = -m_0H[-(\hat{H}\cdot\hat{N})\sin\theta + (\hat{H}\cdot\hat{\theta})(\hat{N}\cdot\hat{\theta})\sin\theta + \hat{\theta}\cdot\hat{N}\times\hat{H}\cos\theta] + \tilde{K}\sin\theta \ . \tag{5}$$

Minimization with respect to $\hat{\theta}$ gives

$$0 = \hat{\theta}\times\frac{\partial\epsilon}{\partial\hat{\theta}}\bigg|_{\theta} = -m_0H\hat{\theta}\times\{[\hat{H}(\hat{N}\cdot\hat{\theta}) + \hat{N}(\hat{H}\cdot\hat{\theta})](1-\cos\theta) + (\hat{N}\times\hat{H})\sin\theta\} \ . \tag{6}$$

One solution of (5) and (6) is to have $\hat{\theta}$ along $\hat{N}\times\hat{H}$ with θ finite. This is the planar solution mentioned above. Then (6) is identically satisfied, and (5) determines θ; letting

$$\vec{H} = H(\hat{N}\cos\beta + \hat{Q}\sin\beta) \ , \tag{7}$$

so $\hat{N}\times\hat{H} = -\hat{P}\sin\beta$, Eq. (5) yields

$$\tan\theta = \frac{m_0H\sin\beta}{K + m_0H\cos\beta} \ . \tag{8}$$

Another solution of (5) and (6) is to have $\sin\theta = 0$ and $\hat{\theta}$ in the $\hat{H}$-$\hat{N}$ plane. Then (5) is identically satisfied, and (6) is satisfied either for $\cos\theta = 1$ (so $\theta = 0$) or for $\hat{\theta}$ midway between $\hat{N}$ and $\hat{H}$ (so $\theta = \pi$). We can rule out the $\theta = 0$ solution because then $\hat{\theta}$ is arbitrary, and cannot be restricted to the $\hat{H}$-$\hat{N}$ plane (small deviations along $\hat{N}\times\hat{H}$ show an instability toward the planar solution). Thus only the $\theta = \pi$ solution, which is the π-rotation solution mentioned above, is relevant. We will later see that $K_2 > K_1$ is necessary for the π-rotation solution to be stable.

From our earlier discussion with $\hat{n}$ and ψ as variables, one can see that the planar and π-rotation solutions are the *only* ones. The planar solution has been studied in detail by Henley, Sompolinsky, and Halperin,[6] who obtain the ESR frequencies and discuss the associated hysteresis loop for $\vec{H}$ along $\hat{N}$. They find that $\tilde{K} > 0$ for stability of the planar phase. We will perform a similar analysis for the π-rotation solution.

Note that if $K_1 = 0$, and $\vec{H}$ is not along $\vec{H}_c$, the π-rotation solution is always the minimum of energy, with both the Zeeman and anisotropy energies minimized. Thus, if one can find a material with a small enough K_1, the π-rotation solution should be realizable. For example, let a sample of SG be prepared in $\vec{H}_c$, so that it begins in the $\theta = 0$ planar state. Now apply an $\vec{H}$ opposite to $\vec{H}_c$, with $|\vec{H}| > (K_1 + K_2)m_0^{-1}$. It is straightforward to show that the $\theta = 0$ planar state becomes unstable, and $\hat{n}$ flips into $-\hat{N}$, via a 180° rotation about any axis in the $\hat{P}$-$\hat{Q}$ plane. In this case, the $\theta = \pi$ planar state and the π-rotation state are degenerate. If $K_2 > K_1$, only the π-rotation state is stable. Thus, if one now makes $|\vec{H}|$ very small, and rotates $\vec{H}$ back toward $\vec{H}_c$, one finds that $\hat{n}$ points along $\hat{N}$. On the other hand, if $K_2 < K_1$, only the planar state is stable, and this sequence of operations would end with $\hat{n}$ along $-\hat{N}$. Magnetization measurements should be able to easily distinguish between these two possibilities.

III. DYNAMICS (ESR FREQUENCIES, LOCAL STABILITY)

Let us now consider small oscillations $\delta\vec{m}$ and $\delta\vec{\theta}$ about the energy minimum for the π-rotation state. The equations of motion are (neglecting dissipation)[2,6,8]

$$\frac{\partial\vec{m}}{\partial t} = -\gamma\frac{\partial\epsilon}{\partial\vec{\theta}} = \gamma\vec{m}\times\vec{H} + \gamma\tilde{K}\frac{\partial\cos\theta}{\partial\vec{\theta}} \ , \tag{9}$$

$$\frac{\partial\vec{\theta}}{\partial t} = \gamma\frac{\partial\epsilon}{\partial\vec{m}} = \gamma\left(\frac{\vec{m}}{\chi} - \vec{H} - \frac{m_0}{\chi}\hat{n}\right) \ . \tag{10}$$

Now

$$\frac{\partial\cos\theta}{\partial\vec{\theta}} = \hat{\theta}\frac{\partial\cos\theta}{\partial\theta} = -\hat{\theta}\sin\theta \simeq \delta\theta\hat{\theta}_0 \ , \tag{11}$$

where $\delta\theta = \delta\vec{\theta}\cdot\hat{\theta}_0$ and we have linearized about $\theta_0 = \pi$. With $\delta\hat{n} = \delta\vec{\theta}\times\hat{n}_0 = \delta\vec{\theta}\times\hat{H}$, the linearized versions of (9) and (10) become

$$\frac{\partial}{\partial t}\delta\vec{m} = \gamma\delta\vec{m}\times\vec{H} + \gamma\tilde{K}(\delta\theta)\hat{\theta}_0 \ , \tag{12}$$

$$\frac{\partial}{\partial t}\delta\vec{\theta} = \frac{\gamma}{\chi}(\delta\vec{m} - m_0\delta\vec{\theta}\times\hat{H}) \ . \tag{13}$$

Because of the cross products involving $\hat{H}$, it is convenient to take components of $\delta\vec{m}$ and $\delta\vec{\theta}$ in the $(\hat{H},\hat{J},\hat{K})$ frame, where

$$\hat{H} = \hat{N}\cos\beta + \hat{Q}\sin\beta, \quad \hat{J} = \hat{P}, \quad \hat{K} = \hat{Q}\cos\beta - \hat{N}\sin\beta \ . \tag{14}$$

Then, with $\langle \cdots \rangle$ denoting the unit vector of the enclosed quantity,

$$\hat{\theta}_0 = \langle (\hat{N} + \hat{H}) \rangle = \hat{H} \cos\frac{\beta}{2} - \hat{K} \sin\frac{\beta}{2} \ , \quad (15)$$

$$\delta\theta = \delta\vec{\theta} \cdot \hat{\theta}_0 = \delta\theta_H \cos\frac{\beta}{2} - \delta\theta_K \sin\frac{\beta}{2} \ . \quad (16)$$

Letting $\delta\vec{m}$ and $\delta\vec{\theta}$ vary as $e^{-i\omega t}$, (12)–(16) then imply that the eigenfrequencies ω satisfy

$$0 = \omega^6 - \omega^4(\omega_L^2 + \omega_0^2 + \tilde{\omega}_a^2) + \omega^2[\omega_L^2\omega_0^2 + (\omega_0^2 + \omega_L^2)\tilde{\omega}_a^2 - \tilde{\omega}_a^2 S^2(\omega_0^2 + \omega_L^2 - \omega_0\omega_L)] - \omega_L^2\omega_0^2\tilde{\omega}_a^2(1 - S^2) \ , \quad (17)$$

where $S = \sin(\beta/2)$ and

$$\omega_L \equiv \gamma H, \quad \omega_0 \equiv \gamma m_0/\chi, \quad \tilde{\omega}_a^2 \equiv -\gamma^2\tilde{K}/\chi = \gamma^2(K_2 - K_1)/\chi \ . \quad (18)$$

For $\beta = 0$ (so $\vec{H}$ is along $\vec{H}_c$), Eq. (17) yields

$$\omega^2 = \omega_L^2, \omega_0^2, \tilde{\omega}_a^2 \quad (\vec{H} \parallel \vec{H}_c) \ . \quad (19)$$

The transverse roots $(\delta\vec{m} \perp \hat{H})\omega_L^2$ and ω_0^2 correspond to those for a ferrimagnet with no transverse anisotropy, ω_0 associated with the "optical" mode wherein the magnetization "beats" against the internal degrees of freedom, and ω_L associated with the Larmor precession. There is no transverse anisotropy for this geometry because, for triad anisotropy associated with $\theta_0 \neq 0$, the anisotropy torque can point only along $\hat{\theta}_0$, which here is $\hat{H}$. In addition to the transverse modes, there is the longitudinal mode $(\delta\vec{m} \parallel \hat{H})$ at $\tilde{\omega}_a$, driven only by the anisotropy. Note that $K_2 - K_1$ appears in $\tilde{\omega}_a^2$ because K_2 provides a restoring force, whereas K_1 tries to make the state go unstable ($\omega^2 < 0$ for an unstable state).

For larger values of β, the roots of (17) become more complicated, so that one cannot clearly label a given motion as purely longitudinal or purely transverse. At $\beta = \pi$, the π-rotation state and the planar state become degenerate, so the ESR frequencies should be identical. Indeed, we find, in agreement with Ref. 6, that for $\beta = \pi$ one mode is purely longitudinal at

$$\omega^2 = 0 \quad (\vec{H} \parallel -\vec{H}_c) \ , \quad (20)$$

and the transverse modes are given by

$$\omega^2 = \tfrac{1}{2}(\omega_L^2 + \omega_0^2 + \tilde{\omega}_a^2) \pm \tfrac{1}{2}[(\omega_L^2 + \omega_0^2 + \tilde{\omega}_a^2)^2 - 4(\omega_L^2\omega_0^2 + \tilde{\omega}_a^2\omega_L\omega_0)]^{1/2} \ . \quad (21)$$

The $\omega^2 = 0$ longitudinal mode signifies that the planar and π-rotation states have become degenerate: There is a distortion of each state (i.e., $\delta\hat{\theta}$ in the $\hat{P}$-$\hat{Q}$ plane) for which there is no restoring force. Note that the transverse modes here are more complicated than for the case of single-axis anisotropy (Ref. 7) because in the present case the anisotropy torque points along only $\hat{Q}(=\hat{\theta}_0)$, rather than along both $\hat{Q}$ and $\hat{P}$. The resulting equations of motion thus have less symmetry.

From (17) we can learn something about the stability of the π-rotation state. Use of Newton's rule tells us that, for $\tilde{\omega}_a^2 > 0$ (i.e., $K_2 > K_1$), there are no negative roots. Further, in the absence of dissipation, we expect ω^2 to be purely real. Thus for $K_2 > K_1$ we expect all three roots to be positive, and the π-rotation state to be locally stable. As a consequence, once the system enters the π-rotation part of $(\vec{m}, \vec{\theta})$ space, any transitions back into the planar part of $(\vec{m}, \vec{\theta})$ space must take place via a first-order transition. Such transitions are well known to be metastable, and therefore not easily reproduced. Indeed, if anything characterizes the return-flip data of Ref. 5, it is that there is a great deal of scatter in the data, not inconsistent with metastability.

IV. TORQUE AND TRANSVERSE SUSCEPTIBILITY

The results expected fom torque measurements are easily obtained. Since the anisotropy torque is zero in the π-rotation state, torque measurements should yield a null result when the SG is in that state.

Similarly, the results expected from transverse susceptibility measurements are easily obtained. Since $\vec{m}$ follows $\vec{H}$ in the π-rotation state, we have $\delta m_\perp/m = \delta H_\perp/H$, or

$$\chi_\perp = \frac{\delta m_\perp}{\delta H_\perp} = \frac{m}{H} = \chi + \frac{m_0}{H} \ . \quad (22)$$

For small H, this is considerably larger than what one obtains for the planar state, where the anisotropy prevents $\vec{m}$ from completely following $\vec{H}$. Specifically, for the planar state with $\vec{H}$ along $\vec{H}_c$, one has[9]

$$\chi_\perp = \chi + \frac{m_0^2}{\tilde{K} + m_0 H} \ , \quad (23)$$

where here $\tilde{K} = K_1 + K_2$.

V. DISCUSSION

To our knowledge, there is not yet any explicit experimental evidence to establish that anisotropy in spin-glasses is specified by an anisotropy triad, rather than a single anisotropy axis. Study of the ESR frequencies of the planar state (Ref. 6) can do this. However, the planar state possesses the same statics as does a uniaxial ferromagnet. The π-rotation state discussed here is particularly interesting because it is possible only because of the additional degree of freedom that spin-glasses possess. In addition, as we suggest, the reverse-flip hysteresis data (Ref. 5) may require that we invoke the π-rotation state. Thus, if the condition $K_1 < K_2$ can be satisfied, the π-rotation state should be stable, and should be realizable in the laboratory (as discussed in Sec. III). Note that $K_2 = 4K_1$ if the microscopic interaction causing anisotropy is symmetric under spin interchange.[6,10]

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