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Anisotropy-Triad Dynamics in Spin-Glasses

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In Heisenberg spin-glasses, the orientation of a state is specified by a reference spin triad $(\hat{n}, \hat{p}, \hat{q})$, and its anisotropy is specified by a rotation $\vec{\psi}$ of this state away from its equilibrium orientation. Rotation of $(\hat{n}, \hat{p}, \hat{q})$ by $-\vec{\psi}$ defines an *anisotropy triad* $(\hat{N}, \hat{P}, \hat{Q})$, whose dynamics is derived. This dynamics is exclusively dissipative, consistent with torque and ESR measurements. Analogy to ordinary glasses suggests a Vogel-Fulcher law for the associated relaxation time.

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A valuable picture of the spin-glass (SG) state has emerged from the numerical simulations of Walker and Walstedt for a prototype SG, *Cu*Mn.[1] These authors randomly substitute Mn atoms for Cu, and study the local energy minima for a Ruderman-Kittel-Kasuya-Yosida (RKKY) coupling. They find that, in each local minimum, the Mn spins point in all three spin directions, in a fashion that overall would appear random to the eye. Individual spin orientations are thus specified with respect to an orthonormal triad, which I call the *spin triad* $(\hat{n},\hat{p},\hat{q})$.[2] The orientation of the state as a whole is given by specifying $(\hat{n},\hat{p},\hat{q})$, where $\hat{n}$ is taken along the remanent magnetization $\vec{m}_0$; the orientation of $\hat{p}$ and $\hat{q}$ (about $\hat{n}$), however, cannot be associated with any macroscopic observable.

Because the anisotropy energy of real SG's is found to be independent of the magnitude $m_0 \equiv |\vec{m}_0|$, SG anisotropy has been somewhat puzzling.[3,4] Indeed, Schultz *et al.*,[4] who successfully interpreted a large part of their ESR data in terms of a uniaxial anisotropy involving $\hat{n}\cdot\hat{N}$, comment that "$\hat{N}$ is a fixed (in space) direction whose origin remains a mystery to us." Even more mysterious is the fact that torque experiments show, under certain circumstances, that the anisotropy axis $\hat{N}$ is *not* fixed, but rotates with respect to the crystal.[5,6]

If anisotropy in SG's were specified only by rotations about axes which are normal to $\hat{N}$, it would be the purpose of this paper to derive the dynamics of $\hat{N}$. However, the microscopic calculations of Fert, Levy, and Morgan-Pond[7] have shown that an anisotropy torque develops on rotating an equilibrium SG state about *any* axis $\hat{\psi}$. This torque is along $\hat{\psi}$, and its magnitude (as well as the anisotropy energy) depends only on the magnitude ψ of the rotation angle. $\vec{\psi} \equiv \psi\hat{\psi}$ gives the relative orientation between the apparent lattice orientation (as given by the density matrix which specifies the state) and the actual lattice orientation (as given by the Hamiltonian). Although it is sufficient to specify the anisotropy by $\vec{\psi}$, as in Ref. 7, for the present purposes it is more convenient to work with $(\hat{n},\hat{p},\hat{q})$ and a generalization of the anisotropy axis, which I call the *anisotropy triad* $(\hat{N},\hat{P},\hat{Q})$. Given $(\hat{n},\hat{p},\hat{q})$ and $\vec{\psi}$, one obtains $(\hat{N},\hat{P},\hat{Q})$ on rotating $(\hat{n},\hat{p},\hat{q})$ by $-\vec{\psi}$. Thus, in equilibrium, where there is no anisotropy torque, $(\hat{n},\hat{p},\hat{q})$ coincides with $(\hat{N},\hat{P},\hat{Q})$. [To obtain $\vec{\psi}$ from $(\hat{n},\hat{p},\hat{q})$ and $(\hat{N},\hat{P},\hat{Q})$, one employs the rotation matrix taking $(\hat{N},\hat{P},\hat{Q})$ to $(\hat{n},\hat{p},\hat{q})$:

$$R_{\alpha\beta}=\hat{n}_\alpha\hat{N}_\beta+\hat{p}_\alpha\hat{P}_\beta+\hat{q}_\alpha\hat{Q}_\beta. \tag{1}$$

The relation $\epsilon_{\alpha\beta\gamma}R_{\alpha\beta}=2\hat{\psi}_\gamma\sin\psi$ then gives ψ and $\hat{\psi}$.]

It is thus the purpose of this paper to derive the macroscopic dynamics (or hydrodynamics[8]) of the anisotropy triad $(\hat{N},\hat{P},\hat{Q})$. I find its behavior to be exclusively dissipative, and discuss some implications of the theory for torque[5,6] and ESR[4,9] experiments, which already show such dissipative behavior. Finally, I suggest an analogy between the anisotropy of SG's and the shear rigidity of ordinary glasses.

Of the large number of spin degrees of freedom in a SG, at a macroscopic level we consider only the magnetization $\vec{m}$ and the triads $\{\vec{u}^{(i)}\}\equiv(\hat{n},\hat{p},\hat{q})$ and $\{\vec{v}^{(i)}\}\equiv(\hat{N},\hat{P},\hat{Q})$. In terms of these and the entropy density S, the differential of the energy density ϵ is taken to be

$$d\epsilon = TdS+(h_\alpha-H_\alpha)dm_\alpha+\lambda_\alpha{}^{(i)}du_\alpha{}^{(i)}+\Gamma_{\alpha\beta}dR_{\alpha\beta}. \tag{2}$$

We take $h_\alpha=\chi^{-1}(m_\alpha-m_0\hat{n}_\alpha)$,[4] so that $\vec{m}=m_0\hat{n}+\chi\vec{H}$ in equilibrium can account for the remanent and induced parts of the magnetization. The requirement of rotational invariance [i.e., $d\epsilon=0$ under uniform rotations of $\vec{m}$, $(\hat{n},\hat{p},\hat{q})$, *and* $(\hat{N},\hat{P},\hat{Q})$] then gives $\lambda_\alpha{}^{(1)}=-(m_0/\chi)m_\alpha$, $\lambda_\alpha{}^{(2)}=\lambda_\alpha{}^{(3)}=0$. The last term of Eq. (2) we obtain after introducing the infinitesimal relative-rotation vector $d\psi_\mu\equiv d\theta_\mu-R_{\mu\nu}d\Theta_\nu$, where $d\theta_\mu$ rotates $(\hat{n},\hat{p},\hat{q})$ and $d\Theta_\nu$ rotates $(\hat{N},\hat{P},\hat{Q})$. Then $dR_{\alpha\beta}=\epsilon_{\alpha\mu\nu}d\psi_\mu R_{\nu\beta}$, and so

$$\Gamma_{\alpha\beta}dR_{\alpha\beta}=-\Gamma_\mu d\psi_\mu,\quad \Gamma_\mu\equiv-\epsilon_{\mu\nu\alpha}R_{\nu\beta}\Gamma_{\alpha\beta}. \tag{3}$$

The experiments of Gullikson, Schultz, and Fredkin,[10] when extended to triad anisotropy, indicate that the anisotropy torque is

$$\Gamma_\mu=-(K_1\sin\psi+\tfrac{1}{2}K_2\sin2\psi)\hat{\psi}_\mu, \tag{4}$$

which can be understood from theoretical considerations.[11-13]

To obtain the dynamical behavior, the following equations of motion are assumed:

$$\begin{aligned}&\dot{\epsilon}+\partial_i j_i{}^\epsilon=0,\quad \dot{S}+\partial_i j_i{}^S=R\geq0,\\ &\dot{m}_\alpha=-\gamma\,\partial\epsilon/\partial\theta_\alpha+J_\alpha,\\ &\dot{\vec{u}}^{(i)}=\vec{\omega}\times\vec{u}^{(i)},\quad \dot{\vec{v}}^{(i)}=\vec{\Omega}\times\vec{v}^{(i)},\end{aligned} \tag{5}$$

where $j_i{}^\epsilon$, $j_i{}^S$, R, J_α, $\omega_\alpha\equiv\dot{\theta}_\alpha$, $\Omega_\alpha\equiv\dot{\Theta}_\alpha$ are unknown (γ is the gyromagnetic ratio). In computing $\partial\epsilon/\partial\theta_\alpha$ we must rotate $\vec{m}$ and $(\hat{n},\hat{p},\hat{q})$, but *not* $(\hat{N},\hat{P},\hat{Q})$, so that

$$-\gamma\,\partial\epsilon/\partial\theta_\alpha=\gamma(\vec{m}\times\vec{H})_\alpha+\gamma\Gamma_\alpha. \tag{6}$$

With $R_{\mu\nu}\Gamma_\nu=\Gamma_\mu$ [since $\vec{\Gamma}$ is along $\hat{\psi}$, by Eq. (4)], and $\vec{\Gamma}'\equiv\vec{\Gamma}-(m_0/\chi)\vec{m}\times\hat{n}$, Eqs. (2)–(6) yield

$$0\leq TR=-\partial_i(j_i{}^\epsilon-Tj_i{}^S)-j_i{}^S\partial_iT-J_\alpha(h_\alpha-H_\alpha)+[\vec{\omega}-\gamma(\vec{h}-\vec{H})]\cdot\vec{\Gamma}'-\vec{\Omega}\cdot\vec{\Gamma}. \tag{7}$$

This involves, as expected, a pure divergence term and the product of unknown thermodynamic fluxes (e.g., $j_i{}^S$) with known thermodynamic forces (e.g., ∂_iT). Thus $\vec{h}-\vec{H}$ is the "force" associated with $\vec{m}$, $\vec{\Gamma}'$ is associated with $(\hat{n},\hat{p},\hat{q})$, and $\vec{\Gamma}$ is associated with $(\hat{N},\hat{P},\hat{Q})$. In equilibrium, when these "forces" are all zero, $\vec{m}$, $\hat{n}$, $\hat{N}$, and $\vec{H}$ are all parallel, and $(\hat{n},\hat{p},\hat{q})$ coincides with $(\hat{N},\hat{P},\hat{Q})$. By writing the fluxes as linear combinations of the "forces," and employing the Onsager symmetry principle, we find that

$$\begin{aligned}&j_i=Tj_i{}^S,\quad j_i{}^S=-(\kappa/T)\partial_iT,\\ &J_\alpha=-\gamma D(h_\alpha-H_\alpha),\\ &\omega_\alpha-\gamma(h_\alpha-H_\alpha)=\gamma E\Gamma_\alpha{}'+\gamma E'\Gamma_\alpha,\\ &\Omega_\alpha=-\gamma F\Gamma_\alpha-\gamma E'\Gamma_\alpha{}'.\end{aligned} \tag{8}$$

Here we neglect a possible $\vec{m}\times\vec{\Gamma}'$ term in J_α, and its associated $\vec{m}\times(\vec{h}-\vec{H})$ term in ω_α, as well as other related terms ($m_0\hat{n}\times\vec{\Gamma}'$, etc.). To be consistent with $R\geq0$, we must make the (dissipative) transport coefficients satisfy $\kappa,D,E,F,(EF-E'^2)\geq0$.

Observe that for J_α, Γ_α, and m_0 all zero (as assumed in Ref. 14), the first four of Eqs. (8) agree with Ref. 14. It is the last of Eqs. (8) which contains the (new) dynamics of the anisotropy triad $(\hat{N},\hat{P},\hat{Q})$, via Eq. (5).

We immediately note that the dynamics of $(\hat{N},\hat{P},\hat{Q})$ is exclusively dissipative, since in Eq. (8) the individual terms in $\vec{\Omega}$ are even under time reversal T, and hence $\dot{\vec{v}}^{(i)}$ and $\vec{\Omega}\times\vec{v}^{(i)}$ behave differently under T. [The neglected $\vec{m}\times(\vec{h}-\vec{H})$ and

$m_0\hat{n}\times(\vec{h}-\vec{H})$ terms are also dissipative.] This is consistent with the zero-field–cooled (ZFC) torque measurements of Ref. 6. These authors apply a field $\vec{H}$ to the ZFC sample, and then slowly rotate $\vec{H}$ about a perpendicular axis $\hat{\omega}_0$. They find that the torque initially builds up, but that eventually it saturates, even though $\vec{H}$ continues to rotate about $\hat{\omega}_0$. This indicates that $\vec{H}$ has brought $(\hat{n},\hat{p},\hat{q})$ into rotation, and $(\hat{n},\hat{p},\hat{q})$ has brought $(\hat{N},\hat{P},\hat{Q})$ into rotation, but that there develops a constant phase lag. In addition, in the torque measurements of Ref. 5, when data points were retaken, with $\vec{H}$ held fixed for an additional time interval, the torque was found to have decreased.[15] This I interpret to indicate that the phase lag has relaxed. A third example of this sort arises in the angle dependence of the ESR resonance field $\vec{H}_r$ observed in Ref. 4. There it is found that ZFC samples show no dependence on the angle between $\vec{H}_r$ and the cooling field $\vec{H}_c$ ($H_c < 1$ G), although there is a marked dependence for samples cooled in kilogauss fields. I take this to indicate that $(\hat{N},\hat{P},\hat{Q})$ relaxes more quickly in ZFC samples than in non-ZFC samples. We now turn to a brief analysis of these phenomena, using Eqs. (4)–(6) and (8).

The ZFC torque experiments correspond to a steady-state solution with $\dot{\vec{m}}=\vec{\omega}_0\times\vec{m}$, $\vec{\omega}=\vec{\omega}_0$ and $\vec{\Omega}=\vec{\omega}_c$, where $\vec{\omega}_0$ is the rotation rate of the field $\vec{H}$. Such a solution holds so long as ω_0 is sufficiently small that we may linearize the $\dot{\vec{m}}$ equation with $\vec{m}\approx(m_0+\chi H)\hat{H}$. Using $\hat{n}\approx\hat{H}+\delta\hat{n}$, $\delta\vec{h}\equiv\vec{h}-\vec{H}$, and $\vec{\Gamma}$ as variables, one can show that $\vec{\Gamma}\cdot\hat{H}=\delta\vec{h}\cdot\hat{H}=0$. (This holds even including the $\vec{m}\times\vec{\Gamma}'$ and related terms.) Thus there are six quantities $(\hat{H}\times\delta\hat{n},\hat{H}\times\delta\vec{h},\hat{H}\times\vec{\Gamma})$ to be solved for. In general, one expects $\vec{\Gamma}\cdot\hat{\omega}_0\times\hat{H}\neq 0$. However, the equations simplify considerably if we take $E'\approx 0$. Then (8) gives $\vec{\Gamma}=-(\gamma F)^{-1}\vec{\Omega}$, and so if $\vec{\Omega}$ is along $\hat{\omega}_0$, the torque $\vec{\Gamma}$ lies along the rotation axis $\hat{\omega}_0$. As a consequence, if $\hat{n}$ and $\hat{N}$ initially are normal to $\hat{\omega}_0$, they will always be normal to $\hat{\omega}_0$, and Eq. (4) then gives $\vec{\Gamma}=-(K_1+K_2)(\theta_n-\theta_N)\hat{\omega}_0$, where θ_n and θ_N are measured about the $\hat{\omega}_0$ axis and $\theta_n-\theta_N$ is taken to be small. Since $\dot{\theta}_N=\vec{\Omega}\cdot\hat{\omega}_0$ here, there is a characteristic relaxation rate τ^{-1} determined by

$$\dot{\theta}_N=-\tau^{-1}(\theta_N-\theta_n),\quad \tau^{-1}\equiv\gamma F(K_1+K_2). \tag{9}$$

(This is true for any $\vec{\Omega}$ always along $\hat{\omega}_0$, and $\theta_n-\theta_N$ small.) Because both the rotation rate ω_0 and the torque Γ are needed to extract γF ($=\omega_0/\Gamma$ here), and ω_0 is not given along with the measured values of Γ, it is not possible from the literature to determine characteristic values for γF, and thus τ. However, from the previously mentioned contrasting behavior of ZFC and non-ZFC samples, it is clear that $\tau_{\text{ZFC}}^{-1}\gg\tau_{\text{non-ZFC}}^{-1}$, so that τ is a sensitive function of H_c.

One can also study, for $E'\approx 0$, the situation where $\vec{H}$ changes on a time scale slow enough for $\vec{m}$ and $(\hat{n},\hat{p},\hat{q})$ to be in local equilibrium [so $\vec{h}\approx\vec{H}$ and $\vec{\Gamma}\approx(m_0/\chi)\vec{m}\times\hat{n}$], but too fast for $(\hat{N},\hat{P},\hat{Q})$ to be in local equilibrium (so $\vec{\Gamma}\neq\vec{0}$). This corresponds to the non-ZFC situation.[3b,5,6] We will restrict ourselves to rotation (perhaps not constant) only about $\hat{\omega}_0$, so that the system may be described solely in terms of angles about $\hat{\omega}_0$. For a given $\vec{H}$ and $\hat{N}$, the local equilibrium solution for θ_n is obtained from $\vec{\Gamma}\cdot\hat{\omega}_0=(m_0/\chi)\vec{m}\times\hat{n}\cdot\hat{\omega}_0$:

$$K_1\sin(\theta_n-\theta_N)+\tfrac{1}{2}K_2\sin 2(\theta_n-\theta_N)=m_0H\sin(\theta_H-\theta_n). \tag{10}$$

The adjustment of θ_N to the motion of θ_n and θ_H is obtained from (10) and the finite $\theta_n-\theta_N$ version of (9): with (10), it is

$$\delta\theta_N=\gamma F m_0 H\int_0^t\sin(\theta_H-\theta_n)dt. \tag{11}$$

θ_n, in turn, adjusts to $\delta\theta_N$, via (10). By varying both θ_n and θ_N in Eq. (10), I obtain

$$\delta\theta_n=\tilde{K}[\tilde{K}+m_0H\cos(\theta_H-\theta_n)]^{-1}\delta\theta_N,\qquad \tilde{K}\equiv K_1\cos(\theta_n-\theta_N)+K_2\cos 2(\theta_n-\theta_N). \tag{12}$$

As a consequence, the torque $\vec{\Gamma}$ has, in addition to its value for θ_N frozen [given by the negative of either the left- or right-hand side of Eq. (10)], an additional contribution

$$\delta\vec{\Gamma}=-\hat{\omega}_0\tilde{K}(\delta\theta_n-\delta\theta_N). \tag{13}$$

This expression can be applied either to the case of a constant, rapid rotation of $\vec{H}$ about $\hat{\omega}_0$[5,6] or the case of a rapid rotation of $\vec{H}$ about $\hat{\omega}_0$ to a new fixed value. In the first case one sets $\theta_N=\theta_H=0$ as initial conditions (so $\theta_n=0$ initially), then solves Eq. (10) for θ_n with $\theta_H=\omega_0 t$, and then employs (11)–(13) to obtain $\delta\theta_N(t)$, $\delta\theta_n(t)$, and $\delta\vec{\Gamma}(t)$. In the second case one sets $\theta_N=0$ and $\theta_H=\theta_0$ as initial conditions [with θ_n determined by (10)], and then (11)–(13) are used to obtain the time development.

One more case is of interest: that where both θ_n and θ_N are small and vary as $e^{-i\omega t}$, such as occurs in transverse ac susceptibility measurements,[3b] or ESR measurements with $\vec{H}$ along $\vec{H}_c$[4,9] [for $\vec{H}$ not along $\vec{H}_c$, the motion is more complicated than simple rotation about a single axis normal to $\vec{H}$ (Ref. 12)]. In this case, Eq. (9)

implies that $\theta_N = \theta_n(1 - i\omega\tau)^{-1}$, so that

$$\vec{\Gamma}\cdot\hat{\omega}_0 = -(K_1+K_2)(1+i/\omega\tau)^{-1}\theta_n \equiv -K(\omega)\theta_n. \quad (14)$$

This provides a frequency-dependent anisotropy constant, with characteristic time τ.

Note that both ordinary glasses and fluids have a shear modulus with a frequency dependence well represented by the form $(1+i/\omega\tau)^{-1}$, as in Eq. (14). For fluids τ has no sharp T dependence, whereas for ordinary glasses as one approaches the temperature $T_g{}^*$ from above, one finds the Vogel-Fulcher law, $\tau \propto \exp[A/(T - T_g{}^*)]$. Tholence has discussed, from an experimental viewpoint, the applicability of this "law" to the time dependence of m_0.[16] For nondilute RKKY SG's it seems to hold, whereas for dilute RKKY SG's and for insulating SG's, an Arrhenius law [$\tau \propto \exp(B/T)$, with a wide range of activation energies B (Ref. 17)] seems to hold. The question of Vogel-Fulcher versus Arrhenius behavior is clearly relevant to the relaxation of the anisotropy triad. Analogy to the shear response of ordinary glasses suggests Vogel-Fulcher behavior but this remains to be seen. (Note that Vogel-Fulcher behavior probably would indicate a more collective effect than would Arrhenius behavior.)

To summarize, I have constructed a phenomenological theory of spin-glasses which permits the anisotropy triad to move. Consistent with experiment, this motion is found to be purely dissipative. Detailed confirmation of the structure of the theory would permit quantitative evaluation of the associated relaxation time (as a function, e.g., of H_c, T, H, and spin concentration), and suggest mechanisms responsible for this relaxation.

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