Dissipation in Macroscopic Magnetization Tunneling

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The effect of magnetoelastic dissipation upon the tunneling and coherence of the total magnetization in small ($\sim$100-Å) single-domain ferromagnetic particles is investigated. Such tunneling would be an example of a *macroscopic* quantum phenomenon. It is shown that the dissipation has a super-Ohmic spectral density, $J(\omega) \sim \omega^3$, and that the dissipation is weak for reasonable material parameters. Corrections to the rates, and the damping rate for coherent oscillations, are obtained. A key feature is the inclusion of the elastic waves in the medium surrounding the particle.

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Consider a small, single-domain ferromagnetic particle at a temperature low enough to freeze out spin waves due to crystalline anisotropy. At such temperatures, the exchange interactions align the individual magnetic moments nearly perfectly, and the spontaneous magnetization, $\mathbf{M}$, is close to its saturation value, M_0, in magnitude. The only interesting degree of freedom left is then the direction of $\mathbf{M}$, and the question arises whether it is possible to see quantum-mechanical effects in its dynamical behavior. Since a sphere of radius $a \sim 50$ Å contains about 10^5–10^6 moments, such effects would provide another instance of *macroscopic* quantum phenomena, e.g., tunneling and coherence (MQT and MQC, respectively), which so far have been studied with any seriousness only for the rf SQUID and the closely related current-biased Josephson junction.[1-4]

The issue of macroscopic magnetization tunneling (MMT) was recently raised by Chudnovsky and Gunther[5] (CG) (see also Refs. 6 and 7), who observed that *both* the barrier to tunneling and the mechanism for it would be provided by the magnetocrystalline anisotropy[8] which is always present. Except for a few passing remarks, however, they have not considered the dissipative effect of the environment, and it is the purpose of this paper to do so. Dissipation generally suppresses quantum effects, and it is therefore clearly desirable to assess its importance for MMT before contemplating real experiments. The only source of dissipation that we shall consider in this paper is the magnetoelastic coupling of $\mathbf{M}$ to the phonons. This is perhaps the most obvious intrinsic source, and is also the one mentioned by CG.[9]

The setup for MQT[2] consists of a biaxial crystal, magnetized along an easy axis which we label x, and a field $\mathbf{H}$ opposite to $\mathbf{M}$.[10] The Hamiltonian for $\mathbf{M}$ can be taken to be the experimentally determined anisotropy energy itself since this guarantees the correct semiclassical dynamics. In polar coordinates, the energy *density* can be taken to be

$$E(\theta,\phi) = K_1\cos^2\theta + (K_1+K_2)\sin^2\theta\sin^2\phi - HM_0(1-\sin\theta\cos\phi)\,, \qquad (1)$$

with $K_{1,2} > 0$. The x direction ($\theta=\pi/2$, $\phi=0$) is metastable for $H < H_c = 2K_1/M_0$.

The setup for MQC[3] consists of a uniaxial or biaxial crystal with an easy axis labeled z, and $\mathbf{H}$ in the x direction (model II of Ref. 5). Now,

$$E(\theta,\phi) = K\sin^2\theta - HM_0\sin\theta\cos\phi + H^2M_0^2/4K\,, \qquad (2)$$

and for $H < H_c = 2K/M_0$, there are two degenerate minima at $\phi=0$, $\theta=\pi/2 \pm \delta_0$, $\delta_0 = \cos^{-1}(H/H_c)$.

As discussed by CG, in order to get an appreciable tunneling rate, we must have $\epsilon \equiv 1 - H/H_c \ll 1$. For specificity, we shall use the parameters they quote, $K \sim K_1 \sim K_2 \sim 5\times10^6$ ergs/cm^3, $M_0 \sim 500$ G, and take a spherical particle of radius $a = 50$ Å. Then for MQT, in the absence of dissipation, the WKB exponent, $S_0/\hbar$, is given by

$$\frac{S_0}{\hbar} = v_0\epsilon^{3/2}\frac{8M_0}{3\hbar\gamma}\left(\frac{K_1}{K_2}\right)^{1/2}, \qquad (3)$$

and the small precession frequency in either well, $\omega_{\rm pr}$, is given by $(2\gamma/M_0)(K_1K_2\epsilon)^{1/2}$ to leading order in ϵ. Here, v_0 is the volume of the particle, and $\gamma = g\mu_B/\hbar$, with g being the g-factor and μ_B the Bohr magneton. To get a value of 5 for $S_0/\hbar$, we need $\epsilon \sim 2\times10^{-3}$ which corresponds to tunneling through an angle of 7° or so, and gives $\omega_{\rm pr} \sim 10^{10}$ sec^{-1}, and a tunneling rate of about 10^8 sec^{-1}. The results for MQC are very similar: $S_0/\hbar$ is given by $v_0(2M_0/3\hbar\gamma)(2\epsilon)^{3/2}$, and $\omega_{\rm pr}$ by $(2K\gamma/M_0)(2\epsilon)^{1/2}$, with similar numerical values.[11]

The chief result of our work is that magnetoelastic dissipation is "weak," having a strength characterized by a dimensionless parameter α, given by

$$\alpha = (K_{\rm ME}/K)(a\omega_H/c_s)^2\,. \qquad (4)$$

Here, $K_{\rm ME}$ is what Kanamori [Ref. 8(a), Sec. 5] calls the strain-induced anisotropy coefficient, a is the radius of the particle, $\omega_H \equiv \gamma H_c$, and c_s is an average measure of the sound speed. Since $K_{\rm ME}/K$ is typically 10^{-2}–10^{-4}, and $a\omega_H/c_s$ is about 0.1, α is of order of 10^{-4}–10^{-6}. More precisely, we shall show that the relative size of the dissipative correction to the WKB exponent is proportional to α with a proportionality constant which is difficult to evaluate precisely, but can be estimated to be of order unity.

In the presence of dissipation, the tunneling rate for either MQC or MQT may be found by path-integral methods developed by Leggett and co-workers.[2,3] To do this we need the action for $\mathbf{M}$ and the phonons. The important phonons are those with frequencies less than ω_{pr}, which can be treated via continuum elasticity theory. We are thus led to consider the (Euclidean) action[12]

$$S[\hat{\mathbf{M}}(\tau),\mathbf{u}(\mathbf{x},\tau)]=S_0[\hat{\mathbf{M}}]-\int d\tau\int d^3x\,(a_{ijkl}u_{ij}M_kM_l-L_E^{\rm glas})\,, \tag{5}$$

where $\hat{\mathbf{M}}$ is the direction of $\mathbf{M}$, and $\mathbf{u}$ and u_{ij} are the displacement and strain fields. $S_0[\hat{\mathbf{M}}]$ is the "bare" action for $\mathbf{M}$ without dissipation,[5,13]

$$S_0[\hat{\mathbf{M}}(\tau)]=v_0\int[E(\theta,\phi)-i\gamma^{-1}M_0\cos\theta\dot{\phi}(\tau)]d\tau\,, \tag{6}$$

and $L_E^{\rm glas}$ is the Euclidean Lagrangian for the elastic degrees of freedom alone. Finally, a_{ijkl} is the magnetoelastic tensor ($a_{ijkl}=a_{jikl}=a_{ijlk}$). The fundamental quantity to be calculated is $S_{\rm cl}$, the value of the action along the "classical" or extremal paths. In terms of this, the tunneling rate (or the tunnel splitting for MQC) $\sim\omega_0\times\exp(-S_{\rm cl}/\hbar)$, where ω_0 is (for moderate damping) a small oscillation frequency (ω_{pr} in our case).

Since phonons with frequencies of the order of ω_{pr} typically have wavelengths much greater than a, it is obviously insufficient to consider the phonons of the isolated magnetic grain. The experimental conditions for MMT, should they ever be realized, will almost certainly be such[7] that the grain is embedded in a nonmagnetic solid medium. Hence, the Lagrangian $L_E^{\rm glas}$ should be chosen so as to describe both the grain and the outer medium.[14] In general, of course, this can be very complicated, particularly if there are large stresses at the interface, leading to local modes. In keeping with our goal of a low-frequency description, and in order to keep the problem simple, we will simply ignore these. We will take the simplest choice for $L_E^{\rm glas}$, namely, one that leads to the equations for freely propagating elastic waves in a uniform solid in each medium, and Newton's third law at the interface.

Although it is now possible to formally integrate out the phonons completely, and obtain an effective action for $\mathbf{M}$ alone, the resulting expressions are unwieldy and not useful. For $\epsilon\ll 1$ the classical path for $\mathbf{M}$ lies very close to the x-z plane.[15] The most important coupling term in Eq. (5) is the one that differs most in the two configurations between which tunneling occurs, i.e., the $a_{xzxz}u_{xz}M_xM_z$ term, since the M_z^2 term and the terms involving M_y are small, *a priori*, and the M_x^2 term is almost a constant, and has very little *dynamical* coupling to the phonons in the course of the tunneling. To obtain the tunneling exponent to leading order in ϵ it suffices to drop all these other terms, further approximate M_xM_z by $M_0^2\cos\theta(\tau)$, and expand S to second order in $\phi(\tau)$. One can then perform the resulting Gaussian path integral over $\phi(\tau)$.[6] Denoting $\cos\theta(\tau)$ by $q(\tau)$, and, keeping only leading order terms in ϵ, we obtain an action

$$S[q(\tau),\mathbf{u}(\mathbf{x},\tau)]=v_0\int d\tau[\tfrac{1}{2}m\dot{q}^2+V(q)]-g_c\int d\tau\int_{v_0}d^3x\,qu_{xz}+\int d\tau\int d^3x\,L_E^{\rm glas}\,. \tag{7}$$

Here, $g_c=4a_{xzxz}M_0^2$, m equals $M_0^2/2K_2\gamma^2$ for MQT, $M_0^2/2K\gamma^2$ for MQC, and $V(q)$ is given by

$$V(q)=\begin{cases}K_1(\epsilon q^2-q^4/4) & (\text{MQT})\,,\\ K(\epsilon-q^2/2)^2 & (\text{MQC})\,.\end{cases} \tag{8}$$

Note finally that since the relevant range of q is of order $\epsilon^{1/2}$, we can ignore the fact that q is defined on a circle and extend it to the entire line $(-\infty,+\infty)$. The problem is now reduced to that of a particle of mass m moving in a one-dimensional potential $V(q)$, and coupled to an oscillator bath. One (minor) bonus of this reformulation is that the prefactor for the tunneling rate without dissipation is known; this is $\sqrt{12}(S_0/2\pi\hbar)^{1/2}\omega_{pr}$ for MQT, and twice this value for MQC.

The next step is to integrate out the oscillators and obtain the spectral density[2,3] $J(\omega)$. For phonon wavelengths $\lambda\gg a$, this can be done for any choice of elastic constants for the particle and the surrounding medium. The most crucial feature of $J(\omega)$, however, is the power law with which it grows as $\omega\to 0$. This can be deduced simply by taking both the particle and the outside medium to be *identical*, isotropic, elastic media.[16] Different wave vectors $\mathbf{k}$ then correspond to different oscillator modes, and the coupling constants $c_{\mathbf{k}}$, oscillator masses $m_{\mathbf{k}}$, and frequencies $\omega_{\mathbf{k}}$, as defined in Ref. 2(a) (with the label α replaced by $\mathbf{k}$) can be explicitly calculated. For longitudinal waves, for example, $m_{\mathbf{k}}=\rho$, $\omega_{\mathbf{k}}=c_lk$, and $c_{\mathbf{k}}=g_cv_0k_xk_zh(ka)/k$, where ρ is the density, c_l is the longitudinal sound speed, and

$$h(x)=3(\sin x-x\cos x)/x^3\,. \tag{9}$$

The longitudinal contribution $J_l(\omega)$ is then given by

$$J_l(\omega)=\frac{\pi}{2}\int\frac{d^3k}{(2\pi)^3}\frac{c_{\mathbf{k}}^2}{m_{\mathbf{k}}\omega_{\mathbf{k}}}\delta(\omega-c_lk)\,. \tag{10}$$

This is easily evaluated, as is the transverse part $J_t(\omega)$. Finally, $J(\omega)\equiv J_l+J_t$ is given by

$$J(\omega)=\frac{(v_0g_c)^2}{4\pi\rho}\left[\frac{h^2(x_l)}{15c_l^5}+\frac{h^2(x_t)}{10c_t^5}\right]\omega^3\,, \tag{11}$$

where $x_l=\omega a/c_l$, etc. Note that since $h(0)=1$, $J(\omega)$

$\sim\omega^3$ for small ω. This is an example of "super-Ohmic" dissipation,[3(b)] and cannot be characterized by a frequency-independent friction coefficient. The factors of h^2 act as high-frequency cutoffs.

A better model is obtained by taking the inner sphere to be a different, but still isotropic, elastic medium. The calculation of $J(\omega)$ is now most simply done by using the method of Leggett.[17] One considers the real time [as opposed to the "imaginary" time variable τ appearing in Eq. (5)] dynamical equations for $q(t)$ and $\mathbf{u}(\mathbf{x},t)$, defines Fourier transforms,

$$\tilde{q}(\zeta)=\int_{-\infty}^{\infty} q(t)e^{-i\zeta t}dt\,,\quad \mathrm{Im}\zeta<0\,, \tag{12}$$

and writes the equation for q in the form

$$K(\zeta)\tilde{q}(\zeta)=-\tilde{V}_q(\zeta)\,, \tag{13}$$

where $\tilde{V}_q(\zeta)$ is the Fourier transform of dV/dq. Then

$$J(\omega)=\lim_{\epsilon\to 0+}\mathrm{Im}[K(\omega-i\epsilon)]\,. \tag{14}$$

It is easy to see that if $q(t)$ is regarded as a driving or source term for the phonons, then the power lost at frequency ω is proportional to $\omega J(\omega)|\tilde{q}(\omega)|^2$.[18] The source now produces *elastic* dipole radiation (both transverse and longitudinal), for which, just as in the *electric* dipole case, the power lost varies as ω^4. It follows that $J(\omega)\sim\omega^3$ once again.

To obtain the scale of $J(\omega)$, we Fourier transform the inhomogeneous elastic wave equation obeyed by $\tilde{\mathbf{u}}(\mathbf{x},\zeta)$ over space. The integral equation so obtained can be solved in the limit $k\to 0$.[19] When this solution is substituted in the equation for $\tilde{q}(\zeta)$, the results for $K(\zeta)$ and $J(\omega)$ can be read off. The net effect of this procedure is that Eq. (11) gets multiplied by a prefactor $(1+\eta)/(1+2\eta)^2$, where

$$\eta=(3+2c_t^2/c_l^2)(\mu'-\mu)/15\mu\,, \tag{15}$$

where μ' and μ are the adiabatic shear moduli of the particle and the outer medium, respectively, and all other quantities pertain to the latter. This prefactor is bounded above by 35/8 (since $c_t^2/c_l^2<3/4$) and is generally of order unity. We do not known the precise form of the cutoff, but we do not consider this to be important, and in the following analysis we will simply replace the h^2 factors in Eq. (11) by $e^{-\omega/\omega_l}$, etc., with $\omega_l=bc_l/a$, where $b\sim 1$ is some number.

The effective action $S_{\mathrm{eff}}[q]$ for q can now be written as S_0+S_1, where S_0 is the bare action, and

$$S_1[q]=\frac{1}{2\pi^2}\int_{-\infty}^{\infty}d\omega\int_0^{\infty}d\bar{\omega}\frac{\omega^2}{\bar{\omega}(\omega^2+\bar{\omega}^2)}|q(\omega)|^2 J(\bar{\omega})\,, \tag{16}$$

where $q(\omega)$ is the Fourier transform of $q(\tau)$. [This is a simple rewriting of Eqs. (4.22–24) of Ref. 2(b).] In general, one must solve anew for the extremal path of $S_{\mathrm{eff}}[q]$, but if the dissipation is weak, one can use the path found in its absence, $q_0(\tau)$, to estimate S_1 in Eq. (16). We shall see that this approach is justified for the present problem. It is easily shown that for the MQT problem, $q_0(\tau)=2\sqrt{\epsilon}\,\mathrm{sech}(\omega_{\mathrm{pr}}\tau)$, so that

$$q_0(\omega)=(2\pi\sqrt{\epsilon}/\omega_{\mathrm{pr}})\,\mathrm{sech}(\pi\omega/2\omega_{\mathrm{pr}})\,. \tag{17}$$

The integral in Eq. (16) is then easily evaluated, and its longitudinal part $S_{1,l}$ is

$$S_{1,l}\simeq v_0\epsilon\frac{8b^3}{135\pi}\frac{g_c^2}{\rho c_l^2}\frac{\omega_{\mathrm{pr}}}{\omega_l^2}\,. \tag{18}$$

$S_{1,t}$ is similarly given. The factor $g_c^2/\rho c_l^2$ more or less defines K_{ME}, the strain-induced anisotropy coefficient [see Ref. 8(a), Sec. 5]. Using Eq. (3) for S_0, it follows that S_1/S_0 is proportional (with a constant of order unity) to the dimensionless quantity α defined in Eq. (4). The effect of dissipation is thus to multiply the tunneling rate by a factor $\exp(-S_1/\hbar)$. (We have not attempted to evaluate the modification of the prefactor since this is a much smaller effect owing to the smallness of the dissipation.)

For the case of MQC, one cannot, in general, define a single "tunneling rate," but must enquire about the full dynamical behavior of $\hat{\mathbf{M}}(t)$. Since the dissipation is super-Ohmic and weak, however [see Ref. 3(b), Secs. 2, 3A, and 3B, and Appendix *A*], this behavior is a damped oscillation at all temperatures much less than $\hbar\omega_{\mathrm{pr}}/k_B$, and the oscillation frequency Δ can be computed in exactly the same way as the tunneling rate of the MQT problem. In other words, Δ is given by its value for zero dissipation times a factor $\exp(-S_1/\hbar)$. (Once again, the modification of the prefactor is insignificant.) In this case, $q_0(\omega)=2i\pi/\omega_H\sinh(\pi\omega/\omega_{\mathrm{pr}})$, and the final results for S_1 are very similar; e.g., the 8 in Eq. (18) becomes a 4. It is also of interest to ask for the damping rate of these oscillations. This is found from Eq. (3.11) of Ref. 3(b) to be of the order of $v_0K_{\mathrm{ME}}(\Delta a/c_s)^3\coth(\beta\hbar\Delta/2)$. For the parameters we have been using, this is about 100 sec^{-1} for $k_BT\ll\hbar\Delta$.

We conclude this Letter by noting that for nonspherical particles, an important contribution to $E(\theta,\phi)$, which has been omitted in all the above analysis, is the shape anisotropy (or demagnetization) energy E_{dem}. For ellipsoids,

$$E_{\mathrm{dem}}=\tfrac{1}{2}(N_aM_a^2+N_bM_b^2+N_cM_c^2)\,, \tag{19}$$

where a,b,c are the axes of the ellipsoid, and N_a, etc., are the demagnetization factors, normalized to add up to 4π.[20] Equation (19) continues to hold for *uniformly magnetized samples of any shape*,[21] but the principle axes a,b,c, of the N tensor are now not simply specifiable. The effective anisotropy coefficient is of the order of πM_0^2, which for $M_0\sim 500$ G is about 8×10^5 ergs/cm^3, which is not negligible compared to K, K_1, etc.

This contribution to the energy has the effect of misaligning the easy and hard axes of magnetization from the crystalline axes. While all our calculations are formally valid with the understanding that the applied field is perpendicular to the new easy axis for the case of MQC, or along it for MQT, it is clear that producing these configurations can be a vexing experimental complication if one does not have some way of determining the magnetic axes as opposed to the crystalline axes.

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[9]Other possible sources include the dipolar couplings between different grains and the hyperfine coupling between electronic and nuclear spins.

[10]This is model III of Ref. 5. MQT is also possible for a *uniaxial* crystal, but the tunneling Hamiltonian must then contain at least the fourth- or sixth-order invariant in $E(\mathbf{M})$ which breaks rotational symmetry in the hard plane, and the rate is much smaller.

[11]Note that our value for $S_0/\hbar$ is $\sqrt{2}/3$ times that given in Ref. 5.

[12]The inclusion of a counterterm [as defined in Ref. 2(b), Sec. 2] in Eq. (5) effectively changes the coefficients K, K_1, etc., by an amount of the order of K_{ME} [see Eq. (4)], which is typically 10^{-2} to 10^{-4} times K_i.

[13]This can be derived as in Ref. 5, or by appealing to spin-coherent states.

[14]In order for Eq. (5) to make sense as written, the $\mathbf{x}$ integral should be over all space with $\mathbf{M}\equiv 0$ outside the grain.

[15]Indeed, one can see from Ref. 5, that for the undamped classical paths, $\phi(\tau)$ is $O(\epsilon)$, while $\theta(\tau)-\pi/2$ is $O(\epsilon^{1/2})$.

[16]If the media are different, and the normal modes are described by an incident plus a scattered wave, the scattered intensity varies as k^4 for $ka \ll 1$ (Rayleigh scattering). To leading order in $\mathbf{k}$, we do not expect the *exponent* in $c_{\mathbf{k}}$, $m_{\mathbf{k}}$ or the density of states to change.

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[18]In other words, $J(\omega)$ is proportional to the imaginary part of the dynamic susceptibility of the mode to which $q(t)$ couples. For another illustration of this general result, see Eq. (2.14) of A. Garg, J. N. Onuchic, and V. Ambegaokar, J. Chem. Phys. **83**, 4491 (1985).

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