Dissipation by Nuclear Spins in Macroscopic Magnetization Tunneling

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(Received 14 August 1992)

Magnetic systems are currently considered attractive candidates in which to look for quantum phenomena such as the tunneling of the total magnetization of a small (~100 Å diameter) particle out of a metastable easy direction or between degenerate easy directions. The effect of nuclear spins as a dissipative environment for such a particle is considered and shown to be significant. Two dimensionless parameters characterize the dissipation: a coupling strength varying as the number of nuclear spins, and the ratio of the nuclear and electronic Larmor frequencies.

PACS numbers: 75.60.Jp, 03.65.Db, 76.60.Jx

In the last five years, several authors, including this one, have suggested that magnetic systems may be attractive candidates in which to look for macroscopic quantum tunneling and coherence (MQT and MQC, collectively referred to as MQP—"P" for phenomena) [1]. The phenomena examined theoretically include magnetization reversal in small grains, both ferromagnetic [2–6] and antiferromagnetic [7], nucleation in bulk magnetic materials [8], and the motion of domain walls [9]. Experimental observation of some of these effects has been claimed [10,11], but the question is still open in my opinion, as the evidence is very indirect. The author has questioned the interpretation of MQC in Ref. [11].

Since, as is now established [12–14], dissipation, or the coupling of the macrovariable to its environment, generally suppresses quantum effects, a basic requirement for the observability of MQP is that dissipation must be weak. In previous papers [3,5], magnetoelastic dissipation was shown to be negligibly small in the problem of magnetization reversal in small grains. In this paper we shall consider a new dissipative mechanism, the coupling between electronic and nuclear magnetic moments. We shall show that this form of dissipation *is* significant, and may be quite strong depending on the strength of the hyperfine fields and the number of nuclear moments. To keep the discussion focused, we shall limit ourselves to magnetization reversal in ferromagnetic grains, but such dissipation is also important for the other problems mentioned above.

The system under study consists of a single-domain, insulating [15], ferromagnetic grain, about 50 Å in radius, at a temperature well below the anisotropy gap. The individual electronic moments are then nearly perfectly aligned, and the magnetization $\mathbf{M}$ equals its saturation value M_0. The direction $\hat{\mathbf{M}}$ of $\mathbf{M}$ is variable, however, and quantum effects in its dynamics could justifiably be called macroscopic as there are 10^5–10^6 moments in the particle. In an external field $\mathbf{H}$, the Hamiltonian for an isolated grain can be taken to be the anisotropy energy,

$$H_a(\hat{\mathbf{M}}) = v_0(-K_1\hat{M}_z^2 + K_2\hat{M}_y^2 - \mathbf{M}\cdot\mathbf{H}) , \tag{1}$$

where v_0 is the particle volume, and K_1 and K_2 are anisotropy coefficients ($K_{1,2} > 0$) [16]. The particle is initially magnetized along its easy axis, z, and $\mathbf{H} \parallel -\mathbf{M}$. The magnetization can tunnel out of the z direction, but the rate is very small unless $\epsilon \equiv 1 - H/H_c \ll 1$, where $H_c = 2K_1/M_0$ is the field which renders the z axis classically unstable. (We need $\epsilon \sim 10^{-3}$–10^{-2} to get tunneling through a few degrees for typical anisotropies and magnetizations.) Up to a constant, for $M_{x,y} \ll M_z$, we can write

$$H_a(\theta,\phi) = K_1 v_0 \theta^2(\epsilon - \theta^2/4) + K_2 v_0 \theta^2 \sin^2\phi , \tag{2}$$

where θ and ϕ are polar angles.

The small size of the particle ensures that all the moments will tunnel together, and an instanton calculation [2] gives the WKB exponent in the tunneling rate Γ for the isolated particle as

$$S_0^{\rm cl}/\hbar = v_0 \epsilon^{3/2}(8M_0/3\hbar\gamma)(K_1/K_2)^{1/2} , \tag{3}$$

where γ is the magnetogyric ratio for the electronic moments. (See also Refs. [17,18] for early work on spin tunneling problems.) The small precession frequency in the metastable well ω_p is $(2\gamma/M_0)(K_1K_2\epsilon)^{1/2}$, and the barrier height U is $K_1 v_0 \epsilon^2$. It is useful to note that $S_0^{\rm cl}/\hbar = 16U/3\hbar\omega_p$. For $K_1 \sim K_2 \sim 5\times10^6$ ergs/cm^3, $M_0 \sim 500$ G, a particle radius of 50 Å, and $\epsilon \sim 2\times10^{-3}$, we have $\omega_p \sim 10^{10}$ sec^{-1} and $\Gamma \sim 10^8$ sec^{-1}. There is obviously a large variability in this rate with the material parameters K_1, K_2, and M_0, and especially v_0, the particle volume.

When interactions with nuclear spins are included, the tunneling rate can again be calculated using instanton methods. An immediate (and not fully resolved) question arises about the temperature that should be used in such a calculation. In a real experiment (at a temperature $\Theta \sim 10$ mK, say), even though $k_B\Theta \ll \hbar\omega_p$, $k_B\Theta$ is likely to be huge on the nuclear moment energy scale. The relaxation time for the nuclear spins (T_1), however, is typically several seconds or more at such low Θ, which far exceeds the tunneling or "bounce" time τ_b. ($\tau_b \sim \omega_p^{-1}$ without dissipation, and we will estimate it below when dissipation is included.) Thus the nuclear spins cannot exchange energy with each other during the tunneling process, and can be regarded as being in a single well-defined state (as opposed to a density matrix) at the start of a tunneling event. Formal $\Theta \neq 0$ calculations of the imaginary part of the free energy [19] or of a WKB-like escape rate [20] are based on the opposite assumption of a rapidly relaxing bath (see Sec. 5.3 of Ref. [1(a)], and so should not be used. They would falsely include the effects of incoherence among the microstates of the nuclear spin system. It is more accurate perhaps to do a $\Theta = 0$ calculation, as this better mimics a single initial nuclear spin state. Such a calculation also has the advantage of being simpler and better defined, and giving the maximum depression of the escape rate (including thermal effects) due to dissipative effects. By comparing the exponent in this rate with $U/k_b\Theta$, we can also estimate the crossover temperature between thermal activation and quantum tunneling.

The nuclear moments can be divided into three classes: (1) those in the magnetic atoms, (2) those in nonmagnetic atoms inside the grain, and (3) those in (nonmagnetic) atoms in the medium surrounding the grain. Here, we shall treat in detail only type 1 nuclear spins and qualitatively discuss the treatment and effects of the other two types. The interaction Hamiltonian for one such nuclear spin, denoted **I**, with the electronic spin on the same atom **J**, and the magnetic field, is given by

$$H_n = \mathbf{I} \cdot A \cdot \mathbf{J} - \hbar \gamma_n \mathbf{I} \cdot \mathbf{H}_{\rm ext} , \tag{4}$$

where $\mathbf{H}_{\rm ext} = -H_c \hat{\mathbf{z}} - (4\pi/3)\mathbf{M}$, γ_n is the nuclear magnetogyric ratio, and A is the hyperfine interaction tensor, whose principle axes we will take to be x, y, and z [21]. If there are N nuclear spins in the grain (and N electronic spins, assuming only one magnetic species for simplicity), we have $M_0 v_0 = N\hbar\gamma J$, and we can write

$$H_n = \hbar\omega_n I_z + A'_x I_x J_x + A'_y I_y J_y , \tag{5}$$

where

$$\begin{aligned} \omega_n &= A_z J/\hbar + \gamma_n (H_c + 4\pi M_0/3) , \\ A'_{x,y} &= A_{x,y} + 4\pi N \hbar^2 \gamma\gamma_n / 3v_0 . \end{aligned} \tag{6}$$

Let the initial state of the combined system be denoted by $|\hat{\mathbf{z}}, \{-I\}\rangle$, where $\hat{\mathbf{z}}$ is the orientation of $\hat{\mathbf{M}}$, and $\{-I\}$ indicates that $I_z = -I$ for all nuclear spins. The tunneling rate Γ can be found by calculating the quantity

$$Q(T) = \langle \hat{\mathbf{z}}, \{-I\} | e^{-HT/\hbar} | \hat{\mathbf{z}}, \{-I\} \rangle \tag{7}$$

as $T \to \infty$, and comparing the result with $\exp(-E_0 + i\Gamma/2)T/\hbar$. Here H is the total Hamiltonian. Up to an irrelevant normalization factor, $Q(T)$ is given by the path integral

$$Q(T) = \int [d\hat{\mathbf{M}}] \{\exp - S_0[\hat{\mathbf{M}}(\tau)]/\hbar\} \Lambda_n^N [\hat{\mathbf{M}}(\tau)] , \tag{8}$$

where

$$\Lambda_n[\hat{\mathbf{M}}] = \left\langle -I \middle| \mathcal{T} \exp\left[-\int_0^T H_n(\tau) d\tau/\hbar \right] \middle| -I \right\rangle . \tag{9}$$

Here, $\mathcal{T}$ denotes time ordering, and S_0 is the "bare" action for the isolated grain, given by

$$S_0[\hat{\mathbf{M}}] = \int [H_a(\theta,\phi) - i\gamma v_0 M_0 \cos\theta \dot{\phi}(\tau)] d\tau . \tag{10}$$

To evaluate $Q(T)$ in the semiclassical approximation, we first ignore dissipation (effectively setting $\Lambda_n = 1$). For small ϵ, it is easily shown [2] that the classical path lies nearly in the x-z plane: $M_x \sim \epsilon^{1/2}$, $M_y \sim \epsilon$. We can thus perform the path integration in Eq. (8) over $\phi(\tau)$ in the Gaussian approximation, reducing S_0 to the action for a one-dimensional problem:

$$S_0[\theta] = K_1 v_0 \int d\tau [\epsilon \omega_p^{-2} \dot{\theta}^2 + \theta^2 (\epsilon - \theta^2/4)] . \tag{11}$$

The classical solution or "bounce" is given by

$$\theta_{\rm cl}(\tau) = 2\epsilon^{1/2} {\rm sech}(\omega_p \tau) . \tag{12}$$

To evaluate Λ_n, we can therefore ignore the fluctuations in $J_y(\tau)$ compared to $J_x(\tau)$. We expect the magnitude of the interaction term in Eq. (9), $\epsilon^{1/2} J A'_x \tau_b/\hbar$ to be very small compared to unity, so we can use perturbation theory to find Λ_n. Setting the ground state energy for the nuclear spin to zero, we get

$$\Lambda_n[\hat{\mathbf{M}}] = 1 + \frac{IJ^2}{2\hbar^2} A'^2_x \int_0^T d\tau_1 \int_0^{\tau_1} d\tau_2 \theta(\tau_1)\theta(\tau_2) e^{-\omega_n \tau_{12}} , \tag{13}$$

with $\tau_{12} = \tau_1 - \tau_2$.

At first sight it seems that, in our contradiction to our earlier statements, the coupling to nuclear spins enhances the tunneling rate since $\Lambda_n > 1$ by Eq. (13). This, however, is not so. We have not been careful enough in our treatment of the bare tunneling rate. If we define $\theta_i = \theta(\tau_i)$, and write

$$2\theta_1\theta_2 = \theta_1^2 + \theta_2^2 - (\theta_1 - \theta_2)^2 , \tag{14}$$

the terms involving θ_1^2 and θ_2^2 can be reduced to single time integrals. If we write Λ_n as an exponential, they amount to a renormalization of the θ^2 term in Eq. (11), or a lowering of the potential energy for $\hat{\mathbf{M}}$ in Eq. (2). Indeed since the coupling to nuclear spins cannot be physically turned off, a measurement of the small angle precession frequency ω_p (by ferromagnetic resonance, e.g.) automatically includes this renormalization. Therefore, the quantities that we have been calling ω_p, K_1, and U in Eqs. (1)-(3), (11), and (12) should be understood to include the effects of coupling to nuclear spins. It is not difficult to see that the renormalized potential for $\hat{\mathbf{M}}$ is obtained by allowing the nuclear spins to adjust instantaneously or adiabatically to their minimum energy state for given $\hat{\mathbf{M}}$. True dissipative effects are due solely to the last term in Eq. (14) [22].

The upshot of the above arguments is that the effective action $S_{\rm eff}[\theta]$ equals $S_0 + S_1$, where S_0 is given by Eq. (11) (with ω_p now properly understood), and

$$S_1[\theta] = N \frac{IJ^2}{8\hbar^2} A'^2_x \int_0^T \int_0^T [\theta(\tau_1) - \theta(\tau_2)]^2 \times e^{-\omega_n |\tau_{12}|} d\tau_1 d\tau_2 . \tag{15}$$

Note that this is precisely of the Caldeira-Leggett form [12], with a spectral density $J(\omega) \sim \delta(\omega - \omega_n)$.

It is apparent that we can treat type 2 and 3 nuclear spins in the same way. For type 2 spins, the Hamiltonian can be written entirely as $-\hbar\gamma_{n2}\mathbf{I}_2\cdot\mathbf{H}_{\rm loc}$ instead of Eq. (4). Here, $\mathbf{H}_{\rm loc}$ is the field at the nucleus, and consists of $\mathbf{H}_{\rm ext}$ and fields produced by nearby atomic moments. The latter can be quite strong, and are proportional to M. In the path integral for the matrix element $Q(T)$, we should therefore include another factor Λ_{n2} similar to Eq. (9). To evaluate Λ_{n2}, we divide $\mathbf{H}_{\rm loc}$ into static and dynamic pieces, the latter being proportional in magnitude to $M_x(\tau)$ or $\theta(\tau)$. The part of the dynamic piece that is parallel to the static piece is much smaller in magnitude [because $\theta(\tau)$ is small] and can be neglected, and only the transverse dynamic piece need be kept. The calculation then proceeds as for Λ_n, and yields an additive contribution S_2 to the effective action that is similar to S_1 [Eq. (15)], with N, I, A_x', and ω_n being replaced with quantities $N_2,\ldots,\omega_{n2}$, appropriate to type 2 spins. We note here that since local fields in magnetic materials are often quite strong (1–50 T) at *all* the nuclei [23], A_{x2}' and A_x' may well be comparable.

The evaluation of the correction to the action S_3 from type 3 spins is conceptually similar. The local field now varies in space, as it consists of the applied field $-H_c\hat{\mathbf{z}}$ and the dipole field created by $\mathbf{M}$. The analogs of ω_n and A_x' are also varied with position, and instead of just multiplying by N as we did to get S_1 [Eq. (15)], we have to integrate the single-spin correction to $S_{\rm eff}$ over all space, with a suitable density of type 3 spins. We have not been able to write S_3 in any simple useful form, but we note that the corresponding spectral density can extend down to very low frequencies, including zero if $4\pi M_0/3 > H_c/2$, which is possible for soft magnetic materials. Strong low frequency dissipation can suppress MQP quite severely, so the effect of type 3 spins is potentially dangerous. The overall strength of S_3 is likely to be smaller than that of S_1 or S_2, as the local nuclear field outside the grain is weaker, but we do not have quantitative estimates.

To quantitatively analyze the effects of dissipation on the tunneling rate, we will consider only type 1 spins. We will obtain approximate forms for the minimum value of S_0+S_1 in limiting cases. The S_2 contribution will be easily incorporable once this is done. We proceed as in Sec. 5 of Ref. [12(b)], and define $u=\omega_p\tau$, $\theta(\tau)=2\epsilon^{1/2}\times z(u)$, and

$$\sigma[z]=(\hbar\omega_p/4U)S_{\rm eff}[\theta(\tau)]\,. \tag{16}$$

(Recall that U is the barrier height.) The dissipation is characterized by two dimensionless parameters $\eta=\omega_n/\omega_p$, and μ, given by

$$\mu=NI\epsilon(A_x'/A_z')^2\hbar\omega_p/U\,. \tag{17}$$

We shall denote the minimum value of $\sigma[z]$ by $b(\mu,\eta)$. (Thus, when dissipation is absent, $b=\frac{4}{3}$, which is achieved for $z=\operatorname{sech}u$.) Assuming a hyperfine field at the nucleus of 0.5–50 T, and $\omega_p\sim10^{10}\ \sec^{-1}$, we get $\eta\sim0.001$–1. Taking $N\sim10^5$, $\epsilon\sim10^{-3}$, $A_x'/A_z'\sim1$, and $\hbar\omega_p/U\sim\frac{1}{5}$ (any smaller value would lead to a very small bare tunneling rate), we estimate $\mu\gtrsim10$.

In terms of these dimensionless variables, we can write

$$\sigma[z]=\int du\,(\dot z^2+z^2-z^4)+\frac{\mu\eta^2}{8}\int\int[z(u)-z(u')]^2e^{-\eta|u-u'|}du\,du'\,. \tag{18}$$

We will analyze σ approximately, rather than attempt a comprehensive numerical minimization (which could be efficiently done following Ref. [24]). We will assume that the bounce $z_0(u)$ which minimizes σ varies on a time scale u_b that is alternately much greater (case A) and much lesser (case B) than η^{-1}, and self-consistently verify the conditions on μ and η under which these assumptions hold. [Note that z has a single maximum at $u=0$, and $z(\pm\infty)=0$.] Let us denote the two terms in Eq. (18) by σ_0 and σ_1. In terms of the Fourier transform $\tilde z(\omega)$ of $z(u)$ we have

$$\sigma_1[z]=\frac{\mu\eta}{4\pi}\int d\omega|\tilde z(\omega)|^2\frac{\omega^2}{\omega^2+\eta^2}\,. \tag{19}$$

Consider case A first. Then $\tilde z(\eta)\ll1$, and we can put $\omega=0$ in the $\omega^2+\eta^2$ denominator in Eq. (19). If we revert to an integral over u, we see that σ_1 then has the effect of renormalizing the coefficient of the $\dot z^2$ term in σ_0 to $1+\mu/2\eta$. The resulting action has the same form as the bare action, and a simple scaling argument yields

$$u_b=(1+\mu/2\eta)^{1/2},\quad b_A=\tfrac{4}{3}u_b\,. \tag{20}$$

The subscript denotes case A. The self-consistency condition for this solution to be a minimum is $u_b\gg1/\eta$, i.e., $\eta^{1/2}\gg(2/\mu)^{1/2}$. The bounce is given by $z_0=\operatorname{sech}(u/u_b)$.

Now consider case B. The range of $\tilde z(\omega)$ is then much greater η, and we can approximate the $\omega^2+\eta^2$ denominator in Eq. (19) by ω^2. σ_1 then renormalizes the z^2 term in σ_0, and again a scaling argument gives

$$u_b=(1+\mu\eta/2)^{-1/2},\quad b_B=\tfrac{4}{3}u_b^{-3}\,. \tag{21}$$

The bounce is $u_b^{-1}\operatorname{sech}(u/u_b)$. The self-consistency condition is $u_b\ll1/\eta$, i.e., $\eta^{1/2}\ll(\mu/2)^{1/2}$.

Before discussing the ranges of applicability of these two cases further, we note that in both cases dissipation causes the action to increase, and hence the tunneling rate to decrease. In case A, dissipation effectively increases the inertia for $\hat{\mathbf{M}}$, while leaving the potential unchanged, while in case B, the inertia is unchanged, but the height and width of the barrier are both effectively increased. Either way, the tunneling rate decreases. Somewhat counterintuitively, in case B, the tunneling time $\tau_b\simeq u_b/\omega_p$, or the time spent "under the barrier," decreases. The suppression in the tunneling rate is similar to the Franck-Condon suppression of electronic transition rates in molecules, insofar as the nuclear spins, like the vibrational degrees of freedom, are slow and cannot adjust to the rapidly changing electronic spins. It is not possible, however, to easily identify a Franck-Condon overlap integral in our formulas.

For $\eta \leq 1$, case B has a much greater region of validity than case A. For $\mu = 10$, $\eta = 0.1$, e.g., case A is inapplicable, and case B gives $u_b = 0.82$ (self-consistently correct), and $b = \frac{4}{3}(1.36)$. This is a 36% increase in the WKB exponent, and could suppress tunneling quite severely if the bare rate were already deemed to be small. For $\mu = 10$, $\eta = 0.01$, $b = \frac{4}{3}(1.076)$.

The self-consistency conditions for both cases hold if $(\mu/2)^{1/2} \gg \eta^{1/2} \gg (2/\mu)^{1/2}$. One can then show that $b_A < b_B$ only if $\eta > (2/\mu)^{1/2}$. If $\eta < 1$, as we expect, this condition limits the region where the best path is given by solution A to values of $\mu \gtrsim 100$. On the other hand, neither solution is self-consistently valid if μ is small and η is moderate, e.g., $\mu = \eta = 0.1$. In such cases, however, b can be well estimated perturbatively, i.e., by substituting the undamped solution in Eq. (18) for $\sigma[z]$. This approach fails if μ is small and η is large (but still much less than $2/\mu$), e.g., $\mu = 0.1$, $\eta = 5$. Such parameters are unlikely to be physically relevant, however.

The effect of type 2 spins can be treated in the same way. In fact if μ, η, and the corresponding parameters μ_2 and η_2 for the type 2 spins were such as to both favor a solution of type B, for instance, the least action b could be obtained simply by adding a term $\mu_2\eta_2/2$ under the radical for u_b in Eq. (21).

We have shown that nuclear spin dissipation can suppress MQT in magnetic particles quite severely, and cannot in any case be neglected for a quantitative understanding of the problem. Does it make it totally hopeless, however, to look for these effects? We do not believe so. It may not be out of the question to work with isotopes that have no nuclear moments whatsoever. The magnetic elements ^{56}Fe and ^{52}Cr fall in this class, as do naturally abundant isotopes of several other elements that could be used to make the magnetic compound, as well as the substrate or embedding medium for the particles. Even if high isotopic purity is not feasible, dissipation might be kept small by having a small value of μ. [This might be achieved, e.g., if the magnetic ion was Fe. The natural abundance of ^{57}Fe is 2.25%, effectively reducing N in Eq. (17) by a factor of 50.]

Some recent papers have shown that topological interference effects due to the second term (which is a Berry phase) in Eq. (10) lead to a quenching of the bare MQC tunnel splitting for half-integer and some other special values of the total spin J [25], or for any J, but special values of an external magnetic field applied so as to preserve two classically degenerate ground states [26]. It is likely that nuclear spin dissipation will lead to a partial unquenching of the splitting.

I am indebted to W. P. Halperin and J. B. Ketterson for useful discussions. This work is supported by the National Science Foundation through Grant No. DMR-9102707.

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