Conservation of Angular Momentum in the Problem of Tunneling of the Magnetic Moment

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Tunneling of the magnetic moment has some unique features not encountered in other tunneling problems. The conservation of energy and angular momentum prohibits transitions between degenerate magnetic states in a free single-domain magnetic particle. For such transitions to occur, the particle must be firmly coupled with a large solid matrix that absorbs the change in the angular momentum. We show that the contribution of this effect to the tunneling rate is determined by the ratio of the magnetic anisotropy energy to the shear modulus of the matrix. An experiment is suggested that can test this prediction.

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Tunneling of the magnetic moment between equilibrium orientations in magnets has been the subject of intensive recent theoretical and experimental studies [1]. In these studies one important aspect of the problem has been ignored. The macroscopic magnetic moment $\mathbf{M}$ is due to the ordering of electron spins, or orbital momenta, or combined. Consequently, it has an angular momentum associated with it, $\gamma^{-1}\mathbf{M}$ ($\gamma = ge/2mc$ being the gyromagnetic ratio). For that reason, a freely suspended body, on being magnetized, begins to rotate (the Einstein-de Haas effect). This suggests that any spontaneous transition of $\mathbf{M}$ between equilibrium magnetic states is only possible if the system finds the way to compensate the corresponding change in the angular momentum; otherwise, the transition is strictly prohibited by the conservation law. Consider a free single-domain magnetic particle in the absence of the magnetic field, which is characterized by the magnetic moment $\mathbf{M}$ and the mechanical angular momentum $\mathbf{L}$. As usual, we shall assume that $\mathbf{M}$ is formed by the strong exchange interaction and can only change its orientation, but not the absolute value. The dynamics of $\mathbf{L}$ and $\mathbf{M}$ must preserve the total angular momentum,

$$\mathbf{L} + \gamma^{-1}\mathbf{M} = \text{const}. \tag{1}$$

On the other hand, the symmetry with respect to the time reversal dictates that the free energy of the particle is the even form of vectors $\mathbf{M}$ and $\mathbf{L}$,

$$E = A_{ik} M_i M_k + B_{ik} L_i L_k + C_{ik} M_i L_k + \cdots . \tag{2}$$

If the position of the particle was fixed, that is, if $\mathbf{L}$ was strictly zero, the ground state would be degenerate with respect to the transformation $\mathbf{M} \rightarrow -\mathbf{M}$, with the equilibrium orientation of $\mathbf{M}$, relative to the particle, being determined by the magnetic anisotropy. For a free particle, however, (2) is only degenerate with respect to the simultaneous change of sign of $\mathbf{M}$ and $\mathbf{L}$. Together with (1), this means that the transition between $\mathbf{M}$ and $-\mathbf{M}$ states cannot occur in a free *nonrotating* particle. Such a transition would cause the rotation in the final state with $\mathbf{L} = 2\gamma^{-1}\mathbf{M}$ and the kinetic energy $E_r = L^2/2I$, I being the moment of inertia of the particle. For a nanometer size magnetic particle, E_r would be of the order of a few kelvin (or even greater), that is, much higher than any uncertainty in the energy of the particle at low temperature. To compensate this energy change, the particle should have been prepared in a metastable magnetic state, by, e.g., placing it in a sufficiently large magnetic field. If this is not the case, Eqs. (1) and (2) dictate that the free particle can change the direction of its magnetic moment only if it has zero total angular momentum, that is, in the state with $\mathbf{L} = -\gamma^{-1}\mathbf{M}$. Any rotation of $\mathbf{M}$ will then be accompanied by the rotation of $\mathbf{L}$ such that $\mathbf{L} + \gamma^{-1}\mathbf{M} = 0$. Note that if $\mathbf{M}$ is of spin origin, this condition becomes $\mathbf{L} + \mathbf{S} = 0$, which can be satisfied only for an integer total spin of the magnetic particle, but not for a half-integer spin. This is in accordance with the Kramers theorem and its recent path-integral derivation [2,3], which say that the magnetic moment should be frozen in a half-integer-spin particle.

Consider now a small magnetic particle which is imbedded in a large nonmagnetic *absolutely* rigid solid matrix. "Absolutely rigid" means infinite elastic moduli; that is, the matrix can only move as a whole, but is not allowed to develop any local elastic deformations. Again, the exact degeneracy of the ground state with respect to the transformation $\mathbf{M} \rightarrow -\mathbf{M}$ requires zero total angular momentum of the system, that is, the mechanical rotation with the angular momentum $\mathbf{L} = -\gamma^{-1}\mathbf{M}$. However, the kinetic energy associated with this rotation is now $E_r = L^2/2I_m$, which is inversely proportional to the fifth power of the size of the matrix (I_m being the moment of inertia of the matrix). Thus, in the limit of an infinite stationary matrix, the magnetic degeneracy of the ground state becomes precise, and spontaneous transitions between $\mathbf{M}$ and $-\mathbf{M}$ states can satisfy conservation of energy and angular momentum. In fact, the physical limitation on the minimal size of the matrix is that E_r is negligible compared to the tunneling splitting of the ground-state energy level. One may notice the difference from the conventional double-well problem (e.g., coherent tunneling between degenerate configurations of ammonia molecule), where the size of the system is irrelevant.

Let us finally turn to the physical case of a particle imbedded in a large stationary solid matrix that has finite elastic moduli. The imaginary-time dynamics of the system now involves the elastic twist in the matrix, produced by the rotational recoil due to tunneling of **M**. Equation (1) tells us that to conserve the total angular momentum tunneling from **M** to $-\mathbf{M}$ must be accompanied by the creation of $2M/\gamma\hbar$ transversal phonons of spin 1. However, the energy of these phonons in the final state of an infinite system will be zero, so that no real-time excitations will be created. One should note the direct analogy with the zero-phonon line in Mössbauer spectroscopy. This makes tunneling between degenerate magnetic states (macroscopic quantum coherence) theoretically possible. A more general case (which also should be easier to detect) is quantum decay of a metastable magnetic state, created, e.g., in a small particle by the external magnetic field [4]. As we will see, the balance of the total angular momentum is important for this problem as well. It should also enter the problem of tunneling of a domain wall [5,6]. In all these problems, the ability of the atomic background to absorb the angular momentum produced by the change in magnetization depends on the shear modulus of the material. One should expect that the cases of zero and infinite shear modulus reflect, respectively, our cases of a free particle and a particle in an absolutely rigid matrix. In what follows we will demonstrate this explicitly by calculating the effect of the rotational recoil on the tunneling rate.

The Lagrangian of the system consists of three parts,

$$\mathcal{L}=\mathcal{L}_m+\mathcal{L}_e+\mathcal{L}_{\text{int}}\,, \tag{3}$$

where $\mathcal{L}_m$ is associated with the dynamics of the magnetic moment, $\mathcal{L}_e$ is the Lagrangian of the elastic matrix, and $\mathcal{L}_{\text{int}}$ describes the interaction between **M** and elastic deformations. The magnetic Lagrangian has the form [1-4]

$$\mathcal{L}_m=\gamma^{-1}M\dot{\phi}(\cos\theta-1)-E_a(\theta,\phi)+\mathbf{M}\cdot\mathbf{H}\,, \tag{4}$$

where $\theta(\mathbf{r},t)$ and $\phi(\mathbf{r},t)$ are spherical coordinates of a fixed-length vector **M**, $E_a(\theta,\phi)$ is the energy of the magnetic anisotropy (in which we absorb the magnetic dipole energy as well), and H is the magnetic field. Note that $\mathcal{L}_m$ is proportional to the volume of the particle, V, through $\mathbf{M},E_a\propto V$. The simplest choice of E_a that exhibits tunneling and at the same time provides a reasonable description of the anisotropy of a fine particle is

$$E_a=V[K_\perp M_x^2/M^2-K_\parallel M_z^2/M^2]\,, \tag{5}$$

where $K_\perp$ and $K_\parallel$ are volume-independent anisotropy constants. This form of the anisotropy corresponds to the Y-Z easy plane and the Z easy axis in that plane. The second term in Eq. (5) creates two equilibrium orientations of **M** (parallel and antiparallel to the Z axis) and the energy barrier between them. The first term is responsible for the nonconservation of M_z, that is, its noncommutation with the Hamiltonian in the quantum problem. The tunneling then occurs between up and down orientations of the magnetic moment.

For further consideration, it is convenient to introduce the magnetization of the system,

$$\mathbf{m}(\mathbf{r},t)=\begin{cases}\mathbf{M}(t)/V, & \text{for } \mathbf{r}\in V\,,\\ 0, & \text{otherwise}\,.\end{cases} \tag{6}$$

We will regard the diameter of the particle, d, as the smallest size in the problem. This is justified by the fact that any excitations (magnons, phonons, etc.) inside the particle cost too much energy to contribute to tunneling. Our treatment of the rotational recoil should, therefore, equally apply to the case of an atom having a very large magnetic moment **M**. In this sense, Eq. (6) is equivalent to

$$\mathbf{m}(\mathbf{r},t)=\mathbf{M}(t)\delta(\mathbf{r})\,. \tag{7}$$

The anisotropy of the elastic properties of the system is irrelevant for our conclusions. For that reason we will use the isotropic elastic theory with the Lagrangian

$$\mathcal{L}_e=\int d^3r\left[\frac{1}{2}\rho\dot{u}_i^2-\mu u_{ij}^2-\frac{\lambda}{2}u_{kk}^2\right], \tag{8}$$

where ρ is the density of the system, $\mathbf{u}(\mathbf{r},t)$ is the displacement field, $u_{ij}=\frac{1}{2}(\partial_i u_j+\partial_j u_i)$ is the strain tensor, and μ and $\lambda+\frac{2}{3}\mu$ are the shear modulus and the compression modulus, respectively. Their relation to velocities of transversal and longitudinal phonons is $c_t=(\mu/\rho)^{1/2}$ and $c_l=[(\lambda+2\mu)/\rho]^{1/2}$. For a particle imbedded in a matrix, the density and the elastic moduli of the system, generally speaking, change at the surface of the particle. This effect becomes irrelevant, however, in the limit of small d, when it is sufficient to treat the magnetic moment of the particle as a δ source of the elastic twist in the matrix (see below). We shall assume, therefore, that all coefficients in Eq. (8) refer to the matrix.

Finally, we need the form of the interaction between magnetic and elastic degrees of freedom. Based upon symmetry arguments, the magnetostriction in ferromagnets is approximated by the interaction $a_{iklm}m_im_ku_{lm}$, where $\hat{a}$ is the tensor of magnetoelastic coefficients. In connection with the problem of spin tunneling, this form of coupling has been studied in Ref. [7] (see also Ref. [8]). It was demonstrated that its contribution to the tunneling exponent disappears in the limit of $d\rightarrow 0$. For nanometer size particles its relative contribution is of the order of 10^{-8}-10^{-5}. This result should be taken with caution, however, since the form of the interaction used to derive it only applies to low frequencies and large wavelengths. As we will see, in the limit of small d, the main effect comes from the linear coupling between **M** and **u** which is demanded by the dynamics of the angular momentum. A nice property of this coupling is that it can be written exactly, without any phenomenological constants. According to Eq. (1), the rotation of $\mathbf{M}$ in a small particle must produce the mechanical torque in the matrix,

$$\mathbf{T}(\mathbf{r},t) = -\gamma^{-1}\dot{\mathbf{M}}(t)\delta(\mathbf{r}). \tag{9}$$

On the other hand, the local twist in the material is

$$\mathbf{\Phi}(\mathbf{r},t) = \tfrac{1}{2}\nabla\times\mathbf{u}. \tag{10}$$

This gives the following contribution to the Lagrangian of the system:

$$\mathcal{L}_{\rm int} = \int d^3r\,\mathbf{T}\cdot\mathbf{\Phi} = -\frac{1}{2\gamma}\int d^3r\,\dot{\mathbf{m}}\cdot(\nabla\times\mathbf{u}). \tag{11}$$

The equivalent form, which differs from (11) by a total time derivative, is

$$\mathcal{L}_{\rm int} = \gamma^{-1}\int d^3r\,\mathbf{m}\cdot\dot{\mathbf{\Phi}}, \tag{12}$$

which simply reflects the fact that rotation is equivalent to the magnetic field,

$$\mathbf{h} = \gamma^{-1}\dot{\mathbf{\Phi}}. \tag{13}$$

The boundary term due to the total time derivative can, in principle, be important in the quantum Lagrangian, as it generates an additional phase of the tunneling amplitude. However, for an infinitely large matrix, this term is zero because no elastic twist occurs in the final state. Note that $\dot{\mathbf{u}}/2\gamma$ plays the role of the vector potential in our problem. Based upon this fact, one may try to construct a Chern-Simons term in the Lagrangian. This possibility will be studied elsewhere.

To obtain the tunneling rate, one must evaluate the imaginary-time path integral,

$$\int D\{\mathbf{u}\}D\{\mathbf{M}\}\exp\left[-\frac{1}{\hbar}\int d\tau\,\mathcal{L}_E(\mathbf{u},\mathbf{M})\right], \tag{14}$$

over $\mathbf{u}$ and $\mathbf{M}$ configurations which lead from the initial to the final state. Here $\mathcal{L}_E$ is the imaginary-time version of the total Lagrangian (3). Since we are only interested in the magnetic characteristics of the initial and final states, we should first integrate over $\mathbf{u}$. This is easy because the integral over $\mathbf{u}$,

$$\int D\{\mathbf{u}\}\exp\left\{-\frac{1}{\hbar}\int d\tau\int d^3r\left[\frac{i}{2\gamma}\dot{\mathbf{m}}\cdot(\nabla\times\mathbf{u})+\frac{1}{2}\rho\dot{u}_i^2+\mu u_{ij}^2+\frac{\lambda}{2}u_{kk}^2\right]\right\}, \tag{15}$$

is Gaussian (*dot* now means derivative on τ). This integration leads to the nonlocal in τ, Caldeira-Leggett type, action [9] for $\mathbf{M}$. We will simplify the calculation by assuming that the size of the particle, d, is small compared to c_t/ω_i, where ω_i is the characteristic frequency of the instanton that carries out tunneling. This condition is in accordance with our general line that declares d the smallest scale in the problem. Under this condition, the inertial term $\rho\dot{\mathbf{u}}^2/2$ in Eq. (15) can be neglected, and the standard integration over $\mathbf{u}$ gives the following expression for the effective Euclidean magnetic action,

$$\int d\tau\,\mathcal{L}_{\rm eff} = \int d\tau\,\mathcal{L}_m + \frac{m_0^2 V}{8\gamma^2\mu}\int d\tau\,\dot{\mathbf{n}}^2, \tag{16}$$

where $m_0 = M/V$ and $\mathbf{n} = \mathbf{M}/M$.

According to Eq. (16), the effect of the interaction given by Eqs. (11) and (12) is equivalent to the effective moment of inertia

$$I_e = m_0^2 V/4\gamma^2\mu \tag{17}$$

associated with the rotation of $\mathbf{M}$. The important observation is that this moment of inertia is inversely proportional to the shear modulus of the matrix, making the magnetic moment "too heavy" to tunnel in the limit of $\mu\rightarrow 0$. To assess this effect quantitatively, one should compare I_e with the effective moment of inertia

$$I_m = m_0^2 V/2\gamma^2 K_\perp \tag{18}$$

due to the dynamics of $\mathbf{M}$ itself [4], that is, in the limit of an absolutely rigid atomic background. Comparing Eqs. (17) and (18), one finds that the relative contribution of the rotational recoil to the tunneling rate depends on the ratio $K_\perp/\mu$. More rigorously, the WKB exponent of tunneling

$$B = \frac{1}{\hbar}\int d\tau\,\mathcal{L}_E \tag{19}$$

is proportional to the square root of the effective moment of inertia for $\mathbf{M}$ [4]. Consequently,

$$B \propto \left(\frac{1}{K_\perp}+\frac{1}{2\mu}\right)^{1/2}. \tag{20}$$

The full expression depends on a particular tunneling problem. Consider, e.g., the situation when the external field $\mathbf{H}$ is applied in the direction opposite to $\mathbf{M}$. The most interesting case of a small barrier [4] corresponds to the fine tuning of H to the critical field, $H_c = 2K_\parallel/m_0$, at which the barrier disappears; that is, $H = (1-\epsilon)H_c$ and $\epsilon\rightarrow 0$. In this case,

$$B = \frac{8m_0 V}{3\hbar\gamma}\epsilon^{3/2}\left(\frac{K_\parallel}{K_\perp}+\frac{K_\parallel}{2\mu}\right)^{1/2}. \tag{21}$$

The temperature below which quantum tunneling dominates over thermal transitions is determined by the instanton frequency which is proportional to $(I_m+I_e)^{-1/2}$. This gives

$$k_B T_c \sim \hbar\omega_i \propto \left(\frac{1}{K_\perp}+\frac{1}{2\mu}\right)^{-1/2}. \tag{22}$$

In the case of a small barrier, the exact expression is

$$\omega_i = \frac{2\gamma}{m_0}\epsilon^{1/2}K_\parallel^{1/2}\left(\frac{1}{K_\perp}+\frac{1}{2\mu}\right)^{-1/2}. \tag{23}$$

Let us now check the validity of the condition $d \ll c_t/\omega_i$, under which Eq. (16) was derived. Note that the crossover temperature T_c is independent of the volume of the particle, while the tunneling exponent is proportional to V. Together with the dependence of B and T_c on $K_\perp$, this means that one needs small d and large $K_\perp$ to observe tunneling on a reasonable time scale in an experimentally accessible range of temperatures. Typical values of d and ω_i are [1] $d \sim 30$ Å and $\omega_i \sim 10^9$–10^{10} s^{-1}. The condition $d \ll c_t/\omega_i$ is, therefore, satisfied with a large safety margin in the limit of $K_\perp \ll \mu$. In the opposite limit of $\mu \ll K_\perp$, it becomes the condition on ϵ, $\epsilon \ll m_0^2/8\gamma^2 K_\parallel \rho d^2$.

We have shown that the conservation of the angular momentum in the problem of spin tunneling leads to the model-independent linear interaction of the magnetic moment with the elastic background. This interaction results in the dependences of the tunneling rate and the crossover temperature given by Eqs. (20) and (22). If the magnetic anisotropy and the shear modulus differ on the order of magnitude, *the one which is smaller* determines B and T_c. The smallest of the two ($K_\perp$ or μ) should be large enough, however, to provide sufficiently high values of $\exp(-B)$ and T_c. The magnetic anisotropy in small particles is due to their atomic structure and shape. The shape anisotropy, which originates from the magnetic dipole interaction, puts the lower limit on $K_\perp$ of the order of $\pi m_0^2 \sim 10^5$–10^7 ergs/cm^3. The value of the transversal magnetocrystalline anisotropy depends on the symmetry of the lattice. At low temperature it can be as high (in, e.g., Dy and Tb particles) as 10^8 ergs/cm^3. Thus, in ferromagnetic particles, one should expect $K_\perp \sim 10^5$–10^8 ergs/cm^3. Typical values of μ in conventional solids are 10^{10}–10^{12} ergs/cm^3. Consequently, for such solids, the relative effect of the rotational recoil on the tunneling exponent should not exceed 1%. However, since the experiments on magnetic tunneling require low temperature, which is usually attained by placing the sample in liquid helium, it may not be out of the question to work with small magnetic particles frozen in a helium crystal. The shear modulus of the helium crystal is of the order of 10^8 ergs/cm^3 [10], that is, comparable with the highest values of the magnetic anisotropy in small particles. In this case, the effect of the rotational recoil on tunneling may become large. Note that by varying the pressure, one can produce a quite appreciable change in the shear modulus of the helium crystal. Because of the exponential dependence of the tunneling rate on μ, this could result in the remarkable increase of tunneling on pressure even for weak-anisotropy particles. Measurements of quantum magnetic relaxation in an ensemble of small particles frozen in a helium crystal should enable one to observe this effect. Tunneling should disappear at the melting transition when the matrix loses its ability to absorb the change in the angular momentum. This effect should be especially dramatic when melting into a superfluid phase takes place, which will be the case for the transition on pressure at low temperature [11]. Such an experiment would provide a rare example of macroscopic quantum tunneling in which gradual decoupling of the tunneling variable from the background reduces the tunneling rate.

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