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Renormalization of the tunnel splitting in a rotating nanomagnet

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We study spin tunneling in a magnetic nanoparticle with biaxial anisotropy that is free to rotate about its anisotropy axis. Equations of motion are derived that couple spin and mechanical degrees of freedom and the exact instanton of these equations is obtained. We show that mechanical freedom of the particle renormalizes magnetic anisotropy and increases the tunnel splitting.

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The macroscopic dynamics of a fixed-length magnetic moment \mathbf{M} of a single-domain ferromagnetic particle is described by the Landau-Lifshitz equation.^{1,2} When dissipation (which is usually weak) is neglected this equation reads

$$\frac{\partial \mathbf{M}}{\partial t} = \gamma \mathbf{M} \times \mathbf{B}_{\text{eff}}, \quad \mathbf{B}_{\text{eff}} = -\frac{\delta \mathcal{E}}{\delta \mathbf{M}}, \quad (1)$$

where \mathcal{E} is the classical magnetic energy of the particle which depends on the orientation of \mathbf{M} . It was shown long ago^{3,4} that Eq. (1), besides the real-time solutions, also possesses imaginary-time solutions—instantons—that describe macroscopic quantum tunneling of \mathbf{M} between classically degenerate energy minima (see also the books Refs. 2 and 5). They provide the tunnel splitting of classically degenerate spin states. Magnetic instantons have been intensively studied for various symmetries of the crystal field Hamiltonian and also for problems involving spatial dependence of the spin field, such as, e.g., tunneling of domain walls and nucleation of magnetic bubbles.⁵ The effect of interaction of the tunneling spin with the dissipative environment (phonons, nuclear spins, etc.) has been another active field of study.

In early experiments on spin tunneling⁵ single-domain magnetic particles were always frozen in a solid matrix so that their physical position and orientation were fixed and only rotation of the magnetic moment was allowed. Later, beams of small magnetic clusters were investigated,^{7–12} and more recently free magnetic nanoparticles confined within solid nanocavities have been studied.⁶ Experimentalists have also worked with molecular nanomagnets deposited on surfaces^{13–16} or carbon nanotubes,¹⁷ as well as with single magnetic molecules bridged between metallic electrodes.^{18–22} In such experiments the particles retain some mechanical freedom. This inspired recent theoretical work on the quantum mechanics of rotating magnets.^{23–26} In this Brief Report we study the tunnel splitting in a nanomagnet that is free to rotate about its magnetic anisotropy axis. An interesting example of exact magnetic instanton will be given that mixes the spin and mechanical degrees of freedom. Our results may be relevant to recent experiments with single magnetic molecules.

As is well known, even the classical problem of the rotation of a rigid body is rather involved. In a quantum problem the energy levels related to the rotational degrees of freedom can be obtained analytically only for a symmetric body.²⁷ Even this simplified problem becomes significantly more complicated when the rotating body possesses a spin. Nevertheless, as was recently demonstrated in Ref. 23, the exact low-energy eigenstates can be obtained analytically for a nanomagnet

that, due to a large magnetic anisotropy, can be described as a two-state spin system and is free to rotate about its magnetic anisotropy axis. It was shown that the crucial role is played by the parameter

$$\alpha = 2(\hbar S)^2 / (I \Delta). \quad (2)$$

Here S is the dimensionless total spin of the nanomagnet, I is its moment of inertia, and Δ is the spin-tunnel splitting. At $\alpha < \alpha_1 \equiv [1 - 1/(2S)^2]^{-1}$ the ground state and the first excited state are respectively symmetric and antisymmetric superpositions of $\mathbf{J} = \mathbf{L} + \mathbf{S} = \mathbf{0}$ states shown in Fig. 1. Such superpositions are characterized by zero magnetic moment. At $\alpha > \alpha_1$ the nanomagnet develops a finite magnetic moment. In the limit of $\alpha \rightarrow \infty$ the spin localizes in one of the two directions along the magnetic anisotropy axis.²³ This can be understood qualitatively based upon the following argument. Delocalization of \mathbf{S} in the spin space due to tunneling transitions between $|S\rangle$ and $|-S\rangle$ lowers the ground state energy by $\Delta/2$. However, for a magnet that is free to rotate this can only occur if the spin tunneling is accompanied by cotunneling of \mathbf{L} , so that $\mathbf{J} = \mathbf{L} + \mathbf{S}$ is conserved (Fig. 1). If the corresponding mechanical energy $(\hbar S)^2 / (2I)$ is large compared to $\Delta/2$, which is the case of large α , then tunneling cannot lower the energy and the spin localizes in one of the $|\pm S\rangle$ states.

In this Brief Report we ask a question that has not been previously addressed to our knowledge. How does the mechanical freedom of the nanomagnet change the tunnel splitting Δ ? When rotations are treated semiclassically the answer to this question can be obtained by studying instantons that connect the two states shown in Fig. 1. This problem is rather interesting as the corresponding instantons should mix spin and mechanical degrees of freedom. It also has practical implications for experimental studies of magnetic molecules and atomic clusters that are free to rotate. Such molecules or clusters should have very weak coupling to the environment, which eliminates or greatly reduces the effect of dissipation on tunneling. Below we find the exact instanton solution of the equations of motion describing the dynamics of the magnetic moment and the rotation of the nanomagnet. This solution shows that mechanical freedom renormalizes the tunnel splitting Δ . However, this renormalization is small unless Δ is very large and α is close to α_1 .

Consider a high-spin magnetic particle with biaxial anisotropy that is free to rotate about its easy axis, which we choose to be the X axis. This can be, e.g., a small atomic cluster

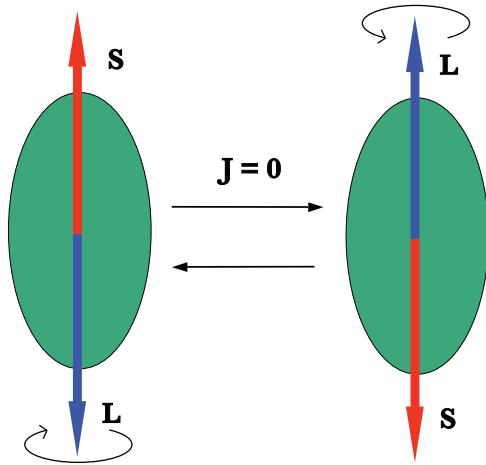


FIG. 1. (Color online) Spin and angular momentum cotunneling in a magnetic nanoparticle that is free to rotate about its anisotropy axis. To conserve the total angular momentum $\mathbf{J} = \mathbf{S} + \mathbf{L}$, tunneling of a spin \mathbf{S} should be accompanied by cotunneling of the mechanical angular momentum \mathbf{L} . The resulting $J = 0$ quantum state is the ground state of the nanomagnet at $\alpha < \alpha_1$.

held inside an asymmetric trap or a molecule held between two tips. Our choice of X as the rotation axis is dictated by necessity to simplify the mathematics of the problem. We define coordinate systems of the laboratory frame (x, y, z) and particle frame (X, Y, Z) . In the particle frame the x axis is along the easy axis in the xy easy plane, and the z axis is the hard axis. The laboratory frame is centered at the same origin such that the X axis of the laboratory frame coincides with the x axis of the particle frame. The particle is free to rotate about this axis. At some initial time $t = 0$ we choose the two coordinate frames to coincide. The angle of rotation of the y and z axes with respect to the Y and Z axes is $\varphi(t)$.

The initial state of the particle is such that its total angular momentum is zero, $\mathbf{J} = \mathbf{S} + \mathbf{L} = \mathbf{0}$. In other words, the total spin (magnetization) vector points along the easy axis and the magnet rotates about this axis such that these angular momenta are equal in magnitude and opposite in direction. The exchange interaction between individual spins is strong, so the magnitude of the total spin of the particle is a constant. The magnetic energy will be expressed below in terms of \mathbf{M} which is proportional to the spin, $\hbar\mathbf{S} = \mathbf{M}/\gamma$. Here $\gamma = -e/2mc < 0$ is the electron gyromagnetic ratio. The orbital angular momentum is associated with the rotational motion of the particle itself, $\hbar\mathbf{L} = I\dot{\varphi}$, where I is the particle's moment of inertia and $\dot{\varphi}$ is its angular velocity.

The anisotropy energy is naturally defined in the particle frame: $\mathcal{E}_A = k_{\perp}M_z^2 - k_{\parallel}M_x^2$. It can be written in terms of spherical polar coordinates (θ, ϕ) which are defined with respect to the particle-frame axes. Up to an unessential constant,

$$\mathcal{E}_A(\theta, \phi) = \frac{1}{2}\mu_0 M_0^2 V [(K_{\perp} + K_{\parallel} \cos^2 \phi) \cos^2 \theta + K_{\parallel} \sin^2 \phi]. \quad (3)$$

Here μ_0 is the magnetic permeability of vacuum, V is the volume of the particle, and $M_0 = M/V$ represents the magnetization, which is a constant of the ferromagnetic material. The anisotropy constants $K_{\perp} > 0$ and $K_{\parallel} > 0$ have

been redefined to show the explicit proportionality of the anisotropy energy to the volume. They are dimensionless numbers, typically of order unity.

The rotational kinetic energy of the particle is $\mathcal{E}_R = \frac{1}{2}I\dot{\varphi}^2$. The Lagrangian of our system consists of the trivial kinetic Lagrangian for the variable φ ,

$$\mathcal{L}_L = \hbar\mathbf{L} \cdot \dot{\varphi} - \mathcal{E}_R = \frac{1}{2}I\dot{\varphi}^2, \quad (4)$$

and the magnetic Lagrangian,

$$\mathcal{L}_S = \left(\frac{M_0 V}{\gamma}\right) \dot{\phi} \cos \theta - \mathcal{E}'_A(\theta, \phi), \quad (5)$$

for the variables θ and ϕ . The first term in Eq. (5) follows from the fact that $\hbar S_z = \hbar S \cos \theta$ is the generalized momentum for the coordinate ϕ . The second term is the effective magnetic energy in the rotating frame,

$$\mathcal{E}'_A = \mathcal{E}_A(\theta, \phi) - \hbar\mathbf{S} \cdot \dot{\varphi}. \quad (6)$$

The last term in this equation is related to the fact that in the particle frame the rotation is equivalent to the magnetic field $\mathbf{B} = \dot{\varphi}/\gamma$.

The total Lagrangian is a sum of \mathcal{L}_L and \mathcal{L}_S :

$$\mathcal{L} = \hbar(\mathbf{L} + \mathbf{S}) \cdot \dot{\varphi} + \frac{M_0 V}{\gamma} \dot{\phi} \cos \theta - \left[\frac{1}{2}I\dot{\varphi}^2 + \mathcal{E}_A(\phi, \theta) \right]. \quad (7)$$

The first term reflects the fact that in the presence of a spin the generator of rotations is $\mathbf{J} = \mathbf{L} + \mathbf{S}$. The explicit form of the total Lagrangian in terms of the generalized coordinates θ , ϕ , and φ is

$$\begin{aligned} \mathcal{L} = & \frac{1}{2}I\dot{\varphi}^2 + \left(\frac{M_0 V}{\gamma}\right) \dot{\phi} \cos \theta + \left(\frac{M_0 V}{\gamma}\right) \dot{\phi} \sin \theta \cos \phi \\ & - \frac{1}{2}\mu_0 M_0^2 V [(K_{\perp} + K_{\parallel} \cos^2 \phi) \cos^2 \theta + K_{\parallel} \sin^2 \phi]. \end{aligned} \quad (8)$$

The equations of motion are Euler-Lagrange equations for θ , ϕ , and φ :

$$\frac{d\phi}{d\tilde{t}} = (K_{\perp} + K_{\parallel} \cos^2 \phi) \cos \theta + \dot{\varphi} \frac{\cos \theta \cos \phi}{\sin \theta}, \quad (9)$$

$$\frac{d(\cos \theta)}{d\tilde{t}} = -K_{\parallel} \cos \phi \sin \phi \sin^2 \theta - \dot{\varphi} \sin \theta \sin \phi, \quad (10)$$

$$\frac{d}{d\tilde{t}} \left[I\dot{\varphi} + \frac{V}{\mu_0 \gamma^2} \cos \phi \sin \theta \right] = 0, \quad (11)$$

where we have introduced dimensionless time $\tilde{t} = \gamma\mu_0 M_0 t$ and $\dot{\varphi} = d\varphi/d\tilde{t}$.

Note that the equations of motion for ϕ and θ can also be obtained from the Landau-Lifshitz equation with $\mathcal{E} = \mathcal{E}'_A$. Indeed, the equations for ϕ and θ that follow from Eq. (1) (see, e.g., Ref. 2),

$$\frac{\partial \phi}{\partial \tilde{t}} = -\frac{\gamma}{M \sin \theta} \left(\frac{\partial \mathcal{E}'_A}{\partial \theta} \right), \quad \frac{\partial \theta}{\partial \tilde{t}} = \frac{\gamma}{M \sin \theta} \left(\frac{\partial \mathcal{E}'_A}{\partial \phi} \right), \quad (12)$$

are identical to Eqs. (9) and (10). The third equation of motion, Eq. (11), is the conservation of the total angular momentum:

$\frac{d}{dt}[L_x + S_x] = \frac{d}{dt}J_x = 0$. At $J_x = 0$ it is equivalent to the constraint

$$I\dot{\phi} = -\frac{M_0 V}{\gamma} \sin\theta \cos\phi. \quad (13)$$

With account of this constraint the equations of motion for ϕ and θ become

$$\frac{d\phi}{d\tilde{t}} = (K_{\perp} + K'_{\parallel} \cos^2\phi) \cos\theta, \quad (14)$$

$$\frac{d(\cos\theta)}{d\tilde{t}} = -K'_{\parallel} \cos\phi \sin\phi \sin^2\theta, \quad (15)$$

where

$$K'_{\parallel} = K_{\parallel} - K_R, \quad K_R = \frac{V}{\mu_0 \gamma^2 I}. \quad (16)$$

We see that for $\mathbf{J} = \mathbf{0}$ the effect of rotations reduces to the renormalization of the easy-axis anisotropy K_{\parallel} . This is easy to understand from the following consideration. In a state with $J_x = 0$, equilibrium vectors \mathbf{S} and \mathbf{L} point in opposite directions along the x axis. If \mathbf{S} deviates from the x axis, S_x decreases and so should L_x to preserve the condition $J_x = 0$. The decrease of L_x corresponds to a decrease of the rotational energy $(\hbar L_x)^2/(2I)$, mandated by $J_x = 0$. Thus, effectively, the magnetic anisotropy energy associated with the deviation of \mathbf{S} from the easy axis becomes smaller when mechanical rotation is allowed.

We should now look for solutions of Eqs. (14) and (15). We first notice that

$$\mathcal{E} = \frac{1}{2}\mu_0 M_0^2 V [(K_{\perp} + K'_{\parallel} \cos^2\phi) \cos^2\theta + K'_{\parallel} \sin^2\phi] \quad (17)$$

is the integral of motion. This is easy to see by differentiating this equation with respect to time and substituting in the resulting equation the time derivatives $\dot{\phi}$ and $\dot{\theta}$ from Eqs. (14) and (15). Not surprisingly, up to a constant, Eq. (17) equals the total energy of the particle in the laboratory frame, $\mathcal{E} = \mathcal{E}_A + \frac{1}{2}I\dot{\phi}^2$, with account of the constraint (13). Equation (14) gives

$$\cos\theta = \frac{d\phi/d\tilde{t}}{K_{\perp} + K'_{\parallel} \cos^2\phi}. \quad (18)$$

This allows us to express \mathcal{E} in terms of the angle ϕ and its time derivative:

$$\mathcal{E} = \frac{1}{2}M_0^2 V \left[\frac{(d\phi/d\tilde{t})^2}{K_{\perp} + K'_{\parallel} \cos^2\phi} + K'_{\parallel} \sin^2\phi \right]. \quad (19)$$

Since this expression is positively defined [see the discussion after Eq. (26)], the classical energy minima occur at $\mathcal{E} = 0$. They correspond to the stationary magnetization pointing in either direction along the easy axis, i.e., $\phi = 0, \pi$ with $\cos\theta = 0$ in accordance with Eq. (18).

Equation $\mathcal{E} = 0$ has no real-time solutions for ϕ that connect the two degenerate classical energy minima. However, in imaginary time, $\tilde{\tau} = i\tilde{t}$, the equation $\mathcal{E} = 0$ is equivalent to

$$\left(\frac{d\phi}{d\tilde{\tau}} \right)^2 = K'_{\parallel} \sin^2\phi (K_{\perp} + K'_{\parallel} \cos^2\phi). \quad (20)$$

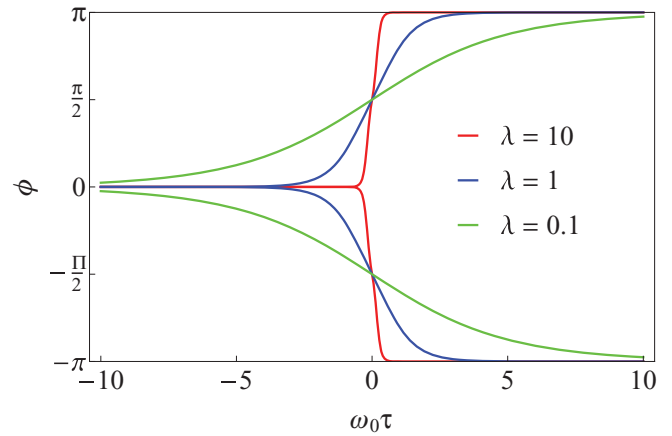


FIG. 2. (Color online) Dependence of ϕ on τ for clockwise and counterclockwise underbarrier rotations of \mathbf{M} .

Such equation has instanton solutions that connect the classical energy minima:

$$\phi(\tau) = \pm \arccos \left\{ \frac{\sinh(\omega_0 \tau)}{\sqrt{\lambda + \cosh^2(\omega_0 \tau)}} \right\}. \quad (21)$$

The τ dependence of θ is given by Eq. (18), and the τ dependence of ϕ is given by Eq. (13). Here

$$\lambda = K'_{\parallel}/K_{\perp}, \quad \omega_0 = |\gamma| \mu_0 M_0 \sqrt{K'_{\parallel}(K'_{\parallel} + K_{\perp})}. \quad (22)$$

Positive and negative signs correspond to the two possible trajectories, which are counterclockwise and clockwise rotations of the magnetization from $\phi = 0$ at $\tau = -\infty$ to $\phi = \pm\pi$ at $\tau = +\infty$, respectively; see Fig. 2.

The tunnel splitting has the form $\Delta = Ae^B$, where A is of the order of quantized oscillations near the minimum of the potential well and $B = \frac{i}{\hbar} \int_{-\infty}^{+\infty} dt \mathcal{L}$ is the WKB exponent. Substituting here \mathcal{L} of Eq. (7) at $\mathbf{J} = \mathbf{0}$, one obtains for the instanton trajectory

$$B = -S \ln \left(\frac{\sqrt{1+\lambda} + \sqrt{\lambda}}{\sqrt{1+\lambda} - \sqrt{\lambda}} \right). \quad (23)$$

In a microscopic theory the easy-axis crystal field is presented as $-DS_z^2$. The connection between D and the parameter K_{\parallel} of the macroscopic theory is²

$$K_{\parallel} = \left(2 - \frac{1}{s} \right) \frac{DV_0}{\mu_0(\hbar\gamma)^2}, \quad (24)$$

where s is the spin per unit cell of the crystal and V_0 is the volume of the unit cell. (The singularity at $s = 1/2$ reflects the fact that single-ion magnetic anisotropy does not exist for spin $1/2$.) The total spin of a ferromagnetic particle can be presented as $S = s(V/V_0)$. Consequently,

$$\frac{K_R}{K_{\parallel}} = \frac{S\hbar^2}{(2s-1)ID} = \frac{\Delta}{(2s-1)2SD} \alpha, \quad (25)$$

where we have used Eqs. (2) and (16). Renormalization of the easy-axis anisotropy by rotations can be presented in the form $K'_{\parallel} = K_{\parallel}(1 - \epsilon)$, where

$$\epsilon = \left(\frac{\alpha}{2S}\right) \left(\frac{1 - \frac{1}{2S}}{1 - \frac{1}{2s}}\right) \frac{\Delta}{E_1} \quad (26)$$

and $E_1 = (2S - 1)D$ is the energy of the first excited spin state at $\Delta = 0$.

The low-energy limit that we are studying corresponds to $\Delta \ll E_1$ and $\alpha < \alpha_1$. In this limit ϵ is small. Consider, e.g., the case of large S and large λ (small tunneling rate). According to Eq. (23) in this case $B = -S \ln(4\lambda)$ so that $\Delta \propto \exp[-S \ln(4\lambda)]$. It is easy to see from this expression

that mechanical rotation renormalizes Δ by a factor $\exp(\epsilon S)$. Normally it would not be large compared to 1. However, since small ϵ in the exponent is multiplied by a large S , it is not out of the question that at sufficiently large Δ a slight increase of the tunnel splitting would be observable in ESR studies of spin clusters that are free to rotate. For, e.g., a typical magnetic molecule with $S = 10$, $I \sim 10^{-35} \text{ g cm}^2$, and $E_1 \sim 10 \text{ K}$, the condition $\alpha = \alpha_1$ corresponds to $\Delta \sim 0.1 \text{ K}$ and $\epsilon S \sim 0.1$, which would result in a small but experimentally detectable difference in the tunnel splitting for the molecule firmly coupled to a solid and the molecule that is free to rotate.

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