

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. Exact instanton of the coupled equations of motion is found that connects degenerate classical energy minima. We show that mechanical freedom of the particle renormalizes magnetic anisotropy and increases the tunnel splitting.

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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

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

where \mathcal{E} is the classical magnetic energy of the particle that 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 books: Refs. 2 and 5). 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 investigated⁷⁻¹² and more recently free magnetic nanoparticles confined within solid nanocavities have been studied⁶. Experimentalists have also worked with molecular nanomagnets deposited on surfaces¹³⁻¹⁶ or carbon nanotubes¹⁷, as well as with single magnetic molecules bridged between metallic electrodes¹⁸⁻²². In such experiments the particles retain some mechanical freedom. This inspired recent theoretical work on quantum mechanics of rotating magnets²³⁻²⁶. In this paper we obtain exact magnetic instanton for a single-domain particle that is free to rotate about its anisotropy axis.

General analytical solution for the rotational quantum levels of a rigid body does not exist²⁷. Spin degree of freedom makes this problem even less tractable. However, as was recently demonstrated in Ref. 23, the exact eigenstates and exact energy levels 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. Such a system at rest is described by the Hamiltonian $\hat{\mathcal{H}}_\sigma = -(\Delta/2)\sigma_x$, where σ_x is a Pauli matrix and Δ is the tunnel splitting of spin-up and spin-down states, with z being the anisotropy axis. It arises from the terms in the full Hamiltonian that do not commute with the z -component of the total spin \mathbf{S} . Rotation of the particle by the angle φ about the Z -axis transforms the two-state

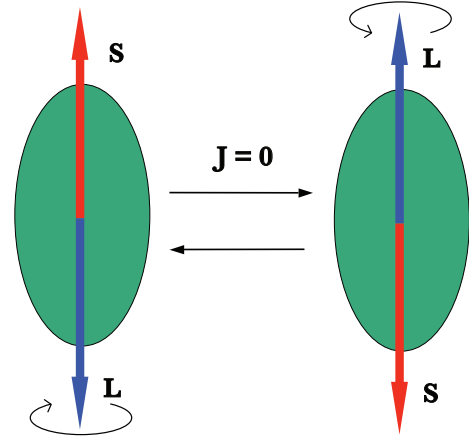


FIG. 1. Color online: Spin and angular momentum tunneling in a rotating magnetic nanoparticle.

Hamiltonian into²³

$$\hat{\mathcal{H}}'_\sigma = -\frac{\Delta}{2} [\cos(2S\varphi)\sigma_x + \sin(2S\varphi)\sigma_y], \quad (2)$$

(S is dimensionless). Exact eigenvalues of the full Hamiltonian of a nanomagnet rotating about its anisotropy axis, $\hat{\mathcal{H}} = (\hbar L_z)^2/(2I) + \hat{\mathcal{H}}'_\sigma$ (where L_z is the z -component of the dimensionless mechanical angular momentum and I is the moment of inertia), were obtained in Ref. 23, where it was shown that parameter

$$\alpha = 2(\hbar S)^2/(I\Delta), \quad (3)$$

determines low energy states of the particle. 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 $\mathbf{J} = \mathbf{L} + \mathbf{S} = 0$ states shown in Fig. 1, with energies $E_\pm = \frac{\hbar^2 S^2}{2I} \pm \frac{\Delta}{2}$.

Derivation of these results was based upon the assumption that the parameter Δ is the same for a stationary magnetic particle and for a particle that is free to rotate. Instanton method allows one to test this assumption. Below we find the exact instanton solution of the equations

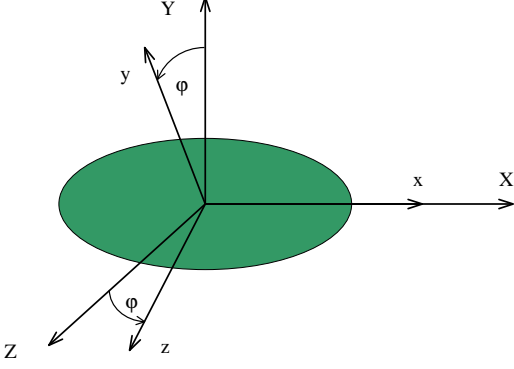


FIG. 2. Geometry of particle and laboratory frames.

of motion describing the dynamics of the magnetic moment and the rotation of the particle. It shows that mechanical freedom does renormalize 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. The initial state of the particle is such that its total angular momentum is zero, $\mathbf{J} = \mathbf{S} + \mathbf{L} = 0$. In other words, the total spin (magnetization) vector points along the easy axis and the particle rotates about this axis such that these angular momenta are equal in magnitude and opposite in direction, see Fig. 1. 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, $\hbar\mathbf{L} = I\dot{\boldsymbol{\varphi}}$, where I is the particle's moment of inertia and $\dot{\boldsymbol{\varphi}}$ is its angular velocity.

We define coordinate systems of the lab frame (x, y, z) and particle frame (X, Y, Z) as shown in Fig. 2. 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 lab frame is centered at the same origin such that the X -axis of the lab 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 x - and z -axes with respect to the X - and Z -axes is $\varphi(t)$. Notice the change of the easy axis as compared to the choice of Ref. 23, which is dictated by the mathematics of the problem.

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 de-

finied with respect to the particle-frame axes,

$$\mathcal{E}_A(\theta, \phi) = \frac{1}{2}\mu_0M_0^2V[(K_{\perp} + K_{\parallel}\cos^2\phi)\cos^2\theta + K_{\parallel}\sin^2\phi]. \quad (4)$$

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

$$\mathcal{L}_L = \hbar\mathbf{L} \cdot \dot{\boldsymbol{\varphi}} - \mathcal{E}_R = \frac{1}{2}I\dot{\boldsymbol{\varphi}}^2, \quad (5)$$

and the magnetic Lagrangian,

$$\mathcal{L}_S = \left(\frac{M_0V}{\gamma}\right)\dot{\phi}\cos\theta - \mathcal{E}'_A(\theta, \phi), \quad (6)$$

for the variables θ and ϕ . The first term in Eq. (6) 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,

$$\mathcal{E}'_A = \mathcal{E}_A(\theta, \phi) - \hbar\mathbf{S} \cdot \dot{\boldsymbol{\varphi}}. \quad (7)$$

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

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

$$\mathcal{L} = \hbar(\mathbf{L} + \mathbf{S}) \cdot \dot{\boldsymbol{\varphi}} + \left(\frac{M_0V}{\gamma}\right)\dot{\phi}\cos\theta - \left[\frac{1}{2}I\dot{\boldsymbol{\varphi}}^2 + \mathcal{E}_A(\phi, \theta)\right]. \quad (8)$$

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

$$\begin{aligned} \mathcal{L} = & \frac{1}{2}I\dot{\boldsymbol{\varphi}}^2 + \left(\frac{M_0V}{\gamma}\right)\dot{\phi}\cos\theta + \left(\frac{M_0V}{\gamma}\right)\dot{\varphi}\sin\theta\cos\phi \\ & - \frac{1}{2}\mu_0M_0^2V[(K_{\perp} + K_{\parallel}\cos^2\phi)\cos^2\theta + K_{\parallel}\sin^2\phi]. \end{aligned} \quad (9)$$

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

$$\frac{d\phi}{dt} = (K_{\perp} + K_{\parallel}\cos^2\phi)\cos\theta + \dot{\varphi}\frac{\cos\theta\cos\phi}{\sin\theta} \quad (10)$$

$$\frac{d(\cos\theta)}{dt} = -K_{\parallel}\cos\phi\sin\phi\sin^2\theta - \dot{\varphi}\sin\theta\sin\phi \quad (11)$$

$$\frac{d}{dt}\left[I\dot{\boldsymbol{\varphi}} + \frac{V}{\mu_0\gamma^2}\cos\phi\sin\theta\right] = 0, \quad (12)$$

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

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),

$$\frac{\partial\phi}{\partial t} = -\frac{\gamma}{M \sin\theta} \left(\frac{\partial\mathcal{E}'_A}{\partial\theta} \right), \quad \frac{\partial\theta}{\partial t} = \frac{\gamma}{M \sin\theta} \left(\frac{\partial\mathcal{E}'_A}{\partial\phi} \right), \quad (13)$$

are identical to the equations (10) and (11). The third equation of motion, Eq. 12, is the conservation of the total angular momentum:

$$\frac{d}{dt}[L_X + S_X] = \frac{d}{dt}J_X = 0. \quad (14)$$

At $J_X = 0$ it is equivalent to the constraint:

$$I\dot{\phi} = -\frac{M_0 V}{\gamma} \sin\theta \cos\phi. \quad (15)$$

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

$$\frac{d\phi}{dt} = (K_{\perp} + K'_{\parallel} \cos^2\phi) \cos\theta \quad (16)$$

$$\frac{d(\cos\theta)}{dt} = -K'_{\parallel} \cos\phi \sin\phi \sin^2\theta, \quad (17)$$

where

$$K'_{\parallel} = K_{\parallel} - K_R, \quad K_R = \frac{V}{\mu_0\gamma^2 I}. \quad (18)$$

We see that for $\mathbf{J} = 0$ 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} look in the 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 the 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 equations (16) and (17). We first notice that

$$\mathcal{E} = \frac{1}{2}\mu_0 M_0^2 V [(K_{\perp} + K'_{\parallel} \cos^2\phi) \cos^2\theta + K'_{\parallel} \sin^2\phi] \quad (19)$$

is the integral of motion. This is easy to see by differentiating this equation on time and substituting in the resulting equation the time derivatives ϕ and θ from equations (16) and (17). Not surprisingly, up to a constant, Eq. (19) 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 (15). Eq. (16) gives

$$\cos\theta = \frac{d\phi/d\tilde{t}}{K_{\perp} + K'_{\parallel} \cos^2\phi}. \quad (20)$$

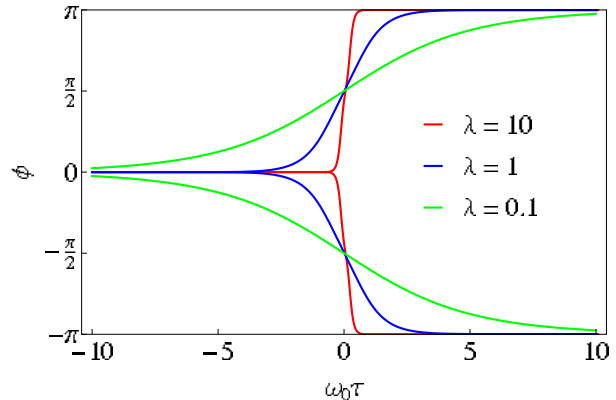


FIG. 3. Dependence of ϕ on τ for clockwise and counterclockwise underbarrier rotations of \mathbf{M} .

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

$$\mathcal{E} = \frac{1}{2}M_0^2 V \left[\frac{(d\phi/d\tilde{t})^2}{K_{\perp} + K'_{\parallel} \cos^2\phi} + K'_{\parallel} \sin^2\phi \right]. \quad (21)$$

Since this expression is positively defined, 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. (20).

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

$$\left(\frac{d\phi}{d\tilde{\tau}} \right)^2 = K'_{\parallel} \sin^2\phi (K_{\perp} + K'_{\parallel} \cos^2\phi). \quad (22)$$

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

$$\phi(\tau) = \pm \arccos \left\{ -\frac{\sinh(\omega_0\tau)}{\sqrt{\lambda + \cosh^2(\omega_0\tau)}} \right\}. \quad (23)$$

The τ -dependence of θ is given by Eq. (20), and the τ -dependence of φ is given by Eq. (15). Here

$$\lambda = K'_{\parallel}/K_{\perp}, \quad \omega_0 = |\gamma|\mu_0 M_0 \sqrt{K'_{\parallel}(K'_{\parallel} + K_{\perp})}. \quad (24)$$

The 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. 3.

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

$$B = \frac{i}{\hbar} \int_{-\infty}^{+\infty} dt \mathcal{L} \quad (25)$$

is the WKB exponent. Substituting here \mathcal{L} of Eq. (8) at $\mathbf{J} = 0$, one obtains for the instanton trajectory

$$B = -S \ln \left(\frac{\sqrt{1+\lambda} + \sqrt{\lambda}}{\sqrt{1+\lambda} - \sqrt{\lambda}} \right). \quad (26)$$

To see the effect of the mechanical freedom of the particle on spin tunneling we define a dimensionless parameter $\alpha' = K_R/K_{\parallel}$. 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

$$K_{\parallel} = \left(2 - \frac{1}{s} \right) \frac{DV_0}{\mu_0(\hbar\gamma)^2}, \quad (27)$$

where s is spin per unit cell of the crystal and V_0 is the volume of the unit cell. (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,

$$\alpha' = \frac{S\hbar^2}{(2s-1)ID} = \frac{\Delta}{(2s-1)2SD} \alpha, \quad (28)$$

where we have used Eqs. (3) and (18). Renormalization of the easy-axis anisotropy by rotations can be presented in the form $K'_{\parallel} = K_{\parallel}(1 - \epsilon)$, where

$$\epsilon = \left(\frac{\alpha}{2s} \right) \left(\frac{1 - \frac{1}{2S}}{1 - \frac{1}{2s}} \right) \frac{\Delta}{E_1} \quad (29)$$

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 $\alpha < \alpha_1 \equiv [1 - 1/(2S)^2]^{-1}$, see Ref. 23. In this limit ϵ is small. Consider, e.g., the case of large S and large λ (small tunneling rate). According to Eq. (26) 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 one. However, since small ϵ in the exponent is multiplied by a large S , it is not out of question that at sufficiently large Δ a slight increase of the tunnel splitting would be observable in spin clusters that are free to rotate.

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