Suppression of Tunneling by Interference in Half-Integer-Spin Particles

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Within a wide class of ferromagnetic and antiferromagnetic systems, quantum tunneling of magnetization direction is spin-parity dependent: it vanishes for magnetic particles with half-integer spin, but is allowed for integer spin. A coherent-state path-integral calculation shows that this topological effect results from interference between tunneling paths.

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The search for new systems which exhibit quantum phenomena at the mesoscopic scale has led to a great deal of activity in recent years on nanometer-size magnetic particles. In such particles it is possible for the electronic spins to be locked together into a well-ordered state, either aligned (ferromagnetic) or antialigned (antiferromagnetic), but whose direction can rotate. For common forms of magnetic anisotropy the magnetic vector has two or more low-energy directions. Several recent investigations have focused on the possibility that the magnetic vector will pass between these directions by quantum tunneling [1, 2]. While the tunneling rates are predicted theoretically to be exponentially small in the size of the magnetic particles, recent experiments [3, 4] suggest that quantum tunneling is observable in particles with several thousand spins, which are available to present technology.

In this Letter we show that for a wide range of systems, *quantum tunneling is completely suppressed if the total spin of the magnetic particle is half integral but is allowed in integral-spin particles* [5]. An important experimental implication of this result is that in ensembles of magnetic particles in which the exact number of spins per particle is not precisely controlled (the typical case with present technology), half of those particles (those with an odd number of electrons) will not exhibit quantum tunneling. Such parity effects are well known in atomic physics, but have not been previously noticed for magnetic particles [6]. We show in several specific examples below that this suppression has a topological origin and arises as a destructive interference between different tunneling paths. Thus we find that one quantum effect (tunneling) is suppressed by another (interference), leading to "classical" behavior in half-integer spin systems.

We begin with a reanalysis of the tunneling behavior of a small ferromagnetic particle with easy-plane–easy-axis anisotropy in the superparamagnetic limit where it behaves like a single large spin—referred to as "model I" in [7]. The classical anisotropy energy E has the form $E(\hat{\mathbf{n}}) = E(\theta,\phi) = K_z\sigma^2\cos^2\theta + K_y\sigma^2\sin^2\theta\sin^2\phi$, where $\hat{\mathbf{n}}$ is the magnetization direction and $K_z > K_y > 0$; this energy corresponds to a quantum spin Hamiltonian,

$$H = K_z\sigma_z^2 + K_y\sigma_y^2, \tag{1}$$

where σ is the particle's total spin, and $M_0 = \mu_B\sigma$ is its magnetic moment, with μ_B the Bohr magneton. We are interested in the tunneling of the magnetization direction $\hat{\mathbf{n}}$ between its two equivalent low-energy directions at the points $\theta=\pi/2$, $\phi=0$ and $\theta=\pi/2$, $\phi=\pi$, corresponding to the coherent-state kets $|0\rangle$ and $|\pi\rangle$, respectively. To compute the tunneling rate P [8] we consider the imaginary time transition amplitude expressed as a coherent-state path integral for spins [9–12],

$$\langle\pi|e^{-\beta H}|0\rangle = \int_{\theta(0),\phi(0)}^{\theta(\beta),\phi(\beta)} D\Omega e^{-S_E}, \tag{2}$$

where β^{-1} is the temperature, $D\Omega \sim \prod_\tau d\phi_\tau d\theta_\tau \sin\theta_\tau$, and where the integral runs over all paths (i.e., magnetization directions) connecting one minimum to the other. The Lagrangian L occurring in the Euclidean action $S_E = \int_0^\beta d\tau L$ is given by

$$L = i\sigma\dot{\phi} - i\sigma\dot{\phi}\cos\theta + E \equiv i\sigma\dot{\phi} + L_0. \tag{3}$$

The first two terms of Eq. (3) define the Wess-Zumino term [10] which is of crucial importance in the following. For any path on the 2-sphere S_2, parametrized by $\phi(\tau)$ and $\theta(\tau)$, this contribution to the action is equal to $i\sigma$ times the area swept out on S_2 between the path and the north pole; for closed paths this has exactly the form of the Berry phase [12–14].

The first term of Eq. (3) has some special features which require discussion. It is a total derivative term, which, when integrated, gives the boundary contribution

$$i\sigma\int_0^\beta d\tau\dot{\phi} = i\sigma[\phi(\beta) - \phi(0) + 2n\pi] \tag{4}$$

to S_E. Here n is a winding number counting the number of times which the path wraps around the north pole. As a total derivative, this term has no effect on the classical (Bloch) equations of motion, which can be derived by extremizing just the L_0 piece of the action [7]. As a consequence, this boundary term is commonly ignored. We show here, however, that this term is crucial for the quantum properties of the magnetic particle, making the tunneling behavior of integral and half-integral spins strikingly different.

This result is most clearly seen by treating the transition amplitude of Eq. (2) within an instanton approximation (i.e., saddle-point evaluation) [8]. In the easy-plane–easy-axis model, the saddle-point paths remain near $\theta = \pi/2$ (cf. [15]), so they can be characterized by their winding in the ϕ variable only. This model has the important feature that the passage from $\phi = 0$ to $\phi = \pi$ can be accomplished either by an instanton or an anti-instanton path, i.e., a clockwise or counterclockwise winding over the barrier; see Fig. 1(a). Then the propagator of Eq. (2) is approximated by a sum over paths comprising a sequence of instantons and anti-instantons winding over the barrier. The expression for low temperatures is

$$\langle \pi | e^{-\beta H} | 0 \rangle \propto e^{-\beta E_0} \sum_{m,l \geq 0}^{m+l \text{ odd}} \frac{(D\beta)^{l+m}}{m!\, l!} e^{i\sigma\pi(m-l)} e^{-S_0^{\text{cl}}(m+l)} = e^{-\beta E_0} \sinh[2D\beta \cos(\pi\sigma) e^{-S_0^{\text{cl}}}]. \quad (5)$$

Here D is the fluctuation determinant (without zero mode) [8], l is the number of instantons, and m the number of anti-instantons in the path, E_0 is the zero-point energy in one well, $\exp(-S_0^{\text{cl}})$ is the instanton contribution to the path integral, and the constraint on the sum reflects the requirement that $\phi(\beta) = \pi[\text{mod}(2\pi)]$. Note that the action S_0^{cl} is exactly the same for the instanton and anti-instanton, because this action is unchanged if ϕ is replaced by $-\phi$ [15]. From Eq. (5) we can now read off the tunneling rate P (energy level splitting):

$$P = 4D |\cos(\pi\sigma)| e^{-S_0^{\text{cl}}}, \quad e^{-S_0^{\text{cl}}} = \left(\frac{1-\sqrt{\lambda}}{1+\sqrt{\lambda}} \right)^{\sigma}. \quad (6)$$

Here $\lambda \equiv K_y/K_z$. Evidently, the tunneling rate vanishes for half-integral spins because of the $\cos(\pi\sigma)$ factor which arises directly from the topological boundary term of Eq. (4). This factor represents an interference between the instanton and anti-instanton contributions to tunneling. If the spin σ is an integer, then the interference is constructive, and the total tunneling rate is of order of the single-instanton rate. But if the spin is half integral, then $\cos(\pi\sigma) = 0$; there is *destructive interference* between the instanton and anti-instanton, and the tunneling rate is *zero*. Note that this spin-parity effect is of topological origin and thus independent of the magnitude of the spin.

A graphic illustration of the difference between integer and half-integer spin systems is afforded by the numerical energy spectra of Hamiltonian (1) presented in Fig. 2. Results for $\sigma = 10\frac{1}{2}$, 10 are shown as a function of K_y, for $K_z = 1$. As K_y is increased, a barrier between the two easy-axis directions is developed, and tunneling occurs for the integer-spin case. The tunneling rate, which is proportional to the energy splitting between the two lowest energy levels, decreases with increasing K_y [see Eq. (6)], eventually vanishing when $K_y = K_z$, since at this point the x component of the magnetization commutes with the Hamiltonian and so is conserved. In the half-integer-spin case, by contrast, it is easy to show from direct consideration of Hamiltonian (1) that the space of states decomposes into two independent subspaces with identical energy spectra. Thus all states are strictly doubly degenerate, and there is no tunneling, consistent with the arguments above.

The result that tunneling is suppressed for half-integer spin has much greater generality than is suggested by the above analysis. The suppression can be derived within the coherent-state path-integral formalism independent of any approximation. Any arbitrary path $\theta(\tau)$, $\phi(\tau)$ in Eq. (2) (not just the saddle-point path) can always be paired with another path, $\pi - \theta(\tau)$, $-\phi(\tau)$, which has the same L_0 in Eq. (3), while the winding-number term of Eq. (3) is reversed; thus, the destructive interference for half-integral spin occurs term by term in Eq. (2). Furthermore, this pairing is possible for much more general Hamiltonians than Eq. (1). One generalization involves adding any additional terms to H which preserve the symmetry of rotation around the x axis (e.g., $\sigma_z\sigma_y$), in-

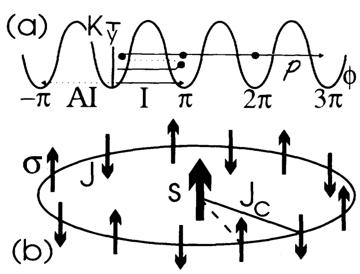

FIG. 1. (a) Anisotropy energy vs ϕ at $\theta = \pi/2$, showing the path for the instanton (I), the anti-instanton (AI), and a more general path $\mathcal{P}$ containing one anti-instanton and four instantons. (b) Antiferromagnetic ring coupled to an excess spin [Eq. (7)].

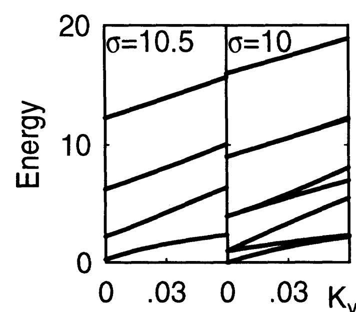

FIG. 2. Low-lying eigenenergies as a function of K_y for $K_z = 1$ for the model of Eq. (1) for spin-$10\frac{1}{2}$ and spin-10. All levels are doubly degenerate for $\sigma = 10\frac{1}{2}$.

cluding odd-order terms like $B_{\mathrm{ext}}\sigma_x$. Of course, if such an external-field term is present, then the degeneracy between the two wells is broken; nevertheless, the vanishing of the propagator of Eq. (2) for half-integer spin still implies vanishing hybridization between the two (inequivalent) wells, and an absence of tunneling. The other interesting generalization of H involves adding *any* even-order term (e.g., σ_z^6, $\sigma_x\sigma_y\sigma_z^2$), but excluding odd-order terms. Then H has time-reversal (T) invariance; absence of tunneling [i.e., vanishing of the transition amplitude (5)] can again be proved by a pairing of paths [this time, the paths $\hat{\mathbf{n}}(\tau)$, $-\hat{\mathbf{n}}(\beta-\tau)$]. In this case, the vanishing of tunnel splitting is directly related to the Kramers degeneracy [16]. We can say more generally that for anisotropy Hamiltonians which have two equivalent energy minima, the absence of tunneling for half-integral spin follows directly from the Kramers theorem [17].

We now analyze a problem raised by a recent experiment on tunneling in small *antiferromagnetic* particles [3]. The particles were not perfect antiferromagnets, but carried an excess magnetic moment, presumed to arise from surface effects, of a few percent of the total number of spins. In applying to the experiment the theory developed in Ref. [18] for the tunneling of a Néel-ordered antiferromagnetic particle through an anisotropy barrier, it was assumed [3] (as suggested in [18]) that the excess spins simply follow adiabatically the direction of the Néel vector without affecting the tunneling dynamics. We show now that, within a simple extension of the existing models, this expectation is false: As above, a *half-integer* excess spin of any size quenches tunneling; an integer excess spin also influences the tunneling rate, though more modestly.

Our model is a 1D antiferromagnetic ring with N spins $\boldsymbol{\sigma}^{(j)}$ (spin magnitude σ) and periodic boundary conditions; all the spins $\boldsymbol{\sigma}$ are coupled to a central excess spin $\mathbf{s}$ with coupling constant $J_c^{(j)} \equiv (-1)^j J_c$. Thus the central spin will prefer to remain aligned with the Néel vector of the spins on the ring [see Fig. 1(b)]. The Hamiltonian of the system may be written

$$\mathcal{H} = \sum_{j=1}^{N}[J\boldsymbol{\sigma}^{(j)}\cdot\boldsymbol{\sigma}^{(j+1)} + H(\boldsymbol{\sigma}^{(j)}) + J_c^{(j)}\boldsymbol{\sigma}^{(j)}\cdot\mathbf{s}]. \quad (7)$$

Here N is even; odd N would produce a different problem involving frustration and nonzero total spin on the ring. H is the anisotropy Hamiltonian (1), and leads to two degenerate states, the one shown in Fig. 1(b), and the one where all the spin directions are reversed. The problem is to compute the tunneling rate between these two states.

This model may be considerably simplified if we restrict our consideration to the continuum and semiclassical (large σ) limit. For this we use again the coherent-state path-integral representation, and, applying standard manipulations [10, 12], we then arrive at a generalized nonlinear σ model in terms of a Néel unit vector $\hat{\boldsymbol{l}}$, and a central spin unit vector $\hat{\mathbf{n}}_s$ [19]. We take $\hat{\boldsymbol{l}}$ to be uniform around the ring, obtaining the following effective Euclidean action:

$$S_E = \int_0^\beta d\tau\left(\frac{\chi_\perp}{8\mu_B^2}(\dot{\theta}_l^2 + \dot{\phi}_l^2\sin^2\theta_l) + NE(\theta_l,\phi_l) + \sigma N J_c\hat{\boldsymbol{l}}\cdot\hat{\mathbf{n}}_s + is\dot{\phi}_s(1-\cos\theta_s)\right), \quad (8)$$

where $\hat{\boldsymbol{l}}$ and $\hat{\mathbf{n}}$ are expressed in polar coordinates, and the transverse susceptibility $\chi_\perp$ is related to the parameters of Eq. (7) by $\chi_\perp = N\mu_B^2/J$.

As before, we find that if the excess spin s of the antiferromagnet is half integer, then the tunneling rate is exactly zero. The proof follows in the same way: we consider an arbitrary path $\{\theta_l(\tau),\phi_l(\tau),\theta_s(\tau),\phi_s(\tau)\}$ and its partner with $\theta_{l,s}\to\pi-\theta_{l,s}$, $\phi_{l,s}\to-\phi_{l,s}$. Then in the path-integral expression analogous to Eq. (2), and with $\hat{\boldsymbol{l}}$ and $\hat{\mathbf{n}}_s$ having the same boundary conditions, these two paths have opposite winding-number contributions $\pm is\pi(1+2n)$ [cf. Eq. (3)], and the same values of L_0. Thus, a factor $\cos(\pi s)$ appears exactly as in Eq. (5), implying complete destructive interference and a vanishing of the tunneling rate if the central spin s is half integral.

Also as in the ferromagnetic case, this vanishing can be seen to be related to the Kramers degeneracy. Again, the model Eq. (7), and therefore Eq. (8), has time-reversal invariance (since all terms contain an even number of spin operators). Thus, the ground state is a Kramers doublet so long as the total spin of the model is half integral, which, since N is even, requires that s is half integral. Again, suppression of tunneling is related to the absence of a tunnel splitting in the ground state. However, we caution as we did above that there is not a one-to-one correspondence between the Kramers theorem and absence of tunneling for antiferromagnetic models.

Finally, we consider the question of how strongly a nonzero but *integer s* modifies the tunneling dynamics of Eq. (8). For $s=0$ this model can be solved in the instanton approximation; the saddle-point solution happens to be identical to a different (uniaxial-anisotropy) model considered in [18]. The saddle-point path has $\theta_l=\pi/2$ everywhere, while ϕ_l passes from 0 to π. As in [18], the tunneling rate is given by $P\sim\omega e^{-\sqrt{2\chi_\perp k_y}/\mu_B}$, with $\omega\equiv\mu_B(8k_y/\chi_\perp)^{\frac{1}{2}}$, and with the notation $k_{y,z}\equiv\sigma^2NK_{y,z}$ for the anisotropy constants of the whole ring.

A full solution of Eq. (8) for $s\neq0$ seems difficult. We can make progress in the adiabatic approximation [13, 14], in which the spin $\hat{\mathbf{n}}_s$ simply follows the instantaneous direction of the Néel vector $\hat{\boldsymbol{l}}$ at every point along the path. In Eq. (8), this simply involves removing the $\hat{\boldsymbol{l}}\cdot\hat{\mathbf{n}}_s$ term (since it is just a constant) and setting $\theta_s=\theta_l$, $\phi_s=\phi_l$ [20].

We can now find an approximate solution for the tunneling rate if we assume that $k_y\ll k_z$ and that therefore θ_l does not fluctuate very far away from $\pi/2$. If we write $\theta_l=\pi/2+\vartheta$ and expand the Lagrangian to second order in ϑ, we obtain

$$L \approx \frac{\chi_\perp}{8\mu_B^2}(\dot{\vartheta}^2 + \dot{\phi}^2) + \vartheta^2\left(k_z - k_y \sin^2\phi - \frac{\chi_\perp}{8\mu_B^2}\dot{\phi}^2\right) + k_y \sin^2\phi + is\dot{\phi}(1+\vartheta). \quad (9)$$

We find [21] that for $k_y \ll k_z$ we have $\dot{\vartheta}^2 \ll \dot{\phi}^2 \ll 8\mu_B^2 k_z/\chi_\perp$. Keeping then only the leading terms in L we find for the extremal ϑ path, $\vartheta \approx -is\dot{\phi}/2k_z$, obtaining an effective Lagrangian for the ϕ variable, $L \approx (\chi_\perp/8\mu_B^2 + s^2/4k_z)\dot{\phi}^2 + k_y \sin^2\phi + is\dot{\phi}$. This Lagrangian is now identical to the one for a pure antiferromagnet (see [18]) with $s = 0$, but with a topological boundary term and a modified value of the transverse susceptibility (the "moment of inertia of the rigid rotor" [11]): $\chi_\perp^{\text{eff}} = \chi_\perp + 2s^2\mu_B^2/k_z$. Thus the tunneling rate is finally given by

$$P \sim |\cos(\pi s)|\omega e^{-\sqrt{2\chi_\perp^{\text{eff}} k_y}/\mu_B}. \quad (10)$$

Again, for half-integral excess spin s tunneling is suppressed. For integral s we can define a crossover excess spin s_c for which the contribution of the excess spin to the moment of inertia becomes comparable to that of the antiferromagnetic ring: $s_c = \sqrt{\chi_\perp k_z/2}/\mu_B$. If, e.g., $k_z \approx 10k_y$, then s_c is approximately 1.6 times the WKB exponent of Eq. (10) for $s = 0$. As Ref. [18] has pointed out, for practical reasons this WKB exponent cannot be much larger than about 25 if tunneling is to be observed in a small particle. Thus, the magnitude of the excess spin is quite restricted ($s < 1.6 \times 25$) if pure antiferromagnetic dynamics is to be observed.

In summary, we have demonstrated that tunneling in a wide class of magnetic particles is strongly parity dependent, being completely suppressed for half-integral spins. Preliminary results indicate that similar effects are to be expected in the tunneling of domain walls [2]. We expect that these phenomena can still exist in the presence of moderate dissipation [11].

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