Phase interference of spin tunneling in an arbitrarily directed magnetic field

Rong Lü*, Jia-Lin Zhu, Yi Zhou, and Bing-Lin Gu

Center for Advanced Study, Tsinghua University, Beijing 100084, People’s Republic of China

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Abstract

We present an exact analytic study on the topological phase interference effect in resonant quantum tunneling of the magnetization between degenerate excited levels for biaxial ferromagnets in an arbitrarily directed magnetic field. We show that the topological phase interference effect depends on the orientation of the field distinctly. The transition from classical to quantum behavior is also discussed.

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*Electronic address: rlu@castu.tsinghua.edu.cn
During the last decade, there has been great interest in the problem of quantum tunneling in nanometer-scale magnets.\textsuperscript{1} One notable subject is that the topological Wess-Zumino (or Berry) phase\textsuperscript{2} can lead to remarkable spin-parity effects.\textsuperscript{3}–\textsuperscript{5} It was theoretically shown that the tunnel splitting is suppressed to zero for half-integer total spins in single-domain biaxial ferromagnetic (FM) particles due to the destructive interference of the Berry phase between two tunneling paths of opposite windings.\textsuperscript{3} However, the interference is constructive for integer spins, and hence the splitting is nonzero. While spin-parity effects are sometimes be related to the Kramers degeneracy, they typically go beyond the Kramers theorem in a rather unexpected way.\textsuperscript{4}–\textsuperscript{6} The auxiliary particle method was proposed to study the spin-parity effects in one model of a single large spin subject to the external and anisotropy fields.\textsuperscript{6} Recently, the spin-phase interference and quantum oscillation effects were studied extensively in FM particles in the presence of a magnetic field, with the field along either the hard,\textsuperscript{4} easy,\textsuperscript{8} or medium axis.\textsuperscript{8} Similar spin-parity effects were also found in antiferromagnetic (AFM) particles\textsuperscript{9,10} and in the quantum propagation of Bloch walls in quasi-one-dimensional ferromagnets\textsuperscript{11} and antiferromagnets.\textsuperscript{12} By applying the effective Hamiltonian approach, the effect of magnetic field and quantum interference of the magnetization vector (or the Néel vector) were studied in FM (or AFM) particles with different symmetry.\textsuperscript{13}

Experiment on the molecules Fe\textsubscript{8} showed a direct evidence of the role of the topological phase in spin dynamics.\textsuperscript{11} Recent theoretical and experimental studies include the spin tunneling in a swept magnetic field,\textsuperscript{14} the thermally activated resonant tunneling based on the perturbation theory\textsuperscript{15} and the exact diagonalization\textsuperscript{16} the discrete WKB method and a nonperturbation calculation\textsuperscript{17} the non-adiabatic Landau-Zener model\textsuperscript{18} the calculation based on exact spin-coordinate correspondence\textsuperscript{19} and the effects caused by the higher order term and the nuclear spins on the tunnel splitting of Fe\textsubscript{8}.\textsuperscript{20,21}

It is noted that the previous studies on FM spin-parity effects\textsuperscript{4,7,9,14} were mostly focused on the phase interference between two opposite winding ground-state tunneling paths. Moreover, the previous works\textsuperscript{4,9} have been confined to the condition that the magnetic field be applied along the easy, medium, or hard axis, separately. In this paper we study the
topological phase interference effects for single-domain biaxial FM particles in an arbitrarily directed magnetic field. Our study provides a nontrivial generalization of the Kramers degeneracy for equivalent double-well system to coherently spin tunneling at ground states as well as low-lying excited states for FM system with asymmetric twin barriers caused by the arbitrarily directed magnetic field. Integrating out the momentum in the path integral, the spin tunneling problem is mapped onto a particle moving problem in one-dimensional periodic potential $U(\phi)$. By applying the periodic instanton method, we obtain exactly the splittings between degenerate excited levels of neighboring wells. $U(\phi)$ is regarded as a one-dimensional superlattice with a periodically recurring asymmetric twin barriers. The general translation symmetry results in the energy band structure, and the low-lying energy level spectrum is obtained by using the Bloch theorem. Our result shows that the excited-level splittings depend significantly on the parity of total spins of FM particles, and this spin-parity effect depends on the orientation of the field distinctly. The transition from quantum to classical behavior is also studied, and the second-order phase transition is shown.

For a spin tunneling problem, the tunneling rate is determined by the Euclidean transition amplitude

$$K_E = \langle f | e^{-HT} | i \rangle = \int D\Omega \exp \left( -S_E \right),$$

where $D\Omega = \sin \theta d\theta d\phi$, and the Euclidean action is

$$S_E(\theta, \phi) = \frac{V}{\hbar} \int d\tau \left[ \frac{M_0}{\gamma} \left( \frac{d\phi}{d\tau} \right)^2 - i \frac{M_0}{\gamma} \left( \frac{d\phi}{d\tau} \right) \cos \theta + E(\theta, \phi) \right].$$

$M_0V = |\vec{M}|V = \hbar \gamma S$, where $V$ is the volume of the particle, $\gamma$ is the gyromagnetic ratio, and $S$ is the total spins. The first two terms in Eq. (2) define the topological Wess-Zumino term which arises from the nonorthogonality of spin coherent states. The total derivative has no effect on the classical equations of motion, but is crucial for the spin-parity effects.

The system of interest has the biaxial symmetry, with $\hat{x}$ being the easy axis, $\hat{y}$ being the medium axis, and $\hat{z}$ being the hard axis. The magnetic field is applied in the ZY plane, at an angle in the range of $\frac{\pi}{2} \leq \theta_H \leq \pi$. Now $E(\theta, \phi)$ in Eq. (2) is
\[ E(\theta, \phi) = K_\perp \cos^2 \theta + K_\parallel \sin^2 \theta \sin^2 \phi - M_0 H_y \sin \theta \sin \phi - M_0 H_z \cos \theta, \quad (3) \]

where \( H_y = H \sin \theta_H, H_z = H \cos \theta_H, K_\parallel \) and \( K_\perp \) are the longitudinal and the transverse anisotropy coefficients satisfying \( K_\perp \gg K_\parallel > 0 \). As \( K_\perp \gg K_\parallel > 0 \), the deviations of \( \theta \) about \( \theta_0 \) are small. Introducing \( \theta = \theta_0 + \alpha, |\alpha| \ll 1 \), Eq. (3) reduces to

\[ E(\alpha, \phi) = K_\perp \sin^2 \theta_0 \alpha^2 + K_\parallel \sin^2 \theta_0 (\sin \phi - \sin \phi_0)^2, \quad (4) \]

where \( \cos \theta_0 = \frac{M_0 H_y}{2K_\perp}, \sin \phi_0 = \frac{M_0 H_y}{2K_\parallel \sin \theta_0} = \frac{\hbar \sin \theta_H}{\sqrt{1 - (\lambda h \cos \theta_H)^2}}, \lambda = \frac{K_\parallel}{K_\perp}, h = \frac{H}{H_0}, \) and \( H_0 = \frac{2K_\parallel}{M_0} \).

Performing the Gaussian integration over \( \alpha \), we obtain the transition amplitude as

\[ K_E = \exp \left\{ -i S(1 - \cos \theta_0)(\phi_f - \phi_i) \right\} \int d\phi \exp \left\{ - \int d\tau \left[ \frac{1}{2} m \left( \frac{d\phi}{d\tau} \right)^2 + U(\phi) \right]\right\}, \quad (5) \]

with \( m = \frac{\hbar^2}{2K_\perp}, \) and \( hU(\phi) = K_\parallel V \sin^2 \theta_0 (\sin \phi - \sin \phi_0)^2. \) Since the configuration space of this problem is a circle, calculations are restricted to the first twin barrier at \( \phi = \frac{\pi}{2} \) and \( \frac{3\pi}{2}. \)

We use \( A \) to denote the instanton passing through the small barrier at \( \phi = \frac{\pi}{2} \) with the height \( \hbar U_S = K_\parallel V \sin^2 \theta_0 (1 - \sin \phi_0)^2 \), and \( B \) through the large barrier at \( \phi = \frac{3\pi}{2} \) with the height \( \hbar U_L = K_\parallel V \sin^2 \theta_0 (1 + \sin \phi_0)^2 \). Correspondingly, there are two kinds of anti-instantons: \( A^- \) and \( B^- \).

The periodic instanton configuration \( \phi_p \) which minimizes the Euclidean action in Eq. (5) at an energy \( E > 0 \) satisfies the equation of motion

\[ \frac{1}{2} m \left( \frac{d\phi_p}{d\tau} \right)^2 - U(\phi_p) = -E. \quad (6) \]

Then we obtain the periodic instanton \( A \) solution as

\[ \sin \phi_A = \frac{1 - \xi_1 \sin^2 (\omega_1 \tau, k_1)}{1 + \xi_1 \sin^2 (\omega_1 \tau, k_1)}, \quad (7) \]

corresponding to the transition of \( \phi \) from \( \arcsin \alpha \) to \( \pi - \arcsin \alpha \), where \( \alpha = \sin \phi_0 + \sqrt{\frac{\hbar E}{K_\parallel V \sin^2 \theta_0}} \), \( \sin(\omega_1 \tau, k_1) \) is the Jacobian elliptic sine function of modulus \( k_1 \), where \( k_1^2 = \frac{(1 - \alpha)(1 + \beta)}{(1 + \alpha)(1 - \beta)} \), \( \beta = \sin \phi_0 - \sqrt{\frac{\hbar E}{K_\parallel V \sin^2 \theta_0}}, \xi_1 = \frac{1 - \alpha}{1 + \alpha}, \omega_1 = \frac{\omega_0}{g_1}, \omega_0 = 2 \sqrt{\frac{K_\perp K_\parallel}{\hbar S}} \sin \theta_0, \) and \( g_1 = \frac{2}{\sqrt{(1 + \alpha)(1 - \beta)}}. \) The classical action or the WKB exponent in the tunnel splitting is obtained
by integrating the Euclidean action in Eq. (5) with the above periodic instanton solution. The result for instanton $A$ is $S_A = W_A + 2E\beta$, where

$$W_A = 4m\omega_1 \left[ E(k_1) + \frac{(k_1^2 - \xi_1)}{\xi_1} K(k_1) + \frac{(\xi_1^2 - k_1^2)}{\xi_1} \Pi(k_1, \xi_1) \right].$$

(8)

$K(k_1)$, $E(k_1)$, and $\Pi(k_1, \xi_1)$ are the complete elliptic integral of the first, second, and third kind.

Now we discuss briefly the calculation of the tunnel splitting. For a particle with mass $m$ moving in a smooth double-well-like one-dimensional potential $U(x)$, the instanton approach gives the ground-state tunnel splitting as:

$$\Delta E_0 = 2C \left( \frac{W_0}{2\pi} \right)^{1/2} \exp(-W_0),$$

(9)

where $W_0$ is the classical action for the ground-state tunneling,

$$C = \left\{ \frac{\det(-\partial^2 + \omega^2)}{\det'[-\partial^2 + U''(x_{cl}(\tau))/m]} \right\}^{1/2}$$

(10)

is the ratio of fluctuation determinants, and $x_{cl}(\tau)$ is the instanton solution. The prime on the det indicates that the zero eigenvalue is to omitted, and $\omega$ is the frequency of harmonic amplitude oscillations in the wells about the minima. The general formulas were presented to evaluate the preexponential factor in the tunnel splitting or the tunneling rate.

It was found that what is need for this evaluation is the asymptotic ($\tau \to \pm\infty$) behavior of the instanton velocity,

$$\frac{dx_{cl}}{d\tau} \approx a \exp(\mp \omega \tau), \text{ as } \tau \to \pm\infty.$$ 

(11)

Then the ground-state tunnel splitting is:

$$\Delta E_0 = 2|a| \left( \frac{m\omega}{\pi} \right)^{1/2} \exp(-W_0).$$

(12)

In most physical applications, however, the ground-state tunnel splitting can be best estimated as

$$\Delta E_0 = p_0\omega \left( \frac{W_0}{2\pi} \right)^{1/2} \exp(-W_0),$$

(13)
where the dimensionless prefactor $p_0$ is often relevant to experiments. It is noted that Eq. (12) is based on quantum tunneling at the level of ground state, and the temperature dependence of the tunneling frequency (i.e., tunneling at excited levels) is not taken into account. The instanton technique is suitable only for the evaluation of the tunneling rate or the tunnel splitting at the vacuum level, since the usual (vacuum) instantons satisfy the vacuum boundary conditions. Recently, different types of pseudoparticle configurations (periodic or nonvacuum instantons) are found which satisfy periodic boundary conditions.

For the same tunneling problem, the WKB answer for the splittings of the $n$th degenerate excited levels or the imaginary parts of the metastable levels is:

$$\Delta E_n \text{ (or } \text{Im} E_n) = \frac{\omega(E_n)}{\pi} \exp(-W), \quad (14)$$

where $\omega(E_n) = \frac{2\pi}{t(E_n)}$ is the energy-dependent frequency. $t(E_n)$ is the period of the real-time oscillation in the potential well,

$$t(E_n) = \sqrt{\frac{2m}{E_n - U(x)}} \int_{x_1(E_n)}^{x_2(E_n)} \frac{dx}{\sqrt{E_n - U(x)}}, \quad (15)$$

where $x_{1,2}(E_n)$ are the turning points for the particle oscillating inside the potential $U(x)$.

The functional-integral and the WKB method showed that for the potentials parabolic near the bottom the result Eq. (14) should be multiplied by $\sqrt{\frac{\pi}{2}} \frac{(2n+1)^{n+1/2}}{2^n n!}$ This factor is very close to 1 for all $n$: 1.075 for $n = 0$, 1.028 for $n = 1$, 1.017 for $n = 2$, etc. Stirling’s formula for $n!$ shows that this factor trends to 1 as $n \to \infty$. Therefore, this correction factor, however, does not change much in front of the exponentially small action term in Eq. (14).

It is noted that Eq. (14) can be obtained by using the connection formulas near a linear turning point, or by matching the WKB wavefunction in the classically forbidden region to the exact harmonic oscillator wavefunction near the classical turning point. It was shown that the WKB method is equivalent to the instanton method for the ground-state tunnel splitting. Liang et al. showed that the WKB method and the periodic instanton method give the same result for the excited-state tunnel splitting.

For the present problem, we find that $\Delta \varepsilon_A = \frac{2}{t_A(E)} \exp(-W_A)$, where $t_A(E) =$
The same method can be applied to the instanton $B$ passing through the large barrier. And the result is $\Delta \varepsilon_B = \frac{2}{\omega_1(E)} \exp (-W_B)$ for $0 < E < U_S$, where $t_B(E) = t_A(E)$, and $W_B$ has the same expression as Eq. (8) but taking $\xi_2$ as $\xi_2 = \frac{1+\beta}{1-\beta}$.

For $U_S \leq E \leq U_L$, the imaginary part of the metastable level is $\text{Im} E = \frac{2}{t_B(E)} \exp (-2\tilde{W}_B)$, where $\tilde{t}_B(E) = \frac{2}{\omega_2(E)} K(k_2')$,

$$
\tilde{W}_B = 2m \xi_3 (1 + \alpha) \omega_2 \left[ \frac{1}{k_2^2 - \xi_3} E(k_2) - \frac{1}{\xi_3} K(k_2) + \frac{\xi_3}{k_2^2} + \frac{2k_2^2}{\xi_3} \Pi(k_2, \xi_3) \right],
$$

(16)

with $k_2' = \sqrt{1 - k_2^2}$, $\omega_2 = \frac{\omega_0}{g_2}$, $g_2 = \sqrt{\frac{2}{\alpha - \beta}}$, $\xi_3 = \frac{1+\beta}{\alpha - \beta}$, and $k_2' = \frac{(\alpha-1)(1+\beta)}{2(\alpha - \beta)}$. Now we discuss the low energy limit of the level splitting. With the help of harmonic oscillator approximation for energy near the bottom of the potential well, $\varepsilon_n = \left( n + \frac{1}{2} \right) \Omega$, and

$$
\Omega = \sqrt{\frac{1}{m} \left( \frac{d^2 U}{d\phi^2} \right)}_{\phi = \phi_0} = \sqrt{\frac{2K \| V}{\hbar m}} \sin \theta_0 \cos \phi_0,
$$

$W_{A(B)}$ can be expanded as

$$
W_{A(B),n} = W_{A(B),0} - \left( n + \frac{1}{2} \right) + \left( n + \frac{1}{2} \right) \ln \left( \frac{n + 1/2}{8\sqrt{\lambda} S \sin \theta_0 \cos^3 \phi_0} \right), \quad (17a)
$$

$$
W_{A(B),0} = 2\sqrt{\lambda} S \sin \theta_0 \left( \cos \phi_0 \pm 2 \sin \phi_0 \arcsin \sqrt{\frac{1 \mp \sin \phi_0}{2}} \right), \quad (17b)
$$

where “−” for the instanton $A$, and “+” for the instanton $B$. Therefore, the tunnel splittings for instantons $A$ and $B$ are

$$
\hbar \Delta \varepsilon_{A(B),n} = \frac{2^{3/2}}{\sqrt{\pi n!}} (K \| V) \sqrt{\lambda S^{-1}} \sin \theta_0 \cos \phi_0 \left( 8\sqrt{\lambda} S \sin \theta_0 \cos^3 \phi_0 \right)^{n+1/2} \exp \left(-W_{A(B),0}\right).
$$

(18)

Equation (17b) shows that the WKB exponent for instanton $A$ is smaller than that for instanton $B$ at finite magnetic field because the barrier through which instanton $B$ must tunnel is higher than that for instanton $A$. For the case of ground-state resonance, i.e., $n = 0$, Eq. (18) reduces to

$$
\Delta \varepsilon_{A(B),0} = \frac{4}{\sqrt{\pi}} \lambda^{1/4} S^{1/2} \left( \sin \theta_0 \cos^3 \phi_0 \right)^{1/2} \Omega \exp \left(-W_{A(B),0}\right). \quad (19a)
$$
It is not particularly illuminating to write out the prefactor $p_0$ (see Eq. (13)), although it can be seen that it is dimensionless and independent of the volume $V$ of the particle. Note that Eq. (13) is the approximate formula for ground-state tunnel splitting with exponential accuracy. In most physical applications, the preexponential factor can be best estimated as an attempt frequency. The apparent disagreement with Eq. (13) was also found in other spin-tunneling problems. However, in the case of zero magnetic field Eq. (19a) reduces to

$$
\Delta \varepsilon_{A,0} (H = 0) = \Delta \varepsilon_{B,0} (H = 0) \\
= 4 \Omega_0 \left( \frac{W_0}{2\pi} \right)^{1/2} \exp (-W_0),
$$

(19b)

where $\Omega_0 = \Omega (H = 0)$, and $W_0 = W_{A,0} (H = 0) = W_{B,0} (H = 0) = 2\sqrt{\lambda S}$. Compared with Eq. (13), $p_0 = 4$ for this case.

It is noted that $\hbar \Delta \varepsilon_{A(B),n}$ is only the level shift induced by tunneling between degenerate excited states through a single barrier. $U (\phi)$ can be regarded as a one-dimensional superlattice with the sublattices $A$ and $B$. The Bloch states for sublattices $A$ and $B$ are

$$
\Phi_A (\xi, \phi) = \frac{1}{\sqrt{L}} \sum_n e^{i\xi\phi_n} \varphi_A (\phi - \phi_n),
$$

(20a)

and

$$
\Phi_B (\xi, \phi) = \frac{1}{\sqrt{L}} \sum_n e^{i(\phi_n + a)} \varphi_B (\phi - \phi_n - a),
$$

(20b)

where $\phi_n = 2n\pi + \phi_0$, $L = N (a + b)$, $a = \pi - 2\phi_0$, and $b = \pi + 2\phi_0$. The total wavefunction is

$$
\Psi (\phi) = a_A (\xi) \Phi_A (\xi, \phi) + a_B (\xi) \Phi_B (\xi, \phi).
$$

(21)

Including the Wess-Zumino phase, we derive the secular equation in the tight-binding approximation as

$$
\begin{bmatrix}
\varepsilon_n - E (\xi) & e^{i(\xi - \mu)a} \Delta \varepsilon_{A,n} + e^{-i(\xi - \mu)b} \Delta \varepsilon_{B,n} \\
e^{-i(\xi - \mu)a} \Delta \varepsilon_{A,n} + e^{i(\xi - \mu)b} \Delta \varepsilon_{B,n} & \varepsilon_n - E (\xi)
\end{bmatrix}
\begin{bmatrix}
a_A (\xi) \\
a_B (\xi)
\end{bmatrix} = 0,
$$

(22)

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where $\mu = S (1 - \cos \theta_0)$, and the Bloch wave vector $\xi = 0$ in the first Brillouin zone. Then the tunnel splitting of the $n$th excited level is

$$\Delta \varepsilon_n = 2 \sqrt{(\Delta \varepsilon_{A,n})^2 + (\Delta \varepsilon_{B,n})^2 + 2 (\Delta \varepsilon_{A,n}) (\Delta \varepsilon_{B,n}) \cos [2\pi S (1 - \cos \theta_0)]}, \quad (23)$$

Another approach to obtain Eq. (23) is to calculate the transition amplitude directly. The subtle point in evaluating the transition amplitude is how to arrange the instantons and anti-instantons appropriately to satisfy the boundary condition. Note that one configuration starting from $|\theta_0, \phi_0\rangle$ and ending at $|\theta_0, \phi_0\rangle$ can be an arbitrary permutation of the pairs $(AB)$, $(AA^-)$, $(B^-B)$, and $(B^-A^-)$. Introducing $s$, $t$, $p$, and $q$ as the numbers of $A$, $B$, $A^-$, and $B^-$ in the instantons and anti-instantons pairs, and $i$, $j$, $k$, and $l$ as those of $(AB)$, $(AA^-)$, $(B^-B)$, and $(B^-A^-)$, we have

$$i + j = s, \quad i + k = t, \quad j + l = p, \quad k + l = q. \quad (24)$$

Therefore, $s + q = t + p$, and only three variables are independent. Now the transition amplitude is

$$K_E = \sqrt{\frac{\Omega}{\pi \hbar}} e^{-\left((n + \frac{1}{2})\Omega\right)} \sum_{s,t,p,q} N(s, t, p, q) \frac{N(s, t, p, q)}{(s + t + p + q)!} \left[ (\hbar \Delta \varepsilon_{A,n} T)^{s+p} (\hbar \Delta \varepsilon_{B,n} T)^{t+q} \times e^{-iS(1-\cos \theta_0)(\pi-2\phi_0)(s-p)} e^{-iS(1-\cos \theta_0)(\pi+2\phi_0)(t-q)} \right] \delta_{s+q,t+p}, \quad (25)$$

where $N(s, t, p, q)$ represents the number of different configurations for a given set of $(s, t, p, q)$. It can be calculated as

$$N(s, t, p, q) = \sum_{i=\max(0, t-q)}^{\min(s, t)} \frac{(i + j + k + l)!}{i!j!k!l!}, \quad (26)$$

which has a simple result

$$N(s, t, p, q) = \frac{[(s + q)!]^2}{s!t!p!q!}, \quad (27)$$

Then we have

$$K_E = \sqrt{\frac{\Omega}{\pi \hbar}} e^{-\left((n + \frac{1}{2})\Omega\right)} \cosh \left[ \sqrt{(\Delta \varepsilon_{A,n})^2 + (\Delta \varepsilon_{B,n})^2 + 2 (\Delta \varepsilon_{A,n}) (\Delta \varepsilon_{B,n}) \cos [2\pi S (1 - \cos \theta_0)] T} \right], \quad (28)$$
and we can read off the splitting of the \( n \)th excited level shown in Eq. (23) directly.

Finally we discuss the phase transition from classical to quantum behavior. It was found that for a particle moving in a double-well potential \( U(x) \), the behavior of the energy-dependent period of oscillations \( P(E) \) in the Euclidean potential \( -U(x) \) determines the order of the quantum-classical transition. If \( P(E) \) monotonically increases with the amplitude of oscillations, i.e., with decreasing energy \( E \), the transition is of second order. The corresponding crossover temperature is
\[
T_0^{(2)} = \frac{\tilde{\omega}_0}{2\pi}, \quad \tilde{\omega}_0 = \sqrt{\frac{1}{m} |U''(x_{sad})|}
\]
is the frequency of small oscillations near the bottom of \( -U(x) \), where \( x_{sad} \) corresponds to the top (the saddle point) of the barrier. If, however, the dependence of \( P(E) \) is non-monotonic, the first order crossover takes place. For this case, the period of the instanton \( A \) is
\[
P_A(E) = \frac{4}{\omega_1} K(k_1).
\]
The monotonically decreasing behavior of \( P_A(E) \) in the domain \( 0 \leq E \leq U_S \) is shown in Fig. 1 for several values of \( \sin \phi_0 \), which shows that the second-order phase transition takes place, and the crossover temperature is
\[
k_B T_0^{(2),A} = \frac{\sqrt{K_\parallel K_\perp V}}{\pi \hbar S} \sin \theta_0 \sqrt{1 - \sin \phi_0}. \tag{29a}
\]
The period of the instanton \( B \) is \( P_B(E) = P_A(E) \) for \( 0 \leq E \leq U_S \), while \( P_B(E) = \frac{4}{\omega_2} K(k_2) \) for \( U_S \leq E \leq U_L \). Fig. 2 shows the monotonically decreasing behavior of \( P_B(E) \) for the whole domain \( 0 \leq E \leq U_L \). Again one finds a second-order transition from the thermal to quantum regime, and the crossover temperature is
\[
k_B T_0^{(2),B} = \frac{\sqrt{K_\parallel K_\perp V}}{\pi \hbar S} \sin \theta_0 \sqrt{1 + \sin \phi_0}. \tag{29b}
\]

At zero magnetic field, \( \Delta \varepsilon_{A,n} = \Delta \varepsilon_{B,n} \), the tunnel splitting is suppressed to zero for the half-integer total spins by the destructive interfering Wess-Zumino-Berry phases, which is in good agreement with the Kramers theorem. The presence of a magnetic field perpendicular to the plane of rotation of magnetization yields an additional contribution to the Berry phase, resulting constructive and destructive interferences alternatively for both integer and half-integer spins. Tunneling is thus periodically suppressed. At finite magnetic field, the low-lying tunneling level spectrum depends on the parity of total spins. If \( S \) is an integer,
the tunnel splitting is
\[ \Delta \varepsilon_n = 2 \sqrt{\left( \Delta \varepsilon_{A,n} \right)^2 + \left( \Delta \varepsilon_{B,n} \right)^2 + 2 \left( \Delta \varepsilon_{A,n} \right) \left( \Delta \varepsilon_{B,n} \right) \cos (2\pi S \cos \theta_0)} . \] (30a)

While if $S$ is a half-integer, the splitting is
\[ \Delta \varepsilon_n = 2 \sqrt{\left( \Delta \varepsilon_{A,n} \right)^2 + \left( \Delta \varepsilon_{B,n} \right)^2 - 2 \left( \Delta \varepsilon_{A,n} \right) \left( \Delta \varepsilon_{B,n} \right) \cos (2\pi S \cos \theta_0)} . \] (30b)

Equations (30a) and (30b) clearly show oscillations of the tunnel splitting as a function of the transverse magnetic field, together with the parity effect. Note that the magnetic field along the medium axis does not produce any oscillations. As $\Delta \varepsilon_{A,n} > \Delta \varepsilon_{B,n}$ at finite magnetic field, the tunnel splitting will not be suppressed to zero even if the total spin is a half-integer.

The similar oscillation of tunnel splitting with the magnetic field was also found in Ref. 29, while the system considered in Ref. 29 is a single-domain antiferromagnetic particle with a field along the hard axis. The most interesting observation is that only the $\hat{z}$ component of the magnetic field (i.e., along the hard axis) can lead to the oscillation of the tunnel splitting. As shown in Fig. 3, the splitting depends on the magnitude of the field $H$ and its angle $\theta_H$ with the hard axis, and even a small misalignment of the field with the $\hat{z}$ axis can completely destroy the oscillation effect. For small $\theta_H$ the tunnel splitting oscillates with the field, whereas no oscillation is shown up for large $\theta_H$. In the latter case, a much stronger increase of tunnel splitting with the field is shown. This strong dependence on the orientation of the field can be observed for ground-state resonance as well as excited-state resonance.

As a result, we conclude that the topological phase interference or spin-parity effects depend on the orientation of the external magnetic field distinctly. The distinct angular dependence, together with the oscillation of the tunnel splittings with the field, may provide an independent experimental test for the spin phase interference effects in FM particles. This “Aharonov-Bohm” type of oscillation in magnetic system is analogous to the oscillations as a function of external flux in a SQUID ring. Due to the topological nature of the Berry phase, these spin-parity effects are independent of details such as the magnitude of total spins and the shape of the soliton. The transition from classical to quantum behavior is also studied for the small
and large tunneling barrier, respectively. By calculating the periods of instanton $A$ and $B$ analytically, we find the monotonically decreasing behavior of the periods with increasing energy, which yields a second-order phase transition. Our results should be useful for a quantitative understanding on the spin-parity effects and the quantum-classical crossover for FM particles in an arbitrarily directed magnetic field. It is noted that the theoretical results presented here are based on the instanton method, which is semiclassical in nature, i.e., valid for large spins and in the continuum limit. The theoretical calculations performed in this paper can be extended to the single-domain antiferromagnetic particles where the relevant quantity is the excess spin due to the small noncompensation of two sublattices. Work along this line is still in progress. We hope that the theoretical results obtained in the present work will stimulate more experiments whose aim is observing the topological phase interference effects and the quantum-classical transition in resonant quantum tunneling of magnetization in nanometer-scale single-domain ferromagnets.
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Figure Captions:

Fig. 1 The relative period $P_A(E)/P_A(E = U_S)$ of the periodic instanton $A$ as a function of energy $E/K_{\parallel}V\sin^2\theta_0$ in the domain $0 \leq E \leq U_S$.

Fig. 2 The relative period $P_B(E)/P_B(E = U_S)$ of the periodic instanton $B$ as a function of energy $E/K_{\parallel}V\sin^2\theta_0$ in the domain $0 \leq E \leq U_L$.

Fig. 3 The relative tunnel splitting of the ground level ($n = 0$) $\Delta\varepsilon_0/\Delta\varepsilon_{B,0}$ ($H = 0$) as a function of $H/H_0$ for $\theta_H = 0^\circ, 1^\circ, 3^\circ, 5^\circ$, and $90^\circ$. Here $\lambda = K_{\parallel}/K_{\perp} = 0.1$. 
$P_A(E)/P_A(\text{E}=U_S)$

$E/K_2 V \sin^2 \theta_0$

- $\sin \phi_0 = 0.1$
- $\sin \phi_0 = 0.3$
- $\sin \phi_0 = 0.5$
- $\sin \phi_0 = 0.7$
\[ P_B(E)/P_B(E=U_S) \]

\[ E/K \| V \sin^2 \theta_0 \]

\[ \sin \phi_0 = 0.1 \]
\[ \sin \phi_0 = 0.3 \]
\[ \sin \phi_0 = 0.5 \]
\[ \sin \phi_0 = 0.7 \]
