Magnetization quantum tunneling at excited levels for biaxial spin system in an arbitrarily directed magnetic field

Rong Lü*, Su-Peng Kou, Jia-Lin Zhu, Lee Chang, and Bing-Lin Gu

Center for Advanced Study, Tsinghua University, Beijing 100084, P. R. China

Abstract

The quantum tunneling of the magnetization vector between excited levels are studied theoretically in single-domain ferromagnetic nanoparticles with biaxial crystal symmetry placed in an external magnetic field at an arbitrarily directed angle in the ZX plane. By applying the periodic instanton method in the spin-coherent-state path-integral representation, we calculate the tunnel splittings and the tunneling rates between excited levels in the low barrier limit for different angle ranges of the external magnetic field ($\theta_H = \pi/2$, $\pi/2 \ll \theta_H \ll \pi$, and $\theta_H = \pi$). The temperature dependences of the tunneling frequency and the decay rate are clearly shown for each case. Our results show that the tunnel splittings and the tunneling rates depend on the orientation of the external magnetic field distinctly, which provides a possible experimental test for magnetic quantum tunneling in nanometer-scale single-domain ferromagnets.

PACS number(s): 75.45.+j, 75.50.Ee

*Author to whom the correspondence should be addressed.

Electronic address: rlu@castu.phys.tsinghua.edu.cn
I. INTRODUCTION

Macroscopic quantum tunneling (MQT) and coherence (MQC) of the magnetization were intensively investigated both theoretically and experimentally in recent years. More recently, much attention was attracted to the spin tunneling in the single-domain ferromagnetic (FM) nanoparticles in the presence of an external magnetic field applied at an arbitrary angle. The MQT problem for FM particles with uniaxial crystal symmetry was first studied by Zaslavskii with the help of mapping the spin system onto a one-dimensional particle system. For the same crystal symmetry, Miguel and Chudnovsky calculated the tunneling rate by applying the imaginary-time path integral, and demonstrated that the angular and field dependences of the tunneling exponent obtained by Zaslavskii’s method and by the path-integral method coincide precisely. Kim and Hwang performed a calculation based on the instanton technique for FM particles with biaxial and tetragonal crystal symmetry. Kim extended the tunneling rate for biaxial crystal symmetry to a finite temperature, and presented the numerical results for the WKB exponent below the crossover temperature and their approximate formulas around the crossover temperature. The quantum-classical transition of the escape rate for FM particles with uniaxial crystal symmetry in an arbitrarily directed field was studied by Garanin, Hidalgo and Chudnovsky with the help of mapping onto a particle moving in a double-well potential. The switching field measurement was carried out on single-domain FM nanoparticles of Barium ferrite (BaFeCoTiO) containing about $10^5 - 10^6$ spins. The measured angular dependence of the crossover temperature was found to be in excellent agreement with the theoretical prediction which strongly suggests the MQT of magnetization in the BaFeCoTiO nanoparticles. Lü et al. studied the MQT and MQC of the Néel vector in single-domain antiferromagnetic (AFM) nanoparticles with biaxial, tetragonal, and hexagonal crystal symmetry in an arbitrarily directed field.

It is noted that the previous results of MQT of the magnetization vector at excited levels in an arbitrarily directed field were obtained by numerically solving the equation of motion satisfied by the least trajectory. The purpose of this paper is to present an analytical
investigation of the quantum tunneling at excited levels in the biaxial FM particles in an arbitrarily directed field, based on the periodic instanton method. Both the nonvacuum (or thermal) instanton and bounce solution, the WKB exponents and the preexponential factors are evaluated exactly for different angle ranges of the magnetic field ($\theta_H = \pi/2$, $\pi/2 + O(\epsilon^{3/2}) < \theta_H < \pi - O(\epsilon^{3/2})$, and $\theta_H = \pi$). Our results show that the distinct angular dependence, together with the dependence of the WKB tunneling rate on the strength of the external magnetic field, may provide an independent experimental test for the magnetic tunneling at excited levels in single-domain FM nanoparticles.

This paper is structured in the following way. In Sec. II, we review briefly some basic ideas of MQT and MQC in FM particles. And we discuss the fundamentals concerning the computation of level splittings and tunneling rates of excited states in the double-well-like potential. In Secs. III, we study the spin tunneling at excited levels for FM particles with biaxial crystal symmetry in the presence of an external magnetic field applied in the ZX plane with a range of angles $\pi/2 \leq \theta_H \leq \pi$. The conclusions are presented in Sec. V.

II. MQT AND MQC OF THE MAGNETIZATION VECTOR IN FM PARTICLES

The system of interest is a nanometer-scale single-domain ferromagnet at a temperature well below its anisotropy gap. For such a FM particle, the tunnel splitting for MQC or the tunneling rate for MQT is determined by the imaginary-time transition amplitude from an initial state $|i\rangle$ to a final state $|f\rangle$ as

$$U_{fi} = \langle f | e^{-HT} | i \rangle = \int D\Omega \exp (-S_E), \tag{1}$$

where $S_E$ is the Euclidean action and $D\Omega$ is the measurement of the path integral. In the spin-coherent-state representation, the Euclidean action can be expressed as

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

where $V$ is the volume of the FM particle and $\gamma$ is the gyromagnetic ratio. $M_0 = |\hat{M}| = \hbar \gamma S/V$, where $S$ is the total spin of FM particles. It is noted that the first two terms
in Eq. (2) define the topological Berry or Wess-Zumino, Chern-Simons term which arises from the nonorthogonality of spin coherent states. The Wess-Zumino term has a simple topological interpretation. For a closed path, this term equals $-iS$ times the area swept out on the unit sphere between the path and the north pole. The first term in Eq. (2) is a total imaginary-time derivative, which has no effect on the classical equations of motion, but it is crucial for the spin-parity effects. However, for the closed instanton or bounce trajectory described in this paper (as shown in the following), this time derivative gives a zero contribution to the path integral, and therefore can be omitted.

In the semiclassical limit, the dominant contribution to the transition amplitude comes from finite action solution (instanton or bounce) of the classical equation of motion. The instanton’s contribution to the tunneling rate $\Gamma$ or the tunnel splitting $\Delta$ (not including the topological Wess-Zumino phase) is given by

$$\Gamma \ (\text{or} \ \Delta) = A \omega_p \left( \frac{S_{cl}}{2\pi} \right)^{1/2} e^{-S_{cl}},$$

where $\omega_p$ is the oscillation frequency in the well, $S_{cl}$ is the classical action, and the prefactor $A$ originates from the quantum fluctuations about the classical path. It is noted that Eq. (7) is based on quantum tunneling at the level of ground state, and the temperature dependence of the tunneling frequency is not taken into account. However, 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, Liang et al. developed new types of pseudoparticle configurations which satisfy periodic boundary condition (i.e., periodic instantons or nonvacuum instantons). They found that the tunneling effect indeed increases exponentially with energy in the low-energy region.

For a particle moving in a double-well-like potential $U(x)$, the level splittings of degenerate excited levels or the imaginary parts of the metastable levels at an energy $E > 0$ are given by the following formula in the WKB approximation

$$\Delta E \ (\text{or} \ \text{Im} \ E) = \frac{\omega (E)}{\pi} \exp \left[ -S(E) \right],$$

(8)
and the imaginary-time action is

\[ S(E) = 2\sqrt{2m} \int_{x_1(E)}^{x_2(E)} dx \sqrt{U(x) - E}, \]  

where \( x_{1,2}(E) \) are the turning points for the particle oscillating inside the inverted potential \(-U(x)\). \( \omega(E) = 2\pi/t(E) \) is the energy-dependent frequency, and \( t(E) \) is the period of the real-time oscillation in the potential well,

\[ t(E) = \sqrt{2m} \int_{x_3(E)}^{x_4(E)} \frac{dx}{\sqrt{E - U(x)}}, \]  

where \( x_{3,4}(E) \) are the turning points for the particle oscillating inside the potential \( U(x) \). Recently, the crossover from quantum to classical behavior and the associated phase transition were studied extensively in nanospin systems and other systems.

### III. MQC AND MQT FOR BIAXIAL CRYSTAL SYMMETRY

In this section, we study the tunneling behaviors of the magnetization vector in single-domain FM nanoparticle which has the biaxial crystal symmetry, with \( \pm \hat{z} \) being the easy axes in the absence of an external magnetic field. The magnetic field is applied in the \( ZX \) plane, at an angle in the range of \( \pi/2 \leq \theta_H \leq \pi \). Then the magnetocrystalline anisotropy energy \( E(\theta, \phi) \) can be written as

\[ E(\theta, \phi) = K_1 \sin^2 \theta + K_2 \sin^2 \theta \sin^2 \phi - M_0 H_x \sin \theta \cos \phi - M_0 H_z \cos \theta + E_0, \]  

where \( K_1 \) and \( K_2 \) are the longitudinal and the transverse anisotropy coefficients, respectively, and \( E_0 \) is a constant which makes \( E(\theta, \phi) \) zero at the initial orientation. As the external magnetic field is applied in the \( ZX \) plane, \( H_x = H \sin \theta_H \) and \( H_z = H \cos \theta_H \), where \( H \) is the magnitude of the field and \( \theta_H \) is the angle between the magnetic field and the \( \hat{z} \) axis.

By introducing the dimensionless parameters as

\[ K_2 = K_2/2K_1, H_x/H_0, H_z/H_0, \]  

the \( E(\theta, \phi) \) term of Eq. (11) can be rewritten as
\[ \mathcal{E}(\theta, \phi) = \frac{1}{2} \sin^2 \theta + K_2 \sin^2 \theta \sin^2 \phi - \mathcal{H}_2 \sin \theta \cos \phi - \mathcal{H}_2 \cos \theta + \mathcal{E}_0, \]  
\text{(13)}

where \( \mathcal{E}(\theta, \phi) = 2K_1 \mathcal{E}(\theta, \phi) \), and \( \mathcal{H}_0 = 2K_1/M_0 \). At finite magnetic field, the plane given by \( \phi = 0 \) is the easy plane, on which \( \mathcal{E}(\theta, \phi) \) reduces to

\[ \mathcal{E}(\theta, \phi = 0) = \frac{1}{2} \sin^2 \theta - \mathcal{H} \cos (\theta - \theta_H) + \mathcal{E}_0. \]  
\text{(14)}

We denote \( \theta_0 \) to be the initial angle and \( \theta_c \) the critical angle at which the energy barrier vanishes when the external magnetic field is close to the critical value \( \mathcal{H}_c(\theta_H) \) (to be calculated in the following). Then, \( \theta_0 \) satisfies \([d\mathcal{E}(\theta, \phi = 0)/d\theta]_{\theta = \theta_0} = 0\), \( \theta_c \) and \( \mathcal{H}_c \) satisfy both

\[ [d\mathcal{E}(\theta, \phi = 0)/d\theta]_{\theta = \theta_c, \mathcal{H} = \mathcal{H}_c} = 0 \]  \text{and}  \[ [d^2\mathcal{E}(\theta, \phi = 0)/d\theta^2]_{\theta = \theta_c, \mathcal{H} = \mathcal{H}_c} = 0, \]

which leads to

\[ \frac{1}{2} \sin (2\theta_0) + \mathcal{H} \sin (\theta_0 - \theta_H) = 0, \]  \text{(15a)}

\[ \frac{1}{2} \sin (2\theta_c) + \mathcal{H}_c \sin (\theta_c - \theta_H) = 0, \]  \text{(15b)}

\[ \cos (2\theta_c) + \mathcal{H}_c \cos (\theta_c - \theta_H) = 0. \]  \text{(15c)}

After some algebra, the dimensionless critical field \( \mathcal{H}_c(\theta_H) \) and the critical angle \( \theta_c \) are found to be

\[ \mathcal{H}_c = \left[ \left( \sin \theta_H \right)^{2/3} + |\cos \theta_H|^{2/3} \right]^{-3/2}, \]  \text{(16a)}

\[ \sin (2\theta_c) = \frac{2 |\cot \theta_H|^{1/3}}{1 + |\cot \theta_H|^{2/3}}. \]  \text{(16b)}

Now we consider the limiting case that the external magnetic field is slightly lower than the critical field, i.e., \( \epsilon = 1 - \mathcal{H}/\mathcal{H}_c \ll 1 \). At this practically interesting situation, the barrier height is low and the width is narrow, and therefore the tunneling rate in MQT or the tunnel splitting in MQC is large. Introducing \( \eta \equiv \theta_c - \theta_0 \) (\( |\eta| \ll 1 \) in the limit of \( \epsilon \ll 1 \), expanding

\[ [d\mathcal{E}(\theta, \phi = 0)/d\theta]_{\theta = \theta_0} = 0 \]  about \( \theta_c \), and using the relations \([d\mathcal{E}(\theta, \phi = 0)/d\theta]_{\theta = \theta_c, \mathcal{H} = \mathcal{H}_c} = 0\) and \([d^2\mathcal{E}(\theta, \phi = 0)/d\theta^2]_{\theta = \theta_c, \mathcal{H} = \mathcal{H}_c} = 0\), Eq. (15a) becomes

\[ \sin (2\theta_c) \left( \epsilon - \frac{3}{2} \eta^2 \right) - \eta \cos (2\theta_c) \left( 2\epsilon - \eta^2 \right) = 0. \]  \text{(17)}

Then the potential energy \( \mathcal{E}(\theta, \phi) \) reduces to the following equation in the limit of small \( \epsilon \),
\( \mathcal{E}(\delta, \phi) = K_2 \sin^2 \phi \sin^2 (\theta_0 + \delta) + H_x \sin (\theta_0 + \delta) (1 - \cos \phi) + \mathcal{E}_1(\delta), \) \hspace{1cm} (18)

where \( \delta \equiv \theta - \theta_0 \) (\(|\delta| \ll 1 \) in the limit of \( \epsilon \ll 1 \)), and \( \mathcal{E}_1(\delta) \) is a function of only \( \delta \) given by
\[
\mathcal{E}_1(\delta) = \frac{1}{4} \sin (2\theta_c) (3\delta^2 \eta - \delta^3) + \frac{1}{2} \cos (2\theta_c) \left[ \delta^2 \left( \epsilon - \frac{3}{2} \eta^2 \right) + \delta^3 \eta - \frac{1}{4} \delta^4 \right]. \hspace{1cm} (19)
\]

In the following, we will investigate the tunneling behaviors of the magnetization vector at excited levels in single-domain FM nanoparticles with biaxial crystal symmetry at three different angle ranges of the external magnetic field as \( \theta_H = \pi/2, \pi/2 + O(\epsilon^{3/2}) < \theta_H < \pi - O(\epsilon^{3/2}) \), and \( \theta_H = \pi \), respectively.

**A. \( \theta_H = \pi/2 \)**

For \( \theta_H = \pi/2 \), we have \( \theta_c = \pi/2 \) from Eq. (16b) and \( \eta = \sqrt{2}\epsilon \) from Eq. (17). Eqs (18) and (19) show that \( \phi \) is very small for the full range of angles \( \pi/2 \leq \theta_H \leq \pi \) for FM particles with biaxial crystal symmetry. Performing the Gaussian integration over \( \phi \), we can map the spin system onto a particle moving problem in the one-dimensional potential well. Now the imaginary-time transition amplitude Eqs. (1) and (2) becomes
\[
U_{fi} = \int d\delta \exp \left( -S_E[\delta] \right),
\]
\[
= \int d\delta \exp \left\{ -\int d\tau \left[ \frac{1}{2} m \left( \frac{d\delta}{d\tau} \right)^2 + U(\delta) \right] \right\}, \hspace{1cm} (20)
\]
with
\[
m = \frac{\hbar S^2}{2V [K_2 + K_1 (1 - \epsilon)]},
\]
and
\[
U(\delta) = \frac{K_1 V}{4\hbar} \delta^2 \left( \delta - 2\sqrt{2\epsilon} \right)^2. \hspace{1cm} (21)
\]
The problem is one of MQC, where the magnetization vector resonates coherently between the energetically degenerate easy directions at \( \delta = 0 \) and \( \delta = 2\sqrt{2\epsilon} \) separated by a classically impenetrable barrier at \( \delta = \sqrt{2\epsilon} \).
Now we apply the periodic instanton method\textsuperscript{9,10} to evaluate the level splittings of excited states. The periodic (or thermal) instanton configuration $\delta_p$ which minimizes the Euclidean action in Eq. (20) satisfies the equation of motion

$$\frac{1}{2} m \left( \frac{d\delta_p}{d\tau} \right)^2 - U (\delta_p) = -E,$$  

(22)

where $E > 0$ is a constant of integration, which can be viewed as the classical energy of the pseudoparticle configuration. Then the kink-solution is\textsuperscript{10}

$$\delta_p = \sqrt{2\epsilon} + \sqrt{2\epsilon - \alpha} \text{sn} (\omega_1 \tau, k),$$  

(23)

where $\alpha = 2\sqrt{\hbar E/K_1 V}$, and $\omega_1 = \sqrt{K_1 V/2\hbar m} \sqrt{2\epsilon + \alpha}$. $\text{sn}(\omega_1 \tau, k)$ is the Jacobian elliptic sine function of modulus $k = \sqrt{(2\epsilon - \alpha)/(2\epsilon + \alpha)}$. In the low energy limit, i.e., $E \rightarrow 0$, $k \rightarrow 1$, $\text{sn}(u, 1) \rightarrow \tanh u$, we have

$$\delta_p = \sqrt{2\epsilon} [1 + \tanh (\bar{\omega}_1 \tau)],$$  

(24)

which is exactly the vacuum instanton solution derived in Ref. 4, where $\bar{\omega}_1 = \sqrt{2} (K_1 V/\hbar S)^{1/2} \sqrt{1 - \epsilon + K_2/K_1}$.

The Euclidean action of the periodic instanton configuration Eq. (23) over the domain $(-\beta, \beta)$ is found to be

$$S_p = \int_{-\beta}^{\beta} d\tau \left[ \frac{1}{2} m \left( \frac{d\delta_p}{d\tau} \right)^2 + U (\delta_p) \right] = W + 2E\beta,$$  

(25)

with

$$W = \frac{1}{3\sqrt{2}} \sqrt{\frac{K_1 V m}{\hbar}} (2\epsilon)^{3/2} \frac{1}{\sqrt{1 - k'^2/2}} \left[ E (k) - \frac{k'^2}{2 - k'^2} K (k) \right],$$  

(26)

where $k'^2 = 1 - k^2$. $K (k)$ and $E (k)$ are the complete elliptic integral of the first and second kind, respectively. The general formula Eq. (8) gives the tunnel splittings of excited levels as

$$\Delta E = \frac{\omega (E)}{\pi} \exp (-W),$$  

(27)
where \( W \) is shown in Eq. (26), and \( \omega(E) = 2\pi/t(E) \) is the energy-dependent frequency.

For this case, the period \( t(E) \) is found to be

\[
t(E) = \sqrt{2m} \int_{\delta_1}^{\delta_2} \frac{d\delta}{\sqrt{E - U(\delta)}} = \sqrt{\frac{2\hbar m}{K_1 V}} \frac{1}{\sqrt{2\epsilon + \alpha}} K(k'),
\]

(28)

where \( \delta_1 = \sqrt{2\epsilon + \sqrt{2\epsilon - \alpha}} \), and \( \delta_2 = \sqrt{2\epsilon + \sqrt{2\epsilon + \alpha}} \). Now we discuss the low energy limit where \( E \) is much less than the barrier height. In this case, \( k'^4 = 4\hbar E/K_1 V \epsilon^2 \ll 1 \), so we can perform the expansions of \( K(k) \) and \( E(k) \) in Eq. (26) to include terms like \( k'^4 \) and \( k'^4 \ln(4/k') \),

\[
E(k) = 1 + \frac{1}{2} \left( \ln \left( \frac{4}{k'} \right) - \frac{1}{2} \right) k'^2 + \frac{3}{16} \left( \ln \left( \frac{4}{k'} \right) - \frac{13}{12} \right) k'^4 \ldots,
\]

\[
K(k) = \ln \left( \frac{4}{k'} \right) + \frac{1}{4} \left( \ln \left( \frac{4}{k'} \right) - 1 \right) k'^2 + \frac{9}{64} \left( \ln \left( \frac{4}{k'} \right) - \frac{7}{6} \right) k'^4 \ldots.
\]

(29)

With the help of small oscillator approximation for energy near the bottom of the potential well, \( E_n = (n + 1/2) \Omega_1, \Omega_1 = \sqrt{U''(\delta = \sqrt{2\epsilon})/m} = 2\sqrt{K_1 V \epsilon/\hbar m} \), Eq. (26) is expanded as

\[
W = \frac{8}{3} \sqrt{\frac{K_1 V m}{\hbar}} \epsilon^{3/2} - \left( n + \frac{1}{2} \right) + \left( n + \frac{1}{2} \right) \ln \left[ \frac{1}{64 \epsilon^{3/2}} \sqrt{\frac{\hbar}{K_1 V m}} \left( n + \frac{1}{2} \right) \right].
\]

(30)

Then the general formula Eq. (8) gives the low-lying energy shift of \( n \)-th excited states for FM particles with biaxial crystal symmetry in the presence of an external magnetic field applied perpendicular to the anisotropy axis (\( \theta_H = \pi/2 \)) as

\[
\hbar \Delta E_n = \frac{2}{n! \sqrt{\pi}} (K_1 V) \epsilon^{1/2} S^{-1} \sqrt{1 - \epsilon + \lambda} \left( \frac{2^{11/2} \epsilon^{3/2} S}{\sqrt{1 - \epsilon + \lambda}} \right)^{n+1/2} 
\times \exp \left( -\frac{2^{5/2} S}{3 \sqrt{1 - \epsilon + \lambda} \epsilon^{3/2}} \right),
\]

(31)

where \( \lambda = K_2/K_1 \).

When \( n = 0 \), the energy shift of the ground state is

\[
\hbar \Delta E_0 = \frac{2^{15/4}}{\sqrt{\pi}} (K_1 V) (1 - \epsilon + \lambda)^{1/4} \epsilon^{5/4} S^{-1/2} \exp \left( -\frac{2^{5/2} S}{3 \sqrt{1 - \epsilon + \lambda} \epsilon^{3/2}} \right).
\]

(32)

Then Eq. (31) can be written as

\[
\hbar \Delta E_n = \frac{q^n}{n!} (\hbar \Delta E_0),
\]

(33)
where
\[ q_1 = \frac{2^{11/2}e^{3/2}S}{\sqrt{1 - \epsilon + \lambda}}. \]  
(34)

To see the temperature dependence we take the Boltzmann average of the tunneling frequency \( f = 4\Delta E \) at temperature \( T \),
\[ f (T) = \frac{1}{Z_0} \sum_n 4\Delta E_n \exp (-\hbar E_n \beta), \]  
(35)

where \( Z_0 = \sum_n \exp (-\hbar E_n \beta) \) is the partition function with the harmonic oscillator approximated eigenvalues \( E_n = (n + 1/2) \Omega_1 \). The final result of the tunneling frequency at a finite temperature \( T \) is found to be
\[ f (T) = 4\Delta E_0 (1 - e^{-\hbar \Omega_1 \beta}) \exp (q_1 e^{-\hbar \Omega_1 \beta}), \]  
(36)

where \( \Delta E_0 \) and \( q_1 \) are shown in Eqs. (32) and (34).

### B. \( \pi/2 + O (\epsilon^{3/2}) < \theta_H < \pi - O (\epsilon^{3/2}) \)

For \( \pi/2 + O (\epsilon^{3/2}) < \theta_H < \pi - O (\epsilon^{3/2}) \), the critical angle \( \theta_c \) is in the range of \( O (\epsilon^{3/2}) < \theta_c < \pi/2 - O (\epsilon^{3/2}) \), and \( \eta \approx \sqrt{2\epsilon/3} \). Now the problem can be mapped onto a problem of one-dimensional motion by integrating out \( \phi \), and for this case the effective mass \( m \) and the potential \( U (\delta) \) in Eq. (20) are found to be

\[ m = \frac{\hbar S^2}{2K_1 V \left[ \frac{1 - \epsilon}{1 + \left| \cot \theta_H \right| \eta^{3/2}} + \lambda \right]}, \]

and

\[ U (\delta) = \frac{K_1 V}{2\hbar} \sin 2\theta_c \left( \sqrt{6\epsilon \delta^2} - \delta^2 \right) = 3U_0 q^2 \left( 1 - \frac{2}{3} q \right), \]  
(37)

where \( q = 3\delta/2\sqrt{6\epsilon} \), and \( U_0 = \left( 2^{5/2}/3^{3/2} \right) \left( K_1 V \epsilon^{3/2}/\hbar \right) \sin 2\theta_c \). The problem becomes one of MQT, where the magnetization vector escapes from the metastable state at \( \delta = 0, \phi = 0 \) through the barrier by quantum tunneling.
Now the periodic bounce configuration with an energy $E > 0$ is found to be
\[
\delta_p = \frac{2}{3} \sqrt{6\epsilon} \left[ a - (a - b) \text{sn}^2(\omega_2 \tau, k) \right],
\]  
(38)

where
\[
\omega_2 = \frac{1}{2^{1/4} \times 3^{1/4}} \sqrt{\frac{K_1}{\hbar m}} \sin \frac{2\theta_e}{\sqrt[4]{\epsilon}} \sqrt{a - c}.
\]  
(39)

$a(E) > b(E) > c(E)$ denote three roots of the cubic equation
\[
q^3 - \frac{3}{2} q^2 + \frac{E}{2U_0} = 0.
\]  
(40)

$\text{sn}(\omega_2 \tau, k)$ is the Jacobian elliptic sine function of modulus $k = \sqrt{(a - b) / (a - c)}$. In the low energy limit, i.e., $E \to 0$, $k \to 1$, $\text{sn}(u, 1) \to \tanh u$, $a \to 3/2$, $b \to 0$, we have
\[
\delta_p = \frac{\sqrt{6\epsilon}}{\cosh^2(\tilde{\omega}_2 \tau)},
\]  
(41)

where
\[
\tilde{\omega}_2 = \frac{3^{1/4}}{2^{1/4}} \left( \frac{K_1}{\hbar S} \right) \epsilon^{1/4} \frac{\left| \cot \theta_H \right|^{1/6}}{1 + \left| \cot \theta_H \right|^{2/3}} \sqrt{1 - \epsilon + \lambda \left( 1 + \left| \cot \theta_H \right|^{2/3} \right)}.
\]

Eq. (41) agrees well with the vacuum bounce solution obtained in Ref. 4.

The classical action of the periodic bounce configuration Eq. (38) is
\[
S_p = \int_{-\beta}^{\beta} d\tau \left[ \frac{1}{2} m \left( \frac{d\delta_p}{d\tau} \right)^2 + U(\delta_p) \right] = W + 2E\beta,
\]  
(42)

with
\[
W = \frac{2^{9/2}}{5 \times 3^{3/2}} \sqrt{m \epsilon U_0 (a - c)^{5/2}} \left[ 2 \left( k^4 - k^2 + 1 \right) E(k) - \left( 1 - k^2 \right) \left( 2 - k^2 \right) K(k) \right].
\]  
(43)

The period $t(E)$ of this case is found to be
\[
t(E) = \sqrt{2m} \int_{c}^{b} \frac{d\delta}{\sqrt{E - U(\delta)}} = 4 \sqrt{\frac{2\epsilon m}{3U_0 (a - c)}} K(k'),
\]  
(44)

where $k'^2 = 1 - k^2$. Then the general formula Eq. (8) gives the imaginary parts of the metastable energy levels as
\[\text{Im} \ E = \frac{\omega (E)}{\pi} \exp (-W), \quad (45)\]

where \(\omega (E) = 2\pi /t (E)\), and \(W\) is shown in Eq. (43).

Here we discuss the low energy limit of the imaginary part of the metastable energy levels. For this case, \(E_n = (n + 1/2) \Omega_2, \ \Omega_2 = \sqrt{U''(\delta = 0)/m} = (3/2) \sqrt{U_0/m}\epsilon, \ a \approx (3/2)(1 - k'^2/4), \ b \approx (3k'^2/4)(1 + 3k'^2/4), \ c \approx -(3k'^2/4)(1 + k'^2/4), \text{ and } k'^4 = 16E/27U_0 \ll 1. \) Therefore, Eqs (43) and (45) reduce to

\[
W = \frac{2^{17/4} \times 3^{1/4}}{5} S\epsilon^{5/4} \left[ \frac{|\cot \theta_H|^{1/6}}{\sqrt{1 - \epsilon + \lambda (1 + |\cot \theta_H|^{2/3})}} \right]^{n+1/2} \\
+ \left( n + \frac{1}{2} \right) \ln \left[ \frac{2^{25/4} \times 3^{11/4} S\epsilon^{5/4} |\cot \theta_H|^{1/6}}{(n + \frac{1}{2}) \sqrt{1 - \epsilon + \lambda (1 + |\cot \theta_H|^{2/3})}} \right], \quad (46)
\]

and

\[
\hbar \text{Im} \ E_n = \frac{3^{1/4} \times 2^{3/4}}{n! \sqrt{\pi}} \epsilon^{1/4} S^{-1} (K_1 V) \left[ \frac{|\cot \theta_H|^{1/6}}{1 + |\cot \theta_H|^{2/3}} \right]^{n+1/2} \\
\times \left( \frac{2^{25/4} \times 3^{11/4} S\epsilon^{5/4} |\cot \theta_H|^{1/6}}{\sqrt{1 - \epsilon + \lambda (1 + |\cot \theta_H|^{2/3})}} \right)^{n+1/2} \\
\times \exp \left( -\frac{2^{17/4} \times 3^{1/4}}{5} S\epsilon^{5/4} \left[ \frac{|\cot \theta_H|^{1/6}}{\sqrt{1 - \epsilon + \lambda (1 + |\cot \theta_H|^{2/3})}} \right]^{n+1/2} \right). \quad (47)
\]

For vacuum bounce case \(n = 0\), we have

\[
\hbar \text{Im} \ E_0 = \frac{3^{13/9} \times 2^{31/8}}{\sqrt{\pi}} (K_1 V) \epsilon^{7/8} S^{-1/2} \left[ \frac{|\cot \theta_H|^{1/6}}{1 + |\cot \theta_H|^{2/3}} \right]^{1/4} \left[ 1 - \epsilon + \lambda (1 + |\cot \theta_H|^{2/3}) \right]^{1/4} \\
\times \exp \left( -\frac{2^{17/4} \times 3^{1/4}}{5} S\epsilon^{5/4} \left[ \frac{|\cot \theta_H|^{1/6}}{\sqrt{1 - \epsilon + \lambda (1 + |\cot \theta_H|^{2/3})}} \right]^{1/4} \right). \quad (48)
\]

Then Eq. (47) can be written as

\[
\hbar \text{Im} \ E_n = \frac{q^n}{n!} (\hbar \text{Im} \ E_0), \quad (49)
\]

where
\[ q_2 = \frac{2^{25/4} \times 3^{11/4} S \epsilon^{5/4} |\cot \theta_H|^{1/6}}{\sqrt{1 - \epsilon + \lambda \left(1 + |\cot \theta_H|^{2/3}\right)}}. \]  

(50)

At finite temperature \( T \) the decay rate \( \Gamma = 2 \text{Im} \, E \) can be easily found by averaging over the Boltzmann distribution

\[ \Gamma (T) = \frac{2}{Z_0} \sum_n \text{Im} \, E_n \exp \left(-E_n \beta\right), \]  

(51)

where \( Z_0 = \sum_n \exp (-\hbar E_n \beta) \) is the partition function with the harmonic oscillator approximated eigenvalues \( E_n = (n + 1/2) \Omega_2 \). The final result of the decay rate at a finite temperature \( T \) is found to be

\[ \Gamma (T) = 2 \text{Im} \, E_0 \left(1 - e^{-\hbar \Omega_2 \beta}\right) \exp \left(q_2 e^{-\hbar \Omega_2 \beta}\right), \]  

(52)

where \( \text{Im} \, E_0 \) and \( q_2 \) are shown in Eqs. (48) and (50).

In Fig. 1 we plot the temperature dependence of the tunneling rate for the typical values of parameters for nanometer-scale single-domain ferromagnets: \( S = 5000, \epsilon = 1 - \pi / \pi c = 2 \times 10^{-3}, \lambda = K_2 / K_1 = 10, \) and \( \theta_H = 3\pi / 4 \). From Fig. 1 we easily see the crossover from purely quantum tunneling to thermally assisted quantum tunneling. The temperature \( T_0^{(0)} \) characterizing the crossover from quantum to thermal regimes can be estimated as \( k_B T_0^{(0)} = \Delta U / S_0 \), where \( \Delta U \) is the barrier height, and \( S_0 \) is the WKB exponent of the ground-state tunneling. It is shown that in the cubic potential \( (q^2 - q^3) \), the usual second-order phase transition from the thermal to the quantum regimes occurs as the temperature is lowered. The second-order phase transition temperature is given by \( k_B T_0^{(2)} = \hbar \omega_b / 2\pi \), where \( \omega_b = \sqrt{|U''(x_b)|} / m \) is the frequency of small oscillations near the bottom of the inverted potential \( -U(x) \), and \( x_b \) corresponds to the bottom of the inverted potential. For the present case, \( \delta_b = 2 \sqrt{6\epsilon / 3} \),

\[ \hbar \omega_b = 2^{5/4} \times 3^{1/4} (K_1 V) S^{-1} \epsilon^{1/4} \frac{|\cot \theta_H|^{1/6}}{1 + |\cot \theta_H|^{2/3}} \sqrt{1 - \epsilon + \lambda \left(1 + |\cot \theta_H|^{2/3}\right)}, \]

\[ S_0 = \frac{2^{17/4} \times 3^{1/4}}{5} S \epsilon^{5/4} \frac{|\cot \theta_H|^{1/6}}{\sqrt{1 - \epsilon + \lambda \left(1 + |\cot \theta_H|^{2/3}\right)}}. \]
\[ h \Delta U = \frac{2^{7/2}}{3^{3/2}} (K_1 V) \epsilon^{3/2} \frac{|\cot \theta_H|^{1/6}}{1 + |\cot \theta_H|^{2/3}}. \]

Then it is easy to obtain that
\[ k_B T_0^{(2)} = \frac{2^{1/4} \times 3^{1/4}}{\pi} (K_1 V) S^{-1} \epsilon^{1/4} \frac{|\cot \theta_H|^{1/6}}{1 + |\cot \theta_H|^{2/3}} \sqrt{1 - \epsilon + \lambda (1 + |\cot \theta_H|^{2/3})}, \]
and \[ k_B T_0^{(0)} = (5\pi/18) k_B T_0^{(2)} \approx 0.87 k_B T_0^{(2)}. \]

C. \( \theta_H = \pi \)

In case of \( \theta_H = \pi \), we have \( \theta_c = 0 \) and \( \eta = 0 \). Working out the integration over \( \phi \), the spin tunneling problem is mapped onto the problem of a particle with effective mass
\[ m = \hbar S^2/2K_2V \]
moving in the one dimensional potential well \( U(\delta) = (K_1 V/\hbar) (\epsilon \delta^2 - \delta^4/4) \).
Now the problem is one of MQT, and the nonvacuum bounce at a given energy \( E > 0 \) is found to be
\[ \delta_p = \sqrt{2\epsilon} \left( 1 + \sqrt{1 - \frac{\hbar E}{K_1 V \epsilon^2}} \right)^{1/2} \text{dn}(\omega_3 \tau, k), \] (53)
where
\[ \omega_3 = \sqrt{\frac{K_1 V}{\hbar m}} \left( 1 + \sqrt{1 - \frac{\hbar E}{K_1 V \epsilon^2}} \right)^{1/2}, \]
\[ k^2 = 1 - \left( \frac{1 - \sqrt{1 - \frac{\hbar E}{K_1 V \epsilon^2}}}{1 + \sqrt{1 - \frac{\hbar E}{K_1 V \epsilon^2}}} \right)^2. \]
In the low energy limit, i.e., \( E \to 0, k \to 1, \text{dn}(u, 1) \to 1/\cosh u \), we have
\[ \delta_p = \frac{2\sqrt{\epsilon}}{\cosh (\bar{\omega}_3 \tau)}, \] (54)
where \( \bar{\omega}_3 = 2\sqrt{K_1 K_2 \epsilon V/\hbar S} \). Eq. (54) is in good agreement with the vacuum bounce solution derived in Ref. 4.

The classical action of the nonvacuum bounce Eq. (53) is
\[ S_p = \int_{-\beta}^{\beta} d\tau \left[ \frac{1}{2} m \left( \frac{d\delta_p}{d\tau} \right)^2 + U(\delta_p) \right] = W + 2E\beta, \quad (55) \]

with

\[ W = \frac{4}{3} m e \left( 1 + \sqrt{1 - \frac{\hbar E}{K_1 V \epsilon^2}} \right)^2 \omega_3 \left[ (2 - k^2) E(k) - 2k'^2 K(k) \right], \quad (56) \]

where \( k'^2 = 1 - k^2 \). Then the imaginary parts of the metastable energy levels are

\[ \text{Im} E = \frac{\omega(E)}{\pi} \exp(-W), \quad (57) \]

where \( \omega(E) = 2\pi/t(E) \), and the period \( t(E) \) for this case is found to be

\[ t(E) = 4\sqrt{\frac{\hbar m}{K_1 V \epsilon}} \frac{1}{1 + \sqrt{1 - \frac{\hbar E}{K_1 V \epsilon^2}}} K(k'), \quad (58) \]

Now we consider the low energy limit of the imaginary part of the metastable energy level. For this case, \( E_n = (n + 1/2) \Omega_3 \), \( \Omega_3 = \sqrt{U''(\delta = 0)/m} = \sqrt{2K_1 V \epsilon / \hbar m} \), \( k'^2 = (1/2^{3/2} \epsilon^{3/2}) \sqrt{\hbar / K_1 V m} (n + 1/2) \ll 1 \), then

\[ W = \frac{8}{3} \sqrt{\frac{K_1}{K_2}} S \epsilon^{3/2} - \left( n + \frac{1}{2} \right) - \left( n + \frac{1}{2} \right) \ln \left[ \frac{32 S \epsilon^{3/2}}{3 \sqrt{\lambda}} \right], \quad (59) \]

and

\[ \hbar \text{Im} E_n = \left( K_1 V \right) \lambda^{1/2} S^{-1/2} \epsilon^{1/2} \left( \frac{32 S \epsilon^{3/2}}{3 \sqrt{\lambda}} \right)^{n+1/2} \exp \left( -\frac{8 S \epsilon^{3/2}}{3 \sqrt{\lambda}} \right). \quad (60) \]

In the case of \( n = 0 \), the imaginary part of the metastable ground state reduces to

\[ \hbar \text{Im} E_0 = \frac{8}{\sqrt{\pi}} \left( K_1 V \right) S^{-1/2} \epsilon^{5/4} e^{-\frac{8 S \epsilon^{3/2}}{3 \sqrt{\lambda}}} \exp \left( -\frac{8 S \epsilon^{3/2}}{3 \sqrt{\lambda}} \right). \quad (61) \]

Then Eq. (60) can be written as

\[ \hbar \text{Im} E_n = \frac{q_n}{n!} (\hbar \text{Im} E_0), \quad (62) \]

where

\[ q_3 = \frac{32 S \epsilon^{3/2}}{3 \sqrt{\lambda}}. \quad (63) \]
And the final result of the decay rate at finite temperature $T$ is found to be

$$
\Gamma (T) = 2 \text{Im} \, E_0 \left( 1 - e^{-\hbar \Omega_3 \beta} \right) \exp \left( q_3 e^{-\hbar \Omega_3 \beta} \right).
$$

(64)

The temperature dependence of the decay rate is shown in Fig. 2. It can be shown that the double-well potential $(q^2 - q^4)$ yields the second-order phase transition from the thermal to the quantum regimes as the temperature is lowered. For this case, the position of the energy barrier is $\delta_b = \sqrt{2} \epsilon$, the frequency of small oscillations near the bottom of the inverted potential is $\hbar \omega_b = 2^{3/2} \left( \sqrt{K_1 K_2 V} \right) \epsilon^{1/2} S^{-1}$, the WKB exponent of the ground-state tunneling is $S_0 = \left( 8/3 \sqrt{\lambda} \right) S \epsilon^{3/2}$, and the height of barrier is $\hbar \Delta U = (K_1 V) \epsilon^2$. Therefore, $k_B T_0^{(2)} = \left( \sqrt{2}/\pi \right) \left( \sqrt{K_1 K_2 V} \right) \epsilon^{1/2} S^{-1}$, and $k_B T_0^{(0)} = \left( 3\pi / 8 \sqrt{2} \right) k_B T_0^{(2)} \approx 0.83 k_B T_0^{(2)}$.

**IV. CONCLUSIONS**

In summary we have investigated the quantum tunneling of the magnetization vector between excited levels in single-domain FM nanoparticles with biaxial crystal symmetry in the presence of an external magnetic field at arbitrarily directed angle. By applying the periodic instanton method in the spin-coherent-state path-integral representation, we obtain the analytic formulas for the tunnel splitting between degenerate excited levels in MQC and the imaginary parts of the metastable energy levels in MQT of the magnetization vector in the low barrier limit for the external magnetic field perpendicular to the easy axis ($\theta_H = \pi/2$), for the field antiparallel to the initial easy axis ($\theta_H = \pi$), and for the field at an angle between these two orientations ($\pi/2 + O(\epsilon^{3/2}) < \theta_H < \pi - O(\epsilon^{3/2})$). The temperature dependences of the tunneling frequency and the decay rate are clearly shown for each case. One important conclusion is that the tunneling rate and the tunnel splitting at excited levels depend on the orientation of the external magnetic field distinctly. Even a small misalignment of the field with $\theta_H = \pi/2$ and $\pi$ orientations can completely change the results of the tunneling rates. Another interesting conclusion concerns the field strength dependence of the WKB exponent in the tunnel splitting or the tunneling rate. It is found
that in a wide range of angles, the $\epsilon \left( = 1 - \frac{H}{H_c} \right)$ dependence of the WKB exponent is given by $\epsilon^{5/4}$, not $\epsilon^{3/2}$ for $\theta_H = \pi/2$, and $\theta_H = \pi$. As a result, we conclude that both the orientation and the strength of the external magnetic field are the controllable parameters for the experimental test of the phenomena of macroscopic quantum tunneling and coherence of the magnetization vector between excited levels in single-domain FM nanoparticles at sufficiently low temperatures. The theoretical calculations performed in this paper can be extended to the FM particles with a much more complex structure of magnetocrystalline anisotropy energy, such as trigonal, tetragonal, and hexagonal crystal symmetries. Work along this line is still in progress. We hope that the theoretical results presented in this paper may stimulate more experiments whose aim is observing macroscopic quantum tunneling and coherence in nanometer-scale single-domain ferromagnets.

ACKNOWLEDGMENTS

R.L. would like to acknowledge Dr. Hui Hu, Dr. Yi Zhou, Dr. Jian-She Liu, Professor Zhan Xu, Professor Jiu-Qing Liang and Professor Fu-Cho Pu for stimulating discussions. R. L. and J. L. Zhu would like to thank Professor W. Wernsdorfer and Professor R. Sessoli for providing their paper (Ref. 15).
REFERENCES

1 For a review, see Quantum Tunneling of Magnetization, edited by L. Gunther and B. Barbara (Kluwer, Dordrecht, 1995); and E. M. Chudnovsky and J. Tejada, Macroscopic Quantum Tunneling of the Magnetic Moment (Cambridge University Press, 1997).

2 O. B. Zaslavskii, Phys. Rev. B 42, 992 (1990).

3 M. -G. Miguel and E. M. Chudnovsky, Phys. Rev. B 54, 388 (1996).

4 G. -H. Kim and D. S. Hwang, Phys. Rev. B 55, 8918 (1997).

5 G. -H. Kim, Phys. Rev. B 57, 10688 (1998).

6 D. A. Garanin, X. M. Hidalgo, and E. M. Chudnovsky, Phys. Rev. B 57, 13639 (1998).

7 W. Wernsdorfer, E. B. Orozco, K. Hasselbach, A. Benoit, D. Mailly, O. Kubo, H. Nakano, and B. Barbara, Phys. Rev. Lett. 79, 4014 (1997).

8 Rong Lü, Jia-Lin Zhu, Xiao-Bing Wang, and Lee Chang, Phys. Rev. B 60, 4101 (1999).

9 J. -Q. Liang, Y. -B. Zhang, H. J. W. Müller-Kirstein, Jian-Ge Zhou, F. Zimmerschied, F. -C. Pu, Phys. Rev. B 57, 529 (1998); J. -Q. Liang, H. J. W. Müller-Kirstein, and Jian-Ge Zhou, Z. Phys. B 102, 525 (1997); S. P. Kou, J. Q. Liang, Y. B. Zhang, and F. C. Pu, Phys. Rev. B 59, 11792 (1999).

10 J. -Q. Liang, H. J. W. Müller-Kirstein, and D. H. Tchrakian, Phys. Lett. B 282, 105 (1992); J. -Q. Liang and H. J. W. Müller-Kirstein, Phys. Rev. D 46, 4685 (1992); 50, 6519 (1994); 51, 718 (1995).

11 D. Loss, D. P. DiVicenzo, and G. Grinstein, Phys. Rev. Lett. 69, 3232 (1992).

12 J. V. Delft and G. L. Henley, Phys. Rev. Lett. 69, 3236 (1992).

13 A. Garg, Europhys. Lett. 22, 205 (1993).

14 H. B. Braun and D. Loss, Europhys. Lett. 31, 555 (1995).
15 W. Wernsdorfer and R. Sessoli, Science 284, 133 (1999).

16 L. D. Landau and E. M. Lifshita, *Quantum Mechanics* (Pergamon, London, 1965).

17 E. M. Chudnovsky and D. A. Garanin, Phys. Rev. Lett. 79, 4469 (1997).

18 E. M. Chudnovsky, Phys. Lett. A 46, 8011 (1992); D. A. Garanin and E. M. Chudnovsky, Phys. Rev. B 56, 11102 (1997).

19 C. S. Park, S. -K. Yoo, D. K. Park, and D. -H. Yoon, cond-mat/9807344; C. S. Park, S. -K. Yoo, and D. -H. Yoon, cond-mat/9909217.

20 J. -Q. Liang, H. J. W. Müller-Kirstein, D. K. Park, and F. Zimmerschied, Phys. Rev. Lett. 81, 216 (1998); S. Y. Lee, H. J. W. Müller-Kirstein, D. K. Park, and F. Zimmerschied, Phys. Rev. B 58, 5554 (1998); H. J. W. Müller-Kirstein, D. K. Park, and J. M. S. Rana, cond-mat/9902184.

21 D. A. Gorokhov and G. Blatter, Phys. Rev. B 56, 3130 (1997); 57, 3586 (1998); 58, 5486 (1998).

Figure Captions:

Fig. 1 The temperature dependence of the relative decay rate \( \Gamma (T) / \Gamma (T = 0K) \) for FM particles in a magnetic field with a range of angles \( \pi / 2 + O (\epsilon^{3/2}) < \theta_H < \pi - O (\epsilon^{3/2}) \). Here, \( S = 5000, \epsilon = 1 - H / H_c = 2 \times 10^{-3}, \lambda = K_2 / K_1 = 10, \) and \( \theta_H = 3\pi/4 \).

Fig. 2 The temperature dependence of the relative decay rate \( \Gamma (T) / \Gamma (T = 0K) \) for FM particles in a magnetic field along \( \pi \). Here, \( S = 5000, \epsilon = 1 - H / H_c = 0.03, \) and \( \lambda = K_2 / K_1 = 10 \).
\( S = 5000 \)
\( \varepsilon = 0.002 \)
\( \lambda = 10 \)
\( \theta_H = \frac{3\pi}{4} \)
$S = 5000$
$\varepsilon = 0.03$
$\lambda = 10$