Topological phase interference induced by a magnetic field along hard anisotropy axis in nanospin systems with different crystal symmetries

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Based on the instanton technique in the spin-coherent-state path-integral representation, the spin-parity effects induced by the topological Wess-Zumino or Berry phase are studied theoretically in nanometer-scale single-domain ferromagnets in the presence of an external magnetic field along the hard anisotropy axis. We consider the magnetocrystalline anisotropy with the biaxial, trigonal, tetragonal, and hexagonal crystal symmetry, respectively. Both the Wentzel-Kramers-Brillouin (WKB) exponent and the preexponential factors are evaluated in the instanton's contribution to the ground-state tunnel splittings. The Euclidean transition amplitudes between energetically degenerate easy directions are obtained with the help of the dilute instanton gas approximation. The effective Hamiltonian approach is applied to present the final results of ground-state tunnel splittings for each kind of crystal symmetry. The Euclidean transition amplitudes and the ground-state tunnel splittings are found to depend on the parity of total spins of ferromagnets and oscillate with the external magnetic field for both the integer and half-integer total spins. We show that the topological phase interference or spin-parity effects can reflect in thermodynamic quantities of magnetic tunneling states. Possible relevance to experiments is also discussed.

I. INTRODUCTION

It is well known that in principle quantum mechanics is applicable to both macroscopic and microscopic systems. Macroscopic systems are essentially classical, in the sense that in such systems phenomena typical of quantum mechanics, such as barrier penetration or Schrödinger's cat, do not occur in general. However, Caldeira and Leggett predicted that quantum tunneling could occur on the macroscopic scale, provided that the dissipative interactions with the environment were small enough.[1,2] By applying the instanton technique in the imaginary-time path integral, they concluded that the rate of quantum tunneling was reduced by the dissipation in general.[1–4] Macroscopic quantum phenomena can take place in the Josephson systems and the superconducting quantum interference device (SQUID).[5]

Recently, there has been great theoretical and experimental interest in studying the nanometer-scale ferromagnetic[6–13] (FM) and antiferromagnetic (AFM) particles[13–19] which exhibit macroscopic quantum tunneling (MQT) and coherence (MQC). Theoretical study based on the instanton technique was performed by Chudnovsky and Gunther[6] with an exponential accuracy, and Enz and Schilling[7] developed a more sophisticated version of the instanton technique to obtain the ground-state tunnel splitting with the preexponential factors. Van Hemmen and Süto[8] formulated the WKB method and calculated the tunneling rates and the corresponding splittings of excited states. Scharf, Wreszinski, and van Hemmen[9] proposed an approach based on a particle mapping with subsequent application of the WKB method to refine the results for the splittings of excited levels with moderate spin. By applying the instanton technique in the spin-coherent-state path integral, Garg and Kim[10] extended the previous WKB calculation to include the preexponential factors for various forms of the magnetocrystalline anisotropy. Zaslavskii[11] described the spin tunneling based on the exact correspondence between the spin system and a particle moving in a potential field. And he obtained the tunneling exponent, the preexponential factors, and their temperature dependences. Barnes proposed the auxiliary particle method[12] to study the model for a single large spin subject to the external and anisotropy fields.[13] By introducing auxiliary spinless fermions, Barnes mapped the spin model to two tight binding models of spinless fermions, then obtained the tunnel splitting and discussed the spin-parity effects.[13] For AFM particles with an easy-plane anisotropy, Barnes[13] showed that the tunnel splitting is quasiperiodic in the magnitude of a field applied perpendicular to a principal anisotropy axis. He also found that in one dimension there are periodic regions on the field axis for which the model is quantum critical, while in two or three dimensions criticality is reduced to points. Both the Zaslavskii method and the auxiliary particle method gave a clear picture to calculate the splitting of excited levels which was not easily obtained by using the path integral method. Now much attention was attracted to the spin tunneling in the presence of an arbitrarily directed magnetic field[20–23] and the quantum-classical phase transition problem.[24,25] Experiments were carried out to investigate the small magnets either via relaxation measurements,[26,27] or via measurements of the noise spectrum and the ac susceptibility,[28,29] which can be considered as signals of magnetic quantum tunneling. Besides its importance in understanding the transition from quantum to classical physics, the spin tunneling is crucial to the reliability of small magnetic units in memory devices and the design of possible quantum computers.[30]

One notable subject in magnetic quantum tunneling is that the topological Berry phase,[31] or Wess-Zumino, Chern-Simons term[32] in the action can lead to remarkable parity effects for some spin systems with high symmetries. It was theoretically shown that the tunnel splitting is suppressed to zero for half-integer total spins in FM particles with biaxial crystal symmetry in the absence of a magnetic field.[33,34] Such a effect is known as the topological quenching.[35] However, the phase interference is constructive for integer spins, and hence the splitting is nonzero.[33,34] Similar effects were found in AFM particles, where only the integer excess spins can tunnel but not the half-integer ones.[21,22] While spin-parity effects in the absence of magnetic field are sometimes related to the Kramers degeneracy,[33,34] they typically go beyond the Kramers theorem in a rather unexpected way.[35,36] The effects of magnetic field were studied extensively in FM and AFM particles with biaxial crystal symmetry.[13,35,37–41] One recent experimental method based on the Landau-Zener model was developed by Wernsdorfer and Sessoli[42] to measure the tunnel splittings in the molecular Fe_8 cluster with a spin ground state of $S=10$. They observed a clear oscillation of the tunnel splitting as a function of the magnetic field along the hard axis, which is a direct evidence of the role of the topological Berry phase in spin dynamics of these molecules. They also observed an oscillatory rate in the presence of a dc field along the easy axis that is such as to align the ground level in one well with an excited level in the other. Whether the instanton technique can be applied in studying the spin dynamics in molecule with $S=10$, and under what conditions the Landau-Zener model is appropriate for the quantum relaxation of unstable excited states become two important questions. Recent theoretical studies include the quantum relaxation in magnetic molecules,[43,44] the spin tunneling in a swept magnetic field,[45] the thermally activated resonant tunneling with the help of the perturbation theory[46] and the exact diagonalization,[47] the auxiliary particle method,[12,13] the discrete WKB method,[48] and the nonadiabatic Landau-Zener model.[49] Barnes[12,13] showed that the intermediate spin, which occurs in the theory of anyons, can be exhibited by magnets in a suitable directed magnetic field. By using the auxiliary particle method, Barnes obtained the tunnel splitting of Fe_8 and showed that the parity is periodic for a field which is along either the easy or the hard axis or a suitable combination of the two.[13] Prokof'ev and Stamp found that the higher order term $c\ \ (S_+^4+S_-^4)$ makes an important contribution to the period of oscillation and markedly affect the tunnel splitting of Fe_8, and the nuclear spins also affect the tunnel splitting.[50]

It is noted that previous results[35] about the effects of the magnetic field along the hard anisotropy axis on topological phase interference were obtained for the simplest possible form of the magnetocrystalline anisotropy (the biaxial crystal symmetry). The purpose of this paper is to extend the result of FM particles with biaxial crystal symmetry[35] to that with a more complex structure, such as trigonal, tetragonal, and hexagonal crystal symmetries around the $\hat{z}$ axis, which have three, four, and six degenerate easy directions, respectively. The theoretical study based on the path-integral method consists of two major steps. The first step is to find the classical, or least-action path (instanton) from the equation of motion, and then to obtain the contribution of the associated instanton to the tunnel splittings. Garg and Kim[10] studied the general formulas for evaluating the instanton's contribution to the tunneling rate or the tunnel splitting. In Sec. II, we explain briefly the basic ideas of this evaluation, and then apply this method to calculate the instanton's contribution to the ground-state tunnel splittings for FM particles with biaxial, trigonal, tetragonal, and hexagonal crystal symmetry at finite field in Secs. III–VI, respectively. The second step is to study the effects caused by the topological Wess-Zumino phase, and then obtain the final results of the ground-state splittings. For FM particles with simple biaxial crystal symmetry, this step is easily done by summing up the contributions of clockwise and counterclockwise tunneling paths.[35] However, for FM particles with complex symmetry, this step turns out to be more difficult. By using the dilute instanton-gas approximation, we obtain the transition amplitudes between degenerate states, which lead to some direct results concerning the topological phase interference effects. Furthermore, we propose an approach of the effective Hamiltonian to obtain the low-lying tunneling level spectrum and discuss the degeneracies of tunneling levels. The topological quenching for half-integer spins in FM particles with biaxial crystal symmetry at zero field can be rederived by applying this approach. Our results show that the tunnel splittings depend significantly on the parity of total spins for each kind of crystal symmetries. And the structure of low-lying tunneling level spectrum for the trigonal, tetragonal, and hexagonal crystal symmetry is much more complex than that for the biaxial crystal symmetry. We also find that the tunnel splittings oscillate with the field for both integer and half-integer spins, and the oscillation behavior for integer spins are much different from that for half-integer spins. Thermodynamic properties (such as the specific heat or the magnetic susceptibility) of the tunneling levels are evaluated, and are found to be strongly dependent on the parity of total spins and oscillate with the field. This may provide an experimental test for spin-parity effects in FM particles.

II. BERRY PHASE IN SPIN-COHERENT-STATE PATH INTEGRAL

In this section, we review briefly the magnetic MQT and MQC based on the instanton method in the spin-coherent-state path integral. We emphasize that the Wess-Zumino phase which is crucial for the spin-parity effects arises from the nonorthogonality of spin coherent states, and the gauges are determined by the single-valuedness of spin coherent states.

In the spin-coherent-state path-integral representation, the Euclidean action for a FM particle is given by

$$S_E(\theta,\phi)=\frac{V}{\hbar}\int d\tau\left[i\frac{M_0}{\gamma}\left(\frac{d\phi}{d\tau}\right)-i\frac{M_0}{\gamma}\left(\frac{d\phi}{d\tau}\right)\cos\theta+E(\theta,\phi)\right], \tag{1}$$

where $M_0=|\vec{M}|=\hbar\gamma S/V$, V is the volume of the particle, γ is the gyromagnetic ratio, and S is the total spins. The spin coherent state is defined as the maximum eigenstates of $\hat{S}_z$, $|S,M=S\rangle$, rotated into the direction of the unit vector $\vec{n}=(\sin\theta\cos\phi,\sin\theta\sin\phi,\cos\theta)$,

$$|\vec{n}\rangle\equiv|\theta,\phi\rangle\equiv e^{-i\hat{S}_z\phi}e^{-i\hat{S}_y\theta}e^{-i\hat{S}_z\chi}|S,S\rangle, \tag{2}$$

where $\vec{S}$ is the spin operator. The spin coherent state Eq. (2) obeys the eigenvalue equation $\vec{n}\cdot\vec{S}|\vec{n}\rangle=S|\vec{n}\rangle$. The spin coherent state can be expanded in terms of $|S,M\rangle$ by applying Wigner's formula,[51] and the result is[52]

$$|\vec{n}\rangle=e^{-iS\chi}\sum_{M=-S}^{S}\sqrt{\frac{(2S)!}{(S+M)!(S-M)!}}\times e^{-iM\phi}\left(\cos\frac{\theta}{2}\right)^{S+M}\left(\sin\frac{\theta}{2}\right)^{S-M}|S,M\rangle. \tag{3}$$

One has the freedom to define χ arbitrarily. This is a gauge freedom, which can be eliminated by fixing χ. χ has to be fixed by the requirement that the spin coherent state be single valued upon $\phi\rightarrow\phi+2\pi n$, where n is an integer.[53] Therefore, χ takes the values

$$\chi=\left(\frac{m}{nS}+1\right)\phi, \tag{4}$$

for arbitrary integers m and n. For $m=-2nS$, $\chi=-\phi$, and this is the case of north-pole gauge which we will adopt thought this paper. For $m=0$, $\chi=\phi$, and this is the case of south-pole gauge. And the results obtained in either of the gauges is physically equivalent.

It is noted that the first two terms in Eq. (1) define the topological Berry or Wess-Zumino, Chern-Simons term which arises from the nonorthogonality of spin coherent states,

$$\langle\vec{n}'|\vec{n}\rangle=\left(\cos\frac{\theta'}{2}\cos\frac{\theta}{2}+\sin\frac{\theta'}{2}\sin\frac{\theta}{2}e^{i(\phi-\phi')}\right)^{2S}. \tag{5}$$

For infinitesimally separated angles, the overlap becomes

$$\langle\vec{n}'|\vec{n}\rangle=1+iS\delta\phi(\cos\theta-1), \tag{6}$$

for the north-pole parametrization $\chi=-\phi$, where $\delta\phi=\phi'-\phi$. 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. (1) 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.[33,34]

In the semiclassical limit, the dominant contribution to the transition amplitude comes from finite action solution (instanton) of the classical equation of motion. The motion of the magnetization vector $\vec{M}$ is determined by the imaginary-time version of classical Landau-Lifshitz equation,

$$i\frac{d\vec{M}}{d\tau}=-\gamma\vec{M}\times\frac{dE(\vec{M})}{d\vec{M}}, \tag{7}$$

which can be expressed as the following equations in the spherical coordinate system:

$$i\left(\frac{d\overline{\theta}}{d\tau}\right)\sin\overline{\theta}=\frac{\gamma}{M_0}\frac{\partial E}{\partial\overline{\phi}}, \tag{8a}$$

$$i\left(\frac{d\overline{\phi}}{d\tau}\right)\sin\overline{\theta}=-\frac{\gamma}{M_0}\frac{\partial E}{\partial\overline{\theta}}, \tag{8b}$$

where $\overline{\theta}$ and $\overline{\phi}$ denote the classical path. The instanton's contribution to the tunneling rate Γ or the tunnel splitting Δ (not including the Wess-Zumino phase) is given by[10]

$$\Gamma\quad(\text{or}\quad\Delta)=A\omega_p\left(\frac{S_{cl}}{2\pi}\right)^{1/2}e^{-S_{cl}}, \tag{9}$$

where ω_p is the oscillation frequency in the well, and S_{cl} is the classical action. The factor A originates from the quantum fluctuations about the classical path, which can be evaluated by expanding the Euclidean action to second order in the small fluctuations.[10]

III. MQC FOR BIAXIAL CRYSTAL SYMMETRY

In this section, we study the topological phase interference or spin-parity effects in single-domain FM nanoparticles with biaxial crystal symmetry. In a magnetic field along the hard anisotropy axis $\hat{z}$, the Hamiltonian of this system can be written as

$$\mathcal{H}=k_1\hat{S}_z^2+k_2\hat{S}_y^2-\gamma H\hat{S}_z, \tag{10}$$

where $k_1\gg k_2>0$ are proportional to the anisotropy coefficients. Then the total energy $E(\theta,\phi)$ of such a FM particle is

$$E(\theta,\phi)=K_1\cos^2\theta+K_2\sin^2\theta\sin^2\phi-M_0H\cos\theta+E_0, \tag{11}$$

where K_1 and K_2 are the transverse and longitudinal anisotropy coefficients, and E_0 is a constant which makes $E(\theta,\phi)$ zero at the initial orientation. Here we assume that the transverse anisotropy coefficient is much larger than the longitudinal one, i.e., $K_1\gg K_2>0$, which satisfies the experimental situation on highly anisotropic materials. The same model was first studied in Ref. 35. However, the main purpose of Ref. 35 was to study the phase interference effects caused by the Wess-Zumino term. The expression of tunnel splitting was not clearly shown in Ref. 35. Here we will evaluate both the WKB exponent and the preexponential factors in the ground-state tunnel splitting at finite magnetic field by applying the instanton technique, which may be helpful for experimental checks. It is noted that the experiment[42] on Fe_8 is not covered in this situation. In particular, whether the instanton method can be applied in a spin system with $S=10$ needs further investigation. However, the theoretical studies[6,37] based on the instanton method showed that the tunneling probability is exponentially small for large spins $S\sim10^2$–10^3 at zero field, unless $K_1\gg K_2>0$. Therefore, the results presented here is useful for the highly anisotropic materials.

After adding a constant, we rewrite Eq. (11) as

$$E(\theta,\phi)=K_1(\cos\theta-\cos\theta_0)^2+K_2\sin^2\theta\sin^2\phi, \tag{12}$$

where $\cos\theta_0 = M_0H/2K_1 = H/H_c$. $H_c = 2K_1/M_0$ is the coercive field at which the initial state becomes classically unstable. Under the assumption that $0 \leqslant H < H_c$, the energy minima of this system are at $\theta = \theta_0$ and $\phi = 0, \ \pi$. We denote $|1\rangle$ as the state at $\theta = \theta_0, \ \phi = 0$, and $|2\rangle$ as the state at $\theta = \theta_0, \ \phi = \pi$, other energy minima repeat the two states with period 2π. For the case of high transverse anisotropy, the magnetization vector is forced to lie in the $\theta = \theta_0$ plane, so the fluctuations of θ about θ_0 are small. Introducing $\theta = \theta_0 + \alpha(|\alpha| \ll 1)$, the total energy of Eq. (12) reduces to

$$E(\alpha,\phi) = K_1 \sin^2\theta_0 \alpha^2 + K_2 \sin^2\theta_0 \sin^2\phi. \tag{13}$$

Substituting Eq. (13) into the classical equations of motion, we obtain the instanton solution

$$\bar{\alpha} = -i\sqrt{\frac{K_2}{K_1}} \frac{1}{\cosh(\omega_0\tau)},$$

$$\sin\bar{\phi} = \frac{1}{\cosh(\omega_0\tau)}, \tag{14}$$

corresponding to the switching of $\vec{M}$ from $\bar{\theta} = \theta_0, \ \bar{\phi} = 0$ at $\tau = -\infty$ to $\bar{\theta} = \theta_0, \ \bar{\phi} = \pi$ at $\tau = \infty$, where $\omega_0 = 2(V/\hbar S)\sin\theta_0\sqrt{K_1K_2}$. In deriving Eqs. (13) and (14), we have used the approximation that $|\alpha| \ll |\theta_0|$. It is easy to show that this approximation is valid for the weak magnetic field. For the strong magnetic field, i.e., $H \to H_c, \ \theta_0 = \sqrt{2\epsilon}$, where $\epsilon = 1 - H/H_c \ll 1$. From Eq. (14), $|\alpha| \approx \sqrt{K_2/K_1} \approx 0.1$ for typical values of parameters for single-domain FM nanoparticles ($K_1 \approx 10^7$ erg/cm^3 and $K_2 \approx 10^5$ erg/cm^3). Therefore, $|\alpha| \ll |\theta_0|$ is valid for almost the whole region of the magnetic field $0 \leqslant H \leqslant 0.9H_c$.

The corresponding classical action can be evaluated by integrating Eq. (1) with the above classical trajectories, and the result is

$$S_{cl} = 2\sqrt{\frac{K_2}{K_1}} S \sin\theta_0. \tag{15}$$

By applying the instanton technique,[10] we obtain the instanton's contribution to the ground-state tunnel splitting as

$$\hbar\Delta = \frac{4}{\pi^{1/2}}(VK_1)\left(\frac{K_2}{K_1}\right)^{3/4}(\sin\theta_0)^{3/2} S^{-1/2} e^{-S_{cl}}. \tag{16}$$

Here we have not included the additional phase factors generated by the topological Wess-Zumino term, but will reinstate them in the final expressions of the tunnel splittings.

With the help of the dilute instanton-gas approximation,[54] the transition amplitude between degenerate states is given by

$$\langle j'|e^{-HT}|j\rangle = \sqrt{\frac{\omega_0}{\pi\hbar}}\, e^{-\omega_0T/2} \sum_{m,n}^{m-n=j-j'(\mathrm{mod}2)} \times \frac{(\hbar\Delta T e^{-i\delta})^m(\hbar\Delta T e^{i\delta})^n}{m!n!}, \tag{17}$$

where $|j\rangle$ and $|j'\rangle$ denote the two degenerate states, and $\delta = S\pi(1-\cos\theta_0)$ is the phase factor generated by the topological Wess-Zumino term for FM particles with biaxial crystal symmetry in a magnetic field along the hard anisotropy axis. After simple algebra, the propagators from $|1\rangle$ to the other degenerate states are found to be

$$\langle 1|e^{-HT}|1\rangle = \sqrt{\frac{\omega_0}{\pi\hbar}}\, e^{-\omega_0T/2}\cosh\{2\hbar\Delta T\cos[S\pi(1-\cos\theta_0)]\},$$

$$\langle 2|e^{-HT}|1\rangle = \sqrt{\frac{\omega_0}{\pi\hbar}}\, e^{-\omega_0T/2}\sinh\{2\hbar\Delta T\cos[S\pi(1-\cos\theta_0)]\}. \tag{18}$$

At zero field, $\theta_0 = \pi/2$, $\langle 2|e^{-HT}|1\rangle$ ($\langle\theta = \pi/2, \phi = \pi|e^{-HT}|\theta = \pi/2, \phi = 0\rangle$) vanishes for half-integer spins, in accordance with the Kramers theorem. However, a magnetic field along the hard axis can lead to a nonzero propagator between the Kramers doublet even if the total spin is a half-integer. This is, however, not the only effect the topological phase interference results in. In fact, we shall present the strikingly different tunnel splittings for integer and half-integer spins with the help of the effective Hamiltonian approach.[55]

In Ref. 4, Leggett *et al.* showed that for a two-level system isolated from its environment, its motion in the two-dimensional Hilbert space can be completely described by a simple Hamiltonian $H = -(1/2)\hbar\Delta_0\sigma_x + (1/2)\varepsilon\sigma_z$, where σ_i are Pauli matrices, and the basis is chosen so that the eigenstate of σ_z with eigenvalue $+1$ (-1) corresponds to the system being localized in the right (left) well. ε is the difference in the ground-state energies of the states localized in the two wells in the absence of tunneling. For degenerate ground states, $\varepsilon = 0$. The quantity $(1/2)\hbar\Delta_0$ is the matrix element for tunneling between the wells. Now the effective Hamiltonian approach in magnetic MQC is a development of the tunneling Hamiltonian of Leggett *et al.*,[4] where the phase factors generated by the topological Wess-Zumino term are properly incorporated. By applying the similar method in Ref. 56, one can show that the effective Hamiltonian approach is equivalent to the dilute instanton-gas approximation.[54] However, this approach has the advantage of being very simple and direct, which permits one to discuss the degeneracies of low-lying tunneling levels in detail.

We introduce an effective Hamiltonian as

$$H_{\mathrm{eff}} = -\hbar\Delta M, \tag{19}$$

where M is a linear operator defined by

$$M|j\rangle = p|j+1\rangle + q|j-1\rangle. \tag{20}$$

Equation (20) can be viewed as a process whereby $|j\rangle$ goes forward to $|j+1\rangle$ with weight p and backward to $|j-1\rangle$ with weight q. For this case the matrix form of M is

$$[M] = \langle j'|M|j\rangle = \begin{bmatrix} 0 & p+q \\ p+q & 0 \end{bmatrix}, \tag{21}$$

where $p = q^* = \exp[-iS\pi(1-\cos\theta_0)]$. A simple diagonalization of H_{eff} shows that the eigenvalues are $E = \pm 2\hbar\Delta\cos[S\pi(1-\cos\theta_0)]$. Therefore, the splitting of ground state is $\hbar\Delta_1 = 4\hbar\Delta|\cos[S\pi(1-\cos\theta_0)]|$. When $H=0$, $\theta_0 = \pi/2$, and the tunnel splitting is $\hbar\Delta_1(H=0) = 4\hbar\Delta_0|\cos(S\pi)|$, where

$$\hbar\Delta_0 = \frac{4}{\pi^{1/2}}(VK_1)\left(\frac{K_2}{K_1}\right)^{3/4} S^{-1/2} e^{-S_{cl}(H=0)}, \tag{22}$$

with

$$S_{cl}(H=0) = 2\sqrt{\frac{K_2}{K_1}}S. \tag{23}$$

The topological quenching for half-integer spins at $H=0$ is rederived by use of the effective Hamiltonian approach, which agrees well with the Kramers theorem. Compared with Eqs. (15), (16) and Eqs. (22), (23), it is easy to show that the tunnel splitting increases with the external magnetic field because the field decreases the barrier between the two degenerate states. The expression of $\hbar\Delta_1$ shows that the tunnel splitting oscillates with the field for both integer and half-integer spins, and therefore $\hbar\Delta_1$ is suppressed to zero whenever

$$H/H_c = (S-n+1/2)/S, \tag{24}$$

where n is an integer.

At the end of this section, we discuss the possible relevance to the experimental test for spin-parity effects in single-domain FM nanoparticles. Since we have already obtained the low-lying tunneling level spectrum, the partition function is

$$Z = \mathrm{Tr}\, e^{-\beta H} = 2\cosh(E_0\beta), \tag{25}$$

where $E_0 = 2\hbar\Delta\cos[S\pi(1-\cos\theta_0)]$, $\hbar\Delta$ is shown in Eqs. (15) and (16), and $\beta = 1/k_BT$ with k_B the Boltzmann constant. The specific heat $c = -T(\partial^2 F/\partial T^2)$, where $F = -k_BT\ln Z$. Then we obtain the specific heat for FM particles with biaxial crystal symmetry in a magnetic field along the hard anisotropy axis as

$$c = \frac{E_0^2}{k_BT^2}\frac{1}{\cosh^2(E_0\beta)}. \tag{26}$$

When $H=0$,

$$c(H=0) = \frac{4\hbar^2\Delta_0^2}{k_BT^2}\frac{1}{\cosh^2(2\hbar\Delta_0\beta)}, \tag{27}$$

for integer spins, while $c(H=0)=0$ for half-integer spins. The magnetic susceptibility $\chi = -\partial^2 F/\partial H^2$, and at zero magnetic field we obtain that

$$\chi(H=0) = -\frac{2k_BT}{H_c^2}(S\pi)^2(\hbar\Delta_0\beta)\tanh(2\hbar\Delta_0\beta), \tag{28a}$$

for integer spins, while

$$\chi(H=0) = \frac{4k_BT}{H_c^2}(S\pi)^2(\hbar\Delta_0\beta)^2, \tag{28b}$$

for half-integer spins. It is clearly shown that the specific heat and the magnetic susceptibility for integer spins are much different from those for half-integer spins, providing a possible experimental method to test the topological phase interference effects.

IV. MQC FOR TRIGONAL CRYSTAL SYMMETRY

In this section, we consider a FM system with trigonal crystal symmetry, i.e., which has three consecutive energy minima in a period. The magnetic field is applied in $\hat{z}$, parallel to the hard anisotropy axis. Now the total energy is

$$\begin{aligned} E(\theta,\phi) &= K_1\cos^2\theta - K_2\sin^3\theta\cos(3\phi) - M_0H\cos\theta + E_0', \\ &= K_1(\cos\theta-\cos\theta_0)^2 - K_2\sin^3\theta\cos(3\phi) + E_0, \end{aligned} \tag{29}$$

where K_1 and K_2 are magnetic anisotropic constants satisfying $K_1 \gg K_2 > 0$. $\cos\theta_0 = H/H_c$, and $H_c = 2K_1/M_0$. As $K_1 \gg K_2 > 0$, the magnetization vector is forced to lie in the $\theta = \theta_0$ plane, and therefore the fluctuations of θ about θ_0 are small. Introducing $\theta = \theta_0 + \alpha(|\alpha| \ll 1)$, the total energy of Eq. (29) reduces to

$$E(\alpha,\phi) = K_1\sin^2\theta_0\alpha^2 + 2K_2\sin^3\theta_0\sin^2(3\phi/2). \tag{30}$$

The ground state corresponds to the magnetization vector pointing in one of the three degenerate easy directions: $\theta = \theta_0$, and $\phi = 0,\ 2\pi/3,\ 4\pi/3$. If we denote the three states as $|1\rangle$, $|2\rangle$, and $|3\rangle$, other energy minima repeat the three states with period 2π.

The classical equations of motion with Eq. (30) has the instanton solution

$$\begin{aligned} \bar{\alpha} &= -i\sqrt{2\frac{K_2}{K_1}}(\sin\theta_0)^{1/2}\frac{1}{\cosh(\omega_1\tau)}, \\ \sin\left(\frac{3}{2}\bar{\phi}\right) &= \frac{1}{\cosh(\omega_1\tau)}, \end{aligned} \tag{31}$$

where $\omega_1 = 3\sqrt{2}(V/\hbar S)\sqrt{K_1K_2}(\sin\theta_0)^{3/2}$. Equation (31) corresponds to the transition of $\vec{M}$ from $|1\rangle$ to $|2\rangle$. In deriving Eqs. (30) and (31), we have used the approximation that $|\alpha| \ll |\theta_0|$. It is easy to show that this approximation is valid for the weak magnetic field. For the strong magnetic field, i.e., $H \to H_c$, $\theta_0 = \sqrt{2\epsilon}$, where $\epsilon = 1 - H/H_c \ll 1$. From Eq. (31), $|\alpha| \approx \sqrt{K_2/K_1}\epsilon^{1/4} \approx 0.01$ for typical values of parameters for FM particles ($K_1 \approx 10^7$ erg/cm^3 and $K_2 \approx 10^5$ erg/cm^3). Therefore, $|\alpha| \ll |\theta_0|$ is valid for almost the whole region of the magnetic field $0 \le H \le 0.99H_c$. The associated classical action is

$$S_{cl} = \frac{2^{5/2}}{3}\sqrt{\frac{K_2}{K_1}}S(\sin\theta_0)^{3/2}, \tag{32}$$

and the instanton's contribution to the tunnel splitting is

$$\hbar\Delta = \frac{2^{9/4}\times 3^{1/2}}{\pi^{1/2}}(VK_1)\left(\frac{K_2}{K_1}\right)^{3/4}(\sin\theta_0)^{9/4}S^{-1/2}e^{-S_{cl}}. \tag{33}$$

At zero field, the splitting $\hbar\Delta_0=\hbar\Delta(H=0)$ agrees well with the result in Ref. 56.

For FM particles with trigonal crystal symmetry, the Euclidean transition amplitude between degenerate states is given by

$$\langle j'|e^{-HT}|j\rangle=\sqrt{\frac{\omega_1}{\pi\hbar}}e^{-\omega_1 T/2}\sum_{m,n}^{m-n=j-j'(\mathrm{mod}\ 3)}\times\frac{(\hbar\Delta T e^{-i\delta})^m(\hbar\Delta T e^{i\delta})^n}{m!n!},\qquad(34)$$

where $|j\rangle$ and $|j'\rangle$ are any two of the three degenerate states, and $\delta=(2/3)S\pi(1-\cos\theta_0)$ is the phase factor generated by the geometric Wess-Zumino term. After some algebra, we obtain the propagators from $|1\rangle$ to other states as

$$\langle 1|e^{-HT}|1\rangle=\frac{1}{3}\sqrt{\frac{\omega_1}{\pi\hbar}}e^{-\omega_1 T/2}[\exp(2\hbar\Delta T\cos\delta)+2\exp(-\hbar\Delta T\cos\delta)\cosh(\sqrt{3}\hbar\Delta T\sin\delta)],$$

$$\langle 2|e^{-HT}|1\rangle=\frac{1}{3}\sqrt{\frac{\omega_1}{\pi\hbar}}e^{-\omega_1 T/2}\{\exp(2\hbar\Delta T\cos\delta)-\exp(-\hbar\Delta T\cos\delta)[\cosh(\sqrt{3}\hbar\Delta T\sin\delta)+i\sqrt{3}\sinh(\sqrt{3}\hbar\Delta T\sin\delta)]\},$$

$$\langle 3|e^{-HT}|1\rangle=\frac{1}{3}\sqrt{\frac{\omega_1}{\pi\hbar}}e^{-\omega_1 T/2}\{\exp(2\hbar\Delta T\cos\delta)-\exp(-\hbar\Delta T\cos\delta)[\cosh(\sqrt{3}\hbar\Delta T\sin\delta)-i\sqrt{3}\sinh(\sqrt{3}\hbar\Delta T\sin\delta)]\}.\qquad(35)$$

For this case, the effective Hamiltonian is found to be

$$H_{\mathrm{eff}}=-\hbar\Delta\begin{bmatrix}0&q&p\\p&0&q\\q&p&0\end{bmatrix},\qquad(36)$$

where $p=q^*=\exp[-i(2/3)S\pi(1-\cos\theta_0)]$. A simple diagonalization of H_{eff} shows that the eigenvalues are

$$E_1=-2\hbar\Delta\cos\left[\frac{2}{3}S\pi(1-\cos\theta_0)\right],\qquad(37a)$$

$$E_2=2\hbar\Delta\cos\left\{\frac{1}{3}\pi[2S(1-\cos\theta_0)+1]\right\},\qquad(37b)$$

$$E_3=2\hbar\Delta\cos\left\{\frac{1}{3}\pi[2S(1-\cos\theta_0)-1]\right\}.\qquad(37c)$$

At zero field, $\theta_0=\pi/2$, and the eigenvalues are $\hbar\Delta_0$ and $-2\hbar\Delta_0$ for integer spins, the former being doubly degenerate. However, the eigenvalues are $2\hbar\Delta_0$ and $-\hbar\Delta_0$ for half-integer spins, the latter being doubly degenerate.

For FM particles with trigonal crystal symmetry in a magnetic field along the hard anisotropy axis, the partition function of low-lying tunneling levels is

$$Z=\mathrm{Tr}\,e^{-\beta H}=e^{-E_1\beta}+e^{-E_2\beta}+e^{-E_3\beta}.\qquad(38)$$

The specific heat at finite magnetic field is found to be

$$c=-\frac{1}{k_BT^2}\frac{1}{Z^2}(E_1e^{-E_1\beta}+E_2e^{-E_2\beta}+E_3e^{-E_3\beta})^2+\frac{1}{k_BT^2}\frac{1}{Z}(E_1^2e^{-E_1\beta}+E_2^2e^{-E_2\beta}+E_3^2e^{-E_3\beta}).\qquad(39)$$

At zero magnetic field, Eq. (39) reduces to

$$c(H=0)=\frac{18\hbar^2\Delta_0^2}{k_BT^2}\frac{e^{\hbar\Delta_0\beta}}{(e^{2\hbar\Delta_0\beta}+2e^{-\hbar\Delta_0\beta})^2},\qquad(40a)$$

for integer spins, while

$$c(H=0)=\frac{18\hbar^2\Delta_0^2}{k_BT^2}\frac{e^{-\hbar\Delta_0\beta}}{(e^{-2\hbar\Delta_0\beta}+2e^{\hbar\Delta_0\beta})^2},\qquad(40b)$$

for half-integer spins. The magnetic susceptibility at zero magnetic field is

$$\chi(H=0)\approx\frac{8k_BT}{3H_c^2}\frac{(\hbar\Delta_0\beta)^2}{Z_0}S^2\pi^2e^{-\hbar\Delta_0\beta}+\frac{8k_BT}{9H_c^2}\frac{(\hbar\Delta_0\beta)}{Z_0}S^2\pi^2(e^{2\hbar\Delta_0\beta}-e^{-\hbar\Delta_0\beta}),\qquad(41a)$$

for integer spins, while

$$\chi(H=0)\approx\frac{8k_BT}{3H_c^2}\frac{(\hbar\Delta_0\beta)^2}{Z_0}S^2\pi^2e^{\hbar\Delta_0\beta}+\frac{8k_BT}{9H_c^2}\frac{(\hbar\Delta_0\beta)}{Z_0}S^2\pi^2(e^{-2\hbar\Delta_0\beta}-e^{\hbar\Delta_0\beta}),\qquad(41b)$$

for half-integer spins, where $Z_0=2e^{-\hbar\Delta_0\beta}+e^{2\hbar\Delta_0\beta}$ for integer spins, and $Z_0=2e^{\hbar\Delta_0\beta}+e^{-2\hbar\Delta_0\beta}$ for half-integer spins.

V. MQC FOR TETRAGONAL CRYSTAL SYMMETRY

In this section, we will study the spin-parity effects in single-domain FM nanoparticles with tetragonal crystal symmetry in a magnetic field along the hard anisotropy axis, which has four consecutive minima in a period. Now the total energy is

$$\begin{aligned}E(\theta,\phi)&=K_1\cos^2\theta+K_2\sin^4\theta-K_2'\sin^4\theta\cos(4\phi)-M_0H\cos\theta+E_0'\\&=K_1(\cos\theta-\cos\theta_0)^2+K_2\sin^4\theta-K_2'\sin^4\theta\cos(4\phi)+E_0,\end{aligned}\qquad(42)$$

where K_1, K_2, and K_2' are magnetic anisotropic constants which satisfy that $K_1\gg K_2,K_2'>0$. In the case of very strong K_1, $\vec{M}$ is forced to lie in the $\theta=\theta_0$ plane, and thus the fluctuations of θ about θ_0 are small. Introducing $\theta=\theta_0+\alpha(|\alpha|\ll 1)$, Eq. (42) reduces to

$$E(\alpha,\phi)=K_1\sin^2\theta_0\alpha^2+2K_2'\sin^4\theta_0\sin^2(2\phi). \tag{43}$$

The easy directions are at $\theta=\theta_0$, and $\phi=0,\ \pi/2,\ \pi,\ 3\pi/2$. We denote the four states as $|1\rangle$, $|2\rangle$, $|3\rangle$, and $|4\rangle$, other energy minima repeat the four states with period 2π.

Substituting Eq. (43) into the classical equations of motion, we obtain the instanton solution mapping from $|1\rangle$ to $|2\rangle$ as

$$\bar{\alpha}=-i\sqrt{2\frac{K_2'}{K_1}}\sin\theta_0\frac{1}{\cosh(\omega_2\tau)},$$

$$\sin(2\bar{\phi})=\frac{1}{\cosh(\omega_2\tau)}, \tag{44}$$

where $\omega_2=2^{5/2}(V/\hbar S)\sqrt{K_1K_2'}\sin^2\theta_0$. Correspondingly, the classical action is

$$S_{cl}=2^{1/2}\sqrt{\frac{K_2'}{K_1}}S\sin^2\theta_0. \tag{45}$$

Based on the formulas in Ref. 10, we obtain the instanton's contribution to the ground-state tunnel splitting as

$$\hbar\Delta=\frac{2^{13/4}}{\pi^{1/2}}(VK_1)\left(\frac{K_2'}{K_1}\right)^{3/4}\sin^3\theta_0S^{-1/2}e^{-S_{cl}}. \tag{46}$$

By use of the dilute instanton-gas approximation, we obtain the Euclidean transition amplitude for FM particles with tetragonal crystal symmetry at finite magnetic field as

$$\langle j'|e^{-HT}|j\rangle=\sqrt{\frac{\omega_2}{\pi\hbar}}e^{-\omega_2T/2}\sum_{m,n}^{m-n=j-j'(\mathrm{mod}\ 4)}\times\frac{(\hbar\Delta Te^{-i\delta})^m(\hbar\Delta Te^{i\delta})^n}{m!n!}, \tag{47}$$

where $\delta=(1/2)S\pi(1-\cos\theta_0)$. The propagators from $|1\rangle$ to the other states are

$$\langle 1|e^{-HT}|1\rangle=\frac{1}{2}\sqrt{\frac{\omega_2}{\pi\hbar}}\,e^{-\omega_2T/2}[\cosh(2\hbar\Delta T\cos\delta)+\cosh(2\hbar\Delta T\sin\delta)], \tag{48a}$$

$$\langle 2|e^{-HT}|1\rangle=\frac{1}{2}\sqrt{\frac{\omega_2}{\pi\hbar}}\,e^{-\omega_2T/2}[\sinh(2\hbar\Delta T\cos\delta)-i\sinh(2\hbar\Delta T\sin\delta)], \tag{48b}$$

$$\langle 3|e^{-HT}|1\rangle=\frac{1}{2}\sqrt{\frac{\omega_2}{\pi\hbar}}\,e^{-\omega_2T/2}[\cosh(2\hbar\Delta T\cos\delta)-\cosh(2\hbar\Delta T\sin\delta)], \tag{48c}$$

$$\langle 4|e^{-HT}|1\rangle=\frac{1}{2}\sqrt{\frac{\omega_2}{\pi\hbar}}\,e^{-\omega_2T/2}[\sinh(2\hbar\Delta T\cos\delta)+i\sinh(2\hbar\Delta T\sin\delta)]. \tag{48d}$$

At zero magnetic field, $\theta_0=\pi/2$, it is shown that if the total spin is a half-integer, the propagators behave like those of two-level system, indicating the degeneracy of the tunneling levels. Moreover, $\langle 3|e^{-HT}|1\rangle$ (i.e., $\langle\theta=\pi/2,\phi=\pi|e^{-HT}|\theta=\pi/2,\phi=0\rangle$) is suppressed to zero for half-integer spins at zero field. This effect is in good agreement with the Kramers theorem, which demands a state with its time-reversed counterpart should be degenerate for half-integer spins if the system has time-reversal invariance.

For the present case, the effective Hamiltonian can be written as

$$H_{\mathrm{eff}}=-\hbar\Delta\begin{bmatrix}0&q&0&p\\p&0&q&0\\0&p&0&q\\q&0&p&0\end{bmatrix}, \tag{49}$$

where $p=q^*=\exp[-i(1/2)S\pi(1-\cos\theta_0)]$. Then the eigenvalues of this system are

$$E_{1,3}=\pm 2\hbar\Delta\cos\left[\frac{1}{2}S\pi(1-\cos\theta_0)\right],$$

$$E_{2,4}=\pm 2\hbar\Delta\sin\left[\frac{1}{2}S\pi(1-\cos\theta_0)\right]. \tag{50}$$

When $H=0$, $\theta=\pi/2$, it is easy to show that if S is a half-integer, the energies are $\pm\sqrt{2}\hbar\Delta_0$ with double degenerate, where

$$\hbar\Delta_0=\frac{2^{13/4}}{\pi^{1/2}}(VK_1)\left(\frac{K_2'}{K_1}\right)^{3/4}S^{-1/2}e^{-S_{cl}(H=0)}, \tag{51}$$

and

$$S_{cl}(H=0)=2^{1/2}\sqrt{\frac{K_2'}{K_1}}S. \tag{52}$$

But if S is an integer, the energies are $\pm\hbar\Delta_0$ and 0, the latter being doubly degenerate.

Now the partition function of low-lying tunneling levels is given by

$$Z=2[\cosh(E_1\beta)+\cosh(E_2\beta)], \tag{53}$$

where E_1 and E_2 are shown in Eq. (50) by taking "+" values. The specific heat at finite magnetic field is

$$c=\frac{4}{k_BT^2}\frac{1}{Z^2}\{(E_1^2+E_2^2)[1+\cosh(E_1\beta)\cosh(E_2\beta)]-2E_1E_2\sinh(E_1\beta)\sinh(E_2\beta)\}. \tag{54}$$

At zero magnetic field, Eq. (54) reduces to

$$c(H=0)=\frac{2}{k_BT^2}\frac{(\hbar\Delta_0)^2}{\cosh(\sqrt{2}\hbar\Delta_0\beta)}, \tag{55a}$$

for half-integer spins, while

$$c(H=0)=\frac{4}{k_B T^2}\frac{(\hbar\Delta_0)^2}{[1+\cosh(2\hbar\Delta_0\beta)]^2}, \qquad (55b)$$

for integer spins. The magnetic susceptibility at zero magnetic field is found to be

$$\chi\approx\frac{k_BT}{H_c^2}\frac{(\hbar\Delta_0\beta)^2}{[1+\cosh(2\hbar\Delta_0\beta)]}S^2\pi^2 - \frac{k_BT}{2H_c^2}\frac{(\hbar\Delta_0\beta)}{[1+\cosh(2\hbar\Delta_0\beta)]}\sinh(2\hbar\Delta_0\beta)S^2\pi^2, \qquad (56a)$$

for integer spins, while

$$\chi\approx\frac{k_BT}{2H_c^2}(\hbar\Delta_0\beta)^2S^2\pi^2 - \frac{k_BT}{2\sqrt{2}H_c^2}(\hbar\Delta_0\beta)\tanh(\sqrt{2}\hbar\Delta_0\beta)S^2\pi^2, \qquad (56b)$$

for half-integer spins. The tunneling level spectrum, the specific heat and the magnetic susceptibility for integer spins are much different from those for half-integer spins.

VI. MQC FOR HEXAGONAL CRYSTAL SYMMETRY

In this section, we will study the tunneling behaviors of FM particles with hexagonal crystal symmetry, which has six degenerate easy directions in a period. In the presence of a magnetic field along the hard anisotropy axis $\hat{z}$, the total energy is

$$\begin{aligned}E(\theta,\phi)&=K_1\cos^2\theta+K_2\sin^4\theta+K_3\sin^6\theta\\&\quad-K_3'\sin^6\theta\cos(6\phi)-M_0H\cos\theta+E_0'\\&=K_1(\cos\theta-\cos\theta_0)^2+K_2\sin^4\theta+K_3\sin^6\theta\\&\quad-K_3'\sin^6\theta\cos(6\phi)+E_0,\end{aligned} \qquad (57)$$

where K_1, K_2, K_3, and K_3' are magnetic anisotropic constants satisfying $K_1\gg K_2,K_3,K_3'>0$. The magnetization vector is forced to lie in the $\theta=\theta_0$ plane, so the fluctuations of θ about θ_0 are small. Introducing $\theta=\theta_0+\alpha(|\alpha|\ll 1)$, Eq. (57) reduces to

$$E(\alpha,\phi)=K_1\sin^2\theta_0\alpha^2+2K_3'\sin^6\theta_0\sin^2(3\phi). \qquad (58)$$

The easy directions are at $\theta=\theta_0$, and $\phi=0$, $\pi/3$, $2\pi/3$, π, $4\pi/3$, $5\pi/3$. We denote the six states as $|1\rangle$, $|2\rangle$, $|3\rangle$, $|4\rangle$, $|5\rangle$, and $|6\rangle$, other energy minima repeat the six states with period 2π. The classical equations of motion with Eq. (58) has the instanton solution

$$\bar{\alpha}=-i\sqrt{2\frac{K_3'}{K_1}}\sin^2\theta_0\frac{1}{\cosh(\omega_3\tau)},$$

$$\sin(3\bar{\phi})=\frac{1}{\cosh(\omega_3\tau)}, \qquad (59)$$

where $\omega_3=3\times 2^{3/2}(V/\hbar S)\sqrt{K_1K_3'}\sin^3\theta_0$. The associated classical action is

$$S_{cl}=\frac{2^{3/2}}{3}\sqrt{\frac{K_3'}{K_1}}S\sin^3\theta_0. \qquad (60)$$

According to the formulas in Ref. 10, we obtain the instanton's contribution to the tunnel splitting for FM particles with hexagonal crystal symmetry at finite field as

$$\hbar\Delta=\frac{3^{1/2}\times 2^{11/4}}{\pi^{1/2}}(VK_1)\left(\frac{K_3'}{K_1}\right)^{3/4}(\sin\theta_0)^{9/2}S^{-1/2}e^{-S_{cl}}. \qquad (61)$$

The splitting at zero field $\hbar\Delta_0=\hbar\Delta(H=0)$ is consistent with the result in Ref. 56.

The transition amplitude between degenerate states is given by

$$\langle j'|e^{-HT}|j\rangle=\sqrt{\frac{\omega_3}{\pi\hbar}}e^{-\omega_3T/2}\sum_{m,n}^{m-n=j-j'(\mathrm{mod}\ 6)}\times\frac{(\hbar\Delta Te^{-i\delta})^m(\hbar\Delta Te^{i\delta})^n}{m!n!}, \qquad (62)$$

where $\delta=(1/3)S\pi(1-\cos\theta_0)$. After some complicated calculations, the propagators from $|1\rangle$ to the other states are found to be

$$\langle 1|e^{-HT}|1\rangle=\frac{1}{3}\sqrt{\frac{\omega_3}{\pi\hbar}}e^{-\omega_3T/2}[\cosh(2\hbar\Delta T\cos\delta)+2\cosh(\hbar\Delta T\cos\delta)\cosh(\sqrt{3}\hbar\Delta T\sin\delta)], \qquad (63a)$$

$$\langle 2|e^{-HT}|1\rangle=\frac{1}{3}\sqrt{\frac{\omega_3}{\pi\hbar}}e^{-\omega_3T/2}[\sinh(2\hbar\Delta T\cos\delta)+\sinh(\hbar\Delta T\cos\delta)\cosh(\sqrt{3}\hbar\Delta T\sin\delta)-i\sqrt{3}\cosh(\hbar\Delta T\cos\delta)\sinh(\sqrt{3}\hbar\Delta T\sin\delta)], \qquad (63b)$$

$$\langle 3|e^{-HT}|1\rangle=\frac{1}{3}\sqrt{\frac{\omega_3}{\pi\hbar}}e^{-\omega_3T/2}[\cosh(2\hbar\Delta T\cos\delta)-\cosh(\hbar\Delta T\cos\delta)\cosh(\sqrt{3}\hbar\Delta T\sin\delta)-i\sqrt{3}\sinh(\hbar\Delta T\cos\delta)\sinh(\sqrt{3}\hbar\Delta T\sin\delta)], \qquad (63c)$$

$$\langle 4|e^{-HT}|1\rangle=\frac{1}{3}\sqrt{\frac{\omega_3}{\pi\hbar}}e^{-\omega_3T/2}[\sinh(2\hbar\Delta T\cos\delta)-2\sinh(\hbar\Delta T\cos\delta)\cosh\sqrt{3}\hbar\Delta T\sin\delta], \qquad (63d)$$

$$\langle 5|e^{-HT}|1\rangle=\frac{1}{3}\sqrt{\frac{\omega_3}{\pi\hbar}}\,e^{-\omega_3 T/2}[\cosh(2\hbar\Delta T\cos\delta)-\cosh(\hbar\Delta T\cos\delta)\cosh(\sqrt{3}\hbar\Delta T\sin\delta)+i\sqrt{3}\sinh(\hbar\Delta T\cos\delta)\sinh(\sqrt{3}\hbar\Delta T\sin\delta)], \tag{63e}$$

$$\langle 6|e^{-HT}|1\rangle=\frac{1}{3}\sqrt{\frac{\omega_3}{\pi\hbar}}\,e^{-\omega_3 T/2}[\sinh(2\hbar\Delta T\cos\delta)+\sinh(\hbar\Delta T\cos\delta)\cosh(\sqrt{3}\hbar\Delta T\sin\delta)+i\sqrt{3}\cosh(\hbar\Delta T\cos\delta)\sinh(\sqrt{3}\hbar\Delta T\sin\delta)]. \tag{63f}$$

When $H=0$, $\theta_0=\pi/2$, $\langle 4|e^{-HT}|1\rangle$ (i.e., $\langle\theta=\pi/2,\phi=\pi|e^{-HT}|\theta=\pi/2,\phi=0\rangle$) is suppressed to zero for half-integer spins due to the destructive interference of Wess-Zumino phase between the topologically distinct tunneling paths, which is in good agreement with the Kramers theorem. However, the field applied along the hard axis can lead to a nonzero transition amplitude even if the total spin is a half-integer.

Now the effective Hamiltonian can be written as

$$H_{\text{eff}}=-\hbar\Delta\begin{bmatrix}0&q&0&0&0&p\\p&0&q&0&0&0\\0&p&0&q&0&0\\0&0&p&0&q&0\\0&0&0&p&0&q\\q&0&0&0&p&0\end{bmatrix}, \tag{64}$$

where $p=q^*=\exp[-i(1/3)S\pi(1-\cos\theta_0)]$. Then the eigenvalues of the system are

$$E_{1,4}=\pm 2\hbar\Delta\cos\left[\frac{1}{3}S\pi(1-\cos\theta_0)\right],$$

$$E_{2,5}=\pm 2\hbar\Delta\cos\left\{\frac{1}{3}\pi[S(1-\cos\theta_0)+1]\right\},$$

$$E_{3,6}=\pm 2\hbar\Delta\cos\left\{\frac{1}{3}\pi[S(1-\cos\theta_0)-1]\right\}. \tag{65}$$

At zero magnetic field, $\theta_0=\pi/2$, the energies are $\sqrt{3}\hbar\Delta_0$, 0, and $-\sqrt{3}\hbar\Delta_0$ for half-integer spins, all the three levels being doubly degenerate. While the energies are $\pm 2\hbar\Delta_0$ and $\pm\hbar\Delta_0$ for integer spins, the latter two levels being doubly degenerate.

For this case, the partition function of low-lying tunneling levels is given by

$$Z=2[\cosh(E_1\beta)+\cosh(E_2\beta)+\cosh(E_3\beta)], \tag{66}$$

where E_1, E_2, and E_3 are shown in Eq. (65) by taking "+" values. The specific heat at finite field is

$$c=\frac{4}{k_BT^2}\frac{1}{Z^2}\{E_1^2+E_2^2+E_3^2+(E_1^2+E_2^2)\cosh(E_1\beta)\cosh(E_2\beta)+(E_1^2+E_3^2)\cosh(E_1\beta)\cosh(E_3\beta)+(E_2^2+E_3^2)\times\cosh(E_2\beta)\cosh(E_3\beta)-2E_1E_2\sinh(E_1\beta)\sinh(E_2\beta)-2E_1E_3\sinh(E_1\beta)\sinh(E_3\beta)-2E_2E_3\sinh(E_2\beta)\sinh(E_3\beta)\}. \tag{67}$$

At zero magnetic field, Eq. (67) reduces to

$$c(H=0)=\frac{2\hbar^2\Delta_0^2}{k_BT^2}\frac{[4+5\cosh(2\hbar\Delta_0\beta)\cosh(\hbar\Delta_0\beta)-4\sinh(2\hbar\Delta_0\beta)\sinh(\hbar\Delta_0\beta)]}{[\cosh(2\hbar\Delta_0\beta)+2\cosh(\hbar\Delta_0\beta)]^2}, \tag{68a}$$

for integer spins, while

$$c(H=0)=\frac{6\hbar^2\Delta_0^2}{k_BT^2}\frac{2+\cosh(\sqrt{3}\hbar\Delta_0\beta)}{[1+2\cosh(\sqrt{3}\hbar\Delta_0\beta)]^2}, \tag{68b}$$

for half-integer spins. The magnetic susceptibility at zero field is found to be

$$\chi(H=0)=\frac{2k_BT}{3H_c^2}\frac{(\hbar\Delta_0\beta)^2}{[2\cosh(\hbar\Delta_0\beta)+\cosh(2\hbar\Delta_0\beta)]^2}\times S^2\pi^2\cosh(\hbar\Delta_0\beta)-\frac{2k_BT}{9H_c^2}\times\frac{(\hbar\Delta_0\beta)}{[2\cosh(\hbar\Delta_0\beta)+\cosh(2\hbar\Delta_0\beta)]}\times S^2\pi^2[\sinh(\hbar\Delta_0\beta)+\sinh(2\hbar\Delta_0\beta)], \tag{69a}$$

for integer spins, while

$$\chi(H=0)=\frac{2k_BT}{9H_c^2}\frac{(\hbar\Delta_0\beta)^2}{[1+2\cosh(\sqrt{3}\hbar\Delta_0\beta)]^2}\times S^2\pi^2[2+\cosh(\sqrt{3}\hbar\Delta_0\beta)]-\frac{2\sqrt{3}k_BT}{9H_c^2}\frac{(\hbar\Delta_0\beta)}{[1+2\cosh(\sqrt{3}\hbar\Delta_0\beta)]^2}\times S^2\pi^2\sinh(\sqrt{3}\hbar\Delta_0\beta), \tag{69b}$$

for half-integer spins. It has been clearly shown that the specific heat and the magnetic susceptibility for integer spins are significantly different from those for half-integer spins, which provides a possible experimental test for spin-parity effects in FM particles.

VII. DISCUSSIONS AND CONCLUSIONS

In summary, we have investigated the spin-parity effects in resonant coherently quantum tunneling of the magnetization vector in single-domain FM nanoparticles with biaxial, trigonal, tetragonal, and hexagonal crystal symmetries in a magnetic field along the hard anisotropy axis. Both the WKB exponent and the preexponential factors are evaluated in the instanton's contribution to the ground-state tunnel splittings based on the instanton technique in the spin-coherent-state path integral. The Euclidean transition amplitudes between degenerate states are evaluated by use of the dilute instanton-gas approximation, which gives some direct results for the topological phase interference effects. The low-lying tunneling level spectrum is clearly shown by applying the effective Hamiltonian approach.

One important conclusion is that for all four kinds of crystal symmetries, the ground-state tunnel splittings for half-integer spins are significantly different from those for integer spins, resulting from the Berry phase interference between topologically distinct tunneling paths. For FM particles with simple biaxial crystal symmetry at zero magnetic field, the topological quenching for half-integer spins is rederived by use of the effective Hamiltonian approach. However, a magnetic field along the hard axis can lead to a finite tunnel splitting even if the total spin is a half-integer. The low-lying tunneling level spectrum for the trigonal, tetragonal, or hexagonal crystal symmetry is found to be much more complex than that for the biaxial crystal symmetry, and the tunnel splittings at $H=0$ can be nonzero even if the total spin is a half-integer. The transition amplitude from states along $\hat{x}$ to $-\hat{x}$ vanishes for half-integer spins in FM particles with biaxial, tetragonal, or hexagonal crystal symmetry at $H=0$, which is entirely equivalent to the Kramers theorem. Another important observation is that the tunnel splittings oscillate with the magnetic field for both integer and half-integer spins for each kind of crystal symmetry. Note that these spin-parity effects are of topological origin, and therefore are independent of the magnitude of total spins. The heat capacity and the magnetic susceptibility of low-lying tunneling states are evaluated and are found to depend significantly on the parity of total spins, providing a possible experimental method to examine the theoretical results on topological phase interference effects. Our results presented here should be useful for a quantitative understanding on the spin-parity effects in single-domain FM nanoparticles with different crystal symmetries.

The theoretical calculations performed in this paper can be extended to single-domain AFM nanoparticles, 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 results presented here will stimulate more experiments whose aim is observing the topological phase interference or spin-parity effects in nanometer-scale single-domain ferromagnets.

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