Spin-phase interference, coherent superposition, and quantum tunneling at excited levels in nano-antiferromagnets

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Abstract

The spin-phase interference effects are studied analytically in resonant quantum tunneling of the Néel vector between degenerate excited levels in nanometer-scale single-domain antiferromagnets in the absence of an external magnetic field. We consider a model for mesoscopic antiferromagnets with uncompensated excess spins for the more general structure of magnetic anisotropy, such as biaxial, trigonal, tetragonal and hexagonal crystal symmetry. This study provides a nontrivial generalization of the Kramers degeneracy for double-well system to coherently spin tunneling at ground states as well as low-lying excited states in AFM system with \( m \)-fold rotational symmetry around the \( \hat{z} \) axis. The energy level spectrum and the thermodynamic properties of magnetic tunneling states are found to depend significantly on the parity of the excess spins at sufficiently low temperatures. Possible relevance to experiments is also discussed.

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I. INTRODUCTION

Can quantum mechanics be used to describe the behavior of macroscopic objects? This question has fascinated physicists for more than 70 years until the 1980s when it was proposed by Leggett et al.\(^1\) that macroscopic objects could behave quantum mechanically, provided that the dissipative interactions with the environment were small enough. In recent years, macroscopic quantum phenomena have been observed in various systems—for example, quantum tunneling of the phase in a Josephson junction, permanent current in small conductor rings, Bose-Einstein condensation in atomic vapours, and C\(_{60}\) molecules. Recently, tunneling of the magnetization has been intensively studied theoretically and experimentally in nanoparticles and molecular clusters.\(^2\) Two kinds of phenomena can be envisaged. First, macroscopic quantum tunneling (MQT), where the magnetization tunnels through an barrier from one metastable state to a stable one. Second, macroscopic quantum coherence (MQC), where the magnetization oscillates back and forth between the degenerate states. This oscillation should show up in the frequency-dependent magnetic noise and susceptibility spectra.\(^2,3\) Besides its importance from a fundamental point of view, tunneling of the magnetization changes the properties of small magnets, with potential implications for the data-storage technology. It is also very important to the reliability of nanometer-scale magnetic units in memory devices and the designing of quantum computers.\(^4\)

One notable subject in spin quantum coherence is that the topological Wess-Zumino-Berry phase\(^5\) can lead to remarkable spin-parity effects. It was found that the tunnel splitting is suppressed to zero for half-integer total spins in biaxial ferromagnetic (FM) particles due to the destructive phase interference between topologically different tunneling paths.\(^6\) However, the phase interference is constructive for integer spins, and hence the splitting is nonzero.\(^6\) While spin-parity effects are sometimes be related to Kramers degeneracy, they typically go beyond the Kramers theorem in a rather unexpected way.\(^7\) The auxiliary particle method was proposed to study the model for a single large spin subject to the external and anisotropy fields, and to discuss the spin-parity effects.\(^8\) Similar effect was found in
antiferromagnetic (AFM) particles, where only the integer excess spins can tunnel but not the half-integer ones.\textsuperscript{9,10} However, these spin-parity effects are intrinsically absent in the phenomena of MQT and MQC in the Josephson-junction-based superconducting systems, which makes the tunneling phenomena in nanoscale magnets more important for understanding the foundations of quantum mechanics. Theoretical results showed that MQC in AFM particles should show up at higher temperatures and higher frequencies than in FM particles of similar size,\textsuperscript{9–12} which makes AFM particles more interesting for experimental purposes. Recently, topological phase interference effects were investigated extensively in FM and AFM particles in a magnetic field,\textsuperscript{7–9,13–17} and in the systems with different symmetries.\textsuperscript{18,19}

One recent experiment\textsuperscript{20} was performed to measure the tunnel splittings in molecules Fe\textsubscript{8}, and a clear oscillation of the splitting as a function of the field along the hard axis was observed, which is a direct evidence of the role of the topological spin phase (Berry phase) in the spin dynamics. Recent theoretical and experimental studies include the thermally activated resonant tunneling based on the exact diagonalization,\textsuperscript{21} the auxiliary particle method,\textsuperscript{8} the discrete WKB method and a nonperturbation calculation,\textsuperscript{22} the non-adiabatic Landau-Zener model,\textsuperscript{23} and the calculation based on exact spin-coordinate correspondence.\textsuperscript{24}

The importance of the topological interference term of the Berry phase for the problem of spin tunneling and the associated spin-parity effects have been elucidated in Refs. 6, 7 and 10. However, the theoretical studies on AFM systems\textsuperscript{9–12} have been focused on phase interference between two opposite winding ground-state tunneling paths in biaxial particles. The spin-phase interference between excited-level tunneling paths is not clearly shown for AFM particles. Moreover, the previous works on AFM spin tunneling\textsuperscript{9–12} have been confined to the system with biaxial symmetry, which has two energetically degenerate easy directions in the basal plane. The purpose of this paper is to study the quantum tunneling and spin-phase interference \textit{at excited states} for AFM particles in the absence of an external magnetic field. By calculating the nonvacuum instantons, we obtain the analytical results for tunnel splittings at excited levels. To compare theory with experiment, we consider the
AFM particles with the general structure of magnetocrystalline anisotropy, such as biaxial, trigonal, tetragonal, and hexagonal symmetry around \( \hat{z} \), which have two, three, four, and six energetically degenerate easy directions in the basal plane. For AFM particles with biaxial symmetry, the spin-phase interference effect can be studied by summing up the contributions of topologically different tunneling paths of clockwise and counterclockwise instantons.\(^{6,9}\) However, for the system with complex trigonal, tetragonal or hexagonal symmetry, this procedure is hard to evaluate. In this paper, the spin tunneling problem is mapped onto a particle moving problem in one-dimensional periodic potential \( U(\phi) \) by integrating out the momentum in the path integral, and the tunneling level spectrum of excited states is obtained by using the Bloch theorem. Our results show that the excited-level tunnel splittings depend significantly on the parity of the excess spins of AFM particles. The low-energy limits of the nonvacuum instanton and the tunnel splittings of excited levels agree well with the results of ground-state tunneling in biaxial AFM particles.\(^{6,9,10}\) The structure of tunneling level spectrum for the trigonal, tetragonal and hexagonal crystal symmetry is found to be much more complex than that for the biaxial crystal symmetry.\(^{10,12}\) The tunnel splitting for biaxial AFM particles is quenched to zero for half-integer excess spins due to the destructive interference of Berry phase.\(^{6,9}\) While the tunnel splitting can be nonzero even if the excess spin is a half-integer for the trigonal, tetragonal, or hexagonal symmetry at zero magnetic field.

This paper is structured in the following way. In Sec. II, we review briefly some basic ideas of quantum tunneling in AFM particles, and discuss the fundamentals concerning the computation of excited-level splittings in the double-well potential. In Secs. III we study the resonant quantum tunneling of Néel vector between degenerate excited states in AFM particles with the biaxial symmetry in detail, and present the results for trigonal, tetragonal and hexagonal symmetry in Sec. IV. The conclusions are presented in Sec. V.
II. PHYSICAL MODEL

The system of interest is a single-domain AFM particle of about 5~10 nm in radius at a temperature well below its anisotropy gap. According to the two-sublattice model, there is a strong exchange energy \( m_1 \cdot m_2 / \chi_\perp \) between two sublattices, where \( m_1 \) and \( m_2 \) are the magnetization vectors of the two sublattices with large, fixed and unequal magnitudes, and \( \chi_\perp \) is the transverse susceptibility. Under the assumption that the exchange energy between two sublattices is much larger than the magnetocrystalline anisotropy energy, the Euclidean action for the AFM particle (neglecting dissipation with the environment) is

\[
S_E[\theta(x, \tau), \phi(x, \tau)] = \frac{1}{\hbar} \int d\tau \int d^3x \left\{ \frac{m_1 + m_2}{\gamma} \left( \frac{d\phi}{d\tau} \right) + \frac{m}{\gamma} \left( \frac{d\phi}{d\tau} \right) \cos \theta + \frac{\chi_\perp}{2\gamma^2} \left( \frac{d\theta}{d\tau} \right)^2 + \left( \frac{d\phi}{d\tau} \right)^2 \sin^2 \theta \right\},
\]

where \( \gamma \) is the gyromagnetic ratio, \( \alpha \) is the exchange constant (which is also referred to as the stiffness constant, or the Bloch wall coefficient), and \( \tau = it \) is the imaginary-time variable. The \( E(\theta, \phi) \) term includes the magnetocrystalline anisotropy and the Zeeman energies. The polar coordinate \( \theta \) and the azimuthal coordinate \( \phi \), which are the angular components of \( m_1 \) in the spherical coordinate system, can determine the direction of the Néel vector.

As pointed out in Ref. 10, for a nanometer-scale single-domain AFM particle, the Néel vector may depend on the imaginary time but not on coordinates because the spatial derivatives in Eq. (1) are suppressed by the strong exchange interaction between two sublattices. So all the calculations performed in the present work are for the homogeneous Néel vector. Therefore, Eq. (1) reduces to

\[
S_E(\theta, \phi) = \frac{V}{\hbar} \int d\tau \left\{ \frac{m_1 + m_2}{\gamma} \left( \frac{d\phi}{d\tau} \right) + \frac{m}{\gamma} \left( \frac{d\phi}{d\tau} \right) \cos \theta + \frac{\chi_\perp}{2\gamma^2} \left( \frac{d\theta}{d\tau} \right)^2 + \left( \frac{d\phi}{d\tau} \right)^2 \sin^2 \theta \right\} + E(\theta, \phi),
\]

where \( V \) is the volume of the single-domain AFM nanoparticle. \( m = m_1 - m_2 = \hbar \gamma s / V \), where \( s \) is the excess spin due to the noncompensation of two sublattices. Note that the first
term in the Euclidean action is a total imaginary-time derivative. Its integral depends only on the initial and final states and hence has no effect on the classical equations of motion, but yields a boundary contribution to the Euclidean action. However, it was shown that this term, known as the topological phase term, is of central importance for the quantum interference effect and makes the tunneling behaviors of integer and half-integer excess spins strikingly different.\textsuperscript{9–12}

The Euclidean transition amplitude from an initial state $|\theta_i, \phi_i\rangle$ to a final state $|\theta_f, \phi_f\rangle$ can be expressed as the following imaginary-time path integral in the spin-coherent-state representation,

$$
\langle \theta_f, \phi_f | e^{-HT} | \theta_i, \phi_i \rangle = \int D\{\theta\} D\{\phi\} \exp[-S_E(\theta, \phi)],
$$

(3)

where the Euclidean action $S_E(\theta, \phi)$ has been defined in Eq. (2). In the semiclassical limit, the dominant contribution to the transition amplitude comes from finite action solutions of the classical equations of motion (instantons). According to the standard instanton technique, the tunneling rate $\Gamma$ for MQT or the tunnel splitting $\Delta$ for MQC is given by $\Gamma(\text{or } \Delta) = Ae^{-S_{cl}}$,\textsuperscript{26} where $S_{cl}$ is the WKB exponent or the classical action which minimizes the Euclidean action of Eq. (2). The preexponential factor $A$ originates from the quantum fluctuations about the classical path, which can be evaluated by expanding the Euclidean action to the second order in small fluctuations.\textsuperscript{26} It is noted that the above result is based on tunneling at the ground state, and the temperature dependence of the tunneling frequency (i.e., tunneling at excited states) is not taken into account. The instanton technique is suitable only for the evaluation of the tunneling rate at the vacuum level, since the usual (vacuum) instantons satisfy the vacuum boundary conditions. Different types of pseudoparticle configurations were developed which satisfy periodic boundary condition (i.e., periodic instantons or nonvacuum instantons).\textsuperscript{27}

For a particle moving in a double-well-like potential $U(x)$, the WKB method gives the tunnel splitting of $n$th excited states as$^{28}$

$$
\Delta E_n = \frac{\omega(E_n)}{\pi} \exp[-S(E_n)],
$$

(4)
with the imaginary-time action is

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

(5)

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

$$t (E_n) = \sqrt{2m} \int_{x_3(E_n)}^{x_4(E_n)} dx \sqrt{E_n - U (x)},$$

(6)

where $x_{3,4} (E_n)$ are the classical turning points for the particle oscillating inside $U (x)$.

The functional-integral and the WKB method showed that for the potentials parabolic near the bottom the result Eq. (4) should be multiplied by $\sqrt{\pi (2n+1)^{n+1/2}}$. 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. (4).

Recently, the crossover from quantum to classical behavior and the associated phase transition have been investigated extensively in nanospin systems.\cite{29,30–34}

III. MQC FOR BIAXIAL SYMMETRY

In this section, we consider an AFM system with biaxial symmetry, i.e., which has two degenerate easy directions in the biaxial plane. Now the magnetocrystalline anisotropy energy is

$$E (\theta, \phi) = K_1 \cos^2 \theta + K_2 \sin^2 \theta \sin^2 \phi + E_0,$$

(7)

where $K_1$ and $K_2$ are the transverse and longitudinal anisotropic constants satisfying $K_1 \gg K_2 > 0$, and $E_0$ is a constant which makes $E (\theta, \phi)$ zero at the initial state. As $K_1 \gg K_2 > 0$, the Néel vector is forced to lie in the $\theta = \pi/2$ plane, so the fluctuations of $\theta$ about $\pi/2$ are small. Introducing $\theta = \pi/2 + \alpha (|\alpha| \ll 1)$, Eq. (7) reduces to
\[ E(\alpha, \phi) \approx K_1\alpha^2 + K_2\sin^2 \phi. \] (8)

The ground state corresponds to the Néel vector pointing in one of the two degenerate easy directions: \( \theta = \pi/2 \), and \( \phi = 0, \pi \), other energy minima repeat the two states with period \( 2\pi \). Performing the Gaussian integration over \( \alpha \), we can map the spin system onto a particle moving problem in one-dimensional potential well. Now the transition amplitude becomes

\[
U_{fi} = \exp \left[ -iS_{tot}(\phi_f - \phi_i) \right] \int d\phi \exp \left( -S_E[\phi] \right),
\]

\[
= \exp \left[ -iS_{tot}(\phi_f - \phi_i) \right] \int d\phi \exp \left\{- \int d\tau \left[ \frac{1}{2}M \left( \frac{d\phi}{d\tau} \right)^2 + U(\phi) \right] \right\},
\] (9)

with \( S_{tot} = 2S - s \) being the total spins of two sublattices, \( M = \frac{V}{\pi} \left( \frac{1}{\gamma_1} + \frac{m^2}{2K_1\gamma_1} \right) = \frac{\hbar^2}{AV} \left[ 1 + \frac{1}{2} \left( \frac{V}{\gamma_1} \right) \left( \frac{2}{\gamma} \right)^2 \right] \) being the effective mass, and \( U(\phi) = (K_2V/\hbar)\sin^2 \phi \) being the effective potential. \( J \) is the exchange density between two sublattices, which is related to the transverse susceptibility \( \chi_\perp \) by taking the simple estimate as \( \chi_\perp \approx \hbar^2\gamma^2S^2/JV^2 \). \( S = m_1V/\hbar\gamma \) is the total spin in \( m_1 \) sublattice, and \( s = mV/\hbar\gamma \) is the excess spin due to the noncompensation of two sublattices. It is noted that the total derivative in Eq. (2), when integrated, gives an additional phase factor to the transition amplitude Eq. (9) which depends on the initial and final values of \( \phi \). For the biaxial symmetry, this phase factor in Eq. (9) is \( \exp(-i\pi S_{tot}) \). The potential \( U(\phi) \) is periodic with period \( \pi \), and there are two minima in the entire region \( 2\pi \). We may look at \( U(\phi) \) as a superlattice with lattice constant \( \pi \) and total length \( 2\pi \), and we can derive the energy spectrum by applying the Bloch theorem and the tight-binding approximation. The translational symmetry is ensured by the possibility of successive \( 2\pi \) extensions.

The periodic instanton configuration \( \phi_p \) which minimizes the Euclidean action in Eq. (9) satisfies the equation of motion

\[
\frac{1}{2}M \left( \frac{d\phi_p}{d\tau} \right)^2 - U(\phi_p) = -E,
\] (10)

where \( E > 0 \) is a constant of integration, which can be viewed as the classical energy of the pseudoparticle configuration. Then we obtain the kink-solution as
\[
\sin^2 \phi_p = 1 - k^2 \text{sn}^2 (\omega_1 \tau, k),
\]

where \( \text{sn}(\omega_1 \tau, k) \) is the Jacobian elliptic sine function of modulus \( k \),

\[
k^2 = \frac{n_1^2 - 1}{n_1^2},
\]

with \( \omega_1 = \sqrt{\frac{\mathcal{V}}{\hbar S}} \sqrt{\frac{K_2 J}{1+\frac{J}{K_1} \left( \frac{s}{S} \right)^2}} \), and \( n_1 = \sqrt{K_2 V / \hbar E} > 1 \). In the low energy limit, i.e., \( E \to 0, k \to 1, \text{sn}(u, 1) \to \tanh u \), we have

\[
\sin \phi_p = \frac{1}{\cosh (\omega_1 \tau)},
\]

which is exactly the vacuum instanton solution derived in Ref. 10.

The Euclidean action of the periodic instanton configuration Eq. (11) over the domain \((-\beta, \beta)\) is found to be

\[
S_p = \int_{-\beta}^\beta d\tau \left[ \frac{1}{2} M \left( \frac{d\phi_p}{d\tau} \right)^2 + V(\phi_p) \right] = W + 2E\beta,
\]

with

\[
W = 2^{3/2} S \sqrt{\frac{K_2}{J}} \left[ 1 + \frac{1}{2} \left( \frac{J}{K_1} \right) \left( \frac{s}{S} \right)^2 \right] \left[ E(k) - \left( 1 - k^2 \right) K(k) \right],
\]

where \( K(k) \) and \( E(k) \) are the complete elliptic integral of the first and second kind, respectively. Now we take the low energy limit where \( E \) is much less than the barrier height. In this case, \( k'^2 = 1 - k^2 = \hbar E / K_2 V \ll 1 \), so we can perform the expansions of \( K(k) \) and \( E(k) \) in Eq. (15) to include terms like \( k^2 \) and \( k^2 \ln (4/k') \),

\[
E(k) = 1 + \frac{1}{2} \left[ \ln \left( \frac{4}{k'} \right) - \frac{1}{2} \right] k'^2 + \cdots,
\]

\[
K(k) = \ln \left( \frac{4}{k'} \right) + \frac{1}{4} \left[ \ln \left( \frac{4}{k'} \right) - 1 \right] k'^2 + \cdots.
\]

With the help of small oscillator approximation for energy near the bottom of the potential well, \( E = E_{n}^{\text{bia}} = (n + 1/2) \omega_1 \), Eq. (15) is expanded as

\[
W = 2^{3/2} S \sqrt{\frac{K_2}{J}} \left[ 1 + \frac{1}{2} \left( \frac{J}{K_1} \right) \left( \frac{s}{S} \right)^2 \right] \left( n + \frac{1}{2} \right) - \left( n + \frac{1}{2} \right)
\]

\[
+ \left( n + \frac{1}{2} \right) \ln \left[ \frac{\left( n + \frac{1}{2} \right)}{2^{3/2}} \frac{1}{S \sqrt{\frac{K_2}{J}} \left[ 1 + \frac{1}{2} \left( \frac{J}{K_1} \right) \left( \frac{s}{S} \right)^2 \right]} \right].
\]
Then the general formula Eq. (4) gives the low-lying energy shift of $n$th excited states for AFM particles with biaxial crystal symmetry at zero magnetic field as

$$
\frac{\hbar}{2\pi n!} \sqrt{K_2 J V} \left( \left( \frac{K_2}{J} \left[ 1 + \frac{1}{2} \left( \frac{J}{K_1} \left( \frac{s}{S} \right)^2 \right) \right] \right) \right)^{n+1/2} \times \exp \left( -2^{3/2} S \left[ \frac{K_2}{J} \left[ 1 + \frac{1}{2} \left( \frac{J}{K_1} \left( \frac{s}{S} \right)^2 \right) \right] \right) \right). (18)
$$

When $n = 0$, the energy shift of the ground state is

$$
\frac{\hbar}{2^{5/4} \sqrt{\pi}} \sqrt{K_2 J V} \left( \left( \frac{K_2}{J} \left[ 1 + \frac{1}{2} \left( \frac{J}{K_1} \left( \frac{s}{S} \right)^2 \right) \right] \right) \right)^{1/4} \times \exp \left( -2^{3/2} S \left[ \frac{K_2}{J} \left[ 1 + \frac{1}{2} \left( \frac{J}{K_1} \left( \frac{s}{S} \right)^2 \right) \right] \right) \right). (19)
$$

Then Eq. (18) can be written as

$$
\frac{\hbar}{n!} \left( \frac{\hbar}{\Delta E_{0}^{\text{bia}}} \right), (20)
$$

where

$$
q = 2^{7/2} S \left[ \frac{K_2}{J} \left[ 1 + \frac{1}{2} \left( \frac{J}{K_1} \left( \frac{s}{S} \right)^2 \right) \right] \right]. (21)
$$

Now we discuss briefly the dissipation effect and the temperature dependence of the decay rate. It is noted that Eqs. (20) and (21) are obtained under the condition that the levels in the two wells are degenerate. In more general cases, the transition amplitude between two levels separated by the barrier or the decay rate should be sensitive to this resonance condition for the two levels. For a spin tunneling problem, it is important to consider the discrete level structure. It was quantitatively shown that the phenomenon of MQC depends crucially on the width of the excited levels in the right well. Including the effects of dissipation, the decay rate, in particular, is given by

$$
\Gamma_n = \frac{1}{2} \left( \Delta E_n \right)^2 \sum_{n'} \frac{\Omega_{nn'}}{(E_n - E_{n'})^2 + \Omega_{nn'}^2}, (22)
$$
where $\Delta \mathcal{E}_n$ is the level splitting, $n'$ are the levels in the other well and $\Omega_{nn'}$ is the sum of the linewidths of the $n$th and $n'$th levels caused by the coupling of the system to the environment. For the exact resonance conditions, the temperature dependence of the decay rate is

$$\Gamma(T) = \sum_n \frac{(\Delta \mathcal{E}_n)^2}{2\Omega_n} \exp\left(-\frac{\mathcal{E}_n}{\beta}\right), \quad (23)$$

where the level broadening $\Omega_n$ contains all the details of the coupling between the magnet and its environment. If the width caused by the coupling of the system to the environment is sufficiently large, the levels overlap, so that the problem is more or less equivalent to the tunneling into the structureless continuum.

In this case, the results obtained in this paper should be changed by including the dissipation. It is noted that the purpose of this paper is to study the coherently quantum tunneling and spin-phase interference at excited levels for AFM particles in the absence of a magnetic field at sufficiently low temperatures. Strong dissipation is hardly the case for magnetic systems, and thereby our results are expected to hold. It has been argued that the decay rate should oscillate on the applied magnetic field depending on the relative magnitude between the width and the level spacing. However, it is not clear, to our knowledge, what should be the effect of finite temperature in the problem of spin tunneling. The full analysis of spin tunneling onto the precession levels remains an open problem.

Now we consider the transition exponent which is usually addressed by experiments. Transitions between two states in a bistable system or escaping from a metastable state can occur either due to the quantum tunneling or via the classical thermal activation. In the limit of $T \to 0$, the transitions are purely quantum-mechanical and the rate goes as $\Gamma \sim \exp\left(-S_{cl}\right)$, with $S_{cl}$ being the classical action or the WKB exponent which is independent of temperature. As the temperature increases from zero, thermal effects enter in the quantum tunneling process. If the temperature is sufficiently high, the decay from a metastable state is determined by processes of thermal activation, and the transition rate follows the Arrhenius law, $\Gamma \sim \exp\left(-U/k_B T\right)$, with $k_B$ being the Boltzmann constant and $U$ being the
height of energy barrier between the two states. Because of the exponential dependence of the thermal rate on $T$, the temperature $T_c$ characterizing the crossover from quantum to thermal regime can be estimated as $k_B T_c = U/S_{cl}$. For the present case, one can estimate that

$$k_B T_c = \frac{1}{2\sqrt{2}} \left( \sqrt{K_2 JV} \right) S^{-1} \frac{1}{\sqrt{1 + \frac{1}{2} \left( \frac{J}{K_1} \right) \left( \frac{s}{S} \right)^2}}. $$

characterizing the crossover from quantum to classical regime. Typical values of parameters for single-domain AFM nanoparticles are: the sublattice spin $S = 5000$, the excess spin $s = 150$, the longitudinal anisotropy constant $K_2 \sim 10^5$ erg/cm$^3$, the transverse anisotropy constant $K_1 \sim 10^6$ erg/cm$^3$, the exchange energy density between two sublattices $J \sim 10^9$ erg/cm$^3$, and the radius of particle is about 5 nm. By taking these values, we obtain that $T_c \approx 704$ mK, which agrees well with the experimental result for the diluted samples of horse-spleen ferritin.

It is noted that $\hbar \Delta \mathcal{E}^{bia}_n$ is only the level shift induced by tunneling between degenerate excited states through a single barrier. The effective periodic potential $U(\phi) = U(\phi + n\pi)$ can be regarded as a one-dimensional superlattice with lattice constant $\pi$. The tunneling through one barrier leads to the level splitting which extends formally to an energy “band” by translation symmetry (the rotation symmetry in the present case, $U(\phi) = U(\phi + n\pi)$). The energy “band” structure of this problem is formally the same as that of a one-dimensional tight-binding model in solid state physics. Then the energy spectrum of low-lying excited levels can be determined by the Bloch theorem. It is easy to show that if $\mathcal{E}^{bia}_n$ are the degenerate eigenvalues of the system with infinitely high barrier, the energy level spectrum is given by the following formula with the help of tight-binding approximation,

$$E_n^{bia} = \mathcal{E}_n^{bia} - 2\Delta \mathcal{E}_n^{bia} \cos [(S_{tot} + \xi) \pi].$$

(24)

The Bloch wave vector $\xi$ can be assumed to take either of the two values 0 and 1 in the first Brillouin zone. It is noted that in Eq. (24) we have included the contribution of topological phase for AFM particles with biaxial crystal symmetry (i.e., $\pi S_{tot}$). One can easily show
that the low-lying tunneling level spectrum, which corresponds to the splittings of \( n \)th excited state due to the resonant quantum coherence of the Néel vector between energetically degenerate states, depends on the parity of \( S_{\text{tot}} \) (or the excess spin \( s \)) significantly.

At the end of this section, we discuss the possible relevance to the experimental test for spin-parity effects in single-domain AFM nanoparticles. One can easily show that the specific heat for integer excess spins is much different from that for half-integer excess spins at sufficiently low temperatures \( T \sim T_0 = \hbar \Delta \mathcal{E}_0^{\text{bia}} / k_B \). When the temperature is higher \( \hbar \Delta \mathcal{E}_0 < k_B T < \hbar \omega_1 \), the excited energy levels may give contribution to the partition function of tunneling states. Now the partition function without the dissipation is given by

\[
\mathcal{Z} \approx \mathcal{Z}_0 \left[ 1 + \frac{1}{2} \left( 1 - e^{-\beta \hbar \omega_1} \right) \left( \beta \hbar \Delta \mathcal{E}_0^{\text{bia}} \right)^2 I_0 \left( 2q e^{-\beta \hbar \omega_1/2} \right) \right],
\]

(25)

for both integer and half-integer excess spins. \( \mathcal{Z}_0 = 2e^{-\beta \hbar \omega_1/2} \left( 1 - e^{-\beta \hbar \omega_1} \right) \) is the partition function in the well calculated for \( k_B T \ll \Delta U \) over the low-lying oscillator like states with \( \mathcal{E}_n^{\text{bia}} = (n + 1/2) \omega_1 \). \( I_0 (x) = \sum_{n=0} x^{2n} / (n!)^2 \) is the modified Bessel function, and \( q \) is shown in Eq. (21). We define a characteristic temperature \( \bar{T} \) that is solution of equation \( q e^{-\hbar \omega_1/2k_B \bar{T}} = 1 \). Then we obtain the specific heat up to the order of \( (\beta \hbar \Delta \mathcal{E}_0)^2 \) as

\[
c = k_B \left( \beta \hbar \omega_1 \right)^2 \frac{e^{\beta \hbar \omega_1}}{(e^{\beta \hbar \omega_1} - 1)^2} + \frac{1}{2} k_B \left( \beta \hbar \Delta \mathcal{E}_0 \right)^2 \left\{ \left[ 2 \left( 1 - e^{-\beta \hbar \omega_1} \right) + 4 \left( \beta \hbar \omega_1 \right) e^{-\beta \hbar \omega_1} \right] e^{-\beta \hbar \omega_1/2} \right. \\
+ \left. \left( \beta \hbar \omega_1 \right)^2 e^{-\beta \hbar \omega_1} I_0 \left( 2q e^{-\beta \hbar \omega_1/2} \right) - q \left( \beta \hbar \omega_1 \right) \frac{1}{2} \left( 5 e^{-3 \beta \hbar \omega_1/2} - e^{-\beta \hbar \omega_1/2} \right) \right. \\
+ \left. 4 \left( e^{-\beta \hbar \omega_1/2} - e^{-3 \beta \hbar \omega_1/2} \right) \right\} I''_0 \left( 2q e^{-\beta \hbar \omega_1/2} \right) + q^2 \left( \beta \hbar \omega_1 \right)^2 \left( e^{-\beta \hbar \omega_1/2} - e^{-3 \beta \hbar \omega_1/2} \right) \\
\times I''_0 \left( 2q e^{-\beta \hbar \omega_1/2} \right),
\]

(26)

for both integer and half-integer excess spins, where \( I'_0 = -I_1 \), and \( I''_0 = I_2 - I_1 / x \). \( I_\nu (x) = \sum_{n=0} (-1)^n \left( x/2 \right)^{2n+\nu} / n! \Gamma (n + \nu + 1) \), where \( \Gamma \) is Gamma function. The results show that the spin-parity effect will be lost at high temperatures. The specific heat for integer excess spins is almost the same as that for half-integer excess spins.
IV. MQC FOR TRIGONAL, TETRAGONAL AND HEXAGONAL SYMMETRIES

In this section, we will apply the method in Sec. III to study resonant quantum tunneling of the Néel vector in AFM particles with trigonal, tetragonal and hexagonal crystal symmetry. For the trigonal symmetry,

\[ E(\theta, \phi) = K_1 \cos^2 \theta - K_2 \sin^3 \theta \cos(3\phi) + E_0, \]

(27)

where \( K_1 \gg K_2 > 0 \). The energy minima of this system are at \( \theta = \pi/2 \), and \( \phi = 0, 2\pi/3, 4\pi/3 \), and other energy minima repeat the three states with period \( 2\pi \). The spin tunneling problem can be mapped onto a problem of one-dimensional motion by integrating out the small fluctuations of \( \theta \) about \( \pi/2 \), and for this case \( U(\phi) = 2(K_2 V/\hbar) \sin^2(3\phi/2) \). Now \( U(\phi) \) is periodic with period \( 2\pi/3 \), and there are three minima in the entire region \( 2\pi \). The periodic instanton configuration with an energy \( E > 0 \) is \( \sin^2\left(\frac{3}{2}\phi_p\right) = 1 - k^2 \sin^2(\omega_2 \tau, k) \), where \( k = \sqrt{(n_1^2 - 1)/n_1^2} \), \( \omega_2 = 3V/\hbar S \sqrt{1 + \frac{3}{4} \left(\frac{n_1}{S}\right)^2} \), and \( n_1 = \sqrt{2K_2 V/\hbar E} > 1 \). The low energy limit of this periodic instanton configuration agrees well with the vacuum instanton solution obtained in Ref. 18. The associated classical action is \( S_p = W + 2E\beta \), with

\[ W = \frac{8}{3} S \sqrt{\frac{K_2}{J}} \left[ 1 + \frac{1}{2} \left(\frac{J}{K_1}\right) \left(\frac{s}{S}\right)^2 \right] \left[ E(k) - (1 - k^2) K(k) \right]. \]

(28)

The general formula Eq. (4) gives the low-lying energy shift of \( n \)th excited state as

\[ \hbar \Delta E_n^{tri} = \frac{3}{\sqrt{2\pi n!}} S \sqrt{\frac{K_2 J V}{1 + \frac{3}{4} \left(\frac{J}{K_1}\right)^2 \left(\frac{s}{S}\right)^2}} \left(\frac{32}{3} S \sqrt{\frac{K_2}{J}} \left[ 1 + \frac{1}{2} \left(\frac{J}{K_1}\right) \left(\frac{s}{S}\right)^2 \right] \right)^{n+1/2} \]

\[ \times \exp \left( -\frac{8}{3} S \sqrt{\frac{K_2}{J}} \left[ 1 + \frac{1}{2} \left(\frac{J}{K_1}\right) \left(\frac{s}{S}\right)^2 \right] \right). \]

(29)

The periodic potential \( U(\phi) \) can be viewed as a superlattice with lattice constant \( 2\pi/3 \) and total length \( 2\pi \), and the Bloch theorem then gives the energy level spectrum of \( n \)th excited state \( E_n^{tri} = (n + 1/2) \omega_2 \) as \( E_n^{tri} = E_n^{tri} - 2\Delta E_n^{tri} \cos \left[ (S_{tot} + \xi) 2\pi/3 \right] \), where \( \xi = -1, 0, 1 \) in the first Brillouin zone. It is easy to show that the low-lying energy level spectrum is \( \hbar E_n^{tri} - 2\hbar \Delta E_n^{tri} \), and \( \hbar E_n^{tri} + \hbar \Delta E_n^{tri} \) for integer excess spins, the latter being doubly degenerate.
While the level spectrum is \( \hbar \mathcal{E}_{n}^{\text{tri}} - \hbar \Delta \mathcal{E}_{n}^{\text{tri}} \), and \( \hbar \mathcal{E}_{n}^{\text{tri}} + 2 \hbar \Delta \mathcal{E}_{n}^{\text{tri}} \) for half-integer excess spins, the former being doubly degenerate.

For the tetragonal symmetry, the magnetocrystalline anisotropy energy is

\[
E(\theta, \phi) = K_1 \cos^2 \theta + K_2 \sin^4 \theta - K'_2 \sin^4 \theta \cos(4\phi) + E_0,
\]

where \( K_1 \gg K_2, K'_2 > 0 \). The energy minima are at \( \theta = \pi/2 \), and \( \phi = 0, \pi/2, \pi, 3\pi/2 \), and other energy minima repeat the four states with period \( 2\pi \). The problem can be mapped onto a problem of particle moving in one-dimensional potential \( U(\phi) = 2(K'_2 V/\hbar) \sin^2(2\phi) \) by integrating out the small fluctuations of \( \theta \) about \( \pi/2 \). Now \( U(\phi) \) is periodic with period \( \pi/2 \), and there are four minima in the entire region \( 2\pi \). The periodic instanton configuration with an energy \( E > 0 \) is \( \sin^2(2\phi_p) = 1 - k^2 \sin^2(\omega_3 \tau, k) \), where \( k = \sqrt{(n_1^2 - 1)/n_1^2}, \omega_3 = 4 \sqrt{\frac{V}{8S}} \sqrt{\frac{K'_2 J}{1 + \frac{1}{2} \left( \frac{J}{K_1} \right) \left( \frac{s}{S} \right)^2}}, \) and \( n_1 = \sqrt{2K'_2 V/\hbar E} > 1 \). The associated classical action is

\[
S_p = W + 2E\beta, \text{ with}
\]

\[
W = 2S \sqrt{\frac{K'_2}{J}} \left[ 1 + \frac{1}{2} \left( \frac{J}{K_1} \right) \left( \frac{s}{S} \right)^2 \right] \left[ E(k) - (1 - k^2) K(k) \right].
\]

The low-lying energy shift of \( n \)th excited state is

\[
\hbar \Delta \mathcal{E}_{n}^{\text{te}} = \frac{2^{3/2}}{\sqrt{\pi n!}} \sqrt{\frac{K'_2 JV}{S}} \sqrt{1 + \frac{1}{2} \left( \frac{J}{K_1} \right) \left( \frac{s}{S} \right)^2} \left( 8S \sqrt{\frac{K'_2}{J}} \left[ 1 + \frac{1}{2} \left( \frac{J}{K_1} \right) \left( \frac{s}{S} \right)^2 \right] \right)^{n+1/2} \times \exp \left( -2S \sqrt{\frac{K'_2}{J}} \left[ 1 + \frac{1}{2} \left( \frac{J}{K_1} \right) \left( \frac{s}{S} \right)^2 \right] \right).
\]

Now \( U(\phi) \) can be viewed as a superlattice with lattice constant \( \pi/2 \) and total length \( 2\pi \), and the Bloch theorem gives the energy level spectrum of \( n \)th excited state \( \mathcal{E}_{n}^{\text{te}} = (n + 1/2) \omega_3 \) as \( E_{n}^{\text{te}} = \mathcal{E}_{n}^{\text{te}} - 2\Delta \mathcal{E}_{n}^{\text{te}} \cos\left[ (S_{\text{tot}} + \xi) \pi/2 \right] \), where \( \xi = -1, 0, 1, 2 \) in the first Brillouin zone. Then the low-lying energy level spectrum is \( \hbar \mathcal{E}_{n}^{\text{te}} = 2\Delta \mathcal{E}_{n}^{\text{te}} \), and \( \hbar \mathcal{E}_{n}^{\text{te}} \) for integer excess spins, the latter being doubly degenerate. While the level spectrum is \( \hbar \mathcal{E}_{n}^{\text{te}} = \sqrt{2} \hbar \Delta \mathcal{E}_{n}^{\text{te}} \) with doubly degenerate for half-integer excess spins.

For the case of hexagonal symmetry,

\[
E(\theta, \phi) = K_1 \cos^2 \theta + K_2 \sin^4 \theta + K_3 \sin^6 \theta - K'_3 \sin^6 \theta \cos(6\phi) + E_0,
\]
where \( K_1 \gg K_2, K_3, K_3' > 0 \). The easy directions are at \( \theta = \pi/2 \), and \( \phi = 0, \pi/3, 2\pi/3, \pi, 4\pi/3, 5\pi/3 \), and other energy minima repeat the six states with period \( 2\pi \). For the present case, \( U(\phi) = 2(K_3'V/h)\sin^2(3\phi) \) is periodic with period \( \pi/3 \), and there are six minima in the entire region \( 2\pi \). The periodic instanton configuration at a given energy \( E > 0 \) is \( \sin^2(3\phi_0) = 1 - k^2\sin^2(\omega_4\tau, k) \), where \( k = \sqrt{(n_1^2 - 1)/n_1^2} \), \( \omega_4 = 6V\bar{h}\sqrt{1 + \frac{K_3'}{K_1 \left( \frac{J}{K_1} \right)^2}} \), and \( n_1 = \sqrt{2K_3'V/hE} > 1 \). Correspondingly, the classical action is \( S_p = W + 2E\beta \), with

\[
W = \frac{4}{3}S\sqrt{\frac{K_3'}{J} \left[ 1 + \frac{1}{2} \left( \frac{J}{K_1} \right) \left( \frac{s}{S} \right)^2 \right]} \left[ E(k) - (1 - k^2) K(k) \right],
\]

and the low-lying energy shift of \( n \)th excited state is

\[
\hbar \Delta \mathcal{E}_n^{he} = \frac{3 \times 2^{1/2}}{\sqrt{\pi n!}} \frac{\sqrt{K_3'JV}}{S\sqrt{1 + \frac{1}{2} \left( \frac{J}{K_1} \right)^2}} \left( \frac{16}{3} \frac{K_3'}{J} \frac{1}{\sqrt{1 + \frac{1}{2} \left( \frac{J}{K_1} \right)^2}} \left[ 1 + \frac{1}{2} \left( \frac{J}{K_1} \right) \left( \frac{s}{S} \right)^2 \right] \right)^{n+1/2} \times \exp \left( -\frac{4}{3} \frac{K_3'}{J} \left[ 1 + \frac{1}{2} \left( \frac{J}{K_1} \right) \left( \frac{s}{S} \right)^2 \right] \right).
\]

Now \( U(\phi) \) can be regarded as a one-dimensional superlattice with lattice constant \( \pi/3 \). By applying the Bloch theorem and the tight-binding approximation, we obtain the energy level spectrum of \( n \)th excited state \( \mathcal{E}_n^{he} = (n + 1/2) \omega_4 \) as \( E_n^{he} = \mathcal{E}_n^{he} - 2\Delta \mathcal{E}_n^{he} \cos [(S_{tot} + \xi) \pi/3] \), where \( \xi = -2, -1, 0, 1, 2, 3 \). If the excess spin \( s \) is an integer, the low-lying energy level spectrum is \( \hbar \mathcal{E}_n^{he} \pm 2\hbar \Delta \mathcal{E}_n^{he} \), and \( \hbar \mathcal{E}_n^{he} \pm \hbar \Delta \mathcal{E}_n^{he} \), the latter two levels being doubly degenerate. If \( s \) is a half-integer, the level spectrum is \( \hbar \mathcal{E}_n^{he} \pm \sqrt{3}\hbar \Delta \mathcal{E}_n^{he} \), and \( \hbar \mathcal{E}_n^{he} \), all three levels being doubly degenerate.

In brief, the low-lying energy level spectrum for trigonal, tetragonal and hexagonal symmetry are found to depend on the parity of the excess spins of AFM particles distinctly, resulting from the phase interference between topologically different tunneling paths. The structure of low-lying tunneling level spectrum for the trigonal, tetragonal, or hexagonal symmetry is found to be much more complex than that for the biaxial symmetry. The low-lying energy level spectrum can be nonzero even if the excess spin is a half-integer for the trigonal, tetragonal, or hexagonal symmetry. The results of AFM nanoparticles with general structure of magnetocrystalline anisotropy will be helpful for experimental test.
V. CONCLUSIONS

In summary, we have studied the topological phase interference effects in the model for mesoscopic AFM particles with uncompensated excess spin for the more general structure of magnetic anisotropy, such as biaxial, trigonal, tetragonal, and hexagonal crystal symmetries. The low-lying tunnel splittings between \( n \)th degenerate excited states of neighboring wells are evaluated with the help of the periodic instanton method, and the energy level spectrum is obtained by applying the Bloch theorem and the tight-binding approximation in one-dimensional periodic potential. This is the first complete study, to our knowledge, of spin-phase interference between excited-level tunneling paths in AFM particles with general structure of magnetocrystalline anisotropy, which will be useful for experimental check.

One important conclusion is that for all the four kinds of crystal symmetries, the low-lying energy level spectrum for integer excess spins is significantly different from that for half-integer excess spins, resulting from the phase interference between topologically distinct tunneling paths. For AFM particles with simple biaxial symmetry, which has two degenerate easy directions in the basal plane (i.e., the double-well system), the tunnel splitting is suppressed to zero for half-integer excess spins due to the destructive phase interference between topologically different tunneling paths connecting the same initial and final states. However, the structure of low-lying tunneling level spectrum for the trigonal, tetragonal, or hexagonal symmetry is found to be much more complex than that for the biaxial symmetry. The low-lying energy level spectrum can be nonzero even if the excess spin is a half-integer for the trigonal, tetragonal, or hexagonal symmetry. Our analytical study provides a nontrivial generalization of Kramers degeneracy for double-well system to coherently spin tunneling at ground states as well as low-lying excited states for AFM systems with \( m \)-fold rotational symmetry around the \( \hat{z} \) axis. Note that these spin-parity effects are of topological origin, and therefore are independent of the magnitude of excess spins of AFM particles, the shape of the soliton and the tunneling potential. One can easily show that the heat capacity of low-lying magnetic tunneling states depends significantly on the parity of
excess spins for AFM particles with different symmetries at sufficiently low temperatures, 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 topological phase interference or spin-parity effects in resonant quantum tunneling in single-domain AFM particles with different symmetries.

Over the past years a lot of experimental and theoretical works were performed on the spin tunneling in molecular Mn$_{12}$-Ac$_{37}$ and Fe$_{838}$ clusters having a collective spin state $S = 10$ (in this paper $S = 10^3$-$10^4$). More recently, Wernsdorfer and Sessoli have measured the tunnel splittings in the molecular Fe$_8$ clusters, and have found a clear oscillation of the tunnel splitting with the field along hard axis, which is a direct evidence of the role of the Berry phase in the spin dynamics of these molecules. Further experiments should focus on the level quantization of collective spin states of $S = 10^2$-$10^4$ and their quantum spin phases. The theoretical calculations performed in this paper can be extended to the FM and AFM particles in a magnetic field. Similar spin-phase interference effects observed in Fe$_8$ cluster should be found in single-domain AFM nanoparticles in a magnetic field. Work along this line is still in progress. With current technology and fast progress on this field, our study on spin-phase interference and resonant quantum coherence effects in AFM nanoparticles should be experimentally testable in the near future.

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