Angular-dependent spin tunneling in mesoscopic biaxial antiferromagnets

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(Received 6 May 2002; revised manuscript received 25 November 2002; published 26 March 2003)

An imaginary-time path integral study is presented for spin tunneling in the biaxial antiferromagnetic grain with noncompensated sublattices placed in a magnetic field with an arbitrary direction in the plane of easy and medium axes. Different structures of the tunneling barriers can be generated by the magnitude and the orientation of the magnetic field. By calculating the nonvacuum instantons or bounces, we analytically obtain the dependence of decay rates from excited levels as well as ground-state levels and the temperature of the crossover from the thermal to quantum regime on the direction and strength of the field in a wide range of angles of the applied magnetic field. It is found that the WKB exponent and the crossover temperature strongly depend on the orientation of the field, which can be tested with the use of existing experimental techniques. In the large noncompensation limit, our results reduce to spin tunneling in ferromagnetic particles.

DOI: 10.1103/PhysRevB.67.104425 PACS number(s): 75.45.+j, 75.10.Jm, 75.50.Ee

I. INTRODUCTION

Quantum tunneling at mesoscopic or macroscopic scale is one of the most fascinating phenomena in condensed matter physics. During the last decade, the problem of quantum tunneling of magnetization in nanometer-scale magnets has attracted a great deal of theoretical and experimental interest.[1,2] In addition to the importance of the tunneling phenomena in magnets from a fundamental point of view, they are potentially important for the future magnetic devices working at a nanoscale and the designing of quantum computer.[3]

In discussing macroscopic quantum phenomena, it is essential to distinguish between two types of processes: macroscopic quantum coherence (MQC, i.e., coherent tunneling) and macroscopic quantum tunneling (MQT, i.e., incoherent tunneling). In the case of MQC, the system performs coherent NH_3-type oscillations between two degenerate wells separated by a classically impenetrable barrier. The tunneling removes the degeneracy of the original ground states and leads to a level splitting. For the case of MQT, the system escapes from a metastable potential well into a continuum by quantum tunneling, and the tunneling results in an imaginary part of the energy. As emphasized by Leggett, the two phenomena of MQC and MQT are physically very different, particularly from the viewpoint of experimental feasibility.[4] MQC is a far more delicate phenomenon than MQT, as it is much more easily destroyed by an environment,[5] or by very small *c*-number symmetry breaking fields that spoil the degeneracy. Even though some of the dissipative coupling are an unsuspected effect on the quantum tunneling depending on the situation, it has been reported that they are not strong enough to make the phenomena of MQT and MQC unobservable.

More recently, much attention was attracted to the spin tunneling in the single-domain ferromagnetic (FM) nanoparticles in the presence of a magnetic field applied at an arbitrary angle. The MQT problem for a uniaxial FM model was first studied by Zaslavskii with the help of mapping the spin system onto a one-dimensional particle system.[6] For the same model, Miguel and Chudnovsky[7] calculated the tunneling rate by applying the imaginary-time path integral, and demonstrated that the angular and field dependences of the tunneling exponent obtained by Zaslavskii's method and by the path-integral method coincide precisely. Kim and Hwang performed a calculation based on the instanton technique for biaxial and tetragonal FM particles.[8] Kim extended the tunneling rate for biaxial FM particles to a finite temperature, and presented the numerical results for the WKB exponent below the crossover temperature and their approximate formulas around the crossover temperature.[9] The quantum-classical transition of the escape rate for uniaxial FM particles in an arbitrarily directed field was studied by Garanin, Hidalgo and Chudnovsky by mapping onto a particle moving in a double-well potential.[10] Wernsdorfer *et al.* made the switching field measurement on single-domain FM nanoparticles of Barium ferrite (BaFeCoTiO) containing about 10^5–10^6 spins.[11] The measured angular dependance of the crossover temperature was found to be in excellent agreement with the theoretical prediction,[7] which strongly suggests the magnetic quantum tunneling in the BaFeCoTiO nanoparticles.

The phenomenon of quantum tunneling was also found in nanaometer-scale antiferromagnetic (AFM) particles, where the Néel vector is the tunneling entity.[12–19] It was shown that quantum tunneling shall show up at higher temperatures and higher frequencies in AFM particles than in FM particles of similar size. Moreover, most FM systems are actually ferrimagnetic or AFM particles. All these make nanometer-scale antiferromagnets more interesting from experimental and theoretical aspects. Recently, the temperature dependence of spin tunneling has been studied in the biaxial AFM particles.[20]

Up to now theoretical studies on AFM systems have been focused on quantum tunneling at ground-state level. Moreover, most previous works[12–19] have been confined to the condition that the magnetic field be applied along the easy, medium or hard anisotropy axis, separately. However, the generic quantum tunneling problem, and the easiest to implement in practice, is that of AFM particle in a magnetic field applied at some arbitrary angle θ_H to the anisotropy axis. The problem does not possess any symmetry and for that reason is more difficult mathematically. However, it is worth pursuing because of its significance for experiments. The purpose of this paper is to present an theoretical investigation of the quantum tunneling at excited levels in the biaxial AFM particles with an arbitrarily directed field, based on the spin-coherent-state path-integral method. We will show that MQC and MQT can be consecutively observed by changing the direction of magnetic field, and discuss their dependence on the direction and the magnitude of field. Both the nonvacuum (or thermal) instanton and bounce solutions, the WKB exponents and the preexponential factors are evaluated exactly for different angle ranges of the magnetic field [$\theta_H = \pi/2$, $\pi/2 + O(\sqrt{\epsilon}) < \theta_H < \pi - O(\sqrt{\epsilon})$, and $\theta_H = \pi$]. Both variables are expressed as a function of parameters which can be changed experimentally, such as the number of total spins, the effective anisotropy and the exchange interaction constants, and the strength and orientation of applied magnetic field. Our results show that the distinct angular dependence, together with the dependence of the WKB tunneling rate on the strength of the external magnetic field, may provide an independent experimental test for the spin tunneling at excited levels in nanoscale antiferromagnets. The dependence of the crossover temperature T_c and the magnetic viscosity (which is the inverse of WKB exponent at the quantum-tunneling-dominated regime $T \ll T_c$) on the direction and the magnitude of the field, and the magnetic anisotropies is expected to be observed in future experiments on individual single-domain AFM particles with an arbitrarily directed magnetic field. Furthermore, since the model considered here is a general AFM model with noncompensated sublattices, we can easily obtain the results of spin tunneling in FM particles by taking a relatively large noncompensation of sublattices.

This paper is structured in the following way. In Sec. II, we introduce the general formulation for quantum tunneling in AFM particles based on the two-sublattice model and spin-coherent-state path-integral method. And we discuss the fundamentals concerning the computation of level splittings and tunneling rates of excited states in a double-well-like potential. In Secs. III, we study the spin tunneling at excited levels for biaxial AFM particles in the presence of a magnetic field applied in the ZX plane with a range of angles $\pi/2 \leqslant \theta_H \leqslant \pi$. The conclusions and discussions are presented in Sec. V.

II. PHYSICAL MODEL

The system of interest is an nanometer-scale single-domain AFM particle at a temperature well below its anisotropy gap. According to the two-sublattice model,[12,13] there is a strong exchange energy $\mathbf{m}_1 \cdot \mathbf{m}_2 / \chi_\perp$ between two sublattices, where $\mathbf{m}_1$ and $\mathbf{m}_2$ 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 expressed in the spin-coherent-state representation[12–15,17,18]

$$\begin{aligned}\mathcal{S}_E[\theta(\mathbf{x},\tau),\phi(\mathbf{x},\tau)] &= \frac{1}{\hbar}\int d\tau \int d^3x \Bigg\{ i\frac{m_1+m_2}{\gamma}\left(\frac{d\phi}{d\tau}\right) + \frac{m}{\gamma}\left(\frac{d\phi}{d\tau}\right)\cos\theta \\ &\quad + \frac{\chi_\perp}{2\gamma^2}\left[\left(\frac{d\theta}{d\tau}\right)^2 + \left(\frac{d\phi}{d\tau}\right)^2 \sin^2\theta\right] \\ &\quad + \frac{1}{2}\alpha[(\nabla\theta)^2 + (\nabla\phi)^2\sin^2\theta] + E(\theta,\phi)\Bigg\}, \end{aligned} \tag{1}$$

where γ is the gyromagnetic ratio, α is the exchange constant (which is also referred to as the stiffness constant, or the Bloch wall coefficient[21]), and $\tau = it$ is the imaginary-time variable. $m = m_1 - m_2 = \hbar\gamma s/V$, where s is the excess spin due to the noncompensation of two sublattices. The $E(\theta,\phi)$ term includes the magnetocrystalline anisotropy and the Zeeman energies. The polar coordinate θ and the azimuthal coordinate ϕ in the spherical coordinate system with $\mathbf{l}\cdot\hat{z} = \cos\theta$, $\mathbf{l}$ is the Néel vector of unit length and $\hat{z}$ is a unit vector along the $\mathbf{z}$ axis.

As pointed out in Ref. 13, 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

$$\begin{aligned}\mathcal{S}_E(\theta,\phi) &= \frac{V}{\hbar}\int d\tau \Bigg\{ i\frac{m_1+m_2}{\gamma}\left(\frac{d\phi}{d\tau}\right) + \frac{m}{\gamma}\left(\frac{d\phi}{d\tau}\right)\cos\theta \\ &\quad + \frac{\chi_\perp}{2\gamma^2}\left[\left(\frac{d\theta}{d\tau}\right)^2 + \left(\frac{d\phi}{d\tau}\right)^2\sin^2\theta\right] + E(\theta,\phi)\Bigg\}, \end{aligned} \tag{2}$$

where V is the volume of the single-domain AFM nanoparticle. The first term in Eq. (2) is a total imaginary-time derivative, which has no effect on the classical equations of motion, but it is crucial for the spin-phase-interference effects.[5,13,14,17,18,22–25] However, for the closed instanton trajectory described in this paper (as shown in the following), this time derivative gives a zero contribution to the path integral, and therefore can be omitted.

The 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

$$\mathcal{K}_E = \langle \theta_f,\phi_f | e^{-\mathcal{H}T} | \theta_i,\phi_i\rangle = \int \mathcal{D}\{\theta\}\mathcal{D}\{\phi\}\exp[-\mathcal{S}_E(\theta,\phi)], \tag{3}$$

where the Euclidean action $\mathcal{S}_E(\theta,\phi)$ 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 Γ for

MQT or the tunnel splitting Δ for MQC is given by Γ (or $\Delta) = Ae^{-\mathcal{S}_{\rm cl}}$,[26] where $\mathcal{S}_{\rm cl}$ 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.[26,19] 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).[27]

For a particle moving in a double-well-like potential $U(x)$, the WKB method gives the tunnel splitting of degenerate excited levels or the imaginary parts of the metastable levels at an energy $E>0$ as[10,28,29]

$$\Delta E(\text{or Im}\, E) = \frac{\omega(E)}{2\pi}\exp[-\mathcal{S}(E)], \tag{4}$$

with the imaginary-time action

$$\mathcal{S}(E) = \sqrt{2m}\int_{x_1(E)}^{x_2(E)} dx\, \sqrt{U(x)-E}, \tag{5}$$

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

$$t(E) = \sqrt{2m}\int_{x_3(E)}^{x_4(E)} \frac{dx}{\sqrt{E-U(x)}}, \tag{6}$$

where $x_{3,4}(E)$ are the classical turning points for the particle oscillating inside $U(x)$. The functional-integral in the one-loop approximation, the correct WKB method, and the method of Schödinger equation showed that for the potentials parabolic near the bottom the result Eq. (4) should be multiplied by $\sqrt{\pi/e}[(2n+1)^{n+1/2}/2^n e^n n!]$.[27,29,30] The splitting of excited state for a generic double-well potential was obtained in Ref. 31 by using the Rayleigh-Schrödinger perturbation expansion of the eignfunctions, which agrees well with the result based on the one-loop path-integral. This correction 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) for the spin tunneling problem considered in this work.

III. MQC AND MQT IN BIAXIAL AFM PARTICLES

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

$$E(\theta,\phi) = K_1\sin^2\theta + K_2\sin^2\theta\sin^2\phi - mH_x\sin\theta\cos\phi - mH_z\cos\theta + E_0, \tag{7}$$

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

By introducing the dimensionless parameters as

$$\bar{K}_2 = K_2/2K_1, \bar{H}_x = H_x/H_0, \bar{H}_z = H_z/H_0, \tag{8}$$

the $E(\theta,\phi)$ term of Eq. (7) can be rewritten as

$$\bar{E}(\theta,\phi) = \frac{1}{2}\sin^2\theta + \bar{K}_2\sin^2\theta\sin^2\phi - \bar{H}_x\sin\theta\cos\phi - \bar{H}_z\cos\theta + \bar{E}_0, \tag{9}$$

where $E(\theta,\phi) = 2K_1\bar{E}(\theta,\phi)$ and $H_0 = 2K_1/m$. At finite magnetic field, the plane given by $\phi=0$ is the easy plane, on which $\bar{E}(\theta,\phi)$ reduces to

$$\bar{E}(\theta,\phi=0) = \frac{1}{2}\sin^2\theta - \bar{H}\cos(\theta-\theta_H) + \bar{E}_0. \tag{10}$$

We denote θ_0 to be the initial angle and θ_c the critical angle at which the energy barrier vanishes when the external magnetic field is close to the critical value $\bar{H}_c(\theta_H)$ (to be calculated in the following). Then, θ_0 satisfies $[d\bar{E}(\theta,\phi=0)/d\theta]_{\theta=\theta_0}=0$, θ_c and $\bar{H}_c$ satisfy both $[d\bar{E}(\theta,\phi=0)/d\theta]_{\theta=\theta_c,\bar{H}=\bar{H}_c}=0$ and $[d^2\bar{E}(\theta,\phi=0)/d\theta^2]_{\theta=\theta_c,\bar{H}=\bar{H}_c}=0$, which leads to

$$\frac{1}{2}\sin(2\theta_0) + \bar{H}\sin(\theta_0-\theta_H) = 0, \tag{11a}$$

$$\frac{1}{2}\sin(2\theta_c) + \bar{H}_c\sin(\theta_c-\theta_H) = 0, \tag{11b}$$

$$\cos(2\theta_c) + \bar{H}_c\cos(\theta_c-\theta_H) = 0. \tag{11c}$$

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

$$\bar{H}_c = [(\sin\theta_H)^{2/3} + |\cos\theta_H|^{2/3}]^{-3/2}, \tag{12a}$$

$$\sin(2\theta_c) = \frac{2|\cot\theta_H|^{1/3}}{1+|\cot\theta_H|^{2/3}}. \tag{12b}$$

Now we consider the limiting case that the external magnetic field is slightly lower than the critical field, i.e., $\epsilon = 1 - \bar{H}/\bar{H}_c \ll 1$. At this practically interesting situation, the barrier height is low and the width is narrow, and therefore the tunneling rate in MQT or the tunnel splitting in MQC is large. Introducing $\eta\equiv\theta_c-\theta_0$ ($|\eta|\ll 1$ in the limit of $\epsilon\ll 1$), expanding $[d\bar{E}(\theta,\phi=0)/d\theta]_{\theta=\theta_0}=0$ about θ_c, and using the relations $[d\bar{E}(\theta,\phi=0)/d\theta]_{\theta=\theta_c,\bar{H}=\bar{H}_c}=0$ and $[d^2\bar{E}(\theta,\phi=0)/d\theta^2]_{\theta=\theta_c,\bar{H}=\bar{H}_c}=0$, Eq. (11a) becomes

$$\sin(2\theta_c)\left(\epsilon-\frac{3}{2}\eta^2\right)-\eta\cos(2\theta_c)(2\epsilon-\eta^2)=0. \quad (13)$$

Simple calculations show that η is of the order of $\sqrt{\epsilon}$. Thus the order of magnitude of the second term in Eq. (13) is smaller than that of the first term by $\sqrt{\epsilon}$ and the value of η is determined by the first term, which leads to $\eta\simeq\sqrt{2\epsilon/3}$. However, when θ_H is very closed to $\pi/2$ or π, $\sin(2\theta_c)$ becomes almost zero, and the first term is much smaller than the second term in Eq. (13). Then η is determined by the second term when $\theta_H\simeq\pi/2$ or π, which leads to $\eta\simeq\sqrt{2\epsilon}$ for $\theta_H\simeq\pi/2$ and $\eta\simeq 0$ when $\theta_H\simeq\pi$. Since the first term in Eq. (13) is dorminant in the range of values, θ_c, which satisfies $\tan(2\theta_c)>O(\sqrt{\epsilon})$, $\eta\simeq\sqrt{2\epsilon/3}$ is valid for $\pi/2+O(\sqrt{\epsilon})<\theta_H<\pi-O(\sqrt{\epsilon})$ by using Eq. (12b). Therefore, $\eta\simeq\sqrt{2\epsilon}$, 0, and $\sqrt{2\epsilon/3}$ for $\theta_H\simeq\pi/2$, π, and $\pi/2+O(\sqrt{\epsilon})<\theta_H<\pi-O(\sqrt{\epsilon})$, respectively. In this situation the potential energy $\bar{E}(\theta,\phi)$ reduces to the following equation in the limit of small ϵ:

$$\bar{E}(\delta,\phi)=\bar{K}_2\sin^2\phi\sin^2(\theta_0+\delta)+\bar{H}_x\sin(\theta_0+\delta)(1-\cos\phi)+\bar{E}_1(\delta), \quad (14)$$

where $\delta\equiv\theta-\theta_0$ ($|\delta|\ll 1$ in the limit of $\epsilon\ll 1$), and $\bar{E}_1(\delta)$ is a function of only δ given by

$$\bar{E}_1(\delta)=\frac{1}{4}\sin(2\theta_c)(3\delta^2\eta-\delta^3)+\frac{1}{2}\cos(2\theta_c)\left[\delta^2\left(\epsilon-\frac{3}{2}\eta^2\right)+\delta^3\eta-\frac{1}{4}\delta^4\right]. \quad (15)$$

In the following, we will study the quantum tunneling at excited levels in biaxial AFM particles at three different angle ranges of the external magnetic field as $\theta_H=\pi/2$, $\pi/2+O(\sqrt{\epsilon})<\theta_H<\pi-O(\sqrt{\epsilon})$, and $\theta_H=\pi$, respectively.

A. $\theta_H=\pi/2$

For $\theta_H=\pi/2$, we have $\theta_c=\pi/2$ from Eq. (12b) and $\eta=\sqrt{2\epsilon}$ from Eq. (13). Equations (14) and (15) show that ϕ is very small for the full range of angles $\pi/2\leqslant\theta_H\leqslant\pi$ for AFM particles with biaxial crystal symmetry. Performing the Gaussian integration over ϕ, we can map the spin system onto a particle moving problem in the one-dimensional potential well. Now the imaginary-time transition amplitude Eq. (3) becomes

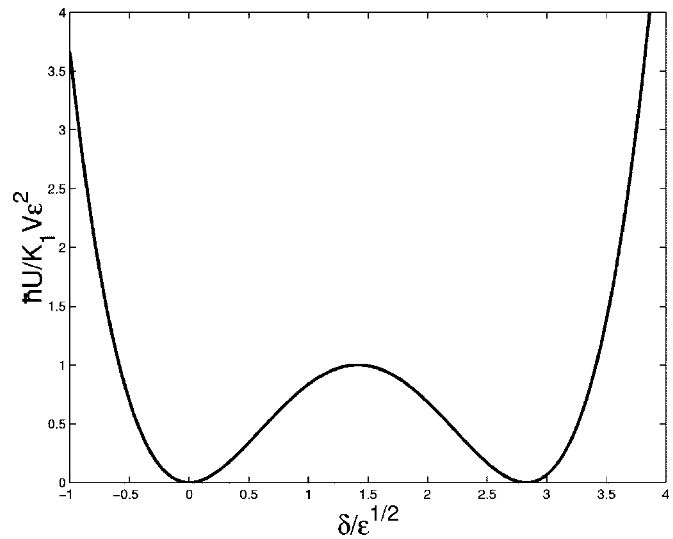

FIG. 1. The $\delta(=\theta-\theta_0)$ dependence of the effective potential $\hbar U/K_1V\epsilon^2$ for $\theta_H=\pi/2$ (the phenomenon of MQC).

$$\mathcal{K}_E=\int d\delta\exp(-\mathcal{S}_E[\delta])=\int d\delta\exp\left\{-\int d\tau\left[\frac{1}{2}\mathcal{M}\left(\frac{d\delta}{d\tau}\right)^2+U(\delta)\right]\right\}, \quad (16)$$

with the effective mass

$$\mathcal{M}=\mathcal{M}_{\mathrm{AFM}}+\mathcal{M}_{\mathrm{FM}},$$

$$\mathcal{M}_{\mathrm{AFM}}=\frac{\chi_\perp V}{\hbar\gamma^2},\quad \mathcal{M}_{\mathrm{FM}}=\frac{m^2V}{2\hbar\gamma^2[K_2+K_1(1-\epsilon)]},$$

and the effective potential

$$U(\delta)=\frac{K_1V}{4\hbar}\delta^2(\delta-2\sqrt{2\epsilon})^2. \quad (17)$$

The problem is one of MQC (as shown in Fig. 1), where the Néel vector resonates coherently between the energetically degenerate easy directions at $\delta=0$ and $\delta=2\sqrt{2\epsilon}$ separated by a classically impenetrable barrier at $\delta=\sqrt{2\epsilon}$.

The nonvacuum (or thermal) instanton configuration δ_p which minimizes the Euclidean action in Eq. (16) satisfies the equation of motion

$$\frac{1}{2}\mathcal{M}\left(\frac{d\delta_p}{d\tau}\right)^2-U(\delta_p)=-\mathcal{E}, \quad (18)$$

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

$$\delta_p=\sqrt{2\epsilon}+\sqrt{2\epsilon-\alpha}\,\mathrm{sn}(\omega_1\tau,k_1), \quad (19a)$$

where $\alpha=2\sqrt{\hbar\mathcal{E}/K_1V}$ and

$$\omega_1=\gamma\sqrt{\frac{K_1}{\chi_\perp}\left(\epsilon+\frac{\alpha}{2}\right)}\left[1+\frac{m^2}{2\chi_\perp[K_2+K_1(1-\epsilon)]}\right]^{-1/2}. \quad (19b)$$

$\mathrm{sn}(\omega_1\tau,k)$ is the Jacobian elliptic sine function of modulus $k_1=\sqrt{(2\epsilon-\alpha)/(2\epsilon+\alpha)}$. The Euclidean action of the nonvacuum instanton configuration Eq. (19) over the domain $(-\beta,\beta)$ is found to be

$$\mathcal{S}_p=\int_{-\beta}^{\beta}d\tau\left[\frac{1}{2}\mathcal{M}\left(\frac{d\delta_p}{d\tau}\right)^2+U(\delta_p)\right]=\mathcal{W}+2\mathcal{E}\beta, \quad (20a)$$

with

$$\mathcal{W}=\frac{2}{3}\epsilon^{3/2}\frac{\sqrt{K_1\chi_\perp}V}{\hbar\gamma}\left[1+\frac{m^2}{2\chi_\perp[K_2+K_1(1-\epsilon)]}\right]^{1/2}\times\frac{1}{\sqrt{1-k_1'^2/2}}\left[E(k_1)-\frac{k_1'^2}{2-k_1'^2}K(k_1)\right], \quad (20b)$$

where $k_1'^2=1-k_1^2$. $K(k_1)$ and $E(k_1)$ are the complete elliptic integral of the first and second kind, respectively. The general formula Eq. (4) gives the tunnel splittings of excited levels as

$$\Delta\mathcal{E}=\gamma\sqrt{\frac{K_1}{\chi_\perp}\left(\epsilon+\frac{\alpha}{2}\right)}\left[1+\frac{m^2}{2\chi_\perp[K_2+K_1(1-\epsilon)]}\right]^{-1/2}\times\frac{1}{2K(k_1')}\exp(-\mathcal{W}), \quad (21)$$

where $\mathcal{W}$ is shown in Eq. (20b).

Now we discuss the low energy limit where $\mathcal{E}$ is much less than the barrier height. In this case, $k_1'^4=4\hbar\mathcal{E}/K_1V\epsilon^2\ll 1$, so we can perform the expansions of $K(k_1)$ and $E(k_1)$ in Eq. (20b) to include terms such as $k_1'^4$ and $k_1'^4\ln(4/k_1')$,

$$E(k_1)=1+\frac{1}{2}\left[\ln\left(\frac{4}{k_1'}\right)-\frac{1}{2}\right]k_1'^2+\frac{3}{16}\left[\ln\left(\frac{4}{k_1'}\right)-\frac{13}{12}\right]k_1'^4+\cdots,$$

$$K(k_1)=\ln\left(\frac{4}{k_1'}\right)+\frac{1}{4}\left[\ln\left(\frac{4}{k_1'}\right)-1\right]k_1'^2+\frac{9}{64}\left[\ln\left(\frac{4}{k_1'}\right)-\frac{7}{6}\right]k_1'^4+\cdots. \quad (22)$$

With the help of small oscillator approximation for energy near the bottom of the potential well $\mathcal{E}_n=(n+1/2)\Omega_1$

$$\Omega_1=\sqrt{U''(\delta=\sqrt{2\epsilon})/\mathcal{M}}=2\gamma\sqrt{\frac{K_1\epsilon}{\chi_\perp}}\left[1+\frac{m^2}{2\chi_\perp[K_2+K_1(1-\epsilon)]}\right]^{-1/2},$$

Eq. (20b) is expanded as

$$\mathcal{W}=\mathcal{W}_0-\left(n+\frac{1}{2}\right)+\left(n+\frac{1}{2}\right)\ln\left[\frac{\hbar\gamma}{32\epsilon^{3/2}\sqrt{K_1\chi_\perp}V}\times\left[1+\frac{m^2}{2\chi_\perp[K_2+K_1(1-\epsilon)]}\right]^{-1/2}\left(n+\frac{1}{2}\right)\right], \quad (23a)$$

where

$$\mathcal{W}_0=\frac{8}{3}\frac{\sqrt{K_1\chi_\perp}V}{\hbar\gamma}\epsilon^{3/2}\left[1+\frac{m^2}{2\chi_\perp[K_2+K_1(1-\epsilon)]}\right]^{1/2}. \quad (23b)$$

Then the low-lying energy shift of nth excited states for biaxial AFM particles in the presence of a magnetic field applied perpendicular to the anisotropy axis ($\theta_H=\pi/2$) as

$$\hbar\Delta\mathcal{E}_n=\frac{32\sqrt{2}}{n!\sqrt{\pi}}(K_1V)\epsilon^2\left(\frac{32\epsilon^{3/2}\sqrt{K_1\chi_\perp}V}{\hbar\gamma}\times\left[1+\frac{m^2}{2\chi_\perp[K_2+K_1(1-\epsilon)]}\right]^{1/2}\right)^{n-1/2}\exp(-\mathcal{W}_0). \quad (24)$$

Comparing with the formula (290) of Ref. 31 by taking the effective mass and effective potential shown in Eq. (17), Eqs. (23b) and (24) give the same result for the WKB exponent, while the preexponential factor differs by a factor of 2^{2n}. Since the level splitting Eq. (24) is derived with the low-energy expansion up to the lowest nonzero order of Eq. (22), one can expert agreement only for the ground-state level. At high energies the formula (21) applies. It is noted that the purpose of this paper is to study the AFM spin tunneling as a function of strength and orientation of the applied magnetic field, which is largely determined by the exponential term of the tunnel splitting, this correction preexponential factor does not change much of the whole physics of spin quantum tunneling. And in most physical applications, this prefactor is best estimated as an ''attempt frequency.''

For the case of large noncompensation ($m\gg\sqrt{\chi_\perp K_1}$), Eq. (24) reduces to the result for quantum tunneling in biaxial FM particles

$$\hbar\Delta\mathcal{E}_n^{\mathrm{FM}}=\frac{(q_1^{\mathrm{FM}})^n}{n!}\hbar\Delta\mathcal{E}_0^{\mathrm{FM}}, \quad (25a)$$

where

$$q_1^{\mathrm{FM}}=16\sqrt{2}\epsilon^{3/2}\sqrt{\frac{K_1}{K_2+K_1(1-\epsilon)}}S', \quad (25b)$$

and S' is the total spin of FM particles. $\hbar\Delta\mathcal{E}_0^{\mathrm{FM}}$ is the tunnel splitting of ground-state level for FM particles

$$\hbar\Delta\mathcal{E}_0^{\mathrm{FM}}=\frac{2^{13/4}}{\sqrt{\pi}}(K_1V)\epsilon^{5/4}\sqrt{1-\epsilon+K_2/K_1}\times S'^{-1/2}\exp(-\mathcal{W}_0^{\mathrm{FM}}), \quad (26a)$$

where

$$\mathcal{W}_0^{\rm FM}=\frac{4\sqrt{2}}{3}\frac{1}{\sqrt{1-\epsilon+K_2/K_1}}S'\epsilon^{3/2}. \tag{26b}$$

The WKB exponent in the tunnel splitting, $\mathcal{W}_0^{\rm FM}$ for $\theta_H=\pi/2$, fully agrees with the result of ground-state tunneling in biaxial FM particles.[8] It is noted that we have extended the results in Ref. 8 to spin tunneling at ground state as well as low-lying excited state in a general AFM system with uncompensated moments in an arbitrarily directed magnetic field.

For the case of small noncompensation ($m\ll\sqrt{\chi_\perp K_1}$), Eq. (24) reduces to the result of AFM spin tunneling

$$\hbar\Delta\mathcal{E}_n^{\rm AFM}=\frac{(q_1^{\rm AFM})^n}{n!}\hbar\Delta\mathcal{E}_0^{\rm AFM}, \tag{27a}$$

where

$$q_1^{\rm AFM}=32S\epsilon^{3/2}\sqrt{\frac{K_1}{J}}. \tag{27b}$$

Note that $\chi_\perp=m_1^2/J$ and $m_1=\hbar\gamma S/V$, where J is the exchange interaction between two sublattices and S is the sublattice spin.[12–14] $\hbar\Delta\mathcal{E}_0^{\rm AFM}$ is the ground-state tunnel splitting of AFM particles

$$\hbar\Delta\mathcal{E}_0^{\rm AFM}=\frac{8}{\sqrt{\pi}}(JV)\left(\frac{K_1}{J}\right)^{3/4}\epsilon^{5/4}S^{-1/2}\exp(-\mathcal{W}_0^{\rm AFM}), \tag{28a}$$

where

$$\mathcal{W}_0^{\rm AFM}=\frac{8}{3}\sqrt{\frac{K_1}{J}}S\epsilon^{3/2}. \tag{28b}$$

B. $\pi/2+O(\sqrt{\epsilon})<\theta_H<\pi-O(\sqrt{\epsilon})$

For $\pi/2+O(\sqrt{\epsilon})<\theta_H<\pi-O(\sqrt{\epsilon})$, the critical angle θ_c is in the range of $O(\sqrt{\epsilon})<\theta_c<\pi/2-O(\sqrt{\epsilon})$, and $\eta\approx\sqrt{2\epsilon/3}$. Now the problem can be mapped onto a problem of one-dimensional motion by integrating out ϕ, and for this case the effective mass $\mathcal{M}$ and the effective potential $U(\delta)$ in the Euclidean action of Eq. (16) are found to be

$$\mathcal{M}=\mathcal{M}_{\rm AFM}+\mathcal{M}_{\rm FM}=\frac{\chi_\perp V}{\hbar\gamma^2}\left(1+\frac{m^2}{2K_1A\chi_\perp}\right) \tag{29a}$$

and

$$U(\delta)=\frac{K_1V}{2\hbar}\sin 2\theta_c(\sqrt{6\epsilon}\delta^2-\delta^3)=3U_0q^2\left(1-\frac{2}{3}q\right), \tag{29b}$$

where $q=3\delta/2\sqrt{6\epsilon}$, and $U_0=(2^{5/2}/3^{3/2})(K_1V\epsilon^{3/2}/\hbar)\times\sin 2\theta_c$. The prefactor A in Eq. (29a) is

$$A=\frac{1-\epsilon}{1+|\cot\theta_H|^{2/3}}+\frac{K_2}{K_1}.$$

The problem becomes one of MQT (as shown in Fig. 2),

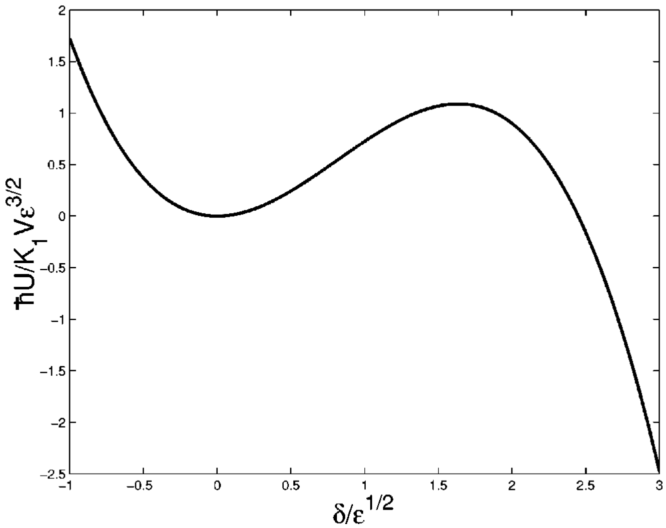

FIG. 2. The $\delta(=\theta-\theta_0)$ dependence of the effective potential $\hbar U/K_1V\epsilon^{3/2}$ for $\theta_H=3\pi/4$ (the phenomenon of MQT).

where the Néel vector escapes from the metastable state at $\delta=0$, $\phi=0$ through the barrier by quantum tunneling.

Now the nonvacuum bounce configuration with an energy $\mathcal{E}>0$ is found to be

$$\delta_p=\frac{2}{3}\sqrt{6\epsilon}[a-(a-b)\mathrm{sn}^2(\omega_2\tau,k_2)], \tag{30a}$$

where

$$\omega_2=\frac{1}{2^{1/4}\times 3^{1/4}}\gamma\sqrt{\frac{K_1}{\chi_\perp}}\left(1+\frac{m^2}{2K_1A\chi_\perp}\right)^{-1/2}\times\sqrt{\sin 2\theta_c}\epsilon^{1/4}\sqrt{a-c}. \tag{30b}$$

$a(\mathcal{E})>b(\mathcal{E})>c(\mathcal{E})$ denote three roots of the cubic equation

$$q^3-\frac{3}{2}q^2+\frac{\mathcal{E}}{2U_0}=0. \tag{31}$$

$\mathrm{sn}(\omega_2\tau,k_2)$ is the Jacobian elliptic sine function of modulus $k_2=\sqrt{(a-b)/(a-c)}$. The classical action of the nonvacuum bounce configuration Eq. (30a) is

$$\mathcal{S}_p=\int_{-\beta}^{\beta}d\tau\left[\frac{1}{2}\mathcal{M}\left(\frac{d\delta_p}{d\tau}\right)^2+U(\delta_p)\right]=\mathcal{W}+2\mathcal{E}\beta, \tag{32a}$$

with

$$\mathcal{W}=\frac{2^{23/4}}{5\times 3^{9/4}}\frac{\sqrt{K_1\chi_\perp}V}{\hbar\gamma}\epsilon^{5/4}\sqrt{\sin 2\theta_c}\left(1+\frac{m^2}{2K_1A\chi_\perp}\right)^{1/2}\times(a-c)^{5/2}[2(k_2^4-k_2^2+1)E(k_2)-(1-k_2^2)(2-k_2^2)K(k_2)]. \tag{32b}$$

Then the general formula Eq. (4) gives the imaginary parts of the metastable energy levels as

$$\mathrm{Im}\,\mathcal{E}=\frac{1}{2^{5/4}\times 3^{1/4}}\epsilon^{1/4}\gamma\sqrt{\frac{K_1}{\chi_\perp}}\sqrt{\sin 2\theta_c}\left(1+\frac{m^2}{2K_1A\chi_\perp}\right)^{-1/2}\times\frac{\sqrt{a-c}}{K(k_2')}\exp(-\mathcal{W}). \tag{33}$$

Here we discuss the low energy limit of the imaginary part of the metastable energy levels. For this case, $\mathcal{E}_n=(n+1/2)\Omega_2$,

$$\Omega_2=\sqrt{U''(\delta=0)/\mathcal{M}}=3^{1/4}\times 2^{1/4}\epsilon^{1/4}\gamma\sqrt{\frac{K_1}{\chi_\perp}}\sqrt{\sin 2\theta_c}\times\left(1+\frac{m^2}{2K_1A\chi_\perp}\right)^{-1/2},$$

$a\approx(3/2)(1-k'^2/4)$, $b\approx(3k'^2/4)(1+3k'^2/4)$, $c\approx-(3k'^2/4)(1+k'^2/4)$, and $k_2'^4=16\mathcal{E}/27U_0\ll 1$. Therefore, Eqs. (32b) reduces to

$$\mathcal{W}=\mathcal{W}_0-\left(n+\frac{1}{2}\right)+\left(n+\frac{1}{2}\right)\ln\left[\frac{2^{25/4}\times 3^{5/4}}{\left(n+\frac{1}{2}\right)}\frac{\sqrt{K_1\chi_\perp}V}{\hbar\gamma}\epsilon^{5/4}\times\sqrt{\sin 2\theta_c}\left(1+\frac{m^2}{2K_1A\chi_\perp}\right)^{1/2}\right], \tag{34a}$$

where

$$\mathcal{W}_0=\frac{2^{19/4}\times 3^{1/4}}{5}\epsilon^{5/4}\frac{\sqrt{K_1\chi_\perp}V}{\hbar\gamma}\frac{|\cot\theta_H|^{1/6}}{\sqrt{1+|\cot\theta_H|^{2/3}}}\times\sqrt{1+\frac{m^2}{2\chi_\perp K_1\left(\frac{1-\epsilon}{1++|\cot\theta_H|^{2/3}}+\frac{K_2}{K_1}\right)}}. \tag{34b}$$

The imaginary part of n-the excited level is found to be

$$\hbar\,\mathrm{Im}\,\mathcal{E}_n=\frac{3^{3/2}\times 2^7}{n!\sqrt{\pi}}\epsilon^{3/2}(K_1V)\frac{|\cot\theta_H|^{1/3}}{1+|\cot\theta_H|^{2/3}}\times\left[2^{27/4}\times 3^{5/4}\epsilon^{5/4}\frac{\sqrt{K_1\chi_\perp}V}{\hbar\gamma}\frac{|\cot\theta_H|^{1/6}}{\sqrt{1+|\cot\theta_H|^{2/3}}}\times\sqrt{1+\frac{m^2}{2\chi_\perp K_1\left(\frac{1-\epsilon}{1++|\cot\theta_H|^{2/3}}+\frac{K_2}{K_1}\right)}}\right]^{n-1/2}\times\exp(-\mathcal{W}_0). \tag{35}$$

For the case of large noncompensation ($m\gg\sqrt{\chi_\perp K_1}$), Eq. (35) reduces to the result for quantum tunneling in FM particles

$$\hbar\,\mathrm{Im}\,\mathcal{E}_n^{\mathrm{FM}}=\frac{(q_2^{\mathrm{FM}})^n}{n!}\hbar\,\mathrm{Im}\,\mathcal{E}_0^{\mathrm{FM}}, \tag{36a}$$

where

$$q_2^{\mathrm{FM}}=\frac{3^{5/4}\times 2^{25/4}\epsilon^{5/4}|\cot\theta_H|^{1/6}S'}{\sqrt{1-\epsilon+\frac{K_2}{K_1}(1+|\cot\theta_H|^{2/3})}}, \tag{36b}$$

and S' is the total spin of FM particles. $\mathrm{Im}\,\mathcal{E}_0^{\mathrm{FM}}$ is the imaginary part of ground-state level for FM particles

$$\hbar\Delta\mathcal{E}_0^{\mathrm{FM}}=\frac{3^{7/8}\times 2^{31/8}}{\sqrt{\pi}}(K_1V)\epsilon^{7/8}S'^{-1/2}\frac{|\cot\theta_H|^{1/4}}{1+|\cot\theta_H|^{2/3}}\times\left[1-\epsilon+\frac{K_2}{K_1}(1+|\cot\theta_H|^{2/3})\right]^{1/4}\exp(-\mathcal{W}_0^{\mathrm{FM}}), \tag{37a}$$

where

$$\mathcal{W}_0^{\mathrm{FM}}=\frac{2^{17/4}\times 3^{1/4}}{5}S'\epsilon^{5/4}\frac{|\cot\theta_H|^{1/6}}{\sqrt{1-\epsilon+\frac{K_2}{K_1}(1+|\cot\theta_H|^{2/3})}}. \tag{37b}$$

The WKB exponent in the imaginary part of ground-state level, $\mathcal{W}_0^{\mathrm{FM}}$ for $\pi/2+O(\sqrt{\epsilon})<\theta_H<\pi-O(\sqrt{\epsilon})$, fully agrees with the result of the ground-state spin tunneling in biaxial FM particles.[8]

For the case of small noncompensation ($m\ll\sqrt{\chi_\perp K_1}$), Eq. (35) reduces to the result of AFM spin tunneling

$$\hbar\,\mathrm{Im}\,\mathcal{E}_n^{\mathrm{AFM}}=\frac{(q_2^{\mathrm{AFM}})^n}{n!}\hbar\,\mathrm{Im}\,\mathcal{E}_0^{\mathrm{AFM}}, \tag{38a}$$

where

$$q_2^{\mathrm{AFM}}=\frac{3^{5/4}\times 2^{27/4}|\cot\theta_H|^{1/6}}{\sqrt{1+|\cot\theta_H|^{2/3}}}S\epsilon^{5/4}\sqrt{\frac{K_1}{J}}. \tag{38b}$$

$\hbar\,\mathrm{Im}\,\mathcal{E}_0^{\mathrm{AFM}}$ is the imaginary part of the ground-state level for AFM particles

$$\hbar\,\mathrm{Im}\,\mathcal{E}_0^{\mathrm{AFM}}=\frac{3^{7/8}\times 2^{29/8}}{\sqrt{\pi}}(JV)\left(\frac{K_1}{J}\right)^{3/4}\epsilon^{7/8}S^{-1/2}\times\frac{|\cot\theta_H|^{1/4}}{(1+|\cot\theta_H|^{2/3})^{3/4}}\exp(-\mathcal{W}_0^{\mathrm{AFM}}), \tag{39a}$$

where

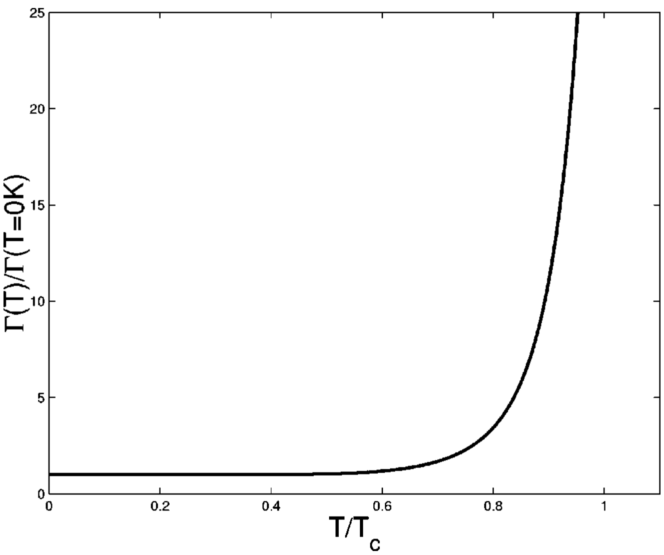

FIG. 3. The temperature dependence of the relative decay rate $\Gamma(T)/\Gamma(T=0\ \mathrm{K})$ for AFM particles in a magnetic field with a range of angles $\pi/2+O(\sqrt{\epsilon})<\theta_H<\pi-O(\sqrt{\epsilon})$. Here, $K_1=10^7$ erg/cm^3, $J=10^{10}$ erg/cm^3, $S=10^4$, $\epsilon=5\times10^{-3}$, and $\theta_H=3\pi/4$.

$$\mathcal{W}_0^{\mathrm{AFM}}=\frac{2^{19/4}\times3^{1/4}}{5}\sqrt{\frac{K_1}{J}}S\epsilon^{5/4}\frac{|\cot\theta_H|^{1/6}}{\sqrt{1+|\cot\theta_H|^{2/3}}}, \quad (39b)$$

and S is the sublattice spin of AFM particles.

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

$$\Gamma(T)=\frac{2}{\mathcal{Z}_0}\sum_n \mathrm{Im}\,\mathcal{E}_n^{\mathrm{AFM}}\exp(-\mathcal{E}_n^{\mathrm{AFM}}\beta), \quad (40)$$

where $\mathcal{Z}_0=\Sigma_n\exp(-\hbar\mathcal{E}_n^{\mathrm{AFM}}\beta)$ is the partition function with the harmonic oscillator approximated eigenvalues $\mathcal{E}_n^{\mathrm{AFM}}=(n+1/2)\Omega_2$. Then the decay rate at a finite temperature T is found to be

$$\Gamma(T)=2\,\mathrm{Im}\,\mathcal{E}_0^{\mathrm{AFM}}(1-e^{-\hbar\Omega_2\beta})\exp(q_2^{\mathrm{AFM}}e^{-\hbar\Omega_2\beta}), \quad (41)$$

where $\mathrm{Im}\,\mathcal{E}_0^{\mathrm{AFM}}$ and q_2^{AFM} are shown in Eqs. (38b) and (39a).

In Fig. 3 we plot the temperature dependence of the tunneling rate for the typical values of parameters for nanometer-scale single-domain antiferromagnets $S=10^4$, $\epsilon=1-\overline{H}/\overline{H}_c=5\times10^{-3}$, $K_1=10^7$ erg/cm^3, $J=10^{10}$ erg/cm^3, and $\theta_H=3\pi/4$. From Fig. 3 one can easily see the crossover from purely quantum tunneling to thermally assisted quantum tunneling. The temperature T_c characterizing the crossover from quantum to thermal regimes can be estimated as $k_BT_c=\Delta U/\mathcal{W}_0$, where ΔU is the barrier height, and $\mathcal{W}_0$ is the WKB exponent of the ground-state tunneling. For this case, one can easily show that the height of barrier is

$$\hbar\Delta U=\frac{2^{7/2}}{3^{3/2}}(K_1V)\epsilon^{3/2}\frac{|\cot\theta_H|^{1/6}}{1+|\cot\theta_H|^{2/3}},$$

and then the crossover temperature is

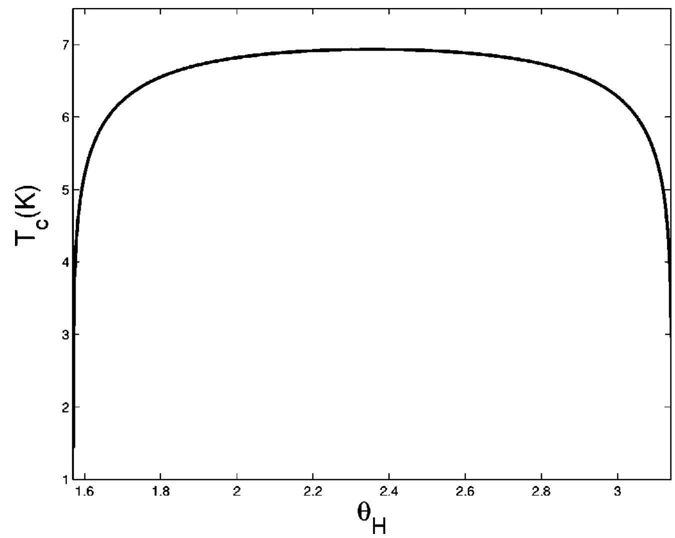

FIG. 4. The θ_H dependence of the crossover temperature T_c for $\pi/2<\theta_H<\pi$. Here, $K_1=10^7$ erg/cm^3, $J=10^{10}$ erg/cm^3, $S=10^4$, the radius of the particle is 5 nm, and $\epsilon=5\times10^{-3}$.

$$k_BT_c=\frac{5}{2^{5/4}\times3^{7/4}}\frac{\sqrt{K_1J}V}{S}\epsilon^{1/4}\frac{|\cot\theta_H|^{1/6}}{\sqrt{1+|\cot\theta_H|^{2/3}}}.$$

In Fig. 4, we plot the θ_H dependence of the crossover temperature T_c for AFM particles in magnetic field with a wide range of angles $\pi/2<\theta_H<\pi$. Figure 4 shows that the maximal value of T_c is about 6.94 K at $\theta_H=2.35$, which is one or two orders of magnitude higher than that for ferromagnets with a similar size.[7,8] Note that, even for ϵ as small as 10^{-3}, the angle corresponding to an appreciable change of the orientation of the Néel vector by quantum tunneling is $\delta_2=\sqrt{6\epsilon}$ rad$>4°$. The maximal value of T_c as well as Γ is expected to be observed in experiment.

C. $\theta_H=\pi$

In case of $\theta_H=\pi$, we have $\theta_c=0$ and $\eta=0$. Working out the integration over ϕ, the spin tunneling problem is mapped

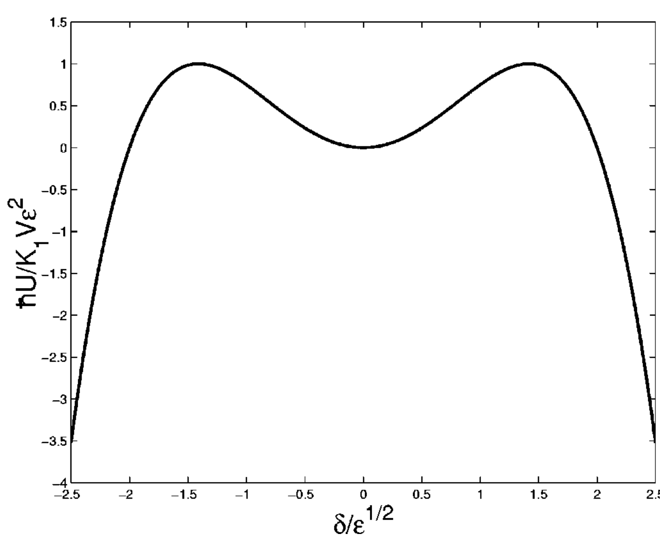

FIG. 5. The $\delta(=\theta-\theta_0)$ dependence of the effective potential $\hbar U/K_1V\epsilon^2$ for $\theta_H=\pi$ (the phenomenon of MQT).

onto the problem of a particle with effective mass $\mathcal{M} = \mathcal{M}_{\mathrm{AFM}} + \mathcal{M}_{\mathrm{FM}} = \chi_\perp V/\hbar\gamma^2 + m^2/2K_2\hbar\gamma^2$ moving in the one-dimensional potential well $U(\delta) = (K_1 V/\hbar)(\epsilon\delta^2 - \delta^4/4)$. Now the problem is one of MQT (as shown in Fig. 5), and the nonvacuum bounce at a given energy $\mathcal{E}>0$ is found to be

$$\delta_p = \sqrt{2\epsilon}\left(1+\sqrt{1-\frac{\hbar\mathcal{E}}{K_1 V\epsilon^2}}\right)^{1/2} \mathrm{dn}(\omega_3\tau, k_3), \quad (42a)$$

where

$$\omega_3 = \gamma\sqrt{\frac{K_1}{\chi_\perp}} \frac{\epsilon^{1/2}}{\sqrt{1+\frac{m^2}{2K_2\chi_\perp}}}\left(1+\sqrt{1-\frac{\hbar\mathcal{E}}{K_1 V\epsilon^2}}\right)^{1/2},$$

$$k_3^2 = 1 - \left(\frac{1-\sqrt{1-\frac{\hbar\mathcal{E}}{K_1 V\epsilon^2}}}{1+\sqrt{1-\frac{\hbar\mathcal{E}}{K_1 V\epsilon^2}}}\right). \quad (42b)$$

The classical action of the nonvacuum bounce Eq. (42b) is

$$\mathcal{S}_p = \int_{-\beta}^{\beta} d\tau \left[\frac{1}{2} m \left(\frac{d\delta_p}{d\tau}\right)^2 + U(\delta_p)\right] = \mathcal{W} + 2\mathcal{E}\beta, \quad (43a)$$

with

$$\mathcal{W} = \frac{4}{3}\epsilon^{3/2}\frac{\sqrt{K_1\chi_\perp}V}{\hbar\gamma}\sqrt{1+\frac{m^2}{2K_2\chi_\perp}}\left(1+\sqrt{1-\frac{\hbar\mathcal{E}}{K_1 V\epsilon^2}}\right)^{3/2} \times [(2-k_3^2)E(k_3) - 2k_3'^2 K(k_3)], \quad (43b)$$

where $k_3'^2 = 1 - k_3^2$. Then the imaginary parts of the metastable energy levels are

$$\mathrm{Im}\,\mathcal{E} = \frac{\omega_3}{4K(k_3')}\exp(-\mathcal{W}). \quad (44)$$

Now we consider the low energy limit of the imaginary part of the metastable energy level. For this case, $\mathcal{E}_n = (n+1/2)\Omega_3$,

$$\Omega_3 = \sqrt{U''(\delta=0)/\mathcal{M}} = \gamma\sqrt{\frac{2K_1}{\chi_\perp}}\frac{\epsilon^{1/2}}{\sqrt{1+\frac{m^2}{2K_2\chi_\perp}}},$$

$k_3'^2 = (1/2^{3/2}\epsilon^{3/2})\sqrt{\hbar/K_1 V\mathcal{M}}(n+1/2) \ll 1$, then

$$\mathcal{W} = \mathcal{W}_0 - \left(n+\frac{1}{2}\right) - \left(n+\frac{1}{2}\right) \times \ln\left(\frac{32\sqrt{2}\epsilon^{3/2}\frac{\sqrt{K_1\chi_\perp}V}{\hbar\gamma}\sqrt{1+\frac{m^2}{2K_2\chi_\perp}}}{n+1/2}\right), \quad (45a)$$

$$\mathcal{W}_0 = \frac{2^{7/2}}{3}\epsilon^{3/2}\frac{\sqrt{K_1\chi_\perp}V}{\hbar\gamma}\sqrt{1+\frac{m^2}{2K_2\chi_\perp}}, \quad (45b)$$

and

$$\mathrm{Im}\,\mathcal{E}_n = \frac{1}{n!\sqrt{\pi}}\gamma\sqrt{\frac{K_1}{\chi_\perp}}\epsilon^{1/2}\frac{1}{\sqrt{1+\frac{m^2}{2K_2\chi_\perp}}} \times\left(32\sqrt{2}\epsilon^{3/2}\frac{\sqrt{K_1\chi_\perp}V}{\hbar\gamma}\sqrt{1+\frac{m^2}{2K_2\chi_\perp}}\right)^{n+1/2} \times\exp(-\mathcal{W}_0). \quad (45c)$$

In the case of large non-compensation ($m \gg \sqrt{\chi_\perp K_1}$), Eq. (45c) reduces to the result for quantum tunneling in FM particles

$$\hbar\,\mathrm{Im}\,\mathcal{E}_n^{\mathrm{FM}} = \frac{(q_3^{\mathrm{FM}})^n}{n!}\hbar\,\mathrm{Im}\,\mathcal{E}_0^{\mathrm{FM}}, \quad (46a)$$

where

$$q_3^{\mathrm{FM}} = 32\epsilon^{3/2}\sqrt{\frac{K_1}{K_2}}S'. \quad (46b)$$

$\hbar\Delta\mathcal{E}_0^{\mathrm{FM}}$ is the imaginary part of ground-state level for FM particles

$$\hbar\Delta\mathcal{E}_0^{\mathrm{FM}} = \frac{8}{\sqrt{\pi}}(K_1 V)S'^{-1/2}\epsilon^{5/4}\left(\frac{K_2}{K_1}\right)^{1/4}\exp(-\mathcal{W}_0^{\mathrm{FM}}), \quad (47a)$$

where

$$\mathcal{W}_0^{\mathrm{FM}} = \frac{8}{3}S'\epsilon^{3/2}\sqrt{\frac{K_1}{K_2}}, \quad (47b)$$

and S' is the total spin of FM particles. The WKB exponent in the imaginary part of ground-state level, $\mathcal{W}_0^{\mathrm{FM}}$ for $\theta_H = \pi$, fully agrees with the result of the ground-state spin tunneling in biaxial FM particles.[8]

For the case of small noncompensation ($m \ll \sqrt{\chi_\perp K_1}$), Eq. (45c) reduces to the result of AFM spin tunneling

$$\hbar\,\mathrm{Im}\,\mathcal{E}_n^{\mathrm{AFM}} = \frac{(q_3^{\mathrm{AFM}})^n}{n!}\hbar\,\mathrm{Im}\,\mathcal{E}_0^{\mathrm{AFM}}, \quad (48a)$$

where

$$q_3^{\mathrm{AFM}} = 32\sqrt{2}S\epsilon^{3/2}\sqrt{\frac{K_1}{J}}. \quad (48b)$$

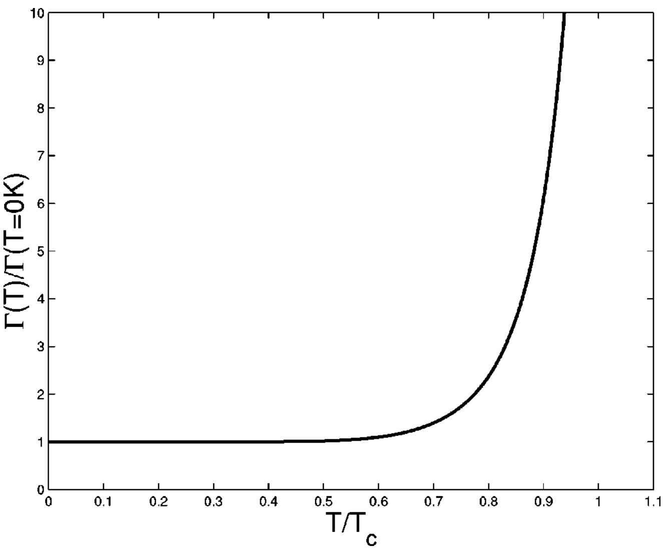

FIG. 6. The temperature dependence of the relative decay rate $\Gamma(T)/\Gamma(T=0\ \mathrm{K})$ for AFM particles in a magnetic field with $\theta_H=\pi$. Here, $K_1=10^7$ erg/cm^3, $J=10^{10}$ erg/cm^3, $S=1.5\times10^4$, and $\epsilon=0.1$.

$\hbar\ \mathrm{Im}\,\mathcal{E}_0^{\mathrm{AFM}}$ is the imaginary part of the ground state for AFM particles,

$$\hbar\ \mathrm{Im}\,\mathcal{E}_0^{\mathrm{AFM}}=\frac{2^{11/4}}{\sqrt{\pi}}(JV)\left(\frac{K_1}{J}\right)^{3/4}\epsilon^{5/4}S^{-1/2}\exp(-\mathcal{W}_0^{\mathrm{AFM}}), \tag{49a}$$

where

$$\mathcal{W}_0^{\mathrm{AFM}}=\frac{2^{7/2}}{3}\sqrt{\frac{K_1}{J}}S\epsilon^{3/2}, \tag{49b}$$

and S is the sublattice spin of AFM particles. And the final result of the decay rate at finite temperature T is found to be

$$\Gamma(T)=2\ \mathrm{Im}\,\mathcal{E}_0(1-e^{-\hbar\Omega_3\beta})\exp(q_3^{\mathrm{AFM}}e^{-\hbar\Omega_3\beta}). \tag{50}$$

The temperature dependence of the decay rate is shown in Fig. 6. For this case, the position of the energy barrier is $\delta_b=\sqrt{2\epsilon}$, the height of barrier is $\hbar\Delta U=(K_1V)\epsilon^2$. Therefore, the crossover temperature is $k_BT_c=(3/8\sqrt{2})\times(\sqrt{K_1J}V)\epsilon^{1/2}S^{-1}$.

IV. CONCLUSIONS

In summary we have investigated the quantum tunneling between excited levels in a general AFM model with noncompensated sublattices in the presence of an external magnetic field at arbitrarily directed angle. By calculating the nonvacuum instantons exactly in the spin-coherent-state path-integral representation, we obtain the analytic formulas for the tunnel splitting between degenerate excited levels in MQC and the imaginary parts of the metastable energy levels in MQT of the Néel vector in the low barrier limit for the external magnetic field perpendicular to the easy axis ($\theta_H=\pi/2$), for the field antiparallel to the initial easy axis ($\theta_H=\pi$), and for the field at an angle between these two orientations $[\pi/2+O(\sqrt{\epsilon})<\theta_H<\pi-O(\sqrt{\epsilon})]$. The temperature dependences of the decay rate are clearly shown for each case. For the case of large noncompensation, the WKB exponents in the tunnel splitting and the tunneling rate fully agree with the results of ground-state spin tunneling in biaxial FM particles.[8] In the comparison with other more accurate methods (e.g., Ref. 31), our result gives the exact same expression of the exponential term in the tunneling rate, while could differ by a factor in the preexponential term. Since our major interest in this work is the angular-dependent spin tunneling in AFM particles, which is largely determined by the exponential term in the tunnel splitting or the tunneling rate, this correction factor does not change much in front of the exponentially small action term.

One important conclusion is that the tunneling rate and the tunnel splitting at excited levels depend on the orientation of the external magnetic field distinctly. Even a small misalignment of the field with $\theta_H=\pi/2$ and π orientations can completely change the results of the tunneling rates. In a wide range of angles the θ_H dependence of the WKB exponent mainly comes from the behavior of the function $|\cot\theta_H|^{1/6}/\sqrt{1+|\cot\theta_H|^{2/3}}$. Another interesting conclusion concerns the field strength dependence of the WKB exponent in the tunnel splitting or the tunneling rate. It is found that in a wide range of angles, the $\epsilon(=1-\overline{H}/\overline{H}_c)$ dependence of the WKB exponent is given by $\epsilon^{5/4}$, not $\epsilon^{3/2}$ for $\theta_H=\pi/2$, and $\theta_H=\pi$. As a result, we conclude that both the orientation and the strength of the external magnetic field are the controllable parameters for the experimental test of the phenomena of macroscopic quantum tunneling and coherence of the Néel vector between excited levels in single-domain AFM nanoparticles at sufficiently low temperatures. If the experiment is to be performed, there are three control parameters for comparison with theory: the angle of the external magnetic field θ_H, the strength of the field in terms of ϵ, and the temperature T.

A detailed comparison between the theory and experiment on quantum tunneling of magnetization remains a challenging task. In order to avoid the complications due to distributions of particle size and shape, some groups have tried to study the temperature and field dependence of magnetization reversal of individual magnets. Wernsdorfer and co-workers have performed the switching field measurements on individual ferrimagnetic and insulating BaFeCoTiO nanoparticles containing about 10^5–10^6 spins at very low temperatures (0.1–6 K).[11] They found that above 0.4 K, the magnetization reversal of these particles is unambiguously described by the Néel-Brown theory of thermal activated rotation of the particle's moment over a well defined anisotropy energy barrier. Below 0.4 K, strong deviations from this model are evidenced which are quantitatively in agreement with the predictions of the MQT theory without dissipation.[7] The BaFeCoTiO nanoparticles have a strong uniaxial magnetocrystalline anisotropy.[11] However, the theoretical results presented here may be useful for checking the general theory in a general AFM systems, in which the magnitude of uncompensation ranges from large (FM case) to small (AFM case). The experimental procedures on single-domain FM nanoparticles of Barium ferrite with uniaxial symmetry[11] may be applied to the general AFM systems. Note that the inverse of the WKB exponent in the tunneling rate B^{-1} is the magnetic viscosity S at the quantum-tunneling-dominated regime $T \ll T_c$ studied by magnetic relaxation measurements.[1,2] Therefore, the spin tunneling phenomena should be checked at any θ_H by magnetic relaxation measurements. Over the past years a lot of experimental and theoretical works were performed on the spin tunneling in molecular Mn_{12}-Ac (Refs. 1,32) and Fe_8 (Refs. 2,33) clusters having a collective spin state $S=10$ (in this paper $S=10^3$–10^5). These measurements on molecular clusters with $S=10$ suggest that quantum phenomena might be observed at larger system sizes with $S \gg 1$. Further experiments should focus on the level quantization of collective spin states of $S=10^2$–10^4.

Finally we discuss briefly the dissipation effect on spin tunneling. 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.[34] Including the effects of dissipation, the decay rate, in particular, is given by[34,35,2]

$$\Gamma_n=\frac{1}{2}(\Delta\mathcal{E}_n)^2\sum_{n'}\frac{\Omega_{nn'}}{(\mathcal{E}_n-\mathcal{E}_{n'})^2+\Omega_{nn'}^2}, \tag{51}$$

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 nth 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

$$\Gamma(T)=\sum_n \frac{(\Delta\mathcal{E}_n)^2}{2\Omega_n}\exp(-\hbar\mathcal{E}_n\beta), \tag{52}$$

where the level broadening Ω_n contains all the details of the coupling between the magnet and its environment. If the width caused by the dissipative coupling 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 spin tunneling at excited levels for single-domain AFM particles in an arbitrarily directed magnetic field at sufficiently low temperatures. Strong dissipation is hardly the case for single-domain magnetic particles,[36] 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.[2,5,24,34,37] 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.

The theoretical calculations performed in this paper can be extended to the AFM particles with a much more complex structure of magnetocrystalline anisotropy energy, such as trigonal, tetragonal, and hexagonal crystal symmetries. Work along this line is still in progress. We hope that the theoretical results presented in this paper may stimulate more experiments whose aim is observing macroscopic quantum tunneling and coherence in nanometer-scale single-domain antiferromagnets.

ACKNOWLEDGMENTS

We are grateful to W. Wernsdorfer for useful discussions. R.L. would like to acknowledge H. Pan, Y. Zhou, X.-B. Wang, Y.-L. Liu, J.-L Zhu, Z. Xu, and J.-Q. Liang for helpful discussions. This work was supported in part by Grant No. De730/1-2 of the Deutsche Forschungsgemeinschaft.

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

[2] For a review of recent developments on spin tunneling in molecular cluster and the dissipation effects, see I. Tupitsyn and B. Barbara, cond-mat/0002180 (unpublished), and references therein: B. Barbara, L. Thomas, F. Lionti, I. Chiorescu, and A. Sulpice, J. Magn. Magn. Mater. **200**, 167 (1999); W. Wernsdorfer, Adv. Chem. Phys. **118**, 99 (2001).

[3] M. N. Leuenberger and D. Loss, Nature (London) **410**, 789 (2001), and references therein.

[4] A. J. Leggett, in *Quantum Tunneling of Magnetization*, edited by L. Gunther and B. Barbara (Kluwer, Dordrecht, 1995).

[5] A. Garg, J. Appl. Phys. **76**, 6168 (1994); Phys. Rev. Lett. **74**, 1458 (1995).

[6] O. B. Zaslavskii, Phys. Rev. B **42**, 992 (1990).

[7] M.-G. Miguel and E. M. Chudnovsky, Phys. Rev. B **54**, 388 (1996).

[8] G.-H. Kim and D. S. Hwang, Phys. Rev. B **55**, 8918 (1997).

[9] G.-H. Kim, Phys. Rev. B **57**, 10 688 (1998).

[10] D. A. Garanin, X. M. Hidalgo, and E. M. Chudonovsky, Phys. Rev. B **57**, 13 639 (1998).

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

[12] B. Barbara and E. M. Chudnovsky, Phys. Lett. A **145**, 205 (1990).

[13] E. M. Chudnovsky, J. Magn. Magn. Mater. **140-144**, 1821 (1995).

[14] J. M. Duan and A. Garg, J. Phys.: Condens. Matter **7**, 2171 (1995).

[15] I. V. Krive and O. B. Zaslavskii, J. Phys.: Condens. Matter **2**, 9457 (1990); E. N. Bogachek and I. V. Krive, Phys. Rev. B **46**, 14 559 (1992).

[16] H. Simanjuntak, J. Phys.: Condens. Matter **6**, 2925 (1994).

[17] G. Levine and J. Howard, Phys. Rev. Lett. **75**, 4142 (1995).

[18] A. Chiolero and D. Loss, Phys. Rev. B **56**, 738 (1997); Phys. Rev. Lett. **80**, 169 (1998).

[19] Rong Lü, Peifang Zhang, Jia-Lin Zhu, and Lee Chang, Phys. Rev. B **56**, 10 993 (1997).

[20] J.-Q. Liang *et al.*, Phys. Lett. A **248**, 434 (1998); Y.-H. Nie *et al.*, Phys. Rev. B **64**, 134419 (2001).

[21] C. Kittle, Rev. Mod. Phys. **21**, 552 (1949); E. P. Wohlfarth, Proc. Phys. Soc., London, Sect. A **65**, 1053 (1952); C. Herring, Phys. Rev. **85**, 1003 (1952).
[22] D. Loss, D. P. DiVicenzo, and G. Grinstein, Phys. Rev. Lett. **69**, 3232 (1992).
[23] J. von Delft and G. L. Henley, Phys. Rev. Lett. **69**, 3236 (1992).
[24] H. B. Braun and D. Loss, Europhys. Lett. **31**, 555 (1995).
[25] W. Wernsdorfer, Science **284**, 133 (1999).
[26] S. Coleman, *Aspects of Symmetry* (Cambridge University Press, Cambridge, England, 1985), Chap. 7.
[27] J.-Q. Liang and H. J. W. Müller-Kirstein, Phys. Rev. D **46**, 4685 (1992); **50**, 6519 (1994); **51**, 718 (1995).
[28] L. D. Landau and E. M. Lifshita, *Quantum Mechanics*, 3rd ed. (Pergamon, New York, 1977), Chap. VII, Sec. 50, Problem 3.
[29] E. Gildener and A. Patrascioiu, Phys. Rev. D **16**, 423 (1977); U. Weiss and W. Haeffner, *ibid.* **27**, 2916 (1983); H. K. Shepard, *ibid.* **27**, 1288 (1983).
[30] For a recent discussion see A. Garg, cond-mat/0003115 (unpublished); H. J. W. Müller-Kirsten, J.-Z. Zhang, and Y. Zhang, J. High Energy Phys. **11**, 11 (2001).
[31] P. Achuthan, H. J. W. Müller-Kirsten, and A. Wiedemann, Fortschr. Phys. **38**, 77 (1990).
[32] C. Paulsen, J.-G. Park, B. Barbara, R. Sessoli, and A. Caneschi, J. Magn. Magn. Mater. **140-144**, 379 (1995); C. Paulsen and J.-G. Park, in *Quantum Tunneling of Magnetization*, edited by L. Gunther and B. Barbara (Kluwer, Dordrecht, 1995); M. A. Novak and R. Sessoli, *ibid.*; J. M. Hernández, X. X. Zhang, F. Luis, J. Bartolomé, J. Tejada, and R. Ziolo, Europhys. Lett. **35**, 301 (1996); L. Thomas, F. Lionti, R. Ballou, D. Gatteschi, R. Sessoli, and B. Barbara, Nature (London) **383**, 145 (1996); J. R. Friedman, M. P. Sarachik, J. Tejada, and R. Ziolo, Phys. Rev. Lett. **76**, 3830 (1996); J. M. Hernńdez, X. X. Zhang, F. Luis, J. Tejada, J. R. Friedman, M. P. Sarachik, and R. Ziolo, Phys. Rev. B **55**, 5858 (1997); F. Lionti, L. Thomas, R. Ballou, B. Barbara, A. Sulpice, R. Sessoli, and D. Gatteschi, J. Appl. Phys. **81**, 4608 (1997); D. A. Garanin and E. M. Chudnovsky, Phys. Rev. B **56**, 11 102 (1997).
[33] A.-L. Barra, P. Debrunner, D. Gatteschi, C. E. Schulz, and R. Sessoli, Europhys. Lett. **35**, 133 (1996); C. Sangregorio, T. Ohm, C. Paulsen, R. Sessoli, and D. Gatteschi, Phys. Rev. Lett. **78**, 4645 (1997).
[34] A. Garg, Phys. Rev. B **51**, 15 161 (1995).
[35] U. Weiss, *Quantum Dissipative Systems* (World Scientific, Singapore, 1999).
[36] A. Garg and G.-H. Kim, Phys. Rev. Lett. **63**, 2512 (1989); Phys. Rev. B **43**, 712 (1991); H. Simanjuntak, J. Low Temp. Phys. **90**, 405 (1992); P. C. E. Stamp, Phys. Rev. Lett. **66**, 2802 (1991); E. M. Chudnovsky, O. Iglesias, and P. C. E. Stamp, Phys. Rev. B **46**, 5392 (1992); A. Garg, Phys. Rev. Lett. **70**, 1541 (1993); **74**, 1458 (1995); J. Appl. Phys. **76**, 6168 (1994); G. Tatara and H. Fukuyama, Phys. Rev. Lett. **72**, 772 (1994).
[37] E. M. Chudnovsky and D. P. DiVincenzo, Phys. Rev. B **48**, 10 548 (1993).