Thermally Assisted Macroscopic Quantum Tunneling of a
Ferromagnetic Particle in a Magnetic Field at an Arbitrary Angle

Gwang-Hee Kim

Department of Physics, Sejong University, Seoul 143-747, KOREA

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Abstract

At finite temperature we study the quantum tunneling of magnetization for a small ferromagnetic particle with the biaxial symmetry placed in a magnetic field at an arbitrary angle. We present numerical WKB exponent below the crossover temperature in which the quantum tunneling is affected by the thermal activation, and the approximate form of the WKB exponent around the crossover region. The effect of quantum fluctuations on the thermal activation rate beyond the crossover regime are discussed.

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I. INTRODUCTION

In the last decade, the problem of quantum tunneling of magnetization (QTM) in a single domain magnetic particle has attracted a great deal of theoretical and experimental interest. There are two reasons for this. One is the rapid development of the new field of nanomagnetism, in which a single domain magnetic particle can be prepared. The other is that such a system meets the main criteria for observing macroscopic quantum tunneling (MQT). There are three conditions for seeing MQT which can be stated qualitatively as follows. First, the height and the width of the barrier should not be large. Second, the effective moment of inertia associated with the magnetization switching should not be too large. These two conditions are required for a tunneling time accessible to current experimental situations. Third, the crossover temperature at which the transition between the thermal activation and the quantum tunneling occurs should be of the order of milliKelvin range or above. Based on the criteria, it has been believed that a magnetic field is a good external parameter to make QTM observable. Also, tunneling dynamics poses many interesting problems, particularly concerning the role played by the thermal fluctuations at finite temperature. This is the main subject of this paper, which deals with thermal corrections to quantum tunneling rate of a single domain ferromagnetic particle in a magnetic field.

Consider a single domain ferromagnetic particle uniformly magnetized along an easy axis determined by magnetocrystalline anisotropy. Since the anisotropy generates two or more energetically equivalent orientations of the magnetization, at extremely low temperature the direction of the magnetization \( \mathbf{M} \) might change from one easy axis to the other easy axis by the quantum tunneling process. However, according to the criteria, the rate of change of \( \mathbf{M} \) is too small to be observed without controlling the height and the width of the barrier as well as the effective moment of inertia intrinsically produced by the magnetic anisotropy energy. Applying an external magnetic field \( \mathbf{H} \) in a proper direction, one of two equivalent orientations becomes metastable and the magnitude of the physical quantities can be controlled in an appropriate way. In this situation the change of \( \mathbf{M} \) with time is expected
to be able to be observed and compared with theoretical results. As temperature increases from zero, thermal effects add to the quantum tunneling process. At sufficiently high temperature, the direction of the magnetization is changed by the pure thermal activation whose rate is proportional to \( \exp(-U/k_B T) \) where \( U \) is an energy barrier related to the magnetic anisotropy energy and the external magnetic field. In the intermediate temperature region, it is expected that there exists a crossover temperature \( T_c \). Around \( T_c \) both thermal fluctuations and quantum tunneling coexist, namely, either the thermally activated quantum tunneling below \( T_c \) or the quantum mechanically assisted thermal activation process above \( T_c \) can occur. Well below the crossover temperature, i.e., at a temperature low enough to neglect the thermal activation process, the rate of change of \( M \) is solely due to quantum tunneling. 

A number of theoretical studies about QTM for the single domain ferromagnetic particle have been performed for several magnetocrystalline anisotropy. The QTM problem with a magnetic field at an arbitrary angle was firstly studied by Zaslavskii who calculated the tunneling rate for the uniaxial symmetry by mapping the spin system onto a one-dimensional particle system. For the same symmetry, Miguel and Chudnovsky have calculated the tunneling rate within the imaginary time path integral method. They have also discussed the tunneling rate at finite temperature and suggested experimental procedures. Kim and Hwang have performed the calculation based on instanton approach for biaxial and tetragonal symmetry. Their work presented the tunneling and oscillation rate between angles \( \pi/2 \leq \theta_H \leq \pi \) at zero temperature, where \( \theta_H \) is the angle between the initial easy axis and the external magnetic field. This paper extends the tunneling rate for the biaxial symmetry to a finite temperature. Within the instanton approach, we present the numerical results for the WKB exponent below the crossover temperature and their approximate formulas around the crossover temperature. Also, we discuss the effect of quantum fluctuations on the thermal activation rate beyond the crossover regime.

This paper is structured in the following way. In Sec. I, we introduce general formulation for the tunneling rate based on the spin coherent state path integral method. We discuss the
angular dependence of the critical field and the critical angle by using approximate formula of the total energy in the small \( \epsilon(= 1 - H/H_c) \) limit, where \( H_c \) is defined as a critical field at which the barrier disappears. In Sec. II, we give a brief survey of the quantum tunneling of magnetization in the presence of the magnetic field with a general direction at zero temperature. In Sec. III, we consider thermal effects on the quantum tunneling rates below \( T_c \) and quantum corrections to thermal activation rates above \( T_c \) and present their numerical results in the entire temperature regime. In Sec. IV we summarize the analytic results discussed and provide actual estimates of such important quantities as the tunneling rate, crossover temperature, and so on, for several real materials.

II. GENERAL FORMULATION

Consider the spin coherent state path integral representation of the partition function given by

\[
Z(\beta \hbar) = \oint D[M(\tau)] \exp(-SE/\hbar),
\]

where \( \beta = 1/k_B T \), the path sum is over all periodic paths \( M(\tau) = M(\tau + \beta \hbar) \), and \( SE \) the Euclidean action which includes the Euclidean version of the magnetic Lagrangian \( L_E \) as

\[
S_E[M(\tau)] = \oint d\tau L_E[M(\tau)].
\]

Due to strong exchange interaction in a single domain ferromagnetic particle, the magnitude of the magnetization is a constant \( M_0 \). For that reason the dynamical variable is the direction of the magnetization, equivalently, \( \theta(\tau) \) and \( \phi(\tau) \) in the spherical coordinates of \( M \). Therefore, up to the normalization the functional measure in Eq. (1) is equivalent to \( D\Omega \) as

\[
D\Omega = \lim_{\epsilon \to 0^+} \prod_{k=1}^{N} \left( \frac{2J + 1}{4\pi} \right) \sin \theta_k d\theta_k d\phi_k,
\]

where \( \epsilon = \max(\tau_{k+1} - \tau_k) \) and \( J = M_0 V/\hbar \gamma \). Here \( \gamma = g\mu_B/\hbar \), with \( g \) being the g-factor, \( \mu_B \) the Bohr magneton, \( M_0 \) the magnitude of the magnetization and \( V \) the volume of the
system. Since the tunneling rate $\Gamma$ of a metastable state is proportional to the imaginary part of the free energy of the system, $\text{Im}F$, and $\text{Im}Z \propto \text{Im}F$, from Eq. (1) the tunneling rate in semiclassical limit, with an exponential accuracy, is

$$\Gamma \propto \exp(-S_{\text{E}}^{\text{min}}(T)/\hbar),$$

(4)

where $S_{\text{E}}^{\text{min}}(T)$ is obtained along the trajectory with period $\beta\hbar$ that minimizes the Euclidean action

$$S_{E} = V \int_{0}^{\beta\hbar} \{ iM_{0}[1 - \cos \theta(\tau)] \frac{d\phi(\tau)}{d\tau} + E[M(\tau)] \} d\tau.$$

(5)

The first term in Eq. (5) is the topological Wess-Zumino term [8,9], and the second term is the energy density, which is composed of the magnetic anisotropy energy $E_{a}$ and the energy given by an external magnetic field $H$, given by

$$E[M(\tau)] = E_{a} - M \cdot H.$$  

(6)

We have selected the biaxial symmetry with magnetic anisotropy energy $E_{a} = K_{1}(\alpha_{1}^{2} + \alpha_{2}^{2}) + K_{2}\alpha_{2}^{2}$. Here $\alpha_i$'s are the directional cosines and $K_{1} > 0$, $K_{2} > 0$ are the parallel and transverse anisotropy constants, respectively, whose relative magnitude can be of any value. Applying the magnetic field in the $xz$ plane [4,5] and expressing $\alpha_{1}$, $\alpha_{2}$ and $\alpha_{3}$ in spherical coordinates of $M$, the energy (6) is expressed as

$$E[\theta, \phi] = K_{1} \sin^{2} \theta + K_{2} \sin^{2} \phi \sin^{2} \theta$$

$$-H_{x}M_{0} \sin \theta \cos \phi - H_{z}M_{0} \cos \theta + E_{0},$$

(7)

where $E_{0}$ is a constant to make $E(\theta, \phi)$ zero at the initial orientation. As will be seen later, while there isn’t an exact analog of kinetic energy in the action (5), there is an effective moment of inertia in the dynamics of a single domain particle that is inversely proportional to a linear combination of $K_{2}$ and $H_{x}$. For this reason we need either the transverse anisotropy constant $K_{2}$ or the magnetic field $H_{x}$ transverse to the initial easy axis for quantum tunneling. Otherwise, QTM is not observable due to the effective moment of inertia whose magnitude becomes infinity.
Introducing the dimensionless constants,

\[ k_2 = \frac{K_2}{K_1}, \quad h_x = \frac{H_x}{H_0}, \quad h_z = \frac{H_z}{H_0}, \quad H_0 = \frac{2K_1}{M_0}, \]  

(8)

we obtain the energy (7) written as

\[ \bar{E}[\theta, \phi] = \frac{1}{2} \sin^2 \theta + \frac{k_2}{2} \sin^2 \phi \sin^2 \theta - h_x \sin \theta \cos \phi - h_z \cos \theta + E_0, \]

(9)

where \( \bar{E}(\theta, \phi) = E(\theta, \phi)/2K_1 \). As noted in Eq. (9), there are two equivalent easy directions in the \( xy \) plane, which are \( \phi = 0 \) and \( \pi \) in the azimuthal angle if \( h_x = 0 \). We treat the problem of the easy directions given by \( \phi = 0 \) in the \( xy \) plane. Even though we start the argument from \( \phi = \pi \), the whole discussion is the same as the one from \( \phi = 0 \) by replacing \( \phi \) by \( \pi + \phi \). So, the classical path becomes \( \phi_{cl}(\tau) = \phi_0(\tau) + n\pi \) where \( n = 0 \) or \( 1 \) where \( \phi_0(\tau) \) is the classical trajectory started from \( \phi = 0 \). [3] However, if the external field is not along the easy axis (\( z \) axis), i.e., \( h_x \neq 0 \), only \( \phi = 0 \) in Eq. (9) is the easy direction in the \( xy \) plane and \( \phi = \pi \) not. Keeping this point in mind, the total energy on the easy plane \( \phi \) becomes

\[ \bar{E}(\theta, 0) = \frac{1}{2} \sin^2 \theta - h \cos(\theta - \theta_H) + E_0, \]

(10)

where we used \( h_x = h \sin \theta_H \) and \( h_z = h \cos \theta_H \). Let us define \( \theta_0 \) to be the angle of metastable state generated by the anisotropy energy and the external magnetic field, and \( \theta_c \) the angle at which the barrier vanishes by the external critical magnetic field \( H_c \). Then, \( \theta_0 \) is determined by \( [d\bar{E}(\theta, 0)/d\theta]_{\theta=\theta_0} = 0 \), and \( \theta_c \) and \( h_c \) by both \( [d\bar{E}(\theta, 0)/d\theta]_{\theta=\theta_c,h=h_c} = 0 \) and \( [d^2\bar{E}(\theta, 0)/d\theta^2]_{\theta=\theta_c,h=h_c} = 0 \). After simple calculations, the dimensionless switching or critical field \( h_c(\equiv H_c/H_0) \) and the critical angle \( \theta_c \) are expressed as [4,5]

\[ h_c = (\sin^{2/3} \theta_H + |\cos \theta_H|^{2/3})^{-3/2}, \]  

(11)

\[ \theta_c = \frac{1}{2} \arcsin\left[ \frac{2}{1 + |\cot \theta_H|^{2/3}} \right], \]  

(12)

where it should be noted that Eq. (11) is the well-known Stoner-Wohlfarth expression. [10] Simple analysis for Eq. (11) reveals that the small value of \( \epsilon \) is favorable for the
magnetization switching because the height and the width of the barrier are proportional to the power of $\epsilon$. The situation can be achieved by applying the external magnetic field close to the critical field. Hence the problem will be considered in the small limit of $\epsilon$, i.e., $\epsilon \ll 1$.

Expanding $[d\bar{E}(\theta, 0)/d\theta]_{\theta=\theta_0} = 0$ about $\theta_c$, and using the relations $[d\bar{E}(\theta, 0)/d\theta]_{\theta=\theta_c, h=h_c} = 0$ and $[d^2\bar{E}(\theta, 0)/d\theta^2]_{\theta=\theta_c, h=h_c} = 0$, we obtain the equation for $\eta(=\theta_c - \theta_0)$ as

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

Simple calculations show that $\eta$ 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 $\eta$ is determined by the first term which leads to $\eta \simeq \sqrt{2\epsilon}/3$. However, when $\theta_H$ is very close to $\pi/2$ or $\pi$, $\sin(2\theta_c)$ becomes close to zero, and the second term is much larger than the first term. Therefore, the value of $\eta$ is obtained from the second term when $\theta_H \simeq \pi/2$ or $\pi$, which leads to $\eta \simeq \sqrt{2\epsilon}$ for $\theta_H \simeq \pi/2$ and $\eta \simeq 0$ for $\theta_H \simeq \pi$ from the detailed analysis of the equations which $\theta_0$ and $\theta_c$ satisfy. Since the first term in (13) is dominant in the range of value $\theta_c$ which satisfies $\tan(2\theta_c) > O(\sqrt{\epsilon})$, $\eta \simeq \sqrt{2\epsilon}/3$ is valid for $\pi/2 + O(\sqrt{\epsilon}) < \theta_H < \pi - O(\sqrt{\epsilon})$ from Eq. (12). Fig. 1 shows that this is checked by performing the numerical calculation for the equations which $\theta_0$ and $\theta_c$ satisfy.

Introducing a small variable $\delta(=\theta - \theta_0)$, we obtain an approximate form of $\bar{E}(\theta, \phi)$ as

$$\bar{E}(\delta, \phi) = \frac{k_2}{2} \sin^2 \phi \sin^2(\theta_0 + \delta) + h_x \sin(\theta_0 + \delta)(1 - \cos \phi) + \bar{E}_1(\delta), \quad (14)$$

where $\bar{E}(\theta, \phi)$ is represented as $\bar{E}(\delta, \phi)$, and $\bar{E}_1(\delta)$ is a function of only $\delta$ given by

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

Although $\cos(2\theta_c)$-term in Eq. (13) looks smaller by a factor of $\eta$ which is of the order of $\sqrt{\epsilon}$, it can not be neglected near $\theta_H = \pi/2$ and $\pi$ because $\sin(2\theta_c)$ is almost zero for these regions of $\theta_H$. In order to evaluate the order of magnitude of each term in action (3) and simplify the calculations for $\epsilon \ll 1$, it is convenient to use new scaled variables
\[ \tau = \epsilon^{\alpha/2} \omega_0 \tau, \quad \tilde{\delta} = \delta / \sqrt{\epsilon}, \quad \omega_0 = 2\gamma K_1 / M_0. \] (16)

Then, the Euclidean action (15) becomes

\[
S_E[\tilde{\delta}(\bar{\tau}), \phi(\bar{\tau})] = \hbar J \epsilon^{-\frac{\alpha}{2}} \int_{0}^{\beta} d\bar{\tau} \left[ i \epsilon^{\frac{\alpha}{2}} \left[ 1 - \cos(\theta_0 + \sqrt{\epsilon} \tilde{\delta}) \right] d\phi / d\bar{\tau} \right.
\]
\[
+ \frac{k_2}{2} \sin^2 \phi \sin^2(\theta_0 + \sqrt{\epsilon} \tilde{\delta}) + h_x \sin(\theta_0 + \sqrt{\epsilon} \tilde{\delta})(1 - \cos \phi)
\]
\[
\left. + \frac{1}{4} \sin(2\theta_c) \epsilon^{\frac{\alpha}{2}}(3\tilde{\delta}^2 \eta / \sqrt{\epsilon} - \tilde{\delta}^3) + \frac{1}{2} \cos(2\theta_c) \epsilon^2 [\tilde{\tau}^2 (1 - 3\eta^2 / 2\epsilon) + \tilde{\delta}^3 \eta / \sqrt{\epsilon} - \tilde{\delta}^4 / 4] \right]. \quad (17)
\]

where \( \tilde{\beta} = \epsilon^{\alpha/2} \omega_0 \beta \hbar \) and the parameters \( \alpha \) is fixed by the analysis of the order of magnitude of each term in the action (17) subject to the situation considered.

### III. QTM AT ZERO TEMPERATURE

Consider the Euclidean action (17) at \( T = 0 \) by performing a simple dimensional analysis. At \( T = 0 \) (\( \beta \to \infty \)) the instanton starts and ends at the metastable state \( \tilde{\delta}_{\text{cl}} = 0 \), \( \phi_{\text{cl}} = 0 \) for \( \pi/2 + O(\sqrt{\epsilon}) < \theta_H < \pi - O(\sqrt{\epsilon}) \), in which \( \bar{E}(\tilde{\delta}_{\text{cl}} = 0, \phi_{\text{cl}} = 0) = 0 \). Thus, from the energy conservation the total energy \( \bar{E}(\tilde{\delta}_{\text{cl}}, \phi_{\text{cl}}) \) in Eq. (14) becomes zero at any \( \bar{\tau} \) for \( T = 0 \). Since \( \sin(2\theta_c) \)-term (\( \sim \epsilon^{3/2} \)) is larger than the last term (\( \sim \epsilon^2 \)) by \( \sqrt{\epsilon} \) in Eq. (17), the first, second and third term should be of the order of \( \epsilon^{3/2} \). Since \( h_x \) is finite in the range of angle and \( \theta_0 \) is not close to 0 and \( \pi/2 \), the small value of \( \phi \) in the third term of Eq. (17) contributes to the path integral such that \( \phi \) should be of the order of \( \epsilon^{3/4} \). This fact is valid for any value of \( k_2 \) because \( h_x \) is not zero in this range of angle and plays the role of the effective transverse anisotropy component which is crucial for QTM. Therefore, from \( \bar{E}(\tilde{\delta}_{\text{cl}}, \phi_{\text{cl}}) = 0 \), the magnitude \( \phi_{\text{cl}} \) is approximately given by

\[
\phi_{\text{cl}}^2 = -\frac{2 \bar{E}_1(\delta_{\text{cl}})}{k_2 \sin^2(\theta_0 + \delta_{\text{cl}}) + h_x \sin(\theta_0 + \delta_{\text{cl}})}.
\] (18)

From Eq. (15) we get \( \bar{E}_1(\delta) \sim \sin(2\theta_c)(3\delta^2 \eta - \delta^3) \) because \( \cos(2\theta_c) \)-term is much smaller than \( \sin(2\theta_c) \)-term. Since \( \eta \simeq \sqrt{2\epsilon / 3} \), and the critical angle \( \theta_c \) and the angle \( \theta_0 \) of metastable state have values between 0 and \( \pi/2 \) in this range of angle, we get \( \phi_{\text{cl}} \sim -i \sqrt{M \cot \theta_c \epsilon^{3/4}} \), where the effective moment of inertia \( M \) is [11].
\[ M = \frac{\sin \theta_c}{h_x + k_2 \sin \theta_c}. \]  

(19)

Thus, the approximate formula of the classical Euclidean action is found to be \( S^c_E \sim i\hbar J \sin \theta_c \sqrt{\epsilon} \delta_{cl} \phi_{cl} \sim \hbar J \epsilon^{5/4} \sqrt{M} \sin \theta_c \cos \theta_c \) from the dynamical part of the first term in Eq. (17), where \( \delta_{cl} \sim O(1) \) and \( E(\delta_{cl}, \phi_{cl}) = 0 \). By using the critical field (11) and the critical angle (12), the classical action is approximately

\[ S^c_E \approx \hbar J \epsilon^{5/4} g(\theta_H), \]  

(20)

where

\[ g(\theta_H) = \frac{|\cot \theta_H|^{1/6}}{\sqrt{1 + \frac{k_2}{K_1}(1 + |\cot \theta_H|^{2/3})}}. \]  

(21)

In the case of \( \theta_H = \pi \), we have \( \theta_c = \theta_0 = \eta = 0 \) and \( 0 \leq \delta_{cl} \leq 2\sqrt{\epsilon} \) from Eq. (15). In this situation, the action (17) becomes

\[ S_E[\tilde{\delta}(\tilde{\tau}), \phi(\tilde{\tau})] = \hbar \epsilon^{-\frac{\alpha}{2}} \int_0^{\tilde{\tau}} d\tilde{\tau}' \left\{ \frac{i}{2} \epsilon^{\frac{\alpha + 2}{2}} \tilde{\delta}^2 \left( \frac{d\phi}{d\tilde{\tau}'} \right) + \frac{k_2}{2} \epsilon \tilde{\delta}^2 \sin^2 \phi + \frac{1}{2} \epsilon^2 (\tilde{\delta}^2 - \frac{\tilde{\delta}^4}{4}) \right\}. \]  

(22)

Since the last term in Eq. (22) is of the order of \( \epsilon^2 \), the order of magnitude of \( k_2 \sin^2 \phi \) in the second term should be \( \epsilon \). If \( k_2 \) becomes much smaller than \( \epsilon \), in which the magnetic particle possesses the uniaxial symmetry, [4,6] QTM is not observable. This is understood from the fact as follows. Consider the action (5) as the dynamical system in the Hamiltonian formulation which consists of the canonical coordinate \( \phi \) and \( p_\phi = 1 - \cos \theta \). The Poisson bracket \( \{p_\phi, H\} \) which determines the dynamics of the spin system with the Lagrangian (5) becomes zero for \( \theta_H = \pi \) because the Hamiltonian \( H \) becomes a function of only \( p_\phi \). Thus, \( \theta \) does not depend on time. [12] If the value of \( k_2 \) is much greater than \( \epsilon \), the small value of \( \phi \), i.e., \( O(\phi) \sim \epsilon^{1/2} \) mainly contributes to the path integral, in which \( \alpha \) is chosen to be 1. In this case, a comparison of \( k_2 \)-term with the last term in Eq. (22) determines \( \phi_{cl} \sim -i\sqrt{K_1 \epsilon/K_2} \), which leads to the least action \( S^c_E \sim i\hbar J \epsilon^{2} \delta_{cl} \phi_{cl} \sim \hbar J \epsilon^{3/2} \sqrt{K_1/K_2} \). The approximate forms of \( S^c_E \) which we have discussed for \( \pi/2 + O(\sqrt{\epsilon}) < \theta_H < \pi - O(\sqrt{\epsilon}) \) and \( \theta_H = \pi \) agree with the exact results in Ref. [3] up to the numerical factor \( N = 16 \times 6^{1/4}/5 \)
and 8/3, respectively. From above estimates we note that the dependence of the WKB exponent on $\theta_H$, $\epsilon$ and $K_2/K_1$ can be deduced from the simple dimensional analysis based on the energy conservation $\bar{E}(\bar{\delta}_{cl}, \phi_{cl}) = 0$ without solving the equation of motion from $\delta S_E = 0$. However, in order to obtain the dependence of the WKB exponent on $K_1$, $K_2$, $\epsilon$, and $\theta_H$ at finite temperature, we cannot make use of the dimensional estimate as above, because $\bar{E}(\bar{\delta}_{cl}, \phi_{cl}) \neq 0$. Thus, we need to calculate the bounce solution by numerically solving the equation of motion satisfied by the least trajectory.

IV. QTM AT FINITE TEMPERATURE

Consider the Euclidean action (17) with a periodic instanton for $\pi/2 + O(\sqrt{\epsilon}) < \theta_H < \pi - O(\sqrt{\epsilon})$ and $\theta_H = \pi$.

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

Since the $\sin(2\theta_c)$-term is much larger than $\cos(2\theta_c)$-term in Eq. (17), the action is simplified as

$$S_E[\bar{\delta}(\tau), \phi(\tau)] = hJ e^{-\frac{\alpha}{2}} \int_0^\bar{\beta} d\bar{\tau} \left\{ -i\epsilon \frac{\alpha + 1}{2} \sin(\theta_0 + \sqrt{\epsilon} \delta) \phi \left( \frac{d\delta}{d\bar{\tau}} \right) + \frac{k_2}{2} \sin^2 \phi \sin^2(\theta_0 + \sqrt{\epsilon} \delta) + 2h_x \sin(\theta_0 + \sqrt{\epsilon} \delta) \sin^2(\phi/2) + \frac{1}{4} \sin(2\theta_c) e^\frac{2}{\epsilon} (3\delta^2 \frac{\eta}{\sqrt{\epsilon}} - \delta^3) \right\},$$

(23)

where $h_x \neq 0$ and $k_2$ has any value, and we performed the integration by part for the first term. Also, the total time derivative is neglected because $\phi_{cl}(-\bar{\beta}/2) = \phi_{cl}(\bar{\beta}/2) = 0$ by noting that from the Euler-Lagrange equation $\phi_{cl}$ is proportional to $d\bar{\delta}_{cl}/d\bar{\tau}$ which is zero at $\bar{\tau} = \pm \bar{\beta}/2$ due to the periodicity of $\bar{\delta}_{cl}(\bar{\tau})$. As is well known for the crystal symmetry such as the form $K_z \alpha_z^2 - K_y \alpha_y^2$, the topological term $d\phi/d\tau$ in Eq. (5) plays important roles in the presence of a clockwise or counterclockwise winding over the barrier along the passage from $\phi = 0$ to $\phi = \pi$ in macroscopic quantum coherence (MQC). Since there is no such
topological situation in our quantum tunneling problem, the total derivative term does not contribute to the classical action. In the action \( \mathcal{A} \) the last term generates the potential barrier in the problem. Thus, the magnitude of each term should be the same order as that of the last term. Noting that the order of magnitude of each term is \( O(\epsilon^{(\alpha+1)/2} \phi) \), \( O(\sin^2 \phi) \), \( O(\sin^2(\phi/2)) \) and \( \epsilon^{3/2} \), respectively, the small values of \( \phi \) contribute to the path integral, and its order of magnitude is expected to be \( \epsilon^{3/4} \), as was discussed at \( T = 0 \). Therefore, an appropriate choice of \( \alpha \) is \( 1/2 \), which makes to each term to be of the order of \( \epsilon^{3/2} \) in the integrand.

Performing the Gaussian integration over \( \phi \) in the partition function \( \mathcal{Z} \) and the measure \( \mathcal{M} \), and introducing the variable \( \bar{\tau} = \tilde{\tau} \sqrt{\sin(2\theta_c)/M} \), the remaining integral is of the form

\[
\int D[\bar{\delta}(\bar{\tau})] \exp(-S_E^{\text{eff}}/\hbar), \tag{24}
\]

where \( \bar{\beta} = \tilde{\beta} \sqrt{\sin(2\theta_c)/M} = \epsilon^{1/4} \omega_0 \beta \hbar \sqrt{\sin(2\theta_c)/M} \) and the effective action is given by

\[
S_E^{\text{eff}}[\bar{\delta}(\bar{\tau})] = \hbar J \epsilon^{5/4} \sqrt{M \sin(2\theta_c)} \int_0^{\bar{\beta}} d\bar{\tau} \left[ \frac{1}{2} \left( \frac{d\bar{\delta}}{d\bar{\tau}} \right)^2 + \bar{U}(\bar{\delta}) \right], \tag{25}
\]

where \( \bar{U}(\bar{\delta}) = (\sqrt{6}\bar{\delta}^2 - \bar{\delta}^3)/4 \).

The classical trajectory \( \bar{\delta}_{\text{cl}}(\bar{\tau}) \) with period \( \bar{\beta} \) which minimizes the effective action satisfies

\[
\frac{d^2 \bar{\delta}_{\text{cl}}}{d\bar{\tau}^2} = \left[ \frac{d\bar{U}}{d\bar{\delta}} \right]_{\bar{\delta}=\bar{\delta}_{\text{cl}}}, \tag{26}
\]

and the corresponding total energy becomes

\[
\bar{E}_{\text{tot}}(\bar{\beta}) = \frac{1}{4}(\sqrt{6}\bar{\delta}^2 - \bar{\delta}^3) - \frac{1}{2} \left( \frac{d\bar{\delta}}{d\bar{\tau}} \right)^2. \tag{27}
\]

At zero temperature the classical trajectory becomes a regular bounce \( \bar{\delta}_{\text{cl}}(\bar{\tau}) = \sqrt{6}/\cosh^2[(3\bar{\beta})^{1/4}\bar{\tau}] \) with \( \bar{E}_{\text{tot}}(\bar{\beta} \to \infty) = 0 \). In order to include thermal corrections to quantum tunneling rate, we need to consider the periodic solutions which satisfy Eq. \( \mathcal{Z} \). In fact, there exist two kinds of periodic solution, \( \bar{\delta}_m(= 2\sqrt{6}/3) \) and the periodic motion \( \bar{\delta}_{\text{cl}}(\bar{\tau}) \) of the particle in the inverted potential with total energy \( \bar{E}_{\text{tot}} \), as is seen in Fig. 2. For the constant solution \( \bar{\delta}_m \), we obtain the classical action from Eq. \( \mathcal{Z} \).
where

$$U_m = \frac{8\sqrt{6}}{9} K_1 V e^{3/2} \left[ \frac{\left| \cot \theta_H \right|^{1/3}}{1 + \left| \cot \theta_H \right|^{2/3}} \right],$$

and the corresponding escape rate

$$\Gamma_0 \propto \exp(-S_0/\bar{h}) = \exp(-U_m/k_B T),$$

which is the Boltzmann formula representing a pure thermal activation. Let us define $T_c$ to be the temperature at which the periodic solution $\delta_{\text{cl}}(\bar{\tau})$ with period $\bar{\beta}$ approaches the limit $\delta_m$. Then, slightly below $T_c$, the thermal bounce $\delta_{\text{cl}}(\bar{\tau})$ reduces to small oscillations near the bottom of the inversed potential $-\bar{U}(\bar{\delta})$ shown in Fig. 2. In this case we take only the first Fourier harmonics for the thermal bounce because the next harmonics are smaller near $T_c$, $\delta_{\text{cl}}(\bar{\tau}) = \bar{\delta}_0(T) + \bar{\delta}_1(T) \cos(\bar{\omega}\bar{\tau}),$

with $\bar{\omega} = 2\pi/\bar{\beta}$. The temperature dependence of $\bar{\delta}_0$ and $\bar{\delta}_1$ is crucial for both the magnitude of $T_c$ and the dependence of $\delta_{\text{cl}}$ on $\bar{\tau}$. If $\bar{\delta}_1$ does not depend on $T$, $\delta_{\text{cl}}$ becomes a function of $\bar{\tau}$ through $\cos(\bar{\omega}_{\text{c}}\bar{\tau})$ at $T = T_c$, which is contrary to the fact that $\bar{\delta}_{\text{cl}}$ is a constant $\delta_m$ at $T_c$. Therefore, $\bar{\delta}_1$ should vanish at $T_c$ and have a finite value below $T_c$. In order to obtain $T_c$ and the bounce solution for $T \lesssim T_c$, we substitute the harmonic type solution (31) into the equation of motion for the thermal bounce, such as Eqs (34) or (48) subject to the situation under consideration. We then obtain the temperature dependence of $\bar{\delta}_0$ and $\bar{\delta}_1$ by taking the constant term and the coefficient of $\cos(\bar{\omega}\bar{\tau})$ to be zero, and the relation

$$\bar{\omega}_{\text{c}}(= 2\pi/\bar{\beta}_c) = -\bar{U}''(\bar{\delta}_m = 2\sqrt{6}/3)$$

which gives

$$k_B T_c = \frac{\hbar \bar{\omega}_0}{\pi} f(\epsilon, \theta_H),$$

where
\[ f(\epsilon, \theta_H) = \left( \frac{3}{8} \right)^{1/4} \epsilon^{1/4} \left[ \frac{\left| \cot \theta_H \right|^{1/6}}{1 + \left| \cot \theta_H \right|^{2/3}} \right] \sqrt{1 + \frac{K_2}{K_1} (1 + \left| \cot \theta_H \right|^{2/3}).} \]  

(33)

Since the thermal bounce (31) satisfies the equation of motion

\[ \frac{d^2 \delta}{d\bar{\tau}^2} - \frac{\sqrt{6}}{2} \delta + \frac{3}{4} \bar{\tau}^2 = 0, \]  

(34)

the temperature dependence of \( \bar{\delta}_0 \) and \( \bar{\delta}_1 \) is given by

\[ \bar{\delta}_0(T) = \frac{\sqrt{6}}{3} [1 + (\frac{T}{T_c})^2], \quad \bar{\delta}_1(T) = \frac{2\sqrt{3}}{3} \sqrt{1 - (\frac{T}{T_c})^4}, \]  

(35)

where at \( T = T_c \) \( \bar{\delta}(\bar{\tau}) = 2\sqrt{6}/3(= \bar{\delta}_m) \) by noting that \( \bar{\delta}_0(T_c) = 2\sqrt{6}/3 \) and \( \bar{\delta}_1(T_c) = 0 \). Substituting in Eq. (23) for the action, the approximate form of the minimal action can be written as

\[ \frac{S_{\text{E}}^{\text{min}}}{\hbar} \approx \frac{U_m}{k_B T} [1 - 3(\frac{T_c - T}{T_c})^2], \]  

(36)

It is important that for \( T \lesssim T_c \) the action \( S_{\text{E}}^{\text{min}} \) of the thermal bounce (31) is smaller than the action \( U_m/k_B T \) of the constant path \( \bar{\delta}_m \). Hence below \( T_c \), the functional integral for the decay is dominated by the thermal bounce. This fact implies that \( T_c \) is the crossover temperature from the quantum-mechanical to the thermally activated decay. Noting that the Boltzmann formula (30) is derived from the constant path \( \bar{\delta}_{\text{cl}}(\bar{\tau}) = \bar{\delta}_m \) and also valid above \( T_c \), the thermal bounce is degenerate into the constant trajectory for \( T \geq T_c \).

In order to obtain the WKB exponent in the entire range of temperatures less than \( T_c \), we employ two equivalent approaches for the numerical calculation of the least action. First, we use the equation of motion (34) for the numerical bounce solution, in which the action has been integrated over \( \phi \) with a periodic boundary condition \( \bar{\delta}_{\text{cl}}(\bar{\tau} = -\bar{\beta}/2) = \bar{\delta}_{\text{cl}}(\bar{\tau} = \bar{\beta}/2) \) where \( \bar{\beta} = 2\pi (2/3)^{1/4}(T_c/T) \). As illustrated in Fig. 3, note that numerical solutions become oscillatory formulas which are of the form (31) at temperatures slightly less than \( T_c \). When inserted into the action (23), the classical action can be obtained by performing the integration numerically. Second, numerically solve the coupled equation of motion of \( \bar{\delta}_{\text{cl}} \) and \( \phi_{\text{cl}} \) derived from the Euler-Lagrange equation for the action (23) by incorporating
the energy conservation $E(\bar{\delta}_{cl}, \phi_{cl}) = 0$. The second method is reduced to the first one if $|\phi|$ is small. However, since the first method is not valid unless $|\phi|$ is small enough to perform the Gaussian integration over $\phi$, the second method is a more general approach to obtain the WKB exponent. Although the first method is enough for the WKB exponent in our cases, the second method is useful for checking the results of the first. Thus, we used both approaches for the WKB exponent and found that the results obtained from the first methods are identical to the one from the second method.

In Fig. 4 we present the phase diagram of $\bar{\delta}$ and $\bar{\phi}(= \phi/\phi_0)$ where $\phi_0 = ig(\theta_H)\sqrt{2(1 + |\cot \theta_H|^2/3)}\epsilon^{3/4}$ in the range of temperature $0 \leq T < T_c$. Note that the orbit is oval for $0 < T << T_c$ and circular near $T_c$ with a center at $(2\sqrt{6}/3, 0)$. Substituting $\bar{\delta}_{cl}(\bar{\tau})$ and $\phi_{cl}$ into the action (23), we obtain the WKB exponent which is shown in Fig. 5. Note that the approximate form (36) is valid at the temperature close to $T_c$, and the WKB exponent is not sensitive to $T$ for temperatures between 0 and $T_c$.

We now discuss the effect of quantum fluctuation on the thermal activation at the temperature above $T_c$. At temperature slightly above $T_c$ quantum correction to the thermal activation becomes important. Its effect is due to the extension of the regular second-order action by terms up to the fourth order, i.e., non-Gaussian terms in the action functional. The quantum fluctuation, therefore, smears the transition in the crossover region $|T_c - T| \ll T_c$, in which the WKB exponent is of the form (36). Even at higher temperature where thermal activation prevails, the quantum effect is still incorporated into the preexponential factor of the thermal activation rate in the form [17]

$$\Gamma = \frac{\omega_p}{2\pi} C_q \exp(-\beta U_m)$$

where $\omega_p = \sqrt{U''(\delta = 0)}$ is the frequency of oscillation around the metastable minimum and $C_q$ a quantum mechanical correction. The escape rate is enhanced by quantum fluctuations. This correction has been studied in great detail [14,17]. Calculation of $C_p$ based on quantum fluctuation, the decay rate which includes the quantum effect in this region has the form

$$\Gamma = \frac{\omega_p}{2\pi} \prod_{n=1}^{\infty} \frac{\omega_n^2 + \omega_p^2}{\omega_n^2 - \omega_p^2} \exp(-\beta U_m),$$

(38)
where $\omega_n = 2\pi n/\beta \hbar$ and $\omega_b = \sqrt{-\ddot{U}'(\delta_m)}$ is the barrier frequency. This barrier frequency characterizes the width of the parabolic top of the barrier. Using the identity

$$\frac{1}{2} \sinh(\beta \hbar \omega / 2) = \frac{1}{\beta \hbar \omega} \prod_{n=1}^{\infty} \frac{\omega_n^2}{\omega^2 + \omega_n^2},$$

the decay rate (38) can be expressed as [17,18]

$$\Gamma = \left( \frac{\omega_b}{\pi} \frac{\sinh(\beta \hbar \omega_b / 2)}{\sin(\beta \hbar \omega_b / 2)} \right) \exp(-\beta U_m), \quad (40)$$

Since $\ddot{U}(0) = -\ddot{U}'(\delta_m)$ in this range of angles, we obtain $\omega_b = \omega_p = 2\omega_0 f(\epsilon, \theta_H)$. At temperatures slightly above the crossover region $T - T_c \ll T_c$, we expand $\sin(\beta \hbar \omega_b / 2)$ in Eq. (40) about $T_c$. Noting that $\beta_c \hbar = 2\pi$ from the crossover temperature (32), we have $\sin(\hbar \omega_b / 2k_B T) \approx \pi(T - T_c)/T + \cdots$ and from Eq. (40) the escape rate is given by

$$\Gamma \approx \frac{\omega_0}{\pi} f^2(\epsilon, \theta_H) \frac{T_c}{T - T_c} \sinh[\beta \hbar f(\epsilon, \theta_H) \omega_0] \exp(-\beta U_m). \quad (41)$$

At temperatures well below the pure thermal activation regime, but well beyond the crossover region $T - T_c \ll T_c$, we need to take the escape rate (40) as an exponential of a sum of logarithms and expand each logarithm in power of $\beta \hbar$. Then, we obtain the approximate form of the escape rate given by

$$\Gamma \approx \frac{\omega_0}{\pi} f^2(\epsilon, \theta_H) \left[ \frac{U_m}{k_B T} - \frac{f^2(\epsilon, \theta_H)}{3} \left( \frac{\hbar \omega_0}{k_B T} \right)^2 \right]. \quad (42)$$

As noted in Eq. (42), the quantum effect which is inversely proportional to $T^2$, is incorporated into the Boltzmann formula ($\beta U_m$) in the exponent which is indicative of quantum-mechanically assisted thermal activation process.

The expression (40) which includes the quantum fluctuation effects is valid in the entire thermal activation regime beyond the crossover region $T - T_c \ll T_c$. Its approximate forms (41) and (42) are valid slightly and well above the crossover temperature, respectively. For a pure thermal activation regime in which $\beta$ becomes much small, we obtain the approximate relation $\sinh(\beta \hbar \omega_p / 2) \approx \sin(\beta \hbar \omega_b / 2)$. In this case the escape rate is given by

$$\Gamma = \frac{\omega_0 f(\epsilon, \theta_H)}{\pi} \exp(-\beta U_m), \quad (43)$$
which is just the Boltzmann formula for the pure thermal activation process from Eq. (40).

Fig. 6 illustrates that we superpose the approximate expressions for the tunneling rates on the numerical results and show the region of the validity of the analytic results. Eq. (42) is applicable in a wide range of temperatures above $T_c$ but not appropriate as $T$ approaches the crossover region which includes $T_c$. Close to the crossover region, the expression (41) is valid in a narrow region. In the crossover region Eq. (41) is not appropriate because of singularity. In this case we need to extend the second-order action by terms up to the higher order in order to regularize the divergence, as previously noted, which leads to the WKB exponent to be of the form (33). When compared with the Boltzmann formula (43) based on the pure thermal activation, quantum fluctuations increase as $T$ becomes close to $T_c$ from the pure thermal regime. Also, note that while the tunneling rate at zero temperature is valid in a wide range of temperature below $T_c$, there exist an appreciable amount of thermal fluctuations for $T \gtrsim T_c$.

B. $\theta_H = \pi$

When the external magnetic field is opposite to the initial orientation, $h_x = \theta_0 = \theta_c = 0$ and the Euclidean action becomes Eq. (22). Estimating the order of magnitude of each term in the integrand of the action (22), $\alpha$ is chosen to be 1 and $\phi \sim O(\epsilon^{1/2})$ for the value of $k_2$ much larger than $\epsilon$, as previously discussed at $T = 0$. Performing the Gaussian integration over $\phi$, the effective action for the integral (24) is given by

$$S_E^{\text{eff}}[\delta(\bar{\tau})] = \frac{\hbar J \epsilon^{3/2}}{\sqrt{k_2}} \int_0^\beta d\bar{\tau} \left[ \frac{1}{2} \left( \frac{d\delta}{d\bar{\tau}} \right)^2 + \frac{1}{2} \left( \delta^2 - \frac{\delta^4}{4} \right) \right], \quad (44)$$

where we introduced $\bar{\tau} = \sqrt{k_2} \tau$ and $\bar{\beta} = \sqrt{k_2} \beta = \epsilon^{1/2} \omega_0 \beta \hbar \sqrt{K_2/K_1}$. At $T = 0$, the least trajectory for the action (14) is $\delta_{cl}(\bar{\tau}) = 2 / \cosh(\bar{\tau})$ with $\bar{E}_{tot} = \bar{U}(\delta_{cl}) - \frac{1}{2} (\frac{d\delta_{cl}}{d\bar{\tau}})^2 = 0$ where $\bar{U}(\delta) = (4\delta^2 - \delta^4)/8$.

As with subsection IV A, the height of barrier $U_m = K_1 V \epsilon^2$, and the crossover temperature are
\[ k_B T_c = \frac{\hbar \omega_0}{\pi} \sqrt{\frac{K_2}{2K_1}} \epsilon^{1/2}. \]  
(45)

For \( T \) slightly less than \( T_c \), the thermal bounce is represented as

\[ \delta_{cl}(\tau) = \sqrt{\frac{10}{5}} \sqrt{1 + 4\left(\frac{T}{T_c}\right)^2} + \frac{4}{\sqrt{15}} \sqrt{1 - \left(\frac{T}{T_c}\right)^2 \cos(\bar{\omega} \tau)}, \]
(46)

and its corresponding action

\[ \frac{S_{E}^{cl}}{\hbar} \approx \frac{U_m}{k_B T_c} \left[ 1 - \frac{32}{15} \left( \frac{T_c - T}{T_c} \right)^2 \right]. \]
(47)

In the entire temperature range less than the crossover temperature, the WKB exponent is found by using the numerical solution for the equation given by

\[ \frac{d^2 \delta}{d\tau^2} - \delta + \frac{\delta^3}{2} = 0, \]
(48)

with a periodic boundary condition with a period \( \beta = \frac{(2\pi/\sqrt{2})}{(T_c/T)}. \) Its result is similar to the one illustrated in Fig. 4. Using this solution, we obtain the WKB exponent whose form is the same as Fig. 4.

For the thermal activation regime beyond the crossover region \( T - T_c \ll T_c \), it is necessary to consider the escape rate (40) by including quantum fluctuation with the frequency \( \omega_p = \epsilon^{1/2} \omega_0 \sqrt{K_2/K_1} (= \omega_b/\sqrt{2}) \). Then, the rate is approximately given by

\[ \Gamma \approx \sqrt{\frac{2\omega_0}{\pi^2}} s(\epsilon) \frac{T_c}{T - T_c} \sinh[\beta \hbar s(\epsilon) \omega_0] \exp(-\beta U_m), \]
(49)

for the quantum correction regime slightly beyond the crossover region \( T - T_c \ll T_c \), and

\[ \Gamma \approx \frac{\omega_0}{\pi} s(\epsilon) \exp\left\{ -\left[ \frac{U_m}{k_B T} - \frac{s^2(\epsilon)}{2} \left( \frac{\hbar \omega_0}{k_B T} \right)^2 \right] \right\}, \]
(50)

for the temperature regime well beyond the crossover region, where \( s(\epsilon) = \epsilon^{1/2} \sqrt{K_2/2K_1}. \) The shape of \( \ln \Gamma(T)/\Gamma(0) \) is similar to the results shown in Fig. 3 at all temperatures.

V. DISCUSSION AND CONCLUSION

Table 4 summarizes the analytic results for the range of angles discussed. Note that the \( \epsilon \)-dependence of the height of barrier, the crossover temperature and the WKB exponent at
$T = 0$ are different for $\pi/2 + O(\sqrt{\epsilon}) < \theta_H < \pi - O(\sqrt{\epsilon})$ and $\theta_H = \pi$. The exponents of $\epsilon$ in $U_m$, $T_c$ and $B(T = 0)$ are equal to 1.5, 0.25 and 1.25 in a wide range of angles between $\pi/2$ and $\pi$, and increase up to a value of 2, 0.5 and 1.5 if the applied field goes toward the opposite direction of the initial magnetization ($\theta_H = \pi$), respectively. The properties of the former are characteristic of the quadratic-plus-cubic potential whose form is well-known in Josephson systems, and those of the latter are characteristic of the quadratic-plus-quartic potential. [20]

The exponent of $\epsilon$ in the WKB exponent is easily understood by noting that the WKB exponent is proportional to the height of barrier $U_m$ and inversely proportional to the width of the barrier characterized by $\omega_b$, i.e., $B \propto U/\hbar \omega_b$. To be specific, $B \propto \epsilon^{3/2-1/4}$ for $\pi/2 + O(\sqrt{\epsilon}) < \theta_H < \pi - O(\sqrt{\epsilon})$ and $B \propto \epsilon^{2-1/2}$ for $\theta_H = \pi$. Denoting the WKB exponent $B(T = 0)$ to be $c_B U_m/\hbar \omega_b$, $c_B$ is 36/5 for the range of angle $\pi/2 + O(\sqrt{\epsilon}) < \theta_H < \pi - O(\sqrt{\epsilon})$ and 32/3 for $\theta_H = \pi$, which are generally determined by the form of potential near liability.

The ratio of $T_c(\theta_H)$ to $T_c(135^\circ)$ is larger at $k_2 = 0$ than at finite $k_2$ for $\theta_H < 135^\circ$ and smaller for $\theta_H > 135^\circ$ (Fig. 7). The position of the maximum of $\bar{T}_c$ moves from $\theta_H \rightarrow 101^\circ$ at $k_2 \rightarrow 0$ (uniaxial symmetry) to $\theta_H \rightarrow 135^\circ$ at $k_2 \rightarrow \infty$, and its magnitude $\bar{T}_c$ becomes smaller as $k_2$ increases. This behavior is easily understood from Eq. (53) which gives a maximum value of the crossover temperature $T_c$ at

$$\theta_H^{\text{max}} = \pi - \arctan\left(\frac{K_2/K_1}{\sqrt{(K_2/K_1 + 0.5)^2 + 2 - 1.5}}\right)^{3/2},$$

as is illustrated in Fig. 8.

It is noted that the WKB exponent $B$ at zero temperature can have the maximum value, depending on the ratio of $K_2$ to $K_1$. For $K_1 \gg K_2$, there is no maximal point in $B$ at zero temperature. [4,5] Thus, there are some values of the ratio of the anisotropy constants $K_2/K_1$, at which the WKB exponent changes slightly in a wide range of angles. In the crossover region the quantum fluctuation is incorporated into the WKB exponent as a parabolic form with a slightly different curvature with respect to temperature in the range of angle $\pi/2 + O(\sqrt{\epsilon}) < \theta_H < \pi - O(\sqrt{\epsilon})$ and $\theta_H = \pi$, which leads to the smearing of the
WKB exponent, as is seen in Fig. 3. The quantum correction is also reflected in the thermal activation regime with different coefficient of $(T_c/T)^2$ well above the crossover temperature for $\pi/2 + O(\sqrt{\epsilon}) < \theta_H < \pi - O(\sqrt{\epsilon})$ and $\theta_H = \pi$.

To illustrate the above approximate results with concrete numbers we have collected in Table I and II various values for several materials, the example with $K_1 = K_2$, BaFe$_{12-2x}$Co$_x$Ti$_x$O$_{19}$($x = 0.8$), [21] Tb, [22] and Ni [23]. From Table III it is evident that the typical magnitude of $\epsilon$ for the magnetic system to tunnel out of the barrier within reasonable time is of the order of $10^{-3}$ or less, and that the associated crossover temperature $T_c$ is typically of the order of 100 mK or less. Noting that the inverse of the WKB exponent $B^{-1}$ is the magnetic viscosity $S$ studied by magnetic relaxation measurements, [24] the value of $S(T = 0)$ at the angle $\theta_H = 135^\circ$ are of order of 0.1-1 for the parameter $\epsilon$ with the magnitude $10^{-3} - 10^{-4}$. Also, the estimated values of $\theta_H^{\text{max}}$ and $T_c^{\text{max}}$ in Table I can be compared with the experimental measurements and their consistency will suggest the possibility of QTM in these particles.

In conclusion, we have studied thermal effects on quantum tunneling of magnetization placed in a magnetic field at an arbitrary angle. We have presented the analytic forms of quantum tunneling rates at several temperature regimes and performed the numerical calculations in the entire temperature regime. It is found that thermal corrections to quantum tunneling rate is small for $T \ll T_c$, but quite large for $T \sim T_c$. This is because the thermal activation process is strongly influenced by the quantum fluctuation in this regime. Furthermore, the dependence of $T_c$ and magnetic viscosity on the parameters $\theta_H$, $\epsilon$ and $K_2/K_1$ is expected to be observed in future experiments.

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Electronic address: gkim@phy.sejong.ac.kr

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[11] The effective moment of inertia is derived from the partition function (1), the measure (3) and the action (17) by performing the Gaussian integration over \( \phi \).

[12] Alternatively, if we consider \( M \) as a spin operator, the Hamiltonian obtained from Eq. (7) simply commutes with \( M_z \) in the absence of \( K_2 \) and \( H_x \), in which QTM is not expected.

[13] Care should be taken in case of \( k_2 = 0 \) for \( \theta_H = \pi \) because the terms except for the first and last one in Eq. (17) vanish. Since \( \phi_{cl} \) is not small in this situation, we need to solve the coupled equation of motion for \( \bar{\delta}_{cl} \) and \( \phi_{cl} \) from Euler-Lagrange equation. By using \( \bar{E}(\bar{\delta}_{cl}, \phi_{cl}) = 0 \), we get \( \bar{\delta}_{cl} = 0 \) and \( 2 \cdot \phi_{cl} \) can be any value for \( \bar{\delta}_{cl} = 0 \), but it does not contribute to the classical action. Since we obtain \( d\phi/d\tilde{\tau} = -i \epsilon \) for \( \bar{\delta}_{cl} = 2 \), \( S_{cl} \) becomes infinity. Thus, QTM is not observable.

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[19] In Fig. 6, we make use of the tunneling rate in the crossover region which is of the form

\[
\Gamma = \frac{\omega_b}{2\pi} A_0 \frac{\sinh(\beta \hbar \omega_0/2)}{\pi} \sqrt{\pi} \kappa \exp(-\kappa \varepsilon) \exp[(\kappa \varepsilon)^2 - \beta U_m],
\]

where \[ 17 \] \( \kappa^2 = \frac{18}{5} \beta U_m, \varepsilon \equiv 1 - T/T_c, \text{erfc}(x) = \frac{2}{\sqrt{\pi}} \int_x^\infty dt \exp(-t^2), \text{and} A_0 = 1 \text{ for} T \geq T_c \text{ and} T/T_c \text{ for} T \leq T_c. \]
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TABLE I. Summary of the results for the quantum tunneling in the range of angle \(\pi/2 + O(\sqrt{\epsilon}) < \theta_H < \pi - O(\sqrt{\epsilon})\) and \(\theta_H = \pi\). \(B(= S_{E}^{1/\hbar})\) is the WKB exponent at (a) zero temperature \((T = 0), [5]\) (b) crossover region \((T - T_{c} \ll T_{c})\), and (c) thermal activation region with quantum fluctuations \((T - T_{c} \gg T_{c})\). Here \(t = T/T_{c}\), \(m(\theta_H) = |\cot \theta_H|^{1/3}/(1 + |\cot \theta_H|^{2/3})\), \(g(\theta_H)\) is given by Eq. (21) and \(f(\epsilon, \theta_H)\) by Eq. (33).

| Form of potential near metastable point | \(\pi/2 + O(\sqrt{\epsilon}) < \theta_H < \pi - O(\sqrt{\epsilon})\) | \(\theta_H = \pi\) |
|----------------------------------------|------------------------------------------------|------------------|
| \(\alpha\) | \(1/2\) | 1 |
| \(U_{m}/K_{1}V\) | \(8\sqrt{6}/9\epsilon^{3/2}m(\theta_H)\) | \(\epsilon^{2}\) |
| \(\omega_{b}/\omega_{0}\) | \(2f(\epsilon, \theta_H)\) | \(\epsilon^{1/2}\sqrt{2K_{2}/K_{1}}\) |
| \(k_{B}T_{c}/(h\omega_{0}/\pi)\) | \(f(\epsilon, \theta_H)\) | \(\sqrt{K_{2}/2K_{1}}\epsilon^{1/2}\) |
| \(\tilde{\delta}_{0}(T)\) | \(\sqrt{6}/3(1 + t^{2})\) | \(\sqrt{6}/(1 + 4t^{2})^{1/2}\) |
| \(\tilde{\delta}_{1}(T)\) | \(2\sqrt{3}/3(1 - t^{2})^{1/2}\) | \(4/\sqrt{15}(1 - t^{2})^{1/2}\) |
| (a) \(B\) | \(16\times6^{1/4}JE^{5/4}g(\theta_H)\) | \(8\sqrt{3}J\epsilon^{3/2}\sqrt{K_{1}/K_{2}}\) |
| (b) \(B\) | \(U_{m}/k_{B}T[1 - 3(1 - t)^{2}]\) | \(U_{m}/k_{B}T[1 - 32/15(1 - t)^{2}]\) |
| (c) \(B\) | \(U_{m}/k_{B}T - \pi^{2}/4t^{2}\) | \(U_{m}/k_{B}T - \pi^{2}/4t^{2}\) |
TABLE II. Magnetization $M_0$, easy-axis anisotropy constant $K_1$, hard-axis anisotropy constant $K_2$, critical magnetic field $H_c$, and characteristic frequency $\omega_0$ for various materials, where in case of Ni the shape-induced contribution to the hard axis anisotropy is obtained for the slab geometry.

| Material | $M_0$ [emu/cm$^3$] | $K_1$ $10^5$[erg/cm$^3$] | $K_2$ $10^5$[erg/cm$^3$] | $V$ $10^3$[nm$^3$] | $H_c(135^\circ)$ [10$^3$Oe] | $H_c(180^\circ)$ [10$^3$Oe] | $\omega_0$ $10^{11}$/sec |
|----------|------------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| $K_1 = K_2$ | 500 | 100 | 100 | 1 | 20 | 40 | 7.04 |
| BaFe$_{12-2x}$Co$_x$Ti$_x$O$_{19}$ (x = 0.8) | 340 | 12 | $\sim$ 0 | 3.14 | 3.53 | 7.06 | 1.24 |
| Tb | $\sim$ 100 | $\sim$ 10 | $\sim 10^3$ | $\sim$ 1 | 10 | 20 | 3.54 |
| Ni | 508 | 8 | 16 | $\sim$ 1 | 1.575 | 3.15 | 0.557 |
TABLE III. Angle for the maximal WKB exponent $\theta_{H}^{B_{\text{max}}}$ and the maximal crossover temperature $\theta_{H}^{T_{\text{max}}}$, magnetic field parameter $\epsilon$, the height of barrier $U_m$ at $\theta_H = 135^\circ$ and $\theta_H = 180^\circ$, the WKB exponent $B(T = 0)$ at $\theta_H = \theta_{H}^{B_{\text{max}}}$ and $135^\circ$, the crossover temperature $T_c$ at $\theta_H = \theta_{H}^{T_{\text{max}}}$, and inverse tunneling rate $\Gamma^{-1}(T = 0)$ at $\theta_H = 135^\circ$ for various materials.

| Material | $\theta_{H}^{B_{\text{max}}}$ | $\theta_{H}^{T_{\text{max}}}$ | $\epsilon$ | $U_m(135^\circ)$ [K] | $U_m(180^\circ)$ [K] | $B_{\text{max}}$ | $B(135^\circ)$ | $T_c^{\text{max}}$ [mK] | $\Gamma^{-1}(0, 135^\circ)$ [sec] |
|----------|-------------------------------|-------------------------------|----------|---------------------|---------------------|----------------|--------------|-----------------|---------------------|
| $K_1 = K_2$ | 161 | 113 | $10^{-1}$ | $2.50 \times 10^4$ | 725 | $4.50 \times 10^3$ | $4.34 \times 10^3$ | 669 | $6.30 \times 10^{1870}$ |
| | | | $10^{-2}$ | 78.9 | 7.25 | 253 | 244 | 376 | $6.38 \times 10^{92}$ |
| | | | $10^{-3}$ | 2.50 | $7.25 \times 10^{-2}$ | 14.2 | 13.7 | 211 | $4.59 \times 10^{-7}$ |
| | | | $10^{-4}$ | 7.89 $\times 10^{-2}$ | $7.25 \times 10^{-4}$ | 0.799 | 0.772 | 119 | $8.35 \times 10^{-12}$ |
| Ba$_{x}$Fe$_{12-2x}$ & 180 | 101 | $10^{-1}$ | 940 | 273 | $\infty$ | 1.62 $\times 10^4$ | 76.9 | $1.73 \times 10^{7022}$ |
| Co$_x$Ti$_x$O$_{19}$ & | | | $10^{-2}$ | 29.7 | 2.73 | $\infty$ | 909 | 43.2 | $2.08 \times 10^{382}$ |
| (x = 0.8) & | | | $10^{-3}$ | 0.94 | $2.73 \times 10^{-2}$ | $\infty$ | 51.1 | 24.3 | $4.09 \times 10^{10}$ |
| | | | $10^{-4}$ | 2.97 $\times 10^{-2}$ | $2.73 \times 10^{-4}$ | $\infty$ | 2.88 | 13.7 | $3.50 \times 10^{-10}$ |
| Tb & 135 | 135 | $10^{-1}$ | 250 | 72.5 | 107 | 107 | 2.67 K | $4.18 \times 10^{32}$ |
| | | | $10^{-2}$ | 7.89 | 0.725 | 6.00 | 6.00 | 1.50 K | $4.29 \times 10^{-11}$ |
| | | | $10^{-3}$ | 0.25 | $7.25 \times 10^{-3}$ | 0.337 | 0.337 | 845 | 1.12 $\times 10^{-12}$ |
| | | | $10^{-4}$ | 7.89 $\times 10^{-3}$ | $7.25 \times 10^{-5}$ | $1.90 \times 10^{-2}$ | $1.90 \times 10^{-2}$ | 475 | $6.09 \times 10^{-12}$ |
| Ni & 151 | 120 | $10^{-1}$ | 200 | 58.0 | $3.48 \times 10^3$ | $3.44 \times 10^3$ | 67.0 | $9.46 \times 10^{480}$ |
| | | | $10^{-2}$ | 6.31 | 0.580 | 195 | 193 | 37.7 | $4.98 \times 10^{71}$ |
| | | | $10^{-3}$ | 0.2 | $5.80 \times 10^{-3}$ | 11.0 | 10.9 | 21.2 | $3.06 \times 10^{-7}$ |
| | | | $10^{-4}$ | 6.31 $\times 10^{-3}$ | $5.80 \times 10^{-5}$ | 0.617 | 0.612 | 11.9 | $7.83 \times 10^{-11}$ |
FIGURES

FIG. 1. $\eta = \theta_c - \theta_0$ as a function of $\theta_H$ for $\epsilon = (a) 0.01$ and (b) 0.001. Note that $\eta \approx \sqrt{2\epsilon}/3$ is valid in a wide range of angles, i.e., $\pi/2 + O(\sqrt{\epsilon}) < \theta_H < \pi - O(\sqrt{\epsilon})$.

FIG. 2. (a) The shape of the potential $\bar{U}(\delta)$ and (b) the shape of the inverted potential $-\bar{U}(\delta)$ for the range of angle $\pi/2 + O(\sqrt{\epsilon}) < \theta_H < \pi - O(\sqrt{\epsilon})$. In (a) the height of barrier $\bar{U}(\delta_m = 2\sqrt{6}/3)$ is $2\sqrt{6}/9$ at $\delta_m = 2\sqrt{6}/3$.

FIG. 3. The bounce solution obtained by numerically solving Eq. (34) for several temperatures $t = (a) 0.95$, (b) 0.98, (c) 0.99 and (d) 0.999, where $t = T/T_c$. The dashed curves in each temperature are drawn from the oscillatory solution (31) for comparison. Notice that as $T$ becomes close to $T_c$, the approximate formulas from Eq. (31) are compatible with numerical solutions.

FIG. 4. The phase diagram $\bar{\phi}$ vs. $\bar{\delta}$ at $t = (a) 0$, (b) 0.82, (c) 0.97, and (d) 0.999. Note that as $T$ increases from zero, the orbit is changed from an egg-shape to a circle.

FIG. 5. The temperature dependence of the magnetic viscosity, $S(T)(\equiv B(T)^{-1})$: $S(T)/S(0)$ versus $T/T_c$. Here the dotted line indicates the analytic formula (36) in the crossover temperature region, $|T - T_c| << T_c$.

FIG. 6. Plots of $\ln \Gamma/\Gamma(0)$ as a function of $T/T_c$, where $\Gamma(0)$ is the tunneling rate at $T = 0$. (a) the numerical calculation in the entire temperature, (b) Eq. (43) for the pure thermal activation regime, (c) Eq. (42) for the quantum corrections, (d) Eq. (41) for the quantum corrections close to the crossover region, and (e) the rate in the crossover region whose exponent is of the form (36).

FIG. 7. The $\theta_H$ dependence of the scaled crossover temperature $\bar{\theta}_c[= T_c(\theta_H)/T_c(135^\circ)]$ at (a) $k_2 = 0$, (b) 1.0, (c) 2.0, and (d) 100.

FIG. 8. The dependence of $\theta_H^{\max}$ on $k_2$ which is derived from Eq. (51) where $k_2$ is defined in Eq. (8).
\( \Delta \)

\( \bar{\delta} \)
\( \ln \left[ \frac{\Gamma}{\Gamma(0)} \right] \) vs. \( \frac{T}{T_c} \)

(a) 
(b) 
(c) 
(d) 
(e)
