Magnetic Field Dependence of Macroscopic Quantum Tunneling and Coherence of Ferromagnetic Particle

Gwang-Hee Kim\textsuperscript{a} and Dae Sung Hwang\textsuperscript{b}

\textit{Department of Physics, Sejong University, Seoul 143-747, Korea}

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Abstract

We calculate the quantum tunneling rate of a ferromagnetic particle of \(\sim 100\,\text{Å}\) diameter in a magnetic field of arbitrary angle. We consider the magnetocrystalline anisotropy with the biaxial symmetry and that with the tetragonal symmetry. Using the spin-coherent-state path integral, we obtain approximate analytic formulas of the tunneling rates in the small \(\epsilon (= 1 - H/H_c)\)-limit for the magnetic field normal to the easy axis \((\theta_H = \pi/2)\), for the field opposite to the initial easy axis \((\theta_H = \pi)\), and for the field at an angle between these two orientations \((\pi/2 \ll \theta_H \ll \pi)\). In addition, we obtain numerically the tunneling rates for the biaxial symmetry in the full range of the angle \(\theta_H\) of the magnetic field \((\pi/2 < \theta_H \leq \pi)\), for the values of \(\epsilon = 0.01\) and 0.001.

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\textsuperscript{a} e-mail: gkim@phy.sejong.ac.kr

\textsuperscript{b} e-mail: dshwang@phy.sejong.ac.kr

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A quest to understand the macroscopic quantum tunneling of ferromagnetic particles has been an important topical issue of intensive theoretical \[1,2\] and experimental studies. \[3\] Since the magnitude of the total magnetization $\vec{M}$ is frozen up at sufficiently low temperature, the direction $\hat{M}$ of the total magnetization becomes the only dynamical variable. In the absence of an external magnetic field, this direction is subject to the magnetocrystalline anisotropy which possesses its easy directions depending on the crystal symmetry. In this situation, $\hat{M}$ has at least two equivalent stable orientations which leads to the oscillation of $\hat{M}$ between them. This phenomenon is called macroscopic quantum coherence (MQC). \[1\]-\[3\] Simple analysis shows that in this case the height of barrier is too high to observe MQC in experiments. However, by applying a magnetic field $\vec{H}$ along the direction which lies halfway between two equivalent stable orientations, we can control the height and the width of barrier and make MQC observable. On the other hand, if we apply a magnetic field in a proper direction, the symmetry of two easy directions is broken. Then, one of the two stable orientations becomes metastable. In this situation we can obtain the optimal condition for the observation of tunneling, which is called macroscopic quantum tunneling (MQT). \[1\]-\[3\] The ferromagnetic particle is typically a single domain with as many as $10^5 - 10^6$ magnetic moments, which is a macroscopic number of particles. During the dynamical process we can not neglect the influence of the environment such as phonons, \[4\] nuclear spins, \[5\] and Stoner excitations and eddy currents in metallic magnets. \[6\] Even though some of these couplings are noticable, it has been reported \[1\]-\[3\] that they are not strong enough to make MQC or MQT unobservable. A few experimental attempts \[3\] have been made to observe the MQC or MQT of a large single ferromagnetic particle or a collection of magnetic particles. At present, it is not easy to perform a direct comparison between theoretical and experimental results because of stochastic behaviors of the systems.

Recently, Zaslavskii \[7\] studied the uniaxial anisotropy which is the simplest case, mapped the spin problem onto a one-dimensional particle system, and then obtained the tunneling
exponent, the preexponential factor and their temperature dependences in the low barrier limit with a magnetic field applied at some angle to the anisotropy axis. Later, Miguel and Chudnovksy [8] performed the calculation based on the imaginary time path integral method, and demonstrated that their result of the tunneling exponent in MQT coincides with the Zaslavskii’s result in the uniaxial symmetry, for its dependence on the direction and magnitude of the applied magnetic field. They discussed the tunneling rates at finite temperature and suggested the experimental procedures whose results can be compared with their theoretical results. In this paper, we extend the above calculations to the biaxial and tetragonal symmetries [9] by applying the instanton technique. Since the result of the biaxial symmetry is a generalization of that of the uniaxial symmetry studied by Zaslavskii, and by Miguel and Chudnovsky, we can compare our results with theirs by taking the vanishing limit of the transverse anisotropy constant. Also, we will explain that MQC and MQT can be consecutively observed by changing the direction of $\vec{H}$, and discuss their dependence on the direction and the magnitude of $\vec{H}$.

This paper is organized as follows. In Sec. II, we briefly discuss the theory of MQT and MQC in a ferromagnetic particle based on the standard instanton method. In Sec. III and IV, we consider the tunneling rate for biaxial and tetragonal symmetry in a magnetic field with a range of angles $\pi/2 \leq \theta_H \leq \pi$. We derive approximate formulas of the tunneling rates in the small $\epsilon$-limit for three angle ranges ($\theta_H = \pi/2$, $\pi/2 \ll \theta_H \ll \pi$, $\theta_H = \pi$), and present the $\theta_H$-dependence of the WKB exponent in the full range of angles ($\pi/2 < \theta_H \leq \pi$). The conclusions are given in Sec. V.

II. THE MQT AND MQC IN A FERROMAGNETIC SYSTEM

The tunneling rate $\Gamma$ of a ferromagnetic particle escaping from a metastable state in MQT or the oscillation rate $\Delta$ between double wells in MQC has the relation

$$(\Gamma \text{ or } \Delta) \propto \exp(-B). \quad (1)$$
The WKB exponent $B$ in Eq. (1) is approximately given by $U/\hbar\omega_b$, where $U$ is the height of barrier. The barrier frequency $\omega_b$ is the frequency of small oscillations around the minimum of the inverted potential, and characterizes the width of the barrier hindering the decay process. $\omega_b^2$ is inversely proportional to the effective mass of the magnetic particle, where the mass is induced by the transverse component of the magnetic field or by the transverse magnetic anisotropy constant. Therefore, it is necessary that the magnetic particle has a transverse magnetic anisotropy or we apply a transverse magnetic field, in order that MQT or MQC of the magnetic particle is possible. For calculating $\Gamma$ or $\Delta$ in Eq. (1), we introduce the angles $\theta$ and $\phi$ for the direction of $\vec{M}$ in the spherical coordinate system and employ the instanton method, in which the Euclidean action is written as

$$S[\theta(\tau), \phi(\tau)] = V \int [iM_0 \frac{1}{\gamma} (1 - \cos \theta) \frac{d\phi(\tau)}{d\tau} + E(\theta(\tau), \phi(\tau))]d\tau,$$

(2)

where $V$ is the volume of the magnetic particle, $\gamma = g\mu_B/\hbar$, $\mu_B$ the Bohr magneton and $M_0$ the magnitude of the magnetization. The first term in Eq. (2) is the topological Wess-Zumino term $[10]$, and the second term is the energy density which is composed of the magnetic anisotropy energy and the potential energy given by external magnetic field.

The action (2) produces from $\delta S = 0$ the classical equation of motion for $\vec{M}$ called the Gilbert equation $[11]$, which in the Euclidean space is written as

$$i\frac{d\vec{M}}{d\tau} = -\gamma \vec{M} \times \frac{dE}{d\vec{M}}.$$

(3)

We note that the action (2) describes the $1 \oplus 1$ dimensional dynamics in the Hamiltonian formulation, which consists of the canonical coordinates $\phi$ and $p_\phi = 1 - \cos \theta$. $[12][13]$

In the standard instanton method, the tunneling or the oscillation rate is given by the formula

$$\begin{cases} \Gamma \\ \Delta \end{cases} = C_0 \sqrt{\frac{S_{cl}}{2\pi\hbar}} \begin{cases} \omega_t \\ \omega_c \end{cases} \exp(-\frac{S_{cl}}{\hbar}),$$

(4)

where $S_{cl}$ is the classical action evaluated by using the solution of the equation of motion derived from $\delta S = 0$. In Eq. (3) $C_0$ is the preexponential factor which is originated from
the $\delta^2 S$ term, $\omega_t$ a precession frequency in MQT and $\omega_c$ an oscillation frequency in MQC, where $\omega_t$ and $\omega_c$ are of the order of the barrier frequency $\omega_b$.

III. TUNNELING RATE FOR BIAXIAL SYMMETRY

In this section we study a system which has the biaxial symmetry, with $\pm \hat{z}$ axes being the easy axes of the Hamiltonian. As in Ref. [8], we apply the magnetic field in the $xz$-plane and take the $\hat{z}$ axis as the initial easy axis when there is no magnetic field. Then the total energy $E(\theta, \phi)$ of the system is given by

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

where $K_1$ and $K_2$ are the parallel and transverse anisotropy constants, respectively, and $E_0$ is a constant which makes $E(\theta, \phi)$ zero at the initial orientation. As will be shown later, the effective mass of the magnetic particle is inversely proportional to a linear combination of $K_2$ and transverse magnetic field $H_x$ while there is no exact analog of the kinetic energy in the action (2). Denoting $\theta_H$ to be the angle between the magnetic field and $z$-axis, we obtain MQC for $\theta_H = \pi/2$ and MQT for $\pi/2 < \theta_H \leq \pi$ with large possible tunneling rates depending on the magnitude of the applied magnetic field.

It is convenient to introduce the dimensionless parameters

$$\bar{K}_2 \equiv K_2/2K_1, \quad \bar{H}_x \equiv H_x/H_0, \quad \bar{H}_z \equiv H_z/H_0,$$

where $H_0 \equiv 2K_1/M_0$. Then the total energy (3) is written as

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

where $\bar{E}(\theta, \phi) = E(\theta, \phi)/2K_1$. The plane given by $\phi = 0$ is the easy plane, on which $\bar{E}(\theta, \phi)$ is given by

$$\bar{E}(\theta, 0) = \frac{1}{2} \sin^2 \theta - \bar{H} \cos(\theta - \theta_H) + \bar{E}_0,$$
where $H_x = \bar{H} \sin \theta_H$ and $H_z = \bar{H} \cos \theta_H$ since $\bar{H}$ is in $xz$-plane. We define $\theta_0$ to be the initial angle and $\theta_c$ the angle at which the barrier vanishes by the applied critical magnetic field. From Eq. (7) we get the following conditions which $\theta_0$ and $\theta_c$ should satisfy,

\[
\sin \theta_0 \cos \theta_0 + \bar{H} \sin(\theta_0 - \theta_H) = 0,
\]

\[
\sin \theta_c \cos \theta_c + \bar{H}_c \sin(\theta_c - \theta_H) = 0,
\]

\[
\cos(2\theta_c) + \bar{H}_c \cos(\theta_c - \theta_H) = 0,
\]

where $\bar{H}_c$ is the dimensionless critical field with which the barrier just vanishes. From Eqs. (10) and (11) the critical field $\bar{H}_c$ is given by

\[
\bar{H}_c = \frac{1}{\sin^{2/3} \theta_H + | \cos \theta_H |^{2/3}}.
\]

From Eqs. (10)-(12) $\theta_c$ is given by

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

\[
\cos(2\theta_c) = 1 - \frac{2}{1 + |\cot \theta_H|^{2/3}}.
\]

For the special cases $\theta_H = \pi/2, \, 3\pi/4, \, \pi$, we have $\theta_c = \pi/2, \, \pi/4, \, 0$, respectively.

The practically interesting situation is when the barrier height is low and the width is narrow in order to have the tunneling rate large. Such a situation is realized when the value of $\epsilon \equiv 1 - \bar{H}/\bar{H}_c$ is small. For the small value of $\epsilon$, Eq. (12) becomes

\[
\sin(2\theta_c)(\epsilon - \frac{3}{2} \eta^2) - \eta \cos(2\theta_c)(2\epsilon - \eta^2) = 0,
\]

where $\eta = \theta_c - \theta_0$ which is expected to be small for $\epsilon \ll 1$. As will be seen later, detailed calculations show that $\eta$ is of the order of $\sqrt{\epsilon}$. Then 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 to have $\eta \simeq 2\sqrt{\epsilon}/3$, except for the $\theta_H$ values near $\theta_H = \pi/2$ and $\pi$. However, when $\theta_H$ is very close to $\pi/2$ or $\pi$, $\sin(2\theta_c)$ becomes almost zero as shown in Fig. 1, and the first term is much smaller than the second term in Eq. (13). Then the value of $\eta$ is obtained from the second term when $\theta_H \simeq \pi/2$ or $\pi$, and $\eta$ is given by $\eta \simeq \sqrt{2\epsilon}$ for $\theta_H \simeq \pi/2$ and
\[ \eta \simeq 0 \text{ for } \theta_H \simeq \pi \text{ from the detailed analysis of Eqs. (9)-(11).} \] After a little manipulation, we obtain from Eq. (7) an approximate formula of \( \bar{E}(\theta, \phi) \) in the limit of small \( \epsilon \) given by

\[ \bar{E}(\delta, \phi) = \bar{K}_2 \sin^2 \phi \sin^2(\theta_0 + \delta) + \bar{H}_x \sin(\theta_0 + \delta)(1 - \cos \phi) + \bar{E}_1(\delta), \quad (16) \]

where \( \bar{E}(\theta, \phi) \) is written as \( \bar{E}(\delta, \phi) \) by introducing a small variable \( \delta \equiv \theta - \theta_0 \), and \( \bar{E}_1(\delta) \) is a function of only \( \delta \) given by

\[ \bar{E}_1(\delta) = \frac{1}{4} \sin(2\theta_c)(3\delta^2 \eta - \delta^3) + \frac{1}{2} \cos(2\theta_c)[\delta^2(\epsilon - \frac{3}{2} \eta^2) + \delta^3 \eta - \frac{\delta^4}{4}]. \quad (17) \]

As previously discussed for \( \eta \), even though the \( \cos(2\theta_c) \) term in Eq. (17) looks smaller by a factor of \( \eta \) which is of the order of \( \sqrt{\epsilon} \), the second term cannot be neglected near \( \theta_H = \pi/2 \) and \( \pi \) because \( \sin(2\theta_c) \) is almost zero for these regions of \( \theta_H \). When we assume that \( |\phi| \ll 1 \), from the energy conservation \( \bar{E}(\delta, \phi) = 0 \) in Eq. (16), \( \phi \) is approximately given by

\[ \phi^2 = -\frac{\bar{E}_1(\delta)}{\bar{K}_2 \sin^2 (\theta_0 + \delta) + \frac{1}{2} \bar{H}_x \sin (\theta_0 + \delta)}. \quad (18) \]

Since \( \delta \) is of the order of \( \sqrt{\epsilon} \) as can be seen from Eq. (17), \( \bar{E}_1(\delta) \) is \( O(\epsilon^{3/2}) \) or less. Therefore, \( \phi \) is very small from Eq. (18) whose validity is for the full range of angles \( \pi/2 \leq \theta_H \leq \pi \) in biaxial symmetry. However, in case of tetragonal symmetry the magnitude of \( \phi \) is not small about \( \theta_H = \pi \), as can be seen in Eq. (23). In such a situation it is not possible to expand \( \bar{E}(\delta, \phi) \) as powers of \( \phi \) and to reduce the classical equation to the one-dimensional equation like Eq. (23).

Since we confirmed that \( |\phi| \) is very small in case of the biaxial symmetry which we study in this section, we have the approximate formula of \( \bar{E}(\delta, \phi) \) in Eq. (16) given by

\[ \bar{E}(\delta, \phi) = [\bar{K}_2 \sin^2 (\theta_0 + \delta) + \frac{1}{2} \bar{H}_x \sin (\theta_0 + \delta)]\phi^2 + \bar{E}_1(\delta). \quad (19) \]

By introducing a new scaled time variable \( \bar{\tau} \equiv \omega_0 \tau \) with \( \omega_0 \equiv 2\gamma K_1/M_0 \), the Euclidean action (2) becomes

\[ S[\delta(\bar{\tau}), \phi(\bar{\tau})] = \hbar J \int \{i[1 - \cos(\theta_0 + \delta) \frac{d\phi(\bar{\tau})}{d\bar{\tau}}] + \bar{E}(\delta(\bar{\tau}), \phi(\bar{\tau}))\} d\bar{\tau}, \quad (20) \]
where $J \equiv VM_0/h\gamma$. In the following we also use the scaled angular frequencies $\bar{\omega}_i$ defined by $\bar{\omega}_i \equiv \omega_i/\omega_0$ ($i = b, c, t$).

There are three approaches for the calculation of $S_{cl}$ from the action (20). Firstly, if $|\phi|$ is small, we can perform a Gaussian integration over the variable $\phi$ in the path integral and reduce the system to that with only one variable $\delta$. Then it is possible to do the rest of the calculation by using the standard instanton method. However, if $|\phi|$ is not small like in the tetragonal symmetry case with $\theta_H = \pi$, the integrand of Eq. (20) can not be reduced to the form which allows the Gaussian integration over $\phi$. Secondly, we can use the fact that $\bar{E}(\delta_{cl}, \phi_{cl}) = 0$ and that $(d\phi/d\delta)d\delta$ can be substituted for $(d\phi/d\bar{\tau})d\bar{\tau}$ in the Euclidean action (20). Here, we do not need to know the explicit solutions of the classical paths $\delta_{cl}(\bar{\tau})$ and $\phi_{cl}(\bar{\tau})$ for the classical action $S_{cl}$ because we can obtain $d\phi/d\delta = (d\phi/d\bar{\tau})/(d\delta/d\bar{\tau})$ from the Euler-Lagrange equations given by Eq. (21) and (22) below. However, this approach can not give the value of the preexponential factor, for which we need the explicit solution of the bounce in MQC or the instanton in MQT as a function of $\bar{\tau}$. Thirdly, we directly solve the coupled equations of motion for $\delta$ and $\phi$ given by Eqs. (21) and (22) by incorporating $\bar{E}(\delta_{cl}, \phi_{cl}) = 0$. Even though it is hard and sometimes rather tedious to obtain the solution as a function of $\bar{\tau}$ from Eqs. (21) and (22), and $\bar{E}(\delta_{cl}, \phi_{cl}) = 0$, this methods provides a complete result for the tunneling rate $\Gamma$ in MQT or the oscillation frequency $\Delta$ in MQC. In this approach, the preexponential factor can be obtained from the second variational term $\delta^2 S$ of the action in the spin-coherent-path integral, and the classical action $S_{cl}$ is obtained by the direct integration of Eq. (20) over $\bar{\tau}$ with the explicit solution $\delta_{cl}(\bar{\tau})$ and $\phi_{cl}(\bar{\tau})$. While the third method is the most general approach to obtain the tunneling rate, it is reduced to the first one if $|\phi|$ is small enough. Also, the second method is useful for checking the result of the first or the third method for the WKB exponent. In biaxial symmetry, due to $|\phi| \ll 1$ we apply the first method to obtain the explicit instanton or bounce solution as a function of $\bar{\tau}$, which is used to calculate the WKB exponent and the preexponential factor. On the other hand, since the magnitude of $\phi$ is not small in tetragonal symmetry case with $\theta_H = \pi$, we need to make use of the third method for the calculation of $\Gamma$. In addition, by
using the second method we check the results of the WKB exponent from the first or third method for each case.

The classical trajectory of $\delta$ and $\phi$ is determined by the Euler-Lagrange equation derived from the action (21),

$$i \sin(\theta_0 + \delta) \frac{d\phi}{d\bar{\tau}} = -\frac{\partial \bar{E}(\delta, \phi)}{\partial \delta},$$

$$i \sin(\theta_0 + \delta) \frac{d\delta}{d\bar{\tau}} = \frac{\partial \bar{E}(\delta, \phi)}{\partial \phi}.$$  \hfill (22)

From Eqs. (19), (21) and (22), the equation of motion for the instanton $\delta_{cl}(\bar{\tau})$ in the biaxial symmetry case becomes

$$\frac{d\delta_{cl}}{d\bar{\tau}} = \sqrt{\frac{2}{M}} \bar{E}_1(\delta_{cl}),$$

where the effective mass is given by

$$M = \left[ \frac{\bar{H}_x}{\sin(\theta_0 + \delta_{cl})} + 2\bar{K}_2 \right]^{-1}.$$  \hfill (24)

Now let us study the tunneling rates at three different magnetic field directions of $\theta_H = \pi/2$, $\pi$, and $\pi/2 \ll \theta_H \ll \pi$.

**A. $\theta_H = \pi/2$**

For $\theta_H = \pi/2$, $\theta_c = \pi/2$ from Eqs. (13) and (14), and $\eta = \sqrt{2e}$ as explained below Eq. (15). Then, from Eqs. (17) and (24) we get the approximate forms of the effective potential energy $\bar{E}_1(\delta)$ and the effective mass $M$ in the reduced one dimension as

$$\bar{E}_1(\delta) = \frac{1}{2} \delta^2 \left( \frac{\delta}{2} - \sqrt{2e} \right)^2,$$

$$M = (\bar{H}_x + 2\bar{K}_2)^{-1},$$

where $\bar{H}_x \approx \bar{H}_c$. Here we notice that MQC can not be generated without an external field along the $x$-axis or a transverse anisotropy constant $K_2$. Eq. (23) with Eqs. (25) and (26) has the solution called the bounce which is given by
\[ \delta_{cl}(\bar{\tau}) = \sqrt{2\epsilon}[1 + \tanh(\bar{\omega}_c \bar{\tau})], \]  

(27)

where \( \bar{\omega}_c = \sqrt{\epsilon/2\sqrt{1 + K_2/K_1}}. \) Then from Eq. (18) we get the classical path for \( \phi \) given by

\[ \phi_{cl}(\bar{\tau}) = -i\epsilon \frac{1}{\sqrt{1 + \frac{K_2}{K_1}} \cosh^2(\bar{\omega}_c \bar{\tau})}. \]  

(28)

We can calculate the classical action by integrating Eq. (20) with the above classical trajectory, and the result is given by

\[ S_{cl} = (\hbar J) \frac{4\sqrt{2}}{3} \frac{\epsilon^{3/2}}{\sqrt{1 + \frac{K_2}{K_1}}}. \]  

(29)

From Eq. (23) and Fig. 2, we get the height of barrier as \( E_1(= U/2K_1V) = \epsilon^2/2 \) at \( \delta_m = \sqrt{2\epsilon} \) and the oscillation frequency around the minimum of the inverted potential \( -\bar{E}_1(\delta) \) as \( \bar{\omega}_b(\equiv \sqrt{-\bar{E}_1''(\delta_m)}/M) = \sqrt{2} \bar{\omega}_c. \) Then, as mentioned in Sec. I, the WKB exponent \( B(= S_{cl}/\hbar) \) is approximately given by

\[ B \sim \frac{U}{\hbar \omega_b} = \frac{J}{2} \frac{\epsilon^{3/2}}{\sqrt{1 + \frac{K_2}{K_1}}}, \]  

(30)

which agrees up to the numerical factor with the result in Eq. (29) obtained by using the explicit instanton solution.

**B. \pi/2 \ll \theta_H \ll \pi**

For \( \pi/2 \ll \theta_H \ll \pi \), the critical angle \( \theta_c \) is in the range of \( 0 \ll \theta_c \ll \pi/2 \) and \( \eta \simeq \sqrt{2\epsilon/3}. \) Then from Eqs. (13) and (14) we get

\[ \bar{E}_1(\delta) = \frac{1}{4} \sin(2\theta_c)(\sqrt{6\epsilon \delta^2 - \delta^2}), \]  

(31)

\[ M = \left[ \frac{\bar{H}_x}{\sin \theta_c} + 2\bar{K}_2 \right]^{-1}, \]  

(32)

where \( \bar{H}_x = \bar{H}_c \sin \theta_H. \) The classical equation of motion Eq. (28) gives the instanton solution as
\[ \delta_{cl}(\bar{\tau}) = \frac{\sqrt{6\epsilon}}{\cosh^2(\bar{\omega}_t \bar{\tau})}, \quad (33) \]
\[ \phi_{cl}(\bar{\tau}) = -i(6\epsilon)^{3/4} |\cot \theta_H|^{1/6} \frac{\sinh(\bar{\omega}_t \bar{\tau})}{\cosh^4(\bar{\omega}_t \bar{\tau})} \left[ 1 + \frac{K_2}{K_1} \right]^{-1/2}, \quad (34) \]

where Eq. (18) has been used to get \( \phi_{cl}(\bar{\tau}) \), and the dimensionless frequency \( \bar{\omega}_t \) is given by
\[ \bar{\omega}_t = \left( \frac{3}{8} \right)^{1/4} \epsilon^{1/4} \left| \cot \theta_H \right|^{1/6} \left[ 1 + \frac{K_2}{K_1} \right] \left( 1 + |\cot \theta_H|^{2/3} \right)^{-1/2}. \quad (35) \]

Then the classical action is found to be
\[ S_{cl} = (\hbar J) \frac{16 \times 6^{1/4}}{5} \epsilon^{5/4} \frac{\left| \cot \theta_H \right|^{1/6}}{\sqrt{1 + \frac{K_2}{K_1} \left( 1 + |\cot \theta_H|^{2/3} \right)}}. \quad (36) \]

It is noted that Eq. (36) without \( K_2 \) agrees with the classical action in Ref. \[8\] which studied the uniaxial symmetry case. By using \( \bar{E}_1(\delta_m) = 2\sqrt{6\epsilon}/3 = \sin(2\theta_c)(6\epsilon)^{3/2}/27 \) and \( \bar{\omega}_b = \sqrt{-\bar{E}_1''(\delta_m)/M} = 2\bar{\omega}_t \) given from Eq. (31) and Fig. 2, we approximately obtain \( B \) as
\[ \frac{U}{\hbar \omega_b} = 4 \times 6^{1/4} \frac{\epsilon^{5/4}}{9} \frac{\left| \cot \theta_H \right|^{1/6}}{\sqrt{1 + \frac{K_2}{K_1} \left( 1 + |\cot \theta_H|^{2/3} \right)}}, \quad (37) \]

which is consistent with Eq. (36) up to the numerical factor.

**C. \( \theta_H = \pi \)**

In case of \( \theta_H = \pi \), we had \( \theta_c = 0 \) and \( \eta = 0 \). Then we have
\[ \bar{E}_1(\delta) = \frac{1}{2}(\epsilon \delta^2 - \delta^4), \quad (38) \]
\[ M = (2K_2)^{-1} \quad (39) \]

which gives the classical path obtained from Eq. (23) as
\[ \delta_{cl}(\bar{\tau}) = \frac{2\sqrt{\epsilon}}{\cosh(\bar{\omega}_t \bar{\tau})}, \quad (40) \]
\[ \phi_{cl}(\bar{\tau}) = -i \sqrt{\frac{K_1 \epsilon}{K_2}} \tanh(\bar{\omega}_t \bar{\tau}) + n\pi, \quad (41) \]
where \( n = 0 \) or \( 1 \), and \( \bar{\omega}_t = \sqrt{K_2 \epsilon/K_1} \). Here we note that \( \phi_{cl}(\bar{\tau}) \) is obtained from the conservation of energy \( \bar{E}(\delta_{cl}, \phi_{cl}) = 0 \). Then the corresponding classical action is given by

\[
S_{cl} = (\hbar J) \frac{8}{3} \sqrt{\frac{K_1}{K_2}} \epsilon^{3/2}. \tag{42}
\]

We note again that

\[
B \sim \frac{U}{\hbar \omega_b} = \frac{J}{4} \sqrt{\frac{K_1}{K_2}} \epsilon^{3/2}, \tag{43}
\]

which is obtained from the fact that \( \bar{E}(\delta_m = \sqrt{2\epsilon}) = \epsilon^2/2 \) and \( \bar{\omega}_b = \sqrt{2\bar{\omega}_t} \), is consistent with \( S_{cl} \) in Eq. (42) up to the numerical factor.

### D. Prefactor and Discussion

In order to complete our study of MQT and MQC in the biaxial symmetry, we need to calculate the preexponential factor in Eq. (4). Since in the biaxial symmetry the problem can be reduced to the one-dimensional one due to the smallness of \( |\phi| \), the calculation of the preexponential factor can be performed by using the well-known instanton method developed by many authors. However, in many other symmetries it is not possible to reduce the problem to the one-dimensional one by directly integrating over one of two variables such as \( \phi \), because the magnitude of \( \phi \) is not small enough for the action to be expanded as powers of \( \phi \) for Gaussian integration. This situation will be seen in the tetragonal symmetry case with \( \theta_H = \pi \). In order to treat such a case, Garg and Kim studied the formalism for evaluation of the prefactor based on the spin-coherent-state path integral. Here we explain briefly the basic idea of this study. Expanding the action \( S[\theta(\tau), \phi(\tau)] \) in terms of \( \theta_1 \) and \( \phi_1 \) about the classical path, where \( \theta(\tau) = \theta_{cl} + \theta_1 \) and \( \phi(\tau) = \phi_{cl} + \phi_1 \), we obtain the action as \( S[\theta, \phi] \approx S_{cl} + \delta^2 S \) with \( \delta^2 S \) being a functional of \( \theta_1 \) and \( \phi_1 \). Then Gaussian integration can be performed over \( \phi_1 \), and the \( \theta_1 \) path integral is now cast into the standard form of one-dimensional potential problem. What we need in this calculation is the approximate form of \( d\theta_{cl}/d\tau \) for large \( \tau \).
In the biaxial symmetry, by using Eq. (27) for $\theta_H = \pi/2$, Eq. (33) for $\pi/2 \ll \theta_H \ll \pi$, and Eq. (30) for $\theta_H = \pi$, we obtain the complete analytic forms of the tunneling rate for MQT or the oscillation rate for MQC, and the results are summarized in Table II(a). [18,19]

We note that in Table II(a) the preexponential factor is multiplied by a factor of two in case of $\theta_H = \pi$ because there exist two classical paths which give the same action. Also it is noted that the $\epsilon$ behavior of the WKB exponent $B$ is given by $\epsilon^{3/2}$ for $\theta_H = \pi/2$, $\epsilon^{5/4}$ for $\pi/2 \ll \theta_H \ll \pi$, and $\epsilon^{3/2}$ for $\theta_H = \pi$. It is seen by taking $K_2$ to be zero in Table II [20] that the tunneling rate in the uniaxial symmetry vanishes for $\theta_H = \pi$. This situation can be understood from the fact that for $\theta_H = \pi$ the Poisson bracket $\{p_\phi, H\}$ which determines the dynamics of the spin system with the Lagrangian (2) is zero because in this case the Hamiltonian $H$ becomes a function of only $p_\phi$, which gives $dp_\phi/d\tau = d(1 - \cos \theta)/d\tau = 0$ and then $\theta$ is constant in time. [18]

We obtained the instanton solutions for the full range of $\theta_H$ ($\pi/2 < \theta_H \leq \pi$) by solving numerically the equations of motion (21) and (22) or equivalently Eq. (23). Then by using the obtained instanton solutions we calculated the classical action from Eq. (20) to obtain the WKB exponent $B(= S_{cl}/\hbar)$. In Fig. 3 we present the instanton solutions with $\epsilon = 0.001$ and $K_1 = K_2$ for several values of $\theta_H$ which we obtained by numerical calculations. We also obtained the $\theta_H$-dependence of $B$ with $\epsilon = 0.01$ and $\epsilon = 0.001$ for $\pi/2 < \theta_H \leq \pi$, and present the result in Fig. 4. As is noted in the figure, the maximal value of $B$ is at about $\theta_H = 2.78(\approx 159^\circ)$ and the approximate analytic result obtained in Eq. (36) is almost valid in the range of angles $120^\circ \leq \theta_H \leq 170^\circ$.

IV. TUNNELING RATE FOR TETRAGONAL SYMMETRY

In this section we study the tetragonal symmetry whose anisotropy energy is given by

$$E_a(\theta, \phi) = K_1 \sin^2 \theta + K_2 \sin^4 \theta - K'_2 \cos(4\phi) \sin^4 \theta,$$  \hspace{1cm} (44)
where we once again take the easy axis to be $\pm \hat{z}$ for $K_1 > 0$. When we apply $\vec{H}$ in the $xz$-plane as in the previous section, the total energy becomes

$$E(\theta, \phi) = E_a(\theta, \phi) - H_x M_0 \sin \theta \cos \phi - H_z M_0 \cos \theta + E_0,$$

where we assume that $K_1 > 0$ and $|\bar{K}_2 - \bar{K}'_2| \ll 1$. Here we also use the dimensionless parameters defined in Eq. (6). By choosing $K'_2 > 0$, we take $\phi = 0$ to be an easy plane in our calculations. In the $\phi = 0$ plane the scaled total energy is written as

$$\bar{E}(\theta, 0) = \frac{1}{2} \sin^2 \theta + (\bar{K}_2 - \bar{K}'_2) \sin^4 \theta - \bar{H} \cos(\theta - \theta_H) + \bar{E}_0,$$ 

where $\bar{K}'_2 \equiv \bar{K}'_2/2K_1$. The initial angle $\theta_0$ and the critical angle $\theta_c$ defined in the previous section are determined by the equations

$$\sin \theta_0 \cos \theta_0 + \bar{H} \sin(\theta_0 - \theta_H) + 4(\bar{K}_2 - \bar{K}'_2) \sin^3 \theta_0 \cos \theta_0 = 0,$$ 

$$\sin \theta_c \cos \theta_c + \bar{H} \sin(\theta_c - \theta_H) + 4(\bar{K}_2 - \bar{K}'_2) \sin^3 \theta_c \cos \theta_c = 0,$$ 

$$\cos(2\theta_c) + \bar{H} \cos(\theta_c - \theta_H) + 4(\bar{K}_2 - \bar{K}'_2)(3 \sin^2 \theta_c \cos^2 \theta_c - \sin^4 \theta_c) = 0.$$ 

Using Eqs. (48) and (49), we obtain the critical magnetic field as

$$\bar{H}_c = \frac{1}{(\sin^{2/3} \theta_H + |\cos \theta_H|^{2/3})^{3/2}} \left[1 + \frac{4(\bar{K}_2 - \bar{K}'_2)}{1 + |\cos \theta_H|^{2/3}} \right].$$

In the small $\epsilon$-limit ($\epsilon \equiv 1 - \bar{H}/\bar{H}_c$), by using Eqs. (48) and (49) we obtain the approximate equation for $\eta(\equiv \theta_c - \theta_0)$ given by

$$- \epsilon [2\bar{H}_c \sin(\theta_c - \theta_H)] + \eta^2 [3\bar{H}_c \sin(\theta_c - \theta_H) + 6(\bar{K}_2 - \bar{K}'_2) \sin(4\theta_c)]$$

$$+ \eta \{ \epsilon [2\bar{H}_c \cos(\theta_c - \theta_H)] - \eta^2 [\bar{H}_c \cos(\theta_c - \theta_H) + 8(\bar{K}_2 - \bar{K}'_2) \cos(4\theta_c)] \} = 0.$$ 

The order of magnitude of the first two terms in Eq. (51) is higher than that of the last third term by $O(\sqrt{\epsilon})$. Therefore, for $\pi/2 \ll \theta_H \ll \pi$ we obtain $\eta$ from the first two terms of Eq. (51) as

$$\eta = \sqrt{\frac{2\epsilon}{3}} [1 + 4(\bar{K}_2 - \bar{K}'_2) \cos(2\theta_c)],$$

(52)
and $\theta_c$ from Eqs. (48) and (49) as

$$\theta_c = \frac{\pi}{4} + \frac{4(\bar{K}_2 - \bar{K}_2')}{3}.$$  (53)

However, for $\theta_H = \pi/2$ and $\pi$, since $\sin(\theta_c - \theta_H)$ and $\sin(4\theta_c)$ are equal to zero, the first two terms in Eq. (51) vanish. In these cases the value of $\eta$ is determined by the last third term, which gives

$$\theta_c = \frac{\pi}{2}, \quad \eta \simeq \sqrt{2\epsilon}[1 - 4(\bar{K}_2 - \bar{K}_2')] \quad \text{for} \quad \theta_H = \frac{\pi}{2},$$  (54)

$$\theta_c = 0, \quad \eta = 0 \quad \text{for} \quad \theta_H = \pi.$$  (55)

For reference, we note that $\eta$ is approximately given by

$$\eta \simeq \sqrt{2\epsilon}[1 - 4(\bar{K}_2 - \bar{K}_2')] \frac{\cos(4\theta_c)}{\bar{H}_c \cos(\theta_c - \theta_H)}$$  (56)

for the value of $\theta_H$ around $\theta_H = \pi/2$.

In the small $\epsilon$-limit the approximate form of $\bar{E}(\theta, 0)$ in Eq. (16) is written as

$$\bar{E}_1(\delta) = -\frac{1}{2} \bar{H}_c \sin(\theta_c - \theta_H) + 2(\bar{K}_2 - \bar{K}_2') \sin(4\theta_c)(3\delta^2 \eta - \delta^3)$$

$$-\frac{1}{2} \bar{H}_c \cos(\theta_c - \theta_H) + 8(\bar{K}_2 - \bar{K}_2') \cos(4\theta_c)[\delta^2(\epsilon - \frac{3}{2} \eta^2) + \eta \delta^3 - \frac{\delta^4}{4}]$$

$$+ 4(\bar{K}_2 - \bar{K}_2') \cos(4\theta_c) \delta^2 \epsilon,$$  (57)

where $\delta \equiv \theta - \theta_0$ which is small in the small $\epsilon$-limit. Then the total energy becomes

$$\bar{E}(\delta, \phi) = \bar{K}_2'[1 - \cos(4\phi)] \sin^4(\theta_0 + \delta) + \bar{H}_x(1 - \cos \phi) \sin(\theta_0 + \delta) + \bar{E}_1(\delta),$$  (58)

which will be discussed for three angle ranges, $\theta_H = \pi/2, \pi$ and $\pi/2 \ll \theta_H \ll \pi$.

**A. $\theta_H = \pi/2$**

Using the equation of motion for the classical trajectories, Eqs. (21) and (22), the classical path at $\theta_H = \pi/2$ is given by
\[ \delta_d(\bar{\tau}) = \sqrt{2\epsilon}[1 - 2\frac{(K_2 - K_2')}{K_1}] [1 + \tanh(\bar{\omega}_c\bar{\tau})], \quad (59) \]

\[ \phi_c(\bar{\tau}) = -i\epsilon[1 - 2\frac{(K_2 + K_1)}{K_1}][\frac{1}{\cosh^2(\bar{\omega}_c\bar{\tau})}], \quad (60) \]

where \( \bar{\omega}_c = \sqrt{\epsilon/2[1 + 2(K_2 + K_2')/K_1]} \). In this case the approximate form of \( \bar{E}_1(\delta) \) and the effective mass become

\[ \bar{E}_1(\delta) = \frac{[1 + 12(\bar{K}_2 - \bar{K}_2')]}{8} \delta^2 \{ \delta - 2\sqrt{2\epsilon [1 - 4(K_2 - K_2')]} \}^2, \quad (61) \]

\[ M = \frac{1}{H_c + 16\bar{K}_2}. \quad (62) \]

Since \( |\phi_c(\bar{\tau})| \ll 1 \), we can obtain a reduced one-dimensional action by performing the Gaussian integration over \( \phi \). In this case Eq. \((61)\) is the effective potential energy in the equation \((23)\) for the bounce \( \delta_d(\bar{\tau}) \). Using Eqs. \((59)\) and \((60)\), the corresponding classical action is given by

\[ S_{cl} = (\hbar J)^4 \frac{4}{3} \sqrt{2\epsilon}^{3/2}(1 - 4\frac{K_2}{K_1}). \quad (63) \]

From Eq. \((61)\) and Fig. 2, the height of barrier and the barrier frequency for \( \theta_H = \pi/2 \) are given by

\[ \bar{E}_1(\delta_m) = 2\epsilon^2[1 - 4(\bar{K}_2 - \bar{K}_2')] \quad \text{and} \quad \bar{\omega}_b = \sqrt{2}\bar{\omega}_c, \quad (64) \]

where \( \delta_m = \sqrt{2\epsilon [1 - 4(\bar{K}_2 - \bar{K}_2')]} \). Then the approximate form of the WKB exponent becomes

\[ B \sim \frac{U}{\hbar \omega_b} = 2J\epsilon^{3/2}(1 - 4\frac{K_2}{K_1}), \quad (65) \]

which up to the numerical factor is consistent with the action Eq. \((63)\) obtained by the calculation with the explicit bounce solution.

**B. \( \theta_H \ll \theta_H \ll \pi \)**

In this case the potential energy and the effective mass in the reduced one dimension are approximately given by
\[
\bar{E}_1(\delta) = -\frac{1}{2}[\bar{H}_c \sin(\theta_c - \theta_H) + 2(\bar{K}_2 - \bar{K}_2') \sin(4\theta_c)][(\sqrt{6}\epsilon\delta^2 - \delta^3),
\]
\[
M = (1 + |\cot \theta_H|^{2/3})[1 - 8 \frac{K_2'}{K_1} + 2 \frac{3 - 2|\cot \theta_H|^{2/3}}{3(1 + |\cot \theta_H|^{2/3})}(\frac{K_2 - K_2'}{K_1})].
\]

The classical trajectory is given by
\[
\delta_{cl}(\bar{\tau}) = \frac{\sqrt{6}\epsilon}{\cosh^2(\bar{\omega}_t \bar{\tau})},
\]
\[
\phi_{cl}(\bar{\tau}) = -i(6\epsilon)^{3/4}|\cot \theta_H|^{1/6}\frac{\sinh(\bar{\omega}_t \bar{\tau})}{\cosh^3(\bar{\omega}_t \bar{\tau})}
\times \sqrt{1 + |\cot \theta_H|^{2/3}}[1 - 4 \frac{K_2'}{K_1} + 2 \frac{2 - 3|\cot \theta_H|^{2/3}}{3(1 + |\cot \theta_H|^{2/3})}(\frac{K_2 - K_2'}{K_1})],
\]
where the precession frequency \(\bar{\omega}_t\) in MQT is given by
\[
\bar{\omega}_t = (\frac{3}{8})^{1/4}\epsilon^{1/4}(\frac{|\cot \theta_H|^{1/6}}{1 + |\cot \theta_H|^{2/3}})[1 + 4 \frac{K_2'}{K_1} + 2 \frac{5 - 3|\cot \theta_H|^{2/3}}{3(1 + |\cot \theta_H|^{2/3})}(\frac{K_2 - K_2'}{K_1})].
\]

The corresponding classical action becomes
\[
S_{cl} = (hJ) \frac{16 \times 6^{1/4}}{5} \epsilon^{5/4}|\cot \theta_H|^{1/6}[1 - 4 \frac{K_2'}{K_1} + 2 \frac{2 - |\cot \theta_H|^{2/3}}{3(1 + |\cot \theta_H|^{2/3})}(\frac{K_2 - K_2'}{K_1})].
\]

Using the barrier height \(\bar{E}_1(\delta_m = 2\sqrt{6}\epsilon/3) = 4a(6\epsilon)^{3/2}/27\) where \(a = -[\bar{H}_c \sin(\theta_c - \theta_H) + 2(\bar{K}_2 - \bar{K}_2') \sin(4\theta_c)]/2\), and the oscillation frequency \(\bar{\omega}_b = 2\bar{\omega}_t\), we obtain the approximate form of the WKB exponent as
\[
B \sim \frac{U}{\hbar \omega_b} = \frac{4 \times 6^{1/4}}{9} J\epsilon^{5/4}|\cot \theta_H|^{1/6}[1 - 4 \frac{K_2'}{K_1} + 2 \frac{2 - |\cot \theta_H|^{2/3}}{3(1 + |\cot \theta_H|^{2/3})}(\frac{K_2 - K_2'}{K_1})],
\]
which is consistent with Eq. (71) up to the numerical factor.

C. \(\theta_H = \pi\)

For \(\theta_H = \pi\), the total energy is written as
\[
E(\delta, \phi) = K_2'[1 - \cos(4\phi)]\delta^4 + \frac{1}{2}(\epsilon\delta^2 - \frac{1}{4}[1 - 8(K_2 - K_2')\delta^4])\}
\]
From the Euler-Lagrange equation (21) and (22) for \(\delta(\bar{\tau})\) and \(\phi(\bar{\tau})\), the classical trajectory is given by (77)
\[ \delta_{cl}(\bar{\tau}) = \sqrt{\frac{K_1\epsilon}{K + K_2' \cosh(4\bar{\omega}_t \bar{\tau})}} \],
\[ \phi_{cl}(\bar{\tau}) = -i\bar{\omega}_t \bar{\tau} + \frac{n}{2}\pi, \]

where \( n = 0, 1, 2, 3, \bar{K} = K_1/4 - K_2, \) and \( \bar{\omega}_t = \epsilon. \) Eq. (73) shows that in the present case \(|\phi| \ll 1\) is not valid and that we cannot expand \( \bar{E}(\delta, \phi) \) as powers of \( \phi, \) which means that we cannot reduce the problem to the one-dimensional one like Eq. (73). Thus the classical action should be obtained directly from the solution (74) and (75) of the Euler-Lagrange equation, which becomes

\[ S_{cl} = (\hbar J) \frac{\epsilon}{4} \left( \frac{K_1}{K_2} \right) \ln \left( \frac{\bar{K} + K_2}{K_2^2} \right), \]

where \( K_2 = \sqrt{\bar{K}^2 - (K_2')^2}. \) Even though the effective potential energy and the mass in one-dimensional form are not appropriate for the present case, the dependence of \( B \) on \( \epsilon \) can be derived from the values of \( \bar{E}_1(\delta_m) \) and \( \bar{\omega}_b \) in Table I, which gives \( \epsilon^{2-1} \) from \( \bar{E}_1(\delta_m)(\propto \epsilon^2) \) and \( \bar{\omega}_b(\propto \epsilon). \)

**D. Prefactor and Discussion**

Following the calculations used in the previous section, we can obtain the preexponential factor \( C_0 \) in Eq. (4). As can be shown in Table II, for \( \theta_H = \pi/2 \) and \( \pi/2 \ll \theta_H \ll \pi \) the values of \( C_0 \) in the tetragonal symmetry coincide with those in the biaxial symmetry. This situation is understood by the fact that the prefactor \( C_0 \) is determined only by the shape of the effective potential energy in the reduced one dimension, and the shapes are the same for the two symmetries as can be seen in Eqs. (25), (31), (61), and (66). However, the situation is different for \( \theta_H = \pi \) because \(|\phi| \ll 1\) is not valid. Thus, the factor \( C_0 \) should be determined from \( \delta^2 S \) by using the approximate forms of \( \delta_{cl}, E_{\delta\phi}, E_{\phi\phi}, E_{\delta\delta}, \) and so on, in the tetragonal symmetry case with \( \theta_H = \pi, \) which leads to the result of \( C_0 \) in Table II(b). It is possible to perform the same numerical calculation for given values of the parameters...
\( \bar{K}_2 \) and \( \bar{K}'_2 \) in the tetragonal symmetry in the same way as we did in the biaxial symmetry for the full range \( \pi/2 < \theta_H \leq \pi \). The results of the detailed numerical calculation for the dependence of \( B \) on \( \theta_H \) will be presented elsewhere.

V. CONCLUSIONS

In summary we obtained the approximate analytic forms of the oscillation rate (MQC) for \( \theta_H = \pi/2 \) and of the tunneling rate (MQT) for \( \pi/2 \ll \theta_H \ll \pi \), and \( \theta_H = \pi \) in the biaxial and tetragonal symmetries. Also, the \( \theta_H \)-dependence of the WKB exponent \( B \) for the full range \( \pi/2 < \theta_H \leq \pi \) with the values of \( \epsilon = 0.01 \) and 0.001 was calculated numerically in the biaxial symmetry. For future works it will be needed to compare the theoretical results of the \( \theta_H \)-dependence of \( B \) for given \( \epsilon \) and those of the \( \epsilon \)-dependence of \( B \) for given \( \theta_H \) obtained in this paper for the biaxial and tetragonal symmetries, with the experimental results which can be obtained by the same procedures as those suggested in Ref. 8 for the uniaxial symmetry.

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for $K_1 < 0$. Thus the range of $\theta_H$ in which we are interested is $\pi/4 \leq \theta_H \leq \pi/2$ for $K_1 > 0$ and $0 \leq \theta_H \leq \sin^{-1}(1/\sqrt{3})$ for $K_1 < 0$. Even though the anisotropy energy for $K_1 > 0$ can be reduced to the one for the tetragonal symmetry by replacing $\bar{K_2}$ and $\bar{K'_2}$ by $-7/16$ and $1/16$, the approximation which we used in the tetragonal symmetry is not applicable. Also, the situation is much more complicated in case of the cubic symmetry for $K_1 < 0$. For the cubic symmetry, numerical approach should be used to obtain the dependence of tunneling rate on the direction of magnetic field, which will be done elsewhere.

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of 4. We carefully checked that our results is consistent with that of Ref. [19] which is referred in Ref. [8].

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TABLE I. The values appearing in Fig 2 for three ranges of $\theta_H$ (a) in the biaxial symmetry and (b) in the tetragonal symmetry. $\delta_m$ is the angle where the maximum of the barrier locates. Here, $a = -[\bar{H}_c \sin(\theta_c - \theta_H) + 2\bar{K} \sin(4\theta_c)]/2$ where $\bar{K} \equiv (K_2 - K_2')/2K_1$.

| Field angle | $\delta_m$ | $E_1(\delta_m)$ | $\bar{\omega}_b$ |
|-------------|------------|-----------------|------------------|
| $\theta_H = \pi/2$ | $\sqrt{2}\epsilon$ | $\epsilon^2/2$ | $\sqrt{2}\bar{\omega}_c$ |
| $\pi/2 \ll \theta_H \ll \pi$ | $2\sqrt{6}\epsilon/\sqrt{3}$ | $\sin(2\theta_c)(6\epsilon)^{3/2}/27$ | $2\bar{\omega}_t$ |
| $\theta_H = \pi$ | $\sqrt{2}\epsilon$ | $\epsilon^2/2$ | $\sqrt{2}\bar{\omega}_t$ |

(a)

| Field angle | $\delta_m$ | $E_1(\delta_m)$ | $\bar{\omega}_b$ |
|-------------|------------|-----------------|------------------|
| $\theta_H = \pi/2$ | $\sqrt{2}\epsilon(1 - 4\bar{K})$ | $2\epsilon^2(1 - 4\bar{K})$ | $\sqrt{2}\bar{\omega}_c$ |
| $\pi/2 \ll \theta_H \ll \pi$ | $2\sqrt{6}\epsilon/\sqrt{3}$ | $4\epsilon(6\epsilon)^{3/2}/27$ | $2\bar{\omega}_t$ |
| $\theta_H = \pi$ | $\sqrt{K_1\epsilon/2(\bar{K} + K_2')}$ | $K_1\epsilon^2/8(\bar{K} + K_2')$ | $\sim \bar{\omega}_t$ |

(b)
TABLE II. The oscillation rate in MQC for $\theta_H = \pi/2$ and the tunneling rate in MQT for $\pi/2 \ll \theta_H \ll \pi$ (a) in the biaxial symmetry and (b) in the tetragonal symmetry. $C_0$ is preexponential factor, $\omega_b$ in MQC or $\omega_p$ in MQT the frequency of small oscillations around the minimum of the inverted potential, and $B$ the WKB exponent. The complete form of the rate is given in Eq. (4). We define $K_\beta = (\tilde{K} + K_\alpha)/K_2$, $K_\gamma = (K_2 - K_2')/K_1$, $\bar{\omega}_u = (3^4/8 \sqrt{3} / (1 + |\cot \theta_H|^{1/3}) \epsilon^{1/4}$ and $B_u = 16 \times \epsilon^{5/4}$, where $\bar{\omega}_u$ and $B_u$ are the dimensionless frequency and the WKB exponent for $\pi/2 \ll \theta_H \ll \pi$ in the uniaxial symmetry.

| Field angle | $C_0$ | $\omega_I/\omega_0$ (I = c for MQC or t for MQT) | $B (= S_d/h)/J$ |
|-------------|-------|-----------------------------------------------|----------------|
| $\theta_H = \pi/2$ | $8\sqrt{3}$ | $\frac{1}{\sqrt{2}} \sqrt{1 + K_2/K_1}^{1/2}$ | $\frac{4}{3} \sqrt{2} (1 + K_2/K_1)^{-1/2} \epsilon^{3/2}$ |
| $\pi/2 \ll \theta_H \ll \pi$ | $4\sqrt{15}$ | $\bar{\omega}_u [1 + \frac{K_2}{K_1} (1 + |\cot \theta_H|^{1/3})]^{1/2}$ | $\bar{B}_u/[1 + \frac{K_2}{K_1} (1 + |\cot \theta_H|^{1/3})]^{1/2}$ |
| $\theta_H = \pi$ | $4\sqrt{3}$ | $\sqrt{\frac{K_2}{K_1}} \epsilon^{1/2}$ | $\frac{8}{3} \sqrt{\frac{K_1}{K_2}} \epsilon^{3/2}$ |

(a)

| Field angle | $C_0$ | $\omega_I/\omega_0$ (I = c or t) | $B (= S_d/h)/J$ |
|-------------|-------|-----------------------------------|----------------|
| $\theta_H = \pi/2$ | $8\sqrt{3}$ | $\sqrt{\frac{2}{3}} [1 + 2(K_2 + K_2')/K_1]$ | $\frac{4}{3} \sqrt{2} (1 - 4K_2/K_1) \epsilon^{3/2}$ |
| $\pi/2 \ll \theta_H \ll \pi$ | $4\sqrt{15}$ | $\bar{\omega}_u [1 + \frac{K_2'}{K_1} + \frac{2}{3} \frac{(5 - 3)}{1 + |\cot \theta_H|^{1/3}} K_\gamma] \epsilon$ | $B_u[1 - 4 \frac{K_2'}{K_1} + \frac{2}{3} \frac{(2 - 3)}{1 + |\cot \theta_H|^{1/3}} K_\gamma]$ |
| $\theta_H = \pi$ | $16 \sqrt{\frac{K_2 - K_2'/2K_\alpha}{K_2 K_\beta}} \ln K_\beta$ | $\frac{4}{9} \frac{K_2'}{K_1} [\ln K_\beta] \epsilon$ | $\frac{1}{4} \frac{K_2'}{K_1} [\ln K_\beta] \epsilon$ |

(b)
FIGURES

FIG. 1. Comparison of (a) $\sin(2\theta_c)$ with (b) $\cos(2\theta_c)$ in Eqs. (13) and (17). Note that $\sin(2\theta_c) = 0$, $|\cos(2\theta_c)| = 1$ for both $\theta_H = \pi/2$ and $\pi$.

FIG. 2. The plots of the effective potential $\bar{E}_1(\delta)$ as a function of $\delta(=\theta - \theta_0)$ for (a) $\theta_H = \pi/2$ (MQC), (b) $3\pi/4$ and (c) $\pi$ about $\theta_0$ at which the system is metastable (MQT). The position $\delta_m$ and the height $\bar{E}_1(\delta_m)$ of the barrier, and the barrier frequency $\bar{\omega}_b$ are given in Table I for the biaxial and tetragonal symmetries.

FIG. 3. The $\theta_H$-dependence of the instanton solutions in the biaxial symmetry for $\epsilon(= 1 - H/H_c) = 0.001$ and $K_1 = K_2$. Here, (a) $\theta_H = 90.0002^\circ$, (b) $90.01^\circ$, (c) $135^\circ$, (d) $179.99^\circ$, (e) $180^\circ$.

FIG. 4. The $\theta_H$-dependence of the relative WKB exponent $B(\theta_H)/B(3\pi/4)$ in the biaxial symmetry with $K_1 = K_2$ for (a) $\epsilon = 0.01$ and (b) $\epsilon = 0.001$. Here, $B(\theta_H = 3\pi/4, \epsilon = 0.01) = 9.14 \times 10^{-3}$ and $B(\theta_H = 3\pi/4, \epsilon = 0.001) = 5.14 \times 10^{-4}$.
The graph shows the function $\mathcal{E}_1(\delta)$. It has three different curves labeled (a), (b), and (c). The x-axis represents $\delta$, and the y-axis represents $\mathcal{E}_1(\delta)$. Each curve has a distinct pattern and magnitude, indicating different behaviors for each case.
\[ \frac{B(\theta_H)}{B(3\pi/4)} \]