Quantum nucleation in ferromagnets with tetragonal and hexagonal symmetries

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Abstract

The phenomenon of quantum nucleation is studied in a ferromagnet in the presence of a magnetic field at an arbitrary angle. We consider the magnetocrystalline anisotropy with tetragonal symmetry and that with hexagonal symmetry, respectively. By applying the instanton method in the spin-coherent-state path-integral representation, we calculate the dependence of the rate of quantum nucleation and the crossover temperature on the orientation and strength of the field for a thin film and for a bulk solid. Our results show that the rate of quantum nucleation and the crossover temperature depend on the orientation of the external magnetic field distinctly, which provides a possible experimental test for quantum nucleation in nanometer-scale ferromagnets.

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I. INTRODUCTION

The tunneling of macroscopic object, known as Macroscopic Quantum Tunneling (MQT), is one of the most fascinating phenomena in condensed matter physics. In the last decade, the problem of quantum tunneling of magnetization in nanometer-scale magnets has attracted a great deal of theoretical and experimental interest. MQT in magnetic systems are interesting from a fundamental point of view as they can extend our understanding of the transition from quantum to classical behavior. On the other hand, these phenomena are important to the reliability of small magnetic units in memory devices and the designing of quantum computers in the future. And the measurement of magnetic MQT quantities such as the tunneling rates could provide independent information about microscopic parameters such as the magnetocrystalline anisotropies and the exchange constants. All this makes magnetic quantum tunneling an exciting area for theoretical research and a challenging experimental problem.

The problem of quantum nucleation of a stable phase from a metastable one in ferromagnetic films is an interesting fundamental problem which allows direct comparison between theory and experiment. Consider a ferromagnetic film with its plane perpendicular to the easy axis determined by the magnetocrystalline anisotropy energy depending on the crystal symmetry. A magnetic field $\mathbf{H}$ is applied in a direction between perpendicular and opposite to the initial easy direction of the magnetization $\mathbf{M}$, which favors the reversal of the magnetization. The reversal occurs via the nucleation of a critical bubble, which then the nucleus does not collapse, but grows unrestrictedly in volume. If the temperature is sufficiently high, the nucleation of a bubble is a thermal overbarrier process, and the rate of thermal nucleation follows the Arrhenius law $\Gamma_T \propto \exp \left( -U/k_B T \right)$, with $k_B$ being the Boltzmann constant and $U$ being the height of energy barrier. In the limit of $T \to 0$, the nucleation is purely quantum-mechanical and the rate goes as $\Gamma_Q \propto \exp \left( -S_{cl}/\hbar \right)$, with $S_{cl}$ being the classical action or the WKB exponent which is independent of temperature. Because of the exponential dependence of the thermal rate on $T$, the temperature $T_c$ characterizing the
crossover from quantum to thermal regime can be estimated as \( k_B T_c = \hbar U/S_{cl} \).

The problem of quantum nucleation was studied by Privorotskii\(^3\) who estimated the exponent in the rate of quantum nucleation based on the dimensional analysis. Chudnovsky and Gunther\(^4\) studied the quantum nucleation of a thin ferromagnetic film in the presence of an external magnetic field along the opposite direction to the easy axis at zero temperature by applying the instanton method in the spin-coherent-state path-integral representation. Ferrera and Chudnovsky extended the quantum nucleation to a finite temperature.\(^5\) Kim studied the quantum nucleation in a thin ferromagnetic film placed in a magnetic field at an arbitrary angle.\(^6\)

It is noted that the previous results\(^4\)–\(^6\) of quantum nucleation were obtained for ferromagnetic sample with the simplest form of the magnetocrystalline anisotropy energy such as the biaxial symmetry, and the model considered in Refs. 4 and 5 was confined to the condition that the magnetic field be applied along the opposite direction to the easy axis. The purpose of this paper is to extend the previous results of quantum nucleation in ferromagnetic system with simple biaxial symmetry to that of system with a more general symmetry, such as tetragonal and hexagonal symmetry. The generic quantum nucleation problem, however, and the easiest to implement in practice, is that of ferromagnets with a general structure of magnetocrystalline anisotropy in a magnetic field applied at a some angle \( \theta_H \) to the anisotropy axis. This problem does not possess any symmetry and for that reason is more difficult mathematically. It is worth pursuing, however, because of its significance for experiments.\(^1\)

In this paper the magnetic field is applied in an arbitrary direction between perpendicular and opposite to the initial easy axis (\( \hat{z} \) axis). Our interest in studying quantum nucleation of magnetic bubbles with a more general structure of magnetocrystalline anisotropy in an arbitrarily directed magnetic field is stimulated by the fact that the corresponding experiment would be most easy to perform and to interpret. Within the instanton approach, we present the numerical results for the WKB exponent in quantum nucleation of a thin ferromagnetic film with the magnetic field applied in a range of angles \( \pi/2 < \theta_H < \pi \), where \( \theta_H \) is the angle between the initial easy axis (\( \hat{z} \) axis) and the field. We also discuss the \( \theta_H \) dependence of the
crossover temperature $T_c$ from purely quantum nucleation to thermally assisted processes. Our results show that the distinct angular dependence, together with the dependence of the WKB exponent on the strength of the external magnetic field, may provide an independent experimental test for quantum nucleation in a ferromagnetic film. Quantum nucleation (the description involves space-time instantons), being a field theory problem, is more difficult than tunneling of magnetization in single-domain particles, both at the conceptual and at the technical level. Therefore, this paper provides a nontrivial generalization of uniform rotation of magnetization vector (homogeneous spin tunneling) in single-domain magnets to a nonuniform rotation of magnetization in bulk magnets with a more general structure of magnetocrystalline anisotropy in the presence of a magnetic field at an arbitrary angle.

Compared with the tunneling in single-domain particles, a local tunneling event in a bulk magnet can trigger instability on a much greater scale, which leads to really macroscopic consequences. In experiments, it may be easier to monitor single nucleation events in a thin film than to detect the magnetization reversal in a nanometer-scale particle. Therefore, our theoretical results for a general structure of magnetocrystalline anisotropy in an arbitrarily directed field will be more applicable for experimental tests of quantum nucleation. Besides the importance from the fundamental point of view, processes of quantum nucleation and collapse of magnetic bubbles are potentially important for quantum limitations on the density and long-term reliability of the data storage in magnetic memory devices and designing of quantum computer.

This paper is structured in the following way. In Sec. II, we review briefly some basic ideas of quantum nucleation of magnetization in ferromagnets. In Secs. III and IV, we study quantum nucleation of magnetization in ferromagnets with tetragonal and hexagonal symmetry in an external magnetic field applied in the $ZX$ plane with a range of angles $\pi/2 \leq \theta_H < \pi$. The conclusions and discussions are presented in Sec. V.
II. THE PHYSICAL MODEL

For a spin tunneling problem, the rate of magnetization reversal by quantum tunneling is determined by the imaginary-time transition amplitude from an initial state \( |i\rangle \) to a final state \( |f\rangle \) as

\[
U_{fi} = \langle f | e^{-HT} | i \rangle = \int \mathcal{D}\{\mathbf{M}(r, \tau)\} \exp \left(-\mathcal{S}_E/\hbar\right),
\]

where \( \mathcal{S}_E \) is the Euclidean action which includes the Euclidean Lagrangian density \( \mathcal{L}_E \) as

\[
\mathcal{S}_E = \int d\tau d^3r \mathcal{L}_E.
\]

For ferromagnets at sufficiently low temperature, all the spins are locked together by the strong exchange interaction, and therefore only the orientation of magnetization \( \mathbf{M}(r, \tau) \) can change but not its absolute value. For that reason the field \( \mathbf{M}(r, \tau) \) is equivalent to the fields \( \theta(r, \tau) \) and \( \phi(r, \tau) \), which are spherical coordinates of \( \mathbf{M} \). In this case the measure of the path integral \( \mathcal{D}\{\mathbf{M}(r, \tau)\} \) in Eq. (1) is equivalent to

\[
\int \mathcal{D}\{\theta(r, \tau)\} \mathcal{D}\{\phi(r, \tau)\} = \lim_{\varepsilon \to 0} N \prod_{k=1}^{N} \left(\frac{2S + 1}{4\pi}\right) \sin \theta_k d\theta_k d\phi_k,
\]

where \( \varepsilon = \max(\tau_{k+1} - \tau_k) \) and \( S = M_0/\hbar\gamma \) is the total spin of ferromagnet. Here \( \gamma \) is the gyromagnetic ratio and \( M_0 \) is the magnitude of magnetization.

In the spin-coherent-state representation, the magnetic Lagrangian is given by

\[
\mathcal{L}_E = i \frac{M_0}{\gamma} \left(\frac{d\phi(r, \tau)}{d\tau}\right) [1 - \cos \theta(r, \tau)] + E(\theta, \phi).
\]

The first term in Eq. (4) is a total imaginary-time derivative, which has no effect on the classical equations of motion, but it is crucial for the spin-parity effects. However, for the closed instanton trajectory described in this paper (as shown in the following), this time derivative gives a zero contribution to the path integral, and therefore can be omitted.

The energy density in Eq. (4) is

\[
E(\theta, \phi) = E_a(\theta, \phi) + E_{ex}(\theta, \phi),
\]
where \( E_a \) includes the magnetocrystalline anisotropy energy and the energy due to the external magnetic field, and \( E_{ex} \) is the exchange energy

\[
E_{ex} = \frac{\alpha}{2} \left( \partial_i M_j \right)^2 = \frac{\alpha}{2} M_0^2 \left[ (\nabla \theta)^2 + \sin^2 \theta (\nabla \phi)^2 \right],
\]

where \( \alpha \) is the exchange stiffness. The magnetocrystalline anisotropy energy for tetragonal and hexagonal symmetry is shown in Sec. III and IV, respectively. In the semiclassical limit, the rate of quantum nucleation, with an exponential accuracy, is given by

\[
\Gamma_Q \propto \exp \left[ -S_{E}^{\text{min}}/\hbar \right],
\]

where \( S_{E}^{\text{min}} \) is obtained along the trajectory that minimizes the Euclidean action \( S_E \).

### III. FERROMAGNETS WITH TETRAGONAL SYMMETRY

In this section, we study the quantum nucleation of magnetization in ferromagnets with tetragonal symmetry in the presence of a magnetic field at arbitrary angles in the \( ZX \) plane, which has the following magnetocrystalline anisotropy energy

\[
E_a (\theta, \phi) = K_1 \sin^2 \theta + K_2 \sin^4 \theta - K'_2 \sin^4 \theta \cos (4\phi) - M_0 H_x \sin \theta \cos \phi - M_0 H_z \cos \theta,
\]

where \( K_1, K_2 \) and \( K'_2 \) are the magnetic anisotropy coefficients, and \( K_1 > 0 \). In the absence of the magnetic field, the easy axes of this system are \( \pm \widehat{z} \) for \( K_1 > 0 \). And the field is applied in the \( ZX \) plane at \( \pi/2 < \theta_H < \pi \). Then the total energy is given by

\[
E[\theta (r, \tau), \phi (r, \tau)] = K_1 \sin^2 \theta + K_2 \sin^4 \theta - K'_2 \sin^4 \theta \cos (4\phi) + \frac{\alpha}{2} M_0^2 \left[ (\nabla \theta)^2 + \sin^2 \theta (\nabla \phi)^2 \right] - M_0 H_x \sin \theta \cos \phi - M_0 H_z \cos \theta + E_0,
\]

where \( E_0 \) is a constant which makes \( E(\theta, \phi) \) zero at the initial state. By applying the similar method in Ref. 15, we can perform a Gaussian integration over the variable \( \phi \) in the path integral and reduce the system to that with only one variable \( \delta \) (as shown in the following). Then it is possible to perform the rest of the calculation by using the instanton
method. This method simplifies the problem tremendously, compared to the problem where the action depended on \( \theta (\tau) \) and \( \phi (\tau) \), though a complete mathematical equivalence to the initial problem is preserved.

By introducing the dimensionless parameters as

\[
K_2 = K_2 / 2K_1, \quad K_2' = K_2' / 2K_1, \quad H_x = H_x / H_0, \quad H_z = H_z / H_0,
\]

Eq. (9) can be rewritten as

\[
E (\theta, \phi) = \frac{1}{2} \sin^2 \theta + K_2 \sin^4 \theta - K_2' \sin^4 \theta \cos (4\phi) + \frac{\alpha M_0^2}{4K_1} [(\nabla \theta)^2 + \sin^2 \theta (\nabla \phi)^2] \\
- H_x \sin \theta \cos \phi - H_z \cos \theta + E_0,
\]

where \( E (\theta, \phi) = 2K_1 E (\theta, \phi) \), and \( H_0 = 2K_1 / M_0 \). At finite magnetic field, the plane given by \( \phi = 0 \) is the easy plane, on which \( E_a (\theta, \phi) \) reduces to

\[
E_a (\theta, \phi = 0) = \frac{1}{2} \sin^2 \theta + (K_2 - K_2') \sin^4 \theta - H \cos (\theta - \theta_H) + E_0.
\]

The initial angle \( \theta_0 \) is determined by \([dE_a (\theta, 0) / d\theta]_{\theta=\theta_0} = 0\), and the critical angle \( \theta_c \) and the dimensionless critical field \( H_c \) are determined by both \([dE_a (\theta, 0) / d\theta]_{\theta=\theta_c, \pi=\pi_c} = 0\) and \([d^2E_a (\theta, 0) / d\theta^2]_{\theta=\theta_c, \pi=\pi_c} = 0\), which leads to

\[
\frac{1}{2} \sin (2\theta_0) + H \sin (\theta_0 - \theta_H) + 4 \left( K_2 - K_2' \right) \sin^3 \theta_0 \cos \theta_0 = 0, \quad (13a)
\]
\[
\frac{1}{2} \sin (2\theta_c) + H_c \sin (\theta_c - \theta_H) + 4 \left( K_2 - K_2' \right) \sin^3 \theta_c \cos \theta_c = 0, \quad (13b)
\]
\[
\cos (2\theta_c) + H_c \cos (\theta_c - \theta_H) + 4 \left( K_2 - K_2' \right) \left( 3 \sin^2 \theta_c \cos^2 \theta_c - \sin^4 \theta_c \right) = 0. \quad (13c)
\]

Assuming that \( |K_2 - K_2'| \ll 1 \), we obtain the critical magnetic field and the critical angle as

\[
H_c = \frac{1}{\left[ (\sin \theta_H)^{2/3} + |\cos \theta_H|^{2/3} \right]^{3/2}} \left[ 1 + \frac{4 \left( K_2 - K_2' \right)}{1 + |\cot \theta_H|^{2/3}} \right], \quad (14a)
\]
\[
\sin \theta_c = \frac{1}{\left( 1 + |\cot \theta_H|^{2/3} \right)^{1/2}} \left[ 1 + \frac{8}{3} \left( K_2 - K_2' \right) \frac{|\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} \right]. \quad (14b)
\]
In the low barrier limit, i.e., \( \epsilon = 1 - \frac{\mathcal{H}}{\mathcal{H}_c} \to 0 \), by using Eqs. (13b) and (13c) we obtain the approximate equation for \( \eta (\equiv \theta_c - \theta_0) \) in the order of \( \epsilon^{3/2} \),

\[
-\epsilon \mathcal{H}_c \sin (\theta_c - \theta_H) + \eta^2 \left[ \frac{3}{2} \mathcal{H}_c \sin (\theta_c - \theta_H) + 3 \left( K_2 - K'_2 \right) \sin (4\theta_c) \right] + \eta \left\{ \epsilon \mathcal{H}_c \cos (\theta_c - \theta_H) - \eta^2 \left[ \frac{1}{2} \mathcal{H}_c \cos (\theta_c - \theta_H) + 4 \left( K_2 - K'_2 \right) \cos (4\theta_c) \right] \right\} = 0. \tag{15}
\]

Introducing \( \delta \equiv \theta - \theta_0 \) (\( |\delta| \ll 1 \) in the small \( \epsilon \) limit), we derive the energy \( \mathcal{E} (\theta, \phi) \) as

\[
\mathcal{E} (\delta, \phi) = K_2 \left[ 1 - \cos (4\phi) \right] \sin^4 (\theta_0 + \delta) + \mathcal{H}_x (1 - \cos \phi) \sin (\theta_0 + \delta) + \frac{\alpha M_0^2}{4K_1} \left[ (\nabla \theta)^2 + \sin^2 \theta (\nabla \phi)^2 \right] + \mathcal{E}_1 (\delta), \tag{16}
\]

where \( \mathcal{E}_1 (\delta) \) is a function of only \( \delta \) given by

\[
\mathcal{E}_1 (\delta) = \left[ \frac{1}{2} \mathcal{H}_c \sin (\theta_c - \theta_H) + \left( K_2 - K'_2 \right) \sin (4\theta_c) \right] (\delta^3 - 3\delta^2 \eta) + \left[ \frac{1}{8} \mathcal{H}_c \cos (\theta_c - \theta_H) + \left( K_2 - K'_2 \right) \cos (4\theta_c) \right] (\delta^4 - 4\delta^3 \eta + 6\delta^2 \eta^2 - 4\delta^2 \epsilon) + 4 \left( K_2 - K'_2 \right) \epsilon \delta^2 \cos (4\theta_c). \tag{17}
\]

It can be shown that in the region of \( \pi/2 < \theta_H < \pi, 0 < \theta_c < \pi/2, \eta \) and \( \delta \) are of the order of \( \sqrt{\epsilon} \), the second or third term in Eq. (17) is smaller than the first term in the small \( \epsilon \) limit. It is convenient to use dimensionless variables

\[
r' = \epsilon^{1/4} r/r_0, \quad \tau' = \epsilon^{1/4} \omega_0 \tau, \quad \delta = \delta / \sqrt{\epsilon}, \tag{18}
\]

where \( r_0 = \sqrt{\frac{\alpha M_0^2}{2K_1}} \), and \( \omega_0 = 2\gamma K_1/M_0 \). Then the Euclidean action Eq. (2) for \( \pi/2 < \theta_H < \pi \) becomes

\[
\mathcal{S}_E [\delta (r', \tau'), \phi (r', \tau')] = \frac{hS r_0^3}{\epsilon} \int d\tau' d^3 r' \left\{ -i \epsilon^{1/4} \sin (\theta_0 + \sqrt{\epsilon} \delta) \phi \left( \frac{\partial \delta}{\partial \tau'} \right) + 2K_2 \sin^2 (2\phi) \sin^4 (\theta_0 + \sqrt{\epsilon} \delta) + 2\mathcal{H}_x \sin^2 \left( \frac{\phi}{2} \right) \sin (\theta_0 + \sqrt{\epsilon} \delta) + \frac{1}{2} \epsilon^{3/2} \left( \nabla \delta \right)^2 + \frac{1}{2} \epsilon^{1/2} \sin^2 (\theta_0 + \sqrt{\epsilon} \delta) \left( \nabla' \phi \right)^2 + \frac{A}{4} \epsilon^{3/2} \left( \sqrt{6\delta^2 - \delta^3} \right) \right\}, \tag{19}
\]
where
\[
A = 2 \frac{\left| \cot \theta_H \right|^{1/3}}{1 + \left| \cot \theta_H \right|^{2/3}} \left[ 1 + \frac{4}{3} \left( \mathcal{K}_2 - \mathcal{K}'_2 \right) \frac{7 - 4 \left| \cot \theta_H \right|^{2/3}}{1 + \left| \cot \theta_H \right|^{2/3}} \right].
\] (20)

In Eq. (19) we have performed the integration by part for the first term and have neglected the total imaginary-time derivative. In can be showed that for \( \pi/2 < \theta_H < \pi \), only small values of \( \phi \) contribute to the path integral, so that one can replace \( \sin^2 \phi \) in Eq. (19) by \( \phi^2 \) and neglect the term including \( (\nabla' \phi)^2 \) which is of the order \( \epsilon^2 \) while the other terms are of the order \( \epsilon^{3/2} \). Then the Gaussian integration over \( \phi \) leads to
\[
\int \mathcal{D} \{ \delta (r', \tau') \} \exp \left( -\frac{1}{\hbar} S_E^{\text{eff}} \right),
\] (21)

where the effective action is
\[
S_E^{\text{eff}} [\delta (r', \tau')] = \hbar \epsilon^{1/2} r_0^3 \int d\tau' d^3r' \left[ \frac{1}{2} M \left( \frac{\partial \delta}{\partial \tau'} \right)^2 + \frac{1}{2} (\nabla' \delta)^2 + \frac{A}{4} \left( \sqrt{6} \delta^2 - \delta^3 \right) \right].
\] (22)

The effect mass in Eq. (22) is found to be
\[
M = \frac{\left( 1 + \left| \cot \theta_H \right|^{2/3} \right)}{1 - \epsilon + 16K_2 + 4 \left( K_2 - K'_2 \right) \frac{1}{1 + \left| \cot \theta_H \right|^{2/3}} + 128K_2 \left( K_2 - K'_2 \right) \frac{\left| \cot \theta_H \right|^{2/3}}{1 + \left| \cot \theta_H \right|^{2/3}}}. \] (23)

Introducing the variables \( \tau = \tau' \sqrt{A/M} \) and \( \widetilde{r} = r' \sqrt{A} \), the effective action Eq. (22) is simplified as
\[
S_E^{\text{eff}} [\widetilde{\delta} (\widetilde{r}, \tau)] = \hbar \epsilon^{1/2} r_0^3 \sqrt{M} \int d\tau d^3\widetilde{r} \left[ \frac{1}{2} M \left( \frac{\partial \widetilde{\delta}}{\partial \tau} \right)^2 + \frac{1}{2} (\nabla \widetilde{\delta})^2 + \frac{1}{4} \left( \sqrt{6}\widetilde{\delta}^2 - \widetilde{\delta}^3 \right) \right].
\] (24)

For the quantum reversal of magnetization \( \mathbf{M} \) in a small particle of volume \( V \ll r_0^3 \), \( \mathbf{M} \) is uniform within the particle and \( \widetilde{\delta} \) does not depend on the space \( \mathbf{r} \), Eq. (24) reduces to
\[
S_E^{\text{eff}} [\widetilde{\delta} (\tau, \tau)] = \hbar \epsilon^{5/4} \sqrt{MAV} \int d\tau \left[ \frac{1}{2} \left( \frac{d\widetilde{\delta}}{d\tau} \right)^2 + \frac{1}{4} \left( \sqrt{6}\widetilde{\delta}^2 - \widetilde{\delta}^3 \right) \right].
\] (25)

The corresponding classical trajectory satisfies the equation of motion
\[
\frac{d^2 \delta}{d\tau^2} = \frac{1}{2} \sqrt{6} \delta - \frac{3 \delta^2}{4}.
\] (26)

Eq. (26) has the instanton solution
\[ \vec{\delta}(\tau) = \frac{\sqrt{6}}{\cosh^2 \left( 3^{1/4} \times 2^{-5/4} \tau \right)}, \quad (27) \]

corresponding to the variation of \( \vec{\delta} \) from \( \vec{\delta} = 0 \) at \( \tau = -\infty \), to \( \vec{\delta} = \sqrt{6} \) at \( \tau = 0 \), and then back to \( \vec{\delta} = 0 \) at \( \tau = \infty \). Eq. (27) agrees well with the result in Refs. 13 and 15. The associated classical action is found to be

\[ S_{cl} = \frac{2^{17/4} \times 3^{1/4}}{5} \hbar S^5/4 \]
\[ \times \frac{\left| \cot \theta_H \right|^{1/6} \left[ 1 + \frac{2}{3} \left( K_2 - K_2' \right) \frac{\tau - 2|\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} \right]}{\sqrt{1 - \epsilon + 16K_2 + 4 \left( K_2 - K_2' \right) \frac{1}{1 + |\cot \theta_H|^{2/3}} + 128K_2 \left( K_2 - K_2' \right) \frac{|\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}}}}. \quad (28) \]

Now we turn to the nonuniform problem. In case of a thin film of thickness \( h \) less than the size \( r_0/\epsilon^{1/4} \) of the critical nucleus and its plane is perpendicular to the initial easy axis, we obtain the action Eq. (24) after performing the integration over the \( \tau \) variable,

\[ S_{eff}^E \left[ \vec{\delta}(\vec{r}, \tau) \right] = \hbar S^3/4 r_0^2 h \sqrt{\frac{M}{A}} \int d\tau d^2 \vec{r} \left[ \frac{1}{2} \left( \frac{\partial \vec{\delta}}{\partial \tau} \right)^2 + \frac{1}{2} \left( \nabla \vec{\delta} \right)^2 + \frac{1}{4} \left( \sqrt{6\vec{\delta}^2 - \vec{\delta}^4} \right) \right]. \quad (29) \]

At zero temperature the classical solution of the effective action Eq. (29) has \( O(3) \) symmetry in two spatial plus one imaginary time dimensions. Therefore, the solution \( \vec{\delta} \) is a function of \( u \), where \( u = (\rho^2 + \tau^2)^{1/2} \), and \( \rho = (x^2 + y^2)^{1/2} \) is the normalized distance from the \( z \) axis. Now the effective action Eq. (29) becomes

\[ S_{eff}^E \left[ \vec{\delta}(\vec{r}, \tau) \right] = 4\pi \hbar S^3/4 r_0^2 h \sqrt{\frac{M}{A}} \int du u^2 \left[ \frac{1}{2} \left( \frac{d\vec{\delta}}{du} \right)^2 + \frac{1}{4} \left( \sqrt{6\vec{\delta}^2 - \vec{\delta}^4} \right) \right]. \quad (30) \]

The corresponding classical trajectory satisfies the following equation of motion

\[ \frac{d^2 \vec{\delta}}{du^2} + \frac{2}{u} \frac{d\vec{\delta}}{du} \frac{\sqrt{6\vec{\delta}^2 - 3\vec{\delta}^4}}{4} = \frac{\sqrt{6\vec{\delta}^2 - 3\vec{\delta}^4}}{4}. \quad (31) \]

By applying the similar method the instanton solution of Eq. (31) can be found numerically and is illustrated in Fig. 1. The maximal rotation of \( M \) is \( \delta_{\text{max}} \approx 6.8499 \) at \( \tau = 0 \) and \( \vec{\tau} = 0 \). Numerical integration in Eq. (30), using this solution, gives the rate of quantum nucleation for a thin ferromagnetic film as
\[ \Gamma_Q \propto \exp\left(-\frac{SE}{\hbar}\right) \]
\[ = \exp\left\{ -74.39 S e^{3/4 r_0^2 h} \left[ 1 - \frac{2}{3} \left( \frac{1}{1 + |\cot \theta_H|^{2/3}} \right) \frac{7}{1 + |\cot \theta_H|^{2/3}} \right] \right\} \times \frac{1}{\sqrt{1 - \epsilon + 16 K_2^2 + 4 \left( K_2 - K_2' \right) \frac{1}{1 + |\cot \theta_H|^{2/3}} + 128 K_2 \left( K_2 - K_2' \right) \frac{|\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}}}}. \] (32)

At high temperature, the nucleation of \( M \) is due to thermal activation, and the rate of nucleation follows \( \Gamma_T \propto \exp\left(-\frac{W_{\text{min}}}{k_B T}\right) \), where \( W_{\text{min}} \) is the minimal work necessary to produce a nucleus capable of growing. In this case the instanton solution becomes independent of the imaginary-time variable \( \tau \). In order to obtain \( W_{\text{min}} \), we consider the effective potential of the system

\[ U_{\text{eff}} = \int d^3 r E = \int d^3 r \left[ \frac{\alpha}{2} M_0^2 \left( (\nabla \theta)^2 + \sin^2 \theta (\nabla \phi)^2 \right) + E_a(\theta, \phi) \right]. \] (33)

For a cylindrical bubble Eq. (33) reduces to

\[ U_{\text{eff}} = 4\pi K_1 h r_0^2 \int_0^\infty d\rho \left[ \frac{1}{2} \left( \frac{d\delta}{d\rho} \right)^2 + \frac{1}{4} \left( \sqrt{6 \delta^2 - \delta} \right)^2 \right]. \] (34)

From the saddle point of the functional the shape of the critical nucleus satisfies

\[ \frac{d^2 \delta}{d\rho^2} + \frac{1}{\rho} \frac{d\delta}{d\rho} = \frac{\sqrt{6 - 3 \delta^2}}{2} - \frac{3 \delta^2}{4}. \] (35)

The solution can be found by numerical method similar to the one in Refs. 4 and 6. Fig. 2 shows the shape of the critical bubble in thermal nucleation, and the maximal size is 3.906 at \( \tau = 0 \). Using this result, the minimal work corresponding the thermal nucleation is

\[ W_{\text{min}} = 41.3376 K_1 h r_0^2. \] (36)

Comparing this with Eq. (32), we obtain the approximate formula for the temperature characterizing the crossover from thermal to quantum nucleation as

\[ k_B T_c \approx 0.55 \frac{K_1 e^{1/4 r_0^2 h}}{S} \frac{1}{1 + |\cot \theta_H|^{2/3}} \left[ 1 + \frac{2}{3} \left( \frac{1}{1 + |\cot \theta_H|^{2/3}} \right) \frac{7}{1 + |\cot \theta_H|^{2/3}} \right] \times \left[ 1 - \epsilon + 16 K_2^2 + 4 \left( K_2 - K_2' \right) \frac{1}{1 + |\cot \theta_H|^{2/3}} \right]^{1/2} + 128 K_2 \left( K_2 - K_2' \right) \frac{|\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}}. \] (37)
To observe the quantum nucleation one needs a large crossover temperature and not too small a nucleation rate. Eq. (37) shows that ferromagnets with large anisotropy, i.e., small ratio of $K_2'$ to $K_1$, and small saturated magnetization are preferable for experimental study. In Fig. 3, we plot the $\theta_H$ dependence of the crossover temperature $T_c$ for typical values of parameters for nanometer-scale ferromagnets: $K_1 = 10^7$ erg/cm$^3$, $K_2' = 0.1$, $K_2 - K_2' = 0.01$, $M_0 = 500$ emu/cm$^3$, $\epsilon = 0.01$ in a wide range of angles $\pi/2 < \theta_H < \pi$. Fig. 3 shows that the maximal value of $T_c$ is about 225 mK at $\theta_H = 1.743$. The maximal value of $T_c$ as well as $\Gamma_Q$ is expected to be observed in experiment. The similar $\theta_H$ dependence of the crossover temperature $T_c$ was first observed in Ref. 15, while the problem considered in Ref. 15 was homogeneous spin tunneling in single-domain particles with uniaxial symmetry.

IV. FERROMAGNETS WITH HEXAGONAL SYMMETRY

In this section, we study the quantum nucleation of magnetization in nanometer-scale ferromagnets with hexagonal symmetry in an external magnetic field at an arbitrary angle in the ZX plane. Now the magnetocrystalline anisotropy energy $E_a(\theta, \phi)$ can be written as

$$E_a(\theta, \phi) = K_1 \sin^2 \theta + K_2 \sin^4 \theta + K_3 \sin^6 \theta - K_3' \sin^6 \theta \cos (6\phi) - M_0 H_z \sin \theta \cos \phi - M_0 H_\perp \cos \theta,$$

where $K_1$, $K_2$, $K_3$, and $K_3'$ are the magnetic anisotropic coefficients. The easy axes are $\pm \hat{z}$ for $K_1 > 0$. By choosing $K_3' > 0$, we take $\phi = 0$ to be the easy plane, at which the anisotropy energy can be written in terms of the dimensionless parameters as

$$\overline{E}_a(\theta, \phi = 0) = \frac{1}{2} \sin^2 \theta + \overline{K}_2 \sin^4 \theta + \left(\overline{K}_3 - \overline{K}_3'\right) \sin^6 \theta - \overline{H} \cos (\theta - \theta_H) + \overline{E}_0,$$

where $\overline{K}_3 = K_3/2K_1$ and $\overline{K}_3' = K_3'/2K_1$.

Then the initial angle $\theta_0$ is determined by $[d\overline{E}_a(\theta, 0) / d\theta]_{\theta=\theta_0} = 0$, and the critical angle $\theta_c$ and the dimensionless critical field $\overline{H_c}$ by both $[d\overline{E}_a(\theta, 0) / d\theta]_{\theta=\theta_c, \overline{H} = \overline{H}_c} = 0$ and $[d^2\overline{E}_a(\theta, 0) / d\theta^2]_{\theta=\theta_c, \overline{H} = \overline{H}_c} = 0$, which leads to
\[ \frac{1}{2} \sin(2\theta_0) + \mathcal{H} \sin(\theta_0 - \theta_H) + 4K_2 \sin^3 \theta_0 \cos \theta_0 + 6 \left( K_3 - K_3' \right) \sin^5 \theta_0 \cos \theta_0 = 0, \quad (40a) \]

\[ \frac{1}{2} \sin(2\theta_c) + \mathcal{H}_c \sin(\theta_c - \theta_H) + 4K_2 \sin^3 \theta_c \cos \theta_c + 6 \left( K_3 - K_3' \right) \sin^5 \theta_c \cos \theta_c = 0, \quad (40b) \]

\[ \cos(2\theta_c) + \mathcal{H}_c \cos(\theta_c - \theta_H) + 4K_2 \left( 3 \sin^2 \theta_c \cos^2 \theta_c - \sin^4 \theta_c \right) + 6 \left( K_3 - K_3' \right) \left( 5 \sin^4 \theta_c \cos^2 \theta_c - \sin^6 \theta_c \right) = 0, \quad (40c) \]

Under the assumption that \(|\mathcal{H}_2|, |K_3 - K'_3| \ll 1\), we obtain the dimensionless critical field \(\mathcal{H}_c\) and the critical angle as

\[ \mathcal{H}_c = \frac{1}{\left[ \sin(\theta_H)^{2/3} + |\cos(\theta_H)|^{2/3} \right]^{3/2}} \left[ 1 + \frac{4K_2}{1 + |\cot(\theta_H)|^{2/3}} + \frac{6 \left( K_3 - K_3' \right)}{\left( 1 + |\cot(\theta_H)|^{2/3} \right)^2} \right], \quad (41a) \]

\[ \sin(\theta_c) = \frac{1}{\left( 1 + |\cot(\theta_H)|^{2/3} \right)^{1/2}} \left[ 1 + \frac{8K_2}{3} \frac{|\cot(\theta_H)|^{2/3}}{1 + |\cot(\theta_H)|^{2/3}} \right] + 8 \left( K_3 - K_3' \right) \frac{|\cot(\theta_H)|^{2/3}}{\left( 1 + |\cot(\theta_H)|^{2/3} \right)^2}. \quad (41b) \]

In the limit of small \(\epsilon = 1 - \mathcal{H}/\mathcal{H}_c\), Eq. (40a) becomes

\[ -\epsilon \mathcal{H}_c \sin(\theta_c - \theta_H) + \eta^2 \left[ (3/2) \mathcal{H}_c \sin(\theta_c - \theta_H) + 3K_2 \sin(4\theta_c) \right] + 12 \left( K_3 - K_3' \right) \sin^3 \theta_c \cos \theta_c \left( 5 - 8 \sin^2 \theta_c \right) + \eta \left\{ \epsilon \mathcal{H}_c \cos(\theta_c - \theta_H) \right. \]

\[ -\eta^2 \left[ (1/2) \mathcal{H}_c \cos(\theta_c - \theta_H) + 4K_2 \cos(4\theta_c) \right] + 12 \left( K_3 - K_3' \right) \sin^2 \theta_c \left( 5 - 20 \sin^2 \theta_c + 16 \sin^4 \theta_c \right) \right\} = 0, \quad (42) \]

where \(\eta \equiv \theta_c - \theta_0\) which is small for \(\epsilon \ll 1\). By introducing a small variable \(\delta \equiv \theta - \theta_0\) (\(|\delta| \ll 1\) in the limit of \(\epsilon \ll 1\)), the anisotropy energy becomes

\[ \mathcal{E}_a(\delta, \phi) = K_3 \left[ 1 - \cos(6\phi) \right] \sin^6(\theta_0 + \delta) + \mathcal{H}_x (1 - \cos \phi) \sin(\theta_0 + \delta) + \mathcal{E}_1(\delta), \quad (43) \]

where \(\mathcal{E}_1(\delta)\) is a function of only \(\delta\) given by

\[ \mathcal{E}_1(\delta) = \left[ \frac{1}{2} \mathcal{H}_c \sin(\theta_c - \theta_H) + K_2 \sin(4\theta_c) + 4 \left( K_3 - K_3' \right) \left( 5 \sin^3 \theta_c \cos^3 \theta_c - 3 \sin^5 \theta_c \cos \theta_c \right) \right] \]

\[ \times (\delta^3 - 3\delta^2 \eta) + \left[ \frac{1}{8} \mathcal{H}_c \cos(\theta_c - \theta_H) + K_2 \cos(4\theta_c) + 3 \left( K_3 - K_3' \right) \sin^2 \theta_c \left( \sin^4 \theta_c \right. \right. \]

\[ \times \sin^2 \theta_c - \sin^4 \theta_c \right) \]
\[-10 \sin^2 \theta_c \cos^2 \theta_c + 5 \cos^4 \theta_c \] \((\delta^4 - 4\delta^3 \eta + 6\delta^2 \eta^2 - 4\delta^2 \epsilon) + \epsilon \delta^2 \left[4K_2 \cos (4 \theta_c) \right. \\
+12 \left(K_3 - K_3' \right) \sin^2 \theta_c \left(\sin^4 \theta_c - 10 \sin^2 \theta_c \cos^2 \theta_c + 5 \cos^4 \theta_c \right) \right] \] \quad (44)

By applying the similar procedure in Sec. III, we obtain the transition amplitude Eqs. (21) and (22) by integrating out \(\phi\). For this case the effective mass is

\[
M = \left(1 + |\cot \theta_H|^{2/3} \right) \left[1 + \frac{8}{3} K_2 \frac{|\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} + 8 \left(K_3 - K_3' \right) \frac{|\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} \right] \\
\times \left[1 - \epsilon + 36 K_3 \frac{1}{1 + |\cot \theta_H|^{2/3}} + 4 K_2 \left(1 + 120 K_3' \frac{|\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} \right) \frac{1}{1 + |\cot \theta_H|^{2/3}} \right]^{-1} \\
+ 6 \left(K_3 - K_3' \right) \left(1 + 240 K_3' \frac{|\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} \right) \frac{1}{1 + |\cot \theta_H|^{2/3}} \right]^{-1}, \quad (45)
\]

and the prefactor \(A\) is

\[
A = 2 \frac{|\cot \theta_H|^{1/3}}{1 + |\cot \theta_H|^{2/3}} \left[1 + \frac{4}{3} K_2 \frac{7 - 4 |\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} + 2 \left(K_3 - K_3' \right) \frac{11 - 16 |\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} \right] \right] \\
\times \left[1 - \epsilon + 36 K_3' \frac{1}{1 + |\cot \theta_H|^{2/3}} + 4 K_2 \left(1 + 120 K_3' \frac{|\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} \right) \frac{1}{1 + |\cot \theta_H|^{2/3}} \right]^{-1/2} \\
+ 6 \left(K_3 - K_3' \right) \left(1 + 240 K_3' \frac{|\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} \right) \frac{1}{1 + |\cot \theta_H|^{2/3}} \right]^{-1/2}, \quad (46)
\]

In case of a small ferromagnet of volume \(V \ll r_0^3\), the result of quantum nucleation is \(\Gamma_Q \propto \exp \left(-S_{cl}/\hbar\right)\), where the classical action for hexagonal symmetry is found to be

\[
S_{cl} = \frac{2^{17/4} \times 3^{1/4} \times \epsilon^{5/4}}{5} h S \epsilon^{5/4} |\cot \theta_H|^{1/6} \\
\times \left[1 + \frac{2}{3} K_2 \frac{7 - 2 |\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} + \left(K_3 - K_3' \right) \frac{11 - 12 |\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} \right] \\
\times \left[1 - \epsilon + 36 K_3' \frac{1}{1 + |\cot \theta_H|^{2/3}} + 4 K_2 \left(1 + 120 K_3' \frac{|\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} \right) \frac{1}{1 + |\cot \theta_H|^{2/3}} \right]^{-1/2} \\
+ 6 \left(K_3 - K_3' \right) \left(1 + 240 K_3' \frac{|\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} \right) \frac{1}{1 + |\cot \theta_H|^{2/3}} \right]^{-1/2}, \quad (47)
\]

In case of a thin film of thickness \(h\) less than the size \(r_0/\epsilon^{1/4}\) of the critical nucleus, we obtain the quantum nucleation as \(\Gamma_Q \propto \exp \left(-S_E/\hbar\right)\), with the classical action
\[ S_E = 74.39 \varepsilon^{3/4} r_0^2 \hbar \frac{1 + |\cot \theta_H|^{2/3}}{|\cot \theta_H|^{1/6}} \]

\[ \times \left[ 1 - \frac{2}{3} \overline{K}_2 \frac{7 - 2 |\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} - (\overline{K}_3 - \overline{K}_3') \frac{11 - 12 |\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} \right] \]

\[ \times \left[ 1 - \epsilon + 36 \overline{K}_3 \frac{1}{1 + |\cot \theta_H|^{2/3}} + 4 \overline{K}_2 \left( 1 + 120 \overline{K}_3 \frac{|\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} \right) \frac{1}{1 + |\cot \theta_H|^{2/3}} \right] \]

\[ + 6 \left( \overline{K}_3 - \overline{K}_3' \right) \left( 1 + 240 \overline{K}_3 \frac{|\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} \right) \frac{1}{1 + |\cot \theta_H|^{2/3}} \right]^{-1/2} \tag{48} \]

And the crossover temperature for hexagonal symmetry is found to be

\[ k_B T_c \approx 0.55 \frac{K_1 \varepsilon^{1/4}}{S} \frac{1}{1 + |\cot \theta_H|^{2/3}} \]

\[ \times \left[ 1 + \frac{2}{3} \overline{K}_2 \frac{7 - 2 |\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} + (\overline{K}_3 - \overline{K}_3') \frac{11 - 12 |\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} \right] \]

\[ \times \left[ 1 - \epsilon + 36 \overline{K}_3 \frac{1}{1 + |\cot \theta_H|^{2/3}} + 4 \overline{K}_2 \left( 1 + 120 \overline{K}_3 \frac{|\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} \right) \frac{1}{1 + |\cot \theta_H|^{2/3}} \right] \]

\[ + 6 \left( \overline{K}_3 - \overline{K}_3' \right) \left( 1 + 240 \overline{K}_3 \frac{|\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} \right) \frac{1}{1 + |\cot \theta_H|^{2/3}} \right]^{1/2} \tag{49} \]

V. CONCLUSIONS AND DISCUSSIONS

In summary we have investigated the quantum nucleation of magnetization in nanometer-scale ferromagnets in the presence of an external magnetic field at arbitrary angle. We consider the magnetocrystalline anisotropy with tetragonal symmetry and that with hexagonal symmetry, respectively. By applying the instanton method in the spin-coherent-state path-integral representation, we obtain the analytical formulas for quantum reversal of magnetization in small magnets and the numerical formulas for quantum nucleation in thin ferromagnetic film in a wide range of angles \( \pi/2 < \theta_H < \pi \). The temperature characterizing the crossover from the quantum to thermal nucleation is clearly shown for each case. Our
results show that the rate of quantum nucleation and the crossover temperature depend on the orientation of the external magnetic field distinctly. Therefore, both the orientation and the strength of the external magnetic field are the controllable parameters for the experimental test of quantum nucleation of magnetization in nanometer-scale ferromagnets. If the experiment is to be performed, there are three control parameters for comparison with theory: the angle of the external magnetic field $\theta_H$, the strength of the field in terms of $\epsilon$, and the temperature $T$. Our results show that ferromagnetic samples with large anisotropy and small saturated magnetization are suitable for experimental study the phenomenon of quantum nucleation.

Recently, Wernsdorfer and co-workers have performed the switching field measurements on individual ferrimagnetic and insulating BaFeCoTiO nanoparticles containing about $10^5$-$10^6$ spins at very low temperatures (0.1-6K). They found that above 0.4K, the magnetization reversal of these particles is unambiguously described by the Néel-Brown theory of thermal activated rotation of the particle’s moment over a well defined anisotropy energy barrier. Below 0.4K, strong deviations from this model are evidenced which are quantitatively in agreement with the predictions of the MQT theory without dissipation. It is noted that the observation of quantum nucleation in ferromagnets would be extremely interesting as the next example, after single-domain nanoparticles, of macroscopic quantum tunneling. The theoretical results presented here may be useful for checking the general theory in a wide range of systems, with more general symmetries. The experimental procedures on single-domain ferromagnetic nanoparticles of Barium ferrite with uniaxial symmetry may be applied to the systems with more general symmetries. Note that the inverse of the WKB exponent $B^{-1}$ is the magnetic viscosity $S$ at the quantum-tunneling-dominated regime $T \ll T_c$ studied by magnetic relaxation measurements. Therefore, the quantum nucleation of magnetization should be checked at any $\theta_H$ by magnetic relaxation measurements. Over the past years a lot of experimental and theoretical works were performed on the spin tunneling in molecular Mn$_{12}$-Ac and Fe$_8$ clusters having a collective spin state $S = 10$ (in this paper $S = 10^6$). Further experiments should focus on the level quantization of collective
spin states of \( S = 10^2 - 10^4 \).

The ferromagnet is typically an insulating particle with as many as \( 10^3 \sim 10^6 \) magnetic moments. For the dynamical process, it is important to include the effect of the environment on quantum tunneling caused by phonons,\(^3\) nucleation spins,\(^2\) and Stoner excitations and eddy currents in metallic magnets.\(^1\) However, many studies showed that even though these couplings might be crucial in macroscopic quantum coherence, they are not strong enough to make the quantum tunneling unobservable.\(^1\) The theoretical calculations performed in this paper can be extended to the AFM bubbles, where the relevant quantity is the excess spin due to the small noncompensation of two sublattices. Work along this line is still in progress. We hope that the theoretical results presented in this paper may stimulate more experiments whose aim is observing quantum nucleation in nanometer-scale ferromagnets.

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Figure Captions:

Fig. 1 The instanton, corresponding to subbarrier bubble formation in a thin film by quantum tunneling, for $\tau = 0$, $\tau = \pm 0.5$, $\tau = \pm 1$, $\tau = \pm 1.5$, and $\tau = \pm 2$.

Fig. 2 The shape of the critical bubble in a thermal nucleation of magnetization.

Fig. 3 The $\theta_H$ dependence of the crossover temperature $T_c$ for $\pi/2 < \theta_H < \pi$. Here,
\[ K_1 = 10^7 \text{ erg/cm}^3, \quad K_2' = 0.1, \quad K_2 - K_2' = 0.01, \quad M_0 = 500 \text{ emu/cm}^3, \text{ and } \epsilon = 0.01. \]
\[ \delta \rho \]

\[ \tau = 0 \]
\[ \tau = 0.5 \]
\[ \tau = 1 \]
\[ \tau = 2 \]
\[ \delta \rho \]
