Quantum-classical crossover of the escape rate in the cubic nanospin system with a longitudinal field

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The quantum-classical crossover of the escape rate is studied in the cubic spin model based on the spin-coherent-state path integral with the help of the instanton technique. It is found that for all values of a longitudinal field the crossover is of the first order in the easy axis [001], while of the second order in the easy axis [111]. This feature is expected to be observable in existing experimental techniques.

Recently the nanospin systems have been good candidates to display macroscopic quantum phenomena (MQP).[1] One of several candidates is a single domain ferromagnetic particle with the magnetization **M** whose direction is determined by the magnetocrystalline anisotropy energy depending on the crystal symmetry. The direction of **M** is changed by two processes: thermal activation over the energy barrier and quantum tunneling through it. The former dominates at high temperature, while the latter does so at low temperature. Thus, there is a crossover temperature T_0 between two regimes. In this situation, whether the crossover about T_0 can be sharp (the first-order crossover) or smooth (the second-order crossover, SC) is determined by the magnetic anisotropy constants and the external magnetic field. For the dynamical situation, it is important to include the effect of the environment on the escape rate caused by phonons,[2] nuclear spins,[3] and Stoner excitations and eddy currents in metallic magnets.[4] However, many studies have shown that the effects are not strong enough to make the escape rate extremely small.

The question of the crossover of the escape rate was studied by Affleck,[5] and Larkin and Ovchinnikov,[6] who showed that SC can occur at T_0 by using the standard instanton technique. Chudnovsky[7] gave the criterion to determine first- or second-order crossover (FSC) based on the behavior of the period of oscillations in the inverted potential. Since then, many theoretical studies have been performed in nanospin systems with uniaxial or biaxial symmetry (UBS) based on the simple mapping of the spin problem onto the particle one[8–11] and the periodic instantons.[12] However, the methods used in early calculations for UBS cannot be applied to other models of practical interest such as cubic symmetry, because in such a case it is not possible to obtain the one-dimensional functional form of the action. Noting that in various anisotropy energies MQP can be treated by using the spin-coherent-state path-integral (SCSPI) method,[1] it will be highly required to have a new approach for the crossover in SCSPI. In this paper, employing the nonlinear perturbation near the top of the barrier in one dimension studied in Ref. 13, we briefly present the theoretical method to study the crossover for the system in SCSPI and apply it to the problem of the crossover in the cubic system with a longitudinal field. In general, great care must be taken in SCSPI with obtaining the exact tunneling rate, because the terms of $O(S^0)$ in the exponent which give the prefactor in the tunneling rate are incorrectly obtained.[14] However, whether the system with large $S(\gg 1)$ becomes FSC is mostly determined by the terms of $O(S)$ in the exponent of the tunneling rate.[9] Accordingly, our consideration will be focused on the behavior of the WKB exponent. Considering two cases in cubic symmetry, we find that FSC in this system is determined by the sign of the anisotropy constant and there is only the first-order crossover in the easy axis [001]. This result is of interest in the following perspective. In general, it is well known that a large magnetic field but slightly less than the critical field is a prerequisite for observing MQP in large spin systems $(S\gg 10)$.[1] Meanwhile, the previous theoretical studies about the crossover have shown that the first-order regime in UBS can be observed in a small magnetic field and substantially decreases with increasing the magnetic field. In order to observe the first-order crossover in those systems, their total spins should not be large. Thus, the molecular clusters with $S=10$ such as Mn_{12} (Ref. 18) and Fe_8 (Ref. 19) might be good candidates for observing FSC in UBS. If the total spin is larger $(S\gg 10)$, it is difficult to observe the first-order crossover in UBS. However, in the cubic system the situation will be completely different. We shall show that in the case of the easy axis [001] the first-order crossover can be found for all values of the longitudinal field. This indicates that such a large spin system can be also a good candidate for observing the first-order crossover, which is deemed as a practical advantage in future experiments.

Consider the SCSPI representation of the partition function given by

$$Z(\beta\hbar)=\oint D[\mathbf{M}(\tau)]\exp(-S_E/\hbar), \tag{1}$$

where $\beta=1/k_BT$, the path sum is over all periodic paths $\mathbf{M}(\tau)=\mathbf{M}(\tau+\beta\hbar)$, and S_E is the action which includes the Euclidean version of the magnetic Lagrangian L_E as

$$S_E=\int_0^{\beta\hbar}\left\{i\frac{M}{\gamma}[1-\cos\theta(\tau)]\frac{d\phi(\tau)}{d\tau}+E[\mathbf{M}(\tau)]\right\}d\tau, \tag{2}$$

where $\gamma=g\mu_B/\hbar$ and μ_B is the Bohr magneton. Quite generally, the energy E in Eq. (2) includes the magnetic anisotropy energy E_a, the exchange energy $E_{\rm ex}$, the magnetic dipole-dipole interaction $E_{\rm dd}$, and the Zeeman energy E_H, which determine the switching process of the system. In many cases $E_{\rm dd}$ can be contained in E_a, which leads to the effective anisotropy energy. In ferrimagnetic or antiferromagnetic particles the exchange energy should be enhanced. Using the two sublattice model,[20] the effect of the exchange interaction on the crossover can be neglected in the limit of $y=(K/J)\times(s_2/s_1)\ll 1$, where K is the anisotropy constant, J is the exchange constant, and $s_{1,2}$ is the number of spin in each sublattice.[21] For $K\sim 1$ K, $J\sim 100$ K, and $s_2/s_1<1$, we obtain $y\ll 1$, which is prevalent in many ferromagnetic particles. In this paper we will not take the exchange interaction into account and will simplify the system with $E=E_a+E_H$.

Now, from Eq. (2) the classical trajectory of θ and ϕ is determined by the Euler-Lagrange equation as

$$iMs_\theta\dot{\phi}=-\gamma E_\theta, \quad iMs_\theta\dot{\theta}=\gamma E_\phi, \tag{3}$$

where $s_\theta\equiv\sin\theta$, $\dot{\phi}=d\phi/d\tau$, $E_\theta=\partial E/\partial\theta$ and so on. In order to study the criterion of FSC for the escape rate, we need to decompose θ (ϕ) into the position of the top of the barrier $\overline{\theta}$ ($\overline{\phi}$) and a fluctuation term $\delta\theta$ ($\delta\phi$), i.e., $\theta=\overline{\theta}+\delta\theta$ ($\phi=\overline{\phi}+\delta\phi$) for the behavior of the weakly time-dependent solutions. Here the solutions of the equation of motion are parametrized by the amplitude a of the oscillations, which quantifies the difference between the thermal and the time-dependent solutions near the top of the barrier. Our goal is to solve Eq. (3) for $\delta\theta(\tau)$ and $\delta\phi(\tau)$ and find the correction to the oscillation period away from the thermal saddle point. Denoting $\delta\mathbf{\Omega}(\tau)\equiv[\delta\theta(\tau), \delta\phi(\tau)]$, we have $\delta\mathbf{\Omega}(\tau+\beta\hbar)=\delta\mathbf{\Omega}(\tau)$ at finite temperature and write it as the Fourier series $\delta\mathbf{\Omega}(\tau)=\sum_{n=-\infty}^{\infty}\delta\mathbf{\Omega}_n\exp(i\tilde{\omega}_n\tau)$, where $\tilde{\omega}_n=2\pi n/\beta\hbar$. Proceeding the perturbation of $\delta\mathbf{\Omega}$, we will obtain the correction $\delta\omega(\equiv\omega-\omega_0)$ at higher order where ω_0 is a small oscillation frequency in the lowest order near the top of the barrier. Meanwhile, Chudnovsky[7] proposed that, if the oscillation period is a nonmonotonic (monotonic) function of a where a is a function of E in the absence of dissipation, the system exhibits the first-order crossover (the second-order crossover). Following the criterion, the corrected period $2\pi/\omega$ should be less (greater) than the period $2\pi/\omega_0$ in the lowest order, i.e., $\delta\omega>0$ for the first-order crossover and $\delta\omega<0$ for SC.

Now, we shall apply the formalism to the cubic symmetry whose anisotropy energy is $E_a=K(\alpha_x^2\alpha_y^2+\alpha_y^2\alpha_z^2+\alpha_z^2\alpha_x^2)$ where α's are the direction cosines of $\mathbf{M}$ and K the anisotropy constant. In the case of $K>0$ we have [001], $[00\overline{1}]$, [010], $[0\overline{1}0]$, [100], and $[\overline{1}00]$ for the easy axes. Without an external field these directions are stable, whereas in the presence of the longitudinal field the initial orientation of $\mathbf{M}$ becomes metastable, as is shown in Fig. 1. Choosing its initial direction to be $\hat{z}$ and applying the field along the $-\hat{z}$, we obtain the total energy

$$E(\theta,\phi)=K\sin^2\theta-\frac{K}{8}[7+\cos(4\phi)]\sin^4\theta+MH\cos\theta, \tag{4}$$

and the classical trajectory

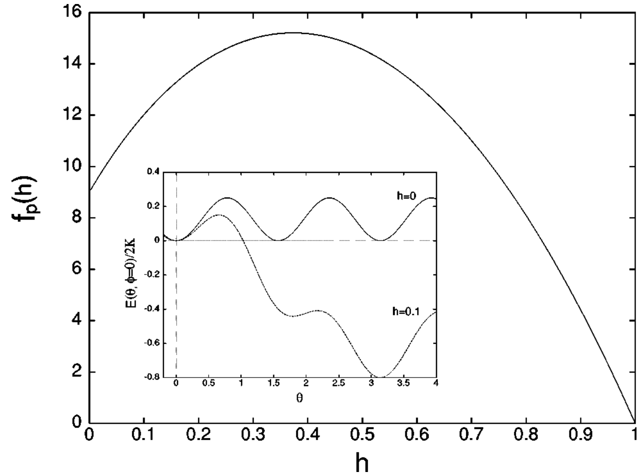

FIG. 1. $f_p(h)$ vs h in cubic symmetry with $K>0$. Inset: shape of energy (4) in the easy plane, $\phi=0$, where h is defined in the text.

$$2in_p\dot{\phi}+4c_\theta-(7+c_{4\phi})s_\theta^2c_\theta-4h=0, \tag{5}$$

$$2in_p\dot{\theta}-s_{4\phi}s_\theta^3=0, \tag{6}$$

where $M/(\gamma K)=n_p$, $H_c=2K/M$, and $h=H/H_c$. Here H_c is the critical magnetic field at which the barrier vanishes. In the thermal activation regime the solutions of Eqs. (5) and (6) are $\overline{\theta}=\theta_0$ and $\overline{\phi}=0$, where

$$\cos\theta_0=\frac{(9h-\sqrt{81h^2-6})^{1/3}+(9h+\sqrt{81h^2-6})^{1/3}}{6^{2/3}}. \tag{7}$$

Note that θ_0 decreases monotonically with increasing h and $0\leq h<1$ corresponds to $0<\theta_0\leq\pi/4$. In order to study FSC, we need to expand Eqs. (5) and (6) into a series around $(\theta_0,0)$ as $\theta=\theta_0+\delta\theta$ and $\phi=\delta\phi$. Simple analysis for Eqs. (4)–(6) shows that $\delta\theta$ is real and $\delta\phi$ imaginary. Thus, to lowest order in perturbation theory, we can write $\delta\theta\simeq a\theta_{p1}\cos(\omega\tau)$ and $\delta\phi\simeq ia\phi_{p1}\sin(\omega\tau)$. Substituting them into Eqs. (5) and (6) while neglecting terms of order higher than a, we have

$$\phi_{p1}/\theta_{p1}=(10s_{\theta_0}-12s_{\theta_0}^3)/(n_p\omega_0)=-n_p\omega_0/(2s_{\theta_0}^3), \tag{8}$$

where $\omega_0=\gamma H_cs_{\theta_0}^2\sqrt{5-6s_{\theta_0}^2}$.

In order to see the behavior of the oscillation period via the frequency we need to investigate Eqs. (5) and (6) by writing $\delta\theta\simeq a\theta_{p1}\cos(\omega\tau)+\delta\theta_2$, and $\delta\phi\simeq ia\phi_{p1}\sin(\omega\tau)+i\delta\phi_2$, where $\delta\theta_2$ and $\delta\phi_2$ are of the order of a^2. Neglecting terms of order higher than a^2, we find $\omega=\omega_0$, and the corresponding perturbations $\delta\theta_2=t_1+t_2\cos(2\omega\tau)$ and $\delta\phi_2=f\sin(2\omega\tau)$ with

$$t_1=-a^2\theta_{p1}^2c_{\theta_0}(25-42s_{\theta_0}^2)/[4s_{\theta_0}(5-6s_{\theta_0}^2)], \tag{9}$$

$$t_2=a^2\theta_{p1}^2c_{\theta_0}(15-22s_{\theta_0}^2)/[4s_{\theta_0}(5-6s_{\theta_0}^2)], \tag{10}$$

$$f=a^2\theta_{p1}^2(2c_{\theta_0})/\sqrt{5-6s_{\theta_0}^2}. \tag{11}$$

This implies that there is no shift in the oscillation frequency. In order to find the change of the oscillation period, we pro-

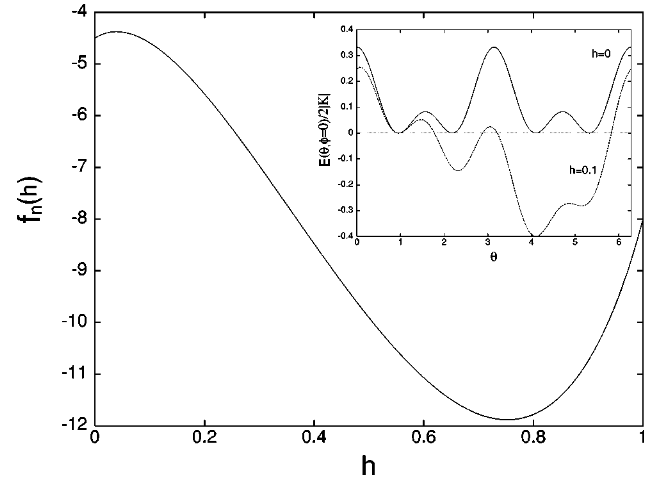

FIG. 2. $f_n(h)$ vs h in cubic symmetry with $K<0$. Inset: shape of energy (14) in the easy plane, $\phi=0$, where h is defined in the text.

ceed to the third order of perturbation theory by writing $\delta\theta \simeq a\,\theta_{p1}\cos(\omega\tau)+\delta\theta_2+\delta\theta_3$, and $\delta\phi\simeq i a\,\phi_{p1}\sin(\omega\tau)+i\delta\phi_2+i\delta\phi_3$. Inserting them again into Eqs. (5) and (6) and retaining only terms up to $O(a^3)$, we have for the change of the frequency

$$\omega^2-\omega_0^2=a^2\theta_{p1}^2(\gamma H_c/2)^2 f_p(\theta_0), \tag{12}$$

where

$$f_p(\theta)=\frac{3s_\theta^2(436+747c_{2\theta}+340c_{4\theta}+77c_{6\theta})}{40-48s_\theta^2}. \tag{13}$$

Using Eqs. (7) and (12), we obtain the correction of ω by varying h (Fig. 1) and $\omega>\omega_0$, i.e., $\tau<\tau_0$ where τ and τ_0 are the period of the instanton just below and on the top of the barrier, respectively. Since $\tau(E)$ begins to decrease with decreasing E from the top of the barrier but $\tau(E_0)$ eventually goes to ∞ where E_0 is the bottom of the metastable well, there should be a minimum in some energy between the bottom of the metastable well and the top of the barrier. This indicates that the whole shape of τ is nonmonotonic. Therefore, we can use Chudnovsky's criterion and find that the first-order crossover occurs at all value of h. In quantum tunneling of magnetization the practically interesting situation is when the barrier height is small and the width is narrow in order to have the escape rate large. Such a situation is realized when $\epsilon(\equiv 1-H/H_c)\ll 1$. In the meantime, in order to observe the first-order crossover in UBS, H should be as small as possible. If the total spin increases, e.g., $S \gg 10$, the escape rate becomes extremely small in a small magnetic field. The first-order regime vanishes in UBS with a large spin. Therefore, the result obtained in the cubic system with $K>0$ is very interesting because it gives some possibility of observing the first-order crossover in the system with a large spin ($S\gg 10$).

Let us now consider the crossover for $K<0$, i.e., the cubic system with an easy axes, e.g., $[111]$, $[\overline{11}1]$, $[11\overline{1}]$, and so on. In the absence of the field there are eight equivalent easy axes. However, applying the longitudinal field, the initial direction of **M** becomes metastable, which is illustrated in Fig. 2. Performing the coordinate transformation defined by $\alpha_x =(\alpha_{x'}-\alpha_{y'})/\sqrt{2}$, $\alpha_y=(\alpha_{x'}+\alpha_{y'})/\sqrt{2}$, and $\alpha_z=\alpha_{z'}$ and denoting θ and ϕ in $x'y'z'$, the total energy is represented as

TABLE I. Approximate expressions in the cubic symmetry for $H\lesssim H_c$, where $\delta=\theta-\theta_i$ and θ_i is the position of the metastable state.

	$K>0$	$K<0$
$E(\delta,0)/\vert K\vert$	$\epsilon\delta^2-(5/4)\delta^4$	$(2\epsilon\delta^2-\sqrt{2}\delta^3)/3$
θ_0	$\sqrt{2\epsilon/5}$	$\theta_i+2\sqrt{2}\epsilon/3$
$\omega_0/(\gamma\vert K\vert/M)$	$4\epsilon/\sqrt{5}$	$4\epsilon/\sqrt{3}$
t_1/a^2	$-5\theta_{p1}^2/4\theta_0$	$-\theta_{n1}^2/\sqrt{2}\epsilon$
t_2/a^2	$3\theta_{p1}^2/4\theta_0$	$\sqrt{2}\theta_{n1}^2/4\epsilon$
f/a^2	$2\theta_{p1}^2/\sqrt{5}$	$-2\theta_{n1}^2/4\epsilon$
$f_p(\theta_0)$ or $f_n(\theta_0)$	$120\ \ \theta_0^2$	-8

$$E=-|K|\left[\sin^2\theta-\frac{1}{8}(7-\cos(4\phi))\sin^4\theta\right] +MH\sin\theta_i\sin\theta\cos\phi+MH\cos\theta_i\cos\theta, \tag{14}$$

where the direction of the field is $\theta_i=\arccos(1/\sqrt{3})$ and $\phi=0$. The relation between h and the θ position of the top of the barrier ($\overline{\theta}=\theta_0$) is given by

$$h(\equiv H/H_c)=3s_{2\theta_0}(2-3s_{\theta_0}^2)/(8s_{\theta_i-\theta_0}), \tag{15}$$

where $H_c=4|K|/3M$. Also, it is noted that θ_0 decreases monotonically with increasing h and $0\leqslant h<1$ corresponds to $\theta_i<\theta_0\leqslant\pi/2$. The correction of the frequency for this case can be calculated in the same fashion. As before we perform the perturbation to third order and obtain the following equation for the shift in the oscillation frequency:

$$\omega^2-\omega_0^2=a^2\theta_{n1}^2(3\gamma H_c/4)^2 f_n(\theta_0), \tag{16}$$

where we used $\delta\theta\simeq a\,\theta_{n1}\cos(\omega\tau)$. Even though we have derived the analytic expression of ω_0 and $f_n(\theta_0)$, it is not illuminating to present their detailed forms which are complicated. We just illustrate the relation between $f_n(\theta_0)$ and h in Fig. 2. Evidently $f_n(\theta_0)<0$ for $h<1$. This implies that there is no first-order crossover in this case.

In Table I we summarize the quantities considered in the cubic symmetry for $\epsilon\ll 1$. The approximate potential form at easy plane ($\phi=0$) is $\epsilon\delta^2-(5/4)\delta^4$ for $K>0$. Quite generally, if the potential is of the form x^2-x^3 or x^2-x^4 in one dimension, the crossover is always second order.[7] However, in Eq. (12) we get $f_p(\theta_0)>0$, which implies that the crossover is first order for $H\lesssim H_c$. In fact, noting that such a potential is not obtained by the effective action in one dimension, it is not certain whether the form, $\delta^2-\delta^4$ gives rise to SC. In other words, if the action in Eq. (1) is reduced to the one with one-dimensional functional form in which the effective potential is of the form $\delta^2-\delta^4$, the crossover is always second order. However, it is not possible to obtain such an effective action and the corresponding potential in the cubic system for $K>0$. Therefore, our detailed analysis has shown that even though the form is $\delta^2-\delta^4$ in Table I, the system displays the first-order crossover.

To illustrate the above results with concrete numbers, we use the typical physical parameters. For K/V

$\sim 10^6$ erg/cm^3, $M/V \sim 100$ emu/cm^3, and $\epsilon \sim 10^{-2} - 10^{-3}$,[22] we have $T_0 \sim 30-3$ mK with $H_c \sim 2$ T. However, two cases in the cubic system are greatly different in the temperature range (ΔT) of the crossover region. Qualitative analysis shows that $\Delta T/T_0 \simeq 1/S$ in the first-order crossover and $\Delta T/T_0 \simeq 1/S^{1/2}$ in SC. For, e.g., $S \sim 10^2$, we obtain $\Delta T/T_0 \sim 0.01$ for the former and 0.1 for the latter, respectively. Thus, the larger spin, the more favorable to test FSC in real experiments.

In conclusion, using the spin-coherent-state path integral, we have studied the crossover in cubic systems with a longitudinal field. Considering two cases, we have found that the crossover is of the first order for $K>0$, while of the second order for $K<0$. The result is of interest theoretically and experimentally in three respects. First, until now most of theoretical studies have been performed in UBS. Whether the crossover becomes first or second order in those systems is determined by the relative magnitude of two anisotropy constants or the ratio of the field to the anisotropy constant. However, in the cubic system FSC is solely determined by the sign of the anisotropy constant. This is a unique feature which has not yet been found. Second, in UBS the first-order regime decreases greatly with increasing the field. This implies that the number of total spin should not be so large. However, in the cubic system the first-order crossover occurs for all values of the longitudinal field. Third, since the crossover region ($\Delta T/T_0$) is inversely proportional to the total spin, the sharpness of the first-order crossover will be more dramatic in larger spin system. These make the cubic system a good candidate for the experimental study.

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