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J. Phys.: Condens. Matter **14** (2002) 849–856 PII: S0953-8984(02)31334-1

Quantum–classical crossover in nanomagnetic systems

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Received 28 November 2001
Published 18 January 2002
Online at stacks.iop.org/JPhysCM/14/849

Abstract
Using the nonlinear perturbation method, we study a crossover between quantum and classical regimes for the escape rate. We present a general formula for determining whether the escape rate changes smoothly around the crossover temperature or not. Applying it to tetragonal and hexagonal symmetries, it is found that the crossover is mostly of first order.

1. Introduction

In recent years, nanospin systems have aroused considerable interest since the discovery that they provide instances which exhibit different types of crossover between thermal activation over the energy barrier and quantum tunnelling under the barrier [1–7]. One good subject is a single-domain ferromagnetic particle with the magnetization M whose direction is changed by the magnetic anisotropy energy depending on the crystal symmetry. At sufficiently high temperature the direction of M is changed by a classical process, in which the changing rate of M, i.e., the so-called escape rate Γ, obeys the Arrhenius law, $\Gamma \sim \exp(-U/k_B T)$, with U being the height of the energy barrier. At a temperature low enough for ignoring thermal fluctuation, M is changed by quantum tunnelling whose rate is $\Gamma \sim \exp(-U/\hbar\omega)$, where ω is the oscillation frequency in the well. Hence, there exists a temperature T_0 ($\sim\hbar\omega/k_B$) at which crossover between two regimes occurs. Up to now, two possible types of quantum–classical crossover, first- and second-order crossovers, have been suggested. At the first-order crossover the system crosses the energy barrier, changing abruptly with temperature, which leads to a steep change of the escape rate around T_0. However, at the second-order crossover the energy of the system changes smoothly with temperature, and the crossover occurs over a broad interval of temperature. Whether the crossover is a first- or second-order one in nanospin systems is mainly determined by the anisotropy energy and the external magnetic field.

The criterion of first- or second-order crossover for the escape rate was proposed by Chudnovsky, who showed that the escape rate changes in a broad or narrow interval of temperature around T_0 depending on the oscillation period $\tau(E)$ of the instanton [1] where E is the energy of the instanton. Many theoretical studies have been performed on nanospin systems that are uniaxial or biaxial systems, by employing a mapping of the spin problem onto a particle one and periodic instantons [2–4]. However, such methods cannot be applied to symmetries other than uniaxial or biaxial symmetry, because there are no one-dimensional functional forms of the actions in such cases. Recently, the present author has developed a theoretical method [8] for dealing with the crossover in cubic or ferrimagnetic nanospin systems by using nonlinear perturbation near the top of the barrier [9]. In this situation it would be interesting to know whether there is a general approach for treating the phenomena in major crystal symmetries. In this paper I will show that it is possible to establish such a general method in the situation where the easy plane is constant, e.g., $\phi = 0$. Also, choosing specific examples such as tetragonal and hexagonal symmetries, we will present complete analytic results on the phase boundary between first- and second-order crossovers.

2. Formulation of the problem

In this section we briefly discuss the basic formalism used to study the quantum–classical crossover of the escape rate in a ferromagnetic particle based on the spin-coherent-state path integral. In this case the Euclidean action in terms of the imaginary time ($\tau = \mathrm{i}t$) becomes

$$S_E(\theta, \phi) = V \int \mathrm{d}\tau \left[\mathrm{i}\frac{m}{\gamma}(1 - \cos\theta)\frac{\mathrm{d}\phi}{\mathrm{d}\tau} + E(\theta, \phi) \right] \tag{1}$$

where V is a volume of the particle, m the magnetization, γ the gyromagnetic ratio, and θ, ϕ spherical coordinates of the magnetization. Also, $E(\theta, \phi)$ is the total energy which is composed of the anisotropy energy and the energy given by an external magnetic field. The classical trajectory of θ and ϕ is determined by

$$\mathrm{i}n\dot{\phi}\sin\theta = -E_\theta \qquad \mathrm{i}n\dot{\theta}\sin\theta = E_\phi \tag{2}$$

where $n = m/\gamma$, $\dot{\phi} = \mathrm{d}\phi/\mathrm{d}\tau$, $\dot{\theta} = \mathrm{d}\theta/\mathrm{d}\tau$, $E_\theta = \partial E/\partial\theta$, and $E_\phi = \partial E/\partial\phi$. Employing the perturbation method for the criterion of first- or second-order crossover, the classical trajectory of θ (ϕ) is decomposed into the position of the barrier $\bar{\theta}$ ($\bar{\phi}$) and a fluctuation term $\delta\theta$ ($\delta\phi$), i.e., $\theta = \bar{\theta} + \delta\theta$ ($\phi = \bar{\phi} + \delta\phi$) for the behaviour of the weakly time-dependent solutions. 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 equation (2) for $\delta\theta(\tau)$ and $\delta\phi(\tau)$ and find the correction to the oscillation period away from the thermal saddle point. Using the notation $\delta\mathbf{\Omega}(\tau) \equiv (\delta\theta(\tau), \delta\phi(\tau))$, we have $\delta\mathbf{\Omega}(\tau + \beta\hbar) = \delta\mathbf{\Omega}(\tau)$ at finite temperature and we write it as Fourier series $\delta\mathbf{\Omega}(\tau) = \sum_{j=-\infty}^{\infty} \delta\mathbf{\Omega}_j \exp(\mathrm{i}\tilde{\omega}_j\tau)$ where $\tilde{\omega}_j = 2\pi j/\beta\hbar$ and $\beta = 1/k_\mathrm{B}T$. Proceeding with 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. According to Chudnovsky's criterion, we have $\delta\omega > 0$ for the first-order crossover and $\delta\omega < 0$ for the second-order one. Thus, we will discuss whether the corrected period $2\pi/\omega$ is less than the period $2\pi/\omega_0$ or not.

Now, let us apply our considerations to the anisotropy energy with a constant easy plane, e.g., $\phi = 0$. Writing $\bar{\theta} = \theta_0$, equation (2) is expressed in terms of $\delta\theta$ and $\delta\phi$, which results in

$$\mathrm{i}n(\delta\dot{\phi}) + A_1(\delta\theta) + A_2(\delta\theta)^2 + A_3(\delta\phi)^2 + A_4(\delta\theta)^3 + A_5(\delta\theta)(\delta\phi)^2 = 0 \tag{3}$$

$$\mathrm{i}n(\delta\dot{\theta}) + B_1(\delta\phi) + B_2(\delta\theta)(\delta\phi) + B_3(\delta\phi)^3 + B_4(\delta\theta)^2(\delta\phi) = 0 \tag{4}$$

where $\delta\dot{\phi} = \mathrm{d}(\delta\phi)/\mathrm{d}\tau$, $\delta\dot{\theta} = \mathrm{d}(\delta\theta)/\mathrm{d}\tau$, and

$$\begin{aligned} A_1 &= E_{\theta\theta}\,\mathrm{cosec}\,\theta_0 \\ A_2 &= \tfrac{1}{2}E_{\theta\theta\theta}\,\mathrm{cosec}\,\theta_0 - E_{\theta\theta}\cot\theta_0\,\mathrm{cosec}\,\theta_0 \\ A_3 &= \tfrac{1}{2}E_{\phi\phi\theta}\,\mathrm{cosec}\,\theta_0 \end{aligned} \tag{5}$$

$$A_4 = \tfrac{1}{6}E_{\theta\theta\theta\theta}\operatorname{cosec}\theta_0 - \tfrac{1}{2}E_{\theta\theta\theta}\cot\theta_0\operatorname{cosec}\theta_0 + E_{\theta\theta}(\tfrac{1}{2}+\cot^2\theta_0)\operatorname{cosec}\theta_0 \quad (6)$$

$$A_5 = \tfrac{1}{2}E_{\theta\theta\phi\phi}\operatorname{cosec}\theta_0 - \tfrac{1}{2}E_{\phi\phi\theta}\cot\theta_0\operatorname{cosec}\theta_0 \quad (7)$$

$$\begin{aligned} B_1 &= -E_{\phi\phi}\operatorname{cosec}\theta_0 \\ B_2 &= -E_{\theta\phi\phi}\operatorname{cosec}\theta_0 + E_{\phi\phi}\cot\theta_0\operatorname{cosec}\theta_0 \\ B_3 &= -\tfrac{1}{6}E_{\phi\phi\phi\phi}\operatorname{cosec}\theta_0 \end{aligned} \quad (8)$$

$$B_4 = -\tfrac{1}{2}E_{\theta\theta\phi\phi}\operatorname{cosec}\theta_0 + E_{\theta\phi\phi}\cot\theta_0\operatorname{cosec}\theta_0 - E_{\phi\phi}(\tfrac{1}{2}+\cot^2\theta_0)\operatorname{cosec}\theta_0. \quad (9)$$

Further, we introduce $E_{\theta\theta} = [\partial^2 E/\partial\theta^2]_{\theta=\theta_0,\phi=0}$, $E_{\phi\phi\theta} = [\partial^3 E/\partial\phi^2\partial\theta]_{\theta=\theta_0,\phi=0}$, and so on. Considering the system in which $\delta\theta$ is real and $\delta\phi$ imaginary, we can write $\delta\theta \simeq a\theta_1\cos(\omega\tau)$ and $\delta\phi \simeq \mathrm{i}a\phi_1\sin(\omega\tau)$ to lowest order in perturbation theory. Substituting them into equations (3) and (4) while neglecting terms of order higher than a, we obtain

$$\frac{\phi_1}{\theta_1} = \frac{A_1}{n\omega_0} = \frac{n\omega_0}{B_1} \quad (10)$$

where it is noted that $\omega_0 = \sqrt{A_1 B_1}/n$.

In order to find the change of the oscillation frequency, we need to investigate equations (3) and (4) by choosing $\delta\theta \simeq a\theta_1\cos(\omega\tau) + \delta\theta_2$, and $\delta\phi \simeq \mathrm{i}a\phi_1\sin(\omega\tau) + \mathrm{i}\delta\phi_2$, 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 $\delta\theta_2 = a^2\theta_1^2[t_1 + t_2\cos(2\omega\tau)]$ and $\delta\phi_2 = a^2\theta_1^2[f_1 + f_2\sin(2\omega\tau)]$ with

$$t_1 = \frac{A_1A_3 - A_2B_1}{2A_1B_1} \quad (11)$$

$$t_2 = \frac{2A_1B_2 + A_2B_1 + A_1A_3}{6A_1B_1} \quad (12)$$

$$f_1 = 0 \quad (13)$$

$$f_2 = \frac{A_1B_2 + 2A_2B_1 + 2A_1A_3}{6n\omega_0B_1}. \quad (14)$$

Since the oscillation frequency does not change in this order, the higher order should be taken into account by writing $\delta\theta \simeq a\theta_1\cos(\omega\tau) + \delta\theta_2 + \delta\theta_3$ and $\delta\phi \simeq \mathrm{i}a\phi_1\sin(\omega\tau) + \mathrm{i}\delta\phi_2 + \mathrm{i}\delta\phi_3$, where $\delta\theta_2$ and $\delta\phi_2$ are of the order of a^3. Inserting them again into equations (3) and (4) and retaining only terms up to $\mathrm{O}(a^3)$, we have for the shift of the frequency

$$\omega^2 - \omega_0^2 = \left(\frac{a\theta_1}{n}\right)^2 (g_1 + g_2 + g_3) \quad (15)$$

where

$$g_1 = 2A_2B_1(t_1 + \tfrac{1}{2}t_2) - A_3(f_2 n\omega_0) \quad (16)$$

$$g_2 = \tfrac{1}{2}B_2(f_2 n\omega_0) + A_1B_2(t_1 - \tfrac{1}{2}t_2) \quad (17)$$

$$g_3 = \frac{1}{4}\left(3A_4B_1 - A_1A_5 + A_1B_4 - 3\frac{A_1^2B_3}{B_1}\right). \quad (18)$$

Now, we shall apply the formalism to the crossover in the presence of a longitudinal magnetic field for tetragonal and hexagonal symmetries.

3. Crossover in tetragonal symmetry

Choosing $\hat{z}$ to be the initial easy axis for the anisotropy constant $K_1 > 0$, the energy is expressed as

$$E(\theta,\phi) = K_1\sin^2\theta + [K_2 - K_2'\cos(4\phi)]\sin^4\theta \quad (19)$$

where $K_1 \gg K_2, K_2' > 0$. Introducing the dimensionless parameters $k = K_2/K_1$ and $q = K_2'/K_1$, the energy on the easy plane $\phi = 0$ is represented as

$$E(\theta, \phi = 0) = K_1[\sin^2\theta + (k - q)\sin^4\theta]. \tag{20}$$

Noting that $\theta_0 = \pi/2$ in equation (20), we obtain from equations (5)–(9)

$$A_1 = -2 - 4(k - q) \qquad A_2 = A_3 = 0 \qquad A_4 = \frac{1 + 14(k - q)}{3} \tag{21}$$

$$A_5 = -32q \qquad B_1 = -16q \qquad B_2 = 0 \qquad B_3 = \tfrac{128}{3}q \qquad B_4 = 24q. \tag{22}$$

Substituting them into equations (10)–(15), the shift of the frequency becomes

$$\omega^2 - \omega_0^2 = 8\left(\frac{a\theta_1}{n}\right)^2 [4k^2 + k(4 - 22q) - 8q + 18q^2 + 1]. \tag{23}$$

Simple analysis shows that $\omega > \omega_0$ for $q < 12/49$. In the meantime, since $q \ll 1$ is the region that we are interested in, we have $\omega > \omega_0$, i.e., $\tau < \tau_0$ where τ and τ_0 are the period of the instanton just below and at 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 at some energy between the bottom of the metastable well and the top of the barrier. This indicates that the shape of τ is not monotonic. Accordingly, the first-order crossover always occurs in this situation.

Next, we consider the crossover in the presence of a longitudinal magnetic field. Applying the magnetic field along the $-\hat{z}$-axis, the total energy with $\boldsymbol{H} = -H\hat{z}$ is represented as

$$E(\theta, \phi) = K_1[\sin^2\theta + (k - q\cos(4\phi))\sin^4\theta + 2h_z(\cos\theta - 1)] \tag{24}$$

where $h_z = H_z/H_0$, $H_0 = 2K_1/m$, and a constant is introduced to make $E(\theta, \phi = 0)$ zero at $\theta = 0$. In this case it is easily seen that $\theta = 0$ is the position of the metastable state for $h_z < 1$. However, there also exists a metastable state even for $h_z > 1$. In order to study this property, we expand $E(\theta, \phi = 0)$ in the limit of $h_z \simeq 1$, which leads to equation (24), approximately given by

$$E(\theta, \phi = 0) \simeq K_1\left[(1 - h_z)\theta^2 + \left(-\frac{1}{3} + k - q + \frac{h_z}{12}\right)\theta^4\right]. \tag{25}$$

In this case, assuming that $k > q + (4 - h_z)/12$ (>0.33), we get the metastable position

$$\theta \simeq \frac{h_z - 1}{\sqrt{2[k - q - \frac{1}{3}(1 - h_z/4)]}} \tag{26}$$

even for $h_z > 1$. However, since $k \ll 1$ is an interesting region, the problem will be considered in the range of field $h_z < 1$.

Before calculating the oscillation frequency, we need to find the position of the barrier θ_0 which is a function of k, q, and h_z. In order to do that, we calculate $\mathrm{d}E(\theta, \phi = 0)/\mathrm{d}\theta$ from equation (24) and obtain

$$h_z = -2(k - q)\cos^3\theta_0 + [2(k - q) + 1]\cos\theta_0 \tag{27}$$

whose behaviour is illustrated in figure 1 for given k and q. Note that $\theta_0 = \pi/2$ for $h_z = 0$ and $\theta_0 = 0$ for $h_z = 1$.

Now, we calculate the parameters for the crossover by using equations (5)–(9):

$$A_1 = 2[-1 + k - q + 3(k - q)\cos(2\theta_0)]\sin\theta_0 \tag{28}$$

$$A_2 = [-1 - 5(k - q) + 9(k - q)\cos(2\theta_0)]\cos\theta_0 \tag{29}$$

$$A_3 = 32q\cos\theta_0\sin^2\theta_0 \tag{30}$$

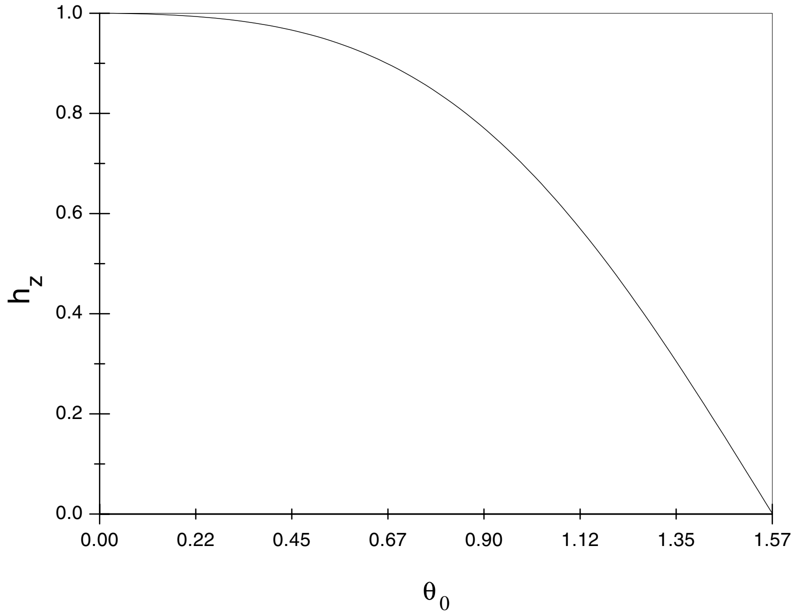

Figure 1. h_z versus θ_0 for $k = 0.2$ and $q = 0.007\,525$.

$$A_4 = [1 - 13(k - q) - 27(k - q)\cos(2\theta_0)]\sin\theta_0/3 \tag{31}$$

$$A_5 = -8q[\sin\theta_0 - 3\sin(3\theta_0)] \tag{32}$$

$$B_1 = -16q\sin^3\theta_0 \tag{33}$$

$$B_2 = -48q\cos\theta_0\sin^2\theta_0 \tag{34}$$

$$B_3 = (128/3)q\sin^3\theta_0 \tag{35}$$

$$B_4 = -12q[1 + 3\cos(2\theta_0)]\sin\theta_0. \tag{36}$$

Inserting them into equations (10)–(15), the change of frequency is determined by three quantities:

$$\begin{aligned} g_1 = -2q\cos^2\theta_0\sin^2\theta_0[&14 + 44(k - q) + 293(k - q)^2 \\ &- 12(k - q)(13 + 25(k - q))\cos(2\theta_0) \\ &+ 231(k - q)^2\cos(4\theta_0)]/[1 - k + q - 3(k - q)\cos(2\theta_0)] \end{aligned} \tag{37}$$

$$g_2 = -3q[13 - (k - q)(3 + 49\cos(2\theta_0))]\sin^2(2\theta_0) \tag{38}$$

$$\begin{aligned} g_3 = 2[&4 - 8k + 22k^2 + 14q - 83kq + 61q^2 + 2(12k^2 - 2k(6 + 19q) \\ &+ q(23 + 26q))\cos(2\theta_0) + 9(2k^2 - 9kq + 7q^2)\cos(4\theta_0)]\sin^2\theta_0. \end{aligned} \tag{39}$$

As discussed previously, $g_1 + g_2 + g_3 = 0$ determines the phase boundary between the first- and the second-order crossover. Using equation (27), we get the phase boundary which depends on k, q, and h_z. As is shown in figure 2, the first-order crossover is dominant for the typical value of q which we are interested in, and the second-order one is confined to the range of field $h_z \lesssim 1$ and $k \simeq 0.3$. As q decreases, the range of the second-order crossover tends to diminish in the phase diagram. Therefore, in order to observe the second-order crossover in the tetragonal symmetry, q and h_z should be relatively large.

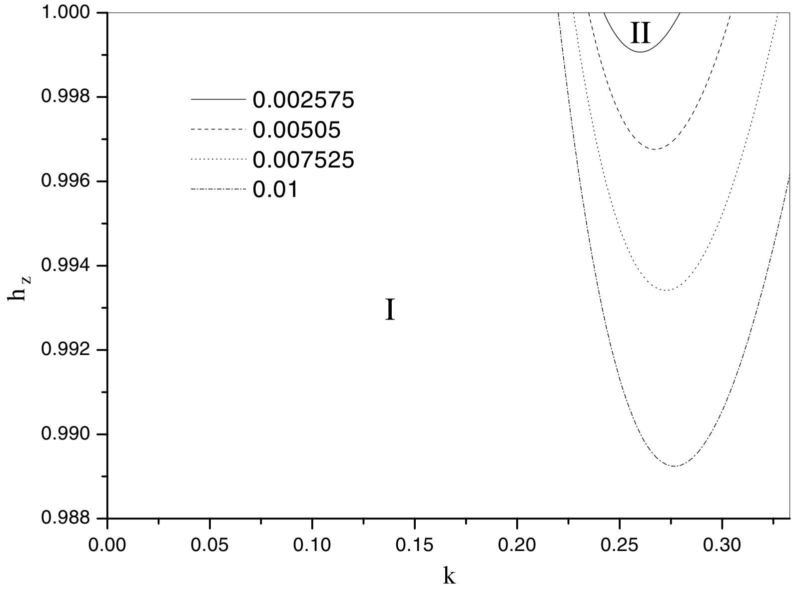

Figure 2. The phase diagram, h_z versus k, obtained from equation (39) for the tetragonal symmetry, where the range of q is $0.002\,573 \sim 0.01$ and the symbol I indicates the first-order regime and II the second-order one. Note that the actual range of h_z is $0 \leqslant h_z < 1$.

4. Crossover in hexagonal symmetry

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

$$E(\theta, \phi) = K_1 \sin^2\theta + K_2 \sin^4\theta + [K_3 - K_3' \cos(6\phi)] \sin^6\theta \tag{40}$$

where the easy axis is chosen to be $\hat{z}$, and $K_1 \gg K_2, K_3, K_3' > 0$. Defining $k_2 = K_2/K_1$, $k_3 = K_3/K_1$, and $q = K_3'/K_1$, the energy at $\phi = 0$ can be written as

$$E(\theta, \phi = 0) = K_1[\sin^2\theta + k_2 \sin^4\theta + (k_3 - q)\sin^6\theta]. \tag{41}$$

In this case, since the position of the barrier is $\theta_0 = \pi/2$, we have

$$A_1 = -2[1 + 2k_2 + 3(k_3 - q)] \qquad A_2 = A_3 = 0 \qquad A_4 = \frac{1 + 14k_2 + 39(k_3 - q)}{3} \tag{42}$$

$$A_5 = -108q \qquad B_1 = -36q \qquad B_2 = 0 \qquad B_3 = 216q \qquad B_4 = 90q \tag{43}$$

which leads to the change of the frequency given by

$$\omega^2 - \omega_0^2 = 18\left(\frac{a\theta_1}{n}\right)^2 [4k_2^2 + 2k_2(2 + 6k_3 - 15q) + 9k_3^2 + k_3(6 - 54q) + 1 - 12q + 45q^2]. \tag{44}$$

Noting that k_2, k_3, and $q \ll 1$, we always have $\omega > \omega_0$, and thereby expect that only the first-order crossover occurs in this situation.

Applying an external field along the $-\hat{z}$-axis, $H_z m \cos\theta$ is added to equation (40). In this situation equations (5)–(9) are more complicated; the results are

$$A_1 = -2\sin\theta_0[1 + 2k_2 \sin^2\theta_0 + 3(k_3 - q)\sin^4\theta_0 - 4\cos^2\theta_0(k_2 + 3(k_3 - q)\sin^2\theta_0)] \tag{45}$$

$$A_2 = \cos\theta_0[-1 - 14k_2 \sin^2\theta_0 - 39(k_3 - q)\sin^4\theta_0 + 4\cos^2\theta_0(k_2 + 9(k_3 - q)\sin^2\theta_0)] \tag{46}$$

$$A_3 = 108q\cos\theta_0\sin^4\theta_0 \tag{47}$$

$$\begin{aligned}A_4 = [&8 - 104k_2 + 69(k_3 - q) - 12(18k_2 + 11(-k_3 + q))\cos(2\theta_0)\\ &+ 375(k_3 - q)\cos(4\theta_0)]\sin\theta_0/24\end{aligned} \tag{48}$$

$$A_5 = 54q\sin^3\theta_0[3 + 5\cos(2\theta_0)] \tag{49}$$

$$B_1 = -36q\sin^5\theta_0 \tag{50}$$

$$B_2 = -180q\cos\theta_0\sin^4\theta_0 \tag{51}$$

$$B_3 = 216q\sin^5\theta_0 \tag{52}$$

$$B_4 = -45q[3 + 5\cos(2\theta_0)]\sin^3\theta_0. \tag{53}$$

As before, we plug them into equations (10)–(15) and obtain the following results which determine the shift of the oscillation frequency:

$$\begin{aligned}g_1 = 72q\cos\theta_0\sin^5\theta_0\{&[(8 + 40k_2 + 81(k_3 - q) - 12(6k_2 + 13(k_3 - q))\cos(2\theta_0)\\ &+ 75(k_3 - q)\cos(4\theta_0))(72 + 8k_2 + 113(k_3 - q)\\ &+ (-296k_2 - 348(k_3 - q))\cos(2\theta_0)\\ &+ 235(k_3 - q)\cos(4\theta_0))\cot\theta_0]/[64(-8 + 8k_2 + 3k_3 - 3q\\ &+ 12(2k_2 + k_3 - q)\cos(2\theta_0) - 15(k_3 - q)\cos(4\theta_0))]\\ &- 3\cos\theta_0[-2k_2 - k_3 + q + 5(k_3 - q)\cos(2\theta_0)]\sin\theta_0\}\end{aligned} \tag{54}$$

$$\begin{aligned}g_2 = \tfrac{45}{8}q\cos^2\theta_0\sin^4\theta_0\{&-152 + 72k_2 - 83(k_3 - q) + 4[134k_2 + 117(k_3 - q)]\cos(2\theta_0)\\ &- 385(k_3 - q)\cos(4\theta_0)\}\end{aligned} \tag{55}$$

$$\begin{aligned}g_3 = -\tfrac{9}{8}q\sin^6\theta_0[&8 - 104k_2 + 69(k_3 - q) - 12\,(18k_2 - 11(k_3 - q))\cos(2\theta_0)\\ &+ 375(k_3 - q)\cos(4\theta_0)] + \tfrac{99}{2}q\sin^4\theta_0[3 + 5\cos(2\theta_0)][1 + 2k_2\sin^2\theta_0\\ &+ 3(k_3 - q)\sin^4\theta_0 - 4\cos^2\theta_0(k_2 + 3(k_3 - q)\sin^2\theta_0)]\\ &+ 18\sin^2\theta_0[1 + 2k_2\sin^2\theta_0 + 3(k_3 - q)\sin^4\theta_0\\ &- 4\cos^2\theta_0(k_2 + 3(k_3 - q)\sin^2\theta_0)]^2.\end{aligned} \tag{56}$$

In this situation theoretical analysis becomes more cumbersome because g_1, g_2, and g_3 depend on three physical quantities k_2, k_3, and q. If such quantities are experimentally estimated, the analytic result $\delta\omega$ ($\propto g_1 + g_2 + g_3$) from equations (54)–(56) gives the guideline for determining whether the crossover is first or second order. In fact, the practically interesting situation is where the height of barrier is small and its width is narrow, which leads to large tunnelling rate. Such a situation is realized in the range of field $H \lesssim H_c$ where H_c is a critical field at which the barrier disappears. Defining $\epsilon \equiv 1 - H/H_c$ where $H_c = 2K_1/m$, the approximate form of the total energy is represented as

$$E(\theta,\phi) \simeq K_1\left[\epsilon\theta^2 - \frac{1}{4}\left(1 - 4k_2 + \frac{\epsilon}{3}\right)\theta^4 + \frac{1}{24}(1 - 16k_2 + 24k_3 - 24q\cos(6\phi))\theta^6\right] \tag{57}$$

where $\epsilon \ll 1$. Noting that the position of barrier is $\theta_0 \simeq \sqrt{2\epsilon/(1 - 4k_2)}$, we obtain the change of frequency given by

$$\omega^2 - \omega_0^2 \simeq \left(\frac{a\theta_1}{n}\right)^2 36(1 - 4k_2)\epsilon. \tag{58}$$

It is evident that the first-order crossover occurs in the range $k_2 < 1/4$. In general, if the potential is of the form $\theta^2 - \theta^4 + \theta^6$ in one dimension, the crossover is always second order. However, equation (58) gives $\omega > \omega_0$ which leads to first-order crossover. Actually, in the case where such a potential is not derived from the effective action, it is not certain that equation (57) gives rise to second-order crossover. To put this another way, if the action in equation (1) is reduced to the one with the one-dimensional functional form in which the effective potential is of the form $\theta^2 - \theta^4 + \theta^6$, the crossover is expected to be second order. However, it is not possible to obtain such an effective action and the corresponding potential in this system. Hence, even though the potential looks like $\theta^2 - \theta^4 + \theta^6$ in the limit of small ϵ, the system displays first-order crossover.

5. Conclusions

We have studied quantum–classical crossover in nanomagnetic systems with a longitudinal field. We have presented a general formula for determining whether the system exhibits first- or second-order crossover, and applied it to specific examples such as tetragonal and hexagonal symmetries. The result is of interest theoretically and experimentally in two respects. First, in uniaxial or biaxial symmetry, which have been much studied in the literature, the first-order regime decreases greatly with increasing field. Thus, in order to observe the sharp change of the escape rate around the crossover temperature in uniaxial or biaxial symmetry, the magnitude of an external field should be small and thereby the number of total spins should be moderate. However, in tetragonal or hexagonal symmetry the first-order crossover is expected to occur even in the range of large magnetic field. Hence, a system with a large spin can also be a good candidate for showing first-order crossover. Second, qualitative analysis shows that $\Delta T/T_0 \simeq 1/S$ in the first-order crossover and $1/\sqrt{S}$ in the second-order one. For, e.g., $S \sim 100$ we can obtain $\Delta T/T_0 \sim 0.10$ for the former and 0.1 for the latter. From this viewpoint, the larger the spin, the more likely one is to see a dramatic change of the escape rate in real experiments.

Acknowledgment

This work was supported by the Korea Research Foundation (2000-015-DP0138).

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