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## Spin tunnelling of trigonal and hexagonal ferromagnets in an arbitrarily directed magnetic field

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**Abstract.** The quantum tunnellings of the magnetization vector in single-domain ferromagnetic nanoparticles placed in an external magnetic field at an arbitrarily directed angle in the  $ZX$ -plane are studied theoretically. We consider magnetocrystalline anisotropies with trigonal and hexagonal crystal symmetry. By applying the instanton technique in the spin-coherent-state path-integral representation, we calculate the tunnel splittings, the tunnelling rates, and the crossover temperatures in the low-barrier limit for different angle ranges of the external magnetic field ( $\theta_H = \pi/2$ ,  $\pi/2 \ll \theta_H \ll \pi$ , and  $\theta_H = \pi$ ). Our results show that the tunnel splittings, the tunnelling rates, and the crossover temperatures depend distinctly on the orientation of the external magnetic field, which provides a possible experimental test for magnetic quantum tunnelling in nanometre-scale single-domain ferromagnets.

### 1. Introduction

Recently, there has been a great experimental and theoretical effort to observe and interpret macroscopic quantum tunnelling (MQT) and coherence (MQC) in nanometre-scale magnets at sufficiently low temperature [1]. Theoretical investigations based on the spin-coherent-state path integral were performed for single-domain ferromagnetic (FM) nanoparticles; these showed that MQT and MQC were possible in magnets containing as many as  $10^5$ – $10^6$  spins. Several experiments involving resonance measurements, magnetic relaxation, and hysteresis loop study for various systems showed either temperature-independent relaxation phenomena or a well-defined resonance depending exponentially on the total number of spins, which supported the idea of magnetic quantum tunnelling [1].

More recently, the tunnelling behaviours of the magnetization vector were studied extensively for single-domain FM nanoparticles in the presence of an external magnetic field applied at an arbitrary angle. The MQT problem for FM particles with uniaxial crystal symmetry was first studied by Zaslavskii who calculated the tunnelling exponent, and the pre-exponential factors and their temperature dependences in the low-barrier limit with the help of a mapping of the spin system onto a one-dimensional particle system [2]. For the same crystal symmetry, Miguel and Chudnovsky [3] calculated the tunnelling rate by applying the imaginary-time path integral, and demonstrated that the angular and field dependences of the tunnelling exponent obtained by Zaslavskii's method and by the path-integral method coincide precisely. They also discussed the tunnelling rate at finite temperature and suggested experimental procedures [3]. Kim and Hwang performed a calculation based on the instanton

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technique for FM particles with biaxial and tetragonal crystal symmetry [4], and Kim extended the tunnelling rate for biaxial crystal symmetry to a finite temperature [5]. The quantum–classical transition of the escape rate for FM particles with uniaxial crystal symmetry in an arbitrarily directed field was investigated by Garanin, Hidalgo, and Chudnovsky with the help of a mapping onto a particle moving in a double-well potential [6]. The switching-field measurement was carried out on single-domain FM nanoparticles of barium ferrite (BaFeCoTiO) containing about  $10^5$ – $10^6$  spins [7]. The measured angular dependence of the crossover temperature was found to be in excellent agreement with the theoretical prediction [3], which strongly suggests MQT of the magnetization in the BaFeCoTiO nanoparticles. Lü *et al* studied the MQT and MQC of the Néel vector in single-domain antiferromagnetic (AFM) nanoparticles with biaxial, tetragonal, and hexagonal crystal symmetry in an arbitrarily directed field [8].

In this paper, we extend the previous theoretical results obtained for the single-domain FM particles with biaxial and tetragonal symmetry to ones for FM particles with a much more complex structure placed in an external magnetic field at an arbitrarily directed angle in the  $ZX$ -plane, on the basis of the instanton technique in the spin-coherent-state path-integral representation. We consider magnetocrystalline anisotropies with trigonal and hexagonal crystal symmetry. Both the Wentzel–Kramers–Brillouin (WKB) exponents and the pre-exponential factors in the tunnelling rates for MQT and the tunnel splittings for MQC in FM particles are evaluated analytically for different angle ranges of the external magnetic field ( $\theta_H = \pi/2$ ,  $\pi/2 + O(\epsilon^{3/2}) < \theta_H < \pi - O(\epsilon^{3/2})$ , and  $\theta_H = \pi$ ), and the temperature which corresponds to the crossover from the thermal to the quantum regime is clearly indicated for each case. Our results show that the distinct angular dependence, together with the dependence of the WKB tunnelling rate and the crossover temperature on the strength of the external magnetic field, may provide an independent experimental test for the magnetic tunnelling in single-domain FM nanoparticles. The calculations performed in this paper are semiclassical in nature, i.e., valid for large spins and in the continuum limit. We analyse the validity of the semiclassical approximation, and find that the semiclassical approximation is rather good for typical values of the parameters for single-domain FM nanoparticles.

This paper is structured in the following way. In section 2, we briefly review the basic ideas of the MQT and MQC in single-domain FM particles. In sections 3 and 4, we study the quantum tunnelling of the magnetization vector for FM particles with trigonal and hexagonal crystal symmetry in the presence of an external magnetic field applied in the  $ZX$ -plane with a range of angles  $\pi/2 \leq \theta_H \leq \pi$ . The conclusions are presented in section 5. In appendix A, we explain briefly the computation of the pre-exponential factors in the WKB tunnelling rate, and then apply this approach to obtain the tunnel splittings for FM particles with trigonal crystal symmetry in a magnetic field applied perpendicular to the anisotropy axis ( $\theta_H = \pi/2$ ) in detail.

## 2. MQT and MQC of the magnetization vector in FM particles

In this section we briefly review some basic ideas concerning MQT and MQC of the magnetization vector in single-domain FM nanoparticles, on the basis of the instanton technique applied to the spin-coherent-state path integral [1, 11, 12].

The system of interest is a nanometre-scale single-domain ferromagnet at a temperature well below its anisotropy gap. For such a FM particle, the tunnel splitting for MQC and the tunnelling rate for MQT are determined by the imaginary-time amplitude for transition from

an initial state  $|i\rangle$  to a final state  $|f\rangle$ :

$$U_{fi} = \langle f | e^{-HT} | i \rangle = \int D\Omega \exp(-S_E) \quad (1)$$

where  $S_E$  is the Euclidean action and  $D\Omega$  is the measure of the path integral. In the spin-coherent-state path-integral representation, the Euclidean action can be expressed as

$$S_E(\theta, \phi) = \frac{V}{\hbar} \int d\tau \left[ i \frac{M_0}{\gamma} \left( \frac{d\phi}{d\tau} \right) - i \frac{M_0}{\gamma} \left( \frac{d\phi}{d\tau} \right) \cos \theta + E(\theta, \phi) \right] \quad (2)$$

where  $V$  is the volume of the FM particle and  $\gamma$  is the gyromagnetic ratio.  $M_0 = |\vec{M}| = \hbar\gamma S/V$ , where  $S$  is the total spin of FM particles. It is noted that the first two terms in equation (2) define the topological Berry or Wess–Zumino, Chern–Simons term which arises from the nonorthogonality of spin coherent states. The Wess–Zumino term has a simple topological interpretation. For a closed path, this term equals  $-iS$  times the area swept out on the unit sphere between the path and the north pole. The first term in equation (2) is a total imaginary-time derivative, which has no effect on the classical equations of motion, but is crucial for the spin-parity effects [9, 10]. However, for the closed instanton or bounce 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.

In discussing macroscopic quantum phenomena, it is essential to distinguish between two types of process: MQC (i.e., coherent tunnelling) and MQT (i.e., incoherent tunnelling). In the case of MQC, the system in question performs coherent  $\text{NH}_3$ -type oscillations between two degenerate wells separated by a classically impenetrable barrier. Tunnelling between neighbouring degenerate vacua can be described by the instanton configuration with nonzero topological charge and leads to a level splitting of the ground states [11]. The tunnelling removes the degeneracy of the original ground states, and the true ground state is a superposition of the previous degenerate ground states. For the case of MQT, the system escapes from a metastable potential well into a continuum by quantum tunnelling at sufficiently low temperatures, and the tunnelling results in an imaginary part of the energy which is dominated by the so-called bounce configuration with zero topological charge [11]. As emphasized by Leggett, the two phenomena of MQC and MQT are physically very different, particularly from the viewpoint of experimental feasibility [20]. MQC is a far more delicate phenomenon than MQT, as it is much more easily destroyed by the environment [21], and by symmetry-breaking fields with very small  $c$ -numbers that spoil the degeneracy.

In the semiclassical limit, the dominant contribution to the transition amplitude comes from the finite-action solution (instanton) of the classical equation of motion. The motion of the magnetization vector  $\vec{M}$  is determined by the Landau–Lifshitz equation

$$i \frac{d\vec{M}}{d\tau} = -\gamma \vec{M} \times \frac{dE(\vec{M})}{d\vec{M}} \quad (3)$$

which can also be expressed as the following equations in the spherical coordinate system:

$$i \left( \frac{d\bar{\theta}}{d\tau} \right) \sin \bar{\theta} = \frac{\gamma}{M_0} \frac{\partial E}{\partial \bar{\phi}} \quad (4a)$$

$$i \left( \frac{d\bar{\phi}}{d\tau} \right) \sin \bar{\theta} = -\frac{\gamma}{M_0} \frac{\partial E}{\partial \bar{\theta}} \quad (4b)$$

where  $\bar{\theta}$  and  $\bar{\phi}$  denote the classical path. Note that the Euclidean action equation (2) describes the  $(1 \oplus 1)$ -dimensional dynamics in the Hamiltonian formulation with canonical variables  $\phi$  and  $P_\phi = S(1 - \cos \theta)$ . The instanton's contribution to the tunnelling rate  $\Gamma$  for MQT or the

tunnel splitting  $\Delta$  for MQC (not including the topological Wess–Zumino or Berry phase) is given by [11, 12]

$$\Gamma \text{ (or } \Delta) = A\omega_p \left( \frac{S_{cl}}{2\pi} \right)^{1/2} e^{-S_{cl}} \quad (5)$$

where  $\omega_p$  is the frequency of small oscillations near the bottom of the inverted potential, and  $S_{cl}$  is the classical action. The pre-exponential factor  $A$  originates from the quantum fluctuations about the classical path, which can be evaluated by expanding the Euclidean action to second order in the small fluctuations [11, 12]. In reference [12], Garg and Kim studied the general formalism for calculating both the exponent and the pre-exponential factors in the WKB tunnelling rates for MQT and MQC in single-domain FM nanoparticles. In appendix A, we explain briefly the basic idea of this calculation, and then apply this approach to calculate the instanton's contribution to the tunnel splittings for MQC of the magnetization vector in FM particles with trigonal crystal symmetry in an external magnetic field perpendicular to the anisotropy axis (considered in section 3) in detail.

### 3. MQC and MQT for trigonal crystal symmetry

In this section, we study the tunnelling behaviours of the magnetization vector in single-domain FM nanoparticles with trigonal crystal symmetry. The external magnetic field is applied in the  $ZX$ -plane, at an angle in the range of  $\pi/2 \leq \theta_H < \pi$ . Now the total energy  $E(\theta, \phi)$  can be written as

$$E(\theta, \phi) = K_1 \sin^2 \theta - K_2 \sin^3 \theta \cos(3\phi) - M_0 H_x \sin \theta \cos \phi - M_0 H_z \cos \theta + E_0 \quad (6)$$

where  $K_1$  and  $K_2$  are the magnetic anisotropy constants satisfying  $K_1 \gg K_2 > 0$ , and  $E_0$  is a constant which makes  $E(\theta, \phi)$  zero at the initial orientation. As the magnetic field is applied in the  $ZX$ -plane,  $H_x = H \sin \theta_H$  and  $H_z = H \cos \theta_H$ , where  $H$  is the magnitude of the field and  $\theta_H$  is the angle between the magnetic field and the  $\hat{z}$ -axis.

In the absence of an external magnetic field, the system reduces to one with threefold rotational symmetry around the  $\hat{z}$ -axis and reflection symmetry in the  $XY$ -plane. The unit vectors  $\hat{z}$  and  $-\hat{z}$  define the two classical ground-state configurations. The amplitude for transition between degenerate ground states can be suppressed to zero as a result of the destructive Wess–Zumino phase if the system has time-reversal invariance at zero magnetic field [10]. However, for the closed instanton or bounce trajectory described in this paper (as shown in the following) the phase term in equation (2), proportional to  $d\phi/d\tau$  (not the  $(d\phi/d\tau) \cos \theta$  term) gives a zero contribution to the integral equation (2) and, therefore, can be omitted.

By introducing the dimensionless parameters

$$\bar{K}_2 = K_2/2K_1 \quad \bar{H}_x = H_x/H_0 \quad \bar{H}_z = H_z/H_0 \quad (7)$$

equation (6) can be rewritten as

$$\bar{E}(\theta, \phi) = \frac{1}{2} \sin^2 \theta - \bar{K}_2 \sin^3 \theta \cos(3\phi) - \bar{H}_x \sin \theta \cos \phi - \bar{H}_z \cos \theta + \bar{E}_0 \quad (8)$$

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

$$\bar{E}(\theta, \phi = 0) = \frac{1}{2} \sin^2 \theta - \bar{K}_2 \sin^3 \theta - \bar{H} \cos(\theta - \theta_H) + \bar{E}_0. \quad (9)$$

We denote as  $\theta_0$  the initial angle and as  $\theta_c$  the critical angle at which the energy barrier vanishes when the external magnetic field is close to the critical value  $\bar{H}_c(\theta_H)$  (to be calculated in the

following). Then, the initial angle  $\theta_0$  satisfies  $[d\bar{E}(\theta, \phi = 0)/d\theta]_{\theta=\theta_0} = 0$ , and the critical angle  $\theta_c$  and the dimensionless critical field  $\bar{H}_c$  satisfy both  $[d\bar{E}(\theta, \phi = 0)/d\theta]_{\theta=\theta_c, \bar{H}=\bar{H}_c} = 0$  and  $[d^2\bar{E}(\theta, \phi = 0)/d\theta^2]_{\theta=\theta_c, \bar{H}=\bar{H}_c} = 0$ , which leads to

$$\frac{1}{2} \sin(2\theta_0) - 3\bar{K}_2 \sin^2 \theta_0 \cos \theta_0 + \bar{H} \sin(\theta_0 - \theta_H) = 0 \quad (10a)$$

$$\frac{1}{2} \sin(2\theta_c) - 3\bar{K}_2 \sin^2 \theta_c \cos \theta_c + \bar{H}_c \sin(\theta_c - \theta_H) = 0 \quad (10b)$$

$$\cos(2\theta_c) - 3\bar{K}_2(2 \sin \theta_c \cos^2 \theta_c - \sin^3 \theta_c) + \bar{H}_c \cos(\theta_c - \theta_H) = 0. \quad (10c)$$

After some algebra,  $\bar{H}_c(\theta_H)$  and  $\theta_c$  are found to be

$$\bar{H}_c = \frac{1}{[(\sin \theta_H)^{2/3} + |\cos \theta_H|^{2/3}]^{3/2}} \times \left[ 1 + 3\bar{K}_2 \frac{1}{(1 + |\cot \theta_H|^{2/3})^{1/2}} + 6\bar{K}_2 \frac{1}{(1 + |\cot \theta_H|^{2/3})^{3/2}} \right] \quad (11a)$$

$$\sin^2 \theta_c = \frac{1}{1 + |\cot \theta_H|^{2/3}} \left[ 1 - 2\bar{K}_2 \frac{|\cot \theta_H|^{2/3}}{(1 + |\cot \theta_H|^{2/3})^{1/2}} - 4\bar{K}_2 \frac{|\cot \theta_H|^{2/3}}{(1 + |\cot \theta_H|^{2/3})^{3/2}} \right]. \quad (11b)$$

Now we consider the limiting case where the external magnetic field is slightly lower than the critical field, i.e.,  $\epsilon = 1 - \bar{H}/\bar{H}_c \ll 1$ . In this practically interesting situation, the barrier height is low and the width is narrow, and therefore the tunnelling rate in MQT or the tunnel splitting in MQC is large. Introducing  $\eta \equiv \theta_c - \theta_0$  ( $|\eta| \ll 1$  in the limit of  $\epsilon \ll 1$ ), expanding  $[d\bar{E}(\theta, \phi = 0)/d\theta]_{\theta=\theta_0} = 0$  about  $\theta_c$ , and using the relations  $[d\bar{E}(\theta, \phi = 0)/d\theta]_{\theta=\theta_c, \bar{H}=\bar{H}_c} = 0$  and  $[d^2\bar{E}(\theta, \phi = 0)/d\theta^2]_{\theta=\theta_c, \bar{H}=\bar{H}_c} = 0$ , we obtain the approximation equation for  $\eta$  to the order of  $\epsilon^{3/2}$ :

$$-\epsilon \bar{H}_c \sin(\theta_c - \theta_H) - \eta^2 \left( \frac{3}{4} \sin 2\theta_c + 3\bar{K}_2 \cos 3\theta_c \right) + \eta \left[ \epsilon \bar{H}_c \cos(\theta_c - \theta_H) + \eta^2 \left( \frac{1}{2} \cos 2\theta_c - 3\bar{K}_2 \sin 3\theta_c \right) \right] = 0. \quad (12)$$

Then  $\bar{E}(\theta, \phi)$  reduces to the following equation in the limit of small  $\epsilon$ :

$$\bar{E}(\delta, \phi) = 2\bar{K}_2 \sin^2(3\phi/2) \sin^3(\theta_0 + \delta) + \bar{H}_x \sin(\theta_0 + \delta)(1 - \cos \phi) + \bar{E}_1(\delta) \quad (13)$$

where  $\delta \equiv \theta - \theta_0$  ( $|\delta| \ll 1$  in the limit of  $\epsilon \ll 1$ ), and  $\bar{E}_1(\delta)$  is a function of only  $\delta$ , given by

$$\begin{aligned} \bar{E}_1(\delta) = & -\frac{1}{2} \left[ \bar{H}_c \sin(\theta_c - \theta_H) - \bar{K}_2(\cos^3 \theta_c - \frac{3}{2} \sin^2 \theta_c \cos \theta_c) \right] (3\delta^2 \eta - \delta^3) \\ & - \frac{1}{2} \left[ \bar{H}_c \cos(\theta_c - \theta_H) - 3\bar{K}_2(\sin^3 \theta_c - 4 \sin \theta_c \cos^2 \theta_c) \right] \\ & \times \left[ \delta^2 \left( \epsilon - \frac{3}{2} \eta^2 \right) + \delta^3 \eta - \frac{1}{4} \delta^4 \right] - \frac{3}{2} \bar{K}_2 (\sin^3 \theta_c - 4 \sin \theta_c \cos^2 \theta_c) \delta^2 \epsilon. \end{aligned} \quad (14)$$

In the following, we will investigate the tunnelling behaviours of the magnetization vector in FM particles with trigonal crystal symmetry for different angle ranges of the external magnetic field:  $\theta_H = \pi/2$  and  $\pi/2 < \theta_H < \pi$ .

3.1.  $\theta_H = \pi/2$

For  $\theta_H = \pi/2$ , we have  $\theta_c = \pi/2$  from equation (11b) and  $\eta = \sqrt{2\epsilon}(1 + 9\bar{K}_2/2)$  from equation (12). Then  $\bar{E}_1(\delta)$  of equation (14) reduces to

$$\bar{E}_1(\delta) = \frac{1}{8}\delta^2(\delta - 2\eta)^2. \tag{15}$$

A plot of the effective potential  $\bar{E}_1(\delta)$  as a function of  $\delta (= \theta - \theta_0)$  for  $\theta_H = \pi/2$  is shown in figure 1. Now the problem is one of MQC, where the magnetization vector resonates coherently between the energetically degenerate easy directions at  $\delta = 0$  and  $\delta = 2\sqrt{2\epsilon}(1 + 9\bar{K}_2/2)$  separated by a classically impenetrable barrier at  $\delta = \sqrt{2\epsilon}(1 + 9\bar{K}_2/2)$ . Substituting equation (15) into the classical equations of motion, we obtain the classical solution called the instanton:

$$\begin{aligned} \bar{\phi} &= i\epsilon \left( 1 + 3\bar{K}_2 + \frac{1}{2}\epsilon \right) \frac{1}{\cosh^2(\bar{\omega}_c \bar{\tau})} \\ \bar{\delta} &= \sqrt{2\epsilon} \left( 1 + \frac{9}{2}\bar{K}_2 \right) [1 + \tanh(\bar{\omega}_c \bar{\tau})] \end{aligned} \tag{16}$$

where  $\bar{\omega}_c = \sqrt{\epsilon/2}(1 + 21\bar{K}_2/2 - \epsilon/2)$ ,  $\bar{\tau} = \omega_0 \tau$ , and  $\omega_0 = 2K_1 V/\hbar S$ . We can calculate the classical action by integrating the Euclidean action equation (2) with the above classical trajectory, and the result is found to be

$$S_{cl} = \frac{2^{5/2}}{3}\epsilon^{3/2}S \left( 1 + \frac{15}{2}\bar{K}_2 + \frac{1}{2}\epsilon \right). \tag{17}$$

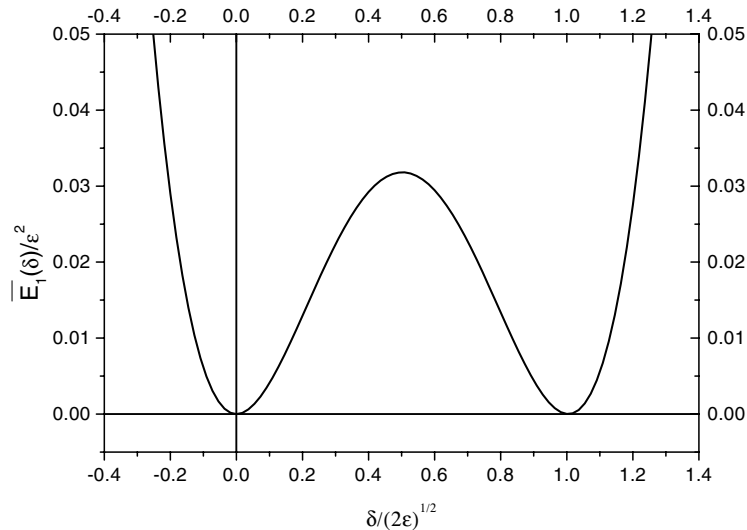


Figure 1. The dependence on  $\delta (= \theta - \theta_0)$  of the effective potential  $\bar{E}_1(\delta)$  for  $\theta_H = \pi/2$  (MQC).

Now we consider the transition exponent which is usually addressed by experiments. Transitions between two states in a bistable system or escaping from a metastable state can occur either due to the quantum tunnelling or via the classical thermal activation. In the limit of temperature  $T \rightarrow 0$ , the transitions are purely quantum mechanical and the rate goes as  $\Gamma \sim \exp(-S_{cl})$ , with  $S_{cl}$  being the classical action or the WKB exponent which is independent of temperature. As the temperature increases from zero, thermal effects enter

in the quantum tunnelling process. If the temperature is sufficiently high, the decay from a metastable state is determined by processes of thermal activation, and the transition rate follows the Arrhenius law,  $\Gamma \sim \exp(-U/k_B T)$ , with  $k_B$  being the Boltzmann constant and  $U$  being the height of the energy barrier between the two states. Because of the exponential dependence of the thermal rate on  $T$ , the temperature  $T_c$  characterizing the crossover from the quantum to the thermal regime can be estimated as  $k_B T_c = U/S_{cl}$ . For a quasiparticle with the effective mass  $M$  moving in one-dimensional potential  $U(x)$ , a more accurate definition of the crossover temperature in the absence of any dissipation was presented by Goldanskii [13, 14]:  $k_B T'_c = \hbar\omega_b/2\pi$ , where

$$\omega_b = \sqrt{-U''(x_b)/M}$$

is the frequency of small oscillations near the bottom of the inverted potential,  $-U(x)$ , and  $x_b$  corresponds to the bottom of the inverted potential. Below  $T'_c$ , thermally assisted quantum tunnelling occurs from the excited levels, that further reduces to the quantum tunnelling from the ground-state level as the temperature decreases to zero. Above  $T'_c$ , quantum tunnelling effects are small and the transitions occur due to the thermal activation to the top of the barrier. For the MQT problem, i.e., the problem of decay from the metastable state, both  $T_c$  and  $T'_c$  can be used as definitions of the crossover temperature corresponding to the crossover from classical to quantum behaviour, since the quantum escaping from a metastable state is a process of incoherent tunnelling. However, for the MQC problem, i.e., the problem of resonance between degenerate states, the situation is different. As the temperature grows from zero, three kinds of transition should be taken into account: quantum coherence between the degenerate ground-state levels (coherent tunnelling), quantum tunnelling from the excited levels (thermally assisted tunnelling or incoherent tunnelling), and classical over-barrier transition (incoherent transition). Two kinds of crossover temperature can be defined to distinguish the three regimes. The Goldanskii definition  $T'_c$  for the MQC problem corresponds to the crossover from quantum coherence between the degenerate ground-state levels (coherent tunnelling) to quantum tunnelling from the excited levels (thermally assisted tunnelling or incoherent tunnelling), while  $T_c$  corresponds to the crossover from quantum coherence between the degenerate ground-state levels (coherent tunnelling) to classical over-barrier transition (incoherent transition). Experiments involving magnetic relaxation and resonance measurements for various systems have shown either temperature-independent relaxation phenomena (in MQT) or a well-defined resonance (in MQC) below some crossover temperature, which strongly support the existence of quantum tunnelling processes [1]. And more recently, the crossover from quantum to classical behaviour and the associated phase transition have been investigated extensively in MQT and MQC in single-domain FM particles [14–18]. It is noted that the sharpness of the crossover between the thermal and quantum regimes also depends on the strength of the dissipation in the environment. In the case of low dissipation, which is common for magnetic systems, its effect on the crossover is small [13, 14].

For the single-domain FM nanoparticle in a magnetic field applied at  $\theta_H = \pi/2$ , the magnetization vector resonates coherently between the energetically degenerate easy directions at  $\delta = 0$  and  $\delta = 2\sqrt{2}\epsilon(1 + 9\bar{K}_2/2)$  separated by a classically impenetrable barrier at  $\delta = \sqrt{2}\epsilon(1 + 9\bar{K}_2/2)$ , and the height of energy barrier is found to be  $U = K_1 V \epsilon^2(1 + 18\bar{K}_2)$ . Then, equating  $S_{cl}$  to  $U/k_B T$ , we obtain that the crossover from quantum coherence between the degenerate ground-state levels (coherent tunnelling) to classical over-barrier transition (incoherent transition) occurs at

$$k_B T_c = \frac{3}{2^{5/2}} \epsilon^{1/2} \frac{K_1 V}{S} \left( 1 + \frac{21}{2} \bar{K}_2 - \frac{1}{2} \epsilon \right). \quad (18)$$



For this MQC problem, the Goldanskii definition of  $T'_c$  corresponding to the crossover from quantum coherence between the degenerate ground-state levels (coherent tunnelling) to quantum tunnelling from excited levels (thermally assisted tunnelling or incoherent tunnelling) becomes  $k_B T'_c = \hbar \omega_b / 2\pi$ , where  $\omega_b = \bar{\omega}_b \omega_0$ , with

$$\bar{\omega}_b \equiv \sqrt{-E''(\delta_m)/M}$$

the frequency of small oscillations of the magnetization vector near the bottom of the inverted potential,  $M^{-1} = (1 + 12\bar{K}_2 - \epsilon)$ , and  $\delta_m$  the position of the energy barrier. For the present case,  $\delta_m = \sqrt{2\epsilon}(1 + 9\bar{K}_2/2)$  and  $\bar{\omega}_b = \sqrt{\epsilon}(1 + 21\bar{K}_2/2 - \epsilon/2) = \sqrt{2\bar{\omega}_c}$ . Then it is easy to obtain that

$$k_B T'_c = \frac{1}{\pi} \epsilon^{1/2} \frac{K_1 V}{S} \left( 1 + \frac{21}{2} \bar{K}_2 - \frac{1}{2} \epsilon \right). \quad (19)$$

Comparison of equations (18) and (19) shows that  $T_c \approx 1.67 T'_c$ , which is consistent with the physical interpretation for quantum–classical transition in the MQC problem.

It is noted that the quantum tunnellings of the magnetization vector in single-domain FM nanoparticles are studied with the help of the instanton technique in the spin-coherent-state path-integral representation, which is semiclassical in nature, i.e., valid for large spins and in the continuum limit. Therefore, one should analyse the validity of the semiclassical approximation. It is well known that for this approach to be valid, the tunnelling rate must be small, which indicates that the WKB exponent or the classical action  $S_{cl} \gg 1$ . Moreover, the energy  $\hbar \omega_b$  of zero-point oscillations around the minimum of the inverted potential  $-\bar{E}_1(\delta)$  should be sufficiently small compared to the height of the barrier,  $U = 2K_1 V \bar{E}_1(\delta_m)$ . For the single-domain FM nanoparticle with trigonal crystal symmetry in a magnetic field applied at  $\theta_H = \pi/2$ , it is easy to show that the WKB exponent is approximately given by

$$B \sim \frac{U}{\hbar \omega_b} = \frac{1}{2} \epsilon^{3/2} S \left( 1 + \frac{15}{2} \bar{K}_2 + \frac{1}{2} \epsilon \right) \quad (20)$$

which agrees up to the numerical factor with the result for the classical action in equation (17) obtained by applying the explicit instanton solution. For typical values of the parameters for single-domain FM nanoparticles,  $K_1 \sim 10^8$  erg cm<sup>-3</sup>,  $K_2 \sim 10^5$  erg cm<sup>-3</sup>, and the total spin  $S = 10^6$ , we obtain that  $B \sim U/\hbar \omega_b \approx 15.8$  from equation (20) and  $S_{cl} \approx 59.6$  for  $\epsilon = 0.001$  from equation (17). In this case the semiclassical approximation should be already rather good.

By applying the instanton technique for FM particles in the spin-coherent-state path-integral representation [11, 12], we obtain the instanton's contribution to the tunnel splitting as (for a detailed calculation see appendix A)

$$\hbar \Delta_0 = \frac{2^{13/4}}{\pi^{1/2}} (K_1 V) \epsilon^{5/4} S^{-1/2} \left( 1 + \frac{57}{4} \bar{K}_2 - \frac{1}{4} \epsilon \right) e^{-S_{cl}} \quad (21)$$

where the WKB exponent or the classical action  $S_{cl}$  has been presented in equation (17).

Now we apply the effective Hamiltonian approach to evaluate the ground-state tunnel splitting [19]. For the present case, the effective Hamiltonian can be written as

$$H_{eff} = \begin{bmatrix} 0 & -\hbar \Delta_0 \\ -\hbar \Delta_0 & 0 \end{bmatrix}. \quad (22)$$

A simple diagonalization of  $H_{eff}$  shows that the eigenvalues of this system are  $\pm \hbar \Delta_0$ . Therefore, the splitting of the ground state due to resonant coherently quantum tunnelling of the magnetization vector between energetically degenerate states is  $\hbar \Delta = 2\hbar \Delta_0$ , where  $\hbar \Delta_0$  is shown in equation (21) with equation (17) for single-domain FM particles with trigonal crystal symmetry in a magnetic field applied perpendicular to the anisotropy axis ( $\theta_H = \pi/2$ ).

3.2.  $\pi/2 < \theta_H < \pi$ 

For  $\pi/2 < \theta_H < \pi$ , the critical angle  $\theta_c$  is in the range of  $0 < \theta_c < \pi/2$ , and  $\eta \approx \sqrt{2\epsilon/3}$ . Then  $\bar{E}_1(\delta)$  of equation (14) reduces to

$$\bar{E}_1(\delta) = \frac{1}{2} \frac{|\cot \theta_H|^{1/3}}{1 + |\cot \theta_H|^{2/3}} \left[ 1 - \frac{15}{2} \bar{K}_2 \frac{1}{(1 + |\cot \theta_H|^{2/3})^{1/2}} \right] (\sqrt{6\epsilon} \delta^2 - \delta^3). \quad (23)$$

The dependence of the effective potential  $\bar{E}_1(\delta)$  on  $\delta$  ( $=\theta - \theta_0$ ) for  $\theta_H = 3\pi/4$  is plotted in figure 2. Here,  $\bar{K}_2 = 0.001$ . Now the problem becomes one of MQT, where the magnetization vector escapes from the metastable state at  $\delta = 0$ ,  $\phi = 0$  through the barrier by quantum tunnelling. Substituting equation (23) into the classical equations of motion, the classical solution called bounce is found to be

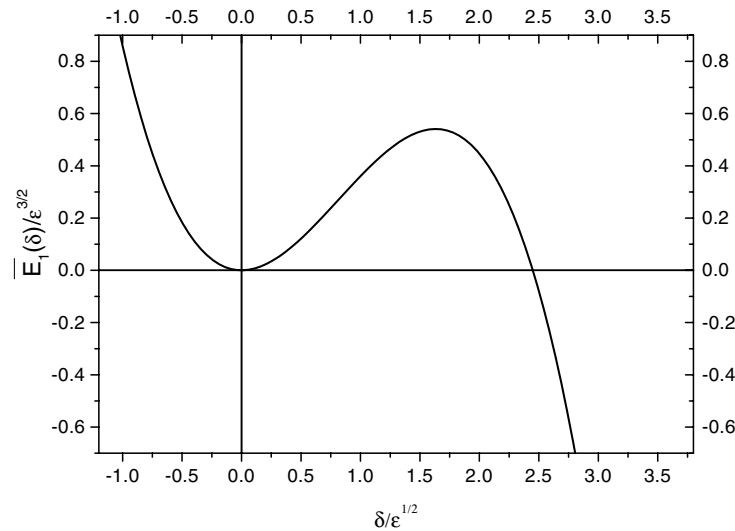
$$\bar{\phi} = i(6\epsilon)^{3/4} |\cot \theta_H|^{1/6} (1 + |\cot \theta_H|^{2/3})^{1/2} \left[ 1 + \frac{\epsilon}{2} - \frac{9}{2} \bar{K}_2 (1 + |\cot \theta_H|^{2/3})^{1/2} + \frac{\bar{K}_2}{4} \frac{2|\cot \theta_H|^{2/3} - 9}{(1 + |\cot \theta_H|^{2/3})^{1/2}} + \bar{K}_2 \frac{|\cot \theta_H|^{2/3} - 3}{(1 + |\cot \theta_H|^{2/3})^{3/2}} \right] \frac{\sinh(\bar{\omega}_c \bar{\tau})}{\cosh^3(\bar{\omega}_c \bar{\tau})} \quad (24)$$

and

$$\bar{\delta} = \sqrt{6\epsilon} / \cosh^2(\bar{\omega}_c \bar{\tau})$$

which corresponds to the variation of  $\delta$  from  $\delta = 0$  at  $\tau = -\infty$  to the turning point  $\delta = \sqrt{6\epsilon}$  at  $\tau = 0$ , and then back to  $\delta = 0$  at  $\tau = +\infty$ , where

$$\bar{\omega}_c = 3^{1/4} 2^{-3/4} \epsilon^{1/4} \frac{|\cot \theta_H|^{1/6}}{1 + |\cot \theta_H|^{2/3}} \left[ 1 - \frac{\epsilon}{2} + \frac{9}{2} \bar{K}_2 (1 + |\cot \theta_H|^{2/3})^{1/2} + \frac{\bar{K}_2}{4} \frac{2|\cot \theta_H|^{2/3} - 21}{(1 + |\cot \theta_H|^{2/3})^{1/2}} + \bar{K}_2 \frac{|\cot \theta_H|^{2/3} + 3}{(1 + |\cot \theta_H|^{2/3})^{3/2}} \right].$$



**Figure 2.** The dependence on  $\delta$  ( $=\theta - \theta_0$ ) of the effective potential  $\bar{E}_1(\delta)$  for  $\theta_H = 3\pi/4$  (MQT). Here,  $\bar{K}_2 = 0.001$ .

The associated classical action is then given by

$$S_{cl} = \frac{3^{1/4} 2^{17/4}}{5} S \epsilon^{5/4} |\cot \theta_H|^{1/6} \left[ 1 + \frac{\epsilon}{2} - \frac{9}{2} \bar{K}_2 (1 + |\cot \theta_H|^{2/3})^{1/2} - \frac{\bar{K}_2}{2} \frac{|\cot \theta_H|^{2/3} + 9/2}{(1 + |\cot \theta_H|^{2/3})^{1/2}} - \bar{K}_2 \frac{|\cot \theta_H|^{2/3} + 3}{(1 + |\cot \theta_H|^{2/3})^{3/2}} \right]. \quad (25)$$

For this case, the barrier height is

$$U = 2K_1 V \bar{E}_1(\delta_m) = \frac{2^{7/2}}{3^{3/2}} \frac{|\cot \theta_H|^{1/3}}{1 + |\cot \theta_H|^{2/3}} \left[ 1 - \frac{15}{2} \bar{K}_2 \frac{1}{(1 + |\cot \theta_H|^{2/3})^{1/2}} \right] \epsilon^{3/2} (K_1 V)$$

at  $\delta_m = 2(6\epsilon)^{1/2}/3$ , and the frequency of small oscillations of the magnetization vector around the bottom of the metastable well is

$$\begin{aligned} \bar{\omega}_b &= 3^{1/4} 2^{1/4} \epsilon^{1/4} \frac{|\cot \theta_H|^{1/6}}{1 + |\cot \theta_H|^{2/3}} \left[ 1 - \frac{\epsilon}{2} + \frac{9}{2} \bar{K}_2 (1 + |\cot \theta_H|^{2/3})^{1/2} \right. \\ &\quad \left. + \frac{\bar{K}_2}{4} \frac{2|\cot \theta_H|^{2/3} - 21}{(1 + |\cot \theta_H|^{2/3})^{1/2}} + \bar{K}_2 \frac{|\cot \theta_H|^{2/3} + 3}{(1 + |\cot \theta_H|^{2/3})^{3/2}} \right] \\ &= 2\bar{\omega}_c. \end{aligned}$$

Then the WKB exponent or the classical action  $B$  is approximately given by

$$B \sim \frac{U}{\hbar \omega_b} = \frac{2^{9/4}}{3^{7/4}} S \epsilon^{5/4} |\cot \theta_H|^{1/6} \left[ 1 + \frac{\epsilon}{2} - \frac{9}{2} \bar{K}_2 (1 + |\cot \theta_H|^{2/3})^{1/2} - \frac{\bar{K}_2}{2} \frac{|\cot \theta_H|^{2/3} + 9/2}{(1 + |\cot \theta_H|^{2/3})^{1/2}} - \bar{K}_2 \frac{|\cot \theta_H|^{2/3} + 3}{(1 + |\cot \theta_H|^{2/3})^{3/2}} \right] \quad (26)$$

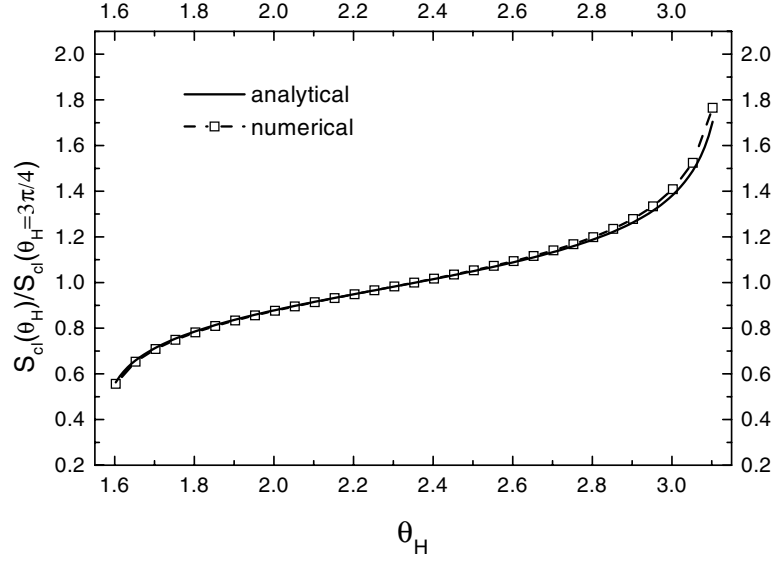
which is consistent with equation (25) up to the numerical factor. After a simple calculation, we obtain the crossover temperature as

$$\begin{aligned} k_B T_c &= \frac{5}{2^{3/4} 3^{7/4}} \epsilon^{1/4} \frac{K_1 V}{S} \frac{|\cot \theta_H|^{1/6}}{1 + |\cot \theta_H|^{2/3}} \left[ 1 - \frac{\epsilon}{2} + \frac{9}{2} \bar{K}_2 (1 + |\cot \theta_H|^{2/3})^{1/2} \right. \\ &\quad \left. + \frac{\bar{K}_2}{2} \frac{|\cot \theta_H|^{2/3} - 21/2}{(1 + |\cot \theta_H|^{2/3})^{1/2}} + \bar{K}_2 \frac{|\cot \theta_H|^{2/3} + 3}{(1 + |\cot \theta_H|^{2/3})^{3/2}} \right] \quad (27) \end{aligned}$$

corresponding to the transition from the quantum to the thermal regime. For a nanometre-scale single-domain FM particle, typical values of the parameters for the magnetic anisotropy coefficients are  $K_1 = 10^8$  erg cm<sup>-3</sup>, and  $K_2 = 10^5$  erg cm<sup>-3</sup>. The radius of the FM particle is about 12 nm and the sublattice spin is  $10^6$ . If  $\epsilon = 0.001$ , we obtain that  $T_c(135^\circ) \sim 203$  mK corresponding to the crossover from the quantum to the classical regime, which compares well with the experimental result of 0.31 K for single-domain FM nanoparticles of barium ferrite (BaFeCoTiO) [7]. Note that, even for  $\epsilon$  as small as  $10^{-3}$ , the angle corresponding to an appreciable change of the orientation of the magnetization vector by quantum tunnelling is  $\delta_2 = \sqrt{6\epsilon}$  rad  $> 4^\circ$ .

The classical action  $S_{cl}$  can be obtained by solving numerically the equations of motion (4a) and (4b). In figure 3 we present the  $\theta_H$ -dependence of  $S_{cl}$  with  $\epsilon = 0.001$  and  $\bar{K}_2 = 0.001$  for  $\pi/2 < \theta_H < \pi$  obtained by numerical and analytical calculations. As is noted in the figure, the analytical result obtained from equation (25) is almost valid over the whole range of angles  $\pi/2 < \theta_H < \pi$ .

By applying the formulae in reference [12], and using equation (25) for the WKB exponent or the classical action, we obtain the tunnelling rate  $\Gamma$  of the magnetization vector in single-domain FM nanoparticles with trigonal crystal symmetry in a magnetic field applied in the



**Figure 3.** The  $\theta_H$ -dependence of the relative classical action  $S_{cl}(\theta_H)/S_{cl}(\theta_H = 3\pi/4)$  in the trigonal symmetry with  $\epsilon = 0.001$  and  $\bar{K}_2 = 0.001$  obtained by numerical and analytical calculations.

range of  $\pi/2 < \theta_H < \pi$  as

$$\Gamma = \frac{2^{31/8} 3^{7/8}}{\pi^{1/2}} \frac{V}{\hbar} K_1 S^{-1/2} \epsilon^{7/8} \frac{|\cot \theta_H|^{1/4}}{1 + |\cot \theta_H|^{2/3}} \left[ 1 - \frac{\epsilon}{4} + \frac{9}{4} \bar{K}_2 (1 + |\cot \theta_H|^{2/3})^{1/2} + \frac{\bar{K}_2}{4} \frac{|\cot \theta_H|^{2/3} - 51/2}{(1 + |\cot \theta_H|^{2/3})^{1/2}} + \frac{\bar{K}_2}{2} \frac{|\cot \theta_H|^{2/3} + 3}{(1 + |\cot \theta_H|^{2/3})^{3/2}} \right] e^{-S_{cl}}. \quad (28)$$

#### 4. MQC and MQT for hexagonal crystal symmetry

In this section, we study the quantum tunnelling of the magnetization vector in single-domain FM particles with hexagonal crystal symmetry whose magnetocrystalline anisotropy energy  $E_a(\theta, \phi)$  at zero magnetic field can be written as

$$E_a(\theta, \phi) = K_1 \sin^2 \theta + K_2 \sin^4 \theta + K_3 \sin^6 \theta - K'_3 \sin^6 \theta \cos(6\phi) \quad (29)$$

where  $K_1$ ,  $K_2$ ,  $K_3$ , and  $K'_3$  are the magnetic anisotropic coefficients. The easy axes are  $\pm \hat{z}$  for  $K_1 > 0$ . When we apply an external magnetic field at an arbitrarily directed angle in the  $ZX$ -plane, the total energy of this system is given by

$$E(\theta, \phi) = E_a(\theta, \phi) - M_0 H_x \sin \theta \cos \phi - M_0 H_z \cos \theta + E_0. \quad (30)$$

By choosing  $K'_3 > 0$ , we take  $\phi = 0$  to be the easy plane, at which the potential energy can be written in terms of dimensionless parameters as

$$\bar{E}(\theta, \phi = 0) = \frac{1}{2} \sin^2 \theta + \bar{K}_2 \sin^4 \theta + (\bar{K}_3 - \bar{K}'_3) \sin^6 \theta - \bar{H} \cos(\theta - \theta_H) + \bar{E}_0 \quad (31)$$

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

Then the initial angle  $\theta_0$  is determined by  $[d\bar{E}(\theta, 0)/d\theta]_{\theta=\theta_0} = 0$ , and the critical angle  $\theta_c$  and the dimensionless critical field  $\bar{H}_c$  by both  $[d\bar{E}(\theta, 0)/d\theta]_{\theta=\theta_c, \bar{H}=\bar{H}_c} = 0$  and

$[d^2\bar{E}(\theta, 0)/d\theta^2]_{\theta=\theta_c, \bar{H}=\bar{H}_c} = 0$ , which leads to

$$\frac{1}{2} \sin(2\theta_0) + \bar{H} \sin(\theta_0 - \theta_H) + 4\bar{K}_2 \sin^4 \theta_0 + 6(\bar{K}_3 - \bar{K}'_3) \sin^5 \theta_0 \cos \theta_0 = 0 \quad (32a)$$

$$\frac{1}{2} \sin(2\theta_c) + \bar{H}_c \sin(\theta_c - \theta_H) + 4\bar{K}_2 \sin^4 \theta_c + 6(\bar{K}_3 - \bar{K}'_3) \sin^5 \theta_c \cos \theta_c = 0 \quad (32b)$$

$$\begin{aligned} \cos(2\theta_c) + \bar{H}_c \cos(\theta_c - \theta_H) + 4\bar{K}_2(3 \sin^2 \theta_c \cos^2 \theta_c - \sin^4 \theta_c) \\ + 6(\bar{K}_3 - \bar{K}'_3)(5 \sin^4 \theta_c \cos^2 \theta_c - \sin^6 \theta_c) = 0. \end{aligned} \quad (32c)$$

Under the assumption that  $|\bar{K}_2|, |\bar{K}_3 - \bar{K}'_3| \ll 1$ , we obtain the dimensionless critical field  $\bar{H}_c$  as

$$\bar{H}_c = \frac{1}{[(\sin \theta_H)^{2/3} + |\cos \theta_H|^{2/3}]^{3/2}} \left[ 1 + \frac{4\bar{K}_2}{1 + |\cot \theta_H|^{2/3}} + \frac{6(\bar{K}_3 - \bar{K}'_3)}{(1 + |\cot \theta_H|^{2/3})^2} \right]. \quad (33)$$

In the limit of small  $\epsilon = 1 - \bar{H}/\bar{H}_c$ , equation (32a) becomes

$$\begin{aligned} -\epsilon \bar{H}_c \sin(\theta_c - \theta_H) + \eta^2 [(3/2)\bar{H}_c \sin(\theta_c - \theta_H) + 3\bar{K}_2 \sin(4\theta_c) \\ + 12(\bar{K}_3 - \bar{K}'_3) \sin^3 \theta_c \cos \theta_c (5 - 8 \sin^2 \theta_c)] + \eta \{ \epsilon \bar{H}_c \cos(\theta_c - \theta_H) \\ - \eta^2 [(1/2)\bar{H}_c \cos(\theta_c - \theta_H) + 4\bar{K}_2 \cos(4\theta_c) \\ + 12(\bar{K}_3 - \bar{K}'_3) \sin^2 \theta_c (5 - 20 \sin^2 \theta_c + 16 \sin^4 \theta_c)] \} = 0 \end{aligned} \quad (34)$$

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

$$\bar{E}(\delta, \phi) = \bar{K}'_3 [1 - \cos(6\phi)] \sin^6(\theta_0 + \delta) + \bar{H}_c (1 - \cos \phi) \sin(\theta_0 + \delta) + \bar{E}_1(\delta) \quad (35)$$

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

$$\begin{aligned} \bar{E}_1(\delta) = \left[ \frac{1}{2} \bar{H}_c \sin(\theta_c - \theta_H) + \bar{K}_2 \sin(4\theta_c) \right. \\ \left. + 4(\bar{K}_3 - \bar{K}'_3)(5 \sin^3 \theta_c \cos^3 \theta_c - 3 \sin^5 \theta_c \cos \theta_c) \right] (\delta^3 - 3\delta^2 \eta) \\ + \left[ \frac{1}{8} \bar{H}_c \cos(\theta_c - \theta_H) + \bar{K}_2 \cos(4\theta_c) \right. \\ \left. + 3(\bar{K}_3 - \bar{K}'_3) \sin^2 \theta_c (\sin^4 \theta_c - 10 \sin^2 \theta_c \cos^2 \theta_c + 5 \cos^4 \theta_c) \right] \\ \times (\delta^4 - 4\delta^3 \eta + 6\delta^2 \eta^2 - 4\delta^2 \epsilon) + \epsilon \delta^2 \left[ 4\bar{K}_2 \cos(4\theta_c) \right. \\ \left. + 12(\bar{K}_3 - \bar{K}'_3) \sin^2 \theta_c (\sin^4 \theta_c - 10 \sin^2 \theta_c \cos^2 \theta_c + 5 \cos^4 \theta_c) \right]. \end{aligned} \quad (36)$$

In the following we investigate the MQC and MQT of the magnetization vector in FM particles with hexagonal crystal symmetry for different angle ranges of the external magnetic field:  $\theta_H = \pi/2, \pi/2 + O(\epsilon^{3/2}) < \theta_H < \pi - O(\epsilon^{3/2})$ , and  $\theta_H = \pi$ .

#### 4.1. $\theta_H = \pi/2$

For  $\theta_H = \pi/2$ , i.e., the external magnetic field is applied perpendicular to the anisotropy axis, we obtain that  $\theta_c = \pi/2$  and  $\eta = \sqrt{2\epsilon}[1 - 4\bar{K}_2 - 12(\bar{K}_3 - \bar{K}'_3)]$ . Now  $\bar{E}_1(\delta)$  becomes

$$\bar{E}_1(\delta) = \frac{1}{8} \left[ 1 + 12\bar{K}_2 + 30(\bar{K}_3 - \bar{K}'_3) \right] \delta^2 \left\{ \delta - 2\sqrt{2\epsilon} \left[ 1 - 4\bar{K}_2 - 12(\bar{K}_3 - \bar{K}'_3) \right] \right\}^2. \quad (37)$$

Substituting equation (37) into the classical equations of motion, we obtain the following instanton solution:

$$\begin{aligned}\bar{\phi} &= i\epsilon \left[ 1 + \frac{\epsilon}{2} - 4\bar{K}_2 - 18\bar{K}'_3 - 6(\bar{K}_3 - \bar{K}'_3) \right] \frac{1}{\cosh^2(\bar{\omega}_c \bar{\tau})} \\ \bar{\delta} &= \sqrt{2\epsilon} \left[ 1 - 4\bar{K}_2 - 12(\bar{K}_3 - \bar{K}'_3) \right] [1 + \tanh(\bar{\omega}_c \bar{\tau})]\end{aligned}\quad (38)$$

which corresponds to the variation of  $\delta$  from  $\delta = 0$  at  $\tau = -\infty$  to

$$\delta = 2\sqrt{2\epsilon} \left[ 1 - 4\bar{K}_2 - 12(\bar{K}_3 - \bar{K}'_3) \right]$$

at  $\tau = +\infty$ , where

$$\bar{\omega}_c = \sqrt{\frac{\epsilon}{2}} \left[ 1 - \frac{\epsilon}{2} + 4\bar{K}_2 + 18\bar{K}'_3 + 6(\bar{K}_3 - \bar{K}'_3) \right].$$

We can calculate the classical action by integrating the Euclidean action of equation (2) with the above instanton solution, and the result is found to be

$$S_{cl} = \frac{2^{5/2}}{3} S \epsilon^{3/2} \left[ 1 + \frac{\epsilon}{2} - 8\bar{K}_2 - 18\bar{K}'_3 - 24(\bar{K}_3 - \bar{K}'_3) \right]. \quad (39)$$

From equation (37) we obtain that the height of barrier is

$$U = 2K_1 V \bar{E}_1(\delta_m) = K_1 V \epsilon^2 \left[ 1 - 4\bar{K}_2 - 18(\bar{K}_3 - \bar{K}'_3) \right]$$

at  $\delta_m = \sqrt{2\epsilon} [1 - 4\bar{K}_2 - 12(\bar{K}_3 - \bar{K}'_3)]$ , and the frequency of oscillation around the minimum of the inverted potential  $-\bar{E}_1(\delta)$  is

$$\bar{\omega}_b = \sqrt{\epsilon} \left[ 1 - \frac{\epsilon}{2} + 4\bar{K}_2 + 18\bar{K}'_3 + 6(\bar{K}_3 - \bar{K}'_3) \right] = \sqrt{2\bar{\omega}_c}.$$

Then the WKB exponent is approximately given by

$$B \sim \frac{U}{\hbar \omega_b} = \frac{1}{2} S \epsilon^{3/2} \left[ 1 + \frac{\epsilon}{2} - 8\bar{K}_2 - 18\bar{K}'_3 - 24(\bar{K}_3 - \bar{K}'_3) \right] \quad (40)$$

which agrees up to the numerical factor with equation (39) obtained by applying the explicit instanton solution. The temperature corresponding to the crossover from the quantum coherence between the degenerate ground-state levels (coherent tunnelling) to the classical over-barrier transition (incoherent transition) is found to be

$$k_B T_c = \frac{3}{2^{5/2}} \epsilon^{1/2} \frac{K_1 V}{S} \left[ 1 - \frac{\epsilon}{2} + 4\bar{K}_2 + 18\bar{K}'_3 + 6(\bar{K}_3 - \bar{K}'_3) \right] \quad (41)$$

and the temperature corresponding to the crossover from quantum coherence between the degenerate ground-state levels (coherent tunnelling) to quantum tunnelling from excited levels (thermally assisted tunnelling or incoherent tunnelling) is found to be

$$k_B T'_c = \frac{1}{\pi} \epsilon^{1/2} \frac{K_1 V}{S} \left[ 1 - \frac{\epsilon}{2} + 4\bar{K}_2 + 18\bar{K}'_3 + 6(\bar{K}_3 - \bar{K}'_3) \right]. \quad (42)$$

By applying the instanton technique for single-domain FM particles in the spin-coherent-state path-integral representation [12], we obtain the instanton's contribution to the tunnel splitting,  $\hbar \Delta_0$ , as

$$\hbar \Delta_0 = \frac{2^{13/4}}{\pi^{1/2}} (V K_1) S^{-1/2} \epsilon^{5/4} \left[ 1 - \frac{\epsilon}{4} + 9\bar{K}'_3 - 6(\bar{K}_3 - \bar{K}'_3) \right] e^{-S_{cl}} \quad (43)$$

where the WKB exponent or the classical action  $S_{cl}$  is clearly shown in equation (39). Then the splitting of the ground state due to resonant coherently quantum tunnelling of the magnetization vector between energetically degenerate states is found to be  $\hbar \Delta = 2\hbar \Delta_0$  for FM particles with hexagonal crystal symmetry in a magnetic field applied perpendicular to the anisotropy axis ( $\theta_H = \pi/2$ ) with the help of the effective Hamiltonian approach.

4.2.  $\pi/2 + O(\epsilon^{3/2}) < \theta_H < \pi - O(\epsilon^{3/2})$

For this case,  $\eta \approx \sqrt{2\epsilon}/3$  and the critical angle  $\theta_c$  is found to be

$$\sin \theta_c = \frac{1}{(1 + |\cot \theta_H|^{2/3})^{1/2}} \left[ 1 + \frac{8\bar{K}_2}{3} \frac{|\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} + 8(\bar{K}_3 - \bar{K}'_3) \frac{|\cot \theta_H|^{2/3}}{(1 + |\cot \theta_H|^{2/3})^2} \right].$$

Now  $\bar{E}_1(\delta)$  becomes

$$\begin{aligned} \bar{E}_1(\delta) = & \frac{1}{2} \frac{|\cot \theta_H|^{1/3}}{1 + |\cot \theta_H|^{2/3}} \left[ 1 - \frac{4\bar{K}_2}{3} \frac{7 - 4|\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} \right. \\ & \left. + 2(\bar{K}_3 - \bar{K}'_3) \frac{11 - 16|\cot \theta_H|^{2/3}}{(1 + |\cot \theta_H|^{2/3})^2} \right] (\sqrt{6\epsilon}\delta^2 - \delta^3). \end{aligned} \tag{44}$$

Then the classical equations of motion have the following bounce solution:

$$\begin{aligned} \bar{\phi} = & i(6\epsilon)^{3/4} |\cot \theta_H|^{1/6} (1 + |\cot \theta_H|^{2/3})^{1/2} \left[ 1 + \frac{\epsilon}{2} - \frac{4\bar{K}_2}{3} \frac{5 - |\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} \right. \\ & \left. - 18\bar{K}'_3 \frac{1}{1 + |\cot \theta_H|^{2/3}} - 2(\bar{K}_3 - \bar{K}'_3) \frac{7 - 6|\cot \theta_H|^{2/3}}{(1 + |\cot \theta_H|^{2/3})^2} \right] \frac{\sinh(\bar{\omega}_c \bar{\tau})}{\cosh^3(\bar{\omega}_c \bar{\tau})} \end{aligned} \tag{45}$$

and

$$\bar{\delta} = \sqrt{6\epsilon} / \cosh^2(\bar{\omega}_c \bar{\tau})$$

corresponding to the variation of  $\delta$  from  $\delta = 0$  at  $\tau = -\infty$  to the turning point  $\delta = \sqrt{6\epsilon}$  at  $\tau = 0$ , and then back to  $\delta = 0$  at  $\tau = +\infty$ , where

$$\begin{aligned} \bar{\omega}_c = & \left( \frac{3}{8} \right)^{1/4} \epsilon^{1/4} \frac{|\cot \theta_H|^{1/6}}{1 + |\cot \theta_H|^{2/3}} \left[ 1 - \frac{\epsilon}{2} + \frac{4\bar{K}_2}{3} \frac{5 - 3|\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} \right. \\ & \left. + 18\bar{K}'_3 \frac{1}{1 + |\cot \theta_H|^{2/3}} + 2(\bar{K}_3 - \bar{K}'_3) \frac{7 - 10|\cot \theta_H|^{2/3}}{(1 + |\cot \theta_H|^{2/3})^2} \right]. \end{aligned}$$

The classical action associated with this bounce solution is found to be

$$\begin{aligned} S_{cl} = & \frac{2^{17/4} 3^{1/4}}{5} S \epsilon^{5/4} |\cot \theta_H|^{1/6} \left[ 1 + \frac{\epsilon}{2} + \frac{4\bar{K}_2}{3} \frac{2 - |\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} \right. \\ & \left. - 18\bar{K}'_3 \frac{1}{1 + |\cot \theta_H|^{2/3}} + 4(\bar{K}_3 - \bar{K}'_3) \frac{2 - 3|\cot \theta_H|^{2/3}}{(1 + |\cot \theta_H|^{2/3})^2} \right]. \end{aligned} \tag{46}$$

For this case, the barrier height  $U (= 2K_1 V \bar{E}_1(\delta_m = 2\sqrt{6\epsilon}/3))$  is given by

$$\begin{aligned} U = & \frac{2^{7/2}}{3^{3/2}} (K_1 V) \epsilon^{3/2} \frac{|\cot \theta_H|^{1/3}}{1 + |\cot \theta_H|^{2/3}} \left[ 1 + \frac{4\bar{K}_2}{3} \frac{7 - 4|\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} \right. \\ & \left. + 2(\bar{K}_3 - \bar{K}'_3) \frac{11 - 16|\cot \theta_H|^{2/3}}{(1 + |\cot \theta_H|^{2/3})^2} \right] \end{aligned}$$

and the frequency of small oscillations of the magnetization vector around the minimum of the inverted potential  $-\bar{E}_1(\delta)$  is

$$\begin{aligned} \bar{\omega}_b = & 3^{1/4} 2^{1/4} \epsilon^{1/4} \frac{|\cot \theta_H|^{1/6}}{1 + |\cot \theta_H|^{2/3}} \left[ 1 - \frac{\epsilon}{2} + \frac{4\bar{K}_2}{3} \frac{5 - 3|\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} \right. \\ & \left. + 18\bar{K}'_3 \frac{1}{1 + |\cot \theta_H|^{2/3}} + 2(\bar{K}_3 - \bar{K}'_3) \frac{7 - 10|\cot \theta_H|^{2/3}}{(1 + |\cot \theta_H|^{2/3})^2} \right] \\ = & 2\bar{\omega}_c. \end{aligned}$$

Then the WKB exponent is approximately given by

$$B \sim \frac{U}{\hbar\omega_b} = \frac{2^{9/4}}{3^{7/4}} S \epsilon^{5/4} |\cot \theta_H|^{1/6} \left[ 1 + \frac{\epsilon}{2} + \frac{4}{3} \bar{K}_2 \frac{2 - |\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} - 18 \bar{K}_3' \frac{1}{1 + |\cot \theta_H|^{2/3}} + 4(\bar{K}_3 - \bar{K}_3') \frac{2 - 3|\cot \theta_H|^{2/3}}{(1 + |\cot \theta_H|^{2/3})^2} \right] \quad (47)$$

which agrees with equation (46) up to the numerical factor. Equating the classical action  $S_{cl}$  to  $U/k_B T_c$ , where  $U$  is the barrier height, we obtain that the crossover from quantum to classical behaviour occurs at

$$k_B T_c = \frac{5}{2^{3/4} 3^{7/4}} \epsilon^{1/4} \frac{K_1 V}{S} \frac{|\cot \theta_H|^{1/6}}{1 + |\cot \theta_H|^{2/3}} \left[ 1 - \frac{\epsilon}{2} + \frac{4}{3} \bar{K}_2 \frac{5 - 3|\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} + 18 \bar{K}_3' \frac{1}{1 + |\cot \theta_H|^{2/3}} + 2(\bar{K}_3 - \bar{K}_3') \frac{7 - 10|\cot \theta_H|^{2/3}}{(1 + |\cot \theta_H|^{2/3})^2} \right]. \quad (48)$$

On the basis of the instanton technique [12], we obtain the tunnelling rate corresponding to the escaping of the magnetization vector from the metastable state for single-domain FM nanoparticles with hexagonal crystal symmetry in a magnetic field applied in the range of  $\pi/2 + O(\epsilon^{3/2}) < \theta_H < \pi - O(\epsilon^{3/2})$  as the following equation:

$$\Gamma = \frac{2^{31/8} 3^{7/8}}{\pi^{1/2}} \frac{V}{\hbar} K_1 S^{-1/2} \epsilon^{7/8} \frac{|\cot \theta_H|^{1/4}}{1 + |\cot \theta_H|^{2/3}} \left[ 1 - \frac{\epsilon}{4} + 9 \bar{K}_3' \frac{1}{1 + |\cot \theta_H|^{2/3}} - \frac{2}{3} \bar{K}_2 \frac{12 - 7|\cot \theta_H|^{2/3}}{1 + |\cot \theta_H|^{2/3}} + 2(\bar{K}_3 - \bar{K}_3') \frac{9 - 13|\cot \theta_H|^{2/3}}{(1 + |\cot \theta_H|^{2/3})^2} \right] e^{-S_{cl}} \quad (49)$$

where the WKB exponent or the classical action  $S_{cl}$  has been clearly shown in equation (46).

#### 4.3. $\theta_H = \pi$

Finally, we study the MQT of the magnetization vector corresponding to the escaping from the metastable state in single-domain FM nanoparticles with hexagonal crystal symmetry in a magnetic field applied at  $\theta_H = \pi$ , i.e., antiparallel to the anisotropy axis. Now the total energy becomes

$$\bar{E}(\delta, \phi) = \bar{K}_3' [1 - \cos(6\phi)] \delta^6 + \frac{1}{2} \delta^2 \left[ \epsilon - \frac{1}{4} (1 - 8\bar{K}_2) \delta^2 \right] - \frac{1}{24} \delta^4 \left\{ \epsilon - \frac{1}{2} \left[ 1 - 32\bar{K}_2 + 48(\bar{K}_3 - \bar{K}_3') \right] \delta^2 \right\}. \quad (50)$$

The classical equations of motion have the bounce solution

$$\bar{\phi} = -i\bar{\omega}_c \bar{\tau} + \frac{n\pi}{3} \quad (51)$$

$$\bar{\delta} = \sqrt{\frac{4\epsilon}{1 - 8\bar{K}_2 - \epsilon[32\bar{K}_3'(1 - \cosh(6\bar{\omega}_c \bar{\tau})) + \frac{1}{3}(1 - 48\bar{K}_2 + 96(\bar{K}_3 - \bar{K}_3'))]}}$$

where  $n = 0, 1, 2, 3, 4, 5$ , and  $\bar{\omega}_c = \epsilon$ . The corresponding classical action is found to be

$$S_{cl} = \frac{2}{3} S \epsilon \frac{1}{\Delta_1} \ln \left( \frac{2\Delta_1}{\Delta_2} \right) \quad (52)$$

with

$$\Delta_1 = 1 - 8\bar{K}_2 - 32\bar{K}_3' \epsilon - \frac{1}{3} \epsilon \left[ 1 - 48\bar{K}_2 + 96(\bar{K}_3 - \bar{K}_3') \right] \quad (53)$$



and

$$\Delta_2 = 32\overline{K}_3'\epsilon. \quad (54)$$

According to the formulae in reference [12], we obtain the tunnelling rate of the magnetization vector escaping from the metastable state for single-domain FM nanoparticles with hexagonal crystal symmetry in a magnetic field applied antiparallel to the anisotropy axis ( $\theta_H = \pi$ ) as

$$\Gamma = \frac{2^{13/2}3^{1/2}}{\pi^{1/2}} \frac{V}{\hbar} K_1 S^{-1/2} \epsilon (1 + 4\overline{K}_2) \times \frac{1}{1 - 16\overline{K}_2 - 64\overline{K}_3'\epsilon - \frac{2}{3}[1 - 48\overline{K}_2 + 96(\overline{K}_3 - \overline{K}_3')]} e^{-S_{cl}} \quad (55)$$

where the WKB exponent or the classical action  $S_{cl}$  is shown in equation (52). Equation (50) shows that in this case  $|\phi| \ll 1$  is not valid, and therefore the problem cannot be reduced to the one-dimensional motion problem. And the effective potential energy and the effective mass in one-dimensional form are not appropriate for the present case.

Now we discuss the range of angles for which equation (46) is valid. Introducing  $\theta_1 = \theta_H - \pi/2$  and  $\theta_2 = \pi - \theta_H$ , from equations (39), (46), and (52), we find that

$$\theta_1 \approx (5^6 2^{-21} 3^{-15/2}) \epsilon^{3/2} \quad \theta_2 \approx (5^{-6} 2^{39/2} 3^{15/2}) \epsilon^{3/2}.$$

This means that equation (46) is almost valid over a wide range of angles  $91^\circ \leq \theta_H \leq 179^\circ$  for  $\epsilon = 0.001$ .

For the single-domain FM nanoparticle with hexagonal crystal symmetry in the presence of an external magnetic field at arbitrarily directed angle, by using equations (39) and (43) for  $\theta_H = \pi/2$ , equations (46) and (49) for  $\pi/2 + O(\epsilon^{3/2}) < \theta_H < \pi - O(\epsilon^{3/2})$ , and equations (52) and (55) for  $\theta_H = \pi$ , we obtain the ground-state tunnel splitting for MQC and the tunnelling rate for MQT of the magnetization vector. Our results show that the tunnel splitting and the tunnelling rate depend distinctly on the orientation of the external magnetic field. When  $\theta_H = \pi/2$ , the magnetic field is applied perpendicular to the anisotropy axis, and when  $\theta_H = \pi$ , the field is antiparallel to the anisotropy axis. It is found that even a very small misalignment of the field with the above two orientations can completely change the results for the tunnelling rates. Another interesting observation concerns the dependence of the WKB exponent or the classical action on the strength of the external magnetic field. Over a wide range of angles, the dependence on  $\epsilon$  ( $=1 - \overline{H}/\overline{H}_c$ ) of the WKB exponent  $S_{cl}$  is given by  $\epsilon^{5/4}$ , not  $\epsilon^{3/2}$ , for  $\theta_H = \pi/2$ , and  $\epsilon$  for  $\theta_H = \pi$ . Therefore, both the orientation and the strength of the external magnetic field are control parameters for an experimental test for MQT and MQC of the magnetization vector in single-domain FM nanoparticles.

## 5. Conclusions

In summary we have investigated the tunnelling behaviours of the magnetization vector in single-domain FM nanoparticles in the presence of an external magnetic field at arbitrarily directed angle. We consider the magnetocrystalline anisotropy with the trigonal crystal symmetry and that with the hexagonal crystal symmetry. By applying the instanton technique in the spin-coherent-state path-integral representation, we obtain both the WKB exponent and the pre-exponential factors in the tunnel splitting between energetically degenerate states in MQC and the tunnelling rate for escaping from a metastable state in MQT of the magnetization vector in the low-barrier limit for the external magnetic field perpendicular to the easy axis ( $\theta_H = \pi/2$ ), for the field antiparallel to the initial easy axis ( $\theta_H = \pi$ ), and for the field at an angle between these two orientations ( $\pi/2 + O(\epsilon^{3/2}) < \theta_H < \pi - O(\epsilon^{3/2})$ ). One

important conclusion is that the tunnelling rate and the tunnel splitting depend distinctly on the orientation of the external magnetic field. Another interesting conclusion concerns the field strength dependence of the WKB exponent or the classical action. We have found that over a wide range of angles, the dependence on  $\epsilon$  ( $=1 - \overline{H}/\overline{H}_c$ ) of the WKB exponent or the classical action  $S_{cl}$  is given by  $\epsilon^{5/4}$ , not  $\epsilon^{3/2}$ , for  $\theta_H = \pi/2$ , and  $\epsilon$  for  $\theta_H = \pi$ . We have obtained the temperatures corresponding to the crossover from the quantum to the thermal regime which are found to depend distinctly on the orientation of the external magnetic field. As a result, we conclude that both the orientation and the strength of the external magnetic field are controllable parameters for an experimental test of the phenomena of macroscopic quantum tunnelling and coherence of the magnetization vector in single-domain FM nanoparticles with trigonal and hexagonal symmetries at a temperature well below the crossover temperature. We have analysed the validity of the semiclassical approximation applied in the present work, and have found that the semiclassical approximation should be already rather good for typical values of the parameters for single-domain FM nanoparticles.

Recently, Wernsdorfer and co-workers performed switching-field measurements on individual ferrimagnetic and insulating BaFeCoTiO nanoparticles containing about  $10^5$ – $10^6$  spins at very low temperatures (0.1–6 K) [7]. They found that above 0.4 K, the magnetization reversal of these particles is unambiguously described by the Néel–Brown theory of thermally activated rotation of the particle's moment over a well-defined anisotropy energy barrier. Below 0.4 K, strong deviations from this model are evidenced which are quantitatively in agreement with the predictions of the MQT theory without dissipation [3]. The BaFeCoTiO nanoparticles have a strong uniaxial magnetocrystalline anisotropy [7]. However, the theoretical results presented here may be useful for checking the general theory for a wide range of systems, with more general symmetries. The experimental procedures for single-domain FM nanoparticles of barium ferrite with uniaxial symmetry [7] may be applied to systems with more general symmetries. Note that the inverse of the WKB exponent  $B^{-1}$  is the magnetic viscosity  $S$  for the quantum-tunnelling-dominated regime  $T \ll T_c$  studied by magnetic relaxation measurements [1]. Therefore, the quantum tunnelling of the magnetization should be checked at any  $\theta_H$  by magnetic relaxation measurements. Over the past few years a lot of experimental and theoretical works have been performed on the spin tunnelling in molecular  $Mn_{12}$ -Ac [22] and  $Fe_8$  [23] 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  $S = 10^2$ – $10^4$ . We hope that the theoretical results presented in this paper may stimulate more experiments whose aim is observing macroscopic quantum phenomena in nanometre-scale single-domain ferromagnets.

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### Appendix A. Evaluation of the pre-exponential factors in the WKB tunnelling rate

In this appendix, we review briefly the procedure for calculating the pre-exponential factors in the WKB rate of quantum tunnelling of the magnetization vector in single-domain FM particles, based on the instanton technique in the spin-coherent-state path-integral representation [12]. The pre-exponential factors in the tunnelling rate (MQT) or the tunnel splitting (MQC) are due to the quantum fluctuations about the classical path, which can be evaluated by expanding the

Euclidean action to second order in small fluctuations. Then we apply this approach to obtain the instanton's contribution to the ground-state tunnel splitting for resonant coherent quantum tunnelling of the magnetization vector in FM particles with trigonal crystal symmetry in an external magnetic field applied perpendicular to the anisotropy axis (considered in section 3) in detail.

In reference [12], Garg and Kim have studied the general formulae for evaluating both the WKB exponent and the pre-exponential factors in the tunnelling rate or the tunnel splitting in the single-domain FM particles on the basis of the instanton technique in the spin-coherent-state path-integral representation, without assuming a specific form of the magnetocrystalline anisotropy and the external magnetic field. Here we explain briefly the basic idea of this calculation. Such a calculation consists of two major steps. The first step is to find the classical, or least-action path (instanton) from the classical equations of motion, which gives the exponent or the classical action in the WKB tunnelling rate. Instantons in one-dimensional field theory can be viewed as pseudoparticles with trajectories existing in the energy barrier, and are therefore responsible for quantum tunnelling. The second step is to expand the Euclidean action to second order in the small fluctuations about the classical path, and then evaluate the Van Vleck determinant of the resulting quadratic form [11, 12]. For single-domain FM particles, writing  $\theta(\tau) = \bar{\theta}(\tau) + \theta_1(\tau)$  and  $\phi(\tau) = \bar{\phi}(\tau) + \phi_1(\tau)$ , where  $\bar{\theta}$  and  $\bar{\phi}$  denote the classical path, one obtains the Euclidean action of equation (2) as  $S_E[\theta(\tau), \phi(\tau)] \approx S_{cl} + \delta^2 S$  with  $S_{cl}$  being the classical action or the WKB exponent and  $\delta^2 S$  being a functional of small fluctuations  $\theta_1$  and  $\phi_1$  [12]:

$$\delta^2 S = -iS \int \frac{d}{d\tau} [\sin \bar{\theta} \theta_1] \phi_1 d\tau + \frac{i}{2} S \int \cos \bar{\theta} \left( \frac{d\bar{\phi}}{d\tau} \right) \theta_1^2 d\tau + \frac{V_0}{2\hbar} \int (E_{\theta\theta} \theta_1^2 + 2E_{\theta\phi} \theta_1 \phi_1 + E_{\phi\phi} \phi_1^2) d\tau \quad (\text{A.1})$$

where

$$\begin{aligned} E_{\theta\theta} &= (\partial^2 E / \partial \theta^2)_{\theta=\bar{\theta}, \phi=\bar{\phi}} \\ E_{\theta\phi} &= (\partial^2 E / \partial \theta \partial \phi)_{\theta=\bar{\theta}, \phi=\bar{\phi}} \\ E_{\phi\phi} &= (\partial^2 E / \partial \phi^2)_{\theta=\bar{\theta}, \phi=\bar{\phi}}. \end{aligned}$$

Under the condition that  $E_{\phi\phi} > 0$ , the Gaussian integration can be performed over  $\phi_1$ , and the remaining path integral over  $\theta_1$  can be cast into the standard form for a one-dimensional motion problem. As usual, there exists a zero mode,  $d\bar{\theta}/d\tau$ , corresponding to a translation of the centre of the instanton, and a negative eigenvalue in the MQT problem [11, 12]. This leads to the imaginary part of the energy, which corresponds to the quantum rate of escape from the metastable state through the classically impenetrable barrier to a stable one. The resonant tunnel splittings of the ground state for the MQC problem can be evaluated by applying a similar technique. What is needed for the calculation of the tunnelling rate (in MQT) and the tunnel splitting (in MQC) is the asymptotic relation of the zero mode,  $d\bar{\theta}/d\tau$ , for large  $\tau$  [11, 12]:

$$d\bar{\theta}/d\tau \approx a e^{-\mu\zeta} \quad \text{as } \zeta \rightarrow \infty. \quad (\text{A.2})$$

The new time variable  $\zeta$  in equation (A2) is related to  $\tau$  as

$$d\zeta = d\tau / 2A(\bar{\theta}(\tau), \bar{\phi}(\tau)) \quad (\text{A.3})$$

where

$$A(\bar{\theta}, \bar{\phi}) = \hbar S^2 \sin^2 \bar{\theta} / 2V E_{\phi\phi}. \quad (\text{A.4})$$

The partial derivatives are evaluated at the classical path. Then the instanton's contribution to the tunnelling rate for MQT or the tunnel splitting for MQC of the magnetization vector in single-domain FM nanoparticles (without the contribution of the topological Wess–Zumino, or Berry phase term in the Euclidean action) is given by [11, 12]

$$|a|(\mu/\pi)^{1/2}e^{-S_{cl}}. \quad (\text{A.5})$$

Therefore, all that is necessary is to differentiate the classical path (instanton) to obtain  $d\bar{\theta}/d\tau$ , then convert from  $\tau$  to the new time variable  $\zeta$  according to equations (A3) and (A4), and read off  $a$  and  $\mu$  by comparison with equation (A2). If the condition  $E_{\phi\phi} > 0$  is not satisfied, one can always perform the Gaussian integration over  $\theta_1$  and end up with a one-dimensional path integral over  $\phi_1$ .

Now we apply this approach to the problem of resonant coherent quantum tunnelling of the magnetization vector between energetically degenerate easy directions in single-domain FM nanoparticles with trigonal crystal symmetry in an external magnetic field applied perpendicular to the anisotropy axis. After some algebra, we find that

$$E_{\phi\phi} \approx 2K_1(1 + 12\bar{K}_2 - \epsilon) \quad (\text{A.6})$$

which is positive. So we can perform the Gaussian integration over  $\phi_1$  directly. The relation between  $\tau$  and the new imaginary-time variable  $\zeta$  for this MQC problem is found to be

$$\tau = \frac{\hbar S^2}{2K_1 V(1 + 12\bar{K}_2 - \epsilon)} \zeta. \quad (\text{A.7})$$

It is easy to differentiate the instanton solution to obtain

$$\frac{d\bar{\delta}}{d\tau} = 8 \frac{K_1 V}{\hbar S} \epsilon \left(1 + 15\bar{K}_2 - \frac{\epsilon}{2}\right) \exp \left[ -\sqrt{2\epsilon} S \left(1 - \frac{3}{2}\bar{K}_2 + \frac{\epsilon}{2}\right) \zeta \right] \quad (\text{A.8})$$

as  $\zeta \rightarrow \infty$ . Thus,

$$|a| = 8 \frac{K_1 V}{\hbar S} \epsilon \left(1 + 15\bar{K}_2 - \frac{\epsilon}{2}\right) \quad (\text{A.9})$$

and

$$\mu = \sqrt{2\epsilon} S \left(1 - \frac{3}{2}\bar{K}_2 + \frac{\epsilon}{2}\right). \quad (\text{A.10})$$

Substituting equations (A9) and (A10) into the general formula (A5), and using equation (17) for the classical action or the WKB exponent, we obtain the instanton's contribution to the tunnel splitting  $\hbar \Delta_0$  as expressed in equation (21) for nanometre-scale single-domain ferromagnets with trigonal crystal symmetry in the presence of an external magnetic field applied perpendicular to the anisotropy axis.

The calculations of the tunnel splitting and the tunnelling rate of the magnetization vector for other MQT and MQC problems considered in the present work can be performed by applying similar techniques, and we will not discuss them any further.

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