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Resonant quantum tunnelling and coherence of the Néel vector for different crystal symmetries

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Abstract. The phenomena of macroscopic quantum tunnelling and coherence of the Néel vector are investigated for small single-domain antiferromagnetic particles. Both the Wentzel–Kramers–Brillouin exponents and the pre-exponential factors are found exactly for the tunnelling rates, for various forms of the magnetocrystalline anisotropy. The calculations are performed on the basis of the two-sublattice model and the instanton method applied to the spin-coherent-state path integral.

1. Introduction

Macroscopic quantum phenomena (MQP) have been studied extensively since Caldeira and Leggett predicted that quantum tunnelling could take place on a macroscopic scale if the dissipative interactions with the environment were small enough [1, 2]. Leggett and co-workers presented a formalism which could include the dissipation by using the imaginary-time path integral and the instanton method, and they found that the rate of quantum tunnelling was reduced by the dissipation in general [1–4]. The Caldeira–Leggett method has been considered extensively for the systems of Josephson junctions [5–7] and superconducting quantum interference devices (SQUIDS) [8].

Recent advances in both materials preparation techniques on nanometre-size magnetic particles and low-temperature magnetometry have made it possible to observe the new MQP in magnetic systems. It has been theoretically pointed out that the magnetization vector can change its direction through an energy barrier by means of quantum tunnelling in small single-domain ferromagnetic (FM) particles at low temperature [9–11]. Similar effects include quantum nucleation of FM bubbles [12] and quantum depinning of domain walls from defects in bulk ferromagnets [13–16]. Several experiments have investigated the quantum tunnelling in small magnetic particles either via relaxation measurements [17–19] or via measurements of the noise spectrum and the ac susceptibility [20, 21]. Experimental results seem to support the idea of magnetic quantum tunnelling.

MQP also exist in the small single-domain antiferromagnetic (AFM) particles in which the Néel vector can tunnel coherently between the easy directions at a temperature well below the anisotropy gap [22–27]. For such quantum tunnelling problems, the difference between an AFM particle and a FM particle originates from the configuration of the spins

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in the classical state. The spins remain exactly parallel in the FM particle. But in the AFM particle, the spins belonging to the two sublattices are inclined with respect to one another, according to the two-sublattice model. Thus, the AFM state is less favourable energetically than the FM state, which leads to a much larger resonance frequency between the wells separated by the magnetic anisotropy in AFM particles than that in FM particles. Formally, the rate Γ for quantum tunnelling can be written as $\Gamma \propto \exp(-U/\hbar\omega_p)$, where U is the energy barrier between the wells and ω_p is the resonance frequency. So the tunnelling rate in an AFM particle is much larger than that in a FM particle. Therefore, an AFM particle is a better candidate as regards the observation of MQP than a FM particle. The quantum tunnelling of the Néel vector was investigated on the basis of the two-sublattice model [22–25, 27] and the anisotropic σ-model [26] independently. Quantum tunnelling is also important in the problems of quantum nucleation of AFM bubbles [25, 26] and quantum depinning of domain walls from defects in bulk antiferromagnets at low temperature [23].

In general, MQP can be classified into macroscopic quantum tunnelling (MQT) and macroscopic quantum coherence (MQC). MQT corresponds to the simple tunnelling of a macroscopic variable through a potential barrier, while MQC corresponds to the resonance of two energetically degenerate states. The tunnelling behaviours of the Néel vectors in MQT and MQC problems will be considered for small single-domain AFM particles in this paper. In previous work, the exponential factors in the Wentzel–Kramers–Brillouin (WKB) rates were calculated for a few simple examples of MQC and MQT of the Néel vector, but the pre-exponential factors in the tunnelling rates were not definitively established [22–24, 27]. The purpose of the present paper is to extend the previous results by calculating both the WKB exponents and the pre-exponential factors in the tunnelling rates (for MQT problems) or the tunnel splittings (for MQC problems) for all major crystal symmetries. So the results obtained in this paper will be more applicable in experimental checks. All of the calculations in this paper are performed in terms of the spin-coherent-state path integral.

This paper is organized as follows. In section 2, we present a formalism for evaluating the exponent and the prefactors in the WKB tunnelling rate for a more general form of the magnetocrystalline anisotropy energy and the Zeeman energy when a magnetic field is applied. In section 3, we apply the general formulae of section 2 to MQT of the Néel vector for biaxial and tetragonal crystal symmetries, and in section 4, to MQC for cubic, uniaxial and hexagonal crystal symmetries. Finally, a summary will be given in section 5.

2. Calculation of the tunnelling rate for the AFM particles

In this section, we will present a formalism for calculating the tunnelling rate (in the MQT problem) and the tunnel splitting (in the MQC problem) for the Néel vector in a small AFM particle on the basis of the two-sublattice model and the instanton method applied to the spin-coherent-state path integral, without assuming a specific form of the magnetocrystalline anisotropy and the Zeeman energies.

According to the two-sublattice model [23], there is a strong exchange energy $\boldsymbol{m}_1 \cdot \boldsymbol{m}_2/\chi_\perp$ for the two sublattices, where $\boldsymbol{m}_1$, $\boldsymbol{m}_2$ are the magnetization vectors of the two sublattices with large, fixed and unequal magnitudes, and $\chi_\perp$ is the transverse susceptibility. Under the assumption that the exchange energy for the two sublattices is much larger than the magnetocrystalline anisotropy energy and the Zeeman energy when a magnetic field is applied, the Euclidean action for a small noncompensated AFM particle (neglecting dissipation) is given by [22, 23]

$$S_E[\theta(\boldsymbol{x},\tau),\phi(\boldsymbol{x},\tau)] = \frac{1}{\hbar}\int \mathrm{d}\tau \int \mathrm{d}^3x \ \left\{\frac{\chi_\perp}{2\gamma^2}\left[\left(\frac{\mathrm{d}\theta}{\mathrm{d}\tau}\right)^2 + \left(\frac{\mathrm{d}\phi}{\mathrm{d}\tau}\right)^2 \sin^2\theta\right]\right.$$
$$\left. + \ \frac{1}{2}\alpha\left[(\nabla\theta)^2 + (\nabla\phi)^2\sin^2\theta\right] + E(\theta,\phi)\right\} \tag{1}$$

where γ is the gyromagnetic ratio, α is the exchange constant associated with the boundary effect of the particle surface and $\tau = \mathrm{i}t$ is the imaginary time. θ and ϕ, which can determine the direction of the Néel vector, are the angular components of $\boldsymbol{m}_1$ in the spherical coordinate system. The magnetocrystalline anisotropy and Zeeman energies are included in the $E(\theta,\phi)$ term in equation (1).

As pointed out in references [22] and [23], for a nanometre-size AFM particle, the Néel vector may depend on the imaginary time but not on the coordinates, because the large spatial derivatives in equation (1) are suppressed by the exchange interaction between two sublattices. So all of the calculations done in the present work are for the homogeneous Néel vector. Therefore, equation (1) reduces to

$$S_E(\theta,\phi) = \frac{V}{\hbar}\int \mathrm{d}\tau \ \left\{\frac{\chi_\perp}{2\gamma^2}\left[\left(\frac{\mathrm{d}\theta}{\mathrm{d}\tau}\right)^2 + \left(\frac{\mathrm{d}\phi}{\mathrm{d}\tau}\right)^2\sin^2\theta\right] + E(\theta,\phi)\right\} \tag{2}$$

where V is the volume of the AFM particle.

To obtain the tunnelling rate for MQT or the tunnel splitting for MQC, the following path integral should be calculated:

$$\int \mathrm{D}\{\theta\}\ \mathrm{D}\{\phi\}\ \exp[-S_E(\theta,\phi)] \tag{3}$$

where the Euclidean action $S_E(\theta,\phi)$ has been defined in equation (2). The paths appearing in the above equation are fixed at the end points $\tau = \pm T/2$.

Now we use the standard instanton method to evaluate the path integral in equation (3). The calculation consists of two major steps. The first step is that of finding the classical or least-action path which gives the WKB exponential factor. The second step is that of evaluating the Van Vleck determinant of the small fluctuations about the classical path, which gives the pre-exponential factors in the tunnelling rate. The calculations for MQT and MQC are very similar, so we will discuss only the former explicitly.

To execute the first step, we must find the classical path $(\bar{\theta},\bar{\phi})$ with the boundary conditions $\bar{\theta}(\tau = \pm T/2) = \theta_\pm$ and $\bar{\phi}(\tau = \pm T/2) = \phi_\pm$. The classical path satisfies the following equations of motion ($\delta S_E = 0$):

$$\begin{aligned} &\frac{\chi_\perp}{\gamma^2}\frac{\mathrm{d}^2\bar{\theta}}{\mathrm{d}\tau^2} = \frac{\chi_\perp}{\gamma^2}\left(\frac{\mathrm{d}\bar{\phi}}{\mathrm{d}\tau}\right)^2\sin\bar{\theta}\cos\bar{\theta} + \frac{\partial E}{\partial\theta} \\ &\frac{\chi_\perp}{\gamma^2}\ \frac{\mathrm{d}}{\mathrm{d}\tau}\left[\left(\frac{\mathrm{d}\bar{\phi}}{\mathrm{d}\tau}\right)\sin^2\bar{\theta}\right] = \frac{\partial E}{\partial\phi}. \end{aligned} \tag{4}$$

In order to evaluate the Van Vleck determinant for small fluctuations about the classical path, we write

$$\theta(\tau) = \bar{\theta}(\tau) + \theta_1(\tau) \qquad \phi(\tau) = \bar{\phi}(\tau) + \phi_1(\tau) \tag{5}$$

and expand the Euclidean action in equation (2) to the second order of θ_1 and ϕ_1, which gives the following expression:

$$S_E(\theta,\phi) = S_{cl} + \delta^2 S \tag{6}$$

where

$$\delta^2 S = \frac{V}{\hbar}\int \mathrm{d}\tau \left[\frac{\chi_\perp}{2\gamma^2}\left(\frac{\mathrm{d}\theta_1}{\mathrm{d}\tau}\right)^2 + \frac{\chi_\perp}{2\gamma^2}\sin^2\bar{\theta}\left(\frac{\mathrm{d}\phi_1}{\mathrm{d}\tau}\right)^2 + \frac{\chi_\perp}{\gamma^2}\sin 2\bar{\theta}\left(\frac{\mathrm{d}\bar{\phi}}{\mathrm{d}\tau}\right)\left(\frac{\mathrm{d}\phi_1}{\mathrm{d}\tau}\right)\theta_1 \right.$$
$$\left. + \frac{\chi_\perp}{2\gamma^2}\cos 2\bar{\theta}\left(\frac{\mathrm{d}\bar{\phi}}{\mathrm{d}\tau}\right)^2\theta_1^2 + \frac{1}{2}(E_{\theta\theta}\theta_1^2 + 2E_{\theta\phi}\theta_1\phi_1 + E_{\phi\phi}\phi_1^2)\right]. \tag{7}$$

S_{cl} in equation (6) is the classical action which minimizes the Euclidean action. $E_{\theta\theta}$, $E_{\theta\phi}$ and $E_{\phi\phi}$ in equation (7) are defined as

$$E_{\theta\theta} = \left.\frac{\partial^2 E}{\partial\theta^2}\right|_{\theta=\bar{\theta},\phi=\bar{\phi}} \qquad E_{\theta\phi} = \left.\frac{\partial^2 E}{\partial\theta\,\partial\phi}\right|_{\theta=\bar{\theta},\phi=\bar{\phi}} \qquad E_{\phi\phi} = \left.\frac{\partial^2 E}{\partial\phi^2}\right|_{\theta=\bar{\theta},\phi=\bar{\phi}}$$

respectively. Assuming that

$$\frac{1}{2}E_{\phi\phi} + \frac{\chi_\perp}{4\gamma^2}\,\frac{\mathrm{d}}{\mathrm{d}\tau}\left[\sin 2\bar{\theta}\left(\frac{\mathrm{d}\bar{\theta}}{\mathrm{d}\tau}\right)\right] > 0$$

we can perform the Gaussian integration over ϕ_1, which leads to the effective action for θ_1 only:

$$I(\theta_1) = \int \mathrm{d}\tau \left[A\left(\frac{\mathrm{d}\theta_1}{\mathrm{d}\tau}\right)^2 + B\theta_1\left(\frac{\mathrm{d}\theta_1}{\mathrm{d}\tau}\right) + C\theta_1^2\right]. \tag{8}$$

Here,

$$A = \frac{V}{\hbar}\frac{\chi_\perp}{2\gamma^2}\left\{1 - \left[\frac{\chi_\perp}{2\gamma^2}\sin^2 2\bar{\theta}\left(\frac{\mathrm{d}\bar{\phi}}{\mathrm{d}\tau}\right)^2\right]\Big/\left(\frac{1}{2}E_{\phi\phi} + \frac{\chi_\perp}{4\gamma^2}\,\frac{\mathrm{d}}{\mathrm{d}\tau}\left[\sin 2\bar{\theta}\left(\frac{\mathrm{d}\bar{\theta}}{\mathrm{d}\tau}\right)\right]\right)\right\} \tag{9a}$$

$$B = \frac{V}{\hbar}\frac{\chi_\perp}{2\gamma^2}\sin 2\bar{\theta}\left(\frac{\mathrm{d}\bar{\phi}}{\mathrm{d}\tau}\right)$$
$$\times \left\{E_{\theta\phi} - \frac{\chi_\perp}{\gamma^2}\frac{\mathrm{d}}{\mathrm{d}\tau}\left[\sin 2\bar{\theta}\left(\frac{\mathrm{d}\bar{\phi}}{\mathrm{d}\tau}\right)\right]\right\}\Big/\left\{\frac{1}{2}E_{\phi\phi} + \frac{\chi_\perp}{4\gamma^2}\,\frac{\mathrm{d}}{\mathrm{d}\tau}\left[\sin 2\bar{\theta}\left(\frac{\mathrm{d}\bar{\theta}}{\mathrm{d}\tau}\right)\right]\right\} \tag{9b}$$

$$C = \frac{V}{\hbar}\left\{\frac{1}{2}E_{\theta\theta} + \frac{\chi_\perp}{2\gamma^2}\left(\frac{\mathrm{d}\bar{\phi}}{\mathrm{d}\tau}\right)^2\cos 2\bar{\theta}\right.$$
$$\left. - \frac{1}{4}\left[E_{\theta\phi} - \frac{\chi_\perp}{\gamma^2}\,\frac{\mathrm{d}}{\mathrm{d}\tau}\left[\sin 2\bar{\theta}\left(\frac{\mathrm{d}\bar{\phi}}{\mathrm{d}\tau}\right)\right]\right]^2\Big/\left[\frac{1}{2}E_{\phi\phi} + \frac{\chi_\perp}{4\gamma^2}\,\frac{\mathrm{d}}{\mathrm{d}\tau}\left[\sin 2\bar{\theta}\left(\frac{\mathrm{d}\bar{\theta}}{\mathrm{d}\tau}\right)\right]\right]\right\}. \tag{9c}$$

We now turn to the normalization factor for the remaining path integral over θ_1. In the spin-coherent-state representation, the measure of the path integral in equation (3) is defined as

$$\int \mathrm{D}\{\theta\}\ \mathrm{D}\{\phi\} = \lim_{n\to\infty}\prod_{k=1}^{n}\left[\frac{2S+1}{4\pi}\right]\int \sin\bar{\theta}_k\ \mathrm{d}\theta_{1,k}\ \mathrm{d}\phi_{1,k} \tag{10}$$

where $\theta_k = \theta(-T/2 + k\eta)$ and $\phi_k = \phi(-T/2 + k\eta)$, and $\eta = T/(n+1)$ is the width of the imaginary-time slices. S in equation (10) is the total spin in one sublattice of the AFM particle. In addition to generating contributions to the B- and C-terms in equation (8), the Gaussian integration over $\phi_{1,k}$ will yield a factor of

$$\left\{2\pi\hbar\Big/\left(\eta V\left[E_{\phi\phi}(\bar{\theta}_k,\bar{\phi}_k) + \frac{\chi_\perp}{2\gamma^2}\,\frac{\mathrm{d}}{\mathrm{d}\tau}\left(\sin 2\bar{\theta}\left(\frac{\mathrm{d}\bar{\theta}}{\mathrm{d}\tau}\right)\right)\bigg|_{\bar{\theta}=\bar{\theta}_k}\right]\right)\right\}^{1/2}. \tag{11}$$

Then the path integral in equation (3) can be written as

$$N' \mathrm{e}^{-S_{cl}} \int [\mathrm{d}\theta_1]\ \mathrm{e}^{-I[\theta_1(\tau)]} \tag{12}$$

where

$$N' = \lim_{n\to\infty} \prod_{k=1}^{n} \left[\frac{2S+1}{2}\right] \times \sqrt{\hbar \Big/ \left\{\eta\, 2\pi V \left[E_{\phi\phi}(\bar\theta_k, \bar\phi_k) + \frac{\chi_\perp}{2\gamma^2}\, \frac{\mathrm{d}}{\mathrm{d}\tau}\left(\sin 2\bar\theta \left(\frac{\mathrm{d}\bar\theta}{\mathrm{d}\tau}\right)\right)\Big|_{\bar\theta=\bar\theta_k}\right]\right\}}\ \sin\bar\theta_k. \tag{13}$$

It is easy to obtain the following relation for the transverse susceptibility $\chi_\perp$ with the exchange energy density J for the two sublattices [24]:

$$\chi_\perp = \frac{\hbar^2\gamma^2}{JV^2} S^2. \tag{14}$$

In the limit of large S, equation (13) reduces to

$$N' = \lim_{n\to\infty} \prod_{k=1}^{n} (A'_k/\pi\eta)^{1/2} \tag{15}$$

where

$$A'_k = J\frac{V}{\hbar}\frac{\chi_\perp}{2\gamma^2} \sin^2\bar\theta_k \Big/ \left[E_{\phi\phi}(\bar\theta_k, \bar\phi_k) + \frac{\chi_\perp}{2\gamma^2}\, \frac{\mathrm{d}}{\mathrm{d}\tau}\left(\sin 2\bar\theta\left(\frac{\mathrm{d}\bar\theta}{\mathrm{d}\tau}\right)\right)\Big|_{\bar\theta=\bar\theta_k}\right]. \tag{16}$$

Next, we change τ to a new time variable ζ, which is defined by

$$\mathrm{d}\zeta = \mathrm{d}\tau/2A'(\bar\theta(\tau), \bar\phi(\tau)). \tag{17}$$

Then, in terms of discretized variables, the path integral in equation (3) can be cast into the standard form for a one-dimensional motion problem [11, 28–30]:

$$\mathrm{e}^{-S_{cl}} \lim_{n\to\infty} \left[\prod_{k=1}^{n} \int \frac{\mathrm{d}\theta_{1,k}}{\sqrt{2\pi\Delta_k}}\right] \exp\left\{-\sum_{k=1}^{n}\left[\frac{1}{2\Delta_k}\left(\frac{A_k}{A'_k}\right)(\theta_{1,k}-\theta_{1,k-1})^2 + 2\Delta_k A'_k C'_k \theta_{1,k}^2\right]\right\} \tag{18}$$

where $\theta_{1,0} = 0$ and Δ_k, the width of the kth imaginary-time slice in ζ, is given by

$$\Delta_k = \zeta_k - \zeta_{k-1} = \eta/2A'_k. \tag{19}$$

We have defined $A_k = A(\bar\theta_k, \bar\phi_k)$ and $C'_k = C'(\bar\theta_k, \bar\phi_k)$ in equation (18), where

$$C' = C - \frac{V}{2\hbar}\frac{\chi_\perp}{\gamma^2}\, \frac{\mathrm{d}}{\mathrm{d}\tau}\left\{\sin 2\bar\theta\left(\frac{\mathrm{d}\bar\phi}{\mathrm{d}\tau}\right) \times \left(E_{\theta\phi} - \frac{\chi_\perp}{\gamma^2}\, \frac{\mathrm{d}}{\mathrm{d}\tau}\left[\sin 2\bar\theta\left(\frac{\mathrm{d}\bar\phi}{\mathrm{d}\tau}\right)\right]\right)\Big/\left(\frac{1}{2}E_{\phi\phi} + \frac{\chi_\perp}{4\gamma^2}\, \frac{\mathrm{d}}{\mathrm{d}\tau}\left[\sin 2\bar\theta\left(\frac{\mathrm{d}\bar\theta}{\mathrm{d}\tau}\right)\right]\right)\right\}. \tag{20}$$

The remaining procedure for evaluating the Van Vleck fluctuation determinant of the quadratic form of θ_1 in equation (18) for the AFM particles is very similar to that for the FM particles [11]. Here we only give a summary of how to evaluate the tunnelling rate or the tunnel splitting for the AFM particle. The first step is to obtain the classical path which satisfies the boundary conditions from the equations of motion. The second step is to differentiate the classical path to obtain $\mathrm{d}\bar{\theta}/\mathrm{d}\tau$, then convert from τ to the new time variable ζ according to the relation in equation (17), which gives

$$\frac{\mathrm{d}\bar{\theta}}{\mathrm{d}\tau} = a\mathrm{e}^{-\mu\zeta} \qquad \text{as } \zeta \to \infty. \tag{21}$$

Then the tunnelling rate Γ is [11, 28–30]

$$\Gamma = k_\zeta |a| (\mu/\pi)^{1/2} \mathrm{e}^{-S_{cl}} \tag{22}$$

where k_ζ is the number of equivalent escape directions, i.e., the number of paths which have the same classical action. Only the asymptotic relation in equation (21) is needed for calculating the tunnelling rate, and this is usually easy to obtain.

In performing the Gaussian integration over ϕ_1, we have assumed that

$$\frac{1}{2}E_{\phi\phi} + \frac{\chi_\perp}{4\gamma^2}\,\frac{\mathrm{d}}{\mathrm{d}\tau}\left[\sin 2\bar{\theta}\left(\frac{\mathrm{d}\bar{\theta}}{\mathrm{d}\tau}\right)\right] > 0. \tag{23}$$

If the above condition is not satisfied, we can always finish the calculation by performing the Gaussian integration over θ_1. In this case, the condition of the positivity of the coefficient of θ_1^2 can be written as

$$\frac{1}{2}E_{\theta\theta} + \frac{\chi_\perp}{2\gamma^2}\cos 2\bar{\theta}\left(\frac{\mathrm{d}\bar{\phi}}{\mathrm{d}\tau}\right)^2 > 0. \tag{24}$$

After performing the Gaussian integration over θ_1, the effective action for ϕ_1 is given by

$$I(\phi_1) = \int \mathrm{d}\tau \;\left[E\left(\frac{\mathrm{d}\phi_1}{\mathrm{d}\tau}\right)^2 + F\phi_1\left(\frac{\mathrm{d}\phi_1}{\mathrm{d}\tau}\right) + G\phi_1^2\right] \tag{25}$$

with

$$\begin{aligned}
E &= \frac{V}{\hbar}\frac{\chi_\perp}{2\gamma^2}\sin^2\bar{\theta}\left[1 - \left\{4\frac{\chi_\perp}{\gamma^2}\cos^2\bar{\theta}\left(\frac{\mathrm{d}\bar{\phi}}{\mathrm{d}\tau}\right)^2\right\}\Big/\left\{E_{\theta\theta} + \frac{\chi_\perp}{\gamma^2}\cos 2\bar{\theta}\left(\frac{\mathrm{d}\bar{\phi}}{\mathrm{d}\tau}\right)^2\right\}\right] \\
F &= -\frac{V}{\hbar}\frac{\chi_\perp}{\gamma^2}\left[E_{\theta\phi}\sin 2\bar{\theta}\left(\frac{\mathrm{d}\bar{\phi}}{\mathrm{d}\tau}\right)\right]\Big/\left[E_{\theta\theta} + \frac{\chi_\perp}{\gamma^2}\cos 2\bar{\theta}\left(\frac{\mathrm{d}\bar{\phi}}{\mathrm{d}\tau}\right)^2\right] \\
G &= \frac{V}{2\hbar}\left[E_{\phi\phi} - E_{\theta\phi}^2\Big/\left\{E_{\theta\theta} + \frac{\chi_\perp}{\gamma^2}\cos 2\bar{\theta}\left(\frac{\mathrm{d}\bar{\phi}}{\mathrm{d}\tau}\right)^2\right\}\right].
\end{aligned} \tag{26}$$

The Van Vleck fluctuation determinant can be evaluated by using the techniques already described, and we will not discuss it any further. In the following two sections, we will apply the formulae derived in this section to calculate both the WKB exponents and the Van Vleck fluctuation determinants of the tunnel splittings (in MQC problems) and the tunnelling rates (in MQT problems) for the Néel vector in small AFM particles for different forms of the magnetocrystalline anisotropy energies and the external magnetic fields.

3. Macroscopic quantum tunnelling

In this section, we will apply the formalism of the previous section to investigate the tunnelling behaviours of the Néel vector in MQT problems with biaxial and tetragonal crystal symmetries, separately.

3.1. Biaxial symmetry

The system that we consider has biaxial symmetry. Let the easy axis be $\boldsymbol{z}$, and the hard axis be $\boldsymbol{x}$. In the presence of an external magnetic field $\boldsymbol{H}$ antiparallel to $\boldsymbol{z}$, the $E(\theta, \phi)$ term in the Euclidean action can be written as

$$E(\theta, \phi) = (K_1 + K_2 \sin^2 \phi) \sin^2 \theta - mH(1 - \cos\theta) \tag{27}$$

where K_1 and K_2 are the longitudinal and transverse anisotropy coefficients, respectively. Like in the problem studied in reference [23], we also assume that the transverse anisotropy coefficient is much larger than the longitudinal one, which agrees with the experimental situation for highly anisotropic materials (such as rare-earth materials). m in equation (27) is defined as $m = m_1 - m_2 = \hbar\gamma s/V \ll m_1$, where s is the excess spin of the AFM particle due to the small noncompensation of the two sublattices.

When $H < H_c = 2K_1/m$, the energy minima of the system are at $\phi = 0$ and $\theta = 0, \pi$. H_c is the coercive field at which the initial state becomes classically unstable. We note that there also exists a spin-flop field which can destroy the spin configuration in an AFM particle. The magnitude of such a field is smaller than that of the coercive field in general. So all of the calculation done in this section is under the condition that the applied magnetic field is smaller than the spin-flop field. Therefore, the two-sublattice configuration is still valid for the AFM particles at $H \neq 0$.

In the presence of a magnetic field applied in the $-\boldsymbol{z}$-direction, there is a metastable state at $\theta = 0$, $\phi = 0$. To decay out of the metastable state, the Néel vector must rotate by the angle $\pm\theta_1$, which satisfies

$$\sin^2\left(\frac{\theta_1}{2}\right) = \epsilon \tag{28}$$

where $\epsilon = 1 - H/H_c$. Substituting equation (27) into the classical equations of motion, we obtain the following bounce solution for $0 < \epsilon < 1$:

$$\bar{\phi} = 0$$
$$\sin^2\left(\frac{\bar{\theta}}{2}\right) = \frac{1 - \tanh^2(\omega_0\sqrt{\epsilon}\tau)}{\lambda - \tanh^2(\omega_0\sqrt{\epsilon}\tau)}$$

where

$$\omega_0 = \frac{V}{\hbar S}\sqrt{2K_1 J} \qquad \text{and} \qquad \lambda = 1/\epsilon \tag{29}$$

corresponding to the variations of $\bar{\theta}$ from $\bar{\theta} = 0$ at $\tau = -\infty$ to the turning points $\bar{\theta} = \pm\theta_1$ at $\tau = 0$, and then back to $\bar{\theta} = 0$ at $\tau = +\infty$. The classical action, $S_{cl}^{B.S.}$, associated with the bounce path for the biaxial symmetry is found to be

$$S_{cl}^{B.S.} = 2^{5/2}\sqrt{\frac{K_1}{J}}S\left[\sqrt{\epsilon} - \frac{(1-\epsilon)}{2}\ln\left(\frac{1+\sqrt{\epsilon}}{1-\sqrt{\epsilon}}\right)\right] \tag{30}$$

where S is the total spin in one sublattice of the AFM particle.

To evaluate the prefactors, we note that

$$\frac{1}{2}E_{\phi\phi} + \frac{\chi_\perp}{4\gamma^2}\frac{\mathrm{d}}{\mathrm{d}\tau}\left[\sin 2\bar{\theta}\left(\frac{\mathrm{d}\bar{\theta}}{\mathrm{d}\tau}\right)\right] = K_2 \sin^2\bar{\theta} + 4K_1 \sin^2\frac{\bar{\theta}}{2}\left(\epsilon - \sin^2\frac{\bar{\theta}}{2}\right)$$
$$+\ K_1 \sin^2\bar{\theta}\left(\epsilon - 10\epsilon\sin^2\frac{\bar{\theta}}{2} - 2\sin^2\frac{\bar{\theta}}{2} + 12\sin^4\frac{\bar{\theta}}{2}\right) \tag{31}$$

which is positive, so we can integrate out ϕ_1. After some complicated calculations, we obtain the following relation between τ and the new time variable ζ:

$$\tau = \frac{\hbar}{2VK_2}S^2\zeta + \frac{\hbar}{VK_2}\sqrt{\frac{2K_1}{J}}S\left[3\sqrt{\epsilon} - (1-\epsilon)\ln\left(\frac{1+\sqrt{\epsilon}}{1-\sqrt{\epsilon}}\right)\right]. \tag{32}$$

And it is easy to differentiate the classical path to obtain

$$\frac{\mathrm{d}\bar{\theta}}{\mathrm{d}\tau} = 4\frac{V}{\hbar S}\sqrt{2K_1J}\frac{\epsilon}{\sqrt{1-\epsilon}}\exp\left\{-\frac{2K_1}{K_2}\left[3\epsilon - \sqrt{\epsilon}(1-\epsilon)\ln\left(\frac{1+\sqrt{\epsilon}}{1-\sqrt{\epsilon}}\right)\right]\right\}$$
$$\times\ \exp\left(-\sqrt{\frac{K_1J\epsilon}{2K_2^2}}S\zeta\right) \qquad \text{as } \zeta\to\infty. \tag{33}$$

Thus,

$$|a| = 4\frac{V}{\hbar S}\sqrt{2K_1J}\frac{\epsilon}{\sqrt{1-\epsilon}}\exp\left\{-\frac{2K_1}{K_2}\left[3\epsilon - \sqrt{\epsilon}(1-\epsilon)\ln\left(\frac{1+\sqrt{\epsilon}}{1-\sqrt{\epsilon}}\right)\right]\right\}$$

and

$$\mu = \sqrt{\frac{K_1J\epsilon}{2K_2^2}}S. \tag{34}$$

Substituting equation (34) into the general formula (22), and using $k_\zeta = 2$ and equation (30) for $S_{cl}^{B.S.}$, we obtain the tunnelling rate for this MQT problem:

$$\Gamma^{B.S.} = \frac{2^{13/4}}{\sqrt{\pi}}\frac{V}{\hbar}K_2\left(\frac{K_1J}{K_2^2}\right)^{3/4}\frac{\epsilon^{5/4}}{\sqrt{1-\epsilon}}\exp\left\{-\frac{2K_1}{K_2}\right.$$
$$\times\ \left.\left[3\epsilon - \sqrt{\epsilon}(1-\epsilon)\ln\left(\frac{1+\sqrt{\epsilon}}{1-\sqrt{\epsilon}}\right)\right]\right\}S^{-1/2}\mathrm{e}^{-S_{cl}^{B.S.}}. \tag{35}$$

The same model has been considered, but only the WKB exponent has been calculated in reference [27] for the limiting case $H\to H_c$. The WKB exponent in reference [27] is consistent with the result in equation (30) of the present work for the small noncompensated AFM particle at $H\to H_c$. Furthermore, both the WKB exponent and the pre-exponential factors in the tunnelling rate are evaluated exactly for $0 < H < H_c$ in the present work.

Suppose that the excess spin of the AFM particle is solely due to the small noncompensation of two sublattices at the surface. It has been argued [22] that for an AFM particle with N spins, $N^{2/3}$ spins are at the surface, and thus the number of excess spins due to statistical fluctuations of the shape is about $(N^{2/3})^{1/2} = N^{1/3}$. For a particle of about 10^3 spins, the number of excess spins would be 10, which is a small fraction of the $N\sim 10^3$ spins in the particle.

Typical values of parameters for the small AFM particle are $K_1 = 10^5$ erg cm^{-3}, $K_2 = 10^7$ erg cm^{-3} and $J = 3.0\times 10^9$ erg cm^{-3}. The particle radius is $R = 30$ Å and the total spin in one sublattice is $S = 5000$. For these values, the MQT rate would be 1.20×10^{-2} s^{-1} for $H/H_c = 0.55$ ($\epsilon = 0.45$) and 1.97×10^5 s^{-1} for $H/H_c = 0.7$ ($\epsilon = 0.3$). The tunnelling rate is found to increase significantly with the external magnetic field because the field decreases the energy barrier between the two nonequivalent wells.

3.2. Tetragonal symmetry

The $E(\theta,\phi)$ term for tetragonal crystal symmetry is given by

$$E(\theta,\phi) = K_1\sin^2\theta - K_2\cos(4\phi)\sin^4\theta. \tag{36}$$

We shall consider the case in which $K_2 > K_1 > 0$ where $\phi = 0$, $\theta = 0$ is a metastable state and $\phi = 0$, $\theta = \pi/2$ is a stable state for the Néel vector. To decay out of the metastable state, the Néel vector must rotate by the angle $\pm\theta_1$, which satisfies

$$\sin^2\theta_1 = 1/\nu \tag{37}$$

where $\nu = K_2/K_1 > 1$. The maximum of $E(\theta, \phi)$ corresponds to $\sin^2\theta_2 = 1/2\nu$. Substituting equation (36) into the classical equations of motion, we obtain the following bounce solution:

$$\begin{aligned} \bar{\phi} &= 0 \\ \sin^2\bar{\theta} &= \frac{1-\tanh^2(\omega_0\tau)}{\nu-\tanh^2(\omega_0\tau)} \end{aligned} \tag{38}$$

corresponding to the variations of $\bar{\theta}$ from $\bar{\theta} = 0$ at $\tau = -\infty$ to the turning points $\bar{\theta} = \pm\theta_1$ at $\tau = 0$, and then back to $\bar{\theta} = 0$ at $\tau = +\infty$. ω_0 in equation (38) is defined as

$$\omega_0 = \frac{V}{\hbar S}\sqrt{2K_1 J}.$$

The classical action, $S_{cl}^{T.S.}$, associated with each bounce path for the tetragonal symmetry is found to be

$$S_{cl}^{T.S.} = \sqrt{\frac{2K_1}{J}}S\left[1 - \frac{\nu-1}{2\sqrt{\nu}}\ln\left(\frac{\sqrt{\nu}+1}{\sqrt{\nu}-1}\right)\right]. \tag{39}$$

Note that

$$\frac{1}{2}E_{\phi\phi} + \frac{\chi_\perp}{4\gamma^2}\,\frac{\mathrm{d}}{\mathrm{d}\tau}\left[\sin 2\bar{\theta}\left(\frac{\mathrm{d}\bar{\theta}}{\mathrm{d}\tau}\right)\right] = K_1\sin^2\bar{\theta}\left[2 + (5\nu-3)\sin^2\bar{\theta} + 4\nu\sin^4\bar{\theta}\right] \tag{40}$$

which is positive, so we can integrate out ϕ_1. The relation between τ and the new time variable ζ is then found to be

$$\tau = \frac{JV\chi_\perp}{4\hbar\gamma^2 K_1}\zeta + \frac{1}{\omega_0}\left[1 - \frac{7\nu-1}{4\sqrt{\nu}}\ln\left(\frac{\sqrt{\nu}+1}{\sqrt{\nu}-1}\right)\right]. \tag{41}$$

As $\zeta \to \infty$, we have

$$\frac{\mathrm{d}\bar{\theta}}{\mathrm{d}\tau} = 2^{3/2}\frac{V}{\hbar S}\sqrt{K_1 J}\frac{1}{\sqrt{\nu-1}}\exp\left[\frac{7\nu-1}{4\sqrt{\nu}}\ln\left(\frac{\sqrt{\nu}+1}{\sqrt{\nu}-1}\right) - 1\right]\exp\left(-\frac{1}{4}\sqrt{\frac{2J}{K_1}}S\zeta\right). \tag{42}$$

Thus,

$$|a| = 2^{3/2}\frac{V}{\hbar S}\sqrt{K_1 J}\frac{1}{\sqrt{\nu-1}}\exp\left[\frac{7\nu-1}{4\sqrt{\nu}}\ln\left(\frac{\sqrt{\nu}+1}{\sqrt{\nu}-1}\right) - 1\right]$$

and

$$\mu = \frac{1}{4}\sqrt{\frac{2J}{K_1}}S. \tag{43}$$

Substituting equation (43) into the general formula (22), and using $k_\zeta = 4$ and equation (39) for the classical action, we finally obtain the tunnelling rate for the tetragonal crystal symmetry:

$$\Gamma^{T.S.} = \frac{2^{11/4}}{\pi^{1/2}}\frac{V}{\hbar}K_1^{1/4}J^{3/4}\frac{1}{\sqrt{\nu-1}}\exp\left[\frac{7\nu-1}{4\sqrt{\nu}}\ln\left(\frac{\sqrt{\nu}+1}{\sqrt{\nu}-1}\right) - 1\right]S^{-1/2}\mathrm{e}^{-S_{cl}^{T.S.}}. \tag{44}$$

For $K_1 = 10^5$ erg cm^{-3}, $J = 3.0 \times 10^9$ erg cm^{-3}, $R = 30$ Å and $S = 5000$, we obtain $\Gamma^{T.S.} = 3.71 \times 10^4$ s^{-1} for $\nu = 1.2$ and $\Gamma^{T.S.} = 2.15 \times 10^8$ s^{-1} for $\nu = 1.5$. It is found that the tunnelling rate is larger for higher ν ($=K_2/K_1$). So we predict that highly anisotropic materials would be likely to exhibit MQP in AFM systems.

4. Macroscopic quantum coherence

In this section we will apply the formalism in section 2 to three examples of MQC. In a small AFM particle, MQC corresponds to the resonance of the Néel vector between the energetically degenerate easy directions. The MQC problems studied in this section are for cubic, uniaxial and hexagonal crystal symmetries, respectively.

4.1. Cubic symmetry

In the absence of an external magnetic field, the $E(\theta, \phi)$ term for cubic crystal symmetry is given by

$$E(\theta, \phi) = K_1(\alpha_x^2\alpha_y^2 + \alpha_y^2\alpha_z^2 + \alpha_z^2\alpha_x^2) \tag{45}$$

where α_x, α_y and α_z are the direction cosines of the Néel vector. In terms of θ and ϕ, equation (45) can be written as

$$E(\theta, \phi) = \frac{1}{8}K_1 \sin^4\theta(1 - \cos 4\phi) + \frac{1}{8}K_1(1 - \cos 4\theta). \tag{46}$$

Here we will consider the tunnelling behaviours of the Néel vector for the cases where $K_1 > 0$ and $K_1 < 0$ cases individually.

If $K_1 > 0$, the energy minima of the system correspond to $\phi = 0$ and $\theta = 0, \pi/2$. Then the easy axis is along [100]. If we denote the two states as $|1\rangle$ and $|2\rangle$, other energy minima will repeat the two states with period π. So the Néel vector can resonate between these energetically degenerate directions. Substituting equation (46) into the classical equations of motion, we obtain the following instanton solution corresponding to the switching of the Néel vector from $\bar{\theta} = 0$ at $\tau = -\infty$ to $\bar{\theta} = \pi/2$ at $\tau = +\infty$:

$$\bar{\phi} = 0$$
$$\sin 2\bar{\theta} = \frac{1}{\cosh(\omega_0\tau)} \tag{47}$$

where ω_0 is the same as in section 3. The classical action, $S_{cl}^{C.S.}$, associated with this instanton for the cubic symmetry is then found to be

$$S_{cl}^{C.S.} = \sqrt{\frac{K_1}{2J}}S. \tag{48}$$

To find the prefactors, we note that

$$\frac{1}{2}E_{\phi\phi} + \frac{\chi_\perp}{4\gamma^2}\ \frac{\mathrm{d}}{\mathrm{d}\tau}\left[\sin 2\bar{\theta}\left(\frac{\mathrm{d}\bar{\theta}}{\mathrm{d}\tau}\right)\right] = K_1\left[\frac{1}{4} + \frac{1}{4}\cos^2 2\bar{\theta} - \frac{1}{2}\cos^3 2\bar{\theta}\right] \tag{49}$$

which is positive, so ϕ_1 can be integrated out. Then we obtain the relation of τ with the new time variable ζ:

$$\tau = \frac{JV\chi_\perp}{2\hbar\gamma^2 K_1}\zeta + \frac{1}{2\omega_0}(2 - \ln 2). \tag{50}$$

It is easy to differentiate the classical path to obtain

$$\frac{\mathrm{d}\bar{\theta}}{\mathrm{d}\tau} = \frac{2}{e}\frac{V}{\hbar S}\sqrt{K_1 J}\exp\left(-\sqrt{\frac{J}{2K_1}}S\zeta\right) \qquad \text{as } \zeta \to \infty. \tag{51}$$

Thus,

$$|a| = \frac{2}{e}\frac{V}{\hbar S}\sqrt{K_1 J}$$

and

$$\mu = \sqrt{\frac{J}{2K_1}} S. \tag{52}$$

Substituting equation (52) into the general formula (22), we obtain one instanton's contribution, $\hbar\Delta_{O.I.}^{C.S.}$, to the tunnel splitting for the cubic symmetry for the $K_1 > 0$ case:

$$\hbar\Delta_{O.I.}^{C.S.} = \frac{2^{3/4}}{e\pi^{1/2}} V K_1^{1/4} J^{3/4} S^{-1/2} \mathrm{e}^{-S_{cl}^{C.S.}}. \tag{53}$$

Now we use the effective-Hamiltonian method [4] to obtain the ground-state tunnelling level splittings for this system. The effective Hamiltonian of the system can be written as

$$H_{eff} = -\hbar\Delta_{O.I.}^{C.S.}\mathcal{M} \tag{54}$$

where $\mathcal{M}$ is a linear operator defined by

$$\mathcal{M}|j\rangle = |j+1\rangle + |j-1\rangle \tag{55}$$

where $|j\rangle$ is one of the two energetically degenerate states. For the present case, the matrix form of H_{eff} is

$$H_{eff} = \begin{bmatrix} 0 & -2\hbar\Delta_{O.I.}^{C.S.} \\ -2\hbar\Delta_{O.I.}^{C.S.} & 0 \end{bmatrix}. \tag{56}$$

Then a simple diagonalization of H_{eff} shows that the energies are $\pm 2\hbar\Delta_{O.I.}^{C.S.}$. Therefore, the tunnel splitting of the ground state is $\Delta^{C.S.} = 4\Delta_{O.I.}^{C.S.}$, which is equivalent to $k_\zeta = 4$ in the general formula (22).

For $K_1 = 10^5$ erg cm^{-3}, $J = 3.0 \times 10^9$ erg cm^{-3}, $R = 30$ Å and $S = 5000$, we obtain the tunnel splitting of the ground state $\Delta^{C.S.} = 4.83 \times 10^5$ s^{-1} for cubic symmetry for the $K_1 > 0$ case.

If $K_1 < 0$, the easy axis is along [111]. Now the energy minima of the system correspond to $\phi = \pi/4$ and $\theta = \theta_1$, $\pi - \theta_1$, where $\sin^2\theta_1 = 2/3$. If we denote the two states as $|1\rangle$ and $|2\rangle$, other energy minima will repeat the two states with period π. A simple analysis of $E(\theta, \phi)$ shows that there are two types of instanton for the present case. We use A to denote the instanton passing through the barrier at $\theta = \pi/2$ from $\theta = \theta_1$ to $\theta = \pi - \theta_1$, and B to denote that passing through the barrier at $\theta = \pi$ from $\theta = \pi - \theta_1$ to $\theta = \pi + \theta_1$ $(=\theta_1)$.

Substituting equation (46) into the classical equations of motion, we obtain the instanton-A solution for the $K_1 < 0$ case:

$$\bar{\phi}_A = \pi/4$$
$$\tan\bar{\theta}_A = -\frac{\tan\theta_1}{\tanh(\omega_1\tau)} \tag{57}$$

corresponding to the transition of the Néel vector from $\bar{\theta} = \theta_1$ at $\tau = -\infty$ to $\bar{\theta} = \pi - \theta_1$ at $\tau = +\infty$, and the instanton-B solution:

$$\bar{\phi}_B = \pi/4$$
$$\tan\bar{\theta}_B = \tan\theta_1 \tanh(\omega_1\tau) \tag{58}$$

corresponding to the transition of the Néel vector from $\bar{\theta} = \pi - \theta_1$ at $\tau = -\infty$ to $\bar{\theta} = \pi + \theta_1$ $(=\theta_1)$ at $\tau = +\infty$, where

$$\omega_1 = \frac{V}{\hbar S}\sqrt{\frac{1}{3}|K_1| J}.$$

And the classical actions for instantons A and B are found to be

$$S_A^{C.S.} = \sqrt{\frac{|K_1|}{3J}}\left[1 - \frac{1}{\sqrt{2}}\arctan\left(\frac{1}{\sqrt{2}}\right)\right]S \tag{59}$$

$$S_B^{C.S.} = \sqrt{\frac{|K_1|}{3J}}\left[1 + \frac{1}{\sqrt{2}}\arctan(\sqrt{2})\right]S \tag{60}$$

for the $K_1 < 0$ case. It is noted that $S_A^{C.S.} < S_B^{C.S.}$ because the energy barrier that the instanton B must tunnel through is higher than that for the instanton A.

Now we turn to the prefactors. Now,

$$\frac{1}{2}E_{\phi\phi} + \frac{\chi_\perp}{4\gamma^2}\frac{\mathrm{d}}{\mathrm{d}\tau}\left[\sin 2\bar{\theta}\left(\frac{\mathrm{d}\bar{\theta}}{\mathrm{d}\tau}\right)\right] = |K_1|\left(\frac{1}{3} - \frac{8}{3}\sin^2\bar{\theta} + \frac{25}{4}\sin^4\bar{\theta} - 3\sin^6\bar{\theta}\right) \tag{61}$$

which is positive for the instanton A or B, so we can integrate out ϕ_1 directly. The relation of τ with the new time variable ζ is then found to be

$$\tau = \frac{3JV\chi_\perp}{4\hbar\gamma^2|K_1|}\zeta + \frac{3}{4}\frac{1}{\omega_1} - \frac{11}{4\sqrt{2}}\frac{1}{\omega_1}\arctan\left(\frac{1}{\sqrt{2}}\right) \tag{62}$$

for the instanton A and

$$\tau = \frac{3JV\chi_\perp}{4\hbar\gamma^2|K_1|}\zeta + \frac{3}{4}\frac{1}{\omega_1} + \frac{11}{4\sqrt{2}}\frac{1}{\omega_1}\arctan(\sqrt{2}) \tag{63}$$

for the instanton B. It is easy to show that, as $\zeta \to \infty$,

$$\frac{\mathrm{d}\bar{\theta}_A}{\mathrm{d}\tau} = \frac{2^{5/2}}{3^{3/2}}\frac{V}{\hbar S}\sqrt{J|K_1|}\exp\left[-\frac{3}{2} + \frac{11}{2^{3/2}}\arctan\left(\frac{1}{\sqrt{2}}\right)\right]\exp\left(-\frac{1}{2}\sqrt{\frac{3J}{|K_1|}}S\zeta\right) \tag{64}$$

and

$$\frac{\mathrm{d}\bar{\theta}_B}{\mathrm{d}\tau} = \frac{2^{5/2}}{3^{3/2}}\frac{V}{\hbar S}\sqrt{J|K_1|}\exp\left[-\frac{3}{2} - \frac{11}{2^{3/2}}\arctan(\sqrt{2})\right]\exp\left(-\frac{1}{2}\sqrt{\frac{3J}{|K_1|}}S\zeta\right). \tag{65}$$

Reading off $|a|$ and μ in equations (64) and (65), and substituting them into the general formula (22), we obtain the contributions to the tunnel splitting of this system, $\hbar\Delta_A^{C.S.}$ and $\hbar\Delta_B^{C.S.}$ corresponding to the instantons A and B respectively:

$$\hbar\Delta_A^{C.S.} = \frac{2^{3/2}}{\pi^{1/2}3^{5/4}}V|K_1|^{1/4}J^{3/4}\exp\left[-\frac{3}{2} + \frac{11}{2^{3/2}}\arctan\left(\frac{1}{\sqrt{2}}\right)\right]S^{-1/2}\mathrm{e}^{-S_A^{C.S.}} \tag{66}$$

$$\hbar\Delta_B^{C.S.} = \frac{2^{3/2}}{\pi^{1/2}3^{5/4}}V|K_1|^{1/4}J^{3/4}\exp\left[-\frac{3}{2} - \frac{11}{2^{3/2}}\arctan(\sqrt{2})\right]S^{-1/2}\mathrm{e}^{-S_B^{C.S.}}. \tag{67}$$

Now the matrix form of the effective Hamiltonian for $K_1 < 0$ is

$$H_{eff} = \begin{bmatrix} 0 & -\hbar(\Delta_A^{C.S.} + \Delta_B^{C.S.}) \\ -\hbar(\Delta_A^{C.S.} + \Delta_B^{C.S.}) & 0 \end{bmatrix}. \tag{68}$$

Then the eigenvalues of the system are found to be $\pm\hbar(\Delta_A^{C.S.} + \Delta_B^{C.S.})$, where $\Delta_A^{C.S.} > \Delta_B^{C.S.}$. Therefore, the tunnel splitting of the ground state is $\Delta^{C.S.} = 2(\Delta_A^{C.S.} + \Delta_B^{C.S.})$.

Taking $|K_1| = 10^5$ erg cm^{-3}, $J = 3.0 \times 10^9$ erg cm^{-3}, $R = 30$ Å and $S = 5000$, the tunnel splitting of the ground state $\Delta^{C.S.}$ would be 5.52×10^{10} s^{-1} for the $K_1 < 0$ case.

4.2. Uniaxial symmetry

Our second example of MQC is a system with an easy axis $\boldsymbol{z}$ and a hard axis $\boldsymbol{x}$. The magnetic field is applied along $\boldsymbol{x}$. Now the $E(\theta,\phi)$ term can be written as

$$\begin{aligned} E(\theta,\phi) &= K_1\sin^2\theta + K_2\sin^2\theta\sin^2\phi - mH\sin\theta\cos\phi + m^2H^2/4K_1 \\ &= K_1(\sin\theta-\sin\theta_0)^2 + 2K_1\sin\theta_0\sin\theta(1-\cos\phi) + K_2\sin^2\theta\sin^2\phi \end{aligned} \tag{69}$$

where $K_2 \gg K_1 > 0$ and $\sin\theta_0 = mH/2K_1$. It is also assumed in this section that the applied magnetic field is smaller than the spin-flop field, which is smaller than the coercive field $H_c = 2K_1/m$ in general. The energy minima of the system are at $\phi = 0$ and $\theta = \theta_0$, $\pi - \theta_0$.

Substituting equation (69) into the classical equations of motion, we obtain the instanton solution

$$\begin{aligned} &\bar\phi = 0 \\ &\sin\bar\theta = \frac{1+\sin\theta_0\cosh(\omega_0\cos\theta_0\tau)}{\sin\theta_0+\cosh(\omega_0\cos\theta_0\tau)} \end{aligned} \tag{70}$$

which corresponds to the variation of $\bar\theta$ from $\bar\theta = \theta_0$ at $\tau = -\infty$ to $\bar\theta = \pi - \theta_0$ at $\tau = +\infty$, where

$$\omega_0 = \frac{V}{\hbar S}\sqrt{2K_1J}.$$

The classical action associated with this instanton for the uniaxial symmetry is found to be

$$S_{cl}^{U.S.} = 2^{3/2}\sqrt{\frac{K_1}{J}}S\cos\theta_0\left[1-2\tan\theta_0\arctan\left(\frac{\cos\theta_0}{1+\sin\theta_0}\right)\right]. \tag{71}$$

To find the prefactors, we note that

$$\begin{aligned} \frac{1}{2}E_{\phi\phi} + \frac{\chi_\perp}{4\gamma^2}\,\frac{\mathrm{d}}{\mathrm{d}\tau}\left[\sin 2\bar\theta\left(\frac{\mathrm{d}\bar\theta}{\mathrm{d}\tau}\right)\right] &= K_2\sin^2\bar\theta + K_1(\sin\bar\theta-\sin\theta_0)^2 \\ &\quad - 2K_1\sin^2\bar\theta(\sin\bar\theta-\sin\theta_0)^2 + K_1\sin^2\bar\theta - K_1\sin^3\bar\theta(\sin\bar\theta-\sin\theta_0) \end{aligned} \tag{72}$$

which is positive, so we can integrate out ϕ_1. After some complicated calculations, we obtain the relation between τ and the new time variable ζ:

$$\begin{aligned} \tau &= \frac{\hbar}{2K_2V}S^2\zeta - \frac{K_1}{K_2}\frac{1}{\omega_0}\ln\left(\frac{1+\sin\theta_0+\cos\theta_0}{1+\sin\theta_0-\cos\theta_0}\right) \\ &\quad + 4\frac{K_1}{K_2}\frac{1}{\omega_0}\cos\theta_0\left[1-\tan\theta_0\arctan\left(\frac{\cos\theta_0}{1+\sin\theta_0}\right)\right] \end{aligned} \tag{73}$$

for $0 < H < H_c$. It is a simple matter to show that, as $\zeta \to \infty$,

$$\begin{aligned} \frac{\mathrm{d}\bar\theta}{\mathrm{d}\tau} &= 2\omega_0\cos^2\theta_0\left(\frac{1+\sin\theta_0+\cos\theta_0}{1+\sin\theta_0-\cos\theta_0}\right)^{(K_1/K_2)\cos\theta_0}\exp\left\{-4\frac{K_1}{K_2}\cos^2\theta_0\right. \\ &\quad\times\left.\left[1-\tan\theta_0\arctan\left(\frac{\cos\theta_0}{1+\sin\theta_0}\right)\right]\right\}\exp\left(-\sqrt{\frac{K_1J}{2K_2^2}}S\cos\theta_0\zeta\right). \end{aligned} \tag{74}$$

Reading off $|a|$ and μ in equation (74), and substituting them into the general formula (22), we obtain one instanton's contribution, $\hbar\Delta_{O.I.}^{U.S.}$, to the tunnel splitting of this system as

$$\begin{aligned} \hbar\Delta_{O.I.}^{U.S.} &= \frac{2^{5/4}}{\sqrt{\pi}}VK_2\left(\frac{K_1J}{K_2^2}\right)^{3/4}S^{-1/2}(\cos\theta_0)^{5/2}\left(\frac{1+\sin\theta_0+\cos\theta_0}{1+\sin\theta_0-\cos\theta_0}\right)^{(K_1/K_2)\cos\theta_0} \\ &\quad\times\exp\left\{-4\frac{K_1}{K_2}\cos^2\theta_0\left[1-\tan\theta_0\arctan\left(\frac{\cos\theta_0}{1+\sin\theta_0}\right)\right]\right\}\mathrm{e}^{-S_{cl}^{U.S.}}. \end{aligned} \tag{75}$$

Now we apply the effective-Hamiltonian method to obtain the ground-state tunnelling level splittings. For this case, the effective Hamiltonian can be expressed as

$$H_{eff} = \begin{bmatrix} 0 & -\hbar\Delta_{O.I.}^{U.S.} \\ -\hbar\Delta_{O.I.}^{U.S.} & 0 \end{bmatrix}. \tag{76}$$

The energies are $\pm\hbar\Delta_{O.I.}^{U.S.}$. Therefore, the tunnel splitting of the ground state is $\Delta^{U.S.} = 2\Delta_{O.I.}^{U.S.}$ for the uniaxial symmetry.

To illustrate this, for the AFM particle with $K_1 = 10^5$ erg cm^{-3}, $K_2 = 10^7$ erg cm^{-3}, $J = 3.0 \times 10^9$ erg cm^{-3}, particle radius $R = 30$ Å and the total spin in one sublattice $S = 5000$, the tunnel splitting would be 6.56×10^{-3} s^{-1} for $H/H_c = 0.4$ ($\epsilon = 0.6$) and 1.06×10^5 s^{-1} for $H/H_c = 0.6$ ($\epsilon = 0.4$). It is clearly shown that the tunnel splitting increases significantly with the external magnetic field because the angle through which the Néel vector must tunnel is decreased by the magnetic field.

4.3. Hexagonal symmetry

Our third example of MQC for the Néel vector is a system with hexagonal crystal symmetry, which has six easy axes in the basal plane. Now the magnetocrystalline anisotropy energy can be written as

$$E(\theta, \phi) = K_1 \sin^2\theta + K_2 \sin^4\theta + K_3 \sin^6\theta - K_3' \sin^6\theta \cos(6\phi). \tag{77}$$

We assume that $K_1 < 0$ and $0 < K_2, K_3, K_3' \ll |K_1|$. The easy directions of this system are at $\theta = \pi/2$ and $\phi = 0, \pi/3, 2\pi/3, \pi, 4\pi/3, 5\pi/3$. We denote these six states as $|1\rangle$, $|2\rangle$, $|3\rangle$, $|4\rangle$, $|5\rangle$ and $|6\rangle$; other energy minima will repeat the six states with period 2π.

Since $|K_1| \gg K_2, K_3, K_3' > 0$, the Néel vector is forced to lie in the x–y plane. We find that the instanton solution of the equations of motion for equation (77) is given by

$$\bar{\theta} = \pi/2 \qquad \sin 3\bar{\phi} = \frac{1}{\cosh(3\omega_0\tau)} \tag{78}$$

corresponding to the variation of $\bar{\phi}$ from $\bar{\phi} = 0$ at $\tau = -\infty$ to $\bar{\phi} = \pi/3$ at $\tau = +\infty$. ω_0 in equation (78) is defined as

$$\omega_0 = 2\frac{V}{\hbar S}\sqrt{K_3' J}.$$

The classical action associated with this instanton for the hexagonal symmetry is found to be

$$S_{cl}^{H.S.} = \frac{4}{3}\sqrt{\frac{K_3'}{J}}S. \tag{79}$$

We now turn to the prefactors. Now,

$$\frac{1}{2}E_{\theta\theta} + \frac{\chi_\perp}{2\gamma^2}\cos 2\bar{\theta}\left(\frac{d\bar{\phi}}{d\tau}\right)^2 = |K_1| - 2K_2 - 3K_3 + 3K_3' - 8K_3'\sin^2(3\phi) = |K_1| + o(|K_1|) > 0 \tag{80}$$

so we can integrate out θ_1. Then the relation between τ and the new time variable ζ is found to be

$$\tau = \frac{JV\chi_\perp}{2\hbar\gamma^2|K_1|}\zeta + \frac{8}{3}\frac{K_3'}{|K_1|}\frac{1}{\omega_0}. \tag{81}$$

It is easy to differentiate the instanton solution to obtain

$$\frac{\mathrm{d}\bar{\phi}}{\mathrm{d}\tau} = 4\frac{V}{\hbar S}\sqrt{JK_3'}\exp\left(-8\frac{K_3'}{|K_1|}\right)\exp\left(-3\sqrt{\frac{JK_3'}{|K_1|^2}}S\zeta\right) \qquad \text{as } \zeta \to \infty. \tag{82}$$

Thus,

$$|a| = 4\frac{V}{\hbar S}\sqrt{JK_3'}\exp\left(-8\frac{K_3'}{|K_1|}\right)$$

and

$$\mu = 3\sqrt{\frac{JK_3'}{|K_1|^2}}S. \tag{83}$$

Substituting equation (83) into the general formula (22), we obtain one instanton's contribution, $\hbar\Delta_{O.I.}^{H.S.}$, to the tunnel splitting of this system as

$$\hbar\Delta_{O.I.}^{H.S.} = \frac{2^{5/2}}{\sqrt{\pi}}V|K_1|\left(\frac{JK_3'}{|K_1|^2}\right)^{3/4}S^{-1/2}\exp\left(-8\frac{K_3'}{|K_1|}\right)\mathrm{e}^{-S_{cl}^{H.S.}}. \tag{84}$$

For this case, the matrix form of the effective Hamiltonian is

$$H_{eff} = -\hbar\Delta_{O.I.}^{H.S.}\begin{bmatrix} 0 & 1 & 0 & 0 & 0 & 1 \\ 1 & 0 & 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 & 0 & 1 \\ 1 & 0 & 0 & 0 & 1 & 0 \end{bmatrix}. \tag{85}$$

The energies are $\pm 2\hbar\Delta_{O.I.}^{H.S.}$ and $\pm\hbar\Delta_{O.I.}^{H.S.}$, the latter two levels being doubly degenerate. Therefore, the greatest tunnel splitting of the ground state is $\Delta^{H.S.} = 4\Delta_{O.I.}^{H.S.}$, which is equivalent to $k_\zeta = 4$ in the general formula (22).

For $K_3' = 10^5$ erg cm^{-3}, $|K_1| = 10^7$ erg cm^{-3}, $J = 3.0 \times 10^9$ erg cm^{-3}, $R = 30$ Å and $S = 5000$, the greatest tunnel splitting of the ground state is found to be 8.47×10^{-3} s^{-1}.

5. Summary

The phenomena of macroscopic quantum tunnelling and coherence of the Néel vector have been considered for small single-domain AFM particles in the present work. The Néel vector can tunnel out of the metastable easy directions or resonate between energetically degenerate easy directions at low temperature. The previously known WKB exponents in the tunnelling rates for these processes are supplemented by calculating the prefactors in this paper. The formalism for evaluating both the WKB exponent and the Van Vleck fluctuation determinant for the tunnelling rate (in the MQT problem) or the tunnel splitting (in the MQC problem) of the Néel vector has been developed by using the spin-coherent-state path integral, on the basis of the two-sublattice model for AFM particles. Then this formalism is applied to investigate the tunnelling behaviours of the Néel vector for all of the major crystal symmetries. Both the WKB exponent and the pre-exponential factors in the tunnelling rate or the tunnel splitting are found exactly for each case with the help of the instanton method applied to the imaginary-time path integral. We hope that the theoretical results obtained in the present work will stimulate more experiments whose aim is observing the macroscopic quantum phenomena in small single-domain antiferromagnets.

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