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Magnetic quantum coherence in trigonal and hexagonal systems

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Abstract. The tunneling behaviors of the magnetization vector are studied in ferromagnetic systems with trigonal and hexagonal crystal symmetries, respectively. The Euclidean transition amplitudes between the energetically degenerate easy directions are evaluated with the help of the dilute instanton-gas approximation. By using the effective Hamiltonian method, the ground-state tunneling level splittings are clearly shown for each kind of symmetry and are found to depend on the parity of the total spin of the ferromagnetic particle. The effective Hamiltonian method is demonstrated to be equivalent to the dilute instanton-gas approximation. Possible relevance to experiments is discussed.

PACS. 73.40.Gk Tunneling – 75.10.Jm Quantized spin models – 75.45.+j Macroscopic quantum phenomena in magnetic systems

1 Introduction

Over recent years, owing mainly to the rapid development in nanostructure physics and in new technology of the highly sensitive SQUID magnetometry, there has been growing interest in studying Macroscopic Quantum Phenomena (MQP) in magnetic systems. At a sufficiently low temperature, all the spins in a grain are locked together by the exchange interaction, and only their global orientation can change. The magnetic MQP can be largely classified into Macroscopic Quantum Tunneling (MQT) and Coherence (MQC). The phenomenon of MQT corresponds to the escaping of the magnetization vector from a metastable state by quantum tunneling in the presence of an external magnetic field, while MQC corresponds to the resonance of the magnetization vector between neighboring degenerate states separated by the magnetocrystalline anisotropy or the applied magnetic field. Particular cases of the magnetic MQP are quantum tunneling of the magnetization vector in single-domain ferromagnetic (FM) nanoparticles [1–6], quantum nucleation of the FM bubble [7,8], quantum depinning of the FM domain wall from defects [9–12], and quantum tunneling of the Néel vector in single-domain antiferromagnetic (AFM) nanoparticles [13–18]. There have been many experiments involving resonance measurements [19], magnetic relaxation [20] and hysteresis loop study [21] for various systems, which seem to support the idea of quantum magnetic tunneling.

One of the most striking effects in magnetic MQP is that for some spin systems with high symmetries, the tunneling behaviors seem sensitive to the parity of the total spin of the magnetic nanoparticle. Because of the magnetocrystalline anisotropies and external magnetic fields, the energy of a single-domain FM nanoparticle depends on the orientation of the magnetization vector. Therefore, there are usually two or more energetically degenerate easy directions, and thus different tunneling paths connecting the same initial and final states. It has been theoretically demonstrated that the Berry phase or the Wess-Zumino term in the magnetic action can lead to the destructive interference between topologically different tunneling paths for the half-integer total spin FM nanoparticle, and the quenching of the ground-state tunneling level splitting in the absence of an external magnetic field [22,23]. This spin-parity effect can be related to Kramers' degeneracy if the magnetic system has time-reversal invariance, but it can also exist in the system without Kramers' degeneracy [24]. A similar effect can take place in the AFM nanoparticle in which only an integer excess spin can tunnel but not a half-integer one [14,15]. Recently, the quantum interference effect has been studied theoretically for the resonant tunneling of the magnetization vector in the FM system with an external magnetic field applied along either the hard [24], easy [25,26] or medium axis [27].

In this paper, we investigate the tunneling behaviors in resonant quantum coherence of the magnetization vector in a single-domain FM nanoparticle with trigonal and hexagonal crystal symmetries, respectively. Both the

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Wentzel-Kramers-Brillouin (WKB) exponent and the preexponential factors in one instanton's contribution to the tunneling level splitting are calculated with the help of the standard instanton method in spin-coherent-state path integral [2]. The Euclidean transition amplitudes between energetically degenerate easy directions are evaluated in the dilute instanton-gas approximation [28]. The final results of the ground-state tunneling level splittings are obtained by using a recently proposed method of the effective Hamiltonian [29,30]. The effective Hamiltonian method is shown to be equivalent to the dilute instanton-gas approximation [28]. The spin-parity or quantum interference effect is also discussed for each kind of symmetry. The thermodynamic properties of the tunneling states (such as the specific heat) are found to depend on the parity of the spin quantum number, which may provide an experimental test for the quantum interference effect in single-domain FM nanoparticles.

This paper is organized as follows. In Section 2, we briefly review some basic ideas of the standard instanton method for MQT and MQC of the magnetization vector in the single-domain FM nanoparticle based on the spin-coherent-state path integral. In Sections 3 and 4, we consider the ground-state tunneling level splittings for the FM systems with trigonal and hexagonal crystal symmetries, respectively. And the conclusions and discussions are presented in Section 5.

2 The instanton method for MQT and MQC in the FM system

The system of interest is a single-domain FM nanoparticle at a temperature well below its anisotropy gap. For such a spin system, the tunneling splitting for MQC or the tunneling rate for MQT of the magnetization vector is determined by the imaginary-time transition amplitude from an initial state $|i\rangle$ to a final state $|f\rangle$ as

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

where S_E is the Euclidean action and $D\Omega$ is the measure of the path integral.

In the spin-coherent-state representation, the Euclidean action for a single-domain FM nanoparticle can be written as

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

where V is the volume of the FM particle and γ is the gyromagnetic ratio. $M_0 = |\mathbf{M}| = \hbar\gamma S/V$ is the magnitude of the magnetization vector $\mathbf{M}$, where S is the total spin of the FM nanoparticle. The polar angle θ and the azimuthal angle ϕ label the spin coherent state $|\theta,\phi\rangle$. The spin coherent state is defined as the maximum eigenstates of S_z, rotated into the direction of the unit vector $\mathbf{n} = (\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta)$. $E(\theta,\phi)$ in equation (2) is the total energy of the FM nanoparticle which includes the magnetocrystalline anisotropy energy and the Zeeman energy when a magnetic field is applied.

It is noted that the Euclidean action is written in the north-pole gauge, and the first two terms in equation (2) define the Wess-Zumino or Berry term which arises from the nonorthogonality of spin coherent states. The Wess-Zumino term has a simple geometrical or 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 gives the boundary contribution to the Euclidean action. However, it has been theoretically demonstrated that this term, known as the topological term, is crucial for the quantum interference effect and makes the tunneling behaviors of integer and half-integer total spins strikingly different [22,23].

In the semiclassical limit, the dominant contribution to the Euclidean transition amplitude comes from finite action solutions of the classical equation of motion (instantons). The classical equation of motion for $\mathbf{M}$ is given by ($\delta S_E = 0$)

$$i\frac{d\mathbf{M}}{d\tau} = -\gamma\mathbf{M}\times\frac{dE(\mathbf{M})}{d\mathbf{M}}, \tag{3}$$

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

$$\begin{aligned} i\left(\frac{d\bar{\theta}}{d\tau}\right)\sin\bar{\theta} &= \frac{\gamma}{M_0}\frac{\partial E}{\partial\phi}, \\ i\left(\frac{d\bar{\phi}}{d\tau}\right)\sin\bar{\theta} &= -\frac{\gamma}{M_0}\frac{\partial E}{\partial\theta}, \end{aligned} \tag{4}$$

where $\bar{\theta}$ and $\bar{\phi}$ denote the classical path. Note that the Euclidean action in equation (2) describes the $(1\oplus 1)$-dimensional dynamics in the Hamiltonian formulation with canonical variables ϕ and $p_\phi = S\left(1-\cos\theta\right)$.

According to the standard instanton method in spin-coherent-state path integral, the instanton's contribution to the tunneling rate Γ for MQT or the tunneling splitting Δ for MQC (not including the phase factor generated by the topological term in the Euclidean action) is given by [2]

$$\Gamma(\text{or }\ \Delta) = A\omega_p(S_{cl}/2\pi)^{1/2}e^{-S_{cl}}, \tag{5}$$

where ω_p is the oscillation frequency in one of the wells separated by the magnetocrystalline anisotropies and external magnetic fields, and S_{cl} is the classical action or the WKB exponent which minimizes the Euclidean action in equation (2). The preexponential 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 [2]. In reference [2], Garg and Kim have presented the general formalism for calculating both the exponent and the preexponential factors in the WKB tunneling rate for the single-domain FM nanoparticles. In Appendix A of the present work, we explain briefly the basic idea of this calculation, and then apply this approach to calculate the tunneling level splitting for the FM system with trigonal crystal symmetry (in Sect. 3) in detail.

3 The MQC for trigonal symmetry

In this section, we investigate the tunneling behaviors of the magnetization vector in a FM system with trigonal crystal symmetry, *i.e.*, threefold rotational symmetry around the $\mathbf{z}$ axis and reflection symmetry in the $x-y$ plane. Then the magnetocrystalline anisotropy energy can be written as [23]

$$E(\theta,\phi) = -K_1\sin^2\theta + [K_2 - K_2'\cos(3\phi)]\sin^3\theta + E_0, \tag{6}$$

where K_1, K_2 and K_2' are the magnetic anisotropy coefficients satisfying $K_1 \gg K_2, K_2' > 0$, and E_0 is a constant which makes $E(\theta,\phi)$ zero at the initial state. The ground state of this system corresponds to the magnetization vector $\mathbf{M}$ pointing in one of the three energetically degenerate easy directions: $\theta = \pi/2$, and $\phi = 0$, $2\pi/3$, $4\pi/3$. If we denote the three states as $|1\rangle$, $|2\rangle$, and $|3\rangle$, other energy minima repeat the three states with period 2π. So the magnetization vector can resonate coherently between the energetically degenerate states.

In the case of very strong anisotropy K_1, the magnetization vector is forced to lie in the $x-y$ plane. So the fluctuations of θ about $\pi/2$ are small. Writing $\theta = \pi/2+\alpha$ ($|\alpha| \ll 1$), and expanding $E(\theta,\phi)$ in equation (6) to second order in α, we obtain

$$E(\alpha,\phi) = K[1-\cos(3\phi)] + \left[K' + \frac{3}{2}K\cos(3\phi)\right]\alpha^2, \tag{7}$$

where $K = K_2'$, $K' = K_1 - (3/2)K_2$, and $K' \gg K > 0$.

Substituting equation (7) into the classical equations of motion, we obtain the instanton solution:

$$\overline{\alpha} = -i\sqrt{\frac{4K}{2K'-3K}}\frac{1}{\cosh(\omega_0\tau)},$$
$$\sin^2\left(\frac{3}{2}\overline{\phi}\right) = \frac{1-\tanh^2(\omega_0\tau)}{1-\lambda\tanh^2(\omega_0\tau)}, \tag{8}$$

which corresponds to the variation of ϕ from $\phi = 0$ at $\tau = -\infty$ to $\phi = 2\pi/3$ at $\tau = +\infty$. ω_0 and λ are defined as $\omega_0 = 3(\gamma/M_0)\sqrt{K(3K+2K')}$ and $\lambda = 6K/(2K'+3K)$. Using the standard instanton method (for detailed calculation see Appendix A), we obtain the contribution to the tunneling splitting (not including the phase factor generated by the topological term in the Euclidean action) for the instanton starting from $|1\rangle$ and ending at $|2\rangle$ as the following expression,

$$\hbar\Delta = \frac{2^{9/4}3^{1/2}}{\pi^{1/2}}(VK')\left(\frac{K}{K'}\right)^{3/4}S^{-1/2}e^{-S_{cl}}, \tag{9}$$

where

$$S_{cl} = \frac{2^{5/2}}{3}\sqrt{\frac{K}{K'}}S. \tag{10}$$

$S = M_0V/\hbar\gamma$ is the total spin of the single-domain FM nanoparticle. Note that both the WKB exponent and the preexponential factors are obtained exactly in equations (9, 10).

Including the phase factors generated by the topological term in the Euclidean action, the Euclidean transition amplitude can be expressed as the following equation with the help of the dilute instanton-gas approximation [28]

$$\langle j'|e^{-HT}|j\rangle = \sqrt{\frac{\omega_0}{\pi\hbar}}e^{-\omega_0T/2} \times \sum_{m,n}^{m-n=j-j'(\mathrm{mod}\,3)} \frac{(\hbar\Delta Te^{-iS2\pi/3})^m(\hbar\Delta Te^{iS2\pi/3})^n}{m!n!}, \tag{11}$$

where $|j\rangle$ and $|j'\rangle$ denote any two of the three energetically degenerate easy directions.

The propagators from $|1\rangle$ to the other states are found to be as follows:

$$\begin{aligned}
\langle 1|e^{-HT}|1\rangle = \frac{1}{3}\sqrt{\frac{\omega_0}{\pi\hbar}}e^{-\omega_0T/2}\Bigg\{&\exp\left[2\hbar\Delta T\cos\left(\frac{2S\pi}{3}\right)\right] \\
&+2\exp\left[-\hbar\Delta T\cos\left(\frac{2S\pi}{3}\right)\right] \\
&\times\cosh\left[\sqrt{3}\hbar\Delta T\sin\left(\frac{2S\pi}{3}\right)\right]\Bigg\}, \\
\langle 2|e^{-HT}|1\rangle = \frac{1}{3}\sqrt{\frac{\omega_0}{\pi\hbar}}e^{-\omega_0T/2}\Bigg\{&\exp\left[2\hbar\Delta T\cos\left(\frac{2S\pi}{3}\right)\right] \\
&-\exp\left[-\hbar\Delta T\cos\left(\frac{2S\pi}{3}\right)\right] \\
&\times\left[\cosh\left(\sqrt{3}\hbar\Delta T\sin\left(\frac{2S\pi}{3}\right)\right)\right. \\
&\left.+i\sqrt{3}\sinh\left(\sqrt{3}\hbar\Delta T\sin\left(\frac{2S\pi}{3}\right)\right)\right]\Bigg\}, \\
\langle 3|e^{-HT}|1\rangle = \frac{1}{3}\sqrt{\frac{\omega_0}{\pi\hbar}}e^{-\omega_0T/2}& \\
\times\Bigg\{&\exp\left[2\hbar\Delta T\cos\left(\frac{2S\pi}{3}\right)\right] \\
&-\exp\left[-\hbar\Delta T\cos\left(\frac{2S\pi}{3}\right)\right] \\
&\times\left[\cosh\left(\sqrt{3}\hbar\Delta T\sin\left(\frac{2S\pi}{3}\right)\right)\right. \\
&\left.-i\sqrt{3}\sinh\left(\sqrt{3}\hbar\Delta T\sin\left(\frac{2S\pi}{3}\right)\right)\right]\Bigg\}.
\end{aligned} \tag{12}$$

Now we use the effective Hamiltonian method [29,30] to evaluate the splittings of the ground state due to the resonant quantum tunneling of the magnetization vector between energetically degenerate states. This approach is a development of the tunneling Hamiltonian of Leggett *et al.* [29], where the phase factor generated by the topological term in the Euclidean action is properly incorporated. The effective Hamiltonian approach is shown to be equivalent to the dilute instanton-gas approximation [28] (see Appendix B), while this approach has the advantage of being very simple and direct. It permits us to obtain the ground-state tunneling level splittings conveniently and discuss the degeneracies of the low-lying tunneling levels in detail.

The effective Hamiltonian of the system can be written as:

$$H_{eff} = -\hbar\Delta M, \tag{13}$$

where M is a linear operator defined by

$$M|j\rangle = p|j+1\rangle + q|j-1\rangle. \tag{14}$$

The above equation can be viewed as by one step $|j\rangle$ goes to $|j+1\rangle$ forward with weight p, and to $|j-1\rangle$ backward with weight q. For the trigonal crystal symmetry, the matrix form of M is found to be

$$[M] = \langle j'|M|j\rangle = \begin{bmatrix} 0 & q & p \\ p & 0 & q \\ q & p & 0 \end{bmatrix}. \tag{15}$$

H_{eff} is Hermitian if $p = q^*$ and can be diagonalized. In fact they should be specified by

$$p = q^* = e^{-iS2\pi/3}. \tag{16}$$

A simple diagonalization of H_{eff} shows that the eigenvalues of this system depend on the parity of the total spin S of the FM nanoparticle. If S is an integer, the energies are $\hbar\Delta$ and $-2\hbar\Delta$, the former being doubly degenerate. If S is a half-integer, the energies are $2\hbar\Delta$ and $-\hbar\Delta$, the latter being doubly degenerate. The energy level spectrum (corresponding to the ground-state tunneling level splittings due to the resonance of the magnetization vector between energetically degenerate easy directions) for the half-integer total spin is significantly different from that for the integer total spin. This spin-parity effect is the result of quantum interference between topologically different tunneling paths.

4 The MQC for hexagonal symmetry

In this section, we consider MQC of the magnetization vector in a FM system with hexagonal crystal symmetry, which has six energetically degenerate easy axes in the basal plane. Now the magnetocrystalline anisotropy energy is [2]

$$\begin{aligned} E(\theta,\phi) = &-K_1\sin^2\theta + K_2\sin^4\theta \\ &+ [K_3 - K_3'\cos(6\phi)]\sin^6\theta + E_0, \end{aligned} \tag{17}$$

where K_1, K_2, K_3 and K_3' are the magnetic anisotropy coefficients satisfying the condition that $K_1 \gg K_2, K_3, K_3' > 0$, and E_0 is a constant which makes $E(\theta,\phi)$ zero at the initial state. The easy directions of this system are at $\theta = \pi/2$, and $\phi = 0$, $\pi/3$, $2\pi/3$, π, $4\pi/3$, $5\pi/3$. We denote the six states as $|1\rangle$, $|2\rangle$, $|3\rangle$, $|4\rangle$, $|5\rangle$, and $|6\rangle$, other energy minima repeat the six states with period 2π.

As $K_1 \gg K_2, K_3, K_3' > 0$, the magnetization vector is forced to lie in the $x-y$ plane. So the fluctuations of θ about $\pi/2$ are small. Introducing $\theta = \pi/2 + \alpha$ ($|\alpha| \ll 1$), $E(\theta,\phi)$ reduces to

$$E(\alpha,\phi) = K[1-\cos(6\phi)] + [K' + 3K\cos(6\phi)]\,\alpha^2, \tag{18}$$

where $K = K_3'$, $K' = K_1 - 2K_2 - 3K_3$, and $K' \gg K > 0$.

Substituting equation (18) into the classical equations of motion, we obtain the instanton solution mapping from $|1\rangle$ to $|2\rangle$ as

$$\begin{aligned} \overline{\alpha} &= -i\sqrt{\frac{2K}{2K'-3K}}\frac{1}{\cosh(\omega_0\tau)}, \\ \sin^2\left(3\overline{\phi}\right) &= \frac{1-\tanh^2(\omega_0\tau)}{1-\lambda\tanh^2(\omega_0\tau)}, \end{aligned} \tag{19}$$

where $\omega_0 = 6(\gamma/M_0)\sqrt{2K(3K+K')}$ and $\lambda = 6K/(K'+3K)$. The associated instanton's contribution to the tunneling splitting is then found to be

$$\hbar\Delta = \frac{2^{11/4}3^{1/2}}{\pi^{1/2}}(VK')\left(\frac{K}{K'}\right)^{3/4}S^{-1/2}e^{-S_{cl}}, \tag{20}$$

where

$$S_{cl} = \frac{2^{3/2}}{3}\sqrt{\frac{K}{K'}}S. \tag{21}$$

By using the dilute instanton-gas approximation, the Euclidean transition amplitude is found to be

$$\begin{aligned} \langle j'|e^{-HT}|j\rangle = &\sqrt{\frac{\omega_0}{\pi\hbar}}e^{-\omega_0T/2} \\ &\times \sum_{m,n}^{m-n=j-j'(\mathrm{mod}\,6)} \frac{(\hbar\Delta Te^{-iS\pi/3})^m(\hbar\Delta Te^{iS\pi/3})^n}{m!n!}, \end{aligned} \tag{22}$$

where $|j\rangle$ and $|j'\rangle$ are any two of the six energetically degenerate easy directions, and the phase factors generated by the topological term in the Euclidean action have been properly included.

After some complicated calculations, we obtain the propagators from $|1\rangle$ to the other states as the following equations,

$$\langle 1|e^{-HT}|1\rangle = \frac{1}{3}\sqrt{\frac{\omega_0}{\pi\hbar}}e^{-\omega_0 T/2}\left\{\cosh\left[2\hbar\Delta T\cos\left(\frac{S\pi}{3}\right)\right] + 2\cosh\left[\hbar\Delta T\cos\left(\frac{S\pi}{3}\right)\right]\cosh\left[\sqrt{3}\hbar\Delta T\sin\left(\frac{S\pi}{3}\right)\right]\right\},$$

$$\langle 2|e^{-HT}|1\rangle = \frac{1}{3}\sqrt{\frac{\omega_0}{\pi\hbar}}e^{-\omega_0 T/2}\left\{\sinh\left[2\hbar\Delta T\cos\left(\frac{S\pi}{3}\right)\right] + \sinh\left[\hbar\Delta T\cos\left(\frac{S\pi}{3}\right)\right]\cosh\left[\sqrt{3}\hbar\Delta T\sin\left(\frac{S\pi}{3}\right)\right] - i\sqrt{3}\cosh\left[\hbar\Delta T\cos\left(\frac{S\pi}{3}\right)\right]\sinh\left[\sqrt{3}\hbar\Delta T\sin\left(\frac{S\pi}{3}\right)\right]\right\},$$

$$\langle 3|e^{-HT}|1\rangle = \frac{1}{3}\sqrt{\frac{\omega_0}{\pi\hbar}}e^{-\omega_0 T/2}\left\{\cosh\left[2\hbar\Delta T\cos\left(\frac{S\pi}{3}\right)\right] - \cosh\left[\hbar\Delta T\cos\left(\frac{S\pi}{3}\right)\right]\cosh\left[\sqrt{3}\hbar\Delta T\sin\left(\frac{S\pi}{3}\right)\right] - i\sqrt{3}\sinh\left[\hbar\Delta T\cos\left(\frac{S\pi}{3}\right)\right]\sinh\left[\sqrt{3}\hbar\Delta T\sin\left(\frac{S\pi}{3}\right)\right]\right\},$$

$$\langle 4|e^{-HT}|1\rangle = \frac{1}{3}\sqrt{\frac{\omega_0}{\pi\hbar}}e^{-\omega_0 T/2}\left\{\sinh\left[2\hbar\Delta T\cos\left(\frac{S\pi}{3}\right)\right] - 2\sinh\left[\hbar\Delta T\cos\left(\frac{S\pi}{3}\right)\right]\cosh\left[\sqrt{3}\hbar\Delta T\sin\left(\frac{S\pi}{3}\right)\right]\right\},$$

$$\langle 5|e^{-HT}|1\rangle = \frac{1}{3}\sqrt{\frac{\omega_0}{\pi\hbar}}e^{-\omega_0 T/2}\left\{\cosh\left[2\hbar\Delta T\cos\left(\frac{S\pi}{3}\right)\right] - \cosh\left[\hbar\Delta T\cos\left(\frac{S\pi}{3}\right)\right]\cosh\left[\sqrt{3}\hbar\Delta T\sin\left(\frac{S\pi}{3}\right)\right] + i\sqrt{3}\sinh\left[\hbar\Delta T\cos\left(\frac{S\pi}{3}\right)\right]\sinh\left[\sqrt{3}\hbar\Delta T\sin\left(\frac{S\pi}{3}\right)\right]\right\},$$

$$\langle 6|e^{-HT}|1\rangle = \frac{1}{3}\sqrt{\frac{\omega_0}{\pi\hbar}}e^{-\omega_0 T/2}\left\{\sinh\left[2\hbar\Delta T\cos\left(\frac{S\pi}{3}\right)\right] + \sinh\left[\hbar\Delta T\cos\left(\frac{S\pi}{3}\right)\right]\cosh\left[\sqrt{3}\hbar\Delta T\sin\left(\frac{S\pi}{3}\right)\right] + i\sqrt{3}\cosh\left[\hbar\Delta T\cos\left(\frac{S\pi}{3}\right)\right]\sinh\left[\sqrt{3}\hbar\Delta T\sin\left(\frac{S\pi}{3}\right)\right]\right\}. \tag{23}$$

It is easy to show that $\langle 4|e^{-HT}|1\rangle$ (*i.e.* $\langle\theta = \pi/2, \phi = \pi|e^{-HT}|\theta = \pi/2, \phi = 0\rangle$) vanishes when S is a half-integer, indicating degeneracy of the states. This suppression turns out to be in good agreement with the Kramers' theorem, which demands that a state with its time-reversed counterpart should be degenerate for the half-integer total spin if the Hamiltonian has time-reversal invariance. This is, however, not the only effect the quantum interference results in. In fact, we present the strikingly different ground-state tunneling level splittings for the integer and half-integer total spin FM nanoparticles in the following.

We now apply the effective Hamiltonian approach to obtain the ground-state tunneling level splittings for this system. For the hexagonal crystal symmetry, the matrix form of M in equation (13) is found to be

$$[M] = \langle j'|M|j\rangle = \begin{bmatrix} 0 & q & 0 & 0 & 0 & p \\ p & 0 & q & 0 & 0 & 0 \\ 0 & p & 0 & q & 0 & 0 \\ 0 & 0 & p & 0 & q & 0 \\ 0 & 0 & 0 & p & 0 & q \\ q & 0 & 0 & 0 & p & 0 \end{bmatrix}, \tag{24}$$

where

$$p = q^* = e^{-iS\pi/3}. \tag{25}$$

Then the energy level spectrum of this system is found to depend on the parity of the total spin of the single-domain FM nanoparticle. If S is a half-integer, the energies are $\sqrt{3}\hbar\Delta$, 0 and $-\sqrt{3}\hbar\Delta$, all the three levels being doubly degenerate. If S is an integer, the energies are $\pm\ 2\hbar\Delta$ and $\pm\ \hbar\Delta$, the latter two levels being doubly degenerate. It is clearly shown that the ground-state tunneling level splittings for the half-integer total spin FM particle are much different form that for the integer total spin one.

5 Conclusions and discussions

In summary, we have investigated the tunneling behaviors in macroscopic quantum coherence of the magnetization vector in single-domain FM nanoparticles based on the standard instanton method in spin-coherent-state path integral. We consider the magnetocrystalline anisotropy with the trigonal crystal symmetry and that with the hexagonal crystal symmetry, which have three and six energetically degenerate easy directions in the basal plane respectively. Both the WKB exponents and the preexponential factors (originated from the small quantum fluctuations about the classical paths) are found exactly for one instanton's contribution to the tunneling splitting. The Euclidean transition amplitudes between any two of the energetically degenerate easy directions are obtained with the help of the dilute instanton-gas approximation. And the final results of the ground-state tunneling level splittings are clearly shown for each kind of symmetry by applying the effective Hamiltonian approach. This Hamiltonian approach is shown to be equivalent to the dilute instanton-gas approximation, which permits us to discuss the tunneling level spectrum conveniently.

One important conclusion is that for both the trigonal and hexagonal crystal symmetries, the ground-state tunneling level splittings for the half-integer total spin FM nanoparticle are significantly different from that for the integer total spin one. For the FM system with biaxial crystal symmetry, which has two energetically degenerate easy directions in the basal plane, it has been theoretically demonstrated that the ground-state tunneling level splitting is suppressed to zero for the half-integer total spin FM nanoparticle [22,23]. However, the tunneling level spectrum for the trigonal or hexagonal symmetry is found to be much more complex than that for the biaxial symmetry. The ground-state tunneling level splittings can be nonzero for the trigonal or hexagonal crystal symmetry even if the total spin of the FM particle is a half-integer. Note that these spin-parity effects are of topological origin and thus independent of the magnitude of the total spin of the single-domain FM nanoparticle.

At the end of this paper, we discuss the possible relevance to the experimental test for the spin-parity effect in single-domain FM nanoparticles. Recently, there has been a focus of renewed interest on studying the quantum tunneling of magnetization in molecular magnets, such as the crystal Mn_{12} acetate ($Mn_{12}Ac$) which has the chemical formula $[Mn_{12}O_{12}(CH_3COO)_{16}(H_2O)_4]\cdot 2CH_3COOH\cdot 4H_2O$ [21,31–37]. Experiments involving magnetization relaxation [31], dynamic susceptibility measurement [32] and hysteresis loop study [21,33–36] indicate that thermally assisted, field-tuned resonant magnetization tunneling takes place between quantum spin states in a large number of identical $Mn_{12}Ac$ molecules. The $Mn_{12}Ac$ molecule contains 12 Mn ions which are strongly bound ferrimagnetically *via* the superexchange through oxygen bridges. The ground state of this molecule has a net spin of $S = 10$ at low temperatures. The magnetic interactions between the spins of different molecules can be negligible since the distance between Mn ions in neighboring molecules is at least 7 Å. Therefore, the $Mn_{12}Ac$ molecule can be viewed as an ensemble of the magnetic tunneling states, in close analogy with the two-level system in amorphous materials. It is then of interest to study the contribution of these magnetic tunneling states to thermodynamic quantities (such as the specific heat) of the system. Here we shall show that the contribution to the specific heat for the integer total spin FM particle is significantly different from that for the half-integer total spin FM particle at low temperatures.

Since we have already obtained the low-lying tunneling level spectrum, the partition function of the tunneling states can be expressed as the following equation for the FM nanoparticle with trigonal crystal symmetry,

$$Z = Tr(e^{-\beta H}) = 2e^{-\hbar\Delta\beta} + e^{2\hbar\Delta\beta}, \text{ if } S \text{ is an integer}, \quad (26)$$

and

$$Z = 2e^{\hbar\Delta\beta} + e^{-2\hbar\Delta\beta}, \text{ if } S \text{ is a half-integer}, \quad (27)$$

where $\hbar\Delta$ has been clearly shown in equations (9, 10), and $\beta = 1/k_B T$ with k_B the Boltzmann constant. Then the specific heat is given by

$$c = -T\frac{\partial^2 F}{\partial T^2}, \quad (28)$$

where

$$F = -k_B T \ln Z. \quad (29)$$

For the trigonal crystal symmetry, we obtain the specific heat after some algebra,

$$c = \frac{18\hbar^2\Delta^2}{k_B T^2}\frac{e^{\hbar\Delta\beta}}{\left(e^{2\hbar\Delta\beta} + 2e^{-\hbar\Delta\beta}\right)^2}, \text{ if } S \text{ is an integer}, \quad (30)$$

while

$$c = \frac{18\hbar^2\Delta^2}{k_B T^2}\frac{e^{-\hbar\Delta\beta}}{\left(e^{-2\hbar\Delta\beta} + 2e^{\hbar\Delta\beta}\right)^2}, \text{ if } S \text{ is a half-integer}. \quad (31)$$

It is clearly shown that the specific heat for the integer total spin FM nanoparticle is much different from that for the half-integer total spin one.

Using the similar method, we obtain the specific heat for the single-domain FM nanoparticle with hexagonal crystal symmetry as the following equations,

$$c = \frac{2\hbar^2\Delta^2}{k_B T^2} \times \frac{\left[4 + 5\cosh(2\hbar\Delta\beta)\cosh(\hbar\Delta\beta) - 4\sinh(2\hbar\Delta\beta)\sinh(\hbar\Delta\beta)\right]}{\left[\cosh(2\hbar\Delta\beta) + 2\cosh(\hbar\Delta\beta)\right]^2}, \quad (32)$$

for the integer total spin FM particle, while

$$c = \frac{3\hbar^2\Delta^2}{k_B T^2}\frac{1}{\cosh^2\left(\sqrt{3}\hbar\Delta\beta\right)}, \quad (33)$$

for the half-integer total spin one, where $\hbar\Delta$ has been shown in equations (20, 21).

In brief, the heat capacity of the magnetic tunneling states is found to depend on the parity of the total spin of the FM nanoparticle, providing a possible experimental method to examine the theoretical results on the spin-parity effect. Our results may be useful in the analysis of further experiments on macroscopic quantum coherence of the magnetization vector in single-domain FM nanoparticles.

The effects of the environment caused by phonons [1,38], nuclear spins [39], Stoner excitations and eddy currents in metallic magnets [11] are crucial in macroscopic quantum coherence of magnetism. And the most important effect is the interaction between the spins of the magnetic nanoparticle and the spins of the environment, since the change of a single 1/2 spin of the system might change the total tunneling picture completely. Whether the quantum interference effect can be observed experimentally is still an interesting problem deserving further investigation. We hope that the theoretical results presented in this paper will stimulate more experiments to observe the quantum interference or spin-parity effect in nanoscale single-domain ferromagnets.

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Appendix A: Evaluation of the preexponential factors

In this appendix we review briefly the procedure to calculate the preexponential factors in the WKB tunneling rate due to the small fluctuations about the classical path, and then apply this procedure to the FM system with trigonal crystal symmetry (considered in Sect. 3).

In reference [2], Garg and Kim have presented general formulas for calculating both the WKB exponent and the preexponential factors in the tunneling rate (MQT) or the tunneling splitting (MQC) for the single-domain FM nanoparticles, without assuming a specific form of the magnetocrystalline anisotropy and 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, which gives the WKB exponent. 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. Writing $\theta(\tau) = \overline{\theta} + \theta_1$ and $\phi(\tau) = \overline{\phi} + \phi_1$, where $\overline{\theta}$ and $\overline{\phi}$ denote the classical path, one obtains the Euclidean action as $S_E[\theta(\tau), \phi(\tau)] \approx S_{cl} + \delta^2 S$ with $\delta^2 S$ being a functional of θ_1 and ϕ_1. Under the assumption that $E_{\theta\theta} - \cot\overline{\theta} E_\theta > 0$, where $E_\theta = \partial E/\partial\theta$ and $E_{\theta\theta} = \partial^2 E/\partial\theta^2$, the Gaussian integration can be performed over θ_1, and the remaining ϕ_1 path integral is casted into the standard form for one-dimensional motion. As usual there exists a zero mode, $d\overline{\phi}/d\tau$, corresponding to a translation of the center of the instanton, and a negative eigenvalue in the MQT problem. This leads to the imaginary part of the energy, which corresponds to the quantum tunneling escape rate from the metastable states of the system. And the resonant tunneling splittings of the ground state in the MQC problem can be evaluated by using the similar method [2]. What is need for the calculation of the tunneling rate (MQT) or the tunneling splitting (MQC) is the asymptotic relation of the zero mode, $d\overline{\phi}/d\tau$, for large τ

$$d\overline{\phi}/d\tau \approx a e^{-\mu\zeta}, \text{as } \zeta \to \infty. \tag{A.1}$$

The new time variable ζ in equation (A.1) is related to τ as

$$d\zeta = d\tau / 2A\left(\overline{\theta}(\tau), \overline{\phi}(\tau)\right), \tag{A.2}$$

where

$$A\left(\overline{\theta}, \overline{\phi}\right) = \hbar S^2 \sin^2\overline{\theta} / 2V\left(E_{\theta\theta} - \cot\overline{\theta} E_\theta\right). \tag{A.3}$$

The partial derivatives are evaluated at the classical path. Then one instanton's contribution to the tunneling rate for MQT or the tunneling splitting for MQC (not including the phase factor generated by the topological term in the Euclidean action) is given by [2]

$$|a| (\mu/\pi)^{1/2} e^{-S_{cl}}. \tag{A.4}$$

Therefore, all that is necessary is to differentiate the classical path (instanton) to get $d\overline{\phi}/d\tau$, then convert from τ to ζ according to equations (A.2, A.3), and read off a and μ by comparison with equation (A.1). If the condition $E_{\theta\theta} - \cot\overline{\theta} E_\theta > 0$ is not satisfied, one can always perform the Gaussian integration over θ_1 and end up with a one-dimensional path integral over ϕ_1.

For the FM system with trigonal crystal symmetry (in Sect. 3), we find that

$$E_{\theta\theta} - \cot\overline{\theta} E_\theta \approx 2K' + O(K) > 0. \tag{A.5}$$

So θ_1 can be integrated out. It is easy to show that as $\zeta \to \infty$,

$$\frac{d\overline{\phi}}{d\tau} = 2^{5/2} \frac{V}{\hbar S} \sqrt{KK'} \exp\left(-\frac{3}{\sqrt{2}} \sqrt{\frac{K}{K'}} S\zeta\right), \tag{A.6}$$

where $K = K_2'$, $K' = K_1 - (3/2) K_2$, and $K' \gg K > 0$. Thus, $|a| = 2^{5/2} (V/\hbar S) \sqrt{KK'}$, and $\mu = \left(3/\sqrt{2}\right) \sqrt{K/K'} S$. Substituting in the general formula (A.4), and using equation (10) for the classical action, we obtain one-instanton's contribution to the tunneling level splitting $\hbar\Delta$ as expressed in equation (9).

The calculation of the tunneling splitting for the FM system with hexagonal crystal symmetry (in Sect. 4) can be performed by using the similar method, and we will not discuss it any further.

Appendix B: The equivalence of the effective Hamiltonian method with the dilute instanton-gas approximation

In this appendix, we show that the effective Hamiltonian approach is equivalent to the dilute instanton-gas approximation for the FM systems with trigonal and hexagonal crystal symmetries, respectively.

For the trigonal crystal symmetry, the eigenstates of the effective Hamiltonian (Eqs. (13, 15, 16)) are found to be

$$\begin{aligned}
|1) &= \frac{1}{\sqrt{3}} \left(|1\rangle + |2\rangle + |3\rangle\right), \\
|2) &= \frac{1}{\sqrt{3}} \left(|1\rangle - e^{-i\pi/3} |2\rangle + e^{i2\pi/3} |3\rangle\right), \\
|3) &= \frac{1}{\sqrt{3}} \left(|1\rangle - e^{i\pi/3} |2\rangle + e^{-i2\pi/3} |3\rangle\right),
\end{aligned} \tag{B.1}$$

with corresponding eigenvalues

$$\begin{aligned}
E = -2\hbar\Delta \cos\left(\frac{2S}{3}\pi\right), 2\hbar\Delta \cos\left(\frac{2S+1}{3}\pi\right), \\
2\hbar\Delta \cos\left(\frac{2S-1}{3}\pi\right),
\end{aligned} \tag{B.2}$$

where $\hbar\Delta$ has been clearly shown in equations (9, 10), and S is the total spin of the single-domain FM nanoparticle. After some algebra, we obtain that

$$\begin{aligned} e^{\mathrm{M}}\left|1\right\rangle &= \frac{1}{\sqrt{3}} e^{\mathrm{M}}\left(|1\rangle + |2\rangle + |3\rangle\right) \\ &= \frac{1}{\sqrt{3}}\Bigg\{\left[e^{(p+q)} + 2e^{-(p+q)/2}\cos\left(\frac{\sqrt{3}}{2}(p-q)\right)\right]|1\rangle \\ &\quad + \Bigg[e^{(p+q)} - e^{-(p+q)/2}\Bigg(\cos\left(\frac{\sqrt{3}}{2}(p-q)\right) \\ &\quad -\sqrt{3}\sin\left(\frac{\sqrt{3}}{2}(p-q)\right)\Bigg)\Bigg]|2\rangle \\ &\quad + \Bigg[e^{(p+q)} - e^{-(p+q)/2}\Bigg(\cos\left(\frac{\sqrt{3}}{2}(p-q)\right) \\ &\quad + \sqrt{3}\sin\left(\frac{\sqrt{3}}{2}(p-q)\right)\Bigg)\Bigg]|3\rangle\Bigg\}, \end{aligned} \tag{B.3}$$

where $p = q^* = e^{-iS2\pi/3}$. Therefore, we obtain the same Euclidean transition amplitudes as shown in equation (12) for the FM nanoparticle with trigonal crystal symmetry.

Now we turn to the FM system with hexagonal crystal symmetry. For this case, the eigenstates of the effective Hamiltonian are found to be

$$\begin{aligned} |1) &= \frac{1}{\sqrt{6}}\left(|1\rangle + |2\rangle + |3\rangle + |4\rangle + |5\rangle + |6\rangle\right), \\ |2) &= \frac{1}{\sqrt{6}}\Big(|1\rangle + e^{i\pi/3}|2\rangle + e^{i2\pi/3}|3\rangle \\ &\quad - |4\rangle + e^{i4\pi/3}|5\rangle + e^{i5\pi/3}|6\rangle\Big), \\ |3) &= \frac{1}{\sqrt{6}}\Big(|1\rangle + e^{-i\pi/3}|2\rangle + e^{-i2\pi/3}|3\rangle \\ &\quad - |4\rangle + e^{-i4\pi/3}|5\rangle + e^{-i5\pi/3}|6\rangle\Big), \\ |4) &= \frac{1}{\sqrt{6}}\left(|1\rangle - |2\rangle + |3\rangle - |4\rangle + |5\rangle - |6\rangle\right), \\ |5) &= \frac{1}{\sqrt{6}}\Big(|1\rangle - e^{i\pi/3}|2\rangle + e^{i2\pi/3}|3\rangle \\ &\quad + |4\rangle + e^{i4\pi/3}|5\rangle - e^{i5\pi/3}|6\rangle\Big), \\ |6) &= \frac{1}{\sqrt{6}}\Big(|1\rangle - e^{-i\pi/3}|2\rangle + e^{-i2\pi/3}|3\rangle \\ &\quad + |4\rangle + e^{-i4\pi/3}|5\rangle - e^{-i5\pi/3}|6\rangle\Big), \end{aligned} \tag{B.4}$$

and the corresponding eigenvalues are

$$\begin{aligned} E = &-2\hbar\Delta\cos\left(\frac{S}{3}\pi\right), 2\hbar\Delta\cos\left(\frac{S+1}{3}\pi\right), \\ &2\hbar\Delta\cos\left(\frac{S-1}{3}\pi\right), 2\hbar\Delta\cos\left(\frac{S}{3}\pi\right), \\ &-2\hbar\Delta\cos\left(\frac{S+1}{3}\pi\right), -2\hbar\Delta\cos\left(\frac{S-1}{3}\pi\right), \end{aligned} \tag{B.5}$$

where $\hbar\Delta$ has been shown in equations (20, 21) for the hexagonal crystal symmetry. Using the similar method, we obtain

$$\begin{aligned} e^{\mathrm{M}}|1\rangle &= \frac{1}{\sqrt{6}} e^{\mathrm{M}}\left(|1\rangle + |2\rangle + |3\rangle + |4\rangle + |5\rangle + |6\rangle\right) \\ &= \frac{1}{\sqrt{6}}\Bigg\{\left[\cosh(p+q) + 2\cosh\left(\frac{1}{2}(p+q)\right)\cos\left(\frac{\sqrt{3}}{2}(p-q)\right)\right]|1\rangle \\ &\quad + \Bigg[\sinh(p+q) + \sinh\left(\frac{1}{2}(p+q)\right)\cos\left(\frac{\sqrt{3}}{2}(p-q)\right) \\ &\quad + \sqrt{3}\cosh\left(\frac{1}{2}(p+q)\right)\sin\left(\frac{\sqrt{3}}{2}(p-q)\right)\Bigg]|2\rangle \\ &\quad + \Bigg[\cosh(p+q) - \cosh\left(\frac{1}{2}(p+q)\right)\cos\left(\frac{\sqrt{3}}{2}(p-q)\right) \\ &\quad + \sqrt{3}\sinh\left(\frac{1}{2}(p+q)\right)\sin\left(\frac{\sqrt{3}}{2}(p-q)\right)\Bigg]|3\rangle \\ &\quad + \left[\sinh(p+q) - 2\sinh\left(\frac{1}{2}(p+q)\right)\cos\left(\frac{\sqrt{3}}{2}(p-q)\right)\right]|4\rangle \\ &\quad + \Bigg[\cosh(p+q) - \cosh\left(\frac{1}{2}(p+q)\right)\cos\left(\frac{\sqrt{3}}{2}(p-q)\right) \\ &\quad - \sqrt{3}\sinh\left(\frac{1}{2}(p+q)\right)\sin\left(\frac{\sqrt{3}}{2}(p-q)\right)\Bigg]|5\rangle \\ &\quad + \Bigg[\sinh(p+q) + \sinh\left(\frac{1}{2}(p+q)\right)\cos\left(\frac{\sqrt{3}}{2}(p-q)\right) \\ &\quad - \sqrt{3}\cosh\left(\frac{1}{2}(p+q)\right)\sin\left(\frac{\sqrt{3}}{2}(p-q)\right)\Bigg]|6\rangle\Bigg\}, \end{aligned} \tag{B.6}$$

where $p = q^* = e^{-iS\pi/3}$. Then the same results are obtained for the Euclidean transition amplitudes as shown in equation (23) for the FM nanoparticle with hexagonal crystal symmetry.

In this appendix, it has been shown that the effective Hamiltonian approach is equivalent to the dilute instanton-gas approximation by calculating the Euclidean transition amplitudes between the energetically degenerate easy directions for the trigonal and hexagonal crystal symmetries. The same results are obtained for each case. However, as shown in Sections 3 and 4, and in this appendix, the effective Hamiltonian approach has the advantage of being very simple and direct. Using which one immediately obtains the eigenvalues and eigenstates, and the degeneracies of the eigenstates can be analyzed in detail.

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