Eur. Phys. J. B **16**, 507–513 (2000)

THE EUROPEAN PHYSICAL JOURNAL B
EDP Sciences

Resonant quantum coherence of magnetization at excited states in nanospin systems with different crystal symmetries

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Received 15 December 1999

Abstract. The quantum interference effects induced by the Wess-Zumino term, or Berry phase are studied theoretically in resonant quantum coherence of the magnetization vector between degenerate states in nanometer-scale single-domain ferromagnets in the absence of an external magnetic field. We consider the magnetocrystalline anisotropy with trigonal, tetragonal and hexagonal crystal symmetry, respectively. By applying the periodic instanton method in the spin-coherent-state path integral, we evaluate the low-lying tunnel splittings between degenerate excited states of neighboring wells. And the low-lying energy level spectrum of mth excited state are obtained with the help of the Bloch theorem in one-dimensional periodic potential. The energy level spectrum and the thermodynamic properties of magnetic tunneling states are found to depend significantly on the total spins of ferromagnets at sufficiently low temperatures. Possible relevance to experiments is also discussed.

PACS. 75.45.+j Macroscopic quantum phenomena in magnetic systems – 75.10.Jm Quantized spin models – 03.65.Bz Foundations, theory of measurement, miscellaneous theories (including Aharonov-Bohm effect, Bell inequalities, Berry's phase)

1 Introduction

In recent years there has been great experimental and theoretical effort to observe and interpret macroscopic quantum tunneling (MQT) and coherence (MQC) in nanometer-scale single-domain magnets [1]. One notable subject is that the topological Berry or Wess-Zumino phase [2,3] can lead to remarkable spin-parity effects. Loss *et al.* [4], and von Delft and Henly [5] showed that the tunnel splitting is suppressed to zero for half-integer total spins in biaxial ferromagnetic (FM) particles due to the destructive phase interfererence between topologically different tunneling paths. However, the phase interference is constructive for integer spins, and hence the splitting is nonzero [4,5]. While spin-parity effects are sometimes be related to Kramers degeneracy [4,5], they typically go beyond the Kramers theorem in a rather unexpected way [6,7]. Barnes *et al.* proposed the auxiliary particle method to study the model for a single large spin subject to the external and anisotropy fields, and discussed the spin-parity effects [8]. Similar effect was found in antiferromagnetic (AFM) particles, where only the integer excess spins can tunnel but not the half-integer ones [11,12]. Recently, topological phase interference effects were investigated extensively in FM and AFM particles in a magnetic field, [6,9,10,13,14] and in the systems with different symmetries [15–17]. Spin tunneling and quantum oscillation at excited states were studied for biaxial FM particles at zero magnetic field [18], and at a field along the hard axis [19]. One recent experiment [20] was performed to measure the tunnel splittings in molecules Fe_8, and a clear oscillation of the splitting as a function of the field along the hard axis was observed, which is a direct evidence of the role of the topological spin phase (Berry phase) in the spin dynamics of these molecules.

It is noted that the previous results of topological phase interference effects were obtained for the tunnel splittings of the ground state in FM particles with different crystal symmetries [16], or for the excited states in FM particles with simple biaxial crystal symmetry [18]. The purpose of this paper is to study the spin-parity effects at excited states for FM particles with a more complex (than biaxial) structure, such as trigonal, tetragonal, and hexagonal symmetry around $\widehat{z}$, which have three, four, and six degenerate easy directions in the basal plane. Integrating out the momentum in the path integral, the spin tunneling problem is mapped onto a particle moving problem in one-dimensional periodic potential $V(\phi)$. By applying the periodic instanton method, we obtain the low-lying tunnel splittings between mth degenerate excited states of neighboring wells. The periodic potential $V(\phi)$ can be regarded as a one-dimensional superlattice. The general translation symmetry results in the energy band structure, and the low-lying energy level spectrum of excited states is obtained by using the Bloch theorem and

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the tight-binding approximation. Our results show that the excited-state tunnel splittings depend significantly on the parity of the total spins. And the structure of energy level spectrum for the trigonal, tetragonal and hexagonal crystal symmetry is found to be much more complex than that for the biaxial crystal symmetry. Another important conclusion is that the spin-parity effects can be reflected in thermodynamic quantities of the low-lying tunneling levels. Thermodynamic property (such as the specific heat) of the magnetic tunneling states is evaluated, and is found to be strongly parity dependent on the total spins, which may provide an experimental test for the topological phase interference effects. And the spin-parity effect is lost at high temperatures.

The remaining part of this paper is organized as follows. In Section 2, we review briefly some basic ideas of MQT and MQC in FM particles, and discuss the fundamentals concerning the computation of excited-level splittings in the double-well-like potential. In Sections 3 and 4, we study the spin tunneling between degenerate excited states in FM particles with the trigonal, tetragonal and hexagonal symmetry. The conclusions are presented in Section 5.

2 Spin tunneling in FM particles

For a spin tunneling problem, the tunnel splitting for MQC or the decay rate for MQT is determined by the imaginary-time transition amplitude from an initial state $|i\rangle$ at $\tau = -T/2$ to a final state $|f\rangle$ at $\tau = T/2$ in the spin-coherent-state representation as [3,31,32]

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

where $\mathrm{D}\Omega = \sin\theta \mathrm{d}\theta \mathrm{d}\phi$. The paths appearing in equation (1) are fixed at the end points $\tau = \pm\, T/2$. For a system with equivalent double wells, we let $|E\rangle_+$ and $|E\rangle_-$ be eigenstates of the same energy E in the right- and left-hand wells, respectively. The small contribution due to quantum tunneling leads to the effect of level splitting ΔE, which removes the asymptotic degeneracy. The corresponding eigenstates of the Hamiltonian separate into odd and even states $|E\rangle_{\mathrm{o}}$ and $|E\rangle_{\mathrm{e}}$ which are superpositions of $|E\rangle_+$, $|E\rangle_-$ such that $|E\rangle_{\mathrm{o}} = \frac{1}{\sqrt{2}}\left(|E\rangle_+ - |E\rangle_-\right)$, and $|E\rangle_{\mathrm{e}} = \frac{1}{\sqrt{2}}\left(|E\rangle_+ + |E\rangle_-\right)$ with eigenvalues $E \pm \Delta E$, respectively. In the limit that $T \to \infty$, one expects that the amplitude for the transition from state $|E\rangle_-$ in the left-hand well to the state $|E\rangle_+$ in the right-hand well in the time interval T as ${}_+\langle E| \, \mathrm{e}^{-HT} \, |E\rangle_- \to \exp\left(-ET\right) \sinh\left(\Delta ET\right)$. Therefore the tunnel splitting ΔE is obtained if the transition amplitude can be calculated. The decay rate Γ from the metastable state for MQT can be evaluated by a similar procedure. The Euclidean action S_E in equation (1) is [3,31,32]

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

where $M_0 = |\mathbf{M}| = \hbar\gamma S/V$, V is the volume of the particle, γ is the gyromagnetic ratio, and S is the total spins. It is noted that the first two terms in equation (2) define the Berry or Wess-Zumino term which has a simple topological interpretation. For a closed path, this term equals $-\mathrm{i}S$ 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 it is crucial for the spin-parity effects [4,5].

In the semiclassical limit, the instanton's contribution to Γ or ΔE (not including the topological Wess-Zumino phase) is given by [22]

$$\Gamma \text{ (or } \ \Delta E) = A\omega_{\mathrm{p}}\left(\frac{S_{\mathrm{cl}}}{2\pi}\right)^{1/2} \mathrm{e}^{-S_{\mathrm{cl}}}, \tag{3}$$

where ω_{p} is the oscillation frequency in the well, S_{cl} is the classical action, and the prefactor A originates from the quantum fluctuations about the classical path. It is noted that equation (3) is based on tunneling at the ground state, and the temperature dependence of the tunneling frequency (*i.e.*, tunneling at excited states) is not taken into account. The instanton technique is suitable only for the evaluation of the tunneling rate at the vacuum level, since the usual (vacuum) instantons satisfy the vacuum boundary conditions. Recently, Liang *et al.* [23] developed new types of pseudoparticle configurations which satisfy periodic boundary condition (*i.e.*, periodic instantons or nonvacuum instantons). For a particle moving in a double-well-like potential $U\left(x\right)$, the WKB method gives the tunnel splitting of excited states at an energy $E > 0$ as [24,25,33]

$$\Delta E = \frac{\omega\left(E\right)}{\pi} \exp\left[-S\left(E\right)\right], \tag{4}$$

with the imaginary-time action is

$$S\left(E\right) = 2\sqrt{2m}\int_{x_1(E)}^{x_2(E)} \mathrm{d}x\sqrt{U\left(x\right) - E}, \tag{5}$$

where $x_{1,2}\left(E\right)$ are the turning points for the particle oscillating in the inverted potential $-U\left(x\right) \omega\left(E\right) = 2\pi/t\left(E\right)$ is the energy-dependent frequency, and $t\left(E\right)$ is the period of the real-time oscillation in the potential well,

$$t\left(E\right) = \sqrt{2m}\int_{x_3(E)}^{x_4(E)} \frac{\mathrm{d}x}{\sqrt{E - U\left(x\right)}}, \tag{6}$$

where $x_{3,4}\left(E\right)$ are the classical turning points for the particle oscillating inside $U\left(x\right)$. The functional-integral and the WKB method showed that for the potentials parabolic near the bottom the result (4) should be multiplied by $\sqrt{\frac{\pi}{e}}\frac{(2n+1)^{n+1/2}}{2^n e^n n!}$ [25,33]. This factor is very close to 1 for all n: 1.075 for $n = 0$, 1.028 for $n = 1$, 1.017 for $n = 2$, etc. Stirling's formula for $n!$ shows that this factor trends to 1 as $n \to \infty$. Therefore, this correction factor, however, does not change much in front of the exponentially small action term in equation (4). Recently, the crossover from quantum to classical behavior and the associated phase transition have been investigated extensively in nanospin systems [25–29] and other systems [30].

3 MQC for trigonal crystal symmetry

In this section, we consider a spin system with trigonal crystal symmetry, *i.e.*, which has three consecutive energy minima in a period. Now the total energy is

$$E(\theta,\phi) = K_1\cos^2\theta - K_2\sin^3\theta\cos(3\phi) + E_0, \tag{7}$$

where K_1 and K_2 are the magnetic anisotropic constants satisfying $K_1 \gg K_2 > 0$, and E_0 is a constant which makes $E(\theta,\phi)$ zero at the initial state. As $K_1 \gg K_2 > 0$, the magnetization vector is forced to lie in the $\theta = \pi/2$ plane, so the fluctuations of θ about $\pi/2$ are small. Introducing $\theta = \pi/2+\alpha$ ($|\alpha| \ll 1$), equation (7) reduces to

$$E(\alpha,\phi) \approx K_1\alpha^2 + 2K_2\sin^2(3\phi/2). \tag{8}$$

The ground state corresponds to the magnetization vector pointing in one of the three degenerate easy directions: $\theta = \pi/2$, and $\phi = 0$, $2\pi/3$, $4\pi/3$, other energy minima repeat the three states with period 2π. Performing the Gaussian integration over α, we can map the spin system onto a particle moving problem in one-dimensional potential well. Now the transition amplitude becomes

$$\begin{aligned} U_{\mathrm{fi}} &= \exp[-\mathrm{i}S(\phi_{\mathrm{f}}-\phi_{\mathrm{i}})]\int \mathrm{d}\phi\exp(-S_E[\phi]),\\ &= \exp[-\mathrm{i}S(\phi_{\mathrm{f}}-\phi_{\mathrm{i}})]\\ &\quad\times\int\mathrm{d}\phi\exp\left\{-\int\mathrm{d}\tau\left[\frac{1}{2}m\left(\frac{\mathrm{d}\phi}{\mathrm{d}\tau}\right)^2+V(\phi)\right]\right\}, \end{aligned} \tag{9}$$

with $m = \hbar S^2/2K_1V$, and $V(\phi) = 2(K_2V/\hbar)\sin^2(3\phi/2)$. It is noted that the total derivative in equation (2), when integrated, gives an additional phase factor to the transition amplitude (9) which depends on the initial and final values of ϕ. For the trigonal symmetry, this phase factor in equation (9) is $\exp(-\mathrm{i}2\pi S/3)$. The potential $V(\phi)$ is periodic with period $2\pi/3$, and there are three minima in the entire region 2π. We may look at $V(\phi)$ as a superlattice with lattice constant $2\pi/3$ and total length 2π, and we can derive the energy spectrum by applying the Bloch theorem and the tight-binding approximation. The translational symmetry is ensured by the possibility of successive 2π extensions.

The periodic instanton configuration ϕ_{p} which minimizes the Euclidean action in equation (9) satisfies the equation of motion

$$\frac{1}{2}m\left(\frac{\mathrm{d}\phi_{\mathrm{p}}}{\mathrm{d}\tau}\right)^2 - V(\phi_{\mathrm{p}}) = -E, \tag{10}$$

where $E > 0$ is a constant of integration, which can be viewed as the classical energy of the pseudoparticle configuration. Then we obtain the kink-solution as

$$\sin^2\left(\frac{3}{2}\phi_{\mathrm{p}}\right) = 1 - k^2\mathrm{sn}^2(\omega_1\tau,k), \tag{11}$$

where $\mathrm{sn}(\omega_1\tau,k)$ is the Jacobian elliptic sine function of modulus k,

$$k^2 = \frac{n_1^2-1}{n_1^2}, \tag{12}$$

with $\omega_1 = 3\sqrt{2}(V/\hbar S)\sqrt{K_1K_2}$, and $n_1 = \sqrt{2K_2V/\hbar E} > 1$. In the low energy limit, *i.e.*, $E \to 0$, $k \to 1$, $\mathrm{sn}(u,1) \to \tanh u$, we have

$$\sin^2\left(\frac{3}{2}\phi_{\mathrm{p}}\right) = \frac{1}{\cosh^2(\omega_1\tau)}, \tag{13}$$

which is exactly the vacuum instanton solution derived in reference [16].

The Euclidean action of the periodic instanton configuration equation (11) over the domain $(-\beta,\beta)$ is found to be

$$S_{\mathrm{p}} = \int_{-\beta}^{\beta}\mathrm{d}\tau\left[\frac{1}{2}m\left(\frac{\mathrm{d}\phi_{\mathrm{p}}}{\mathrm{d}\tau}\right)^2 + V(\phi_{\mathrm{p}})\right] = W + 2E\beta, \tag{14}$$

with

$$W = \frac{2^{5/2}}{3}S\sqrt{\frac{K_2}{K_1}}\left[E(k) - (1-k^2)K(k)\right], \tag{15}$$

where $K(k)$ and $E(k)$ are the complete elliptic integral of the first and second kind, respectively. Now we discuss the low energy limit where E is much less than the barrier height. In this case, $k'^2 = 1-k^2 = \hbar E/2K_2V \ll 1$, so we can perform the expansions of $K(k)$ and $E(k)$ in equation (15) to include terms like k'^2 and $k'^2\ln(4/k')$,

$$\begin{aligned} E(k) &= 1 + \frac{1}{2}\left[\ln\left(\frac{4}{k'}\right) - \frac{1}{2}\right]k'^2 + \cdots,\\ K(k) &= \ln\left(\frac{4}{k'}\right) + \frac{1}{4}\left[\ln\left(\frac{4}{k'}\right) - 1\right]k'^2 + \cdots \end{aligned} \tag{16}$$

With the help of small oscillator approximation for energy near the bottom of the potential well, $E = \varepsilon_m^{\mathrm{tri}} = (m+1/2)\omega_1$, equation (15) is expanded as

$$\begin{aligned} W &= \frac{2^{5/2}}{3}\sqrt{\frac{K_2}{K_1}}S - \left(m+\frac{1}{2}\right)\\ &\quad + \left(m+\frac{1}{2}\right)\ln\left[\frac{3}{2^{9/2}}\sqrt{\frac{K_1}{K_2}}\frac{1}{S}\left(m+\frac{1}{2}\right)\right]\cdot \end{aligned} \tag{17}$$

Then the general formula (4) gives the low-lying energy shift of mth excited states for FM particles with trigonal crystal symmetry at zero magnetic field as

$$\hbar\Delta\varepsilon_m^{\rm tri} = \frac{1}{m!}\frac{2^{\frac{9}{2}\left(m+\frac{1}{2}\right)}}{3^{\left(m-\frac{1}{2}\right)}\pi^{\frac{1}{2}}} \times (K_1 V)\,\lambda^{\frac{1}{2}\left(m+\frac{3}{2}\right)} S^{\left(m-\frac{1}{2}\right)} \exp\left(-\frac{2^{5/2}}{3}\lambda^{1/2}S\right), \quad (18)$$

where $\lambda = K_2/K_1$, and S is the total spin.

It is noted that $\hbar\Delta\varepsilon_m^{\rm tri}$ is only the level shift induced by tunneling between degenerate excited states through a single barrier. The periodic potential $V(\phi)$ can be regarded as a one-dimensional superlattice. The general translation symmetry results in the energy band structure, and the energy spectrum could be determined by the Bloch theorem. It is easy to show that if $\varepsilon_m^{\rm tri}$ are the degenerate eigenvalues of the system with infinitely high barrier, the energy level spectrum is given by the following formula with the help of tight-binding approximation,

$$E_m^{\rm tri} = \varepsilon_m^{\rm tri} - 2\Delta\varepsilon_m^{\rm tri}\cos\left[(S+\xi)\,2\pi/3\right]. \quad (19)$$

The Bloch wave vector ξ can be assumed to take either of the three values -1, 0, 1 in the first Brillouin zone. It is noted that in equation (19) we have included the contribution of topological phase for FM particles with trigonal crystal symmetry (*i.e.*, $2\pi S/3$). The low-lying energy level spectrum, which corresponds to the splittings of mth excited state due to the resonant quantum coherence of the magnetization vector between energetically degenerate states, is found to depend on the parity of total spin of FM particle significantly. If S is an integer, the low-lying energy level spectrum is $\hbar\varepsilon_m^{\rm tri} - 2\hbar\Delta\varepsilon_m^{\rm tri}$, and $\hbar\varepsilon_m^{\rm tri} + \hbar\Delta\varepsilon_m^{\rm tri}$, the latter being doubly degenerate. But if S is a half-integer, the low-lying energy level spectrum is $\hbar\varepsilon_m^{\rm tri} - \hbar\Delta\varepsilon_m^{\rm tri}$, and $\hbar\varepsilon_m^{\rm tri} + 2\hbar\Delta\varepsilon_m^{\rm tri}$, the former being doubly degenerate. This spin-parity effect is the result of phase interference between topologically distinct tunneling path.

At the end of this section, we discuss the possible relevance to the experimental test for spin-parity effects in single-domain FM nanoparticles. First we discuss the thermodynamic behavior of this system at very low temperature $T \sim T_0 = \hbar\Delta\varepsilon_0^{\rm tri}/k_{\rm B}$. For FM particles with trigonal crystal symmetry at such a low temperature, the partition function of the ground state is found to be

$$Z = \exp\left(-\beta\hbar\varepsilon_0^{\rm tri}\right) \times \left[\exp\left(\pm 2\beta\hbar\Delta\varepsilon_0^{\rm tri}\right) + 2\exp\left(\mp\beta\hbar\Delta\varepsilon_0^{\rm tri}\right)\right], \quad (20)$$

where upper sign corresponds to integer spins, lower sign corresponds to half-integer spins, and $\varepsilon_0^{\rm tri} = \omega_1/2$. Then the specific heat is $c = -T\left(\partial^2 F/\partial T^2\right)$, with $F = -k_{\rm B}T\ln Z$. For the low temperature case, the result is

$$c = 18k_{\rm B}\left(\beta\hbar\Delta\varepsilon_0^{\rm tri}\right)^2 \times \frac{\exp\left(\pm\beta\hbar\Delta\varepsilon_0^{\rm tri}\right)}{\left[\exp\left(\pm 2\beta\hbar\Delta\varepsilon_0^{\rm tri}\right) + 2\exp\left(\mp\beta\hbar\Delta\varepsilon_0^{\rm tri}\right)\right]^2}. \quad (21)$$

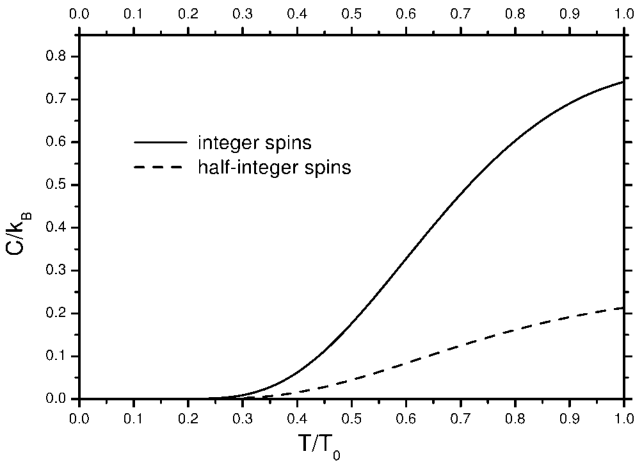

Fig. 1. The temperature dependence of the specific heat for integer and half-integer spins at very low temperature $0 \le T/T_0 \le 1$.

In Figure 1, we plot the temperature dependence of the specific heat for integer and half-integer spins at very low temperature $0 \le T/T_0 \le 1$. It is clearly shown that the specific heat for integer spins is much different from that for half-integer spins at sufficiently low temperatures. When the temperature is higher $\hbar\Delta\varepsilon_0^{\rm tri} \ll k_{\rm B}T < \hbar\omega_1$, the excited energy levels may give contribution to the partition function. Now the partition function is

$$Z \approx Z_0\left[1 + \left(1 - {\rm e}^{-\beta\hbar\omega_1}\right)\left(\beta\hbar\Delta\varepsilon_0^{\rm tri}\right)^2 I_0\left(2q_1{\rm e}^{-\beta\hbar\omega_1/2}\right)\right], \quad (22)$$

for both integer and half-integer spins, where $Z_0 = 3{\rm e}^{-\beta\hbar\omega_1/2}/\left(1-{\rm e}^{-\beta\hbar\omega_1}\right)$ is the partition function in the well calculated for $k_{\rm B}T \ll \Delta U$ over the low-lying oscillatorlike states with $\varepsilon_m^{\rm tri} = (m+1/2)\,\omega_1$, and $\omega_1 = 3\sqrt{2}\,(V/\hbar S)\sqrt{K_1K_2}$. $I_0(x) = \sum_{n=0}(x/2)^{2n}/(n!)^2$ is the modified Bassel function, and $q_1 = \left(2^{9/2}/3\right)\lambda^{1/2}S > 1$. We define a characteristic temperature $\widetilde{T}$ that is solution of equation $q_1{\rm e}^{-\hbar\omega_1/k_{\rm B}\widetilde{T}} = 1$. The temperature $\widetilde{T} = \hbar\omega_1/2\ln q_1$ characterizes the crossover from thermally assisted tunneling to the ground-state tunneling. In Figure 2, we plot the temperature dependence of the specific heat for integer and half-integer spins at high temperature $30 \le T/T_0 \le 60$. The result shows that the spin-parity effect will be lost at high temperatures. The specific heat for integer spins is almost the same as that for half-integer spins.

4 MQC for tetragonal and hexagonal crystal symmetries

In this section, we will apply the method in Section 3 to study spin tunneling in FM particles with tetragonal and

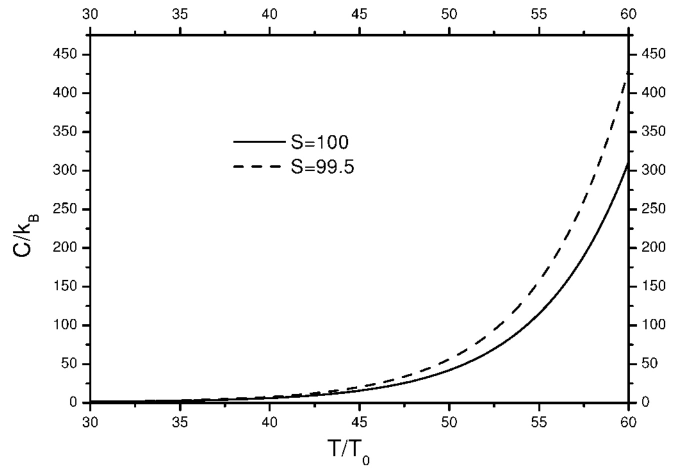

Fig. 2. The temperature dependence of the specific heat for integer and half-integer spins at high temperature $30 \leq T/T_0 \leq 60$. Here $\lambda = 0.001$.

hexagonal crystal symmetry. For the tetragonal symmetry,

$$E(\theta,\phi) = K_1\cos^2\theta + K_2\sin^4\theta - K_2'\sin^4\theta\cos(4\phi) + E_0, \tag{23}$$

where $K_1 \gg K_2,\ K_2' > 0$. The energy minima of this system are at $\theta = \pi/2$, and $\phi = 0,\ \pi/2,\ \pi,\ 3\pi/2$, and other energy minima repeat the four states with period 2π. The problem can be mapped onto a problem of one-dimensional motion by integrating out the fluctuations of θ about $\pi/2$, and for this case $V(\phi) = 2(K_2'V/\hbar)\sin^2(2\phi)$. Now $V(\phi)$ is periodic with period $\pi/2$, and there are four minima in the entire region 2π. The periodic instanton configuration with an energy $E > 0$ is $\sin^2(2\phi_{\rm p}) = 1 - k^2{\rm sn}^2(\omega_2\tau, k)$, where $k = \sqrt{(n_1^2-1)/n_1^2}$, $\omega_2 = 2^{5/2}(V/\hbar S)\sqrt{K_1K_2'}$, and $n_1 = \sqrt{2K_2'V/\hbar E} > 1$. The associated classical action is $S_{\rm p} = W + 2E\beta$, with

$$W = 2^{1/2}S\sqrt{\frac{K_2'}{K_1}}\left[E(k) - \left(1-k^2\right)K(k)\right]. \tag{24}$$

The general formula (4) gives the low-lying energy shift of mth excited state as

$$\hbar\Delta\varepsilon_m^{\rm te} = \frac{1}{m!}\frac{2^{\frac{5}{2}m+\frac{9}{4}}}{\pi^{\frac{1}{2}}} \times (K_1V)\lambda^{\frac{1}{2}\left(m+\frac{3}{2}\right)}S^{\left(m-\frac{1}{2}\right)}\exp\left(-2^{1/2}\lambda^{1/2}S\right), \tag{25}$$

with $\lambda = K_2'/K_1$. The periodic potential $V(\phi)$ can be viewed as a superlattice with lattice constant $\pi/2$ and total length 2π, and the Bloch theorem then gives the energy level spectrum of mth excited state $\varepsilon_m^{\rm te} = (m+1/2)\omega_2$ as $E_m^{\rm te} = \varepsilon_m^{\rm te} - 2\Delta\varepsilon_m^{\rm te}\cos[(S+\xi)\pi/2]$, where $\xi = -1, 0, 1, 2$ in the first Brillouin zone. It is easy to show that the low-lying energy level spectrum is $\hbar\varepsilon_m^{\rm te} \pm 2\hbar\Delta\varepsilon_m^{\rm te}$, and $\hbar\varepsilon_m^{\rm te}$ for integer spins, the latter being doubly degenerate. While the level spectrum is $\hbar\varepsilon_m^{\rm te} \pm \sqrt{2}\hbar\Delta\varepsilon_m^{\rm te}$ with doubly degenerate for half-integer spins. At a very low temperature $T \sim T_0 = \hbar\Delta\varepsilon_0^{\rm te}/k_{\rm B}$, the specific heat is

$$c = 4k_{\rm B}\left(\beta\hbar\Delta\varepsilon_0^{\rm te}\right)^2\frac{1}{1+\cosh\left(2\beta\hbar\Delta\varepsilon_0^{\rm te}\right)}, \tag{26a}$$

for integer spins, while

$$c = 2k_{\rm B}\left(\beta\hbar\Delta\varepsilon_0^{\rm te}\right)^2\frac{1}{\cosh^2\left(\sqrt{2}\beta\hbar\Delta\varepsilon_0^{\rm te}\right)}, \tag{26b}$$

for half-integer spins. The tunneling behavior for integer spins is almost the same as that for half-integer spins at high temperature $\hbar\Delta\varepsilon_0^{\rm te} \ll k_{\rm B}T < \hbar\omega_2$.

For the case of hexagonal symmetry,

$$E(\theta,\phi) = K_1\cos^2\theta + K_2\sin^4\theta + K_3\sin^6\theta - K_3'\sin^6\theta\cos(6\phi) + E_0, \tag{27}$$

where $K_1 \gg K_2, K_3, K_3' > 0$. The easy directions are at $\theta = \pi/2$, and $\phi = 0,\ \pi/3,\ 2\pi/3,\ \pi,\ 4\pi/3,\ 5\pi/3$, and other energy minima repeat the six states with period 2π. For the present case, $V(\phi) = 2(K_3'V/\hbar)\sin^2(3\phi)$ is periodic with period $\pi/3$, and there are six minima in the entire region 2π. The periodic instanton configuration at a given energy $E > 0$ is $\sin^2(3\phi_{\rm p}) = 1 - k^2{\rm sn}^2(\omega_3\tau, k)$, where $k = \sqrt{(n_1^2-1)/n_1^2}$, $\omega_3 = \left(3\times 2^{3/2}\right)(V/\hbar S)\sqrt{K_1K_3'}$, and $n_1 = \sqrt{2K_3'V/\hbar E} > 1$. Correspondingly, the classical action is $S_{\rm p} = W + 2E\beta$, with

$$W = 2^{3/2}S\sqrt{\frac{K_3'}{K_1}}\left[E(k) - \left(1-k^2\right)K(k)\right], \tag{28}$$

and the low-lying energy shift of mth excited state is

$$\hbar\Delta\varepsilon_m^{\rm he} = \frac{1}{m!}\frac{2^{\frac{7}{2}m+\frac{11}{4}}}{3^{m-\frac{1}{2}}\pi^{\frac{1}{2}}} \times (K_1V)\lambda^{\frac{1}{2}\left(m+\frac{3}{2}\right)}S^{\left(m-\frac{1}{2}\right)}\exp\left(-\frac{2^{3/2}}{3}\lambda^{1/2}S\right), \tag{29}$$

with $\lambda = K_3'/K_1$. Now $V(\phi)$ can be regarded as a one-dimensional superlattice with lattice constant $\pi/3$. By applying the Bloch theorem and the tight-binding approximation, we obtain the energy level spectrum of mth excited state $\varepsilon_m^{\rm he} = (m+1/2)\omega_3$ as $E_m^{\rm he} = \varepsilon_m^{\rm he} - 2\Delta\varepsilon_m^{\rm he}\cos[(S+\xi)\pi/3]$, where $\xi = -2,-1,0,1,2,3$. If S is an integer, the low-lying energy level spectrum is $\hbar\varepsilon_m^{\rm he} \pm 2\hbar\Delta\varepsilon_m^{\rm he}$, and $\hbar\varepsilon_m^{\rm he} \pm \hbar\Delta\varepsilon_m^{\rm he}$, the latter two levels being doubly degenerate. If S is a half-integer, the level spectrum is $\hbar\varepsilon_m^{\rm he} \pm \sqrt{3}\hbar\Delta\varepsilon_m^{\rm he}$, and $\hbar\varepsilon_m^{\rm he}$, all three levels being doubly degenerate. Then the specific heat at sufficiently low temperatures is

$$c = 2k_{\rm B}\left(\beta\hbar\Delta\varepsilon_0^{\rm he}\right)^2 \times \frac{\left[4+4\cosh\left(\beta\hbar\Delta\varepsilon_0^{\rm he}\right)+\cosh\left(2\beta\hbar\Delta\varepsilon_0^{\rm he}\right)\cosh\left(\beta\hbar\Delta\varepsilon_0^{\rm he}\right)\right]}{\left[\cosh\left(2\beta\hbar\Delta\varepsilon_0^{\rm he}\right)+2\cosh\left(\beta\hbar\Delta\varepsilon_0^{\rm he}\right)\right]^2}, \tag{30a}$$

for integer spins, while

$$c = 6k_{\mathrm{B}} \left(\beta\hbar\Delta\varepsilon_0^{\mathrm{he}}\right)^2 \frac{2 + \cosh\left(\sqrt{3}\beta\hbar\Delta\varepsilon_0^{\mathrm{he}}\right)}{\left[1 + 2\cosh\left(\sqrt{3}\beta\hbar\Delta\varepsilon_0^{\mathrm{he}}\right)\right]^2}, \quad (30b)$$

for half-integer spins.

In brief, the low-lying energy level spectrum and the heat capacity of the magnetic tunneling states for tetragonal and hexagonal symmetry are found to depend on the parity of the total spins, resulting from the Wess-Zumino phase interference between topologically distinct tunneling paths. And this spin-parity or topological phase interference effect will be lost at high temperatures.

5 Conclusions

In summary, we have investigated the topological phase interference effects in spin tunneling at excited levels for single-domain FM particles with trigonal, tetragonal, and hexagonal crystal symmetries. The low-lying tunnel splittings between mth degenerate excited states of neighboring wells are evaluated with the help of the periodic instanton method, and the energy level spectrum is obtained by applying the Bloch theorem and the tight-binding approximation in one-dimensional periodic potential.

One important conclusion is that for all the three kinds of crystal symmetries, the low-lying energy level spectrum for integer total spins is significantly different from that for half-integer total spins, resulting from the Berry phase interference between topologically distinct tunneling paths. For FM particles with simple biaxial crystal symmetry, which has two degenerate easy directions in the basal plane (*i.e.*, the double-well system), it has been theoretically shown that the tunnel splitting is suppressed to zero for half-integer spins due to the destructive phase interference between topologically different tunneling paths connecting the same initial and final states. However, the structure of low-lying tunneling level spectrum for the trigonal, tetragonal, or hexagonal crystal symmetry is found to be much more complex than that for the biaxial crystal symmetry. The low-lying energy level spectrum can be nonzero even if the total spin is a half-integer for the trigonal, tetragonal, or hexagonal crystal symmetry. Note that these spin-parity effects are of topological origin, and therefore are independent of the magnitude of total spins of FM particles. The heat capacity of low-lying magnetic tunneling states is evaluated and is found to depend significantly on the parity of total spins for FM particles with different crystal symmetries at sufficiently low temperatures, providing a possible experimental method to examine the theoretical results on topological phase interference effects. And the spin-parity effects will be lost at high temperatures. Our results presented here should be useful for a quantitative understanding on the topological phase interference or spin-parity effects in resonant quantum tunneling of magnetization in single-domain FM particles with different crystal symmetries.

More recently, Wernsdorfer and Sessoli [20] have measured the tunnel splittings in the molecular Fe_8 clusters, and have found a clear oscillation of the tunnel splitting with the field along hard axis, which is a direct evidence of the role of the Berry phase in the spin dynamics of these molecules. It is noted that the theoretical results presented in this paper are based on the instanton method, which is semiclassical in nature, *i.e.*, valid for large spins and in the continuum limit. Whether the instanton method can be applied in studying the spin dynamics in molecular clusters with $S = 10$ (such as Fe_8) is an open question.

The theoretical calculations performed in this paper can be extended to the FM and AFM particles in a magnetic field. Work along this line is still in progress. We hope that the theoretical results presented here will stimulate more experiments whose aim is observing the topological phase interference effects in nanometer-scale single-domain magnets.

R.L. and S.P. Kou are grateful to Dr. Y. Zhou, Prof. Z. Xu, Prof. J.-Q. Liang and Prof. F.-C. Pu for stimulating discussions. J.-L. Zhu and R.L. would like to thank Prof. W. Wernsdorfer and Prof. R. Sessoli for providing their paper (Ref. [20]).

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