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Wess–Zumino–Berry phase interference in spin tunnelling at excited levels with a magnetic field

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Abstract. Macroscopic quantum coherence and spin-phase interference between excited levels in single-domain ferromagnetic particles in a magnetic field along the hard-anisotropy axis are studied. The system has a general structure of the magnetocrystalline anisotropy, such as one showing a biaxial, trigonal, tetragonal, or hexagonal symmetry. This study not only yields the previously obtained spin-phase interference results for the ground-state tunnelling, but also provides a generalization of the Kramers degeneracy to coherent spin tunnelling in low-lying excited states. These analytical results are found to be in good agreement with those from numerical diagonalization. We also discuss the transition from quantum to classical behaviour and the possible relevance to experiment.

1. Introduction

Macroscopic quantum phenomena (MQP) in nanoscale magnets have received much attention in recent years both from theorists and from experimentalists [1]. A number of nanoscale samples in different systems have been identified as promising candidates as regards the observation of macroscopic quantum tunnelling (MQT) and coherence (MQC). A perhaps even more interesting finding is that the topological Wess–Zumino term, or Berry phase [2], can lead to remarkable spin-parity effects for some spin systems with high symmetries [1, 3–8]. It has been shown that the ground-state tunnel splitting is completely suppressed to zero for half-integer total spins in biaxial ferromagnetic (FM) particles in the absence of a magnetic field due to the destructive phase interference between topologically different tunnelling paths [3]. However, the phase interference is constructive for integer spins, and hence the splitting is non-zero. The spin-parity effects can be interpreted as Kramers' degeneracy at zero magnetic field, but in the case of a field along the hard-anisotropy axis, these effects are not related to the Kramers theorem since the field breaks the time-reversal symmetry [4, 6]. Experiments on Fe₈ showed that the oscillation of the tunnel splitting as a function of the magnetic field along the hard-anisotropy axis was due to quantum interference of two tunnel paths with opposite windings, which was direct evidence for the topological part of the quantum spin phase (Berry phase) in a magnetic system [7]. Recent theoretical and experimental studies include investigations of the quantum relaxation in magnetic molecules [9], the spin tunnelling in a swept magnetic field [10], the thermally activated resonant tunnelling—with the help of perturbation theory [11] and exact diagonalization [12], the auxiliary-particle method [13], the discrete WKB method and a non-perturbation calculation [14], the non-adiabatic Landau–Zener model [15], calculation based on exact spin-coordinate correspondence [16], and the effects caused by the higher-order term and the nuclear spins on the tunnel splitting of Fe₈ [17].

Up to now, theoretical studies have been focused on spin-phase interference at excited levels in simple biaxial FM particles [8] or at ground states in FM particles with a general structure of the magnetocrystalline anisotropy [7]. However, spin-phase interference between excited-level tunnelling paths is unknown for FM particles with a general structure of the magnetocrystalline anisotropy. The purpose of this paper is to extend the previous results to resonant quantum tunnelling and spin-phase interference at excited levels for single-domain FM particles in the presence of a magnetic field along the hard-anisotropy axis. Moreover, the system considered in this paper shows a general structure of the magnetocrystalline anisotropy, such as one showing a biaxial, trigonal, tetragonal, or hexagonal symmetry around \hat{z} , which has two, three, four, or six degenerate easy directions in the basal plane at zero field. Therefore, our study provides a non-trivial generalization of the Kramers degeneracy to coherent spin tunnelling in ground states as well as low-lying excited states in a magnetic field.

To compute the tunnel splitting, we consider the imaginary-time transition amplitude in the spin-coherent-state path-integral representation. Integrating out the moment in the path integral, the spin-tunnelling problem is mapped onto a moving-particle problem in a one-dimensional periodic potential $U(\phi)$. By applying the periodic instanton method, we obtain the low-lying tunnel splittings of the n th degenerate excited states between neighbouring potential wells. The periodic potential $U(\phi)$ can be regarded as a one-dimensional superlattice. The general translation symmetry results in the energy band structure. By using the Bloch theorem and the tight-binding approximation, we obtain the low-lying energy level spectrum of the excited states. Our results show that the tunnel splittings depend significantly on the parity of the total spins of FM particles. An external magnetic field yields an additional contribution to the Berry phase, resulting in oscillating field dependence of the tunnel splittings for both the integer and half-integer total spins. These analytical results are found to be in good agreement with the exact-diagonalization computation. And the structures of the energy level spectra for the trigonal, tetragonal, and hexagonal symmetries are found to be much more complex than that for biaxial symmetry. The transition from quantum to classical behaviour is also studied and the second-order phase transition is shown. Another important conclusion is that the spin-parity effects can be reflected in thermodynamic quantities for the low-lying tunnelling levels. This may provide an experimental test for the spin-parity or topological phase interference effects in single-domain FM nanoparticles.

2. The physical model

For a spin-tunnelling problem, the tunnel splitting for MQC or the decay rate for MQT is determined by the imaginary-time transition amplitude from an initial state $|i\rangle$ to a final state $|f\rangle$ as follows:

$$\mathcal{K}_E = \langle f | e^{-\mathcal{H}T} | i \rangle = \int \mathcal{D}\Omega \exp(-\mathcal{S}_E) \quad (1)$$

where $\mathcal{D}\Omega = \sin\theta \, d\theta \, d\phi$ is the measure of the path integral. For FM particles at sufficiently low temperature, all the spins are locked together by the strong exchange interaction, and therefore only the orientation of magnetization \mathbf{M} can change and not its absolute value. In the spin-coherent-state representation the Euclidean action \mathcal{S}_E can be written as

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

where $M_0 = |\mathbf{M}| = \hbar\gamma S/V$, S is the total spin, V is the volume of the particle, and γ is the gyromagnetic ratio. The first two terms in equation (2) define the Wess–Zumino term (or Berry phase) which arises from the non-orthogonality of the spin coherent states. The Wess–Zumino

term has a simple topological interpretation. For a closed path, this term equals $-iS$ times the area swept out on the unit sphere between the path and the north pole. The first term in equation (2) is a total imaginary-time derivative, which has no effect on the classical equations of motion, but it is of crucial importance for the spin-parity effects.

It can be noted that for $\delta\mathcal{S}_E = 0$, the action equation (2) reproduces the classical equation of motion whose solution is known as an instanton, and describes the $(1 \oplus 1)$ -dimensional dynamics in the Hamiltonian formulation, which is based on the canonical coordinates ϕ and $P_\phi = S(1 - \cos\theta)$. According to the standard instanton technique in the spin-coherent-state path-integral representation, the tunnelling rate Γ for MQT and the tunnel splitting Δ for MQC are given by [1]

$$\Gamma \text{ (or } \Delta) = A\omega_p \left(\frac{\mathcal{S}_{cl}}{2\pi} \right)^{1/2} e^{-\mathcal{S}_{cl}} \quad (3)$$

where ω_p is the oscillation frequency in the well, \mathcal{S}_{cl} is the classical action, and the prefactor A originates from the fluctuations about the classical path. It can be noted that equation (3) is based on quantum tunnelling at the level of the ground state, and the temperature dependence of the tunnelling frequency (i.e., tunnelling at excited levels) is not taken into account. The instanton technique is suitable only for the evaluation of the tunnelling rate or the tunnel splitting at the vacuum level, since the usual (vacuum) instantons satisfy the vacuum boundary conditions. Recently, different types of pseudoparticle configuration (periodic or non-vacuum instantons) have been found which satisfy periodic boundary conditions [8].

For a particle moving in a double-well-like potential $U(x)$, the WKB approximation gives the tunnel splitting of the n th excited levels as [18]

$$\Delta E_n = \frac{\omega(E_n)}{\pi} \exp[-S(E_n)] \quad (4)$$

and the imaginary-time action is

$$S(E_n) = 2\sqrt{2m} \int_{x_1(E_n)}^{x_2(E_n)} dx \sqrt{U(x) - E_n} \quad (5)$$

where $x_{1,2}(E_n)$ are the turning points for the particle oscillating in the inverted potential $-U(x)$. $\omega(E_n) = 2\pi/t(E_n)$ is the frequency of oscillations at the energy level E_n , and $t(E_n)$ is the period of the real-time oscillation in the potential well:

$$t(E_n) = \sqrt{2m} \int_{x_3(E_n)}^{x_4(E_n)} \frac{dx}{\sqrt{E_n - U(x)}} \quad (6)$$

where $x_{3,4}(E_n)$ are the classical turning points for the particle oscillating inside $U(x)$. The functional-integral and WKB methods show that for the potentials parabolic near the bottom, the result of equation (4) should be multiplied by [19, 20]

$$\sqrt{\frac{\pi}{e}} \frac{(2n+1)^{n+1/2}}{2^n e^n n!}.$$

This factor approaches 1 with increasing n and it 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 tends to 1 as $n \rightarrow \infty$. Therefore, this correction factor does not change much, in front of the exponentially small action term in equation (4).

3. MQC for biaxial symmetry

In this section, we consider an FM system with biaxial symmetry in a magnetic field along the hard-anisotropy axis. The magnetocrystalline anisotropy energy can be written as

$$E(\theta, \phi) = K_1 \cos^2 \theta + K_2 \sin^2 \theta \sin^2 \phi - M_0 H \cos \theta + E_0 \quad (7)$$

where K_1 and K_2 are the longitudinal and the transverse anisotropy coefficients satisfying $K_1 \gg K_2 > 0$. E_0 is a constant which makes $E(\theta, \phi)$ zero for the initial orientation. Although the tunnel splittings and spin-phase interference effects of this system can be easily obtained by direct numerical diagonalization of the Hamiltonian (see references [4] and [7], and in the following), it is of interest to understand these features analytically.

Adding some constants, we rewrite equation (7) as

$$E(\theta, \phi) = K_1(\cos \theta - \cos \theta_0)^2 + K_2 \sin^2 \theta \sin^2 \phi \quad (8)$$

where $\cos \theta_0 = M_0 H / (2K_1)$. As $K_1 \gg K_2 > 0$, the magnetization vector is forced to lie in the $\theta = \theta_0$ plane, and therefore the fluctuations of θ about θ_0 are small. Introducing $\theta = \theta_0 + \alpha$ ($|\alpha| \ll 1$), the total energy $E(\theta, \phi)$ reduces to

$$E(\alpha, \phi) \approx K_1 \sin^2 \theta_0 \alpha^2 + K_2 \sin^2 \theta_0 \sin^2 \phi + 2K_2 \sin \theta_0 \cos \theta_0 \sin^2 \phi \alpha. \quad (9)$$

The ground state of the FM particle with biaxial symmetry corresponds to the magnetization vector pointing in one of the two degenerate easy directions: $\theta = \theta_0$ and $\phi = 0, \pi$; other energy minima repeat the two states with period 2π .

Performing the Gaussian integration over α , we can map the spin system onto a moving-particle problem in a one-dimensional potential well. Now the transition amplitude becomes

$$\mathcal{K}_E = \exp \left\{ -iS \left[1 - \left(1 + \frac{1}{2}\lambda \right) \cos \theta_0 \right] (\phi_f - \phi_i) \right\} \int d\phi \exp(-\mathcal{S}_E[\phi]) \quad (10)$$

with the effective Euclidean action

$$\mathcal{S}_E[\phi] = \int d\tau \left[\frac{1}{2}m \left(\frac{d\phi}{d\tau} \right)^2 + U(\phi) \right] \quad (11)$$

where

$$\lambda = \frac{K_2}{K_1} \quad m = \frac{\hbar S^2}{2K_1 V} \quad U(\phi) = \frac{K_2 V}{\hbar} \sin^2 \theta_0 \sin^2 \phi.$$

The potential $U(\phi)$ is periodic with period π , and there are two minima in the entire region 2π . We may regard the potential $U(\phi)$ as a superlattice with lattice constant π and total length 2π , and we can derive the energy spectrum by applying the Bloch theorem and the tight-binding approximation. The translation symmetry is ensured by the possibility of successive 2π extensions.

Now we apply the periodic instanton method to evaluate the tunnel splittings of excited levels. The periodic instanton configuration ϕ_p which minimizes the Euclidean action of equation (11) satisfies the equation of motion

$$\frac{1}{2}m \left(\frac{d\phi_p}{d\tau} \right)^2 - U(\phi_p) = -E \quad (12)$$

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

$$\sin^2 \phi_p = 1 - k^2 \operatorname{sn}^2(\omega_1 \tau, k). \quad (13)$$

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

$$k^2 = 1 - \frac{\hbar E}{K_2 V \sin^2 \theta_0} \quad (14a)$$

and

$$\omega_1 = 2 \frac{V}{\hbar S} \sqrt{K_1 K_2 \sin \theta_0}. \quad (14b)$$

In the case of resonant quantum tunnelling in the ground state with zero magnetic field, i.e., $E \rightarrow 0$, $k \rightarrow 1$, $\text{sn}(u, 1) \rightarrow \tanh u$, $\lambda \rightarrow 0$, we have

$$\cos \phi_p = \tanh(\omega_1 \tau) \quad (15)$$

which is exactly the vacuum instanton solution derived in reference [21].

The classical action or the WKB exponent can be obtained by integrating the Euclidean action equation (11) with the above periodic instanton solution. The result is found to be

$$S_p = \int_{-\beta}^{\beta} d\tau \left[\frac{1}{2} m \left(\frac{d\phi_p}{d\tau} \right)^2 + U(\phi_p) \right] = W + 2E\beta \quad (16)$$

with

$$W = 2\sqrt{\lambda} S \sin \theta_0 [E(k) - (1 - k^2)K(k)] \quad (17)$$

where $K(k)$ and $E(k)$ are the complete elliptic integrals of the first and second kind, respectively. In the low-energy limit where E is much less than the barrier height, i.e., $k'^2 = 1 - k^2 = \hbar E / K_2 V \sin^2 \theta_0 \ll 1$, we can expand $K(k)$ and $E(k)$ in equation (17) in powers of k' to include terms like k'^2 and $k'^2 \ln(4/k')$:

$$\begin{aligned} E(k) &= 1 + \frac{1}{2} \left[\ln\left(\frac{4}{k'}\right) - \frac{1}{2} \right] k'^2 + \dots \\ K(k) &= \ln\left(\frac{4}{k'}\right) + \frac{1}{4} \left[\ln\left(\frac{4}{k'}\right) - 1 \right] k'^2 + \dots \end{aligned} \quad (18)$$

With the help of the small-oscillator approximation for energy near the bottom of the potential well, $E = \mathcal{E}_n^{bia} = (n + \frac{1}{2})\omega_1$, equation (17) is expanded as

$$W = 2\sqrt{\lambda} S \sin \theta_0 - \left(n + \frac{1}{2}\right) + \left(n + \frac{1}{2}\right) \ln \left[\frac{(n + \frac{1}{2})}{2^3 \sqrt{\lambda} S \sin \theta_0} \right]. \quad (19)$$

Then the general formula, equation (4), gives the low-lying energy shift of the n th excited level for FM particles with biaxial symmetry in a magnetic field along the hard-anisotropy axis as

$$\hbar \Delta \mathcal{E}_n^{bia} = \frac{2^{3/2}}{\sqrt{\pi n!}} \frac{V}{S} \sqrt{K_1 K_2} \sin \theta_0 (8\sqrt{\lambda} S \sin \theta_0)^{n+1/2} \exp(-2\sqrt{\lambda} S \sin \theta_0). \quad (20)$$

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

$$E_n^{bia} = \hbar \mathcal{E}_n^{bia} - 2\hbar \Delta \mathcal{E}_n^{bia} \cos[\pi(\mu + \xi)] \quad (21)$$

where $\mu = S[1 - (1 + \frac{1}{2}\lambda) \cos \theta_0]$, and ξ is the Bloch vector which can be 0 or 1 in the first Brillouin zone. Equation (21) includes the contribution of the Wess–Zumino–Berry phase for FM particles with biaxial symmetry at finite magnetic field. In the absence of a magnetic field, the tunnel splitting is suppressed to zero for half-integer total spins by the destructively interfering Wess–Zumino–Berry phase. This topological quenching effect is in good agreement with the Kramers theorem since the system has time-reversal invariance at zero field. In the presence of even a weak external magnetic field, this strict ‘selection rule’ is relaxed, which leads to a finite tunnel splitting for half-integer total spins. The low-lying energy level

spectrum is $\hbar\mathcal{E}_n^{bia} - 2\hbar\Delta\mathcal{E}_n^{bia}\cos(\pi S\cos\theta_0)$, and $\hbar\mathcal{E}_n^{bia} + 2\hbar\Delta\mathcal{E}_n^{bia}\cos(\pi S\cos\theta_0)$ for integer total spins. However, the low-lying energy level spectrum is $\hbar\mathcal{E}_n^{bia} - 2\hbar\Delta\mathcal{E}_n^{bia}\sin(\pi S\cos\theta_0)$, and $\hbar\mathcal{E}_n^{bia} + 2\hbar\Delta\mathcal{E}_n^{bia}\sin(\pi S\cos\theta_0)$ for half-integer total spins. Therefore the tunnel splitting is $\Delta\mathcal{E}_n = 4\Delta\mathcal{E}_n^{bia}|\cos(\pi S\cos\theta_0)|$ for integer spins, while the tunnel splitting is $\Delta\mathcal{E}_n = 4\Delta\mathcal{E}_n^{bia}|\sin(\pi S\cos\theta_0)|$ for half-integer spins. The tunnel splitting will not be suppressed to zero even if the total spin is a half-integer at finite magnetic field.

In figure 1, we plot the tunnel splitting in the magnetic field at the first excited level ($n = 1$) for integer total spin $S = 100$ obtained by the analytical calculation and the exact-diagonalization calculation. Here we take typical values of parameters for single-domain FM nanoparticles, $K_1 = 10^6 \text{ erg cm}^{-3}$, $\lambda = 0.02$, and the radius of the particle $r = 5 \text{ nm}$. The analytical result is found to be in good agreement with the numerical result, which confirms the theoretical analysis.

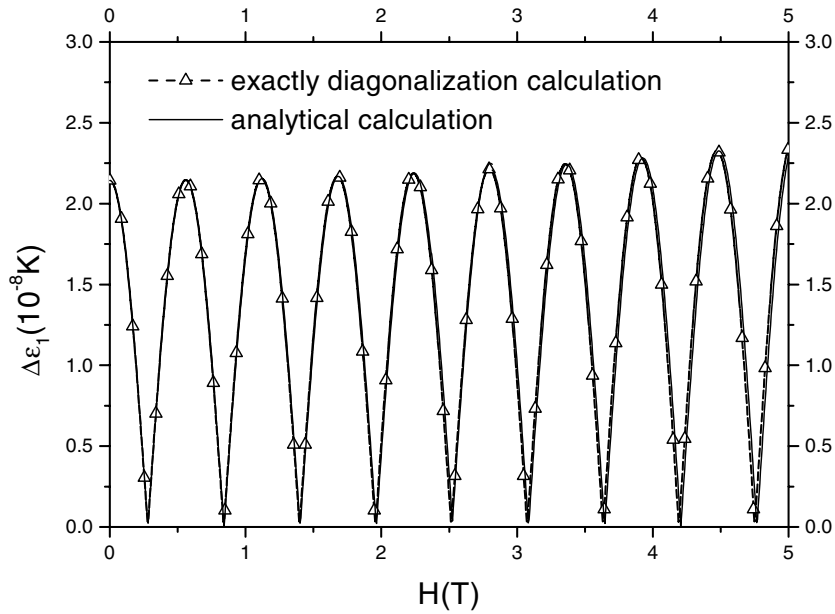


Figure 1. The tunnel splitting $\Delta\epsilon_1$ for biaxial FM particles at the first excited level ($n = 1$) as a function of the field H for integer spins ($S = 100$) obtained by the analytical and the exact-diagonalization calculations. Here $K_1 = 10^6 \text{ erg cm}^{-3}$, $\lambda = 0.02$, and the radius of the particle $r = 5 \text{ nm}$.

In figure 2, we plot the dependence of the first-excited-level splitting on the magnetic field for integer and half-integer total spins, where the oscillation with the field and the spin-parity effects are clearly shown. And in figure 3, we show the tunnel splittings of the ground-state level and the first excited level as functions of the magnetic field for integer total spins $S = 100$. It is clear that the splitting is enhanced by quantum tunnelling at the excited levels.

Recently, spin systems have aroused considerable interest, with the discovery that they provide examples which exhibit first- or second-order transitions between the classical and quantum behaviour of the escape rate [22–24]. In general, transitions in a metastable system can occur via quantum tunnelling through the barrier and the classical thermal activation. It was shown that for a particle with mass m moving in a double-well potential $U(x)$, the behaviour of the energy-dependent period of the oscillations $P(E)$ in the Euclidean potential $-U(x)$ determines the order of the quantum–classical transition [22, 23]. If $P(E)$ increases

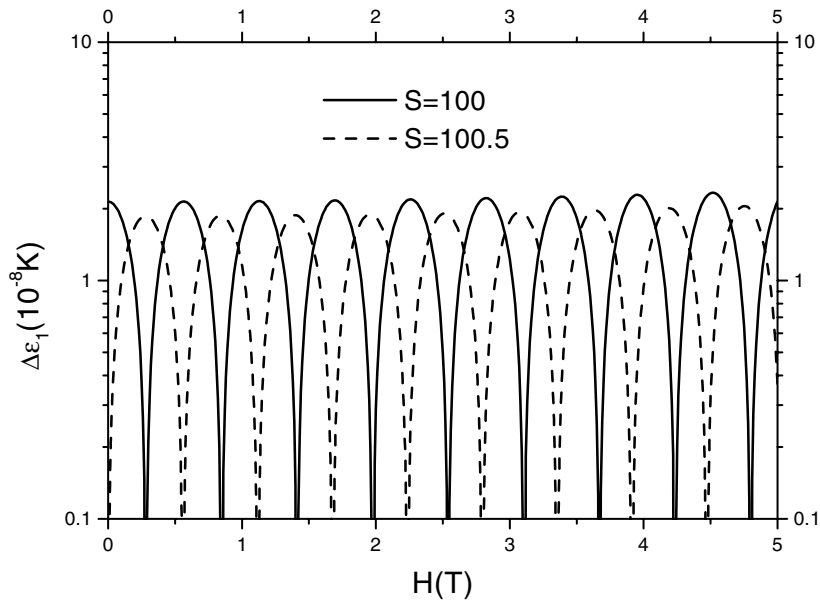


Figure 2. The tunnel splitting $\Delta\varepsilon_1$ for biaxial FM particles at the first excited level ($n = 1$) as a function of the field H , for integer ($S = 100$) and half-integer ($S = 100.5$) total spins. Here $K_1 = 10^6$ erg cm^{-3} , $\lambda = 0.02$, and the radius of the particle $r = 5$ nm.

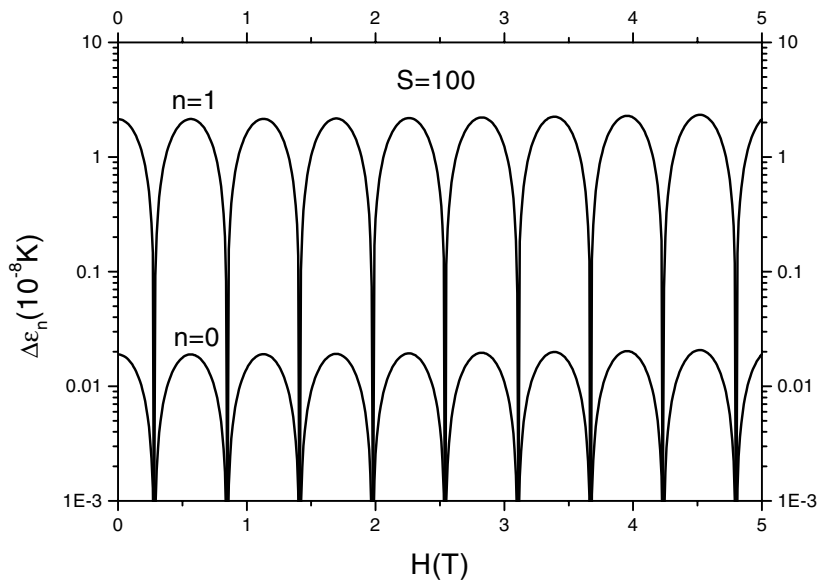


Figure 3. The tunnel splittings $\Delta\varepsilon_n$ for biaxial FM particles at the ground level ($n = 0$) and the first excited level ($n = 1$) as functions of the field H for integer ($S = 100$) total spins. Here $K_1 = 10^6$ erg cm^{-3} , $\lambda = 0.02$, and the radius of the particle $r = 5$ nm.

monotonically with the amplitude of the oscillations, i.e., with decreasing energy E , the transition is of second order. The crossover temperature for the second-order phase transition is $T_0^{(2)} = \tilde{\omega}_0/2\pi$, $\tilde{\omega}_0 = \sqrt{[|U''(x_{sad})|/m]}$, where x_{sad} corresponds to the top (the saddle point)

of the barrier, and $\tilde{\omega}_0$ is the frequency of the small oscillations near the bottom of the inverted potential $-U(x)$. If, however, the dependence of $P(E)$ is non-monotonic, the first-order crossover takes place.

Now we discuss the phase transition from classical to quantum behaviour in this model. For the present case, the period of the periodic instanton is found to be

$$P(E) = \frac{4}{\omega_1} K(k).$$

The monotonically decreasing behaviour of $P(E)$ is shown in figure 4, which shows that a second-order phase transition takes place. We found that the frequency of the small oscillations near the bottom of the inverted potential is

$$\tilde{\omega}_1 = 2 \frac{V}{\hbar S} \sqrt{K_1 K_2} \sin \theta_0.$$

The crossover temperature characterizing the quantum–classical transition is

$$k_B T_0^{(2)} = \frac{V}{\pi S} \sqrt{K_1 K_2} \sin \theta_0. \quad (22)$$

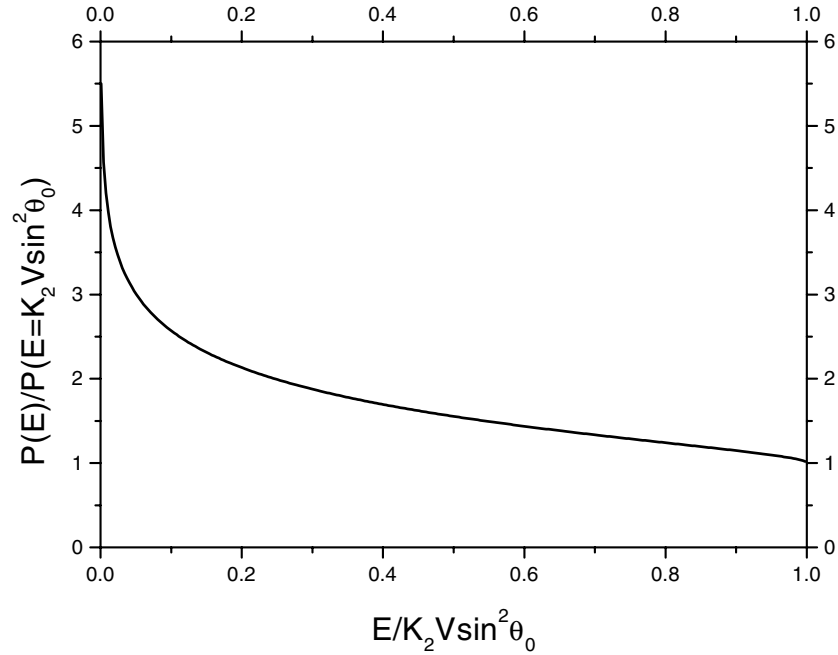


Figure 4. The relative period $P(E)/P(E = K_2 V \sin^2 \theta_0)$ of the periodic instanton as a function of the energy $E/K_2 V \sin^2 \theta_0$ in the domain $0 \leq \hbar E \leq K_2 V \sin^2 \theta_0$.

At the end of this section, we discuss the possible relevance to an experimental test for spin-parity effects in single-domain FM nanoparticles. First we discuss the thermodynamic behaviour of this system at very low temperature $T \sim T_0 = \hbar \Delta \mathcal{E}_0^{bia} / k_B$. For FM particles with biaxial crystal symmetry at such a low temperature, the partition function of the ground state is found to be

$$\mathcal{Z} = 2 \exp(-\beta \hbar \mathcal{E}_0^{bia}) \cosh [2\beta \hbar \Delta \mathcal{E}_0^{bia} \cos(\pi S \cos \theta_0)] \quad (23a)$$

for integer spins, while

$$\mathcal{Z} = 2 \exp(-\beta \hbar \mathcal{E}_0^{bia}) \cosh [2\beta \hbar \Delta \mathcal{E}_0^{bia} \sin(\pi S \cos \theta_0)] \quad (23b)$$

for half-integer spins, where $\mathcal{E}_0^{bia} = \omega_1/2$. Then the specific heat is $c = -T(\partial^2 \mathcal{F}/\partial T^2)$, with $\mathcal{F} = -k_B T \ln \mathcal{Z}$. For FM particles with biaxial crystal symmetry in the presence of a magnetic field along the hard axis at sufficiently low temperatures, we obtain the specific heat as

$$c = 4k_B(\beta\hbar \Delta\mathcal{E}_0^{bia})^2 \frac{[\cos(\pi S \cos \theta_0)]^2}{\{\cosh[2\beta\hbar \Delta\mathcal{E}_0^{bia} \cos(\pi S \cos \theta_0)]\}^2} \quad (24a)$$

for integer spins, while

$$c = 4k_B(\beta\hbar \Delta\mathcal{E}_0^{bia})^2 \frac{[\sin(\pi S \cos \theta_0)]^2}{\{\cosh[2\beta\hbar \Delta\mathcal{E}_0^{bia} \cos(\pi S \cos \theta_0)]\}^2} \quad (24b)$$

for half-integer spins. In figure 5, we show the temperature dependence of the specific heat at $H/H_c = 0.2$ for integer and half-integer total spins. It is clear that the specific heat for integer spins is very different from that for half-integer spins at sufficiently low temperatures.

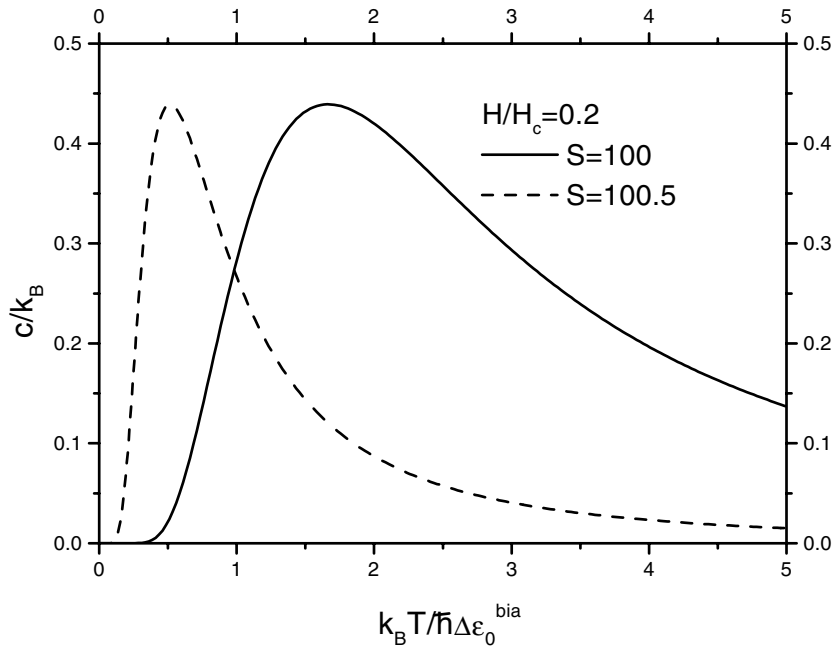


Figure 5. The low-temperature specific heat for biaxial FM particles with integer ($S = 100$) and half-integer ($S = 100.5$) total spins. Here $\lambda = K_2/K_1 = 0.02$, $H/H_c = 0.2$.

When the temperature is higher, $\hbar \Delta\mathcal{E}_0^{bia} \ll k_B T < \hbar\omega_1$, the excited energy levels may give a contribution to the partition function. Now the partition function is found to be

$$\mathcal{Z} \approx \mathcal{Z}_0 \left[1 + (1 - e^{-\beta\hbar\omega_1})(\sqrt{2}\beta\hbar \Delta\mathcal{E}_0^{bia} \cos(\pi\mu))^2 I_0(2q_1 e^{-\beta\hbar\omega_1/2}) \right] \quad (25)$$

for both integer and half-integer spins. Here,

$$\mathcal{Z}_0 = 2e^{-\beta\hbar\omega_1/2} / (1 - e^{-\beta\hbar\omega_1})$$

is the partition function in the well calculated for $k_B T \ll \Delta U$ over the low-lying oscillator-like states with $\mathcal{E}_n^{bia} = (n + 1/2)\omega_1$;

$$I_0(x) = \sum_{n=0}^{\infty} (x/2)^{2n} / (n!)^2$$

is the modified Bessel function, and $q_1 = 2^3 \sqrt{\lambda} S \sin \theta_0 > 1$. We define a characteristic temperature \tilde{T} that is a solution of the equation $q_1 e^{-\hbar\omega_1/2k_B\tilde{T}} = 1$. The temperature $\tilde{T} = \hbar\omega_1/2 \ln q_1$ characterizes the crossover from thermally assisted tunnelling to the ground-state tunnelling. Then we obtain the specific heat up to the order of $(\beta\hbar \Delta\mathcal{E}_0^{bia})^2$ as

$$\begin{aligned}
c = & k_B (\beta\hbar\omega_1)^2 \frac{e^{\beta\hbar\omega_1}}{(e^{\beta\hbar\omega_1} - 1)^2} + k_B (\sqrt{2}\beta\hbar \Delta\mathcal{E}_0^{bia} \cos(\pi\mu))^2 \\
& \times \left\{ [2(1 - e^{-\beta\hbar\omega_1}) + 4(\beta\hbar\omega_1)e^{-\beta\hbar\omega_1} - (\beta\hbar\omega_1)^2 e^{-\beta\hbar\omega_1} I_0(2q_1 e^{-\beta\hbar\omega_1/2})] \right. \\
& - q_1 (\beta\hbar\omega_1) \left[\frac{1}{2} (5e^{-3\beta\hbar\omega_1/2} - e^{-\beta\hbar\omega_1/2}) + 4(e^{-\beta\hbar\omega_1/2} - e^{-3\beta\hbar\omega_1/2}) \right] \\
& \times I'_0(2q_1 e^{-\beta\hbar\omega_1/2}) \\
& \left. + q_1^2 (\beta\hbar\omega_1)^2 (e^{-\beta\hbar\omega_1/2} - e^{-3\beta\hbar\omega_1/2}) I''_0(2q_1 e^{-\beta\hbar\omega_1/2}) \right\} \quad (26)
\end{aligned}$$

for both integer and half-integer spins, where $I'_0 = -I_1$, and $I''_0 = I_2 - I_1/x$.

$$I_\nu(x) = \sum_{n=0}^{\infty} (-1)^n (x/2)^{2n+\nu} / n! \Gamma(n + \nu + 1)$$

where Γ is the Gamma function.

4. MQC for trigonal, tetragonal, and hexagonal symmetries

In this section we will apply the method given in section 3 to study resonant quantum tunnelling of magnetization in single-domain FM nanoparticles with trigonal, tetragonal, and hexagonal symmetry. For the trigonal symmetry, the anisotropy energy is

$$E(\theta, \phi) = K_1 \cos^2 \theta - K_2 \sin^3 \theta \cos(3\phi) - M_0 H \cos \theta + E_0 \quad (27)$$

where $K_1 \gg K_2 > 0$. The energy minima of this system are at $\theta = \theta_0$ and $\phi = 0, \frac{2}{3}\pi, \frac{4}{3}\pi$, and other energy minima repeat the three states with period 2π . This problem can be mapped onto a problem of one-dimensional motion by integrating out the fluctuations of θ about θ_0 , and then the effective potential is

$$U(\phi) = \frac{2K_2 V}{\hbar} \sin^3 \theta_0 \sin^2 \left(\frac{3}{2} \phi \right). \quad (28)$$

Now $U(\phi)$ is periodic with period $\frac{2}{3}\pi$, and there are three minima in the entire region 2π . The periodic instanton configuration with an energy $E > 0$ is found to be

$$\sin^2 \left(\frac{3}{2} \phi_p \right) = 1 - k^2 \operatorname{sn}^2(\omega_2 \tau, k) \quad (29)$$

where

$$k = \sqrt{1 - \frac{\hbar E}{2K_2 V \sin^3 \theta_0}} \quad \omega_2 = 3\sqrt{2} \frac{V}{\hbar S} \sqrt{K_1 K_2 (\sin \theta_0)^{3/2}}.$$

The corresponding classical action is $\mathcal{S}_p = W + 2E\beta$, with

$$W = \frac{2^{5/2}}{3} \sqrt{\lambda} S (\sin \theta_0)^{3/2} [E(k) - (1 - k^2)K(k)]. \quad (30)$$

The low-lying energy shift of the n th excited level is found to be

$$\begin{aligned} \hbar \Delta \mathcal{E}_n^{tri} &= \frac{6}{\sqrt{\pi n!}} \frac{V}{S} \sqrt{K_1 K_2} (\sin \theta_0)^{3/2} \left(\frac{2^{9/2}}{3} \sqrt{\lambda} S (\sin \theta_0)^{3/2} \right)^{n+1/2} \\ &\times \exp \left(-\frac{2^{5/2}}{3} \sqrt{\lambda} S (\sin \theta_0)^{3/2} \right). \end{aligned} \quad (31)$$

The periodic potential $U(\phi)$ can be viewed as a superlattice with lattice constant $\frac{2}{3}\pi$ and total length 2π . Then the Bloch theorem gives the energy level spectrum of the n th excited level $\mathcal{E}_n^{tri} = (n + \frac{1}{2})\omega_2$ as

$$E_n^{tri} = \hbar \mathcal{E}_n^{tri} - 2\hbar \Delta \mathcal{E}_n^{tri} \cos \left[\frac{2\pi}{3} (\mu + \xi) \right] \quad (32)$$

where $\xi = -1, 0, 1$ in the first Brillouin zone. The crossover temperature for the second-order phase transition is

$$k_B T_0^{(2)} = 3\sqrt{2} \frac{V}{\pi S} \sqrt{K_1 K_2} (\sin \theta_0)^{3/2}. \quad (33)$$

For the tetragonal symmetry,

$$E(\theta, \phi) = K_1 \cos^2 \theta + K_2 \sin^4 \theta - K_2' \sin^4 \theta \cos(4\phi) - M_0 H \cos \theta + E_0 \quad (34)$$

where $K_1 \gg K_2, K_2' > 0$. The energy minima are at $\theta = \theta_0$ and $\phi = 0, \frac{1}{2}\pi, \pi, \frac{3}{2}\pi$, and other energy minima repeat the four states with period 2π . The problem can be mapped onto a problem of a particle moving in a one-dimensional potential:

$$U(\phi) = \frac{2K_2' V}{\hbar} \sin^4 \theta_0 \sin^2(2\phi). \quad (35)$$

Now $U(\phi)$ is periodic with period $\frac{1}{2}\pi$, and there are four minima in the entire region 2π . In this case, the periodic instanton solution is

$$\sin^2(2\phi_p) = 1 - k^2 \operatorname{sn}^2(\omega_3 \tau, k) \quad (36)$$

where

$$k = \sqrt{1 - \frac{\hbar E}{2K_2' V \sin^4 \theta_0}} \quad \omega_3 = 4\sqrt{2} \frac{V}{\hbar S} \sqrt{K_1 K_2} \sin^2 \theta_0.$$

The associated classical action is $\mathcal{S}_p = W + 2E\beta$, with

$$W = 2^{1/2} \sqrt{\lambda} S \sin^2 \theta_0 [E(k) - (1 - k^2)K(k)]. \quad (37)$$

The low-lying energy shift of the n th excited level is

$$\hbar \Delta \mathcal{E}_n^{te} = \frac{8}{\sqrt{\pi n!}} \frac{V}{S} \sqrt{K_1 K_2} \sin^2 \theta_0 (2^{5/2} \sqrt{\lambda} S \sin^2 \theta_0)^{n+1/2} \exp(-2^{1/2} \sqrt{\lambda} S \sin^2 \theta_0). \quad (38)$$

The periodic potential $U(\phi)$ can be viewed as a superlattice with lattice constant $\frac{1}{2}\pi$ and total length 2π . For this case the energy level spectrum of the n th excited level $\mathcal{E}_n^{te} = (n + \frac{1}{2})\omega_3$ is

$$E_n^{te} = \hbar \mathcal{E}_n^{te} - 2\hbar \Delta \mathcal{E}_n^{te} \cos \left[\frac{\pi}{2} (\mu + \xi) \right] \quad (39)$$

where $\xi = -1, 0, 1, 2$ in the first Brillouin zone. The crossover temperature is

$$k_B T_0^{(2)} = 2^{5/2} \frac{V}{\pi S} \sqrt{K_1 K_2} \sin^2 \theta_0. \quad (40)$$

For the hexagonal symmetry, the magnetocrystalline anisotropy energy is

$$E(\theta, \phi) = K_1 \cos^2 \theta + K_2 \sin^4 \theta + K_3 \sin^6 \theta - K_3' \sin^6 \theta \cos(6\phi) - M_0 H \cos \theta + E_0 \quad (41)$$

where $K_1 \gg K_2, K_3, K'_3 > 0$. The energy minima are at $\theta = \theta_0$ and $\phi = 0, \frac{1}{3}\pi, \frac{2}{3}\pi, \pi, \frac{4}{3}\pi, \frac{5}{3}\pi$, and other energy minima repeat the six states with period 2π . Now the one-dimensional effective potential is

$$U(\phi) = \frac{2K'_3 V}{\hbar} \sin^6 \theta_0 \sin^2(3\phi). \quad (42)$$

For the present case, $U(\phi)$ is periodic with period $\frac{1}{3}\pi$, and there are six minima in the entire region 2π . The periodic instanton configuration is found to be

$$\sin^2(3\phi_p) = 1 - k^2 \operatorname{sn}^2(\omega_4 \tau, k) \quad (43)$$

where

$$k = \sqrt{1 - \frac{\hbar E}{2K'_3 V \sin^6 \theta_0}} \quad \omega_4 = 6\sqrt{2} \frac{V}{\hbar S} \sqrt{K_1 K_2} \sin^3 \theta_0.$$

Then the corresponding classical action is obtained as $\mathcal{S}_p = W + 2E\beta$, with

$$W = \frac{2^{3/2}}{3} \sqrt{\lambda} S \sin^3 \theta_0 [E(k) - (1 - k^2)K(k)]. \quad (44)$$

Therefore the low-lying energy shift of the n th excited level is found to be

$$\hbar \Delta \mathcal{E}_n^{he} = \frac{3 \times 2^2 V}{\sqrt{\pi} n! S} \sqrt{K_1 K_2} \sin^3 \theta_0 \left(\frac{2^{7/2}}{3} \sqrt{\lambda} S \sin^3 \theta_0 \right)^{n+1/2} \exp\left(-\frac{2^{3/2}}{3} \sqrt{\lambda} S \sin^3 \theta_0\right). \quad (45)$$

The periodic potential $U(\phi)$ can be regarded as a one-dimensional superlattice with lattice constant $\frac{1}{3}\pi$ and total length 2π . By applying the Bloch theorem and tight-binding approximation, we obtain the energy level spectrum of the n th excited level $\mathcal{E}_n^{he} = (n + \frac{1}{2})\omega_4$ as

$$E_n^{he} = \hbar \mathcal{E}_n^{he} - 2\hbar \Delta \mathcal{E}_n^{he} \cos\left[\frac{\pi}{3}(\mu + \xi)\right]. \quad (46)$$

In this case the crossover temperature characterizing the quantum–classical transition is

$$k_B T_0^{(2)} = 6\sqrt{2} \frac{V}{\pi S} \sqrt{K_1 K_2} \sin^3 \theta_0. \quad (47)$$

In brief, the low-lying energy level spectra of the magnetic tunnelling states for trigonal, tetragonal, and hexagonal symmetry are found to depend on the parity of the total spins of the single-domain FM nanoparticles, resulting from the Wess–Zumino–Berry phase interference between topologically distinct tunnelling paths. And the crossover temperature characterizing the quantum–classical transition is also obtained for each case.

5. Conclusions

In summary, we have investigated the spin-phase interference effects in the resonant quantum tunnelling of the magnetization vector between excited levels for single-domain FM nanoparticles in the presence of a magnetic field along the hard-anisotropy axis. The system considered in this paper has a general structure of the magnetocrystalline anisotropy, such as one showing a biaxial, trigonal, tetragonal, or hexagonal symmetry. The low-lying tunnel splittings between the n th degenerate excited levels of neighbouring wells are evaluated with the help of the periodic instanton method in the spin-coherent-state path-integral representation. The low-lying energy level spectrum of the n th excited level is obtained by applying the Bloch theorem and the tight-binding approximation in the one-dimensional periodic potential. This is the first complete study, to our knowledge, of spin-phase interference between excited

levels and effects induced by a magnetic field on FM particles with a general structure of the magnetocrystalline anisotropy.

One important conclusion is that for all of the four kinds of crystal symmetry considered, the low-lying energy level spectrum depends on the spin parity significantly; this results from the Wess–Zumino–Berry phase interference between topologically distinct tunnelling paths. The structures of the low-lying tunnelling level spectra for trigonal, tetragonal, and hexagonal symmetry are found to be much more complex than that for biaxial symmetry. The low-lying energy level spectrum can be non-zero even if the total spin is a half-integer for the trigonal, tetragonal, or hexagonal symmetry at zero magnetic field. Our study provides a generalization of the Kramers degeneracy to coherent spin tunnelling in ground states as well as low-lying excited states. An external magnetic field yields an additional contribution to the Berry phase, resulting in an oscillating field dependence of the tunnel splittings for both the integer and half-integer total spins. This oscillation effect can be tested with the use of existing experimental techniques. Due to the topological nature of the Berry phase, these spin-parity effects are independent of details such as the magnitudes of the total spins, the shape of the soliton, and the tunnelling potential. And the tunnelling effect is enhanced by considering tunnelling at the level of excited states. The transition from quantum tunnelling to thermal activation is also studied. By calculating the oscillation period, we find a monotonically decreasing behaviour of the period with increasing energy, which yields a second-order phase transition. The crossover temperature characterizing the quantum–classical transition is obtained for each case. Comparison with the numerical results gives strong support for the analytical calculations presented in this paper. The heat capacity of low-lying magnetic tunnelling states is evaluated and is found to depend significantly on the parity of total spins for FM particles at sufficiently low temperature. This provides a possible experimental method for examining the theoretical results on spin-phase interference effects. Our results presented here should be useful for a quantitative understanding of the topological phase interference or spin-parity effects in resonant quantum tunnelling of the magnetization in single-domain FM nanoparticles.

The theoretical calculations performed in this paper can be extended to single-domain antiferromagnetic nanoparticles, where the relevant quantity is the excess spin due to the small non-compensation of two sublattices. Work along these lines is still in progress. We hope that the theoretical results obtained in the present work will stimulate more experiments whose aim is observing the topological phase interference or spin-parity effects in resonant quantum tunnelling of the magnetization in nanoscale single-domain ferromagnets.

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