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Instantons and phase transition in quantum interference

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Abstract
In the biaxial nanospin system, quantum tunnelling of the magnetization vector oscillates with increasing magnetic field or temperature. When the number of instanons is equal to the number of anti-instantons, the annihilation of instanton–anti-instanton pair results in a vanished Berry curvature, it leads to the topological quenching point. When the number of instantons and anti-instantons is not equal, berry curvature is nonzero, this suggests that there are extra instantons that revive the tunnelling splitting. So the generation (annihilation) of instanton induced the revival (quenching) of tunnel splitting. This generation or annihilation is accompanied by different phase transitions which can be distinguished by $\frac{H/H_c}{1-K_2/K_1}$. When the system slides from the phase of $H/H_c < 1 - K_2/K_1$ into the phase of $H/H_c > 1 - K_2/K_1$, an instanton will be generated (annihilated), which revives (quenches) the quantum tunnelling. This conclusion also holds when phase transition occurs in the opposite direction, i.e., from $H/H_c > 1 - K_2/K_1$ to $H/H_c < 1 - K_2/K_1$. So there is a phase transition that occurs alternatively in the magnetic molecule system when the topological quenching of the tunnel splitting occurs quasi-periodically.

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(Some figures in this article are in colour only in the electronic version)

1. Introduction

For a single-domain ferromagnetic nanoparticle at sufficiently low temperatures, all the spins are locked together by the strong exchange interaction, and therefore only the orientation of the total magnetization vector can change but not its absolute value. The magnetocrystalline anisotropy and the external applied magnetic field can create easy directions for the total magnetization vector which correspond to local minima of magnetic energy. The vector of the magnetization of spins can coherently tunnel between the minima of magnetic energy.

The behaviours of quantum tunnelling of the magnetization vector are sensitive to the parity of total spin of the single-domain magnet. It has been demonstrated that the ground-state tunnelling level splitting is completely suppressed [1, 2] to zero for the half-integer total spin ferromagnetic nanoparticles with biaxial crystal symmetry. The tunnel splitting oscillates [3] as a function of magnetic field H and vanished at $2J$ field values lying in the interval $(-H^*, H^*)$, where $H^* = (1-\lambda)^{1/2}H_c$. Chudnovsky and Martinez-Hidalgo [4] have found that the switching from oscillations to the monotonic growth of the tunnelling splitting exists beyond the field range of $(-H^*, H^*)$. Due to the cancellation between the real-time motion of the instanton and the contribution of the topological phase, the splitting grows monotonically instead of oscillating in the field range of $H > H^*$. Garg extended the previous calculations of the ground-state tunnel splittings in the presence of a magnetic field along the hard axis and clarified the physical meaning of the monotonic growth from the viewpoint of level crossing [3].

Besides the oscillation of the tunnel splitting of the ground states in the presence of the magnetic field along the hard axis at zero temperature, Kim found that for a given magnetic field along the hard axis the tunnel splitting also oscillates with increasing temperature and it is topologically quenched quasi-periodically [5]. Furthermore, Martinez-Hidalgo and Chudnovsky found a double transition—from thermal activation to thermally activated quantum tunnelling and then to ground-state quantum tunnelling [6]. Using the Landau phase transition theory, Kim also found that the oscillation of the tunnel splitting presents different behaviours in different phases at the phase transition temperature, the different behaviours are marked by different phase transitions in the phase diagram of $H/H_c - (1 - K_2/K_1)$ at the critical temperature.

When we increase external applied magnetic field H in the phase of $H/H_c < 1 - K_2/K_1$ (K is the anisotropy constant, we denote $u = H/H_c$ and $K_2/K_1 = \lambda$ for convenience), the oscillation near the zero temperature has a larger monotonic region and the topological quenching happens slowly. While in the phase of $H/H_c > 1 - K_2/K_1$, the monotonic region near zero temperature becomes smaller and the topological quenching occurs more often [5]. The different behaviours of the tunnelling splitting correspond to different phase transitions. It bears a topological origin in the framework of topological quenching of quantum tunnelling. In this paper, we will present a theoretical explanation to Kim's numerical results of different phase behaviours within a novel instanton approach.

The instanton method has been widely applied to study the quantum tunnelling. The quantum tunnelling is completely suppressed if the total spin of the magnetic particle is half integral but is allowed in integral-spin particle [7], such as tunnelling of the magnetic in small ferromagnetic particles [8], tunnelling of the Neel vector in anti-ferromagnetic particles [9], quantum nucleation of magnetic domains [10] and tunnelling of domain walls [11]. The quantum tunnelling suppression has a topological origin and arises as a destructive interference between different paths. Within an instanton approximation [12], it can be clearly seen that this suppression actually arises as the destructive interference between the instanton and anti-instanton.

In the Yang–Mills field theory, instanton is a kind of soliton, which tunnels through different vacua. While in the molecule magnetic cluster system, the lowest energy levels play the role of two different vacua; in this case, the instantons are the symmetry-related tunnelling paths connecting two classically degenerate minima, on which the action is stationary.

How the interference between the instanton and anti-instanton occurs in different phases is an interesting open question. We will provide a novel approach to the quantum interference from a new field theory of Berry curvature; the instanton path will be treated as topological particles moving in the configuration space. It will be clearly seen how destructive interference between instantons and anti-instantons happens. The phase transition is induced by the generalization and annihilation of instantons, which also indicates the quenching and revival of the quantum tunnelling.

This paper is organized as follows: in section 2, by introducing the topological current of instantons and anti-instanton, we obtain two classes of solutions and studied the topological number of Berry curvature in different phases. In section 3, we studied the two kinds of phase transitions and the total topological charge of instantons. In the last section, a brief summary is presented.

2. Instantons and quantum interference

The tunnelling rate or the tunnelling splitting Γ for macroscopic magnetization tunnelling is given by $\Gamma = A\exp\left(-\frac{I_E}{\hbar}\right)$, here I_E is the Euclidean counterpart of the magnetic action

$$I = \int \mathrm{d}t L = \int \mathrm{d}t\{-\hbar\sigma(\cos\theta - 1)\dot{\phi} - E(\theta,\phi)\}. \tag{1}$$

The first term $\omega[\hat{n}] = \int_0^{\pi} \hbar\sigma(1-\cos\theta)\,\mathrm{d}\phi$ is Wess–Zumino term which is of crucial importance in studying the topological quenching of tunnelling in magnetic molecules. It is intimately related to the Berry phase. For any path on the 2-sphere S^2, the contribution of this term to the action is equal to $\mathrm{i}\sigma$ times the area swept out on S^2 between the path and the north pole; for closed paths this has exactly the form of the Berry phase. When the destructive interference between the instanton and anti-instanton happens, the tunnelling rate Γ becomes zero. In this section, we proposed a topological current which vanishes when the destructive interference happened.

We start from the Hamiltonian with correct anisotropy structure in a magnetic field along $-\hat{z}$,

$$H = K_1 J_x^2 + K_2 J_y^2 + g\mu_B H J_z, \tag{2}$$

this Hamiltonian is invariant below 180° rotations about $\hat{z}$, and so it is best to work in the J_z basis $|m\rangle$. To understand the eigenstates of H, we write its mean value in the coherent state $|\theta,\phi\rangle$ as $E(\theta,\phi)$. Dropping terms of order J compared to J^2, we have

$$E(\theta,\phi) = (K_1\cos^2\phi + K_2\sin^2\phi)J^2\sin^2\theta + g\mu_B H J\cos\theta. \tag{3}$$

For clarity, we denote $\cos\theta = v$, $u = \cos\theta_0 = H/H_c$ and $\lambda = K_2/K_1$, here $H_c = 2K_1/(g\mu_B)$ and $K_1 > K_2$. Adding a constant term, this classical anisotropy energy E can be rewritten as

$$E(u,\phi) = K_1(v-u)^2 + \lambda(1-v^2)\sin^2\phi, \tag{4}$$

where $\vec{n}$ is the magnetization direction and $K_1 > K_2 > 0$. Considering a single-domain ferromagnetic particle with the magnetic momentum $M = 2\mu_B S$ and $S \gg 1$, the dynamical equation for M is

$$\frac{\mathrm{d}M}{\mathrm{d}t} = -\gamma M \times \frac{\delta E}{\delta M}. \tag{5}$$

This equation is called Landau–Lifshitz equation or Bloch equation. It can be expressed by the magnetization direction $\vec{m}$.

The total spin of a single-domain ferromagnetic particle is formed by the ferromagnetic alignment of atomic spins. The exchange coupling, responsible for this alignment, is usually large enough to ensure that, at low temperature, the only relevant dynamics of S in sufficiently small particles is its coherent rotation satisfying

$$s\frac{\mathrm{d}\hat{m}}{\mathrm{d}t} = -\hat{m} \times \frac{\delta E}{\delta \hat{m}}. \tag{6}$$

In terms of coordinates θ and ϕ, the above equation is equivalent to

$$\dot{\theta}\sin\theta = \frac{\gamma}{M_0}\frac{\partial E}{\partial \phi}, \qquad \dot{\phi}\sin\theta = \frac{\gamma}{M_0}\frac{\partial E}{\partial \theta}. \tag{7}$$

Substituting equation (3) into equation (7), we arrive at the Euler–Lagrange equations:

$$\dot{\theta} = \frac{-2\gamma}{M}\lambda\sqrt{(1-v^2)}\sin\phi\cos\phi, \qquad \dot{\phi} = \frac{12\gamma}{M}[(v-u) - \lambda\sin^2\phi\cos\theta]. \tag{8}$$

The action is stationary on the instanton path, we introduce the vector order parameter field from equation (8)

$$\varphi^1 = \frac{-2\gamma}{M}\lambda\sqrt{(1-v^2)}\sin\phi\cos\phi, \qquad \varphi^2 = \frac{12\gamma}{M}[(v-u) - \lambda\sin^2\phi\cos\theta] \tag{9}$$

and denote $\theta = q^1$ and $\phi = q^2$. As we known, the unit vector $\vec{m}$ describes a two-dimensional sphere in configuration space; for convenience, we introduce the tangent vector field on this sphere:

$$n^a = \frac{\varphi^a}{\|\varphi\|}, \qquad n^a n^a = 1, \qquad \|\varphi\| = (\varphi^a\varphi^a)^{\frac{1}{2}}. \tag{10}$$

Based on this tangent vector field, we will obtain a new expression for the Berry curvature. Obviously, the zero points of φ are the singular point of the unit vector field $\vec{n}$. However, singular points are always very important to the topology of the systems. For example, on a two-dimensional sphere, there must be at least two singular points, each of which is assigned with a winding number $W = +1$, to make sure that the Euler characteristic is 2. In fact, in the nonlinear σ model of one-dimensional anti-ferromagnetic ring, the Neel unit vector $\vec{m}$ just describes a unit 2-sphere S^2. The unit vector field $\vec{n} = (n^1, n^2)$ can be viewed as the tangent vector on the sphere S^2, while it will be shown that the topological soliton—instanton—sits at the singular points of $\vec{n}$.

By introducing the $SO(2)$ spin connection $\omega_\mu^{ab}n^b$, we define the covariant derivative of the unit vector field as

$$D_\mu n^a = \partial_\mu n^a - \omega_\mu^{ab}n^b. \tag{11}$$

It is easy to verify that the $SO(2)$ spin connection ω_μ^{ab} can be rewritten as

$$\omega_\mu^{ab} = (\partial_\mu n^a n^b - \partial_\mu n^b n^a) - (D_\mu n^a n^b - D_\mu n^b n^a). \tag{12}$$

Keeping in mind the $U(1)$ gauge potential $A_\mu = \frac{1}{2}\epsilon_{ba}\omega_\mu^{ab}$, we have

$$A_\mu = \epsilon_{ab}n^a\partial_\mu n^b - \epsilon_{ab}D_\mu n^a n^b. \tag{13}$$

If $\epsilon_{ab}n^b = k^a$, i.e., k^a is perpendicular to n^a, then using n^a and k^a, the $U(1)$ connection can be reduced further to $A_\mu = \partial_\mu n^a k^a - D_\mu n^a k^a$. Let u^a be a unit vector field satisfying $D_\mu u^a = 0$ with $u^a = \cos\theta n^a + \sin\theta k^a$, it can be proved that $-k^a D_\mu n^a = \partial_\mu\theta$. Therefore, the covariant derivative part of (13) is identified to the gradient of a phase factor θ, then equation (13) can be expressed as

$$A_\mu = \epsilon_{ab}n^a\partial_\mu n^b + \partial_\mu\theta. \tag{14}$$

We can see that the second term of (14), $\partial_\mu\theta$, behaves as a $U(1)$ gauge transformation of A_μ, it vanishes spontaneously in the gauge field tensor $F_{\mu\nu}$ and can be ignored in $U(1)$ gauge potential decomposition theory [16]. Substituting equation (14) into the $U(1)$ gauge field tensor $F_{\mu\nu} = (\partial_\mu A_\nu - \partial_\nu A_\mu)$, we have

$$F_{\mu\nu} = \epsilon_{ab}\partial_\mu n^a \partial_\nu n^b. \tag{15}$$

It is easy to see that the gauge field 2-form $F = \frac{1}{2}\epsilon^{\mu\nu} F_{\mu\nu}\, \mathrm{d}^2 q = \epsilon^{\mu\nu}\epsilon_{ab}\partial_\mu n^a \partial_\nu n^b\, \mathrm{d}^2 q$ bears the familiar form of a topological current.

In the nonlinear σ model of one-dimensional anti-ferromagnetic ring, the Wess–Zumino term can be defined by the Neel unit vector $\vec{m}$: $\epsilon^{\mu\nu}\vec{m}\cdot(\partial_\mu\vec{m}\times\partial_\nu\vec{m})$, it can be rewritten as $\epsilon^{\mu\nu}\epsilon^{abc}m^a(\partial_\mu m^b \partial_\nu m^c)$. If we choose $\vec{m} = (\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta)$, it is easy to verify that it is identical with the usual Wess–Zumino term $-\mathrm{i}s(1-\cos\theta)\dot{\phi}$. Moreover, instantons which arise from the topological current of gauge field 2-form $F = \frac{1}{2}\epsilon^{\mu\nu}F_{\mu\nu}d^2q$ is inconsistent with the Wess–Zumino term.

Using $\partial_\mu \frac{\varphi^a}{\|\varphi\|} = \frac{\partial_\mu\varphi^a}{\|\varphi\|} + \varphi^a\partial_\mu\frac{1}{\|\varphi\|}$ and the Green function relation in φ-space: $\partial_a\partial_a \ln\|\varphi\| = 2\pi\delta^2(\vec{\varphi})\big(\partial_a = \frac{\partial}{\partial\varphi^a}\big)$, one can prove [16] that

$$F = \epsilon^{\mu\nu}\epsilon_{ab}\partial_\mu n^a\partial_\nu n^b = 2\pi\delta^2(\vec{\varphi})D\left(\frac{\varphi}{q}\right) \tag{16}$$

where $D\big(\frac{\varphi}{q}\big)$ is the Jacobian vector. Equation (16) is an another kind of expression for Berry curvature. In the Yang–Mills gauge field theory, instantons solutions [13] arise from the self-dual equation $F_{\mu\nu} = \frac{1}{2}\epsilon_{\mu\nu\lambda\rho}F_{\lambda\rho}$. When $\vec{\varphi} = 0$, the gauge field is infinite, then the self-dual equation is spontaneously satisfied. Therefore, the solutions of $\vec{\varphi} = 0$ correspond to instanton solutions. In the differential geometry theory, the first Chern number is given by

$$\chi(M) = \frac{1}{2\pi}\int F = \int_M \delta^2(\vec{\varphi})D\left(\frac{\varphi}{q}\right)\mathrm{d}^2q. \tag{17}$$

It can be proved, on a compact oriented two-dimensional manifold, that the Euler number is equal to the first Chern number. As we know, the Euler number of a 2-sphere is 2; if the configuration space of the system forms a 2-sphere, this Chern number is 2. In that case there must exist at least two instantons. The expression (17) provides us an important conclusion: $\chi = 0$, iff $\vec{\varphi} \neq 0$; $\chi \neq 0$, iff $\vec{\varphi} = 0$. In other words, if there exist instantons, the Chern number is nontrivial; if there are no instantons, the Chern number vanishes. So it is necessary to study the zero points of $\vec{\varphi}$ to determine the nonzero solutions of χ. The implicit function theory shows [14] that under the regular condition $D(\varphi/q) \neq 0$, the solutions of equations

$$\varphi^1 = \frac{-2\gamma}{M}\lambda\sqrt{(1-v^2)}\sin\phi\cos\phi = 0, \qquad \varphi^2 = \frac{12\gamma}{M}[v(1-\lambda\sin^2\phi) - u_0] = 0 \tag{18}$$

can be expressed as $q_l = \{\theta_n, \phi_m\}(l = 1, 2, \ldots, N)$. According to the δ-function theory [15, 16], one can expand $\delta(\vec{\varphi})$ at the N solutions $q_l = \{\theta_n, \phi_m\}$ as

$$\delta^2(\vec{\varphi}) = \sum_{l=1}^{N}\beta_l\frac{\delta^2(q-\vec{q}_l)}{\big|D\big(\frac{\varphi}{q}\big)\big|_{q_l}} \tag{19}$$

where l denotes the indices pair $\{n, m\}$. The positive integer $\beta_l = |W_l|$ is called the Hopf index of φ-mapping which means that when the point $\vec{q}$ covers the neighbourhood of the zero point $\vec{q}_n$ once, the vector field $\vec{\varphi}$ covers the corresponding region β_n times. Considering equation (16), the gauge field 2-form can be expanded as

$$F = 2\pi\sum_{l=1}^{N}\beta_l\eta_l\delta^2(q-\vec{q}_l)d^2q \tag{20}$$

where

$$\eta_l = \frac{D\left(\frac{\varphi}{q}\right)_{q_l}}{\left|D\left(\frac{\varphi}{q}\right)\right|_{q_l}} = \mathrm{sgn}\left(D\left(\frac{\varphi}{q}\right)_{q_l}\right) = \pm 1 \tag{21}$$

is called the Brouwer degree of map φ. While $\beta_l \eta_l = W_l$ is just the winding number of the lth zero point. η_l can be obtained by substituting equation (9) into the Jacobian

$$D\left(\frac{\varphi}{q}\right) = \left(\frac{\partial \varphi^1}{\partial \theta}\frac{\partial \varphi^2}{\partial \phi} - \frac{\partial \varphi^2}{\partial \theta}\frac{\partial \varphi^1}{\partial \phi}\right), \tag{22}$$

it yields

$$D\left(\frac{\varphi}{q}\right) = 12\left(\frac{\gamma}{M}\right)^2\left(\frac{K_2}{K_1}\right)^2 \cos\theta \sin^2 2\phi - 24\left(\frac{\gamma}{M}\right)^2\left(\frac{K_2}{K_1}\right)\sin^2\theta \cos 2\phi\left(1 - \frac{K_2}{K_1}\sin^2\phi\right). \tag{23}$$

This is the fundamental equation to find the value of the Brouwer degree η_l at the zero points q_l. In terms of the Brouwer degree η_l and Hopf index β_l, the Euler topological number, equation (17), becomes

$$\chi(M) = \sum_{l=1}^{N} \beta_l \eta_l. \tag{24}$$

2.1. The case for the first solution

There are two classes of solutions of equation (18); first, we present a brief discussion of the first solution

$$\theta = n\pi, \qquad \phi = \arcsin\left(\sqrt{\pm\frac{\cos\theta_0}{1 - \frac{K_2}{K_1}}}\right) + 2m\pi \qquad (n = \pm 1, \pm 2, \ldots, \pm N). \tag{25}$$

Substituting equation (25) into equation (23), we have

$$D_{(1)}\left(\frac{\varphi}{q}\right) = 48\left(\frac{\gamma}{M}\right)^2 \lambda^2 \frac{u}{1-\lambda}\left[(-1)^n - \frac{u}{1-\lambda}\right]. \tag{26}$$

As defined above, $\gamma > 0$, $K_1 > K_2 > 0$, so $\lambda = K_2/K_1 < 1$, then $\left(1 - \frac{1}{\lambda}\right) < 0$; therefore, the Brouwer degree of the first solution is $\eta_l^{(1)} = \mathrm{sgn}(D_1(\varphi/q)) = (-1)^n$. Then equation (20) can be rewritten as

$$F = 2\pi \sum_{n,m=0}^{N} |W_{(n,m)}|(-1)^{n+1}\delta(\theta - n\pi)\delta\left(\phi - \arcsin\sqrt{\frac{K_1}{K_2}} + 2m\pi\right)\mathrm{d}\theta\,\mathrm{d}\phi, \tag{27}$$

when $n = 2p + 1$ (p is integer), the winding number $W_{(n,m)} > 0$, it represents the instantons periodically distributed along the θ-axis when the angle ϕ varies from $\arcsin\sqrt{\frac{K_1}{K_2}}$ to $\arcsin\sqrt{\frac{K_1}{K_2}} + 2m\pi$. When $n = 2p$ (p is integer), the winding number $W_{(n,m)} < 0$, it represents the anti-instantons. Now we see that the topological charge of the instanton and anti-instantons is highly dependent on the parity of periodic number n. Equation (27) indicates that the instantons at $n\pi$ is identical with its parallel transformation to $(n+k)\pi$, so we choose the winding number as $W_{n,m} = +1$ for instanton and $W_{n,m} = -1$ for anti-instantons. The topological number of the first solution is

$$\chi(M) = \begin{cases} \sum_n^N (-1)^{n-1}, & u > (1-\lambda), \\ \sum_n^N (-1)^n, & u < (1-\lambda). \end{cases} \tag{28}$$

Obviously, the topological charge of instantons only depends on the periodic number n of θ.

We first consider the range of $u < (1-\lambda)$. Since the sign of the winding number of instantons only depends on n, the total topological charge relies on the parity of n. When $n = 2p+1$ (p is integer), the number of instantons equals the number of anti-instantons and they possess opposite charge; so, the topological charge of Berry curvature $\chi = 0$. While for $n = 2p$ (p is integer), $\chi = 1$. In the range of $u > (1-\lambda)$, it is the opposite case.

In a wide range of systems, the destructive interference between instanton and anti-instanton results in suppression of quantum tunnelling if the total spin of the magnetic particle is half integer. Equation (28) suggests that the destructive interference between instanton and anti-instanton results in the vanish of topological charge.

2.2. The case for the second solution

Now let us switch to the second solution

$$\theta = \arccos\left(\frac{\cos\theta_0}{1-\frac{K_2}{K_1}}\right) + 2n\pi, \qquad \phi = \frac{m\pi}{2} \qquad (n = \pm 1, \pm 2, \ldots, \pm N). \tag{29}$$

Substituting the above equation into equation (23), we arrive at

$$D_{(2)}\left(\frac{\varphi}{q}\right) = 6\,\omega_0^2\left[1 - \left(\frac{u}{1-\lambda}\right)^2\right](-1)^m(1-\lambda), \tag{30}$$

where $\omega_0 = 2\gamma\sqrt{K_2K_1}/M$.

Since $\lambda = K_2/K_1 < 1$, so $(1-\lambda) > 0$, the Brouwer degree for the second class of solution is decided by $u/(1-\lambda)$ and $(-1)^m$. When $u > (1-\lambda)$, the Brouwer degree of the second solution is $\eta_l^{(2)} = (-1)^{m+1}$, if $u < (1-\lambda)$, $\eta_l^{(2)} = (-1)^m$. So, the topological current is divided into two cases correspondingly; for $u > (1-\lambda)$, it is

$$F = 2\pi \sum_{n,m=0}^{N} |W_{(n,m)}|(-1)^{m+1}\delta\left(\theta - n\pi - \frac{\pi}{2}\right)\delta\left(\phi - \frac{m\pi}{2}\right)\mathrm{d}\theta\,\mathrm{d}\phi, \tag{31}$$

when $u < (1-\lambda)$, it becomes

$$F = 2\pi \sum_{n,m=0}^{N} |W_{(n,m)}|(-1)^{m}\delta\left(\theta - n\pi - \frac{\pi}{2}\right)\delta\left(\phi - \frac{m\pi}{2}\right)\mathrm{d}\theta\,\mathrm{d}\phi. \tag{32}$$

Then, we have

$$\chi(M) = \begin{cases} \sum_{n,m=0}^N (-1)^{m+1}|W_{(n,m)}|, & u > (1-\lambda), \\ \sum_{n,m=0}^N (-1)^{m}|W_{(n,m)}|, & u < (1-\lambda). \end{cases} \tag{33}$$

It presents a similar case as the first solution. The only difference is that the first solution mainly depends on θ, while the second solution depends on ϕ.

The above equation reveals that, when the system transform from $u > (1-\lambda)$ into $u < (1-\lambda)$, the topological charge of instantons will change signs. In other words, there is a topological phase shift of instantons by $\mathrm{e}^{\mathrm{i}\pi}$. Keeping in mind $\lambda = K_2/K_1$ and $u = \cos\theta_0 = H/H_c$, the flipping of instantons may be observed by adjusting the magnetic field and anisotropy constant K_2, K_1.

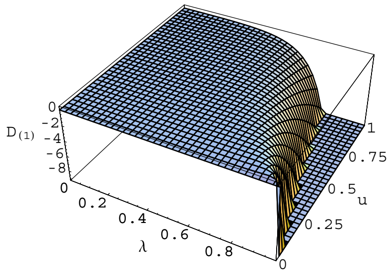

Figure 1. The Jacobian of the first solution $D_{(1)}$ maintains a constant configuration whenever n is an odd number or even number. The solution of $D_{(1)} = 0$ is $u = 1 - \lambda$, which corresponds the curve on which instantons bifurcated. In the regime $u > (1 - \lambda)$, there is crag, the velocity of instanton becomes zero on the edge of the crag. Outside the crag there are no instantons.

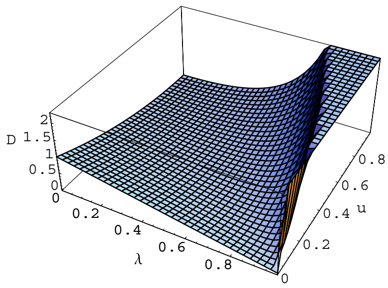

Figure 2. The Jacobian of the second solution $D_{(2)}$ when m is even. The instanton stops when they meet the crag.

So, the first second solution, equation (32), is divided into two phases, the instantons in the phase of $u > (1 - \lambda)$ possess an opposite topological charge to the phase of $u < (1 - \lambda)$. While $u = (1 - \lambda)$ just corresponds to the bifurcation point of instantons, i.e., the instantons are generating or annihilating on the curve $u = (1 - \lambda)$.

3. The phase transition and bifurcation of instantons

In fact, the N isolated solutions in the above are derived under the regular condition $D\left(\frac{\varphi}{q}\right) \neq 0$, when this condition fails, i.e., $D\left(\frac{\varphi}{q}\right) = 0$, the branch process will occur. The velocity of the topological particles in $\theta - \phi$ plane is given by

$$\frac{\mathrm{d}q^i}{\mathrm{d}t} = \left.\frac{D^i\left(\frac{\varphi}{q}\right)}{D^0\left(\frac{\varphi}{q}\right)}\right|_{(q_i^*)} \qquad (i = 1, 2) \tag{34}$$

where $D^0(\varphi/q) = D(\varphi/q) = \epsilon_{ab}\partial_1\varphi^a\partial_2\varphi^b$, $D^1(\varphi/q) = \epsilon_{ab}\partial_2\varphi^a\partial_0\varphi^b$ and $D^2(\varphi/q) = \epsilon_{ab}\partial_0\varphi^a\partial_1\varphi^b$. When $D\left(\frac{\varphi}{q}\right) = 0$, $\mathrm{d}q^i/\mathrm{d}t \to \infty$. This means the velocity of instantons is infinite. So, instantons are generating or annihilating at this point. Furthermore, when $D\left(\frac{\varphi}{q}\right) \to \infty$ and $D^i \neq 0$, the velocity of instantons $\mathrm{d}q^i/\mathrm{d}t \to 0$. From figures 1–3, we see, in the regime $u > 1 - \lambda$, that the Jacobian $D\left(\frac{\varphi}{q}\right)$ dramatically increased to infinity. On the edge of the crag, the velocity of the topological particles goes to zero; in other words, they finally stopped at this critical boundary; outside the critical wall, there is no instantons, so instantons condensate

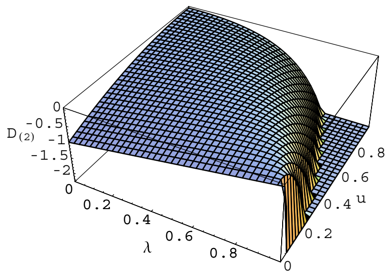

Figure 3. The Jacobian of the second solution $D_{(2)}$ when m is odd. Comparing it with figure 2, we show that the two classes of solutions are in opposite phases, the velocity of the instanton is in opposite direction, but they both stop at the crag. That is the place the instanton transforms into anti-instanton or the opposite process.

on the edge of the crag. From equation (26) and $D_{(1)}\left(\frac{\varphi}{q}\right) = 0$, we have

$$\lambda = 1 \qquad u = (-1)^n(1-\lambda), \tag{35}$$

while for $D_{(2)}\left(\frac{\varphi}{q}\right) = 0$, equation (30) yields

$$\lambda = 1 \qquad u = (1-\lambda). \tag{36}$$

Now one sees that they share a same solution $\lambda = 1$, when n is even number, the other solution is $u = (1-\lambda)$. Recall that $\lambda = K_2/K_1$ and $u = \cos\theta_0$, we arrive at $K_2 = K_1$ and $\cos\theta_0 = (1 - K_2/K_1)$. Now we see that instantons of the second solution bifurcated at $H/H_c = (1 - K_2/K_1)$ and split into two classes which are distinguished by a $\mathrm{e}^{\mathrm{i}\pi}$ phase shift.

According to the phase transition of Landau theory, this bifurcation line is the borderline between two different phases. By performing the Gaussian integration over $\cos\theta$ in the partition function of the biaxial spin system in the presence of temperature and magnetic field, Kim obtained the potential $U(\phi)$ of the effective action [5]

$$U(\phi) = \frac{1}{2}\sin^2\phi\left(1 - \frac{u^2}{1-\lambda\sin^2\phi}\right). \tag{37}$$

This potential shows that there are three different ranges of the field. The position $\phi = \pi/2$ is the maximum for $u < 1-\lambda$, it becomes the local minimum for $1-\lambda < u < \sqrt{1-\lambda}$ and the global minimum for $\sqrt{1-\lambda} < u < 1$. The maximum starts to change from $\pi/2$ either $\phi_m[= \arcsin\sqrt{(1-u)/\lambda}]$ or $\pi - \phi_m$ at $u = 1-\lambda$ and vanishes at $u = 1$ which defines the critical field. By introducing the energy variable $p = (U_{\max} - E)/(U_{\max} - U_{\min})$, where $U_{\max}(U_{\min})$ corresponds to the top (bottom) of the potential, the effective action becomes

$$\frac{1}{\hbar}S_E^{\mathrm{eff}}(p) = \frac{U_{\max} - U_{\min}}{k_B T}[1 + \alpha p + \beta p^2 + O(p^3)]. \tag{38}$$

If the factor β is negative (positive), the system becomes the first- (second-) order transition. There exist two regimes which exhibit the first-order transition, and the phase boundaries are given by $u^{(<)} = (1-\lambda)\sqrt{(1-2\lambda)/(1+\lambda)}$ and

$$u^{\pm}_{(>)} = \frac{16 - 16\lambda + \lambda^2 \pm \lambda\sqrt{\lambda^2 + 32\lambda - 32}}{16 - 2\lambda} \tag{39}$$

where $<$ ($>$) indicates the field region U smaller (larger) than $1-\lambda$. Thus, the first-order regime is surrounded by $u^{(<)} < u < 1-\lambda$ and $u^{-}_{(>)} < u < u^{+}_{(>)}$. While the second-order regimes are given by $u > 1-\lambda$ and $u < u^{(<)}$ (figure 4).

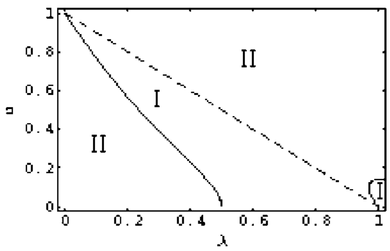

Figure 4. The phase diagram u versus λ, where $\lambda = K_2/K_1$ and $u = \cos\theta_0 = H/H_c$ with $H_c = 2K_1/(g\mu_B)$. I and II indicate the first- and the second-order transitions, respectively.

In fact, the phase transition is intimately related to the generation or annihilation of instantons and anti-instantons. As shown in equation (33), for the second solution, there are two cases: when $u > (1-\lambda)$, it is in the regime of the second-order phase, the topological current of the instantons is $\chi(M) = \sum_{n,m=0}^{N}(-1)^{m+1}|W_{(n,m)}|$; while for the case of $u < (1-\lambda)$, it is in the regime of the first-order transition and the topological number is $\chi(M) = \sum_{n,m=0}^{N}(-1)^{m}|W_{(n,m)}|$. So, we see that the instantons in the regime of the first-order phase transition are distinguished by a $e^{i\pi}$ phase shift to the instantons in the second-order transition. Since the instantons are generating or annihilating at the bifurcation line, the transformation from instanton to anti-instanoton or the reversed process may play a key role during the phase transition.

So, the two kinds of transitions are topologically distinguished. The bifurcation equation just indicates the coexistence line equation; the phase diagram bifurcated at this line and split into the first-order transition and the second-order transition. When $n = 2p$, the branch process of the two kinds of phase transitions takes place at the critical points $K_2 = K_1$ or $u = (1-\lambda)$. For the case of $n = 2p+1$, the coexistence line of the first solution is given by $\lambda = 1$, or $u = -(1-\lambda)$, it intersects the coexistence line of the second solution at $\lambda = 1$. In that case the coexistence line is degenerated to a point.

4. Conclusion

Magnetic molecular clusters of $[(\mathrm{tacn})_6\mathrm{Fe}_8\mathrm{O}_2(\mathrm{OH})_{12}]^{8+}$ are good candidates for experimental studies [3], their anisotropy constants are $K_1 = 0.33K$ and $K_2 = 0.22K$. The tunnel splitting oscillation presents different behaviours with increasing external applied magnetic field in $H/H_c < 1 - K_2/K_1$ and $H/H_c > 1 - K_2/K_1$. In the phase of $H/H_c < 1 - K_2/K_1$, the oscillation near the zero temperature has a larger monotonic region and the topological quenching happens slowly. While in the phase of $H/H_c > 1 - K_2/K_1$, the monotonic region near zero temperature becomes smaller and the topological quenching occurs more often.

When instanton and anti-instanton collide on the critical curve $H/H_c = 1 - K_2/K_1$, they annihilate. New pairs of instanton and anti-instanton are also generating on this critical curve. From our topological current of Berry curvature, one sees that the topological number of Berry curvature is the sum of topological charges of instanton and anti-instanton. It is shown that when the number of instantons equals to the number of anti-instantons, the topological charge vanishes, i.e., the total topological charges of Berry curvature $\chi = 0$. When the number of instantons and anti-instantons is not equal, the total topological charge is $\chi = 1$. The extra instantons revived the quantum tunnelling.

For two different phases, the instantons are divided into two classes which are distinguished by a $e^{i\pi}$ phase shift. The instantons are generating or annihilating on the coexistence line. When the phase transition happens, the instanton (anti-instanton) in one phase annihilates on the coexistence line and transforms into anti-instanton (instanton).

Furthermore, from the configuration of the Jacobian, we see that the speed of both instantons presents a sudden decrease or increase. Further calculations show that this configuration remains the same as we increase u and λ, this is a basic property when a phase transition happens.

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