

**Tunnel splitting and quantum phase interference in biaxial ferrimagnetic particles at excited states**Yi-Hang Nie,<sup>1,2,\*</sup> Yan-Hong Jin,<sup>3</sup> J.-Q. Liang,<sup>1</sup> and F.-C. Pu<sup>3,4</sup><sup>1</sup>*Institute of Theoretical Physics and Department of Physics, Shanxi University, Taiyuan, Shanxi 030006, China*<sup>2</sup>*Department of Physics, Yanbei Normal Institute, Datong, Shanxi 037000, China*<sup>3</sup>*Institute of Physics and Center for Condensed Matter Physics, Chinese Academy of Sciences, Beijing 100080, China*<sup>4</sup>*Department of Physics, Guangzhou Normal College, Guangzhou 510400, China*

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The tunneling splitting in biaxial ferrimagnetic particles at excited states with an explicit calculation of the prefactor of exponent is obtained in terms of periodic instantons that are responsible for tunneling at excited states and is shown as a function of magnetic field applied along an arbitrary direction in the plane of hard and medium axes. Using complex time path integral we demonstrate the oscillation of tunnel splitting with respect to the magnitude and the direction of the magnetic field due to the quantum phase interference of two tunneling paths of opposite windings. The oscillation is gradually smeared and in the end the tunnel splitting monotonously increases with the magnitude of the magnetic field when the direction of the magnetic field tends to the medium axis. The oscillation behavior is similar to the recent experimental observation with Fe<sub>8</sub> molecular clusters. A candidate of possible experiments to observe the effect of quantum phase interference in the ferrimagnetic particles is proposed.

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**I. INTRODUCTION**

The macroscopic quantum phenomenon in spin system at low temperature has attracted considerable attention both theoretically and experimentally for more than a decade.<sup>1-4</sup> The magnetization vector in a single-domain ferromagnetic (FM) grain and the Néel vector in a single-domain antiferromagnetic (AFM) grain can tunnel from a metastable state to a stable one, which is called the macroscopic quantum tunneling, or display a coherent oscillation between two degenerate states, which results in the superposition of macroscopically distinguishable (classically degenerate) states (the understanding of which is a long-standing problem in quantum mechanics) and is called macroscopic quantum coherence (MQC). The geometrical phase (known as the Berry phase) interference plays a crucial role in the MQC. The quenching of MQC can be interpreted by the quantum interference between tunneling paths of opposite windings that possess a phase with obvious geometric meaning.<sup>5-7</sup> The quenching of MQC for half-integer spin has been shown physically to be related to Kramers' degeneracy, however, the effect of geometric phase interference is far richer than that. For example, when the external magnetic field is applied along the hard anisotropy axis, a new quenching of MQC occurs and is not related to Kramers' degeneracy since the external magnetic field breaks the time-reversal symmetry.<sup>8</sup> The Zeeman energy of the biaxial spin particle associated with the external magnetic field produces an additional geometric phase of tunnel paths that leads to the quantum interference, and the tunnel splitting therefore oscillates with respect to the magnetic field. The oscillations of the level splitting for the ferromagnetic particles have been verified by the experiment with molecular clusters Fe<sub>8</sub>, which at low temperature behave like a ferromagnetic particle.<sup>9</sup> The experimental observation of the oscillation of tunnel splitting has triggered off more detailed investigations along this direction.<sup>10-12</sup> Since the tunneling rate in AFM

particles is much higher than that in FM particles of the same volume<sup>13</sup> the AFM particles are expected to be a better candidate for the observation of macroscopic quantum phenomena than the FM particles. The quantum tunneling of the Néel vector in AFM particles has been well studied in terms of the idealized sublattice model<sup>8,14</sup> in which the external magnetic field does not play a role since the net magnetic moment vanishes. The biaxial AFM particles with a small noncompensation of sublattices or in other words biaxial ferrimagnetic particles have to be considered in order to obtain the effect of the external magnetic field on the tunnel splitting. The oscillation of tunnel splitting at ground state of the biaxial ferrimagnetic particles was predicted recently with the magnetic field applied along the hard axis.<sup>15</sup> In the present paper we investigate the effect of quantum phase interference at excited states for a biaxial ferrimagnetic particle in the external magnetic field applied along an arbitrary direction in the plane of hard and medium axis. Since the effect of geometric phase interference has been observed in the experiment of Fe<sub>8</sub> molecular clusters with the magnetic field along an arbitrary direction, the present generalization to the ferrimagnetic particles is not only of theoretical but also of practical interests. At ground state one only considers the paths of imaginary time under barrier. The extension to excited states is highly nontrivial. Paths of complex time have to be taken into account since a path at excited states also approaches the region of potential well and therefore is of real time.

**II. EFFECTIVE LAGRANGIAN OF A BIAxIAL FERRIMAGNETIC PARTICLE IN A MAGNETIC FIELD**

We consider a biaxial AFM particle of two collinear FM sublattices with a small noncompensation. Assuming that the particle possesses an *X* easy axis and *X-Y* easy plane, and the magnetic field *h* is applied along an arbitrary direction in the plane of the hard axis (*Z* axis) and medium axis (*Y* axis), the

Hamiltonian operator of the AFM particle has the form

$$\hat{H} = \sum_{a=1,2} (k_{\perp} \hat{S}_a^{z2} + k_{\parallel} \hat{S}_a^{y2} - \gamma h_z \hat{S}_a^z - \gamma h_y \hat{S}_a^y) + J \hat{S}_1 \cdot \hat{S}_2, \quad (1)$$

where  $k_{\perp}, k_{\parallel} > 0$  are the anisotropy constants,  $J$  is the exchange constant,  $\gamma$  is the gyromagnetic ratio, and the spin operators in two sublattices  $\hat{S}_1$  and  $\hat{S}_2$  obey the usual commutation relation  $[\hat{S}_a^i, \hat{S}_b^j] = i\hbar \epsilon_{ijk} \delta_{ab} \hat{S}_b^k (i, j, k = x, y, z; a, b = 1, 2)$ . In order to obtain the Lagrangian of the system, we begin with the matrix element of the evolution operator in spin-coherent-state representation by means of the spin-coherent state path integrals

$$\langle N_f | e^{-2i\hat{H}T/\hbar} | N_i \rangle = \int \left[ \prod_{k=1}^{M-1} d\mu(N_k) \right] \times \left[ \prod_{k=1}^M \langle N_k | e^{-i\epsilon\hat{H}/\hbar} | N_{k-1} \rangle \right]. \quad (2)$$

Here we define  $|N\rangle = |n_1\rangle |n_2\rangle$ ,  $|N_M\rangle = |N_f\rangle = |n_{1,f}\rangle |n_{2,f}\rangle$ ,  $|N_0\rangle = |N_i\rangle = |n_{1,i}\rangle |n_{2,i}\rangle$ ,  $t_f - t_i = 2T$ , and  $\epsilon = 2T/M$ . The spin-coherent state is defined as

$$|n_a\rangle = e^{i\theta_a \hat{O}_a} |S_a, S_a\rangle, (a=1,2), \quad (3)$$

where  $n_a = (\sin \theta_a \cos \phi_a, \sin \theta_a \sin \phi_a, \cos \theta_a)$  is the unit vector,  $\hat{O}_a = \sin \phi_a \hat{S}_a^x - \cos \phi_a \hat{S}_a^y$  and  $|S_a S_a\rangle$  is the reference spin eigenstate. The measure is defined by

$$d\mu(N_k) = \prod_{a=1,2} \frac{2S_a + 1}{4\pi} \sin \theta_{a,k} d\theta_{a,k} d\phi_{a,k}. \quad (4)$$

Evaluating the path integral on the right-hand side of the Eq. (2) we obtain in the large  $S$  limit<sup>16</sup>

$$\langle N_f | e^{-2i\hat{H}T/\hbar} | N_i \rangle = \int \prod_{a=1,2} D[\theta_a] D[\phi_a] \times \exp \left[ \frac{i}{\hbar} \int_{t_i}^{t_f} (L_0 + L_1) dt \right] \quad (5)$$

with

$$L_0 = \sum_{a=1,2} S_a \dot{\phi}_a (\cos \theta_a - 1) - JS_1 S_2 [\sin \theta_1 \sin \theta_2 \cos(\phi_1 - \phi_2) + \cos \theta_1 \cos \theta_2], \quad (6)$$

$$L_1 = - \sum_{a=1,2} (k_{\perp} S_a^2 \cos^2 \theta_a + k_{\parallel} S_a^2 \sin^2 \theta_a \sin^2 \phi_a - \gamma h_z S_a \cos \theta_a - \gamma h_y S_a \sin \theta_a \sin \phi_a), \quad (7)$$

where  $L_0 + L_1$  denotes the Lagrangian. Since spins  $S_1$  and  $S_2$  in two sublattices are almost antiparallel, we may replace  $\theta_2$  and  $\phi_2$  by  $\theta_2 = \pi - \theta_1 - \epsilon_{\theta}$  and  $\phi_2 = \pi + \phi_1 + \epsilon_{\phi}$ , where  $\epsilon_{\theta}$

and  $\epsilon_{\phi}$  denote small fluctuations. Working out the fluctuation integrations over  $\epsilon_{\theta}$  and  $\epsilon_{\phi}$  the transition amplitude Eq. (5) reduces to

$$\langle N_f | e^{-2i\hat{H}T/\hbar} | N_i \rangle = \int D[\theta] D[\phi] \exp \left( \frac{i}{\hbar} \int_{t_i}^{t_f} \bar{L} dt \right), \quad (8)$$

$$\bar{L} = \Omega \left[ - \frac{M_1 + M_2}{\gamma} \dot{\phi} + \frac{M}{\gamma} \dot{\phi} \cos \theta + \frac{\chi_{\perp}}{2\gamma^2} (\dot{\theta}^2 + \dot{\phi}^2 \sin^2 \theta) \right] - V(\theta, \phi), \quad (9)$$

where  $V(\theta, \phi) = \Omega K_{\perp} (\cos \theta - M h_z / 2K_{\perp})^2 + \Omega K_{\parallel} \sin^2 \theta (\sin \phi - M h_y / 2K_{\parallel} \sin \theta)^2$ , and  $(\theta_1, \phi_1)$  has been replaced by  $(\theta, \phi)$ .  $M_a = \gamma \hbar S_a / \Omega (a=1,2)$ ,  $M = \gamma \hbar (S_1 - S_2) / \Omega$  with  $\Omega$  being the volume of the AFM particle and  $\chi_{\perp} = \gamma^2 / J$ .  $K_{\perp} = 2k_{\perp} S^2 / \Omega$  and  $K_{\parallel} = 2k_{\parallel} S^2 / \Omega$  (setting  $S_1 = S_2 = S$  except in the term containing  $S_1 - S_2$ ) denote the transverse and the longitudinal anisotropy constants, respectively.

We assume a very strong transverse anisotropy, i.e.,  $K_{\perp} \gg K_{\parallel}$ . For this case, the Néel vector is forced to lie on a cone of angle  $2\theta_0$ . Where  $\cos \theta_0 = M h_z / 2K_{\perp} = \delta h_z / h_c$  ( $\delta = K_{\parallel} / K_{\perp}$ ,  $h_c = 2K_{\parallel} / M$ ). Introducing the fluctuation variable  $\eta$  such that  $\theta = \theta_0 + \eta$  and considering  $K_{\perp} \gg K_{\parallel}$  we have  $V(\theta, \phi) = \Omega K_{\perp} \sin^2 \theta_0 \eta^2 + \Omega K_{\parallel} \sin^2 \theta_0 (\sin \phi - b)^2$  ( $b = M h_y / 2K_{\parallel} \sin \theta_0 = h \sin \alpha / \delta \sqrt{(h_c / \delta)^2 - h_z^2}$ , where  $\alpha$  is the angle between the magnetic field and Z axis) and thus the Eq. (9) is written as

$$\bar{L} = \Omega \left[ \frac{1}{2} \left( \frac{M^2}{2K_{\perp} \gamma^2} + \frac{\chi_{\perp}}{\gamma^2} \sin^2 \theta_0 \right) \dot{\phi}^2 - \frac{M_1 + M_2}{\gamma} \times \dot{\phi} + \frac{M}{\gamma} \dot{\phi} \cos \theta_0 - K_{\parallel} \sin^2 \theta_0 (\sin \phi - b)^2 \right] + \Omega \left[ \frac{\chi_{\perp}}{2\gamma^2} \dot{\eta}^2 - K_{\perp} \sin^2 \theta_0 \left( \eta + \frac{M \dot{\phi}}{2K_{\perp} \gamma \sin \theta_0} \right)^2 \right]. \quad (10)$$

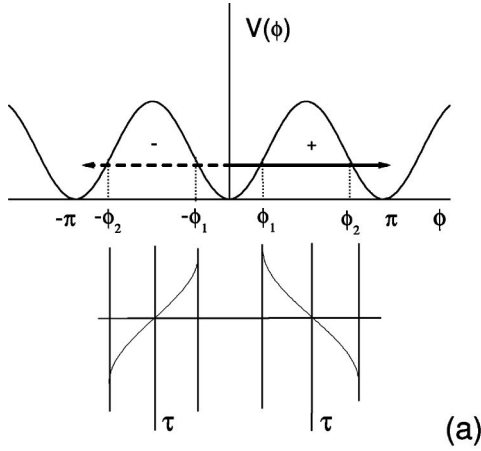
Carrying out the integral over  $\eta$  we obtain

$$\langle N_f | e^{-2\hat{H}\beta/\hbar} | N_i \rangle = \int D[\phi] \exp \left( - \frac{1}{\hbar} \int_{\tau_i}^{\tau_f} L_{eff} d\tau \right), \quad (11)$$

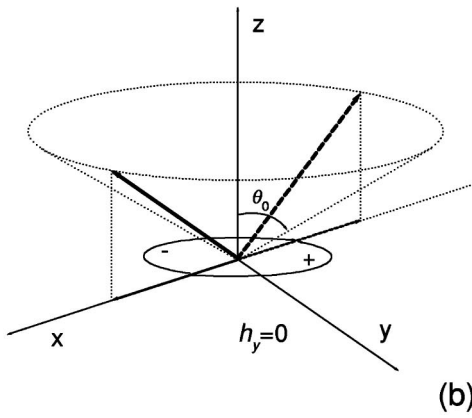
where

$$L_{eff} = \frac{I}{2} \left( \frac{d\phi}{d\tau} \right)^2 + i\Theta \frac{d\phi}{d\tau} + V(\phi) \quad (12)$$

is the effective Euclidean Lagrangian.  $\tau = it$  and  $\beta = iT$ .  $I = I_a + I_f$ , where  $I_f = \Omega M^2 / (2\gamma^2 K_{\perp})$  and  $I_a = \Omega \chi_{\perp} \sin^2 \theta_0 / \gamma^2$  are the effective FM and AFM moments of inertia,<sup>17</sup> respectively.  $V(\phi) = \Omega K_{\parallel} \sin^2 \theta_0 (\sin \phi - b)^2$  is the effective potential and  $\Theta = \hbar (S_0 - d)$  ( $S_0 = S_1 + S_2$  and  $d = h_z / h_0 = h \cos \alpha / h_0$  with  $h_0 = \hbar / \gamma I_f$ ). The second term in the Eq. (12), i.e.,  $i\Theta d\phi/d\tau$  has no effect on the classical equation of motion, however, it leads to a path-dependent phase in Euclidean action. When  $h_y = 0$ ,  $V(\phi) = K_{\parallel} \Omega \sin^2 \theta_0 \sin^2 \phi$  possesses the



(a)



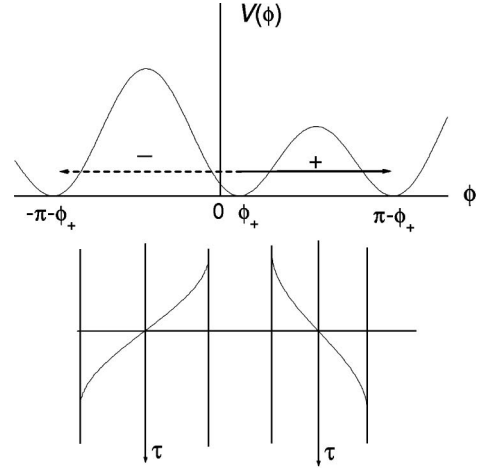
(b)

FIG. 1. (a) The periodic potential and the instanton trajectories. The arrow lines denote two tunnel paths of opposite windings. (b) The equilibrium orientations of the Néel vector in the absence of  $Y$  component of the magnetic field.

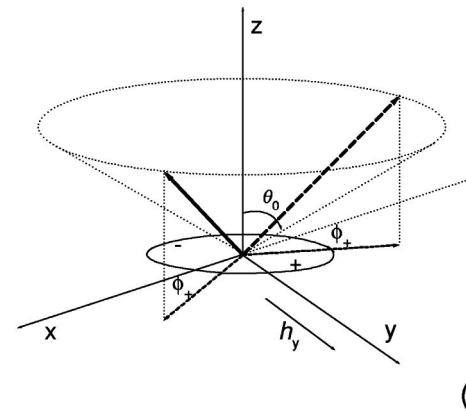
form of the sin-Gordon potential and the directions with  $\theta = \theta_0$ ,  $\phi = 0$  and  $\pi$  are two equilibrium orientations of the Néel vector [Fig. 1(b)] around which the small oscillation frequency of the Néel vector is seen to be  $\omega_0 = \sqrt{2K_{\parallel}\Omega\sin^2\theta_0}/I$ . The quantum tunneling of the Néel vector through two paths of opposite windings results in the quantum phase interference. When  $h_y \neq 0$ , the potential  $V(\phi) = \Omega K_{\parallel} \sin^2\theta_0 (\sin\phi - b)^2$  has an asymmetric twin-barrier [Fig. 2(a)], and the net magnetic moment of the uncompensated sublattices in the applied magnetic field shifts the equilibrium orientations of the Néel vector to  $\phi = \phi_+$  and  $\pi - \phi_+$  ( $\phi_+ = \arcsin b$ ) [Fig. 2(b)] around which the small oscillation frequency of the Néel vector is modified as  $\omega = \omega_0 \sqrt{1 - b^2}$ . The quantum tunneling of the Néel vector through two different barriers leads to the quantum phase interference.

### III. QUANTUM PHASE INTERFERENCE AS $H_Y = 0$

When the external magnetic field is applied along the hard axis ( $Z$  axis), the effective potential is  $V(\phi) = \Omega K_{\parallel} \sin^2\theta_0 \sin^2\phi$ . The quantum tunneling at finite energy  $E$  is dominated by the periodic instantons.<sup>18</sup> From the Euclid-



(a)



(b)

FIG. 2. (a) The potential with asymmetric twin barrier and the instanton trajectories. (b) The equilibrium orientations of the Néel vector in the presence of  $Y$  component of the magnetic field.

ean Lagrangian (12), the equation of motion of the pseudoparticles moving in the classically forbidden region in the barrier is seen to be

$$\frac{I}{2} \left( \frac{d\phi}{d\tau} \right)^2 - V(\phi) = -E. \quad (13)$$

The Néel vector may rotate by tunneling through potential barriers from one orientation ( $\phi = 0$ ) to another ( $\phi = \pi$ ) along clockwise path and anticlockwise path (Fig. 1). The instantons satisfying periodic boundary condition are found to be

$$\phi_c^{\pm} = \pm \frac{\pi}{2} \pm \arcsin[k_1 \text{sn}(\omega_0 \tau)], \quad (14)$$

where “-” denotes the clockwise path and “+” denotes the anticlockwise path (see Fig. 1),  $\text{sn}(\omega_0 \tau)$  is the Jacobian elliptic function with modulus

$$k_1 = \sqrt{1 - \frac{E}{\Omega K_{\parallel} \sin^2\theta_0}}. \quad (15)$$

The two trajectories of instantons  $\phi_c^\pm$  are shown in Fig. 1(a). The Euclidean actions evaluated along the trajectories of periodic instantons are

$$S_e^\pm = W_e + 2E\beta + i\theta_e^\pm, \quad (16)$$

$$\begin{aligned} W_e &= \int_{-\beta}^{\beta} \left[ \frac{I}{2} \left( \frac{d\phi_c^\pm}{d\tau} \right)^2 - V(\phi_c^\pm) \right] d\tau \\ &= \frac{4\Omega K_{\parallel} \sin^2 \theta_0}{\omega_0} [E(k_1) - k_1'^2 \mathcal{K}(k_1)], \end{aligned} \quad (17)$$

$$\theta_e^\pm = \int_{-\beta}^{\beta} \Theta \frac{d\phi_c^\pm}{d\tau} d\tau = \pm \Theta(\pi - 2 \arcsin k_1'), \quad (18)$$

where  $\mathcal{K}(k_1)$  and  $E(k_1)$  are the complete elliptic integrals of the first and the second kinds, respectively.  $k_1'^2 = 1 - k_1^2 = E/\Omega K_{\parallel} \sin^2 \theta_0$ . To investigate the quantum tunneling and related quantum phase interference at excited states, we begin with the instanton induced transition amplitude

$$\begin{aligned} &\sum_{m,n} \left\langle E_n^f \left| \hat{P}_E \exp\left(-\frac{2\hat{H}\beta}{\hbar}\right) \right| E_m^i \right\rangle \\ &= \int d\phi_f d\phi_i \psi_E^*(\phi_f) \psi_E(\phi_i) G(\phi_f, \beta; \phi_i, -\beta). \end{aligned} \quad (19)$$

$\hat{P}_E$  is the operator of projection onto the subspace of fixed energy.<sup>19</sup>  $|E_n^f\rangle$  and  $|E_m^i\rangle$  are two excited states lying on different sides of the barrier. From Eq. (19) the tunnel splitting is written as

$$\begin{aligned} \Delta E &\sim \frac{\exp\left(\frac{2E\beta}{\hbar}\right)}{\beta} \\ &\times \left| \int d\phi_f d\phi_i \psi_E^*(\phi_f) \psi_E(\phi_i) G(\phi_f, \beta; \phi_i, -\beta) \right|, \end{aligned} \quad (20)$$

$$G = \int \mathcal{D}[\phi] \exp\left(-\frac{1}{\hbar} \int_{-\beta}^{\beta} L_{eff} d\tau\right). \quad (21)$$

When the quantum phase interference of tunneling through clockwise and counterclockwise paths is taken into account, the Eq. (20) is written as

$$\Delta E \sim \frac{\exp\left(\frac{2E\beta}{\hbar}\right)}{\beta} |I_1^+ + I_1^-|, \quad (22)$$

$$\begin{aligned} I_1^\pm &= \int d\phi_f^\pm d\phi_i^\pm \psi_E^*(\phi_f^\pm) \psi_E(\phi_i^\pm) G(\phi_f^\pm, \beta; \phi_i^\pm, -\beta) \\ &= \exp\left(-\frac{i\theta_e^\pm}{\hbar}\right) I_0, \end{aligned} \quad (23)$$

$$I_0 = \int d\phi_f d\phi_i \psi_E^*(\phi_f) \psi_E(\phi_i) \bar{G}(\phi_f, \beta; \phi_i, -\beta), \quad (24)$$

$$\bar{G} = \int \mathcal{D}[\phi] \exp\left(-\frac{1}{\hbar} \int_{-\beta}^{\beta} \bar{L}_{eff} d\tau\right), \quad (25)$$

$$\bar{L}_{eff} = \frac{I}{2} \left( \frac{d\phi}{d\tau} \right)^2 + V(\phi). \quad (26)$$

where  $I_0$  is independent of tunnel directions. The path-independent tunneling kernel  $\bar{G}$  is now evaluated with the help of the periodic instantons. Following the procedure of the periodic instanton calculation in Refs. 20 and 21 a general formula for Eq. (24) is found to be

$$I_0 \sim 2\beta \exp\left(-\frac{2E\beta}{\hbar}\right) \left[ \frac{\hbar\omega_0}{4\mathcal{K}(k_1')} \right] \exp\left(-\frac{W_e}{\hbar}\right). \quad (27)$$

To investigate the quantum phase interference at excited state, we have to consider additional phases coming from the real-time paths in the potential well between  $0 \rightarrow \phi_1$  and  $\phi_2 \rightarrow \pi$  ( $0 \rightarrow -\phi_1$  and  $-\phi_2 \rightarrow -\pi$ ). Thus tunnel splitting Eq. (22) is rewritten as

$$\begin{aligned} \Delta E &= \frac{\exp\left(\frac{2E\beta}{\hbar}\right)}{\beta} \left| I_1^+ \exp\left(\frac{iS_r^+}{\hbar}\right) + I_1^- \exp\left(\frac{iS_r^-}{\hbar}\right) \right| \\ &= \frac{\exp\left(\frac{2E\beta}{\hbar}\right)}{\beta} I_0 \left| \exp\left[\frac{i(S_r^+ - \theta_e^+)}{\hbar}\right] + \exp\left[\frac{i(S_r^- - \theta_e^-)}{\hbar}\right] \right|, \end{aligned} \quad (28)$$

where

$$S_r^\pm = \theta_r^\pm + W_r^\pm, \quad (29)$$

$$\theta_r^\pm = -\Theta \int_{[0, \pm\phi_1] \cup [\pm\phi_2, \pm\pi]} d\phi = \mp 2\Theta \arcsin k_1', \quad (30)$$

$$W_r^\pm = \pm \sqrt{\frac{I}{2}} \int_{[0, \pm\phi_1] \cup [\pm\phi_2, \pm\pi]} \frac{E - 2V(\phi)}{\sqrt{E - V(\phi)}} d\phi. \quad (31)$$

It is obvious that  $W_r^+ = W_r^-$ . Substituting Eqs. (27), (29), and (30) into the Eq. (28), we obtain the tunnel splitting

$$\Delta E = \frac{\omega_0 \hbar}{\mathcal{K}(k_1')} \exp\left(-\frac{W_e}{\hbar}\right) |\cos(\Lambda\pi)|, \quad (32)$$

where  $\Lambda = S_0 - d$ . The tunnel splitting  $\Delta E$  is a function of the external magnetic field and energy.

For low lying excited states ( $k_1' = \sqrt{E/\Omega K_{\parallel} \sin^2 \theta_0} \ll 1$ ) in which we are interested, the energy  $E$  may be replaced by the harmonic oscillator approximated eigenvalues  $E_m = (m + \frac{1}{2})\omega_0 \hbar$ . Expanding the complete elliptic integrals  $\mathcal{K}(k_1)$

and  $E(k_1)$  as power series of  $k_1'$  and taking note of limit  $\mathcal{K}(k_1' \rightarrow 0) \rightarrow \pi/2$ , we obtain the tunnel splitting of the  $m$ th excited state,

$$\Delta E_m = \frac{(4B)^m}{m!} \Delta E_0 |\cos(\Lambda \pi)|, \quad (33)$$

where

$$\Delta E_0 = \frac{2\hbar \omega_0}{\sqrt{\pi}} (8B)^{1/2} \exp(-B), \quad (34)$$

with  $B = 4K_{\parallel} \Omega \sin^2 \theta_0 / \hbar \omega_0$  that denotes the tunnel splitting of ground state. It may be worth to estimate the range of validity of our results, i.e., how large  $m$  is.  $\Omega K_{\parallel} \sin^2 \theta_0$  is the barrier height of potential and  $\hbar \omega_0$  is the level space between neighboring levels. For the horse-spleen ferritin reported in Refs. 22 and 23 the residual spin is  $S \sim 100$  (corresponding moment  $M_0 = 217 \mu_B$ ) and volume is  $\Omega \sim 2 \times 10^{-19} \text{ cm}^3$  (diameter 7.5 nm). The longitudinal anisotropy constant and transverse susceptibility are seen to be  $K_{\parallel} = 2 \times 10^5 \text{ erg/cm}^3$  and  $\chi_{\perp} = 10^{-5} \text{ emu/G cm}^3$ , respectively. Using the above parameters we find that the number of the levels in the potential well is about ten as  $\delta \sim 0.03$ . Fig. 3(a) shows the oscillation of tunnel splittings of the lowest three states with respect to the external magnetic field due to the quantum phase interference of two tunneling paths of opposite windings for  $S_0 = \text{integer}$  and halfinteger. From Fig. 3(a) one can find that the magnitude of tunnel splittings at excited states is much higher than that at ground state and may contribute significantly to the experimental observation at finite temperature. When  $d = S_0 - l - \frac{1}{2}$ , i.e.,  $h = (S_0 - l - \frac{1}{2}) h_0$  ( $l$  is an integer), the tunneling splitting  $\Delta E_m$  vanishes. The period of oscillation is

$$\Delta h = \frac{\hbar}{\gamma I_f}, \quad (35)$$

which is independent of the energy.

#### IV. QUANTUM PHASE INTERFERENCE AS $H_Y \neq 0$

When the external magnetic field is applied along an arbitrary direction in the plane of the hard axis and medium axis, the effective potential  $V(\phi) = \Omega K_{\parallel} \sin^2 \theta_0 (\sin \phi - b)^2$  has the asymmetric twin barrier that leads to that Néel vector may rotate from one orientation ( $\phi = \phi_+$ ) to another ( $\phi = \pi - \phi_+$ ) along clockwise underbarrier path and counterclockwise path (Fig. 2). Two different instantons (Fig. 2) corresponding to tunneling through two types of barriers are found as

$$\phi_c^{\pm} = \pm \frac{\pi}{2} \pm 2 \arctan[\lambda_{\pm} \text{sn}(q\tau, k_2)], \quad (36)$$

where

$$k_2 = \left[ \frac{(1-\varepsilon)^2 - b^2}{(1+\varepsilon)^2 - b^2} \right]^{1/2}, \quad \varepsilon = \sqrt{\frac{E}{\Omega K_{\parallel} \sin^2 \theta_0}},$$

$$q = \frac{\omega_0}{2} [(1+\varepsilon)^2 - b^2]^{1/2}, \quad \lambda_{\pm} = \left[ \frac{(1-\varepsilon)^2 - b^2}{(1\pm b)^2 - \varepsilon^2} \right]^{1/2}.$$

Our starting point for investigation of the tunneling and related quantum phase interference at excited states is still the transition amplitude of the barrier penetration projected onto the subspace of fixed energy  $E$ , i.e., the Eq. (19) from which the tunneling splitting is obtained as

$$\Delta E \sim \frac{\exp\left(\frac{2E\beta}{\hbar}\right)}{\beta} \times \left| \int d\phi_f d\phi_i \psi_E^*(\phi_f) \psi_E(\phi_i) G(\phi_f, \beta; \phi_i, -\beta) \right|. \quad (37)$$

The result corresponding to the Eq. (22) is formally the same,

$$\Delta E \sim \frac{\exp\left(\frac{2E\beta}{\hbar}\right)}{\beta} |I_2^+ + I_2^-|. \quad (38)$$

In the present case, however, the two tunneling paths are not symmetric. Thus we find

$$I_2^{\pm} = \exp\left(-\frac{i\delta_e^{\pm}}{\hbar}\right) \bar{I}_2^{\pm}, \quad (39)$$

$$\delta_e^{\pm} = \pm \Theta[\pi \mp 2 \arcsin(b \pm \varepsilon)], \quad (40)$$

$$\bar{I}_2^{\pm} = \int d\phi_f^{\pm} d\phi_i^{\pm} \psi_E^*(\phi_f^{\pm}) \psi_E(\phi_i^{\pm}) \bar{G}^{\pm}(\phi_f^{\pm}, \beta; \phi_i^{\pm}, -\beta), \quad (41)$$

$$\bar{G}^{\pm} = \int \mathcal{D}[\phi] \exp\left(-\frac{1}{\hbar} \int_{-\beta}^{\beta} \bar{L}_{eff}^{\pm} d\tau\right), \quad (42)$$

$$\bar{L}_{eff}^{\pm} = \frac{I}{2} \left( \frac{d\phi_c^{\pm}}{d\tau} \right)^2 + V(\phi_c^{\pm}). \quad (43)$$

$\bar{I}_2^{\pm}$  is now dependent on tunnel direction. The path-dependent tunneling kernel  $\bar{G}^{\pm}$  is evaluated with the help of the periodic instanton. Following the above procedure we obtain

$$\bar{I}_2^{\pm} \sim 2\beta \exp\left(-\frac{2E\beta}{\hbar}\right) \left[ \frac{\hbar \omega_0}{4\sigma \mathcal{K}(k_2')} \right] \exp\left(-\frac{W_e^{\pm}}{\hbar}\right), \quad (44)$$

$$\sigma = [(1+\varepsilon)^2 - b^2]^{-1/2}, \quad k_2' = \sqrt{1 - k_2^2},$$

$$W_e^{\pm} = \frac{4Iq}{\lambda_{\pm}^2} [\lambda_{\pm}^2 E(k_2) + (k_2^2 - \lambda_{\pm}^2) \mathcal{K}(k_2) + (\lambda_{\pm}^4 - k_2^2) \Pi(k_2, \lambda_{\pm}^2)], \quad (45)$$



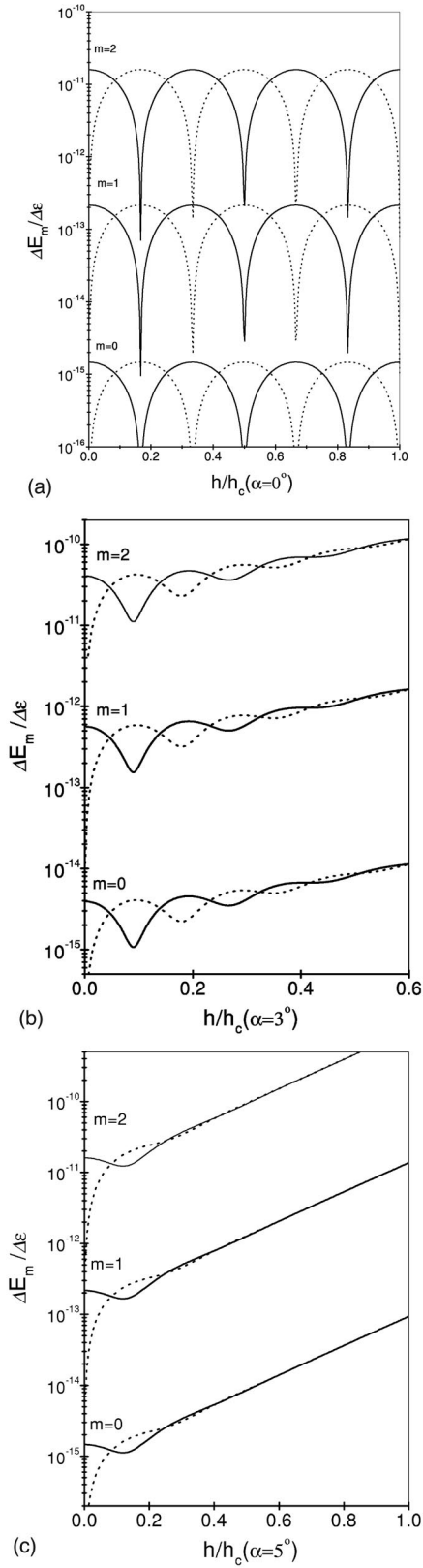


FIG. 3. The level splitting as function of the external magnetic field with angular ( $\Delta\varepsilon = 2\hbar\omega_0/\sqrt{\pi}$ ) (a)  $\alpha=0^\circ$ , (b)  $\alpha=3^\circ$ , and (c)  $\alpha=5^\circ$  for  $S_0 =$  integer (solid line) and  $S_0 =$  half integer (dot line). Here  $S=100$ ,  $\Omega=10^{-19}$  cm<sup>3</sup>,  $\chi_\perp=10^{-5}$ ,  $K_\parallel=10^5$  erg/cm<sup>3</sup> and  $\delta\sim 0.03$ .

where  $\Pi(k_2, \lambda_\pm^2)$  is the complete elliptic integral of the third kind. Considering the additional phase contribution from the real-time paths in potential well the tunnel splitting Eq. (38) is written as

$$\Delta E = \frac{\exp\left(\frac{2E\beta}{\hbar}\right)}{\beta} \left| I_2^+ \exp\left(\frac{iS_r^+}{\hbar}\right) + I_2^- \exp\left(\frac{iS_r^-}{\hbar}\right) \right|, \quad (46)$$

where

$$S_r^\pm = \delta_r^\pm + \Phi_r^\pm, \quad (47)$$

$$\delta_r^\pm = -2\Theta[\arcsin(b \pm \varepsilon) - \arcsin b], \quad (48)$$

$$\begin{aligned} \Phi_r^\pm = 2I\omega_0 \left[ \frac{E(\varphi^\pm, k_2')}{\sigma} - \sigma(1 \mp b)^2 F(\varphi^\pm, k_2') \right. \\ \left. \mp 2b\sigma(1 \mp b - \varepsilon) \Pi(\varphi^\pm, \alpha^\pm, k_2') - \varepsilon \sqrt{\frac{1 \pm b}{1 \mp b}} \right], \end{aligned} \quad (49)$$

$$\varphi^\pm = \arcsin \sqrt{\frac{1 \mp b + \varepsilon}{2(1 \mp b)}}, \quad \alpha^\pm = \sqrt{\frac{2\varepsilon}{1 \mp b + \varepsilon}},$$

where  $F(\varphi^\pm, k_2')$ ,  $E(\varphi^\pm, k_2')$  and  $\Pi(\varphi^\pm, \alpha^\pm, k_2')$  are the incomplete elliptic integrals of the first, the second, and the third kinds, respectively. Inserting Eqs. (39), (44), and (47) into Eq. (46), we obtain the final formula of the tunnel splitting

$$\begin{aligned} \Delta E = \frac{\hbar\omega_0}{2\sigma\mathcal{K}(k_2')} \left\{ \exp\left(-\frac{2W_e^+}{\hbar}\right) + \exp\left(-\frac{2W_e^-}{\hbar}\right) \right. \\ \left. + 2\exp\left(-\frac{W_e^+ + W_e^-}{\hbar}\right) \cos[2\Lambda\pi - (\Phi_r^+ - \Phi_r^-)] \right\}^{1/2}, \end{aligned} \quad (50)$$

which is a function of the external magnetic field and the energy. For low-lying excited states,  $\varepsilon \ll 1, k_2' \ll 1$ , the energy  $E$  is again replaced by harmonic oscillator approximated eigenvalues  $E_m = (m + \frac{1}{2})\hbar\omega$ . Expanding the complete elliptic integrals  $E(k_2)$ ,  $\mathcal{K}(k_2)$ , and  $\Pi(k_2, \lambda_\pm^2)$  in the Eq. (45) as power series of  $k_2'$  we obtain

$$\begin{aligned} W_e^\pm = \frac{4\Omega K_\parallel \sin^2 \theta_0}{\omega_0} \left[ \sqrt{1-b^2} - \frac{1}{16}(1-b^2)^{3/2} k_2'^4 \left( \ln \frac{4}{k_2'} + \frac{1}{4} \right) \right. \\ \left. + b \arcsin b \mp \frac{\pi}{2} \right]. \end{aligned} \quad (51)$$

Substituting the Eq. (51) into the Eq. (50) and taking note of limits  $\mathcal{K}(k_2' \rightarrow 0) \rightarrow \pi/2$ ,  $\sigma(k_2' \rightarrow 0) \rightarrow (1-b^2)^{1/2}$  and  $\Phi_r^+ \approx \Phi_r^-$  at low lying excited states we obtain the tunnel splitting of the  $m$ th excited state as

$$\Delta E_m = \frac{E_2}{m!} [4B(1-b^2)^{3/2}]^m [\cosh(bB\pi) + \cos(2\Lambda\pi)]^{1/2}, \quad (52)$$

$$E_2 = \frac{2\hbar\omega_0}{\sqrt{\pi}} [4B(1-b^2)^{5/2}]^{1/2} \exp[-B(\sqrt{1-b^2} + b \arcsin b)]. \quad (53)$$

Figure 3 shows the oscillation of tunnel splitting at low-lying excited states with respect to the external magnetic field for  $S_0 = \text{integer}$  and half integer, respectively. When  $\Lambda = (2l + 1)/2$  ( $l$  is an integer), tunnel splitting  $\Delta E_m$  tends to a minimum value. The period of oscillation is

$$\Delta h = \frac{h_0}{\cos \alpha}, \quad (54)$$

which is independent of the level, but dependent on the direction of the external magnetic field. When  $\alpha = 0$  and  $m = 0$ , the tunnel splitting  $\Delta E_m$  reduces to the result in Ref. 15. The period increases with the angle  $\alpha$ . When the direction of the magnetic field is along the medium axis ( $\alpha = \pi/2$ ), the

period approaches to infinity, in other words, the oscillation disappears.

## V. CONCLUSION

The effect of the macroscopic quantum phase interference at excited states is studied for the biaxial ferrimagnetic particles with the external magnetic field applied along an arbitrary direction in the plane of hard and medium axis. We present a general formula of tunnel splitting at excited states as a function of the magnetic field and the energy. The oscillation behavior of tunneling splitting at low-lying excited states is similar to that in FM particles observed experimentally in molecular clusters  $\text{Fe}_8$  and should be observed in further experiment with ferrimagnetic particles for which a possible candidate of materials may be horse-spleen ferritin.<sup>22,23</sup>

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