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LETTER TO THE EDITOR

Macroscopic quantum phase interference in antiferromagnetic particles

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Abstract. The tunnel splitting in biaxial antiferromagnetic particles is studied with a magnetic field applied along the hard anisotropy axis. We observe the oscillation of tunnel splitting as a function of the magnetic field due to the quantum phase interference of two tunnelling paths of opposite windings. The oscillation is similar to the recent experimental result with Fe_8 molecular clusters.

The macroscopic quantum phenomenon of magnetic particles at low temperature has attracted considerable attention both theoretically and experimentally in recent years [1–3]. The magnetization vector in solids is traditionally viewed as a classical variable. The quantum transition of the magnetization vector $\boldsymbol{M}$ between different easy directions in a single domain ferromagnetic (FM) grain, in particular the coherent tunnelling between two degenerate orientations of the magnetization called the macroscopic quantum coherence (MQC) [4], has been studied extensively for its exotic characters far from that of classical system. Quenching of MQC for half-integer spin is a fascinating effect [5–8] and can be used to test the macroscopic quantum tunnelling experimentally. The quenching of MQC in spin particles is analysed with the help of the phase interference of spin coherent state-paths which possess a phase with an obvious geometric meaning [5]. Although the quenching of MQC has been interpreted as Kramers' degeneracy, the effect of geometric phase interference is far richer than that. By investigating the quantum tunnelling in biaxial ferromagnetic particles with a magnetic field applied along the hard axis, Garg [9] found a new quenching of tunnelling splitting which is not related to Kramers' degeneracy since the external field breaks the time reversal symmetry. The Zeeman energy of the biaxial spin particle in the external magnetic field results in additional topological phases of the tunnel paths which lead to the quantum phase interference. The tunnelling splitting therefore oscillates with respect to the magnetic field.

According to a recent report [10] the oscillation of tunnelling splitting was observed experimentally in molecular clusters Fe_8 which at low temperature behave like a nanomagnet, namely, a ferromagnetic particle. A more detailed analysis of quantum phase interference with instanton method in the context of spin coherent-state-path-integrals has been given recently [11]. In this letter we investigate the similar effect of quantum phase interference in antiferromagnetic (AFM) particles. Since the tunnelling rate in AFM particles is much higher than that in FM particles of the same volume [12], the AFM particles are expected to be a better candidate for the observation of MQP than the FM particles [12]. The AFM particle is usually described by the Néel vector of the two collinear sublattices whose magnetizations are coupled by strong exchange interaction. The external magnetic field does not play a role since the net magnetic moment vanishes for idealized sublattices. The quantum and classical transitions of the Néel vector in antiferromagnets have been well studied [13] in terms of the idealized sublattice model. The temperature dependence of quantum tunnelling was also given for the same model [14] and the theoretical result agrees with the experimental observation [15]. A biaxial AFM particle with a small non-compensation of sublattices in the absence of an external magnetic field was studied in [16] where it was shown that the uncompensated magnetic moment leads to a modification of the oscillation frequency around the equilibrium orientations of the Néel vector. In this letter we demonstrate that the uncompensated magnetic moment of a small biaxial AFM particle possesses a Zeeman energy in an external magnetic field applied along hard axis and thus a topological phase depending on the magnetic field is introduced similarly to the phase in ferromagnetic particles [9]. The quantum phase interference results in the oscillation of tunnelling splitting as a function of magnetic field which may be regarded as a kind of Aharonov–Bohm effect.

We consider in the following a biaxial AFM particle of two collinear FM sublattices with a small non-compensation. Assuming that the particle possesses a X easy axis and XY easy plane , and the magnetic field h is applied along the hard axis (Z axis), the Hamiltonian operator of the AFM particle has the form

$$\hat{H} = \sum_{a=1,2} \left(k_\perp \hat{S}_a^{z2} + k_{\shortparallel} \hat{S}_a^{y2} - \gamma h \hat{S}_a^z \right) + J \hat{S}_1 \hat{S}_2 \qquad (1)$$

where $k_\perp, k_{\shortparallel} > 0$ are the anisotropy constants, J is the exchange constant, γ is the gyromagnetic ratio, and the spin operators in two sublattices $\hat{S}_1$ and $\hat{S}_2$ obey the usual commutation relation $[\hat{S}_a^i, \hat{S}_b^j] = i\hbar\epsilon_{ijk}\delta_{ab}\hat{S}_b^k$ $(i, j, k = x, y, z; a, b = 1, 2)$. The matrix element of the evolution operator in spin coherent-state representation is

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

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

$$|n_a\rangle = \mathrm{e}^{-\mathrm{i}\theta_a \hat{C}} |S_a, S_a\rangle \qquad (a = 1, 2) \qquad (3)$$

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

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

In the large S limit we obtain [17]

$$\langle N_f | \mathrm{e}^{-2\mathrm{i}\hat{H}T/\hbar} | N_i \rangle = \mathrm{e}^{-\mathrm{i}S_0(\phi_f - \phi_i)} \int \prod a = 1, 2 D[\theta_a] D[\phi_a] \exp\left(\frac{i}{\hbar} \int_{t_i}^{t_f} L \mathrm{d}t \right). \qquad (5)$$

The Lagrangian is defined by $L = L_0 + L_1$ with

$$L_0 = \sum a = 1, 2 S_a \dot{\phi}_a \cos\theta_a \; - J S_1 S_2 \left[\sin\theta_1 \cos\theta_2 \cos(\phi_1 - \phi_2) + \cos\theta_1 \cos\theta_2 \right] \qquad (6)$$

$$L_1 = \sum a = 1, 2 \left(k_\perp S_a^2 \cos^2\theta + k_{\shortparallel} S_a^2 \sin^2\theta_a \sin^2\phi_a - \gamma h S_a \cos\theta_a\right) \tag{7}$$

where $S_0 = S_1 + S_2$ is total spin of two sublattices. Since S_1 and S_2 is almost antiparallel, we may replace θ_2 and ϕ_2 by $\theta_2 = \pi - \theta_1 - \epsilon_\theta$ and $\phi_2 = \pi + \phi_1 + \epsilon_\phi$, where ϵ_θ and ϵ_ϕ denote small fluctuations. Working out the fluctuation integrations over ϵ_θ and ϵ_ϕ the transition amplitude (5) reduces to

$$\langle N_f|\mathrm{e}^{-2\mathrm{i}\hat{H}T/\hbar}|N_i\rangle = \mathrm{e}^{-\mathrm{i}S_0(\phi_f - \phi_i)} \int \prod a = 1, 2 D[\theta] D[\phi] \exp\left(\frac{i}{\hbar}\int_{t_i}^{t_f} \bar{L}\mathrm{d}t\right) \tag{8}$$

$$\bar{L} = \Omega\left[\frac{m}{\gamma}\dot{\phi}\cos\theta + \frac{\chi_\perp}{\gamma} H\dot{\phi}\sin^2\theta + \frac{\chi}{2\gamma^2}\left(\dot{\theta}^2 + \dot{\phi}^2\sin^2\theta\right)\right] - V(\theta, \phi) \tag{9}$$

where $V(\theta, \phi) = \left(\Omega K_\perp \cos^2\theta + K_{\shortparallel} \sin^2\theta \sin^2\phi - mh\cos\theta - \frac{\chi_\perp}{2}h^2\sin^2\theta\right)$,and (θ_1, ϕ_1) has been replaced by (θ, ϕ). $m = \gamma\hbar(S_1 - S_2)/\Omega$ with Ω being the volume of the AFM particle and $\chi_\perp = \frac{\gamma^2}{J}$. $K_\perp = 2k_\perp S^2/\Omega$ and $K_{\shortparallel} = 2k_{\shortparallel} S^2/\Omega$ (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. $K_\perp \gg K_{\shortparallel}$. In this case the Néel vector is forced to lie near the XY plane. Replacing θ by $\frac{\pi}{2} + \eta$ where η denotes the small fluctuation and carrying out the integral over η we obtain

$$\langle N_f|\mathrm{e}^{-2\hat{H}\beta/\hbar}|N_i\rangle = \int D[\phi] \exp\left(-\frac{1}{\hbar}\int_{\tau_i}^{\tau_f} L_{eff}\mathrm{d}\tau\right) \tag{10}$$

where

$$L_{eff} = \frac{I}{2}\left(\frac{\mathrm{d}\phi}{\mathrm{d}\tau}\right)^2 + i\Theta\frac{d\phi}{d\tau} + V(\phi) \tag{11}$$

is the effective Euclidean Lagrangian. $\tau = it$ and $\beta = iT$. $I = \Omega\left(I_a + I_f\right)$ where $I_a = m^2/\left(2\gamma^2 K_\perp\right)$ and $I_f = \chi_\perp/\gamma^2$ are the effective FM and AFM moments of inertia [18], respectively. $V(\phi) = \Omega K_{\shortparallel} \sin^2\phi$ is the effective potential and $\Theta = \hbar S_0 - I\gamma h$. The second term in (11), i.e. $i\Theta(\mathrm{d}\phi/\mathrm{d}\tau)$ which is the total time derivative has no effect on the classical equation of motion, however it leads to a path dependent phase in Euclidean action. The classical equation of motion is seen to be

$$\frac{I}{2}\left(\frac{\mathrm{d}\phi}{\mathrm{d}\tau}\right)^2 - V(\phi) = 0 \tag{12}$$

$\phi = 0$ and π are two equilibrium orientation of the Néel vector. The Néel vector may transit by tunnelling through potential barriers from one orientation ($\phi = 0$) to another ($\phi = \pi$) along clockwise or anticlockwise paths. The instanton solutions of (12) are then obtained as

$$\phi_c^\pm(\tau) = \pm 2\arctan\left(\mathrm{e}^{\omega_0\tau}\right) \tag{13}$$

where $\omega_0 = \sqrt{2K_{\shortparallel}\Omega/I}$ is the small oscillation frequency of the Néel vector around its equilibrium orientation. $\phi_c^-(\tau)$ and $\phi_c^+(\tau)$ denote instanton solutions with clockwise and anticlockwise windings respectively. The Euclidean actions evaluated along the instanton trajectories are seen to be

$$S_E^\pm = \int L_{eff}\mathrm{d}\tau = 2I\omega_0 \pm \Theta\pi. \tag{14}$$

The quantum phase interference of clockwise path '−' and anticlockwise path '+' is seen to be (see figure 1)

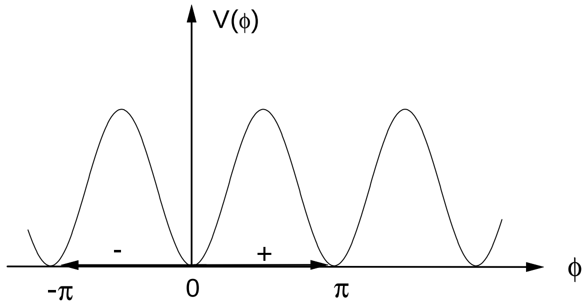

Figure 1. Quantum phase interference of two tunnel paths of opposite windings.

$$\mathrm{e}^{-S_E^+} + \mathrm{e}^{-S_E^-} \sim e^{-2I\omega_0/\hbar} \cos\left(\Lambda\pi\right) \tag{15}$$

where $\Lambda = \frac{\Theta}{\hbar} = S_0 + \frac{h}{h_c}$ with $h_c = \frac{\hbar}{\gamma I}$. Since the potential $V(\phi)$ is periodic and can be regarded as a one-dimensional superlattice. Using the Bloch theory the low-lying energy spectrum could be determined as [19]

$$E_0 = \varepsilon_0 - 2\Delta\varepsilon_0 \cos\pi\left(\Lambda + \xi\right). \tag{16}$$

Where ξ is Bloch wave vector which can be assumed to take either of the two values 0 and 1 [20]. $\Delta\varepsilon_0 = (2\hbar\omega_0 \ddot{y}/\pi\,\mathrm{e}^{-2I\omega_0/\hbar}$ is the usual overlap integral or simply the level shift induced by tunnelling through any one of the barriers. Thus the tunnelling splitting is seen to be

$$\Delta\varepsilon = |2\Delta\varepsilon_0 \cos\pi\left(\Lambda + 1\right) - 2\Delta\varepsilon_0 \cos\pi\Lambda| = 4\Delta\varepsilon_0 \left|\cos\pi\Lambda\right| \tag{17}$$

which is a function of the external magnetic field like in the ferromagnetic particle case [9–11]. When $h = 0$, (17) reduces to the previous result [16] where the tunnelling splitting is quenched when $S_0 = \mathrm{half} - \mathrm{integer}$. With nonzero magnetic field the tunnelling splitting would be quenched whenever $\Lambda = n + \frac{1}{2}$ or $h = \left(S_0 - n - \frac{1}{2}\right)\hbar/I\gamma$ where n is an integer. Figure 2 shows the oscillation of the tunnelling splitting with respect to the external magnetic field. This quenching is due to the quantum phase interference of two tunnelling paths of opposite windings. The period of oscillation is given by

$$\Delta h = \frac{\hbar}{I\gamma}. \tag{18}$$

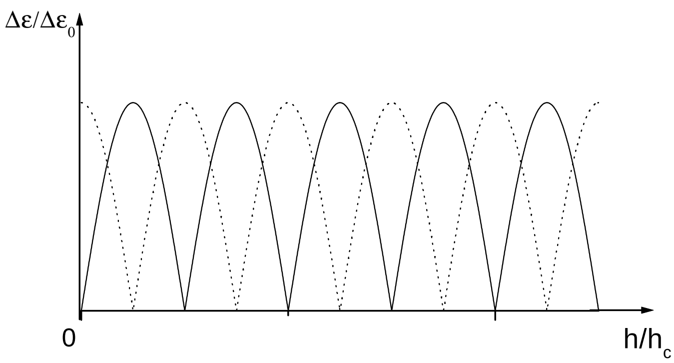

Figure 2. Oscillation of tunnelling splitting as a function of the external field with the solid line for $S_0 = $ half-integer and the dotted line for $S_0 = $ integer.

We have demonstrated a macroscopic quantum interference effect in the tunnelling of the magnetization of antiferromagnetic particles. Such particles thus open a new avenue to test macroscopic quantum interference effects. Experimental tests of our prediction could therefore make an important contribution to our understanding of the transition region between the microscopic and the macroscopic world.

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