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Quantum oscillations in small magnetic particles

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Quantum oscillations in small magnetic particles, induced by external magnetic field, are considered. It is shown that, due to tunneling of the total magnetization M_0, the ground-state energy of the system oscillates as a function of the external-magnetic-field strength with period $\delta H=(\mu/M_0)H_c$ (μ is the magnetic moment; H_c is the coercive field). In small anisotropic antiferromagnetic particles, the magnetic field enhances the Néel-vector tunneling rate and generates oscillations in the soliton rest energy.

I. INTRODUCTION

Magnetic properties of single-domain samples of mesoscopic size have received widespread attention by theorists as well as by experimentalists for a long time (see, e.g., Ref. 1). One of the most interesting features of these systems is the existence of specific demagnetization processes due to thermal or quantum fluctuations of the total magnetization of the sample. At low temperatures, demagnetization is due to the macroscopic quantum tunneling (MQT) of spins. This problem has been studied carefully with use of various approaches.[2–4] For antiferromagnets with "easy-axis" anisotropy, the probability of MQT has been derived within the semiclassical continuum model.[5,6]

In considering macroscopic quantum tunneling, one usually distinguishes between two types of processes: quantum decay and macroscopic quantum coherence. In the first case, it is often assumed that the system initially occupies one of the metastable vacua; the tunneling from the "false" vacuum to the ground state is calculated under the assumption of either quantum nucleation of a new phase domain or homogeneous subbarrier transition (for mesoscopic-sized samples). In the latter case, it is sufficient to calculate the imaginary part of the metastable vacuum energy.

In the phenomenon of macroscopic quantum coherence, tunneling is a multiple-step process. Consequently, the object of the calculation is not the tunneling probability itself but rather the dependence of different properties of the system upon this probability. When studying the macroscopic quantum processes, one usually selects a weakly fluctuating collective degree of freedom, assuming its dynamics can be described quantum mechanically. In this case, the collective quantum dynamics of the macrosystem incorporates different oscillation effects, in complete analogy with conventional quantum-mechanical calculations. The amplitude of these quantum oscillations depends essentially on the size of the system and on the parameters destroying the phase coherence (temperature, dissipation, etc.).

One manifestation of the macroscopic quantum coherence is the effect of the Θ vacuum[7] originating from the oscillating dependence of thermodynamic properties of the system on the "vacuum angle" θ_u (θ_u is the factor with which the total time derivative enters the Lagrangian). In solid-state physics, the parameter θ_v depends on external fields and the "vacuum oscillations" can, in principle, be detected experimentally.

For example, for the Aharonov-Bohm problem in conductors with charge-density waves,[7] θ_v is the normalized magnetic flux. Θ effects have also been discussed for Josephon junctions of mesoscopic sizes.[8] In this case, the vacuum angle depends on the voltage applied to the junction.

The purpose of this paper is to study quantum oscillations in small magnetic particles. We considered a simple semiclassical model of a small-sized anisotropic ferromagnetic in the external magnetic field and showed the existence of quantum oscillations as a function of field strength. For antiferromagnets, Θ-vacuum effects provide oscillatory corrections to the rest energy of topological solitons and stimulate transitions with the flipping over of the Néel vector.

II. QUANTUM OSCILLATIONS VS MAGNETIC FIELD IN SMALL MAGNETIC PARTICLES

Following Ref. 2, we consider a simple semiclassical model of a ferromagnetic particle. The dynamic variables are the components of the total magnetization

$$\mathbf{M}(t)=M_0(\sin\vartheta\sin\Phi,\ \sin\vartheta\cos\Phi,\ \cos\vartheta)\ . \quad (1)$$

In terms of angle variables $\vartheta(t)$ and $\Phi(t)$, the action of the system takes the form (see, e.g., Ref. 2)

$$S=\int dt\left[\frac{M_0}{\mu}\dot{\Phi}\cos\vartheta-E(\vartheta,\Phi)\right]\ , \quad (2)$$

where $E(\vartheta,\Phi)$ is the energy of the particle and μ is the magnetic moment on a site.

Let us consider at first a ferromagnet with "easy-axis" anisotropy in an external magnetic field which is orthogonal to the easy-magnetization plane (along the z axis). Then

$$E(\vartheta,\Phi)=K(\cos\vartheta-H/H_c)^2\ , \quad (3)$$

where K is the energy of magnetic anisotropy and $H_c=2K/M_0$ is the coercive field. The classical ground state of the system is described by the magnetization vector on the "easy cone" $[\theta=\arccos(H/H_c)]$ with the arbitrary value of the azimuthal angle $\Phi=\text{const}$.

Note, however, that the projection of $\mathbf{M}$ on the easy plane is not conserved, and so quantum fluctuations delocalize the azimuthal angular "motion" of the system. Let us derive the shift of the ground-state energy caused by this delocalization. We will be interested not in the total-energy shift, but rather only in the oscillating part of the ground-state energy. For this purpose it is convenient, using the equations of motion, to rewrite the Lagrangian in terms of the azimuthal collective variable,

$$L=\frac{I_{\text{eff}}}{2}\dot{\Phi}^2+\frac{\theta_v}{2\pi}\dot{\Phi}\ , \tag{4}$$

where the effective moment of inertia I_{eff} and the vacuum angle θ_v are determined as follows:

$$I_{\text{eff}}=\left[\frac{M_0}{\mu}\right]^2\frac{1}{2K}\ ,\quad \theta_v=2\pi\frac{M_0}{\mu}\frac{H}{H_c}\ . \tag{5}$$

The Lagrangian (4) describes the quantum dynamics of a particle on a circle and it has been repeatedly studied in the past (see, e.g., Refs. 7, 9, and 10). We use the expression for the oscillating part of the free energy $F(T)$ obtained in Ref. 10:

$$\delta F_{\text{osc}}\simeq\begin{cases}-2T\exp(-2\pi^2 I_{\text{eff}}T)\cos\theta_v\ , & TI_{\text{eff}}\geq 1\ , \quad \text{(6a)}\\ \dfrac{1}{2I_{\text{eff}}}[\min\{(\theta_v/2\pi-\lfloor\theta_v/2\pi\rfloor)\ ,\ (\lceil\theta_v/2\pi\rceil-\theta_v/2\pi)\}]^2\ , & TI_{\text{eff}}\ll 1\ , \quad \text{(6b)}\end{cases}$$

where T is the temperature, $\lfloor x\rfloor$ is the floor function of x and $\lceil x\rceil$ is the ceiling function of x, and so the quantity enclosed in square brackets in Eq. (6b) is the difference between x and the nearest integer. According to Eqs. (5) and (6) the external magnetic field induces quantum oscillations in small anisotropic ferromagnetic particles. The period of oscillations is

$$\delta H=\frac{\mu}{M_0}H_c=2\frac{\mu}{M_0}\frac{K}{M_0} \tag{7}$$

and can be physically interpreted as follows. It is convenient to regard such an anisotropic magnetic particle as a planar rotator with characteristic frequency $\omega_c=\mu H_c$ and a discrete energy spectrum. Then the force-free influence of the external magnetic field results in the shifting ($\Delta E_H=M_0H$) of the rotator-energy levels. At definite values of the magnetic field, when $\Delta E_H=n\omega_c$, the shifted spectrum coincides with the initial one ($H=0$) and thus all characteristics of the system are oscillating functions of $\Delta E_H/\omega_c$. The amplitude of these mesoscopic oscillations decreases exponentially with increasing temperature, in accordance with general properties of quantum coherent phenomena.

The macroscopic quantum coherence in our case manifests itself by the oscillations of the magnetization projection on the magnetic-field sense (see, e.g., Ref. 10)

$$\delta M_z^{\text{osc}}=\frac{\partial F}{\partial H}=-\frac{T}{\delta H}\frac{\vartheta_3'(u,q)}{\vartheta_3(u,q)}\ , \tag{8}$$

where $\vartheta_3(u,q)$ is the Jacobi θ function,

$$u=\frac{H}{\delta H}\ ,\quad q=\exp\left\{-\pi^2\frac{T}{K}\left[\frac{M_0}{\mu}\right]^2\right\}. \tag{9}$$

In terms of the model initial parameters the oscillating part of the magnetization at high temperature $T\geq K(\mu/M_0)^2$ takes the form

$$\delta M_z^{\text{osc}}(H)=T\frac{M_0^2}{\mu K}\exp\left\{-\pi^2\frac{T}{K}\left[\frac{M_0}{\mu}\right]^2\right\}\sin\left[2\frac{\mu KH}{M_0^2}\right]. \tag{10}$$

The characteristic value of the amplitude of oscillations is of the order of μ and thus the precise measurements with the atomic level accuracy are needed to detect the oscillations.

Let us now consider a ferromagnet, in the external magnetic field, which has an easy anisotropy axis (along the x axis) in the easy plane, Ref. 11. Then the energy of the system is

$$E(\vartheta,\Phi)=K_1(\cos\vartheta-\cos\theta_0)^2+K_2\sin^2\vartheta\sin^2\Phi\ , \tag{11}$$

where $K_{1,2}$ (with $K_1>K_2$) are the energies of magnetic anisotropy, $\cos\theta_0=H/H_c$. In zero magnetic field, model (11) has been studied in Ref. 2.

Classical vacua of the system can be described by the two different orientations of the magnetization vector on the "easy cone," namely, $(\vartheta=\theta_0,\ \Phi=0)$ and $(\vartheta=\theta_0,\ \Phi=\pi)$. These vacua are separated by a potential barrier which is finite for a small-sized particle. Due to tunneling, the mean value of the magnetization projection on the "easy axis" vanishes, $\langle M_x\rangle=0$. In Refs. 11 and 2, the probability of the subbarrier transition $|\Phi=0\rangle\Longrightarrow|\Phi=\pi\rangle$, resulting in a tunneling splitting of the ground-state energy level, has been calculated. However, in addition to the double splitting, the energy of each level also decreases (see, e.g., Ref. 12) due to instanton transitions between vacua separated by $2\pi k$ ($k\in\mathbb{Z}$). This energy-level renormalization becomes observable in the presence of an external magnetic field. In particular, for the problem in question it manifests itself for $K_2\gg K_1$ as oscillations of the physical properties of the particle as a function of the magnetic field.

Using Eq. (11) and equations of motion, one can easily obtain the Lagrangian for the azimuthal dynamical variable $\Phi(t)$. In the limit $K_1 \gg K_2$, when classical dynamics of $\vartheta(t)$ is suppressed, the desired Lagrangian takes the form

$$L_\Phi = \frac{I_{\text{eff}}}{2}\dot{\Phi}^2 - I_{\text{eff}}\left[\frac{\omega_0 \sin\theta_0}{2}\right]^2 [1-\cos(2\Phi)] + \frac{\theta_v}{2\pi}\dot{\Phi} , \tag{12}$$

where I_{eff} and θ_v are determined by Eqs. (5), $\omega_0^2 = 4\mu^2 K_1 K_2 / M_0^2$.

If $K_2 = 0$, Lagrangian (12) coincides with the one [Eq. (4)] considered above. In the absence of a magnetic field ($\theta_0 = \pi/2 \Longrightarrow \theta_v = 0$), this model has been discussed thoroughly in Ref. 2.

In the general case, Lagrangian (12) has been studied in Ref. 7, which treated the Aharonov-Bohm effect in conductors with charge-density waves (see also Ref. 10). Thus, formulas for the oscillating part of the ground-state energy of the dynamic system (12) can be taken directly from Ref. 7:

$$\delta E_{\text{osc}} \sim \omega_0 \sin\theta_0 \sqrt{S_0/2\pi}\exp(-S_0)\cos(\pi m_0') , \tag{13}$$

where

$$m_0' = \min\{(m_0 - \lfloor m_0 \rfloor), (\lceil m_0 \rceil - m_0)\} ,$$

with $m_0 = (M_0/\mu)\cos\theta_0$, and

$$S_0 = \frac{M_0}{\mu}\sqrt{(K_2/K_1)(1-H_2/H_c^2)} \tag{14}$$

is the one-instanton action ($\delta\Phi = \pi$). Notice that the semiclassical equation (13) is valid only if $S_0 \gg 1$.

According to Eq. (13), thermodynamic characteristics of the system (11) (e.g., the magnetization) oscillate as a function of the external-magnetic-field strength with the same period δH (7) as for the simple case considered above. We can regard this process as "tunneling precision." The coincidence of oscillation periods comes from the fact that the true symmetry of the system is the 2π symmetry of the azimuthal degree of freedom, not the π symmetry of the classical potential energy (11). However, the oscillation amplitude, even at zero temperature, acquires an additional exponentially small factor associated with the tunneling character of vacuum-vacuum transitions.

III. Θ VACUUM IN MESOSCOPIC ANTIFERROMAGNETS

When studying the properties of small antiferromagnetic (AFM) particles, we will base our calculation upon the nonlinear O(3) σ model. This model can be derived microscopically (see, e.g., Ref. 13) and is suitable for the investigation of nonlinear excitations in quasi-one-dimensional chains. Moreover, the σ model allows for the difference between the dynamical properties of integer and half-integer quantum chains when topological terms are taken into account.

In terms of spherical components of the Néel vector, the Lagrangian of a one-dimensional spin chain takes the form (Ref. 14)

$$\mathcal{L} = g^{-1}\left\{\frac{\sin^2\vartheta}{2}(\partial_\mu\Phi)^2 + \frac{1}{2}(\partial_\mu\vartheta)^2 - \frac{m^2}{2}\sin^2\vartheta\right\} + \frac{\theta_s}{4\pi}\epsilon^{\mu\nu}\partial_\mu(\cos\vartheta\,\partial_\nu\Phi) . \tag{15}$$

Here $g^{-1} = s/2$ (s is a spin on a site in the AFM chain), $\theta_s = 2\pi s$, m is the magnon mass which is determined by the parameters of a microscopic Hamiltonian

$$H = J\sum_i \{\vec{S}_i \cdot \vec{S}_{i+1} + aS_i^z S_{i+1}^z - b(S_i^z)^2\} \tag{16}$$

through the simple equation $m = \Delta^{-1}\sqrt{(a+b)/2}$ (Δ is the period of a lattice). We set the magnon velocity $c = 2SJ\Delta$ equal to unity.

Unlike the previous model, in antiferromagnets the external magnetic field parallel to the anisotropy axis results only in the precession of the Néel vector and can be easily accounted for by the stretched derivative of the azimuthal variable $\Phi(t)$ (see, e.g., Ref. 15), $\dot{\Phi} \to \dot{\Phi} - \omega_H$, where $\omega_H = 2\mu H$ is the precession frequency and μ is the magnetic moment.

The classical vacua ($\theta = 0, \pi$) of the model correspond to two equivalent orientations of the Néel vector along the anisotropy axis. It is evident from Eq. (15) that the magnetic field does not change the classical ground-state energy of the AFM particle. The quantum corrections (spin tunneling) remove the degeneracy of the vacuum and lead to the double splitting of energy levels.[6,7] In what follows we estimate the influence of the magnetic field on this effect.

For this purpose let us derive the effective Lagrangian of a slow (tunneling) degree of freedom $\vartheta(t)$ by "integrating out" the fast variable $\Phi(t)$ (we assume tunneling in small magnetic particles to be homogeneous). The Lagrangian of the latter, with $\vartheta = \text{const}$, is of the form

$$\Delta L = \frac{\sin^2\vartheta}{2g}(\dot{\Phi} - \omega_H)^2 . \tag{17}$$

The generalized momentum p_Φ conjugate to the coordinate Φ equals $p_\Phi = \sin^2\vartheta\,(\dot{\Phi} - \omega_H)/g$ and the Hamiltonian is

$$H_\Phi = \frac{g}{2\sin^2\vartheta}\left[p_\Phi + \frac{\omega_H \sin^2\vartheta}{g}\right]^2 - \frac{\omega_H^2 \sin^2\vartheta}{2g} . \tag{18}$$

The minimum of the energy (18) is attained for $p_\Phi^0 = -\omega_H \sin^2\vartheta / g$. More rigorously, because of the momentum quantization, $p_\Phi = n\omega_H$ and $n_{\min} = -\lfloor \sin^2\vartheta / g \rfloor$. But so far as the validity of the model (15) assumes $g \ll 1$ ($s \gg 1$), it is possible to neglect the effect of quantization for large quantum numbers.

Hence, the quantum corrections induce the additional term in the Lagrangian of the tunneling degree of freedom

$$\Delta L = \frac{\omega_H^2}{2g}\sin^2\vartheta . \tag{19}$$

The effect caused by Eq. (19) is reduced simply to the renormalization of the magnon mass $m^2 \Longrightarrow m_H^2 = m^2 - \omega_H^2$. For macroscopic quantum tunneling, this renormalization leads to a decrease in the single-instanton action, $S_0 \Longrightarrow S_H = L_{mH}/g$ (L being the length of the chain). Consequently, the magnetic field enhances the tunneling rate.

Finally, let us consider the effect of quantum fluctuations of fast variable $\Phi(t)$ in the presence of a background magnetic field on the energy of a topological soliton in the anisotropic AFM chain. The simplest soliton is of the form (Refs. 14 and 16)

$$\cos\vartheta(x) = \tanh(mx) \ . \tag{20}$$

Substituting Eq. (20) into the Lagrangian (15) and (16) and integrating over the spatial coordinate, one can easily obtain the desired effective Lagrangian of the fast variable,

$$L_\Phi = -E_s + \frac{1}{gm}(\dot{\Phi} - \omega_H)^2 + \frac{\theta_s}{2\pi}(\dot{\Phi} - \omega_H) \ , \tag{21}$$

where $E_s = 2m/g$ is the rest energy of the classical soliton.[15] By comparing Eq. (21) with Eq. (4), it is readily seen that the vacuum angle takes the form

$$\theta_v = \theta_s - 4\pi\frac{\omega_H}{gm} = 2\pi\left[s - 2\frac{\omega_H}{gm}\right] \ . \tag{22}$$

As a result, the soliton rest energy acquires an additional term which oscillates as a function of the magnetic field. In particular, at low temperatures, $T \ll T_q = \hbar J\sqrt{2(a+b)}$, the oscillating part of the soliton rest energy is

$$\delta E_s^{\mathrm{osc}} = \tfrac{1}{2} T_q (s'')^2 \tag{23}$$

where $s'' = \min\{(s' - \lfloor s' \rfloor, (\lceil s' \rceil - s')\}$, with $s' \equiv s - \hbar\omega_H / T_q$.

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