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Quantum Tunneling of Magnetization in Small Ferromagnetic Particles

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The probability of tunneling of the magnetization in a single-domain particle through an energy barrier between easy directions is calculated for several forms of magnetic anisotropy. Estimated tunneling rates prove to be large enough for observation of the effect with the use of existing experimental techniques.

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In recent years there has been considerable interest in the phenomenon of macroscopic quantum tunneling (MQT).[1] It corresponds to the tunneling of a macroscopic variable through the barrier between two minima of the effective potential of a macroscopic system. To date MQT has been studied, both theoretically[1] and experimentally, in superconducting devices.[2] In this paper we show that single-domain magnetic particles represent another rich field for MQT study.

As is known,[3] a sufficiently small ferromagnetic particle consists of a single magnetic domain. Equilibrium easy directions of the magnetic moment $\mathbf{M}$ ($\mathbf{M}^2=M_0^2$ being a constant) correspond to the local minima of the energy

$$E=-\mathbf{M}\cdot\mathbf{H}+A_{ik}M_iM_k+B_{iklm}M_iM_kM_lM_m+\cdots, \tag{1}$$

where $\mathbf{H}$ is the magnetic field and A_{ik}, B_{iklm}, etc., are determined by the crystalline anisotropy and by the shape of the particle. Since $\mathbf{M}$ is an axial vector, any minimum of the energy at $H=0$ is at least twice degenerate with respect to two opposite directions of $\mathbf{M}$. If one considers $\mathbf{M}$ as a spin operator, then the projection $\mathbf{M}\cdot\mathbf{e}$ onto one of the easy directions $\mathbf{e}$ does not in general commute with E. This means that eigenvalues of $\mathbf{M}\cdot\mathbf{e}$, in general, are not conserved quantum numbers even at $H=0$, which is not surprising because the magnetic anisotropy appears as a result of relativistic interactions.[4,5] Consequently, $\mathbf{M}$ can tunnel between the energy minima. Tunneling removes the degeneracy of the ground state and puts the particle into a state of a lower energy, wherein

$$\langle\mathbf{M}\rangle=0,\quad \langle\mathbf{M}^2\rangle=M_0^2. \tag{2}$$

Here, angular brackets denote a quantum average. For two successive measurements of $\mathbf{M}$ separated by the time interval Δt one should obtain at $T=0$ and $H=0$, and with neglect of dissipation, the effect of macroscopic quantum coherence[1]

$$\langle\mathbf{M}(t)\mathbf{M}(t+\Delta t)\rangle=M_0^2\cos(2P\Delta t), \tag{3}$$

where $\hbar P$ is the tunneling matrix element. In the presence of a magnetic field, the potential (1) has, in general, one absolute minimum and several local minima, so that the problem of MQT from a metastable state arises. For both macroscopic quantum coherence and MQT the key quantity is the tunneling rate P, which should be calculated in terms of the macroscopic parameters describing single-domain particles.

The first reference to the possibility of quantum tunneling of the magnetic moment in small particles apparently was made by Bean and Livingston.[6] They suggested tunneling as an explanation of the experimental data[7] indicating that transitions between different orientations of the magnetic moment in single-domain nickel particles do not disappear completely with a decrease in temperature to absolute zero. Two mechanisms for the tunneling process were suggested. The first one[8] applies to relatively large particles whose size is greater than the domain-wall width. It consists of the nucleation of a domain wall, which subsequently sweeps across the particle, switching the direction of its magnetization. Since the energy barrier between the two states is proportional to the volume of the particle, the tunneling rate due to this mechanism is extremely small.[9] For particles of size smaller than the domain-wall width, uniform subbarrier rotation of the magnetic moment has been considered[10-12] on the assumption of the existence of an effective moment of inertia associated with the rotation of **M**. The possibility of such an effect due to the dynamical equations for **M** was recently conjectured by one of us (L.G.).[11,12] In this paper we show that an effective inertia and tunneling follow directly from a quasiclassical treatment of the dynamical equations for **M**, and we calculate the tunneling rate $P = A\exp(-S_E/\hbar)$ for some typical cases, S_E being the extremal imaginary-time action for the subbarrier rotation of **M**.

If we neglect dissipation (the effect of dissipation is briefly discussed below) the dynamical equation for **M** is

$$d\mathbf{M}/dt = -\gamma \mathbf{M} \times \delta E/\delta \mathbf{M}, \tag{4}$$

where $\gamma \equiv ge/2mc$ (g is the gyromagnetic ratio). Introducing angles θ,ϕ for the direction of **M** in a spherical coordinate system, one can also obtain Eq. (4) from the action[13]

$$I = \int dt \{(M_0/\gamma)\dot{\phi}\cos\theta - E(\theta,\phi)\}, \tag{5}$$

which is a simple reflection of the fact that

$$x = \phi, \quad p = (M_0/\gamma)\cos\theta = \hbar S_z \tag{6}$$

(S_z is the Z projection of the total spin of the particle) are canonical variables, so that

$$L = p\dot{x} - E \tag{7}$$

is the Langrangean of the system.[14] In terms of the coordinates θ and ϕ, Eq. (4) is equivalent to

$$\dot{\theta}\sin\theta = (\gamma/M_0)\partial E/\partial\phi, \tag{8a}$$

$$\dot{\phi}\sin\theta = -(\gamma/M_0)\partial E/\partial\theta. \tag{8b}$$

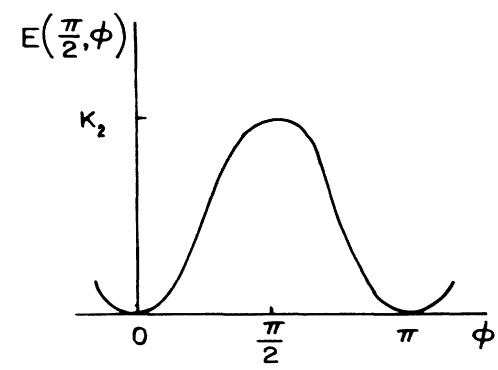

FIG. 1. Energy $E(\theta = \frac{1}{2}\pi, \phi)$ for a model (model I) with an easy plane and an easy axis in the plane.

Let us consider as "model I" the following form of E:

$$E(\theta,\phi) = k_1 M_z^2 + k_2 M_y^2 = K_1\cos^2\theta + K_2\sin^2\theta\sin^2\phi, \tag{9}$$

where $K_1 > K_2 > 0$. This model describes XOY-easy-plane anisotropy with an easy axis along the x direction in the plane. The ground state of the system corresponds, therefore, to **M** pointing in one of the two directions parallel to the X axis; i.e., $\theta = \frac{1}{2}\pi$, $\phi = 0, \pi$ (Fig. 1). In imaginary time, $\tau = it$, the two Eqs. (8), together with Eq. (9), give the following equation for ϕ:

$$\tfrac{1}{2}(d\phi/d\tau)^2 = \omega_0^2(1 - \lambda\sin^2\phi)\sin^2\phi, \tag{10}$$

where

$$\omega_0 \equiv (2\gamma/M_0)(K_1K_2)^{1/2}, \quad \lambda \equiv K_2/K_1. \tag{11}$$

Equation (10) has the instanton solution

$$\phi = \arccos\frac{(1-\lambda)^{1/2}\tanh\omega_0\tau}{(1-\lambda\tanh^2\omega_0\tau)^{1/2}}, \tag{12}$$

corresponding to the switching of **M** from $\phi = \pi$ at $\tau = -\infty$ to $\phi = 0$ at $\tau = \infty$. After the elimination of θ with the use of the equations of motion, the action (5) for this trajectory may be expressed as

$$iI_0 \equiv -S_E = -K_2\int_{-\infty}^{\infty} d\tau\{\omega_0^{-2}(1-\lambda\sin^2\phi)^{-1}(d\phi/d\tau)^2 + \sin^2\phi\}, \tag{13}$$

where the first term can be interpreted as an effective kinetic energy associated with the subbarrier rotation of **M**. Further integration in Eq. (13) gives

$$P \propto \exp\left(\frac{iI_0}{\hbar}\right) = \left(\frac{1-\sqrt{\lambda}}{1+\sqrt{\lambda}}\right)^{M_0/\hbar\gamma}. \tag{14}$$

Notice that $P \to 0$ as $\lambda \to 1$, which follows from the observation that in this limit $E \to K(M_z^2 + M_y^2) = KM_0^2 - KM_x^2$ commutes with M_x. It should also be noticed that for single-domain particles which can be considered as macroscopic, the ratio $M_0/\hbar\gamma$ is large, so that tunneling defined by Eq. (14) can be observed only when $\lambda \ll 1$, i.e., in the case of very strong transverse anisotropy K_1, forcing **M** to lie in the XOY plane, and comparatively small anisotropy K_2 in that plane.

The tunneling rate can increase in the presence of an external magnetic field, which decreases the energy barrier.

Our next example, "model II," corresponds to the simplest case of easy-axis anisotropy along the Z axis and a transverse field applied along the X axis:

$$E=K\sin^2\theta-HM_0\sin\theta\cos\phi+H^2M_0^2/4K. \quad (15)$$

For $H<H_c=2K/M_0$ there are two energy minima ($E=0$) corresponding to $\phi=0$: $\theta=\theta_0$ and $\pi-\theta_0$, where $\sin\theta_0=H/H_c$ (Fig. 2). The energy barrier between the two states is $U=K\epsilon^2$, where $\epsilon\equiv 1-H/H_c$. It disappears at $H=H_c$ when $\theta_0=\frac{1}{2}\pi$ and the two states coincide. From Eqs. (8) and (15) the following equation for θ can be obtained:

$$d^2\theta/d\tau^2=\omega_H^2\cot\theta[1+(d\theta/d\tau)^2/\omega_H^2]-2\omega_1\omega_H\cos\theta[1+(d\theta/d\tau)^2/\omega_H^2]^{1/2}, \quad (16)$$

where $\omega_H=\gamma H$, $\omega_1=\gamma K/M_0$, and $\tau=it$. In the limiting case of a very low field, $\theta_0\rightarrow 0$. In this case the approximate solution of Eq. (16), corresponding to the switching of **M** from $\theta=\pi$ at $\tau=-\infty$ to $\theta=0$ at $\tau=\infty$, is given by

$$\theta=\arccos[\tanh\omega_1\tau]. \quad (17)$$

Calculating the action for this trajectory, we obtain

$$P\propto(H/H_c)^{2M_0/\hbar\gamma}. \quad (18)$$

For $H\rightarrow 0$, P goes to zero because in this limit E of Eq. (15) commutes with M_z.

For another limiting case, $H\rightarrow H_c$, $\epsilon\ll 1$, introducing $\delta=\frac{1}{2}\pi-\theta\ll 1$, we obtain from Eq. (16)

$$d^2\delta/d\tau^2=\omega_H^2(-\epsilon\delta+\tfrac{1}{2}\delta^3), \quad (19)$$

which has the instanton solution

$$\delta=(2\epsilon)^{1/2}\tanh[\sqrt{\epsilon}\omega_H\tau], \quad (20)$$

corresponding to the switching of **M** between the two energy minima at $\delta=\pm(2\epsilon)^{1/2}$. Correspondingly

$$P\propto\exp[-(4M_0/\hbar\gamma)\epsilon^{3/2}]. \quad (21)$$

Note that although the WKB exponent (21) becomes smaller for small ϵ, the observation of MQT in this case is impeded by the closeness of the states with $\theta=\frac{1}{2}\pi\pm(2\epsilon)^{1/2}$ to each other. The latter have appeared because **H** was perpendicular to the anisotropy field.

Our final example, "model III," is described by

$$E=-k_1M_z^2+k_2M_y^2-\mathbf{M}\cdot\mathbf{H}, \quad (22)$$

with **H** being opposite to the easy axis OZ and $k_1,k_2>0$. Up to a constant, this is equivalent to

$$E=(K_1+K_2\sin^2\phi)\sin^2\theta-M_0H(1-\cos\theta). \quad (23)$$

Now the local energy minima are at $\theta=0$ and $\theta=\pi$; the maximum (see Fig. 3) corresponds to $\cos\theta_1=H/H_c$ ($H_c=2K_1/M_0$). The energy barrier between the minima is $U=K_1\epsilon^2$, while quantum transitions are generated by the transverse anisotropy K_2. In the limit $\epsilon\rightarrow 0$, $\theta_1\rightarrow(2\epsilon)^{1/2}, \theta_2\rightarrow 2\sqrt{\epsilon}$. Using Eqs. (8) and (23) one can obtain in this limit

$$d^2\theta/d\tau^2=\omega_0^2(\epsilon\theta-\tfrac{1}{2}\theta^3), \quad (24)$$

where ω_0 is given in Eq. (11). Note that one can interpret this equation as subbarrier rotation of the magnetization with an effective moment of inertia $M^2/2\gamma^2K_2$ due to transverse anisotropy K_2, in the effective potential created by the longitudinal fields K_1 and H. Equation (24) has the instanton solution

$$\theta(\tau)=\theta_2/\cosh(\omega_0\sqrt{\epsilon}\tau), \quad (25)$$

corresponding to the variation of θ from $\theta=0$ at $\tau=-\infty$ to $\theta=\theta_2$ at $\tau=0$, and then back to $\theta=0$ at $\tau=\infty$. Calculating the action for this trajectory, we obtain the WKB exponent for the tunneling from $\theta=0$ to $\theta=\pi$,

$$P\propto\exp[-(8M_0/3\hbar\gamma)(K_1/K_2)^{1/2}\epsilon^{3/2}]$$
$$=\exp(-U/k_BT_c), \quad (26)$$

where

$$T_c=3\hbar\gamma(K_1K_2)^{1/2}\sqrt{\epsilon}/8k_BM_0. \quad (27)$$

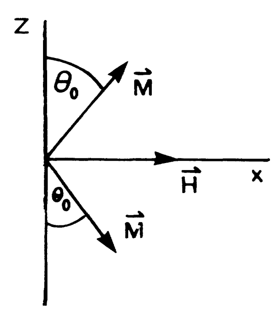

FIG. 2. Directions of the magnetization corresponding to energy minima for a model (model II) having an easy axis and a transverse field along the X axis.

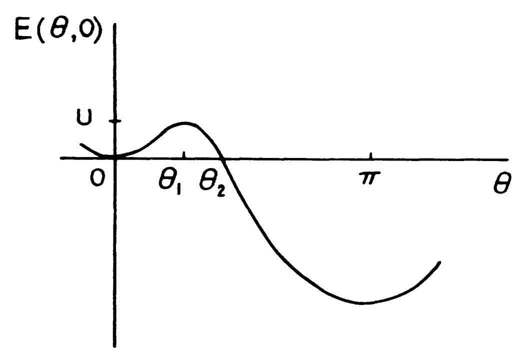

FIG. 3. Energy $E(\theta,\phi=0)$ for a model (model III) with an easy axis, a magnetic field along this axis, and transverse anisotropy.

The prefactor in Eq. (26), as well as in Eqs. (14) and (21), is of the order of $\omega(U/k_BT_c)^{1/2}$, where ω is the characteristic frequency of the instanton solution. Thus, the dependence of the tunneling rate on parameters is dominated by the exponent. On the other hand, the probability of switching of the magnetic moment due to thermal activation is proportional to $\exp(-U/k_BT)$. Hence $T \sim T_c$ corresponds to a crossover from the thermal to the quantum regime, i.e., to the regime where the tunneling rate does not depend upon temperature. Notice that $K_1' = K_1/V$, $K_2' = K_2/V$, and $M_0' = M_0/V$ (V is the volume of the particle) are constants of the material. Thus, T_c depends upon H, but not explicitly on the volume, while the energy barrier U is proportional to V. For $M_0' \simeq 500$ emu/cm^3, $K_1' \simeq K_2' \simeq 5\times10^6$ erg/cm^3, and $\epsilon \sim 0.01$ (which corresponds to an accuracy in the magnetic field control of $\Delta H = \epsilon H_c \simeq 200$ Oe), quantum switching of the magnetic moment can be observed in particles of diameter $\simeq 100$ Å at $T < T_c \simeq 0.06$ K with $H \simeq H_c \simeq 20$ kOe.[15]

In our study of the instanton solutions of the dynamical equation for $\mathbf{M}$ we neglected the effect of dissipation. It can be included by the introduction of the interaction of $\mathbf{M}$ with other degrees of freedom.[12] If one takes into account the interaction with phonons, then Eq. (5), generalized to include the possibility of a nonuniform switching of the magnetization, is

$$I = \int dt \int dV \left\{ \frac{M_0'}{\gamma} \dot{\phi} \cos\theta - E(\mathbf{M}') - \tfrac{1}{2}\alpha \left(\frac{\partial M_i'}{\partial x_k} \right)^2 + \tfrac{1}{2}\rho \dot{\mathbf{u}}^2 - \tfrac{1}{2}\lambda_{iklm} u_{ik} u_{lm} - a_{iklm} u_{ik} M_l' M_m' \right\}, \tag{28}$$

where $\mathbf{M}' = M_0'(\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta)$ is the local magnetization $[\theta = \theta(x,t),\ \phi = \phi(x,t)]$, $u(x,t)$ is the phonon displacement field, $u_{ik} = \frac{1}{2}(\partial_i u_k + \partial_k u_i)$ is the strain tensor, α is the exchange constant, ρ is the mass density of the material, and $\hat{\lambda}$ and $\hat{a}$ are the elastic and magnetoelastic tensors, respectively. Integration over phonon variables $\mathbf{u}(\mathbf{x},t)$ in the path integral then gives the effective potential of the system in the spirit of Caldeira and Leggett.[16] Our preliminary analysis shows that at least for some experimental situations the interaction with phonons may significantly contribute to the probability of switching. A more detailed study of this model, as well as an exact calculation of the prefactor for the tunneling rate, will be presented elsewhere.

In conclusion, we have represented a simple approach which allows us to estimate the rate of quantum switching of the magnetization in a single-domain particle with an arbitrary form of magnetic anisotropy. The WKB exponent has been calculated for several forms of the anisotropy energy. The effect proves to be large enough to be observed with the use of existing experimental techniques.

Note added.—After this work was completed, we learned about the work of Scharf, Wreszinski, and van Hemmen[17] and Enz and Schilling[18] wherein the same quantum problem for a single spin was considered for similar forms of spin Hamiltonians. In the limit of a large spin, the results obtained for models I and II by both the WKB method and numerical diagonalization of the Hamiltonian are in perfect agreement with our results. Our method has the advantage of simplicity and extendibility to inhomogeneous situations which will be presented in a future publication.

[1]For a recent review on MQT see A. J. Leggett *et al.*, Rev. Mod. Phys. **59**, 1 (1987).

[2]W. den Boer and R. De Bruyn Ouboter, Physica (Amsterdam) **98B**, 185 (1980); S. Washburn, R. A. Webb, and S. M. Faris, Phys. Rev. Lett. **54**, 2712 (1985); J. M. Martinus, M. M. Devoret, and J. E. Lukens, Phys. Rev. Lett. **55**, 1547 (1985).

[3]W. F. Brown, Jr., Appl. Phys. **39**, 993 (1968).

[4]L. D. Landau and E. M. Lifshifts, *Electrodynamics of Continuous Media* (Nauka, Moscow, 1982), 2nd ed.

[5]Notice that the possibility of treating **M** as a single quantum variable is a result of the much stronger exchange interaction, which suppresses the dynamics of the individual spins of the particle.

[6]C. P. Bean and J. D. Livingston, J. Appl. Phys. **30**, 120S (1959).

[7]L. Weil, J. Chem. Phys. **51**, 715 (1954).

[8]D. Stauffer, Solid State Commun. **18**, 533 (1976).

[9]Tunneling of a small portion of the domain wall through the energy barrier created by defects in a bulk solid may be significant. Recently such a mechanism has been suggested as an explanation of staircase behavior in the magnetization reversal of $SmCo_{3.5}Cu_{1.5}$ at low temperatures; see M. Uehara *et al.*, Phys. Lett. **114A**, 23 (1986).

[10]E. M. Chudnovsky, Zh. Eksp. Teor. Fiz. **77**, 2157 (1979) [Sov. Phys. JETP **50**, 1035 (1979)].

[11]L. Gunther, unpublished.

[12]See, for example, A. De Franzo *et al.*, in Proceedings of the Thirty-Second Annual Conference on Magnetism and Magnetic Materials, Chicago, Illinois, 9–12 November, 1987 (to be published).

[13]T. L. Gilbert, Phys. Rev. **100**, 1243 (1955).

[14]For a detailed discussion see W. F. Brown, Jr., *Micromagnetics* (Wiley, New York, London, 1963).

[15]For some materials there can be a significant surface effect [see, e.g., A. E. Berkowitz, J. A. Lahut, and C. E. VanBuren, IEEE Trans. Magn. **16**, 184 (1980)] which will be important for extremely small particles. In this case we expect that our theory gives the correct order of magnitude for the WKB exponent.

[16]A. O. Caldeira and A. J. Leggett, Ann. Phys. (N.Y.) **149**, 374 (1983), and **153**, 445E (1984).

[17]G. Scharf, W. F. Wreszinski, and S. L. van Hemmen, J. Phys. A **20**, 4309 (1987).

[18]M. Enz and R. Schilling, J. Phys. C **19**, L711 (1986).