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Quantum interference in small magnetic particles

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We consider tunneling of a large spin between nonequivalent wells in the presence of the magnetic field. Using coherent spin states we show that the quenching of tunneling for a half-integer spin, which occurs at $H=0$, survives up to a reasonably large field. An experiment is proposed which may allow one to observe this quenching. Oscillation of the tunneling rate on the applied field is suggested.

Recently, it has been demonstrated that the vector of the magnetization formed by a large number of spins in magnetic systems can coherently tunnel between the minima of magnetic energy. Particular cases are tunneling of the magnetic moment in small ferromagnetic particles,[1] tunneling of the Néel vector in antiferromagnetic particles,[2] quantum nucleation of magnetic domains,[3] and tunneling of domain walls.[4,5] Theoretical suggestions have led to a number of experiments[6–10] which seem to support the idea of magnetic tunneling. Thus, tunneling of magnetization in solids is becoming another example of quantum phenomena at the mesoscopic scale.[11]

The most distinct effect of that kind, describable in exact mathematical terms,[1,12–17] is tunneling of the magnetic moment of a single-domain magnetic particle, **M**, between two energy minima created by magnetic anisotropy. In the absence of the applied field, the classical magnetic state of the particle is degenerate, $E(\Uparrow)=E(\Downarrow)$. Tunneling removes this degeneracy, leading to the ground state which is a superposition of $|\Uparrow\rangle$ and $|\Downarrow\rangle$. Although instantons of the Landau-Lifshitz equation, used to describe this tunneling, have been known for some time,[1] one fundamental question remained unanswered until very recently. The possibility of tunneling seemed insensitive to the value of the total spin of the particle, S. However, the Kramers theorem dictates that for a half-integer S the degeneracy cannot be removed in the absence of the magnetic field, and, thus, no tunneling should occur. A beautiful solution to this problem has been given in Refs. 16 and 17: It has been demonstrated that the Kramers theorem is recovered via interference of instantons, due to the topological term in the magnetic action.

In real experiments one will always have some weak magnetic field which removes the Kramers degeneracy. Is it, nevertheless, possible to detect the freezing of tunneling in a small magnetic particle with a half-integer total spin? Obviously, this is a question of how large a field is required to remove all traces of the freezing which occurs at $H=0$. In this paper we show that this unquenching field can be sufficiently large to study the effect experimentally, and suggest a possible experiment. We also suggest another effect: oscillation of the tunneling rate with the applied magnetic field.

Consider a single-domain ferromagnetic particle with the magnetic moment $M=2\mu_B\mathbf{S}$ and $S\gg1$. The total spin of such a 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

$$\hbar\frac{d\mathbf{S}}{dt}=-\mathbf{S}\times\frac{\delta E(\mathbf{S})}{\delta\mathbf{S}}\ , \tag{1}$$

where $E(\mathbf{S})$ is the magnetic energy of the particle. Let X be the easy-magnetization axis and XY be the easy-magnetization plane. The simplest form of the corresponding magnetic energy is

$$E=K_\perp\frac{S_z^2}{S^2}-K_\parallel\frac{S_x^2}{S^2}-2\mu_B\mathbf{S}\cdot\mathbf{H}\ , \tag{2}$$

where $K_\perp, K_\parallel>0$ are the anisotropy constants. We will study the situation where **S** initially points along the positive X axis and **H** is applied in the opposite direction (Fig. 1). Then Eq. (2), in spherical coordinates, is equivalent to

$$E(\theta,\phi)=K_\perp\cos^2\theta+K_\parallel(1-\sin^2\theta\cos^2\phi)$$
$$-MH(1-\sin\theta\cos\phi)\ , \tag{3}$$

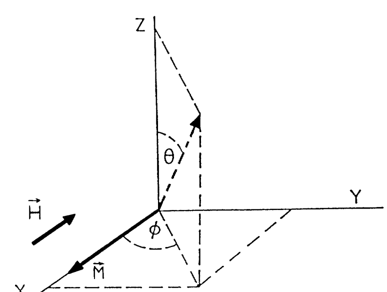

FIG. 1. Single-domain particle in the magnetic field. The field is applied along the easy X axis, opposite to the magnetic moment of the particle.

where a constant has been added to make the energy of the initial state, $E(\pi/2,0)$, equal to zero. The dependence of E on ϕ, when **S** lies in the easy magnetization plane ($\theta=\pi/2$), is shown in Fig. 2. At $H\neq 0$ there is a metastable state $\theta=\pi/2$, $\phi=0$ (i.e., **S** antiparallel to **H**) and the stable state $\theta=\pi/2$, $\phi=\pi$ (i.e., **S** parallel to **H**). The energy barrier U between the two states exists at $H<H_\parallel=K_\parallel/\mu_B S$,

$$U=K_\parallel\epsilon^2\ ,\quad \epsilon(H)=1-\frac{H}{H_\parallel}\ . \tag{4}$$

To escape from the metastable state, **S** must rotate by the angle $\phi=\pm\phi_H$ [Fig. 2(b)], which satisfies

$$\cos\phi_H=2\frac{H}{H_\parallel}-1\ . \tag{5}$$

The classical **S** state characterized by angles θ and ϕ corresponds in quantum mechanics to the coherent spin state,[18]

$$|\theta,\phi\rangle=\left[\cos\frac{\theta}{2}\right]^{2S}\exp\left[\tan\frac{\theta}{2}e^{i\phi}\hat{S}^-\right]|S\rangle\ , \tag{6}$$

where $\hat{S}^-=\hat{S}_x-i\hat{S}_y$, $|S\rangle$ is the eigenstate of $\hat{S}_z$ corresponding to the maximal eigenvalue $S_z=S$. The quantum Hamiltonian $\mathcal{H}$ corresponding to Eq. (2) does not commute with $\hat{S}_x$. This leads to a nonzero amplitude of the underbarrier escape of **S** from the initial Ψ_0 level into the excited Ψ_r level which is in resonance with Ψ_0 [Fig. 2(b)]. In the classical limit, the Ψ_r state corresponds to the precession of **S** about **H**, governed by Eq. (1).

The amplitude of the transition is given by the imaginary-time propagator

$$A=\langle\Psi_0|e^{\beta\mathcal{H}}|\Psi_r\rangle \tag{7}$$

with β^{-1} going to zero in the limit of zero temperature. It can be calculated by inserting the resolution of the identity in Eq. (7),

$$A=\int d\Omega\langle\Psi_0|e^{\beta\mathcal{H}}|\Omega\rangle\langle\Omega|\Psi_r\rangle\ . \tag{8}$$

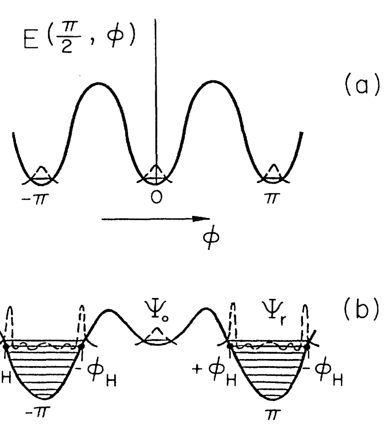

FIG. 2. Magnetic energy in the easy XY plane vs polar angle ϕ. (a) $H=0$, (b) $H\neq 0$. The wave functions associated with the direction of **S** are shown schematically by the dotted curves.

In a semiclassical approximation, the Ψ_0 state is very close to $|\pi/2,0\rangle$, while the integral (8) is dominated by the coherent states $|\pi/2,\pm\phi_H\rangle$. This gives

$$A\propto\left\langle\frac{\pi}{2},0\left|e^{\beta\mathcal{H}}\right|\frac{\pi}{2},-\phi_H\right\rangle\left\langle\frac{\pi}{2},-\phi_H\middle|\Psi_r\right\rangle+\left\langle\frac{\pi}{2},0\left|e^{\beta\mathcal{H}}\right|\frac{\pi}{2},\phi_H\right\rangle\left\langle\frac{\pi}{2},\phi_H\middle|\Psi_r\right\rangle\ . \tag{9}$$

The propagators $\langle\pi/2,0|e^{\beta\mathcal{H}}|\pi/2,\pm\phi_H\rangle$ in Eq. (9) are equivalent to the imaginary-time ($\tau=it$) path integrals

$$\int D\{\mathbf{S}(\tau)\}\exp\left[-\frac{I_E}{\hbar}\right] \tag{10}$$

over the classical $\mathbf{S}(\tau)$ trajectories connecting the $|\pi/2,0\rangle$ and $|\pi/2,\pm\phi_H\rangle$ states. Here I_E is the Euclidean counterpart of the magnetic action

$$I=\int dt[\hbar S(\cos\theta-1)\dot{\phi}-E(\theta,\phi)] \tag{11}$$

that generates Eq. (1). The first term in Eq. (11) corresponds to the Berry phase[19] and is crucial for our considerations. [For a closed $\mathbf{S}(t)$ path it is equal to S times the area swept out by **S** on the surface of the unit sphere.] The path integral (10) is dominated by the instanton of Eq. (1),[1] corresponding to the underbarrier rotation of **S** to the escape point $\phi=\pm\phi_H$. Together with the phase factor generated by Eq. (11) the instanton contribution gives

$$\left\langle\frac{\pi}{2},0\left|e^{\beta\mathcal{H}}\right|\frac{\pi}{2},\pm\phi_H\right\rangle=a(H)e^{-b(H)}e^{\pm i\phi_H S}\ , \tag{12}$$

where $a(H)$ and $b(H)$ are smooth functions of the field. For $H\to 0$ and $H\to H_\parallel$ exact instanton solutions of Eq. (1) and their contribution to the path integral were obtained in Ref. [1].

Let us now turn to the overlap factors $\langle\pi/2,\pm\phi_H|\Psi_r\rangle$. We expand $|\Psi_r\rangle$ as

$$|\Psi_r\rangle=\sum_{S_z}a_{S_z}(H)|S_z\rangle\ . \tag{13}$$

Due to the symmetry of the Hamiltonian (2), a_{S_z} are either all even, $a_{S_z}=a_{-S_z}$, or all odd, $a_{S_z}=-a_{-S_z}$, in S_z. A similar symmetry is possessed by the coherent states: With the help of Eq. (6) we get

$$\left|\frac{\pi}{2},\pm\phi_H\right\rangle=e^{\pm i\phi_H S}\sum_{S_z}e^{\mp i\phi_H S_z}b_{S_z}|S_z\rangle\ , \tag{14}$$

where

$$b_{S_z}=2^{-S}\left[\frac{(2S)!}{(S-S_z)!(S+S_z)!}\right]^{1/2}\ . \tag{15}$$

As follows from Eq. (15), $b_{S_z}=b_{-S_z}$.

Now working out the overlap factors and combining all terms in Eq. (9), we obtain for the tunneling onto an even-symmetry Ψ_r level

$$A=a(H)e^{-b(H)}\sum_{m=0}^{S}C_m\cos(m\phi_H) \tag{16}$$

for an integer S, and

$$A=a(H)e^{-b(H)}\sum_{m=0}^{S-1/2}C_m\cos[(m+\tfrac{1}{2})\phi_H] \tag{17}$$

for a half-integer S. Here $C_m=ka_{S_z}b_{S_z}$, with $S_z=m$ for an integer S and $S_z=m+\frac{1}{2}$ for a half-integer S; $k=2$ if $S_z=0$ and $k=4$ for all $S_z\neq 0$. The amplitude of the tunneling onto an odd Ψ_r level is zero. From Eq. (17), the quenching of tunneling[16,17] for a half-integer total spin at $H=0$ (that is at $\phi_H=\pi$) is evident. Another important observation is that the tunneling rate for both the integer and half-integer S must oscillate with the applied field.

Our main goal is to understand how large a field destroys all traces of the quenching. If one demands that all cosines in Eq. (17) are small, this, according to Eq. (5), would require $H \ll H_\parallel/4S^2$. Note, however, that according to Eq. (15) the coefficients b_{S_z} rapidly go to zero for large S_z. This reflects the fact that the coherent states (14) correspond to the classical vector **S** lying in the XY plane. At $|S_z| \ll S$, with the help of Stirling's formula, we obtain

$$b_{S_z}=(\pi S)^{-1/4}\exp\left[-\frac{3S_z^2}{2S}\right]. \tag{18}$$

It then follows from Eq. (18) that only $m\leq\sqrt{S}$ effectively contribute to A; at $m \gg \sqrt{S}$ coefficients C_m become exponentially small. Thus, the actual limitation on the field becomes $H \ll H_\parallel/S$.

Typical values of the anisotropy field in single-domain particles are $H_\parallel\sim 10^3-10^4$ Oe. Consequently, for $S\sim 10^2-10^3$, the presence of a small magnetic field, $H\sim 1$ Oe, should not be a problem. The more serious problem comes from the fact that in ferromagnetic particles the WKB exponent $b(H)$ is of the order of[1] $(H_\parallel/H_\perp)^{1/2}\epsilon^{3/2}(H)S$, where $H_\perp=K_\perp/\mu_B S$. This makes the tunneling probability exponentially small for a large S, unless $H_\parallel \ll H_\perp$ or $H\rightarrow H_\parallel$. Antiferromagnetic particles with a small noncompensation of sublattices have been shown to be better candidates for experimental study.[2] For such particles the WKB exponent is $(H_\parallel/H_{\rm ex})^{1/2}S$ where the exchange field $H_{\rm ex}$ is orders of magnitude greater than the anisotropy field $H_\parallel$ (S is the total spin of one sublattice). For the topological effects studied in Refs. 16 and 17 and in this work, the relevant quantity in antiferromagnetic particles is the excess spin s due to the noncompensation of sublattices. Then the formalism employed here applies without change to the antiferromagnetic case. The effective freezing of the tunneling for half-integer s will occur for $H \ll H_\parallel/s$. For nanometer-scale antiferromagnetic particles this should be an easier condition to satisfy than in the ferromagnetic case. In a system of a large number of single-domain particles one would expect statistically equal numbers of integer and half-integer spins. If all moments of the particles are initially magnetized in one direction and then the field is switched off, the freezing effect should reveal itself in a longer magnetic relaxation for the half-integer particles. This should result in a peculiar time dependence of the relaxation: a fast drop of the magnetic moment of the system to one-half of the initial value and then slow relaxation to zero. Of course, to observe this effect one must have a very narrow distribution of particle sizes, otherwise the broad distribution of individual lifetimes will smear the relaxation.

The macroscopic tunneling variable **S** must inevitably interact with the dissipative environment. Interactions which violate t invariance can potentially destroy the freezing effect. They include the coupling of **S** to the fluctuations of the electromagnetic field, $V\propto \mathbf{S}\cdot\mathbf{h}$, and the coupling to other spins, $V\propto\sum_i \mathbf{S}\cdot s_i$. The first can be ignored because at low temperature the average statistical fluctuation of the field in the volume of a single-domain particle is much less than the critical field estimated above, $\langle \mathbf{h}^2\rangle^{1/2}\sim 10^{-7}$ Oe at $T\sim 1$ K. Interaction between the spins forming **S** and the spins of the environment should be of major concern[20] since the addition of only one $\frac{1}{2}$ spin to the system may completely change the tunneling picture. The importance of this interaction, of course, largely depends on how strong it is, compared to the exchange interaction between the spins forming **S**: typically 0.1–1 eV. This problem remains to be studied qualitatively. It should be noted that, even in the presence of the freezing at $H=0$, the survival of interference effects at $H\neq 0$ is not obvious and depends on the strength of the dissipation. If **S** is strongly coupled to the dissipative environment, the latter acts as a classical measuring device that determines the direction of tunneling, $+\phi_H$ or $-\phi_H$ [see Fig. 2(b)]. In this case the interference between the instanton and anti-instanton, switching **S** to $+\phi_H$ and $-\phi_H$, respectively, will be destroyed. Note, however, that in insulating materials the width of the ferromagnetic resonance can be extremely narrow. Correspondingly, the precession state which we denote Ψ_r should be a rather good quantum state of a single-domain particle. This makes us believe that quantum interference effects in magnetic particles can be observed experimentally. The approach developed in this work provides the basis for such study and for further theoretical elaborations, including the effect of dissipation.

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