Macroscopic Quantum Coherence in a Magnetic Nanoparticle Above the Surface of a Superconductor

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We study macroscopic quantum tunneling of the magnetic moment in a single-domain particle placed above the surface of a superconductor. Such a setup allows one to manipulate the height of the energy barrier, preserving the degeneracy of the ground state. The tunneling amplitude and the effect of the dissipation in the superconductor are computed.

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Tunneling of the magnetic moment in nanoparticles and molecular clusters has been intensively studied theoretically and experimentally in the last decade [1]. The interest in this problem is twofold. First, magnetic tunneling reveals itself at a quasiclassical level, that is, in situations where all three components of the magnetic moment, **M**, can be rather accurately determined by a macroscopic measurement. The interaction of **M** with microscopic degrees of freedom makes this problem one of tunneling with dissipation [2,3]. Second, tunneling of the magnetic moment changes the magnetic properties of small magnets, with potential implications for the data-storage technology. It also adds nanomagnets to the list of candidates for qubits — the elements of quantum computers.

In zero magnetic field the magnetic state of a classical magnet is symmetric with respect to $\mathbf{M} \to -\mathbf{M}$ due to time-reversal symmetry. In nanoparticles the $|\uparrow\rangle$ and $|\downarrow\rangle$ minima of the energy are separated by a barrier, U, due to the magnetic anisotropy. The thermal rate of switching between the two classical states is proportional to $\exp(-U/T)$. At high temperatures, when the thermal rate is high, the particle is in the superparamagnetic regime. At low temperature, as far as the thermal rate is concerned, the magnetic moment should freeze along one of the anisotropy directions. In particles of sufficiently small size, however, even at $T = 0$ the magnetic moment can switch due to quantum tunneling. The quantum switching rate, Γ, scales with the total spin S of the nanoparticle according to [4–8] $\ln\Gamma \propto -S$. If the switching time, Γ^{-1}, is small compared to the measuring time, the nanoparticle remains superparamagnetic in the limit of $T = 0$. If, in addition, the interaction of **M** with microscopic degrees of freedom (phonons, itinerant electrons, nuclear spins, etc.) is small, the nanoparticle, during a certain decoherence time, can exist in a coherent quantum superposition of two classical states, $\Psi_o = |\uparrow\rangle + |\downarrow\rangle$. In that state the probability that the moment of the particle has a certain orientation oscillates in time as $\cos(\Delta \cdot t/\hbar)$, where Δ is the tunneling splitting. There is some recent experimental evidence of high-frequency coherent quantum spin oscillations in Fe_8 and Mn_{12} molecular nanomagnets of spin 10 [9]. So far, a quantum superposition of truly macroscopic states has been observed only for the flux states in a SQUID [10].

For a magnetic particle of considerable size to be in the quantum superparamagnetic (not necessarily coherent) regime, the energy barrier between the $|\uparrow\rangle$ and $|\downarrow\rangle$ states must be made sufficiently small. Since the barrier scales with the size of the particle, and very small magnetic particles are difficult to measure, the size of the barrier has been the major obstacle in the study of tunneling and coherence in small particles. One way to decrease the barrier is to use an external magnetic field. This method has been used in experiments on magnetic particles and molecular clusters performed to date [9,11–13]. It has a clear drawback if one attempts to create a coherent superposition of the $|\uparrow\rangle$ and $|\downarrow\rangle$ states. Namely, the external field, unless it is applied exactly perpendicular to the anisotropy axis, removes the degeneracy between the $|\uparrow\rangle$ and $|\downarrow\rangle$ states [14]. If the corresponding energy bias is greater than Δ (which requires only a very weak field for a macroscopic system), **M** becomes localized in the $|\uparrow\rangle$ or the $|\downarrow\rangle$ state. In this Letter, we suggest a method of controlling the barrier without breaking the degeneracy. This method is illustrated in Fig. 1. The nanoparticle is placed above the surface of a superconductor at a variable distance controlled by, e.g., a piezoelectric layer or holder. The current induced in the superconductor creates the magnetic image of the nanoparticle. The interaction between the nanoparticle and the superconductor is then equivalent to the dipole interaction between the nanoparticle and its image. The reduction of the barrier is similar to that from an external magnetic field applied opposite to **M**. However, contrary to the situation with the external field, the system (the nanoparticle plus the superconductor) is now degenerate with respect to $\mathbf{M} \to -\mathbf{M}$. It should be emphasized that such a degeneracy is a very general property of the system that is independent of the shape of the particle and the landscape of the superconducting surface. It is rooted in the time-reversal symmetry of the system in the absence of the field. The tunneling rate for the situation shown in Fig. 1 will be computed below.

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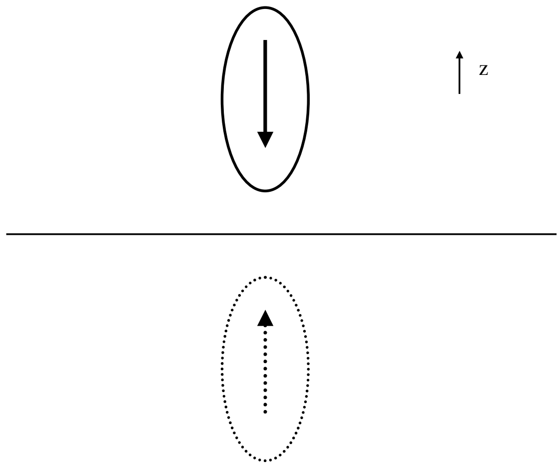

FIG. 1. Magnetic particle above the surface of a superconductor. The effect of the superconductor on the particle is equivalent to the field from the image shown in the figure.

A high tunneling rate does not automatically provide the coherent superposition of quasiclassical states. Different mechanisms of decoherence due to interactions inside the nanoparticle have been worked out in recent years [15–17]. At low temperature, in the absence of nuclear spins and itinerant electrons, the effect of dissipation on the tunneling rate can be very small [1]. Decoherence is a more subtle issue. Generally speaking, macroscopic quantum coherence (MQC), that is, $\cos(\Delta \cdot t/\hbar)$ oscillations of $\mathbf{M}$, occur only if the decohering interactions are small compared to Δ. We will show that this condition can be satisfied at least as far as the interaction between the nanoparticle and the superconductor is concerned. This question is of interest also in the more general context of measuring the rotation of a mesoscopic spin with the help of a superconducting device. Indeed, most of the experiments on individual nanoparticles used SQUIDs. Although the problem studied here is different from those experimental situations, some of the ideas should also apply to those cases.

Let us consider tunneling in a nanoparticle above the flat surface of a superconductor [18], as shown in Fig. 1, in the absence of dissipation. The external magnetic field will be considered zero throughout this paper. To make the classical electrodynamics of the problem less cumbersome, we will make certain simplifying assumptions about the superconductor, the shape of the particle, and its magnetic anisotropy. None of them is important and any generalization can be studied along the same lines. First, we shall assume that the superconductor is in the Meissner regime; that is, the magnetic field at the surface of the superconductor does not exceed H_{c1}. We shall also assume that the characteristic geometrical dimensions of the problem, the size of the particle and its distance to the surface, are greater than the penetration depth λ_L and the coherence length ξ. In that case, it is a good approximation to take the field of the particle at the surface of the superconductor to be parallel to that surface. The effect of the superconductor on the particle is then equivalent to the magnetic dipole interaction with the image shown in Fig. 1.

Next we assume that the particle is of ellipsoidal shape (that is, uniformly magnetized) with crystal fields either small or dominated by the single-ion anisotropy. The total energy of the magnetic anisotropy of such a particle must be quadratic in the magnetization [19],

$$E_{an} = \frac{2\pi}{V} N_{ik} M_i M_k \,, \tag{1}$$

where $\mathbf{M}$ is the total magnetic moment of the particle, V is its volume, and the tensor N_{ik} includes both the demagnetizing effect (that is, shape anisotropy) and the magnetocrystalline anisotropy. In a ferromagnetic particle, M is proportional to V, while N_{ik} is independent of V. The factor V^{-1} in Eq. (1) is, therefore, needed to provide the correct linear scaling of E_{an} with V. We shall assume that the principal axes of N_{ik} coincide with the coordinate axes in Fig. 1, with Z being the easy magnetization direction and $N_{xx} > N_{yy} > N_{zz}$. The magnitude of $\mathbf{M}$ is assumed, as usual, to be formed by a strong exchange interaction and, thus, independent of the orientation. That is, $M_x^2 + M_y^2 + M_z^2 = M^2 = \text{const}$. Recent experiments [20] have demonstrated that small-enough particles, despite their complex structure, flip their magnetization via uniform rotation. The energy of the magnetic anisotropy then becomes

$$E_{an} = \frac{1}{V}(\beta_x M_x^2 - \beta_z M_z^2) \,, \tag{2}$$

where β_x, β_z are positive dimensionless coefficients of order unity.

Equation (2) describes a magnet having a YZ easy magnetization plane with Z being the easy axis in that plane. The two degenerate minima of Eq. (2) correspond to $\mathbf{M}$ looking along and opposite to the Z axis. In the Meissner state of the superconductor, the magnetic field of the particle induces superconducting currents whose field is equivalent to the field of the image shown in Fig. 1. As the particle moves closer to the superconductor, the interaction between the particle and its image increases and the barrier between the two equilibrium orientations of $\mathbf{M}$ decreases. The magnetic moment of the particle, $\mathbf{M}$, and the moment of the image, $\mathbf{m}$, are related through

$$M_x = m_x, \qquad M_y = m_y, \qquad M_z = -m_z \,. \tag{3}$$

With reasonable accuracy the energy of the magnetic dipole interaction between the particle and its image is given by

$$E_{int} = \frac{[\mathbf{M}\cdot\mathbf{m} - 3(\mathbf{n}\cdot\mathbf{M})(\mathbf{n}\cdot\mathbf{m})]}{(2d)^3} \,, \tag{4}$$

where d is the distance from the (center of the) particle to the surface of the superconductor. With the help of relations (3), one obtains [18]

$$E_{int} = \frac{M_z^2}{(2d)^3} \,. \tag{5}$$

The total energy of the system, $E = E_{an} + E_{int}$, then becomes

$$E = \frac{1}{V}(-\beta_z \epsilon M_z^2 + \beta_x M_x^2), \quad (6)$$

where we have introduced $\epsilon = 1 - V/\beta_z(2d)^3$.

According to Eq. (6) the energy barrier between the degenerate $|\Uparrow\rangle$ and $|\Downarrow\rangle$ states of the particle is given by $U = \beta_z \epsilon M_o^2 V$, where $M_o = M/V$ is the volume magnetization of the particle. Our main idea is to manipulate d in such a way that ϵ and, consequently, U become small enough to provide a significant tunneling rate. Note that most deviations from the simplifying assumptions made above will renormalize $\beta_{x,z}$ and d in Eq. (6) but will not change the form of the total energy. It is convenient to introduce the total dimensionless spin of the particle, $\mathbf{S} = \mathbf{M}/\hbar\gamma$ (γ being the gyromagnetic ratio), and two characteristic frequencies,

$$\omega_\parallel = \beta_z \epsilon \gamma M_o, \qquad \omega_\perp = \beta_x \gamma M_o. \quad (7)$$

The total energy can then be written as

$$E = \frac{1}{S}[-\hbar\omega_\parallel S_z^2 + \hbar\omega_\perp S_x^2]. \quad (8)$$

The corresponding tunneling problem has been studied by a number of authors [5–8]. In the limit of $\omega_\parallel \ll \omega_\perp$, i.e., at $\epsilon \ll 1$, the tunneling splitting at $T = 0$ in the absence of dissipation is given by $\Delta = A_o \exp(-B_o)$ with

$$A_o = \frac{16}{\sqrt{\pi}} S^{1/2} \omega_\parallel^{3/4} \omega_\perp^{1/4}, \qquad B_o = 2S\left(\frac{\omega_\parallel}{\omega_\perp}\right)^{1/2}. \quad (9)$$

So far, we have neglected nondissipative terms in the total energy that come from the superconductor. One such term is the kinetic energy of the Cooper pairs,

$$E_{sc1} = \int d^3r \, \frac{n_s m \mathbf{v}_s^2}{2}, \quad (10)$$

where n_s is the concentration of superconducting electrons, m is their mass, and $\mathbf{v}_s = \mathbf{j}_s/en_s$ is their drift velocity expressed in terms of the superconducting current j_s. Equation (10) can be written as

$$E_{sc1} = \frac{4\pi\lambda_L^2}{c^2} \int d^3r \, \mathbf{j}_s^2, \quad (11)$$

where $\lambda_L = mc^2/4\pi e^2 n_s$ is the London penetration depth. The superconducting current is concentrated near the surface, resulting in the surface current [19]

$$\mathbf{g}_s = \int dz \, \mathbf{j}_s = \frac{c}{4\pi} \mathbf{n} \times \mathbf{B}(\mathbf{r}), \quad (12)$$

where $\mathbf{n}$ is the unit vector in the Z direction and $\mathbf{B}(\mathbf{r})$ is the sum of the dipole fields of the magnetic particle and its image at $z = 0$. A somewhat tedious but straightforward calculation then gives

$$E_{sc1} = \frac{\lambda_L}{8\pi} \int d^2r \, \mathbf{B}^2(\mathbf{r}) = \text{const} + \frac{3\lambda_L}{16d^4} M_z^2. \quad (13)$$

The contribution of the kinetic energy of the superconducting electrons to the total energy is small if $d \gg \lambda_L$. Even at $d \sim \lambda_L$, however, it reduces to the renormalization of the d dependence of the interaction between the magnetic particle and the superconductor. Since we are interested in d close to the critical value at which the barrier becomes zero, the form of the Hamiltonian, Eq. (8), and the expressions, Eq. (9), for the tunneling rate remain unaffected by that renormalization.

The next nondissipative term in the energy is the inertia coming from the energy of the electric field,

$$E_{sc2} = \int d^3r \, \frac{\mathbf{E}^2}{8\pi} = \frac{1}{8\pi c^2} \int d^3r \left(\frac{d\mathbf{A}}{dt}\right)^2. \quad (14)$$

where $\mathbf{A} = -(4\pi\lambda_L^2/c)\mathbf{j}_s$ is the vector potential in the superconductor. Then, similar to the previous case, one obtains

$$E_{sc2} = \frac{\lambda_L^3}{16\pi c^2} \int d^2r \left(\frac{d\mathbf{B}}{dt}\right)^2 = \frac{3\lambda_L^3}{32c^2 d^4}(\dot{\mathbf{M}}^2 + \dot{M}_z^2). \quad (15)$$

To estimate the effect of this inertia term on tunneling, it should be compared, at the instanton frequency [1] $\omega_{\text{inst}} = (\omega_\parallel \omega_\perp)^{1/2}$, with E of Eq. (8). The ratio of the energies is

$$E_{sc2}/E \sim (\lambda_L/d)^3 (l/d) (l/\lambda_{\text{inst}})^2, \quad (16)$$

where we have introduced $l = V^{1/3}$ and $\lambda_{\text{inst}} = c/\omega_{\text{inst}}$. Even if the experimental values of l, d, and λ_L do not differ in order of magnitude, λ_{inst} can hardly be less than 1 cm, making the ratio of energies in Eq. (16) negligible for nanoparticles used in tunneling experiments. We may then conclude that Eq. (9) gives a good estimate of the tunneling rate in the absence of dissipation.

Spin tunneling with dissipation due to the interaction of $\mathbf{M}$ with microscopic degrees of freedom inside the nanoparticle has been intensively studied [15,16]. Interactions with phonons, magnons, nuclear spins, etc., have been considered. Here we will study the mechanism of dissipation specific to our problem: the interaction of the magnetic moment with normal quasiparticles in the superconductor. This analysis may also be relevant to experiments on spin tunneling performed using SQUIDs.

We begin with the derivation of the energy dissipation Q in the superconductor due to the rotation of $\mathbf{M}$. With the help of the relations $\mathbf{E} = -c^{-1}\partial\mathbf{A}/\partial t$ and $\mathbf{j} = \sigma\mathbf{E}$, one obtains

$$Q = \int d^3r \, \mathbf{j} \cdot \mathbf{E} = \frac{1}{c^2} \int d^3r \, \sigma(t) \dot{\mathbf{A}}^2. \quad (17)$$

where σ is the conductivity due to quasiparticles. Its Fourier transform can be approximated as $\sigma_\omega = e^2 n_q / m(\nu + i\omega)$, where n_q is the quasiparticle concentration and ν is their scattering rate. The latter is typically 2–3 orders of magnitude greater than the instanton frequency for the tunneling of $\mathbf{M}$. Consequently, the time dependence of σ in Eq. (17) can be neglected and one can use $\sigma = e^2 n_q(T)/m\nu$. Equation (17) then becomes similar to Eq. (14) and the same argument gives the surface integral that is proportional to $(\dot{\mathbf{M}}^2 + \dot{M}_z^2) \propto [\dot{\theta}^2 + (\dot{\theta}^2 + \dot{\phi}^2)\sin^2\theta]$. Because of the smallness of ϵ, the hard-axis anisotropy in Eq. (8) is large compared to the easy-axis anisotropy. Under this condition, quasiclassical trajectories of $\mathbf{M}$ must be close to the easy plane. This means $\phi \approx \pi/2$, while θ for the tunneling trajectory changes from 0 to π. A more rigorous analysis shows that $\dot{\phi}^2 \sim \epsilon\dot{\theta}^2$. Thus, with good accuracy,

$$Q = \frac{\sigma\lambda_L^3}{2c^2}\int d^2r\,\dot{\mathbf{B}}^2 = \frac{3\lambda_L M^2}{16\nu d^4}\left(\frac{n_q}{n_s}\right)\dot{\theta}^2(1 + \sin^2\theta)\,. \tag{18}$$

If Eq. (18) were quadratic in $\dot{\theta}$, it could be interpreted as linear dissipation with a friction coefficient $\eta = 3\lambda_L M^2 n_q/16\nu d^4 n_s$. This would allow the Caldeira-Leggett [2] approach to tunneling with dissipation. The dissipation in the rotation of $\mathbf{M}$ due to its interaction with quasiparticles is nonlinear in θ, however. Nevertheless, since we want to obtain only an estimate of the effect of dissipation on tunneling, and because the $\sin^2\theta$ term in Eq. (18) can hardly change this effect significantly, we shall go ahead and estimate the effective Caldeira-Leggett action as

$$I_{\mathrm{CL}} = \frac{\eta}{4\pi}\int_{\infty}^{\infty} d\tau' \int_{o}^{\infty} d\tau\,\frac{[\theta(\tau) - \theta(\tau')]^2}{(\tau - \tau')^2}\,. \tag{19}$$

Note that the double integral in Eq. (19) is dimensionless.

The measure of the effect of dissipation on tunneling is the ratio $I_{\mathrm{CL}}/\hbar B_o$ where $\hbar B_o$ is the effective action in the absence of dissipation, with B_o given by Eq. (9). After simple algebra, one obtains

$$\frac{I_{\mathrm{CL}}}{\hbar B} \sim \frac{3\pi}{128\sqrt{\epsilon}}\left(\frac{n_q}{n_s}\right)\left(\frac{\lambda_L}{d}\right)\left(\frac{l}{d}\right)^3\left(\frac{\gamma M_o}{\nu}\right). \tag{20}$$

In a typical experiment, one should expect $\lambda_L < d$ and $l < d$, while the ratio $\gamma M_o/\nu$ may hardly exceed 10^{-2}. Consequently, even at $T \sim T_c$, when $n_q \sim n_s$, the effect of the dissipation on the tunneling rate may become visible only at ϵ less than 10^{-5}.

A more rigorous approach along the lines of Ref. [21] shows that the ratio n_q/n_s in Eqs. (18)–(20) should be replaced by the ratio of the coherence factors. Similar to other dissipation problems due to quasiparticles [21], its effect on tunneling must have a maximum at T slightly lower than T_c. Even at the maximum this effect should still be small. Coherence is a more subtle issue. It may be destroyed by a dissipative environment even if the effect of dissipation on tunneling is weak. The mechanism of dissipation discussed above goes down with temperature as $\exp(-\Delta_{sc}/T)$, where Δ_{sc} is the superconducting gap. Consequently, preserving coherent oscillations of $\mathbf{M}$ between $|\uparrow\rangle$ and $|\downarrow\rangle$ states requires $T \ll \Delta_{sc}$.

For actual experiments, Nb is a nearly ideal material for the superconducting surface. It has a large value of $\Delta_{sc} = 18$ K and its surface can be easily polished. Nb also has a small coherence length, $\xi = 38$ nm, and small penetration depth, $\lambda_L \sim 40$ nm. Thus, it can significantly reduce the barrier in a magnetic nanoparticle of comparable dimensions. Another advantage of Nb is that at low temperature its Meissner phase extends to relatively high magnetic fields, $H_{c1} = 1.85$ kOe. The Meissner regime is not destroyed by the proximity of a magnetic particle if its volume magnetization M_0 does not exceed $H_{c1}/4\pi$, which is about 150 emu for Nb. Such particles are readily available. To reduce dissipation inside the particle itself, it should be made of a dielectric material to avoid any electric currents induced by $d\mathbf{M}/dt$. The particle also should be purified isotopically to eliminate the effect of nuclear spins.

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