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MACROSCOPIC QUANTUM TUNNELING IN ANTIFERROMAGNETS

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Starting from the Néel state of a uniaxial antiferromagnetic particle, we show that, due to the tunneling of the Néel vector between easy directions, the ground state of a sufficiently small particle is a quantum superposition of two equivalent Néel states. A certain orientation of the Néel vector becomes frozen as the volume of the particle grows, or the dissipation due to the interaction of the Néel vector with microscopic degrees of freedom increases. For the weak dissipation, which is mostly the case, the crossover from classical to quantum regime occurs at temperature $T^* \sim (\epsilon_a/\epsilon_e)^{1/2} T_N$, where ϵ_a and ϵ_e are anisotropy and exchange constants, T_N is the Néel temperature.

Observation of the quantum behaviour of a macroscopic variable has remained a challenging problem [1] since Schrödinger [2] first discussed it in 1935. To date the only macroscopic systems where significant progress has been made are superconductors. Quantum tunneling between two macroscopically distinct current states, observed in superconductors, is in amazing agreement with theoretical predictions [3]. Recently another field for the study of the macroscopic quantum tunneling (MQT) has been suggested [4,5]. It has been shown that in ferromagnets, a macroscopically large number of spins, coupled by a strong exchange interaction, can coherently tunnel through the energy barrier created by magnetic anisotropy. This effect may reveal itself in the low temperature magnetic relaxation in bulk materials [5–7], or in quantum tunneling of the magnetic moment between easy directions in monodomain ferromagnetic particles [4,8]. In this paper we show that a similar effect exists in antiferromagnetic particles where it may be much greater than in ferromagnets.

In a simplest version of the Néel Model, an antiferromagnetic lattice consists of two ferromagnetic sublattices. Their magnetizations have a fixed length M_0, and, in the absence of the magnetic field, are opposite to each other, $\boldsymbol{M}_1 = -\boldsymbol{M}_2$, so the total magnetization is zero [9]. Antiferromagnetic order is characterized by the Néel vector of a unit length,

$$\boldsymbol{l} = \frac{\boldsymbol{M}_1 - \boldsymbol{M}_2}{2M_0} . \tag{1}$$

Here we are interested in the quantum tunneling of $\boldsymbol{l}$ between two opposite orientations, $|1\rangle = |\uparrow\downarrow\rangle$ and $|-1\rangle = |\downarrow\uparrow\rangle$. Due to this effect a small antiferromagnetic particle must turn to a state with uncertain orientation of the Néel vector,

$$|0\rangle = \frac{1}{\sqrt{2}} (|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle) . \tag{2}$$

As the volume of the particle grows, or the interaction of the Néel vector with microscopic degrees of freedom increases, a certain orientation of $\boldsymbol{l}$ becomes frozen. We believe that a nice mathematical approach to this problem, presented below, may be of general interest for the problem of the ground state of antiferromagnet.

In the absence of the external field and weak Dzyaloshinsky ferromagnetism [#1] the Lagrangian of the uniaxial antiferromagnet is [9]

$$L=\int \mathrm{d}^3x\left[\frac{\chi_\perp}{2\gamma^2}\left(\frac{\mathrm{d}\boldsymbol{l}}{\mathrm{d}t}\right)^2-\frac{\alpha}{2}\left(\frac{\partial l_i}{\partial x_j}\right)^2+\tfrac{1}{2}K(\boldsymbol{n}\cdot\boldsymbol{l})^2\right], \tag{3}$$

where $\chi_\perp$ is the perpendicular susceptibility (with respect to the equilibrium orientation of $\boldsymbol{l}$ along the anisotropy axis $\boldsymbol{n}$), $\gamma=e/mc$, α and K are exchange and anisotropy constants correspondingly. For a small particle $\boldsymbol{l}$ may depend on time but not on coordinates because large spatial derivatives of $\boldsymbol{l}$ are supressed by the exchange interaction. Then representing $\boldsymbol{l}$ in terms of angles θ and ϕ in a spherical coordinate system ($\boldsymbol{n}\cdot\boldsymbol{l}=\cos\theta$), we obtain eq. (3)

$$L=V\left\{\frac{\chi_\perp}{2\gamma^2}\left[\left(\frac{\mathrm{d}\theta}{\mathrm{d}t}\right)^2+\left(\frac{\mathrm{d}\phi}{\mathrm{d}t}\right)^2\sin^2\theta\right]-\tfrac{1}{2}K\sin^2\theta\right\}, \tag{4}$$

where V is the volume of the particle; a constant term, $-\frac{1}{2}KV$, is added for convenience.

Equilibrium orientations of $\boldsymbol{l}$ are $\theta=0$ and $\theta=\pi$ which correspond to the degenerate classical minimum of the energy, $E=0$. The rate of quantum transitions between these states is proportional to the path integral

$$\int \mathrm{D}\{\theta(t)\}\int \mathrm{D}\{\phi(t)\}\exp\left(\frac{\mathrm{i}}{\hbar}\int \mathrm{d}t\,L\right). \tag{5}$$

It may be represented as

$$\Gamma=A\exp(-B/\hbar), \tag{6}$$

where the imaginary time ($\tau=\mathrm{i}t$) action, B, is deduced from the Euclidean action,

$$I=V\int \mathrm{d}\tau\left\{\frac{\chi_\perp}{2\gamma^2}\left[\left(\frac{\mathrm{d}\theta}{\mathrm{d}\tau}\right)^2+\left(\frac{\mathrm{d}\phi}{\mathrm{d}\tau}\right)^2\sin^2\theta\right]+\tfrac{1}{2}K\sin^2\phi\right\}, \tag{7}$$

evaluated along the quasiclassical path $\{\theta(\tau), \phi(\tau)\}$ which connects $\theta=0$ and $\theta=\pi$. The equations of motion following from eq. (7) are

$$\frac{\mathrm{d}\phi}{\mathrm{d}\tau}\sin^2\theta=\text{const}, \tag{8}$$

$$\frac{\chi_\perp}{\gamma^2}\frac{\mathrm{d}^2\theta}{\mathrm{d}\tau^2}=\left[K+\frac{\chi_\perp}{\gamma^2}\left(\frac{\mathrm{d}\phi}{\mathrm{d}\tau}\right)^2\right]\sin\theta\cos\theta. \tag{9}$$

Consequently, a quasiclassical rotation of $\boldsymbol{l}$ may occur in any plane $\phi=$const, and satisfies a sine-Gordon equation,

$$2\frac{\mathrm{d}^2\theta}{\mathrm{d}\tau^2}=\omega_0^2\sin 2\theta, \tag{10}$$

where $\omega_0=\gamma(K/\chi_\perp)^{1/2}$. There is an exact solution of this equation,

$$\theta(\tau)=2\arctan[\exp(\omega_0\tau)], \tag{11}$$

which corresponds to a subbarrier rotation of $\boldsymbol{l}$ from $\theta=0$ at $\tau=-\infty$ to $\theta=\pi$ at $\tau=+\infty$. Evaluating I along this trajectory we obtain for the WKB exponent

$$\frac{B}{\hbar}=\frac{\sqrt{\chi_\perp K}}{\mu_\mathrm{B}}V, \tag{12}$$

where $\mu_\mathrm{B}=\gamma\hbar/2$ is the Bohr magneton. The exact value of the prefactor in eq. (6) is less significant but much more difficult to obtain. It has the form $A=\omega_0 f(B/\hbar)$, where $f(x)$ is a slow (not exponential) dimensionless function of x.

The most essential dependence of the tunneling rate on the volume comes from the exponent (12). The latter must be sufficiently small (say $B/\hbar\leqslant 25$ as accepted, e.g., in the theory of superparamagnetism [10]) to observe the transitions for a reasonable time. According to eq. (12), for typical numbers, $\chi_\perp\sim 10^{-4}$, $K\sim 10^6$ erg/cm^3, quantum tunneling may be significant in particles of diameter $d\leqslant 20$ Å. It should not be a problem since antiferromagnetic particles of that size have been already studied experimentally [11]. Notice, that such particles still contain a macroscopically large number of spins, $N\sim 10^3$. For particles of much greater size the tunneling rate is so small that at zero temperature a certain orientation of the Néel vector becomes frozen.

At high temperature the rate of transitions between different orientations of $\boldsymbol{l}$ is dominated by thermal fluctuations,

[#1] Small magnetic moments due to non-compensating effects are discussed in the end of the paper.

$$\Gamma' = A' \exp(-U/k_B T) , \quad (13)$$

where $U = \frac{1}{2}KV$ is the energy barrier due to magnetic anisotropy. Quantum transitions dominate at $T < T^*$, where T^* satisfies $U/k_B T^* = B/\hbar$. This gives

$$T^* = \frac{\mu_B}{k_B} \sqrt{\frac{K}{\chi_\perp}} . \quad (14)$$

For $\chi_\perp \sim 10^{-4}$, $K \sim 10^6$ erg/cm^3, T^* is of the order of a few Kelvin. In antiferromagnetic materials with lower anisotropy, quantum effects should reveal themselves at lower temperature but in particles of a greater size.

The calculated temperature of the crossover from thermal to quantum regime in antiferromagnets is by one or two orders of magnitude greater than in ferromagnets. It becomes obvious when T^* is expressed in terms of anisotropy and exchange energies per spin, $\epsilon_a \sim \mu_B/M_0$, $\epsilon_e \sim k_B T_N$, where T_N is the Néel temperature. Since magnetic anisotropy is due to relativistic interactions, the ratio ϵ_a/ϵ_e is generally small, typically of the order of 10^{-5}–10^{-4} in 3d transition magnetic systems. Taking for estimation [12] $\chi_\perp \sim \mu_B M_0/k_B T_N$, we obtain

$$T^* \sim \left(\frac{\epsilon_a}{\epsilon_e}\right)^{1/2} T_N . \quad (15)$$

A similar calculation for the tunneling of magnetization through the anisotropy barrier in ferromagnets [4,5] gives $T^* \sim (\tilde{\epsilon}_a/\epsilon_e) T_c$, where $\tilde{\epsilon}_a$ is the geometrical average for the longitudinal and transversal anisotropy, $\epsilon_e \sim k_B T_c$, T_c is the Curie temperature. It shows that quantum fluctuations in antiferromagnets are much stronger than in ferromagnets.

Now let us consider the effect of dissipation on the tunneling rate. The rate of the heat production in antiferromagnets, due to the uniform rotation of $\boldsymbol{l}$, is [12]

$$Q = \frac{M_0}{2\gamma} \int d^3x\, \lambda \dot{\boldsymbol{l}}^2 = \frac{\lambda M_0}{2\gamma} V \dot{\boldsymbol{l}}^2 , \quad (16)$$

where λ is a dissipation constant whose value may be obtained experimentally [13]. According to Leggett [13] eq. (16) allows one to obtain an effective action for the tunneling process, $\tilde{I} = I + I_{dis}$, where

$$I_{dis} = \frac{\lambda M_0}{8\pi\lambda} V \int d\tau \int d\tau' \frac{[\boldsymbol{l}(\tau) - \boldsymbol{l}(\tau')]^2}{(\tau - \tau')^2} \quad (17)$$

is responsible for the interaction of $\boldsymbol{l}$ with microscopic degrees of freedom. Analysis of the extremal trajectory corresponding to $\tilde{I}$ shows that a good approximation for the WKB exponent in the presence of dissipation is $\tilde{B} = B_0(1+\beta)$, where

$$\beta \sim \frac{\lambda}{4\pi} \frac{M_0}{\sqrt{\chi_\perp K}} \sim \frac{\lambda}{4\pi} \left(\frac{\epsilon_e}{\epsilon_a}\right)^{1/2} . \quad (18)$$

Dissipation is important when β is large, which possibly may occur in metals but certainly is not the case in non-metallic antiferromagnets. Note that strong dissipation may result in quenching of the Néel state even in very small particles [#2].

In antiferromagnetic particles of random shape, magnetic moments of sublattices are not totally compensated. For a particle of N spins, $N^{2/3}$ spins are at the surface. Consequently, the number of non-compensated spins due to statistical fluctuations of the shape is $(N^{2/3})^{1/2} = N^{1/3}$. In a particle of 10^3 spins this would produce a magnetic moment of $10\mu_B$. The moment may be greater if a large area of the surface coincides with a crystallographic plane [11]. Small magnetic moments, $M \sim 10^{-3} N\mu_B$, may be also generated by the weak Dzyaloshinsky ferromagnetism. We believe that there are situations where both effects are small enough not to affect significantly our results. At the same time they can be used for the experimental study of MQT in antiferromagnets. As the Néel vector tunnels through the barrier, a non-compensated magnetic moment switches its direction. One can, therefore, study a low temperature magnetic relaxation in antiferromagnetic particles whose magnetic moments were initially oriented by the magnetic field. In such an experiment MQT would reveal itself in the temperature independent relaxation rate below T^*.

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[#2] Qualitatively, the effect of dissipation on the ground state of antiferromagnet has been discussed in terms of MQT in ref. [14].

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