cussion of the procedure of obtaining flat intercrystalline boundaries, to V. F. Gantmakher for a discussion of the results, and to P. L. Kapitza for interest in the work.

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Quantum effects in small ferromagnetic particles

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It is shown that at temperatures $T < 1/2(\varepsilon_a \varepsilon_e)^{1/2}$ (ε_a and ε_e are the anisotropy energy and the exchange energy per magnetic electron) effects related with coherent quantum magnetization fluctuations should appear in sufficiently small ferromagnetic particles.

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1. INTRODUCTION

It is well known (see, for example, Ref. 1) that a sufficiently small particle of ferromagnetic material consists of a single magnetic domain whose magnetization whose orientation is determined by the magnetocrystalline anisotropy of the particles and by the external magnetic field **H**. In the absence of a field, the easiest magnetization of a ferromagnetic particle has several orientations separated by energy barriers. If the temperature of the particle T is comparable with the size of the barrier U_a, then transitions between favorable orientations of the magnetization arise in the particle.

The existence of experimental data indicating that transitions between different orientations of the magnetic moment in small ferromagnetic particles do not disappear completely with a decrease in temperature to absolute zero was noted as far back as in the review by Bean and Livingston.[2] Nevertheless, as far as we know, quantum fluctuations in the magnetization of small particles has not been studied theoretically.

Turning to the study of this problem, we note first that a good approximation to the problem can be obtained if we limit ourselves to coherent magnetic-moment quantum fluctuations in which only the direction and not the magnitude of magnetization changes. This approximation is valid if the energy E_a necessary for the spin flip of one of the magnetic electrons in the particle significantly exceeds the energy $u_a = \varepsilon_a N$ (ε_a is the magnetic anisotropy energy per magnetic electron, N is the number of magnetic electrons in the small particle) required to rotate the particle magnetic moment as a whole

$$\varepsilon_a N \ll \varepsilon_e. \tag{1}$$

Because of the smallness of the ratio $\varepsilon_a/\varepsilon_e \sim 10^{-4} - 10^{-6}$ this condition is satisfied for ferromagnetic particles that are small but still contain a macroscopically large number of atoms.

The magnetic anisotropy energy is the average energy of the relativistic interaction of the electrons. In taking this interaction into account, the operator of the spin angular momentum does not commute with the Hamiltonian and, in general, the ferromagnetic particle is not in a state with a definite angular momentum. This circumstance should manifest itself by the appearance of a nonzero probability for a transition between n easiest-magnetization directions. Such transitions lift the n-fold degeneracy of the ground state with respect to the orientation of the angular momentum, and the ferromagnetic particle goes to a state with a lower energy, in which

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

(M_0 is the saturation magnetic moment). The second equality in (2) corresponds to strict ferromagnetic correlation of relative orientations of the spins of different magnetic electrons of the particle.

2. QUANTUM FLUCTUATIONS AT $H = 0$ AND $T = 0$

We consider a single-domain ferromagnetic particle with a magnetic anisotropy of the easy axis type (for example, a particle of hexagonal cobalt) rigidly fixed in a nonmagnetic matrix. The dependence of the energy E_a of the particle on the angle θ between the direction of its magnetic moment M and the anisotropy axis z is given by the equation

$$E_a = \varepsilon_a N \sin^2 \theta. \tag{3}$$

The presence of two energy minima at $\theta = 0$ and $\theta = \pi$ corresponds to the two easiest magnetization directions,

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along and against the direction of the anisotropy axis.

Let a measurement of the projection of the magnetization on the z axis at the time $t=-A$ give the value $M_z(-A)$. Then the probability amplitude that at the time $t=A$ the measured orientation of the magnetization will be $M_z(A)$ is determined by the integral[3]

$$\langle M_z(-A)M_z(A)\rangle \sim \int D\{\mathbf{M}(t)\}\exp\left(\frac{i}{\hbar}\int d^3x\int_{-A}^{A} dt\,L\{\mathbf{M}(t)\}\right) \quad (4)$$

along all trajectories of the variations of the magnetization $\mathbf{M}(t)$ that satisfy the initial and final conditions indicated above (L is the Lagrangian density of the system). We describe these trajectories by the three Euler angles $\theta(t)$, $\varphi(t)$, and $\psi(t)$, writing the Lagrangian of the magnetic system in the form of the Lagrangian of a spherical top in the anistropy field (3):

$$\mathscr{L}=\int d^3x\,L={}^1/_2J\dot\theta^2+{}^1/_2J\dot\varphi^2\sin^2\theta + {}^1/_2J(\dot\psi+\dot\varphi\cos\theta)^2-\varepsilon_aN\sin^2\theta-\varepsilon_eN, \quad (5)$$

where J is the effective moment of inertia (the last constant term is introduced for convenience).

The mechanical momentum of the top's own rotation

$$\mu=J(\dot\psi+\dot\varphi\cos\theta) \quad (6)$$

is connected with M_0 by the relation

$$M_0=\gamma\mu, \quad \gamma=ge/2mc \quad (7)$$

(g is the gyromagnetic ratio). The third term in (5), which is due to this rotation, equals the exchange energy of the particle

$$\mu^2/2J=\varepsilon_eN. \quad (8)$$

Equations (6) and (7), in accordance with what was said in the Introduction, restrict our analysis to $\mathbf{M}(t)$ trajectories on which the absolute value of the moment μ is equal to a specified value. We emphasize that this condition, being imposed on all the virtual trajectories and not only on trajectories satisfying the equations of motion, contains more than simply the conservation law for μ, which follows, according to (5), from the equation of motion. In paticular, the condition (7) decreases the number of independent dynamic variables and for this reason it is important for calculation of the continual integral in (4).

Using (7) and (8) we find that

$$J=M_0^2/2\gamma^2\varepsilon_eN. \quad (9)$$

One of the classes of trajectories, satisfying the condition of rigidity of the moment and the equation of motion, describes the well-known phenomenon of uniform precession of the moment in the anisotropy field (homogeneous ferromagnetic resonance):

$$\theta=\text{const}, \quad \dot\varphi=-4\varepsilon_a\hbar^{-1}\cos\theta. \quad (10)$$

We made use of the definition of N,

$$M_0={}^1/_2\gamma\hbar N \quad (11)$$

and neglected a small term of the order of $(\varepsilon_a/\varepsilon_e)^{1/2}$.

The minimum of the system energy

$$u={}^1/_2J\dot\theta^2+{}^1/_2J\dot\varphi^2\sin^2\theta+\varepsilon_aN\sin^2\theta \quad (12)$$

corresponds to $\theta=0$ and $\theta=\pi$. For uniform precession the first term in (12) equals zero, while the second is smaller than the last term by a factor $\varepsilon_a/\varepsilon_e$.

Let us note that $it=\tau$ the condition that μ remain constant leads, in addition to precession, to the appearance of extremal sub-barrier trajectories that satisfy the equation

$$0=-{}^1/_2J(d\theta/d\tau)^2+\varepsilon_aN\sin^2\theta, \quad \dot\varphi=0. \quad (13)$$

The single-instanton[4] solution of this equation, describing the transition from the state with $\theta=\pi$ to the state with $\theta=0$, is of the form

$$\theta=\arccos\left\{\text{th}\frac{\tau}{\tau_0}\right\}, \quad \tau_0=\frac{\hbar}{4(\varepsilon_a\varepsilon_e)^{1/2}}. \quad (14)$$

Calculating the Euclidean action on this trajectory, we get at $A\gg\tau_0$ ($\tau_0\sim10^{-12}$ sec), with exponential accuracy

$$S_\tau=\int_{-A}^{A} d\tau\,\mathscr{L}=-N\hbar(\varepsilon_a/\varepsilon_e)^{1/2}. \quad (15)$$

Trajectories close to the extremal trajectory (14) make the main contribution to the transition amplitude (4). A standard calculation[3] leads to the following expression for the transition probability per unit time:

$$W(0,\pi)\approx(T_q/\hbar)\exp\{-u_0/T_q\}, \quad (16)$$

where

$$T_q={}^1/_2(\varepsilon_a\varepsilon_e)^{1/2}. \quad (17)$$

Neglect of the multiple-instanton contributions, together with the condition (1) for the validity of the quasi-classical approach, limits the sizes of the ferromagnetic particles for which Eq. (16) is valid to values of N in the interval

$$(\varepsilon_e/\varepsilon_a)^{1/2}<N<\varepsilon_e/\varepsilon_a. \quad (18)$$

3. SINGLE-DOMAIN PARTICLE WITH $H=0$, $0<T<T_q$

Let us ascertain the extent to which allowance for the temperature $T<T_q$ influences the transition. At a temperature differing from zero, the continual

integral in (4) is defined[3] in the space of trajectories that are periodic in the parameter $\tau = it$ with a period T^{-1} (T is the temperature). From (12) it is easy to see that at $T < T_q$ the periodic trajectories include extremal sub-barrier trajectories that correspond to quasi-classical motion with some average energy $U \ll U_a$ and are described by the equation

$$u = -{}^1/_2 J (d\theta/d\tau)^2 + u_a \sin^2\theta. \tag{19}$$

The solution to this equation, of interest to us, is of the form

$$\theta = \arccos\{k\,\mathrm{sn}(\tau/\tau_0; k)\}, \tag{20}$$

where k is the modulus of the elliptic function,

$$k^2 = 1 - (u/u_a)^2. \tag{21}$$

The equality of the period of the elliptic function to the inverse temperature gives the connection between U and T:

$$4TK(k) = 1 \tag{22}$$

($K(k)$ is a complete elliptic integral of the first kind). In the limit $U \ll U_a$, using the asymptotic form of $K(k)$ as $k \to 1$, we have

$$u = 4u_a \exp\{-2T_q/T\}. \tag{23}$$

A calculation of the effective action on the extremal trajectory (20), within the limits determined by the period of the elliptic function T^{-1}, gives

$$S_T \equiv \oint d\tau\, \mathscr{L} = -\hbar \frac{u_a}{4T_q}[2E(k) + (k^2-1)K(k)] \tag{24}$$

($E(k)$ is a complete elliptic integral of the second kind).

Expanding the expression in the square brackets in powers of $1 - k^2$, with accuracy up to the first nonvanishing term in temperature, we have for the transition probability at $T < T_q$

$$W_T \sim \exp\left\{-\frac{u_a}{T_q}\left[1 - 4\exp\left(-\frac{4T_q}{T}\right)\right]\right\}. \tag{25}$$

A comparison of this formula with (16) shows that at $T < T_q$ the transition probability is described, with good accuracy, by the expression obtained at $T = 0$.

4. QUANTUM EFFECTS IN AN EXTERNAL MAGNETIC FIELD

We now determine the behavior of a single domain particle in an external magnetic field at $T < T_q$. For this purpose, we note that the lifting of the degeneracy on going to a state with $\langle \mathbf{M} \rangle = 0$ is accompanied by a decrease in the energy of the ferromagnetic particle by the amount

$$\Delta \sim T_q \exp\{-N(\varepsilon_a/\varepsilon_e)^{1/2}\}. \tag{26}$$

On the other hand, if the moment differs from zero ($\langle \mathbf{M} \rangle \neq 0$) and is directed along the external magnetic field, then the energy of the particle decreases by the amount

$$u_H = \langle \mathbf{M} \rangle \mathbf{H} \tag{27}$$

(the field is assumed to be applied along the anisotropy axis). For this reason, there exists a critical value of the magnetic field

$$H_c = \Delta / M_0, \tag{28}$$

at which a phase transition from the state with $\langle \mathbf{M} \rangle = 0$ to a state with $\langle \mathbf{M} \rangle = \mathbf{M}_0$ becomes favored. For particles of with minimum size, satisfying the condition (18), H_c is a maximum and of the order of the magnetic anisotropy field.

5. EXPERIMENTAL POSSIBILITIES

At $T > T_q$ the probability of thermal transitions between different easiest-magnetization directions is proportional to $\exp(-U_a/T)$. It was shown above that at $T < T_q$ the transitions are due to purely quantum processes and the temperature has a weak effect on their probability. Thus, satisfaction of the condition $T < T_q$ in an experiment is the first necessary condition for the observation of a quantum (in the sense (2) indicated above) as opposed to a thermal demagnetization of small ferromagnetic particles.

The second condition is a sufficiently large transition probability (16), which depends very strongly ($\sim e^{-r^3}$) on the radius r of the particle. To obtain a sufficiently large number of transitions during the experiment, it is sensible to subject the size of the particles to a restriction $u_a \lesssim 25\,T_q$, similar to that assumed in the theory of supermagnetism.[2] The temperature T_q is a universal parameter for a ferromagnetic substance. The quantities ε_a and ε_e contained in its definition can be taken directly from experiment (see, for example, Ref. 5).

In accordance with (15), quantum effects should manifest thermselves most readily in particles of ferromagnetic materials having a large exchange energy (high Curie temperature) and a small anisotropy. For hexagonal cobalt $T_q \sim 20$ K. The radius of the particle must not exceed $r_q \sim 15$ Å. If Eqs. (16) and (17) are applied to nickel and iron, which have a more complicated magnetic anisotropy, we obtain, respectively, $T_q \sim 2$ K, $r_q \sim 30$ Å and $T_q \sim 5$ K, $r_q \sim 20$ Å.

The main consequence of the quantum fluctuations is the above-described behavior of small ferromagnetic particles in an external magnetic field. In experiments with a large number of particles the study of this effect may be difficult because of the unavoidable differences in the particle sizes. Nevertheless, if the sizes of all ferromagnetic particles are restricted to r_q, the effect of the quantum fluctuations should manifest itself in an absence of magnetic hysteresis at $T < T_q$.

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