Quantum theory of nucleation in ferromagnets

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We consider a ferromagnet whose magnetization is opposite to an applied magnetic field. The rate of quantum nucleation is calculated for a film and is estimated for a bulk solid. The temperature corresponding to the crossover from thermal to quantum nucleation is estimated. The effect may be large enough to be observed in materials with high anisotropy constants.

The general approach to the problem of quantum nucleation of a stable phase from a metastable one consists in the consideration of the instanton solutions of the classical equations of motion for the order parameter.[1] In this manner, or by use of the equivalent Wentzel-Kramers-Brillouin (WKB) method, it has been carried out for different systems, from solids[2,3] to the inflationary universe,[4] but not for a ferromagnet, where the problem is complicated by a special form of the equation of motion for the magnetization. The formulation of the problem is the following: Consider a bulk ferromagnet uniformly magnetized along the easy axis. Magnetic anisotropy is assumed to be strong enough to neglect the probability of spontaneous domain formation in the absence of the magnetic field. Suppose now that a magnetic field **H** is applied in a direction opposite to the direction of the magnetization **M**. There exists a critical field H_c below which there is an energy barrier U between metastable (**M** is antiparallel to **H**) and stable (**M** is parallel to **H**) states. To convert the ferromagnet into a stable state one must overcome the barrier for a sufficiently large nucleus whose surface energy is less than the gain in the volume energy. Then the nucleus does not collapse, but unrestrictedly grows in volume. Of course, there may be also nucleation processes due to thermal fluctuations, with a rate $P \propto \exp(-U/k_BT)$, so that quantum effects can be observed only at low temperatures. Assuming for the quantum nucleation rate[5] $P \propto \exp(-B)$, one can introduce a "crossover temperature," $T_c = U/k_BB$, below which quantum nucleation dominates. To our knowledge, the only attempt to estimate the quantum nucleation rate belongs to Privorotskii,[6] who, based upon dimensional grounds, suggested $B = (M/\hbar\gamma)\delta_0^3 f$, where δ_0 is the domain wall width, $\gamma = ge/2mc$ (g is the gyromagnetic ratio), and f is an unknown dimensionless function of the magnetic field and anisotropy constants. In this paper we present what we believe is a rigorous theory of quantum nucleation in a ferromagnet, and point out the conditions under which the effect can be observed experimentally.

While the theory is applicable in a wide variety of situations, including different forms of magnetic anisotropy, nucleation around defects and surfaces, etc., in this paper we focus our attention solely on nucleation within a homogeneous region of a sample described by the simplest form of the magnetic anisotropy energy density

$$E_a = -k_{\parallel}M_z^2 + k_{\perp}M_y^2 - \mathbf{M}\cdot\mathbf{H} \ . \tag{1}$$

Up to a constant, Eq. (1), in a spherical coordinate system, is equivalent to

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

H being applied in the $-z$ direction. For $H < H_c = 2K_{\parallel}/M$, there are two local energy minima: $\theta = 0$ and $\theta = \pi$; the maximum at $\phi = 0$ (see Fig. 1) corresponds to $\cos\theta_1 = H/H_c$, and defines the barrier between the minima. Notice that transverse anisotropy $K_{\perp}$ is responsible for quantum transitions between the minima. If $K_{\perp} = 0$, M_z, as a quantum operator, commutes with E_a and, therefore, is a conserved quantum number.

Our theory of quantum nucleation is based upon an extension of the theory we have recently developed, using the instanton technique, for quantum tunneling of magnetization in small single-domain ferromagnetic particles[7] (see also, Refs. 8 and 9). For that problem the classical equation for **M**,[10]

$$\frac{\partial \mathbf{M}}{\partial t} = -\gamma \mathbf{M} \times \frac{\delta E_a}{\delta \mathbf{M}} \ , \tag{3}$$

is equivalent to two differential equations for θ and ϕ following from the action[11] for a sample volume V:

$$I = V \int dt \left[\frac{M}{\gamma}\dot{\phi}\cos\theta - E_a(\theta,\phi) \right] . \tag{4}$$

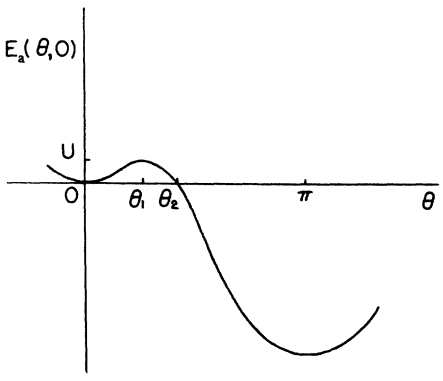

FIG. 1. The energy $E_a(\theta,\phi=0)$.

Using Eqs. (3) and (4), we found the exact instanton solution for the uniform subbarrier rotation of $\mathbf{M}$ between the energy minima, and calculated the extremal imaginary-time action S_E. The case of practical interest corresponds to the small barrier $U=K_{\parallel}V\varepsilon^2$ for small $\varepsilon=1-H/H_c$. In this case we found[7]

$$P\propto\exp\left[-\frac{S_E}{\hbar}\right]=\exp\left[-\frac{8MV}{3\hbar\gamma}\left[\frac{K_{\parallel}}{K_{\perp}}\right]^{1/2}\varepsilon^{3/2}\right]\equiv\exp\left[-\frac{U}{k_BT_c}\right], \tag{5}$$

where the crossover temperature is given by

$$T_c=\frac{3\hbar\gamma\sqrt{K_{\parallel}K_{\perp}}}{8k_BM}\sqrt{\varepsilon}\ . \tag{6}$$

Here we obtain the solution of the nucleation problem, together with the above result, using a slightly different, but straightforward method. For $\mathbf{M}=\mathbf{M}(\mathbf{x},t)$, the energy density includes the exchange term

$$E_e=\tfrac{1}{2}\alpha\left[\frac{\partial M_i}{\partial x_j}\right]^2=\tfrac{1}{2}\alpha M^2[(\nabla\theta)^2+\sin^2\theta(\nabla\phi)^2] \tag{7}$$

(α is the exchange stiffness). The equation of motion is given now by Eq. (3) with $E=E_e+E_a$ instead of E_a. Correspondingly,

$$I=\int dt\int d\mathbf{x}\left[\frac{M}{\gamma}\dot{\phi}\cos\theta-E(\theta,\phi)\right] \tag{8}$$

which is a simple reflection of the fact that $x=\phi$, $p=(M/\gamma)\cos\theta=\hbar S_z$ (S_z is the z projection of the spin density) are canonical variables, so that $L=p\dot{x}-E$ is the Lagrangian density of the system.[11] The amplitude of the quantum transition between two different configurations of $\mathbf{M}(\mathbf{x},t)$ is proportional to the path integral

$$\int D\{\theta(\mathbf{x},t)\}\int D\{\phi(\mathbf{x},t)\}\exp\left[\frac{i}{\hbar}\int dt\int d\mathbf{x}L(\theta,\phi)\right] \tag{9}$$

over all trajectories which connect the initial and final states. The case of practical interest again corresponds to the small barrier for small ε. In this case the shape of the barrier is characterized by $\theta_1=\sqrt{2\varepsilon}$, $\theta_2=2\sqrt{\varepsilon}$ (see Fig. 1), so one can attempt to treat the quantum domain of the problem within a small θ approximation,

$$L=\frac{M}{\gamma}\phi\theta\dot{\theta}-\tfrac{1}{2}\alpha M^2(\nabla\theta)^2-\tfrac{1}{2}\alpha M^2\theta^2(\nabla\phi)^2-K_{\parallel}(\varepsilon\theta^2-\tfrac{1}{4}\theta^4)-K_{\perp}\theta^2\sin^2\phi\ . \tag{10}$$

[We have represented $\dot{\phi}\cos\theta$ as $(d/dt)(\phi\cos\theta)-\phi(d/dt)\cos\theta$ and neglected the total time derivative.] It is convenient to use dimensionless variables $\mathbf{x}'=\mathbf{x}\sqrt{\varepsilon}/\delta_0$, $t'=t\omega_0\sqrt{\varepsilon}$, $\bar{\theta}=\theta/\theta_2$, where $\delta_0=(\alpha M^2/2K_{\parallel})^{1/2}$, $\omega_0=(2\gamma/M)(K_{\parallel}K_{\perp})^{1/2}$. Then

$$I=\frac{8K_{\parallel}\delta_0^3}{\omega_0}\int dt'\int d\mathbf{x}'\left[\left[\frac{K_{\perp}}{K_{\parallel}\varepsilon}\right]^{1/2}\phi\bar{\theta}\frac{d\bar{\theta}}{dt'}-\tfrac{1}{2}\left[\frac{K_{\perp}}{K_{\parallel}\varepsilon}\right]\bar{\theta}^2\sin^2\phi-\tfrac{1}{2}(\nabla'\bar{\theta})^2-\tfrac{1}{2}\bar{\theta}^2(\nabla'\phi)^2-\tfrac{1}{2}(\bar{\theta}^2-\bar{\theta}^4)\right]\ . \tag{11}$$

It is clear now that for $K_{\parallel}\varepsilon/K_{\perp}\ll1$, only small values of ϕ contribute to the path integral (9), so that one can replace $\sin^2\phi$ in Eq. (11) by ϕ^2 and neglect the term $\frac{1}{2}\bar{\theta}^2(\nabla'\phi)^2$ which is of order ε. Then the Gaussian integration over ϕ reduces Eq. (9) to

$$\int D\{\bar{\theta}(\mathbf{x}',\tau')\}\exp\left[-\frac{S}{\hbar}\right], \tag{12}$$

where we have introduced the variable $\tau'\equiv x_4'=it'$ and the Euclidean action

$$S=\int d\tau\int d\mathbf{x}\left[\tfrac{1}{2}J\left[\frac{\partial\theta}{\partial\tau}\right]^2+\tfrac{1}{2}\alpha M^2(\nabla\theta)^2+K_{\parallel}(\varepsilon\theta^2-\tfrac{1}{4}\theta^4)\right]$$

$$=\frac{8K_{\parallel}\delta_0^3}{\omega_0}\int d^4x'[\tfrac{1}{2}(\bar{\nabla}\bar{\theta})^2+\tfrac{1}{2}(\bar{\theta}^2-\bar{\theta}^4)]\ . \tag{13}$$

Here $J\equiv M^2/2\gamma^2K_{\perp}$ can be interpreted as an effective moment of inertia associated with the subbarrier rotation of $\mathbf{M}$, the last term is E_a $(\theta,\phi=0)$ for small θ, and $\bar{\nabla}_i=\partial/\partial x_i'$ $(i=1,2,3,4)$. Trajectories which are in the vicinity of the extremal trajectory determined by the equation

$$\bar{\nabla}^2\bar{\theta}=\bar{\theta}-2\bar{\theta}^3 \tag{14}$$

give the main contribution to the integral (12).

For the subbarrier rotation of $\mathbf{M}$ in a small particle of volume $V\ll\delta_0^3$, $\mathbf{M}$ is uniform within the particle and Eqs. (13) and (14) reduce to

$$S=\frac{4M}{\gamma}\left[\frac{K_{\parallel}}{K_{\perp}}\right]^{1/2}\varepsilon^{3/2}\int d\tau'\left[\tfrac{1}{2}\left[\frac{d\bar{\theta}}{d\tau'}\right]^2+\tfrac{1}{2}(\bar{\theta}^2-\bar{\theta}^4)\right], \tag{15}$$

$$\frac{d^2\bar{\theta}}{d\tau'^2}=\bar{\theta}-2\bar{\theta}^3\ . \tag{16}$$

Equation (16) has the instanton solution $\bar{\theta}=1/\cosh\tau'$ 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 $\tau=\infty$ (see Fig. 1). Substituting this solution into Eq. (15) we immediately obtain Eq. (5).

Turning to the nonuniform problem, consider first the nucleation process in a thin film of thickness h (smaller than the size $\delta_0/\sqrt{\varepsilon}$ of the critical nucleus), whose plane is perpendicular to the easy z axis. If $K_{\parallel}\gg2\pi M^2$, then

we can still use $E=E_e+E_a$ for the energy density, neglecting the demagnetization energy. For $\theta(\mathbf{x},\tau)$ being independent of z, the O(3) symmetry of the problem in two spatial plus one imaginary time dimensions leads one to guess[12] that the solution minimizing the action is given by $\bar{\theta}=\bar{\theta}(u)$, where $u=(\rho'^2+\tau'^2)^{1/2}$, and ρ' is the normalized distance from the z axis. Then Eqs. (13) and (14) reduce to

$$S=16\pi\frac{M}{\gamma}\left[\frac{K_\parallel}{K_\perp}\right]^{1/2}h\delta_0\sqrt{\varepsilon}\int_0^\infty du\,u^2\left[\tfrac{1}{2}\left[\frac{d\bar{\theta}}{du}\right]^2+\tfrac{1}{2}(\bar{\theta}^2-\bar{\theta}^4)\right], \tag{17}$$

$$\frac{d^2\bar{\theta}}{du^2}+\frac{2}{u}\frac{d\bar{\theta}}{du}=\bar{\theta}-2\bar{\theta}^3\,. \tag{18}$$

The instanton solution of Eq. (18) was found numerically and is illustrated in Fig. 2. It corresponds to the inhomogeneous subbarrier rotation of the magnetization in the plane $\phi=0$ within a spatial region of size $\rho\sim\delta_0/\sqrt{\varepsilon}$. The maximal rotation of **M** from the easy axis direction, $\theta_{\max}\approx 3.07\theta_2$ at $\tau=0$, $\rho=0$, is small in the limit of small ε. Numerical integration in Eq. (17), using this solution, gives us the WKB exponent for the subbarrier bubble nucleation in a ferromagnetic film

$$P_2\propto\exp\left[\frac{-S_E}{\hbar}\right]=\exp\left[-37.8\frac{M}{\hbar\gamma}\left[\frac{K_\parallel}{K_\perp}\right]^{1/2}h\delta_0^2\sqrt{\varepsilon}\right]. \tag{19}$$

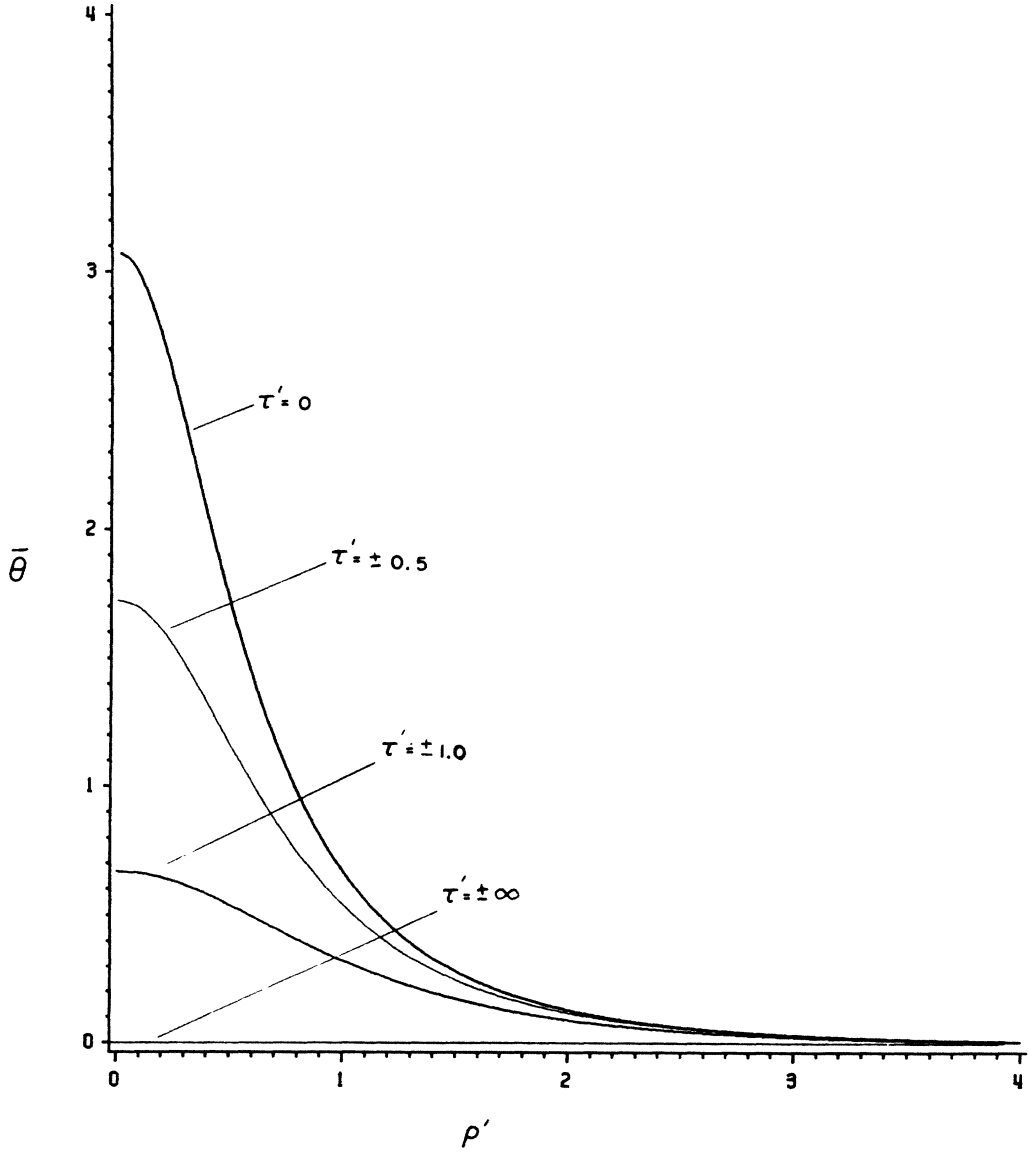

FIG. 2. The instanton, corresponding to subbarrier bubble formation in a film, for various imaginary times τ'. All variables are in reduced units: $\bar{\theta}=\theta/\theta_2$, $\rho'=\rho\sqrt{\varepsilon}/\delta_0$, $\tau'=\tau\omega_0\sqrt{\varepsilon}$.

In real time the nucleation process corresponds to the quantum jump of the classical field $\theta=0$, at $t=0$, to the inhomogeneous state $\theta(\mathbf{x},t=0)=\theta(\mathbf{x},\tau=0)$, $\dot{\theta}(\mathbf{x},t=0)=0$. Afterwards, it evolves according to the classical equation

$$-\frac{\partial^2\bar{\theta}}{\partial t'^2}+\nabla'^2\bar{\theta}=\bar{\theta}-2\bar{\theta}^3 , \tag{20}$$

which is the analytic continuation of the Euclidean equation (14).[1] Correspondingly, $\theta=\theta[v=(\rho'^2-t'^2)^{1/2}]$, determined by Eq. (20), is the analytic continuation of the instanton solution given by Eq. (18). The function $\theta(\rho,\tau=0)$ defines, therefore, the shape of the bubble at the moment of its materialization.

In a thermodynamic theory of nucleation $P\propto\exp(-W_{\min}/k_BT)$, where $W_{\min}$ is the minimal work necessary to produce a nucleus capable of growing. To obtain $W_{\min}$, consider the effective potential of the system

$$U_{\text{eff}}=\int d\mathbf{x}\,E=\int d\mathbf{x}[\tfrac{1}{2}\alpha M^2(\nabla\theta)^2+K_{\parallel}(\varepsilon\theta^2-\tfrac{1}{4}\theta^4)] . \tag{21}$$

For a cylindrical bubble it reduces to

$$U_{\text{eff}}=16\pi K_{\parallel}h\delta_0^2\varepsilon\int_0^{\infty}d\rho'\rho'\left[\frac{1}{2}\left[\frac{d\bar{\theta}}{d\rho'}\right]^2+\frac{1}{2}(\bar{\theta}^2-\bar{\theta}^4)\right] , \tag{22}$$

wherein the shape of the critical nucleus corresponds to a saddle point of this functional:[13]

$$\frac{d^2\bar{\theta}}{d\rho'^2}+\frac{1}{\rho'}\frac{d\bar{\theta}}{d\rho'}=\bar{\theta}-2\bar{\theta}^3 . \tag{23}$$

The solution of Eq. (23) obtained by numerical integration is shown in Fig. 3. Further integration of this solution in Eq. (22) gives $W_{\min}=23.3K_{\parallel}h\delta_0^2\varepsilon$. Representing

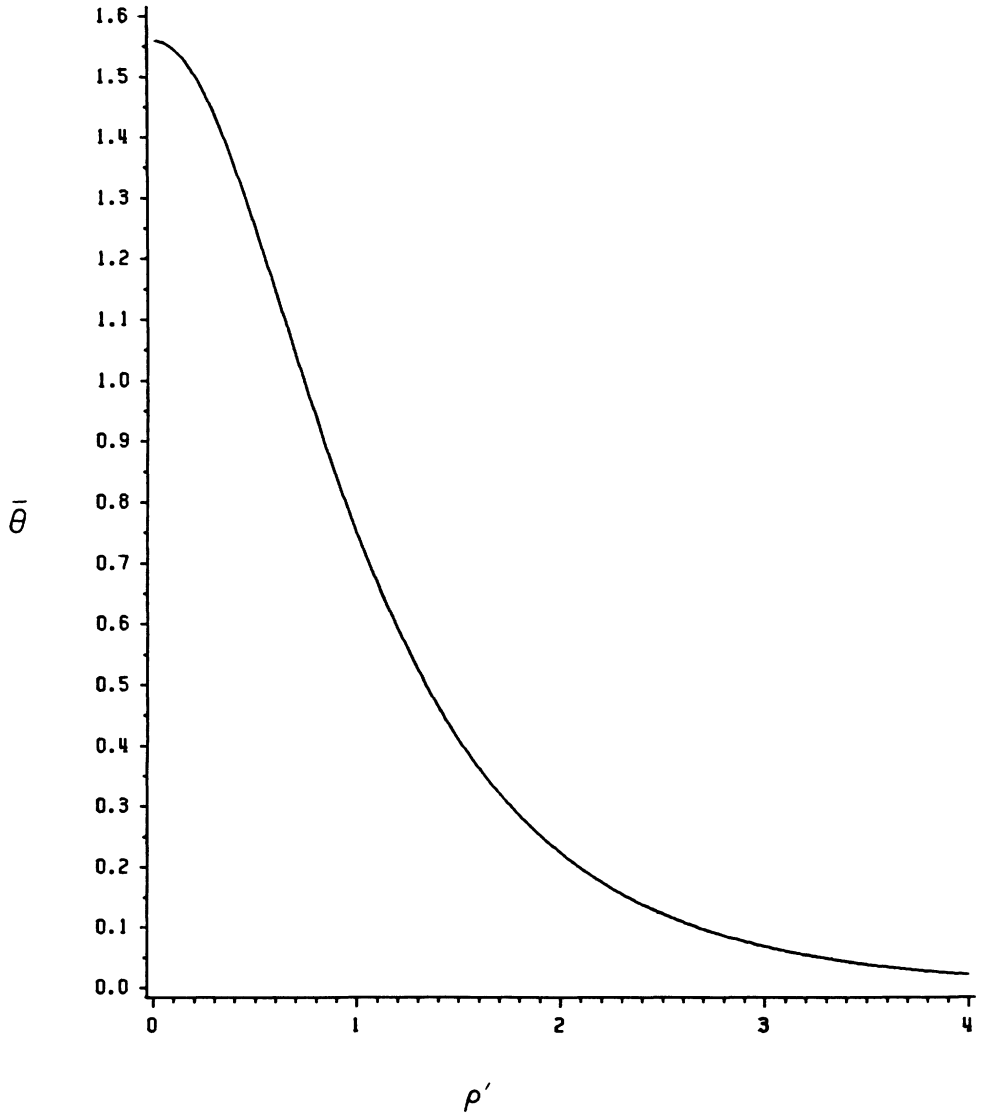

FIG. 3. The shape of the critical bubble in a thermodynamic theory of nucleation.

the exponent in Eq. (19) as $-W_{\min}/k_B T_c$, we obtain the crossover temperature [cf. Eq. (6)]

$$T_c = 0.62 \frac{\hbar\gamma\sqrt{K_\parallel K_\perp}}{k_B M}\sqrt{\varepsilon} \ . \tag{24}$$

It may seem that consideration of quantum nucleation in a bulk ferromagnet reduces to the above analysis in three spatial dimensions, i.e., one should consider Eqs. (15) and (16) with $\theta=\theta(u)$ and $u=(\tau'^2+r'^2)^{1/2}$, where r' is the normalized radius of a spherical nucleus. Unfortunately, numerical analysis of the instanton solution in three (spatial) plus one (imaginary time) dimensions indicates a breakdown of the small θ approximation. Based upon a qualitative analysis, we believe that the WKB exponent and T_c in the bulk, to within an order of magnitude, are given by Eqs. (5) and (6) with $V\sim\delta_0^3\varepsilon^{-3/2}$. Rigorous analysis of the problem involves consideration of the exact potential (1). This is much more laborious and will be done elsewhere.

To observe the effect one needs a large crossover temperature and not too small a nucleation rate. Our formulas show that materials with very large anisotropy, small ratio of $K_\parallel$ to $K_\perp$, and small saturated magnetization are preferable for such study. Recently, a quantum mechanism has been suggested for a staircase behavior in the magnetization reversal of the chemically disordered ferromagnet $SmCo_{3.5}Cu_{1.5}$.[14,15] This material is characterized by having an extremely large uniaxial anisotropy $K_\parallel\sim 1.8\times 10^8$ erg/cm^3 and $M\sim 300$ emu/cm^3. The effect appears in magnetic fields $H\gtrsim 20$ kOe below 2 K. If one assumes that this is due to quantum nucleation[16] within defective regions with $K_\parallel\sim K_\perp\sim 10^7$ erg/cm^3, which corresponds to $H\lesssim H_c$, $\varepsilon\sim 1$, then Eq. (6) gives a crossover temperature on the order of 1 K. The corresponding nucleation rate can be large enough with respect to the time of the experiment for $\delta_0<10$ Å, which is consistent with the experimental data. Although our continuous spin-field approximation cannot be applied to such a thin wall, we believe that it can still be valid as an order of magnitude.

In conclusion, notice that the clearcut observation of quantum nucleation in ferromagnets would be extremely interesting as the next example, after Josephson junctions, of macroscopic quantum tunneling.[17] Based upon our results, we consider thin ferromagnetic films with high anisotropy constants as the best candidates for that study. It should also be noted, that besides being of fundamental interest, processes of quantum nucleation and collapse of magnetic bubbles impose quantum limitations on the density and long-term reliability of the data storage in magnetic memory devices.

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