Macroscopic spin tunneling in weakly ferromagnetic particles of orthorhombic antiferromagnets

V. Yu. Galyshev

Moscow Physical–Technical Institute, 141700 Moscow, Russia

A. F. Popkov

Zelenograd Scientific Institute of Physical Problems, 103460 Moscow, Russia

(Submitted 17 March 1995)
Zh. Éksp. Teor. Fiz. **108**, 1755–1777 (November 1995)

We analyze possible manifestations of macroscopic quantum coherence of states of the antiferromagnetism vector in small particles of a weak ferromagnet with orthorhombic symmetry, and investigate the field aspects of macroscopic quantum tunneling of spins. The probabilities of macroscopic quantum tunneling in the angular antiferromagnetic phases are calculated in the quasiclassical approximation in the region of magnetic hysteresis, taking account of the fluctuation contribution to the path integral. We show that the field dependence of the spontaneous decay rate of the metastable state is nonmonotonic, up to its vanishing in the middle of the hysteresis loop for half-integer spin of the sublattice. In the angular antiferromagnetic phase, the effect of periodic freezing of macroscopic quantum tunneling of the antiferromagnetism vector, associated with interference of instantons, can arise in a magnetic field. Calculations of macroscopic quantum tunneling in $YFeO_3$ and an estimate of the influence on it of a dissipative environment predict the appearance of macroscopic quantum coherence in nanoparticles of this material, even in the absence of a magnetic field. The possibility of detecting macroscopic quantum coherence in an ensemble of small particles of $YFeO_3$ at the temperature peak of the specific heat at low temperatures is indicated.

1. INTRODUCTION

Coherent quantum behavior of spin ensembles has been observed in artificially fabricated ferromagnetic and antiferromagnetic small particles,[1] and also in finely dispersed media, thin films, and bulk crystals.[2,3] It is responsible for spin tunneling in single-domain samples,[4] quantum diffusion of domain boundaries,[5,6] spontaneous domain formation,[7] and other phenomena. Their study is vital to an understanding of the quantum dynamics of spins in mesoscopic magnetically-ordered structures and in connection with the emerging field of nanotechnology.[8] From this point of view, the field properties of spontaneous spin reorientation in small particles of weakly ferromagnetic particles is of unquestioned interest.

So far, the literature has only discussed the influence of a magnetic field on macroscopic quantum tunneling of spins in ferromagnetic[4,9–14] and antiferromagnetic[15–17] single-domain particles. Weakly ferromagnetic materials are of interest from some points of view. First, exchange amplification of the magnetic resonance frequency $\omega_0=\gamma\sqrt{H_AH_E}$ takes place in them, like in antiferromagnets. Here $\gamma=g\mu_B/\hbar$ is the gyromagnetic ratio (g is the Landé factor), H_A is the anisotropy field, and H_E is the exchange field. Thanks to this, the spin tunneling rate $\Gamma \propto \omega_0\exp(-U/k_BT)$, where U is the height of the potential barrier, and the characteristic crossover temperature $T^* \propto \hbar\omega_0$, at which the quantum spin fluctuations begin to predominate over the thermal activation fluctuations, is higher than in ferromagnetic particles of similar size. In addition, thanks to the presence of a weakly ferromagnetic moment in them, in contrast to antiferromagnets, the interaction with a variable external magnetic field is enhanced, which facilitates observations of macroscopic quantum tunneling by resonance methods.

On the other hand, the presence of antisymmetric exchange[18] in weak ferromagnets leads to a substantial difference between the field properties of macroscopic quantum tunneling in this case and the purely ferromagnetic,[4,9] antiferromagnetic,[15–17] and ferrimagnetic cases, in which the sublattices of the antiferromagnet are uncompensated.[19] This has to do, in particular, with the difference in the nature of the magnetic field-induced orientational phase transitions and the properties of the energy degeneracy of the phase states of a weakly ferromagnetic particle. Depending on the orientation of the magnetic field in the crystal, the mechanism of field interference of the instantons, for example, will change.

Interference of instantons suppresses microscopic quantum tunneling of spins in ferromagnetic particles with half-integer spin in the absence of an external magnetic field.[13,14] A magnetic field, which lifts the energy degeneracy by virtue of the Zeeman interaction, can lead to freezing of spin tunneling in such particles.[12] In addition, it causes quantum oscillations of the energy splitting of the degenerate phase states of the ferromagnetic particle, which in turn can lead to field oscillations of its magnetization at low temperatures.[17]

Similarly, field interference of instantons in an antiferro-

 1063-7761/95/110962-12$10.00

magnet causes oscillations of the tunneling rate in the angular phase.[20,21] In this case, the period of the oscillations is determined by the intersublattice exchange field acting on an elementary spin pair, in contrast to a ferromagnetic particle, where it is determined by the anisotropy field.[10]

In a weak ferromagnet the mechanisms of instanton interference will vary, depending on the orientation of the magnetic field in the crystal and the nature of the magnetic anisotropy. Obviously, in connection with this, the field scale of the quantum oscillations of the macroscopic quantum tunneling rate can vary. In addition, we will show that the presence of a spontaneous magnetic moment in a weak ferromagnet does not lead to macroscopic Kramers degeneracy of the levels, in contrast with uncompensated antiferromagnets, in which this latter effect depends on the parity of the uncompensated spin.[19] We analyze these peculiarities for the case of an orthorhombic antiferromagnet with antisymmetric exchange. Such weakly ferromagnetic compounds include rare-earth orthoferrites and orthochromates RMO_3, where M is Fe or Cr. In these compounds, orientational phase transitions, induced by a magnetic field, differ depending on the type of rare-earth ion R^{3+} (Ref. 22). We will direct our attention principally to an analysis of the peculiarities of macroscopic quantum tunneling in compounds of the type $YFeO_3$, $LaFeO_3$, and $LuFeO_3$, which are characterized by a record small line width of the magnetic resonance, which is very important for observing macroscopic quantum tunneling. Aspects of this tunneling in other antiferromagnetic compounds will be discussed briefly.

2. LAGRANGIAN OF A WEAK FERROMAGNET AND THE SPIN TUNNELING RATE

Let us consider an orthorhombic weak antiferromagnet of the type $YFeO_3$ in an external magnetic field. Its thermodynamic potential per unit volume, expanded into components of the ferro- and antiferromagnetism vectors **F** and **G** out to terms of second order, can be represented in the form (see Refs. 22 and 23)

$$\langle \hat{H} \rangle = \langle \hat{H}_0 \rangle + \frac{A}{2}\mathbf{F}^2 - M_0 \mathbf{F}\cdot\mathbf{H} + d_1 F_a G_c + d_3 F_c G_a + b_1 G_a^2 + b_3 G_c^2 + a_1 F_a^2 + a_2 F_c^2, \quad (1)$$

where A and d_j are the isotropic and antisymmetric[18] (Dzyaloshinskĭ) exchange constants, a_j and b_j are the relativistic interaction constants ($j=1,2$), and M_0 is the saturating magnetic moment of the antiferromagnet.

Since usually

$$\frac{F_c}{G_a}, \quad \frac{F_a}{G_c} \propto \frac{d}{A} \propto 10^{-2},$$

the latter terms in Eq. (1) can be neglected. In addition, we can with good accuracy take $d_3 \approx -d_1 \equiv d$. Here, for the anisotropy energy parameters we will assume the following relations to be fulfilled:

$$b_1 < b_3 < \frac{d^2}{A},$$

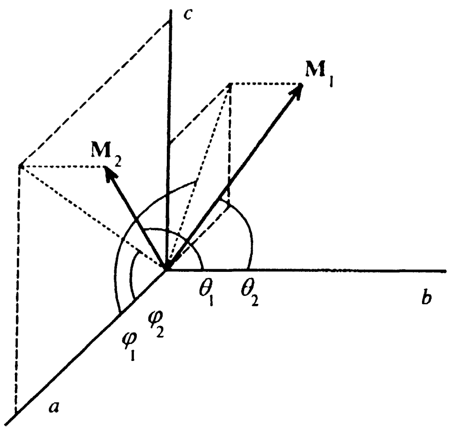

FIG. 1. Angular coordinates of the sublattice magnetizations in the crystallographic coordinate system.

for which the main state of the weak ferromagnet in the absence of a magnetic field is the state $\mathbf{F}=(0,0,F_c)$, $\mathbf{G}=(G_a,0,0)$—the so-called Γ_4 phase.[1)]

In the chosen hierarchy of magnetic parameters, the preferred plane of rotation of the antiferromagnetism vector **G** is the ac plane of the crystal. We will make use of this in our subsequent analysis of the dynamics of spin tunneling in the materials considered.

In the case in which the relativistic interactions and antiferromagnetic exchange are small in comparison with the isotropic exchange interaction (this latter interaction being the main interaction), their influence can be treated as a small perturbation. In this approximation the dynamics of spin tunneling in the adiabatic limit is described according to Ref. 24 by the Lagrangian

$$L = -i\hbar s \sum_j \dot{\varphi}_j (1-\cos\theta_j) + \langle \hat{H} \rangle, \quad (2)$$

where φ_j and θ_j are the polar coordinates of the spins, s is the spin of the magnetic ion, and $\hbar$ is the quantum of action, and the summation is carried out over all magnetic ions.

Inside the magnetic sublattices the spin is bound by the strong exchange interaction. In this context, it is natural to break down the sum in Eq. (2) into a sum over sublattices, assuming the rotation of the spins inside each of them to be coherent. Combining the four sublattices of the weak ferromagnet in two pairs, we arrive at a two-sublattice model which recommends itself for the study of the magnetodynamics of the domain boundaries in orthoferrites.[25,26] In this case $\mathbf{F}=(\mathbf{M}_1+\mathbf{M}_2)/M_0$, $\mathbf{G}=(\mathbf{M}_1-\mathbf{M}_2)/M_0$, where $\mathbf{M}_1$ and $\mathbf{M}_2$ are the magnetizations of the combined sublattices, and $M_0=|\mathbf{M}_1|+|\mathbf{M}_2|$.

We choose a coordinate system with polar axis along the **b** axis of the crystal, and reckon the azimuthal angle from the **a** axis (see Fig. 1). For convenience, we introduce, in analogy with Ref. 25, new dynamic variables for the direction angles of the magnetization of the sublattices:

$$\theta_1 = \theta + \varepsilon, \quad \theta_2 = \pi - \theta + \varepsilon, \quad \varphi_1 = \varphi + \beta,$$

$$\varphi_2 = \pi + \varphi - \beta. \quad (3)$$

In the new variables $\mathbf{\Omega}=(\theta,\varepsilon,\beta,\varphi)$ the components of the vectors **F** and **G** take the form

$$
\begin{aligned}
F_x&=-\sin\theta\sin\varphi\cos\varepsilon\sin\beta\\
&\quad+\cos\theta\cos\varphi\sin\varepsilon\cos\beta,\\
-F_y&=\sin\theta\cos\varphi\cos\varepsilon\sin\beta\\
&\quad+\cos\theta\sin\varphi\sin\varepsilon\cos\beta,\\
F_z&=-\sin\theta\sin\varepsilon,\\
G_x&=\sin\theta\cos\varepsilon\cos\varphi\cos\beta\\
&\quad-\cos\theta\sin\varepsilon\sin\varphi\sin\beta,\\
-G_y&=\sin\theta\sin\varphi\cos\varepsilon\cos\beta\\
&\quad+\cos\theta\cos\varphi\sin\varepsilon\sin\beta,\\
G_z&=\sin\theta\cos\varepsilon,
\end{aligned}
\tag{4}
$$

Consequently, the Euclidean action $S_E=\int_{-\infty}^{\infty} L d\tau$, taking Eqs. (1)–(4) into account, is equal to

$$
\begin{aligned}
S_E&=v_0\int_{-\infty}^{+\infty}\left[-\frac{iM_0}{2\gamma}\sum_{j=1,2}\varphi_\tau^{(j)}(1-\cos\theta^{(j)})+\langle\hat{H}(\mathbf{F},\mathbf{G})\rangle\right]d\tau\\
&=v_0\int_{-\infty}^{+\infty}\left[\frac{iM_0}{\gamma}(\varphi,\sin\varepsilon\sin\theta-\beta_\tau\cos\varepsilon\cos\theta+\varphi_\tau)\right.\\
&\quad+\frac{A}{2}(\sin^2\varepsilon\cos^2\beta+\sin^2\beta\sin^2\theta)+d\sin\beta\cos\beta\\
&\quad\times(\sin^2\varepsilon-\sin^2\theta)+\frac{b_1}{2}(\sin\theta\cos\varepsilon\cos\beta\cos\varphi\\
&\quad-\cos\theta\sin\varepsilon\sin\beta\sin\varphi)^2\\
&\quad+\frac{b_3}{2}(\sin\theta\cos\varepsilon\cos\beta\sin\\
&\quad\left.\times\varphi+\cos\theta\sin\varepsilon\sin\beta\cos\varphi)^2-M_0\mathbf{F}\cdot\mathbf{H}\right]d\tau,
\end{aligned}
\tag{5}
$$

where v_0 is the volume of the particle and $\tau=it$ is imaginary time. The first term in the integrand (5) describes the so-called topological Berry phase[27] responsible for the effects of instanton interference.[13,14] The remaining terms are due to the exchange, relativistic, and Zeeman interactions in the system under consideration.

The amplitude of the conditional probability of a transition of the spins from the state $|b\rangle$ to the state $|e\rangle$ (the propagator) is given, according to theory,[28] by the path integral

$$
K_{bc}=\langle e|\exp[-\hat{H}(\tau-\tau')/\hbar]|b\rangle=N\int_b^e[d\mathbf{\Omega}]\times\exp[-S_E(\mathbf{\Omega})/\hbar],
\tag{6}
$$

where $S_E(\mathbf{\Omega})=\int_0^T L(\mathbf{\Omega})d\tau$ is the action along the path $\mathbf{\Omega}(\tau)$ in imaginary time $\tau=it$, T is the total transition time, and N is a normalization factor. In the quasiclassical limit $|S_E|\gg\hbar$ the main contribution to the integral (6) comes from the instanton trajectories $\mathbf{\Omega_m}$ that minimize the action $S_E(\mathbf{\Omega})$ and satisfy the variational equations $\delta S_E/\delta\mathbf{\Omega}|_{\mathbf{\Omega}=\mathbf{\Omega_m}}=0$. The one-instanton contribution to the probability amplitude in this case is equal to

$$
\tilde{K}_{be}\simeq N\exp[-S_E^{cl}/\hbar]\int[d(\mathbf{\Omega}-\mathbf{\Omega}_m)]\exp[-\delta^2S_E/\hbar],
\tag{7}
$$

where $S_E^{\rm cl}=S_E(\mathbf{\Omega}_m)$ is the classical action on the instanton path and δ^2S_E is the second variation of the action.

According to Ref. 40, it is easy to obtain the quasiclassical expression for the multi-instanton probability amplitude in a system of two symmetric potential wells:

$$
\begin{aligned}
K_{be}&\simeq\exp\left[i\alpha-\frac{E_0T}{\hbar}\right]\sinh\left[\frac{\Delta T}{2\hbar}\right]\\
&=\exp\left[i\alpha-\frac{E_0T}{\hbar}\right]\left\{\frac{\Delta T}{2\hbar}+\frac{1}{3!}\left(\frac{\Delta T}{2\hbar}\right)^3+\cdots-\right\},
\end{aligned}
\tag{8}
$$

where E_0 is the energy of the ground state, Δ is the energy splitting, and α is an arbitrary phase. Then the tunneling rate, expressed in terms of the probability amplitude of a one-instanton transition, is

$$
\Gamma=\frac{\Delta}{2\hbar}=\frac{1}{T}\exp\left[\frac{E_0T}{\hbar}\right]|\tilde{K}_{be}(T)|\simeq A_m\exp\left[-\frac{S_E^{\rm cl}}{\hbar}\right].
\tag{9}
$$

The value of the pre-exponential factor A_m is determined by the fluctuation contribution to the integral (6) near the corresponding tunneling path and, according to theory,[29,30] is

$$
A_m\propto\left(\frac{S_E^{\rm cl}}{2\pi\hbar}\right)^{1/2}\left\{\frac{\det'[\hat{L}_{fl}(\mathbf{\Omega}_m(\tau))]}{\det[\hat{L}_{fl}(\mathbf{\Omega}_m(\pm\infty))]}\right\}^{-1/2},
\tag{10}
$$

where $\hat{L}_{fl}$ is a linear operator associated with the second variation δ^2S_E, and describes the fluctuation spectrum near the corresponding path; $\det'[\hat{L}_{fl}(\mathbf{\Omega}_m(\tau))]$ is the product of the eigenvalues of this operator on the path $\mathbf{\Omega}_m$ (the prime indicates that the zero eigenvalue is to be omitted when calculating the determinant); $\det'[\hat{L}_{fl}(\mathbf{\Omega}_m(\pm\infty))]$ is the product of the eigenvalues of the fluctuation operator near equilibrium states of the system $\mathbf{\Omega}_m(\pm\infty)$.

In the presence of multiple independent instanton trajectories, Eq. (9) generalizes trivially to

$$
\Gamma\simeq\sum_m A_m\exp\left[-\frac{S_E^{cl}(\mathbf{\Omega}_m)}{\hbar}\right].
\tag{11}
$$

We consider three different cases in which the magnetic field acts along one of the crystallographic axes of an orthorhombic crystal.

3. H‖a. MACROSCOPIC QUANTUM TUNNELING IN THE ANGULAR PHASE Γ_{24}

Analysis of the energy (1) shows that (see also Ref. 22) for **H**‖**a** and fields $H<H_c=-H_d/2+\sqrt{(H_d/2)^2+H_AH_E}$ that

are not too strong, where $H_d=d/M_0$, $H_e=A/M_0$, and $H_A=(b_3-b_1)/M_0$, there are two energetically degenerate equilibrium positions $\mathbf{\Omega}_\pm=(\theta=\pi/2,\varepsilon=0,\beta=\beta^0,\varphi=\varphi_\pm^0)$, where $\beta^0=H_d/H_E(1-H^2/H_AH_E)$, and $\sin\varphi_\pm^0=HH_d/H_AH_E(1-H^2/H_AH_E)$. Between these states macroscopic quantum tunneling of spins is possible. For fields $H\geqslant H_c$ a stable phase is the nondegenerate phase Γ_2: $\mathbf{\Omega}_{\Gamma_2}=(\pi/2,0,\beta(H),\pi/2)$, in which macroscopic quantum tunneling of spins is impossible.

Let us calculate the spin tunneling rate for $\mathbf{H}\|\mathbf{a}$ as a function of the magnetic field at zero temperature. The Euler–Lagrange equations in this case have the form

$$\frac{iM_0}{\gamma}(\varphi_\tau\sin\varepsilon\cos\theta+\beta_\tau\cos\varepsilon\sin\theta)+A\sin^2\beta\sin\theta$$
$$\times\cos\theta-2d\sin\beta\cos\beta\sin\theta\cos\theta$$
$$+b_3(\cos\varepsilon\sin\beta\sin\theta$$
$$\times\cos\varphi+\sin\varepsilon\cos\beta\cos\theta\sin\varphi)$$
$$\times(\cos\varepsilon\sin\beta\cos\theta\cos\varphi$$
$$-\sin\varepsilon\cos\beta\sin\theta\sin\varphi)$$
$$+b_1(\cos\varepsilon\sin\beta\sin\theta\sin\varphi$$
$$-\sin\varepsilon\cos\beta\cos\theta\cos\varphi)(\cos\varepsilon\sin\beta\cos\theta\sin\varphi$$
$$+\sin\varepsilon\cos\beta\sin\theta\cos\varphi)$$

$$\frac{iM_0}{\gamma}(\varphi_\tau\cos\varepsilon\sin\theta+\beta_\tau\sin\varepsilon\cos\theta)+A\sin\varepsilon\cos\varepsilon$$
$$\times\cos^2\beta+\sin\varepsilon\cos\varepsilon\sin\beta\cos\beta+b_3(\cos\varepsilon\sin\beta$$
$$\times\sin\theta\cos\varphi+\sin\varepsilon\cos\beta\cos\theta\sin\varphi)$$
$$\times(-\sin\varepsilon\sin\beta\sin\theta\cos\varphi$$
$$+\cos\varepsilon\cos\beta\cos\theta\sin\varphi)$$
$$+b_1(\cos\varepsilon\sin\beta\sin\theta\sin\varphi-\sin\varepsilon\cos\beta\cos\theta$$
$$\times\cos\varphi)(-\sin\varepsilon\sin\beta\sin\theta\sin\varphi-\cos\varepsilon\cos\beta$$
$$\times\cos\theta\cos\varphi)$$
$$+M_0H(-\sin\varepsilon\sin\beta\sin\theta$$
$$\times\sin\varphi-\cos\varepsilon\cos\beta\cos\theta\cos\varphi)=0 \tag{12b}$$

$$\frac{iM_0}{\gamma}(\cos\varepsilon\cos\theta)_\tau+A\sin\beta\cos\beta\,(\sin^2\theta-\sin^2\varepsilon)+d$$
$$\times\cos2\beta(\sin^2\varepsilon-\sin^2\theta)+b_3(\cos\varepsilon\sin\beta\sin\theta\cos\varphi$$
$$+\sin\varepsilon\cos\beta\cos\theta\sin\varphi)(\cos\varepsilon\cos\beta\sin\theta\cos\varphi$$
$$-\sin\varepsilon\sin\beta\cos\theta\sin\varphi)+b_1(\cos\varepsilon\sin\beta\sin\theta\sin\varphi$$
$$-\sin\varepsilon\cos\beta\cos\theta\cos\varphi)(\cos\varepsilon\cos\beta\sin\theta\sin\varphi$$
$$+\sin\varepsilon\sin\beta\cos\theta\cos\varphi)+M_0H(\cos\varepsilon\cos\beta\sin\theta$$
$$\times\sin\varphi+\sin\varepsilon\sin\beta\cos\theta\cos\varphi)=0, \tag{12c}$$

$$\frac{iM_0}{\gamma}(-\sin\varepsilon\sin\theta)_\tau+b_3(\cos\varepsilon\sin\beta\sin\theta\cos\varphi$$
$$+\sin\varepsilon\cos\beta\cos\theta\sin\varphi)(-\cos\varepsilon\sin\beta\sin\theta\sin\varphi$$
$$+\sin\varepsilon\cos\beta\cos\theta\cos\varphi)+b_1(\cos\varepsilon\sin\beta$$
$$\times\sin\theta\sin\varphi-\sin\varepsilon\cos\beta\cos\theta\cos\varphi)(\cos\varepsilon\sin\beta$$
$$\times\sin\theta\cos\varphi+\sin\varepsilon\cos\beta\cos\theta\sin\varphi)$$
$$+M_0H(\cos\varepsilon\sin\beta\sin\theta\cos\varphi$$
$$+\sin\varepsilon\cos\beta\cos\theta\sin\varphi)=0. \tag{12d}$$

Taking into account that the preferred plane of rotation of the antiferromagnetism vector is the ac plane of the crystal, we search for instanton solutions near $\mathbf{\Omega}=(\pi/2,\beta,\varepsilon,\varphi)$. At $\theta=\pi/2$ the first equation (12a) is satisfied out to terms of order $(d/A)^2$. In this same approximation it follows from the second and fourth equations that

$$\varepsilon\cong-i\frac{M_0}{\gamma A}\varphi_\tau+O(d/A)^2, \tag{13}$$

$$\beta\cong\frac{d}{A}-\frac{H}{H_E}\sin\varphi+O(d/A)^2, \tag{14}$$

Substituting Eqs. (13) and (14) into (12c), and noting that $\theta\cong\pi/2+\mathcal{O}(d/A)^2$, we obtain

$$-\frac{\chi_\perp}{\gamma^2}\varphi_{\tau\tau}+\tilde{b}_3(\sin\varphi-\sin\varphi_0)\cos\varphi=0, \tag{15}$$

where $\sin\varphi_0=-H_d/H_E\tilde{b}_3$, $\tilde{b}_3=(b_3-b_1)(1-H^2/H_AH_E)$, and $\chi_\perp=M_0^2/A$.

This equation has the first integral

$$\frac{\chi_\perp}{2\gamma^2}\varphi_\tau^2-\frac{\tilde{b}_3}{2}(\sin\varphi-\sin\varphi_0)^2=E, \tag{16}$$

where E is the constant of integration.

Integral (16) gives the phase portrait of Eq. (15) shown in Fig. 2. The separatrix of the solution, which joins the equilibrium points $\varphi_0=-\pi/2\pm\delta+2\pi n$ $(n=0,\pm1,\pm2,\dots)$, where $\delta=\cos^{-1}(dH/\tilde{b}_3H_E)$, describes the instanton trajectories. There are two types of instanton solutions:

$$\varphi_\pm^{(1)}=-\frac{\tau}{2}\pm2\arctan\left\{\tan\frac{\delta}{2}\tanh\frac{\omega(\tau-\tau_0)}{2}\right\}+2\pi n \tag{17a}$$

and

$$\varphi_\pm^{(2)}=-\frac{\pi}{2}\pm2\arctan\left\{\cot\frac{\delta}{2}\tanh\frac{\omega(\tau-\tau_0)}{2}\right\}+2\pi n, \tag{17b}$$

where $\omega=\gamma\sqrt{\tilde{b}_3/\chi_\perp}\sin\delta$, $n=0,\pm1,\pm2,\dots$

The contributions to the action corresponding to these trajectories are given by

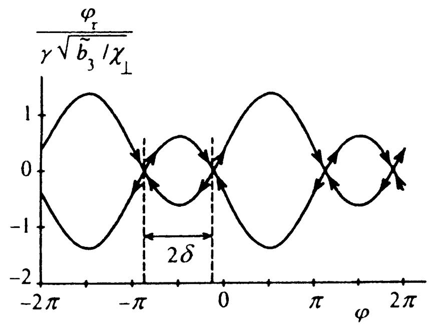

FIG. 2. Phase portrait of Eq. (15).

$$S_E^{(1)}=\frac{2v_0}{\gamma}\sqrt{\tilde{b}_3\chi_\perp}(\sin\delta-\delta\cos\delta)+i\frac{2M_0v_0}{\gamma}\delta \quad \text{(18a)}$$

and

$$S_E^{(2)}=S_E^{(1)}+\frac{2\pi v_0}{\gamma}\sqrt{\tilde{b}_3\chi_\perp}\cos\delta-i\frac{2M_0v_0}{\gamma}\pi. \quad \text{(18b)}$$

The imaginary terms in Eqs. (18a) and (18b) come from the topological Berry phase in the action (5). They define the phase shift in the tunneling probability amplitudes over the corresponding instanton trajectories. But the difference of these two phases is equal to

$$\frac{\mathrm{Im}(S_E^{(1)}-S_E^{(2)})}{\hbar}=\frac{2M_0v_0}{\gamma\hbar}\pi=S_0 4\pi,$$

where S_0, the total spin of the sublattice, is a multiple of 2π. Therefore, the imaginary part of the action has no effect on the total tunneling probability and instanton interference does not arise in this case. By summing over all the instanton and anti-instanton tunneling trajectories (as was done in Ref. 13), it can be shown that the tunneling rate is given by the sum of the one-instanton contributions (9) over the types of trajectories found:

$$\Gamma\cong A^{(1)}\exp\left\{-\frac{S_E^{(1)}}{\hbar}\right\}+A^{(2)}\exp\left\{-\frac{S_E^{(2)}}{\hbar}\right\}. \quad (19)$$

To calculate the prefactors $A^{(1)}$ and $A^{(2)}$, we consider the fluctuation part of the action

$$\delta^2S_E=\frac{v_0}{2}\int d\tau\left\{i\frac{2M_0}{\gamma}\cos\varepsilon(\delta\varphi_\tau\delta_\varepsilon-\delta\theta_\tau\delta\beta)+A\right.$$
$$\times\cos^2\varepsilon(\delta\varepsilon^2+\delta\beta^2)+((b_3-b_1)\cos 2\varphi-M_0H\beta$$
$$\times\sin\varphi)\delta\varphi^2+2(-M_0H\sin\varepsilon\sin\varphi)\delta\varphi\delta\theta$$
$$+\Bigg((2d-M_0H\sin\varphi)\beta-b_1$$
$$\left.\times\cos^2\varphi-b_3\sin^2\varphi-i\frac{M_0}{\gamma}\varphi_\tau\sin\varepsilon\Bigg)\delta\theta^2\right\}. \quad (20)$$

Here, we can complete the square in the angles $\delta\varepsilon$ and $\delta\beta$ and integrate the Gaussian integral in the formula for the transition amplitude (7) over these variables. As a result, we obtain the effective action describing the tunneling dynamics near the instanton path:

$$\delta^2S_{\mathrm{eff}}=\frac{\hbar}{2}\int ds\left[\delta\varphi_s^2+\frac{\chi_\perp v_0^2}{\hbar^2\gamma^2}((b_3-b_1)\cos 2\varphi\right.$$
$$-M_0H\beta\sin\varphi)\delta\varphi^2+2\frac{\chi_\perp v_0^2}{\hbar^2\gamma^2}(-M_0H\sin\varepsilon\sin\varphi)$$
$$\times\delta\varphi\delta\theta+\delta\theta_s^2+\frac{\chi_\perp v_0^2}{\hbar^2\gamma^2}\Bigg((2d-A\beta-M_0H\sin\varphi)\beta$$
$$\left.-b_1\cos^2\varphi-b_3\sin^2\varphi-i\frac{M_0}{\gamma}\varphi_\tau\sin\varepsilon\Bigg)\delta\theta^2\right], \quad (21)$$

where we have replaced the time variable τ by the new variable $s=\hbar\gamma^2\tau/\chi_\perp v_0$.

Thus, in contrast to a ferromagnet,[11] the fluctuation dynamics turns out to be equivalent to two coupled oscillators with potential energies which depend, generally speaking, on the instanton trajectories (17). In the case $H=0$ the oscillators are independent. Let us consider the fluctuation contribution for this case to the path integral. Making the transition to integration over the amplitudes of the eigenmodes of the aforementioned trajectories, as was done, for example, in Ref. 29, we obtain the following expression for the prefactors:

$$A^{(j)}=\frac{\hbar\gamma^2}{\chi_\perp v_0}\left(\frac{\mathrm{Re}\,S_E^{(j)}}{2\pi\hbar}\right)^{1/2}D_\varphi^{(j)}D_\theta^{(j)}, \quad (22)$$

where we have introduced the notation

$$D_\varphi^{(j)}=\left\{\frac{\det'\left[-\partial_s^2+\frac{\chi_\perp v_0^2}{\hbar^2\gamma^2}(b_3-b_1)\cos 2\varphi^{(j)}\right]}{\det\left[-\partial_s^2+\frac{\chi_\perp v_0^2}{\hbar^2\gamma^2}(b_3-b_1)\right]}\right\}^{-1/2}$$

and

$$D_\theta^{(j)}=\left\{\frac{\det\left[-\partial_s^2+\frac{\chi_\perp v_0^2}{\hbar^2\gamma^2}\left(\frac{d^2}{A}-b_1-2(b_3-b_1)\sin^2\varphi^{(j)}\right)\right]}{\det\left[-\partial_s^2+\frac{\chi_\perp v_0^2}{\hbar^2\gamma^2}\left(\frac{d^2}{A}-b_1\right)\right]}\right\}^{-1/2}.$$

Calculation of the first ratio of determinants in Eq. (22) reduces, according to Ref. 30, to finding the asymptotic limit of the corresponding instanton solution $\varphi^{(j)}(s)$ as $s\to\pm\infty$. Specifically, if

$$\lim_{s\to\pm\infty}\frac{d\varphi^{(j)}}{d\tau}=Q^{(j)}e^{-\mu|s|},$$

then the desired ratio is

$$D_\varphi^{(j)}=\left\{\frac{\int_{-\infty}^{+\infty}(\varphi_\tau^{(j)})^2ds}{2(Q^{(j)})^2\mu}\right\}^{-1/2}=\frac{2\chi_\perp v_0}{\hbar\gamma^2}\omega_0, \tag{23}$$

where we take

$$Q^{(j)}=2\omega_0,\quad \mu=\frac{\chi_\perp v_0}{\hbar\gamma^2}\omega_0,\quad \omega_0=\omega\Big|_{H=0}$$
$$=\gamma\sqrt{(b_3-b_1)/\chi_\perp}.$$

Here it should also be noted that the zero mode of the operator associated with the fluctuations in φ arises by virtue of temporal translational invariance relative to the center of the instanton when calculating the transition amplitude.

To calculate the second ratio of determinants, we need to know the asymptotic behavior of the solution of the boundary value problem for the second fluctuational operator, in contrast to the previous case, where it was related to the asymptotic limit of the instanton solution. The corresponding calculations for the case $H=0$ lead to the result (see Appendix A)

$$D_\theta^{(j)}=\left\{\frac{\det\left[-\partial_s^2+\frac{\chi_\perp v_0^2}{\hbar^2\gamma^2}\left(\frac{d^2}{A}-b_1-\frac{2(b_3-b_1)}{\cosh^2(\omega_0\tau)}\right)\right]}{\det\left[-\partial_s^2+\frac{\chi_\perp v_0^2}{\hbar^2\gamma^2}\left(\frac{d^2}{A}-b_1\right)\right]}\right\}^{-1/2}$$

$$=\left\{\frac{\sqrt{\frac{d^2}{A}-b_1}-\sqrt{b_3-b_1}}{\sqrt{\frac{d^2}{A}-b_1}+\sqrt{b_3-b_1}}\right\}^{-1/2}. \tag{24}$$

Taking Eqs. (23) and (24) into account, we obtain the final expression for the tunneling rate in zero field

$$\Gamma_0\cong 4D_\theta\omega_0\sqrt{\frac{S_0^{\rm cl}}{2\pi\hbar}}\exp\left[-\frac{S_0^{\rm cl}}{\hbar}\right], \tag{25}$$

where we have set $S_0^{\rm cl}=2\chi_\perp v_0\omega_0/\gamma^2$. The dependence of the tunneling rate (25) on the volume of the particle for $YFeO_3$ is plotted in Fig. 3. In the calculations we assumed the following values of the material parameters: $H_d=1.4\times10^5$Oe, $H_E=6.4\times10^6$Oe, $H_A=5.3\times10^2$Oe, and $M_0=1800$ G (see Ref. 22).

Note that for $d^2/A\gg b_3$, the influence of the fluctuations of the angle θ on the pre-exponential factor becomes negligible, and calculation of the tunneling rate, as was evident in Eq. (21), reduces to a one-dimensional problem. For simplicity, we assume in what follows that $d^2/A\gg b_3$. In this case $D_\varphi^{(j)}=2\sin\delta(\chi_\perp v_0\omega/\hbar\gamma^2)^{3/2}(\mathrm{Re}S_E^{(j)}/2\hbar)^{-1/2}$, and for an arbitrary magnetic field $H<H_c$ the tunneling rate has the form

$$\Gamma(H)\cong 4\sqrt{\frac{\chi_\perp v_0}{\pi\hbar\gamma^2}}\sin\delta\cdot\omega^{3/2}\sum_{j=1,2}\exp\left[-\frac{S_E^{(j)}}{\hbar}\right]. \tag{26}$$

Figure 4 shows the dependence of the tunneling rate on the magnetic field, calculated for $YFeO_3$ at three values of the particle volume: $v_0=5$, 10, 20 nm^3. The magnitude of the energy barrier separating the equilibrium states decreases with increasing field strength, while the tunneling rate grows exponentially. However, in the immediate vicinity of the critical field of the phase transition from the angular phase Γ_{24} to the phase Γ_2, in conjunction with the vanishing of the

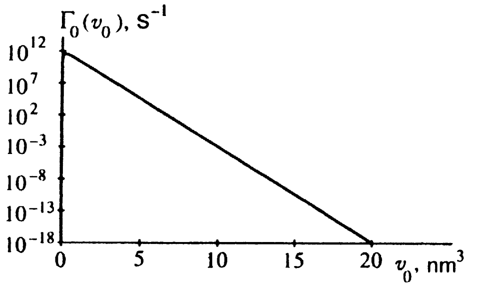

FIG. 3. Dependence of the tunneling rate in the absence of a magnetic field on the volume of the particle of $YFeO_3$.

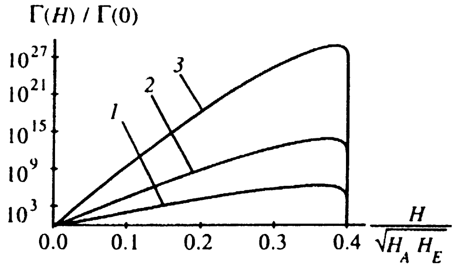

FIG. 4. Field dependence of the tunneling rate of $YFeO_3$ nanoparticles for **H**||**a**: *1*) $v_0=5$ nm^3, *2*) $v_0=10$ nm^3, *3*) $v_0=20$ nm^3.

resonance frequency (the soft mode of the transition $\omega|_{H\to H_c}=0$), the tunneling rate decreases abruptly.

4. H||b. MACROSCOPIC QUANTUM TUNNELING IN THE ANGULAR ANTIFERROMAGNETIC PHASE

In the case **H**||**b**, analysis of the energy (1) shows that the magnetic field fails to lift the energy degeneracy of the equilibrium states with oppositely directed antiferromagnetism vector **G**||±**a** all the way to the field of sublattice collapse $H_{sflip}=H_A+H_E$ (the spin-flip transition field). Therefore, over the entire field range $H<H_{sflip}$, quantum mixing of coherent spin states with **G**||±**a** is possible. The transition matrix element between these states can be calculated in analogy with the previous case.

The variational equations $\delta S_E/\delta\mathbf{\Omega}=0$, as our analysis has shown, have approximate solutions of the following type:

$$\theta\simeq\frac{\pi}{2}+\mathcal{O}\left(\frac{d}{A}\right)^2,$$
$$\beta\simeq\frac{d}{A}+\mathcal{O}\left(\frac{d}{A}\right)^2, \tag{27}$$
$$\sin\varepsilon\simeq -i\frac{M_0}{\gamma A}\varphi_\tau-\frac{H}{H_E}+O\left(\frac{d}{A}\right)^2.$$

Here $\varphi(\tau)$ satisfies the equation

$$-\frac{\chi_\perp}{\gamma^2}\varphi_{\tau\tau}+\tilde{b}_3\sin\varphi\cos\varphi=0, \tag{28}$$

where $\tilde{b}_3=(b_3-b_1)(1-H^2/H_E^2)$. The effective action, obtained by substituting relations (27) into Eq. (5), has the form

$$S_{\text{eff}}=v_0\int_{-\infty}^{+\infty}d\tau\left\{\frac{\chi_\perp}{2\gamma^2}\varphi_\tau^2+i\frac{M_0}{\gamma}\left(1-\frac{H}{H_E}\right)\varphi_\tau+\frac{\tilde{b}_3}{2}\sin^2\varphi\right\}. \tag{29}$$

An analogous action describes the dynamics of the magnetization of a single-axis antiferromagnet in a magnetic field.[15] Antisymmetric exchange in the case **H**||**d** does not have a substantial effect on the spin dynamics. Solving Eq. (28), we obtain the instanton trajectories

$$\varphi_\pm=\pm 2\arctan\{\exp[\omega(\tau-\tau_0)]\}, \tag{30}$$

where $\omega=\gamma\sqrt{\tilde{b}_3/\chi_\perp}$ is the antiferromagnetic resonance frequency. Contributions to the action on these trajectories differ only in their imaginary part

$$S_E^\pm=\frac{2v_0}{\gamma}\sqrt{\tilde{b}_3\chi_\perp}\mp i\frac{M_0v_0}{\gamma}\left(1-\frac{H}{H_E}\right)\pi. \tag{31}$$

The term $\mp iM_0v_0\pi/\gamma=\mp i2\pi\hbar S_0$ (S_0 is the total spin of one sublattice) leads to phase differences that are multiples of 2π in the transition probability amplitudes of alternative trajectories, which does not show up in the instanton interference. However, the field contribution to the imaginary part of the action does result in inteference of instanton amplitudes. Calculating the total probability of the transition from $\varphi=0$ to $\varphi=(2n+1)\pi$ $(n=0,\pm1,\pm2,\dots)$ by summing over all the instanton and anti-instanton trajectories, we obtain the following expression for the tunneling rate:

$$\Gamma(H)\simeq 4\omega\sqrt{\frac{S_{\text{cl}}}{2\pi\hbar}}\left|\cos\left(2\pi S_0\frac{H}{H_E}\right)\right|\exp\left[-\frac{S_{\text{cl}}}{\hbar}\right]D_\theta, \tag{32}$$

where $S_{\text{cl}}=\text{Re } S_E^\pm=2v_0\sqrt{\tilde{b}_3\chi_\perp}/\gamma$ and

$$D_\theta=\left|\frac{\det\left\{-\partial_s^2+\frac{\chi_\perp v_0^2}{\hbar^2\gamma^2}\left(\frac{d^2}{A}+A\frac{H^2}{H_E^2}-b_1\left(1-\frac{H^2}{H_E^2}\right)-\frac{2\tilde{b}_3}{\text{ch}^2(\omega\tau)}+i\frac{2M_0}{\gamma}\frac{H}{H_E}\frac{\omega}{\text{ch}(\omega\tau)}\right)\right\}}{\det\left\{-\partial_s^2+\frac{\chi_\perp v_0^2}{\hbar^2\gamma^2}\left(\frac{d^2}{A}+A\frac{H^2}{H_E^2}-b_1\left(1-\frac{H^2}{H_E^2}\right)\right)\right\}}\right|^{-1/2}.$$

It can be shown that for $H=0$, the ratio of determinants is given by (24), i.e.,

$$D_\theta=\left[\frac{\sqrt{d^2/A-b_1}+\sqrt{b_3-b_1}}{\sqrt{d^2/A-b_1}-\sqrt{b_3-b_1}}\right]^{1/2}$$

and for $d^2/A\gg b_3$ or $H\gg H_A$ we have $D_\theta\simeq 1$.

Thus, in contrast to the case **H**||**a**, the magnetic field creates a phase shift in the tunneling amplitudes over alternative trajectories, as a result of which they interfere. Consequently, the tunneling rate oscillates with period $\Delta H=H_E/2S_0$, where S_0 is the total spin of one sublattice. Interference suppression of tunneling of the magnetization in a weak ferromagnet is reminiscent of oscillations of the current of electrons scattered by a magnetic field in the Bohm–Aharonov effect.[31] Moreover, an action that takes a form similar to (5), as noted in Ref. 17, describes field interference of electrons in a metallic conductor with charge density waves. The magnetic field here plays the role of a normalized magnetic potential. Instanton interference also arises in single-axis ferromagnets, in which the magnetic field is perpendicular to the easy axis.[10] The period of the oscillations, however, in a ferromagnet is determined by the magnetic anisotropy field H_A, while in antiferromagnets, it is determined by the exchange field H_E. Thus, the presence of a spontaneous moment in the given case has no effect on quantum interference of the tunneling amplitudes, in contrast to an uncompensated antiferromagnet, in which the presence of a spontaneous moment can lead to dependence of the macroscopic quantum coherence on the uncompensated spin, as

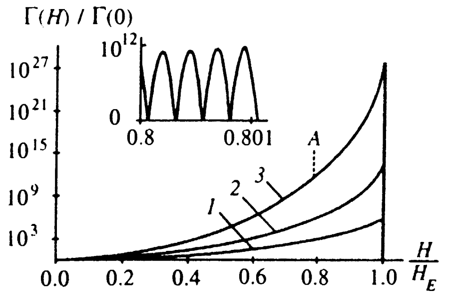

FIG. 5. Field dependence of the tunneling rate of the antiferromagnetism vector on the angular phase for $\mathbf{H}\|\mathbf{b}$: 1) $v_0=5\ \mathrm{nm}^3$, 2) $v_0=10\ \mathrm{nm}^3$, 3) $v_0=20\ \mathrm{nm}^3$. The inset shows an enlarged fragment of the field dependence at the point A.

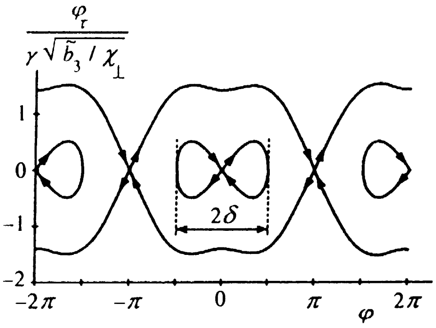

FIG. 6. Phase portrait of the dynamical equations corresponding to the action (34).

in a ferromagnet.[19] Figure 5 plots the field dependence of the tunneling rate for $YFeO_3$, calculated according to Eq. (32). Note that the lack of any influence of the dissipative environment of the spins, lattice vibrations, magnetic impurities, or other factors is essential for the quantum tunneling effects considered here.[2)]

5. H‖c. MACROSCOPIC QUANTUM TUNNELING IN THE REGION OF MAGNETIC HYSTERESIS

The last of the three cases which we consider has to do with spontaneous remagnetization in the region of magnetic hysteresis, when the magnetic field $H<H_c=H_D/2-\sqrt{(H_d/2)^2-H_AH_E}$, is anti-aligned with the equilibrium direction of the magnetization $\mathbf{F}\|\mathbf{c}$ (in this case $\mathbf{G}\|\mathbf{a}$). This state is metastable, and by virtue of quantum fluctuations, can transition to the stable equilibrium state with $\mathbf{F}\|\mathbf{H}$ and $\mathbf{G}\|-\mathbf{a}$. For $H>H_A$, this equilibrium state is nondegenerate due to the antisymmetric exchange interaction, which induces a spontaneous magnetic moment that depends on the direction of the vector $\mathbf{G}$.

In contrast to the two previous cases $\mathbf{H}\|\mathbf{a}$ and $\mathbf{H}\|\mathbf{b}$, in which tunneling leads to quantum mixing of the energetically degenerate phase states of the spin system, here a resonant interaction takes place between the wave function of the metastable state $|b\rangle$ and the wave function $|e\rangle$ of the nearest excited energy level of the deeper potential well with $\mathbf{F}\|\mathbf{H}$. The tunneling process in this case can be arbitrarily divided into two steps: sub-barrier motion up to the turning point and the transition to the resonance level.

The trajectories of sub-barrier motion are found from the variational conditions $\delta S_E/\delta\boldsymbol{\Omega}=0$. Proceeding analogously to the preceding cases, we find the trajectories passing near the ac plane of the crystal. Analysis of the Euler–Lagrange equations shows that the desired solutions have the form

$$\theta\cong\frac{\pi}{2}+\mathcal{O}\left(\frac{d}{A}\right)^2,$$

$$\beta\cong\frac{d}{A}-\frac{H}{H_E}\cos\varphi+\mathcal{O}\left(\frac{d}{A}\right)^2, \tag{33}$$

$$\varepsilon\cong-i\frac{M_0}{A\gamma}\varphi_\tau+\mathcal{O}\left(\frac{d}{A}\right)^2,$$

and the dynamics of the angle φ are described by the effective action

$$S_{\mathrm{eff}}=v_0\int d\tau\left\{\frac{\chi_\perp}{2\gamma^2}\varphi_\tau^2+i\frac{M_0}{\gamma}\varphi_\tau+\frac{\tilde{b}_3}{2}\times(1-\cos\varphi)(\cos\varphi-\cos\delta)\right\}, \tag{34}$$

where $\tilde{b}_3=(b_3-b_1)(1+H^2/H_AH_E)$ and $\delta=\cos^{-1}(-1+2dH/\tilde{b}_3H_E)$. The first integral of the dynamical system defined by the action (34) takes the form

$$\frac{\chi_\perp}{2\gamma^2}\varphi_\tau^2-\frac{\tilde{b}_3}{2}(1-\cos\varphi)(\cos\varphi-\cos\delta)=E. \tag{35}$$

Figure 6 shows the phase portrait of this system, constructed according to Eq. (35). As can be seen from the figure, there are two alternative instanton solutions departing from the point $\varphi=0$, corresponding to the zero value of the first integral and having different turning points $\boldsymbol{\Omega}_\pm=(\pi/2,0,\beta_0,\pm\delta)$, where $\beta_0=d/A-H\cos\delta/H_E$. Integrating relation (35) over time, we obtain the two instanton trajectories

$$\varphi=\pm2\arctan\left\{\frac{2\tan[\delta/2]\exp[\omega(\tau-\tau_0)]}{1+\exp[2\omega(\tau->t_0)]}\right\}, \tag{36}$$

where $\omega=\gamma\sqrt{(1-\cos\delta)\tilde{b}_3/2\chi_\perp}$.

The contribution to the action in the instanton solutions is easily found by evaluating the integral in expression (34), taking account of the integral relation (35)

$$S_E^\pm=\frac{2v_0\sqrt{\tilde{b}_3\chi_\perp}}{\gamma}\left\{\sqrt{\frac{1-\cos\delta}{2}}+\frac{1+\cos\delta}{2}\times\ln\left(\frac{\sqrt{1-\cos\delta}+\sqrt{2}}{\sqrt{1+\cos\delta}}\right)\right\}\pm i\frac{M_0v_0}{\gamma}\delta. \tag{37}$$

Consequently, the contributions to the tunneling rate of the trajectories (36) are

$$\Gamma^\pm(H)\cong A(H)\exp[-S_E^\pm/\hbar], \tag{38}$$

where $A(H)\equiv A^*(H)\cong A^-(H)$.

The transition amplitude to state $|e\rangle$ can be obtained approximately, as in Ref. 12, by expanding it in terms of the transition amplitudes to the coherent states $|\mathbf{\Omega}\rangle$ and noting that the dominant contribution is due to transitions via the states $|\mathbf{\Omega}_{\pm}\rangle$, i.e.,

$$K_{be}=\int d\mathbf{\Omega}K_{b\Omega}\langle\mathbf{\Omega}|e\rangle\cong K_{b\Omega-}\langle\mathbf{\Omega}_{-}|e\rangle+K_{b\Omega+}\langle\mathbf{\Omega}_{+}|e\rangle, \tag{39}$$

where $\langle\mathbf{\Omega}_{\pm}|e\rangle$ is the projection of the resonant level on the coherent states $|\mathbf{\Omega}_{\pm}\rangle$. We can then write down the expression for the decay rate of the metastable state

$$\Gamma=|\Gamma_{-}\langle\mathbf{\Omega}_{-}|e\rangle+\Gamma_{+}\langle\mathbf{\Omega}_{+}|e\rangle|. \tag{40}$$

To analyze expression (40) further, we expand the wave functions $\langle\mathbf{\Omega}_{\pm}|$ and $|e\rangle$ in eigenfunctions of the z-projection operators for the total spins of each sublattice $|S_z^{(j)}\rangle$:

$$\langle\mathbf{\Omega}_{\pm}|=\prod_{j=1,2}\langle\theta_j,\varphi_j|\left(\cos\frac{\theta_j}{2}\right)^{2S^{(j)}}\exp\left[\tan\frac{\theta_j}{2}e^{i\varphi_j}\hat{S}_{-}^{(j)}\right]$$

$$=\prod_{j=1,2}e^{i\varphi_j S_0}\sum_{S_z^{(j)}}\langle S_z^{(j)}|e^{-i\varphi_j S_z^{(j)}}b_{S_z^{(j)}}, \tag{41}$$

where

$$b_{S_z^{(j)}}=2^{-S_0}\left[\frac{(2S_0)!}{(S_0-S_z^{(j)})!(S_0+S_z^{(j)})!}\right]^{1/2},$$

and S_0 is the total spin of the sublattice. Analogously,

$$|e\rangle=\sum_{S_z^{(1)},S_z^{(2)}}a_{S_z^{(1)}S_z^{(2)}}|S_z^{(1)}\rangle|S_z^{(2)}\rangle. \tag{42}$$

The excited state $|e\rangle$ in the quasiclassical approximation corresponds to the resonant eigenmode of the oscillations of the sublattice spins that satisfies the equations $\delta S_E/\delta\mathbf{\Omega}=0$ in real time and has the form $\mathbf{\Omega}=(\pi/2,\varepsilon,\beta,\varphi)$, where ε and β are related to φ by (33), and the angle φ oscillates with amplitude δ. The fact that the projection of the antiferromagnetism vector on the z axis vanishes in the resonant mode implies that $S_z^{(1)}=S_z^{(2)}$, i.e., that the oscillations of the z-components of the spins in the sublattices are identical. This gives us the right to set $a_{S_z^{(1)}S_z^{(2)}}=0$ in expansion (42) for the wave vector $|e\rangle$, if $S_z^{(1)}\neq S_z^{(2)}$. In addition, from the invariance of the master Hamiltonian (1) (for $d_1=-d_3$) under a simultaneous change of sign of the z components of the sublattice spins $S_z^{(1)}$, $S_z^{(2)}\rightarrow -S_z^{(1)},-S_z^{(2)}$, it follows that the wave functions $|e\rangle$ divide into even and odd with respect to such an inversion operation. For the even functions ($|e\rangle_{\rm even}$), $a_{S_z^{(1)}S_z^{(2)}}=a_{-S_z^{(1)}-S_z^{(2)}}$, and for the odd ($|e\rangle_{\rm odd}$), $a_{S_z^{(1)}S_z^{(2)}}=-a_{-S_z^{(1)}-S_z^{(2)}}$. Hence, after substituting relations (41) and (42) into (39) and regrouping terms, we obtain

$$\Gamma=|\Gamma_{\pm}|\sum_{m=0}^{S_0}C_m\cos(2m\delta) \tag{43}$$

for S_0 integer and even wave functions $|e\rangle$, and

$$\Gamma=|\Gamma_{\pm}|\sum_{m=0}^{S_0-1/2}C_{m+1/2}\cos[(2m+1)\delta] \tag{44}$$

for S_0 half-integer and odd wave functions $|e\rangle$. Here $C_0=2b_0^2a_{00}$, $C_m=4b_m^2a_{mm}$, $m\neq 0$.

From these formulas it follows that in the absence of a magnetic field, when $\delta=\pi$, freezing of tunneling due to instanton interference, in contrast to the case of a ferromagnet, takes place neither for Kramers-type nor for non-Kramers-type ions. However, for $H=H_d-\sqrt{H_d^2-H_AH_E}$, when $\delta=\pi/2$, in a weakly ferromagnetic particle with half-integer spin of the sublattices the tunneling rate to a resonant level vanishes. There is no such effect for particles with integer spin, and only nonmonotonic variations of the tunneling rate with varying magnetic field are possible, due to the fact that a set of harmonics drops out of the expansion (43) at certain values of the magnetic field, for example, when $\delta=\pi/4$, $3\pi/4$, etc. Such a dependence of the tunneling rate on the field, as follows from Ref. 12, is also possible in a ferromagnetic particle.

It should be noted that in the presence of a dissipative environment, relaxation processes will influence the character of the instanton interference, going so far as to suppress it. In that case, due to quantum fluctuations, the decay of the metastable state will vary monotonically with the field, according to the envelope of the tunneling rate (38).

6. RESULTS AND DISCUSSION

Thus, we have shown that in a single-axis antiferromagnetic particle with antisymmetric exchange the field dependence of macroscopic quantum tunneling of spins significantly depends on the orientation of the magnetic field in the crystal. When the magnetic field is aligned with the antisymmetric exchange vector ($\mathbf{H}\|\mathbf{d}$), quantum mixing of energetically degenerate states with anti-alignment of their antiferromagnetism vectors ($\mathbf{G}\|\pm\mathbf{a}$) is possible, with resultant energy splitting of the ground state up to the field that collapses the antiferromagnetic sublattices $H_{sflip}=H_E+H_A$. In weakly ferromagnetic particles of the type $YFeO_3$, in which the spins are reoriented in the ac crystal plane, thanks to the presence of alternative instanton trajectories, the corresponding tunneling amplitudes interfere, which leads to periodic freezing of macroscopic quantum tunneling in a magnetic field with period proportional to the exchange interaction of an elementary spin pair $\Delta H\propto H_E/S_0$. A similar analysis for a different magnetic anisotropy, e.g., in compounds of the type $DyFeO_3$ or $TmFeO_3$, in which the preferred plane of rotation in weak magnetic fields is the ab plane of the crystal, shows that quantum interference of instantons is suppressed by the anisotropy. However, in the region of strong fields exceeding the characteristic field $H_{sflop}=\sqrt{H_AH_E}$ (the spin-flop transition field), interference of antiferromagnetic instantons will also occur, as in the first case, since in this field region the ac plane becomes energetically favored. The described mechanism of freezing of macroscopic quantum coherence spin-states in a magnetic field due to instanton interference can also arise in a ferromagnetic particle. However, the periodicity of variation of the tunneling rate in this case is governed by the anisotropy energy of an elementary spin.[10]

If the magnetic field acts perpendicular to the antisymmetric exchange vector ($\mathbf{H}\perp\mathbf{d}$) and does not remove the energy degeneracy (when $\mathbf{H}\|\mathbf{a}$), then tunneling of the antiferromagnetism vector between degenerate angular phases is not accompanied by instanton interference in compounds of the type $YFeO_3$, since in weak fields $H<H_c=-H_d/2+\sqrt{(H_d/2)^2+H_AH_E}$ where (and only where) energy degeneracy of the angular phases in such a particle exists, antiferromagnetic interference is suppressed by anisotropy (in "ab-crystals" it arises when $H>H_{sflop}$).

Finally, when the magnetic field acts along the main crystallographic axis ($\mathbf{H}\|\mathbf{c}$) it is possible to observe spontaneous remagnetization associated with quantum fluctuations in the region of magnetic hysteresis. Here quantum interference of instantons also takes place, but its mechanism differs from the previous one. This is because the intermediate coherent spin states $|\mathbf{\Omega}_{\pm}\rangle$ to which the system transitions as a result of alternative sub-barrier motion are not identical. They can transition one into the other thanks to coherent rotation of the spins in a magnetic field if the conditions of phase synchronism are satisfied. Because of this oscillations of the tunneling rate arise, whose period is determined by the coercion field $\Delta H \propto b/m$. As in a ferromagnet with half-integer total spin,[13,14] in the case under consideration total freezing of macroscopic quantum tunneling is possible if the total spin of an individual sublattice is half-integer. However, it takes place not in zero field, but in a finite field within the magnetic hysteresis range, when $H=H_d-\sqrt{H_d^2-H_AH_E}$ and when the value of the rotation angle of the antiferromagnetism vector at the turning point in the tunneling reaches $\delta=\pi/2$.

The instanton interference effects considered here are strongly related to coherent rotation of the spins. The interaction of macroscopic spins with lattice vibrations, spin-wave excitations at zero temperature, etc., can change the picture of macroscopic quantum tunneling. As was pointed out in Refs. 34 and 35, the quantum coherence of spins may be most greatly affected by the presence of weakly coupled spins in a dissipative environment, creating topological decoherence and being responsible for other mechanisms of decoherence. To minimize such effects, it is necessary to use nonmagnetic bonding media, and as the magnetic ions in the particle itself, one should use isotopes with zero nuclear spin such as ^{56}Fe and ^{59}Ni. The influence of a dissipative environment can be crudely taken into account by assuming the absence of magnetic impurities on the basis of the model proposed in Ref. 36 of a dynamic system with one degree of freedom, linearly interacting with the field of oscillators. If the dissipative function of the system has the form $R=\eta\varphi_\tau^2$, where η is the characteristic damping parameter, then, according to this model, sub-barrier motion of such a system is described by the effective action

$$S_{\text{eff}}=S_E(\eta=0)+\frac{\eta}{4\pi}\int\int d\tau d\tau'\frac{[\varphi(\tau)-\varphi(\tau')]^2}{(\tau-\tau')^2}. \tag{45}$$

In antiferromagnets the dissipative function can be written as

$$R=\sum_{j=1,2}\alpha\frac{M_0v_0}{4\gamma}[(\theta_\tau^{(j)})^2+(\varphi_\tau^{(j)})^2\sin^2\theta^{(j)}],$$

where α is Gilbert's magnetic relaxation parameter. Therefore, for trajectories that pass close to the ac-plane, we have

$$R\simeq\alpha\frac{M_0v_0}{2\gamma}\varphi_\tau^2+O\left(\frac{d}{A}\right)^2.$$

Consequently, in our case the effective action takes the form

$$S_{\text{eff}}=S_E(\alpha=0)+\alpha\frac{M_0v_0}{8\pi\gamma}\int\int d\tau d\tau'\frac{[\varphi(\tau)-\varphi(\tau')]^2}{(\tau-\tau')^2}. \tag{46}$$

Mobility measurements on domain boundaries in $YFeO_3$ at $T=4$ K (Ref. 37) yield $\alpha\cong5\times10^{-4}$. Therefore, the last term in the action (46) can be treated as a perturbation and can be. estimated using the instanton solutions obtained above. Thus, for $H=0$, when $\varphi(\tau)=\pm2\arctan[\exp(\omega\tau)]$, we obtain $S_{\text{eff}}\simeq S_E(\alpha=0)+\alpha M_0v_0/2\gamma$. For particles with volume $v_0=5$ nm^3 the correction to the tunneling rate is

$$\frac{\Gamma(\alpha)}{\Gamma(\alpha=0)}=\exp\left[-\alpha\frac{M_0v_0}{4\hbar\gamma}\right]\approx0.89.$$

In compounds such as $TmFeO_3$, in which $\alpha=0.4$, it is no longer possible to treat the dissipative term as a perturbation. In this case the tunneling probability becomes exponentially small.

Thus, $YFeO_3$ is a very promising weakly-ferromagnetic material for observing macroscopic quantum tunneling. However, it should be noted that the estimates we have made here are quite crude and do not take account of the "freezing out" of phonons and spin excitations at zero temperature (and possibly other effects as well). It is possible that at very low temperatures the influence of a dissipative environment in orthoferrites with strongly anisotropic ions is not so great. But in this case it must be borne in mind that in $RFeO_3$ compounds with ions of the type Dy, Tb, the spin tunneling dynamics can differ substantially from that considered here: because of the strong relativistic interaction in rare-earth ions, compared with the exchange interaction, they behave like "pseudo-Ising" ions. To describe the spin dynamics of such ions, it is necessary to allow for their interaction with multipole moments, as was done, for example, in Ref. 39.

In real experiments, decompensation of the magnetic sublattices can arise in the particles due to fluctuations of the magnetic ion density.[38] Some aspects of macroscopic quantum tunneling in decompensated antiferromagnets were considered by Chudnovsky in Ref. 19. If the spread of the magnetic moments is not large, specifically, if $\Delta M/M_0\ll\sqrt{H_A/H_E}$, which in $YFeO_3$ is roughly 3%, then the particles will behave as if they are weakly ferromagnetic, since the dominant influence on spin reorientation then comes from the spontaneous weakly-ferromagnetic moment $M_d/M_0\propto H_d/H_E$. In this case, however, changes in the instanton interference pattern are possible, associated with possible additional topological contributions to the action (in this regard, see Ref. 19). In particular, for $\mathbf{H}\|\mathbf{a}$ the presence of a half-integer uncompensated spin leads to freezing of macroscopic quantum tunneling over the entire range of magnetic fields. For $\mathbf{H}\|\mathbf{b}$, in the region of weak magnetic fields $H\ll H_{sflop}$, the field dependence of the tunneling rate for integer and half-integer uncompensated spin will differ

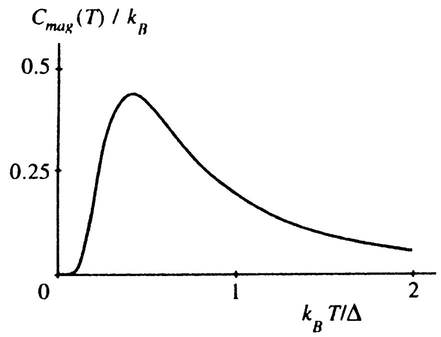

FIG. 7. Temperature dependence of the magnetic specific heat of a nanoparticle in the macroscopic quantum coherence state.

by a field shift of a half-period of the quantum oscillations, starting from zero for half-integer spin. At high magnetic fields, the interference pattern remains essentially unchanged. In general, however, to describe the peculiarities of the influence of decompensation on macroscopic quantum tunneling in a weak ferromagnet a separate analysis is necessary.

In conclusion, we note one more possibility of experimentally detecting macroscopic quantum coherence. In addition to existing methods for observing macroscopic quantum tunneling by low-temperature magnetic relaxation[2,3] and the resonant response to a weak external field,[1,38] we consider possibilities for observing this effect based on the thermodynamic properties of the behavior of small particles with two close-lying energy levels at superlow temperatures $T<T^*=\hbar\omega/k_B$. As is well known, for systems with splitting of the ground state into two close-lying levels with width $\Delta=2\hbar\Gamma$, the expression for the magnetic contribution to the partition function has the form

$$Z_{\text{mag}}\simeq\exp\left[-\frac{E_0}{k_BT}\right]\left[1+\exp\left(-\frac{\Delta}{k_BT}\right)\right], \tag{47}$$

where E_0 is the ground state energy. Hence we obtain the entropy of the system

$$S_{\text{mag}}=k_B\frac{\partial}{\partial T}(T\ln Z_{\text{mag}})\simeq k_B\ln\left[1+\exp\left(-\frac{\Delta}{k_BT}\right)\right]+\frac{\Delta}{T}\frac{\exp[-\Delta/k_BT]}{1+\exp[-\Delta/k_BT]} \tag{48}$$

and the magnetic specific heat

$$C_{\text{mag}}=T\frac{\partial}{\partial T}S_{\text{mag}}\simeq k_B\left(\frac{\Delta}{k_BT}\right)^2\frac{\exp[\Delta/k_BT]}{(1+\exp[\Delta/k_BT])^2}. \tag{49}$$

From an analysis of (49) it follows that the specific heat $C_{\text{mag}}(T)$ has a maximum at $T^*=0.417\Delta/k_B$ equal to $C_{\text{mag}}(T^*)=0.44k_B$ (Fig. 7). Thus, after measuring the specific heat of a magnetic particle, it is possible to obtain a numerical estimate of the energy splitting and the tunneling rate by determining the temperature at which it reaches its maximum value. Thus, for a $YFeO_3$ particle of volume $5\ \text{nm}^3$ in an external field corresponding to the maximum value of the energy splitting, $T^*\approx0.12$ K (in zero field $T^*\approx0.46\times10^{-6}$ K). For temperatures of the order of a hundred millikelvins the phonon specific heat of the lattice in the case of one particle can be estimated from $C_{\text{phon}}(T)=(4\pi^4/5)C_{\text{phon}}(\infty)(T/\Theta_D)^3$, where $\Theta_D=(\hbar\langle u\rangle/k_B)(6\pi^2N_0)^{1/3}\approx678$ K ($\langle u\rangle\approx4.3\times10^5$cm/s is the average speed of sound in the crystal, $N_0\approx1.5\times10^{23}\text{cm}^{-3}$ is the number of atoms per unit volume). Since $C_{\text{phon}}(0.12\ \text{K})\approx2.6\times10^{-8}k_B$ in the case under consideration, the phonon contribution to the specific heat can be neglected in comparison with the magnetic contribution. These estimates demonstrate that the field dependence of the frequency splitting in the state of macroscopic quantum coherence of antiferromagnetic nanoparticles can, in principle, be used for magnetic adiabatic cooling to superlow temperatures, as in the paramagnetic nuclear relaxation effect.[41]

ACKNOWLEDGMENTS

The authors thank the Russian Fund for Fundamental Research for support of this work (Grant No. 95-02-03737-a).

APPENDIX A

We consider the eigenvalue problem for the fluctuation operator

$$\hat{L}(s)=-\partial_s^2+\varepsilon^2-2/\cosh^2 s. \tag{A1}$$

Such an operator describes the fluctuation dynamics of the angle θ in normalized variables whose spectrum determines the magnitude of the pre-exponential factor in (22) and (24) in the absence of a magnetic field. The determinant of the matrix for the operator $\hat{L}(s)$ is equal to the product of the eigenvalues λ_j of the Sturm–Liouville problem $\hat{L}\psi_j=\lambda_j\psi_j$ with boundary conditions $\partial_s\psi(\pm T/2)=0$, i.e., $\det\hat{L}\equiv\Pi_j\lambda_j$. It follows from Ref. 30 that the ratio of determinants $D=\det\hat{L}(s)/\det\hat{L}(\infty)$ of the matrices of the fluctuation operators $\hat{L}(s)$ and $\hat{L}(\infty)=-\partial_s^2+\varepsilon^2$ can be expressed in terms of the asymptotic limit of the solutions of the linear equations

$$\hat{L}(s)\psi_1(s)=0 \tag{A2}$$

and

$$\hat{L}(\infty)\psi_2(s)=0 \tag{A3}$$

with the boundary condition

$$\psi_j(-T/2)=0,\quad \partial_s\psi_j(-T/2)=1, \tag{A4}$$

specifically,

$$D=\frac{\psi_1(T/2)}{\psi_2(T/2)}. \tag{A5}$$

The solution of Eq. (A3) has the form

$$\psi_2(s)=\frac{\sinh[\varepsilon(s+T/2)]}{\varepsilon}.$$

To find the solution of Eq. (A2) we make the substitution $\psi_1=(1-\xi^2)^{\varepsilon/2}u$, where $\xi=\tanh s$, and change of variable $x=(1-\xi)/2$. As a result, Eq. (A2) reduces to hypergeometric form

$$x(1-x)u_{xx}+(\varepsilon+1)(1-2x)u_x-(\varepsilon+2)(\varepsilon-1)u=0. \tag{A6}$$

The general solution of this equation is

$$u(x)=C_1u_1(x)+C_2u_2(x), \tag{A7}$$

where

$$u_1(x)=(1-x)^{-\varepsilon}F(2,-1,\varepsilon+1,x)$$
$$=(1-x)^{-\varepsilon}\left(1-\frac{2x}{\varepsilon+1}\right),$$

$$u_2(x)=x^{-\varepsilon}F(-1,2,1-\varepsilon,x)=x^{-\varepsilon}\left(1+\frac{2x}{\varepsilon-1}\right). \tag{A8}$$

Taking the boundary condition (A4) into account, we find the constants C_1 and C_2 and, in the limit $T\to+\infty$, we find

$$\psi_1(T/2)\to\frac{\varepsilon-1}{\varepsilon+1}\frac{\exp[\varepsilon T]}{2\varepsilon},\quad \psi_2(T/2)\to\frac{\exp[\varepsilon T]}{2\varepsilon}.$$

Consequently, the desired ratio of determinants (A5) in the limit $T\to+\infty$ is given by

$$D=\frac{\varepsilon-1}{\varepsilon+1}. \tag{A9}$$

Noting that the operator (A1) corresponds to the fluctuation part of the action (21) for

$$\varepsilon=\sqrt{\frac{d^2/A-b_1}{b_3-b_1}}$$

formula (A9) then gives

$$D=\frac{\sqrt{d^2/A-b_1}-\sqrt{b_3-b_1}}{\sqrt{d^2/A-b_1}+\sqrt{b_3-b_1}}. \tag{A10}$$

Authors are grateful to the Russian Fund for Fundamental Research for their support of this work (grant No. 95-02-03737-a).

[1)] Γ_4 denotes the irreducible representation of the spatial symmetry group of the crystal D_{2h}^{16} (Ref. 23).

[2)] The study of processes whereby a dissipative environment affects macroscopic quantum tunneling of spins is still in its infancy. We direct the reader's attention to two interesting papers[32,33] that consider the influence of acoustic vibrations and nuclear magnetic spins on the tunneling of the magnetic moment in a ferromagnet. As shown in Refs. 34 and 35, the presence in a dissipative environment of isolated spins radically alters the tunneling dynamics and can completely destroy coherence. This being the case, it would be desirable to minimize the presence of magnetic impurities in the particle and the bonding medium according to the recommendations in Ref. 34 (see Appendix A).

[1] D. D. Awschalom, M. A. McCord, and G. Grinstein, Phys. Rev. Lett. **65**, 783 (1990).

[2] B. Barbara, L. C. Sampaio, J. E. Wegrone *et al.*, J. Appl. Phys. **73**, 6703 (1993).

[3] J. Tejada, X. X. Zhang, and L. I. Ballsells, J. Appl. Phys. **73**, 6709 (1993).

[4] E. M. Chudnovsky and L. Gunther, Phys. Rev. Lett. **60**, 661 (1988).

[5] P. C. E. Stamp, Phys. Rev. Lett. **66**, 2802 (1991).

[6] E. M. Chudnovsky, O. Iglesias, and P. C. E. Stamp, Phys. Rev. B **46**, 5392 (1992).

[7] E. M. Chudnovsky and L. Gunther, Phys. Rev. B **37**, 9455 (1987).

[8] L. Gunther, *Studies of Magnetic Properties of Fine Particles and Their Relevance to Materials Science*, Elsevier, Amsterdam (1992), p. 213.

[9] E. M. Chudnovsky, Phys. Rev. B **47**, 9102 (1992).

[10] A. Garg, Europhysics Lett. **22**, 205 (1993).

[11] A. Garg and G.-H. Kim, Phys. Rev. B **45**, 12921 (1991).

[12] E. M. Chudnovsky and D. P. Di Vincenzo, Phys. Rev. B **48**, 10548 (1993).

[13] D. Loss, D. P. Di Vincenzo, and G. Grinstein, Phys. Rev. Lett. **69**, 3232 (1992).

[14] J. von Delft and C. L. Henley, Phys. Rev. Lett. **69**, 3236 (1992).

[15] B. Barbara and E. M. Chudnovsky, Phys. Lett. A **145**, 205 (1990).

[16] I. V. Krive and O. B. Zaslavskii, J. Phys. Condens. Matter **2**, 9457 (1990).

[17] E. N. Bogacheck and I. V. Krive, Phys. Rev. B **46**, 14559 (1992).

[18] I. E. Dzyaloshinskiĭ, Zh. Éksp. Teor. Fiz. **32**, 1547 (1957) [Sov. Phys. JETP **5**, 1259 (1957)].

[19] E. M. Chudnovsky, J. Magn. Magn. Mater. **140–144**, 1821 (1995).

[20] Ji-M. Duan and A. Garg, Physica B **194–196**, 323 (1994).

[21] V. Yu. Golyshev and A. F. Popkov, Europhysics Lett. **29**, 327 (1995).

[22] K. P. Belov, A. K. Zvezdin, A. M. Kadomtseva, and R. Z. Levitin, *Orientational Transitions in Rare-Earth Magnets* [in Russian], Nauka, Moscow (1979).

[23] E. A. Turov, *Physical Properties of Magnetically Ordered Crystals* [in Russian], USSR Academy of Sciences, Moscow (1963).

[24] E. Manousakis, Rev. Mod. Phys. **63**, 1 (1991).

[25] A. K. Zvezdin, Pis'ma Zh. Éksp. Teor. Fiz. **29**, 605 (1979) [JETP Lett. **29**, 553 (1979)].

[26] V. G. Bar'yakhtar, B. A. Ivanov, and M. V. Chetkin, Usp. Fiz. Nauk **146**, 417 (1985) [Sov. Phys. Usp. **28**, 563 (1985)].

[27] M. V. Berry, Proc. R. Soc. London A **392**, 45 (1994).

[28] R. P. Feynman and A. R. Hibbs, *Quantum Mechanics and Path Integrals*, McGraw-Hill, New York (1965).

[29] C. G. Callan and S. Coleman, Phys. Rev. D **16**, 1762 (1977).

[30] S. Coleman, *Aspects of Symmetry*, Cambridge Univ. Press, New York (1985), Ch. 7.

[31] Y. Aharonov and D. Bohm, Phys. Rev. **115**, 485 (1959).

[32] A. Garg and G. H. Kim, Phys. Rev. B **43**, 712 (1991).

[33] A. Garg, Phys. Rev. Lett. **70**, 1541 (1993).

[34] N. V. Prokof'ev and P. C. E. Stamp, J. Phys. Condens. Matter **5**, L667 (1994).

[35] P. C. E. Stamp, Physica B **197**, 133 (1994).

[36] A. O. Caldeira and A. J. Leggett, Ann. Phys. (N. Y.) **149**, 374 (1983).

[37] S. O. Demokritov, A. I. Kirilyuk, and N. M. Kreines *et al.*, J. Magn. Magn. Mater. **102**, 339 (1991).

[38] D. D. Awschalom *et al.*, Phys. Rev. Lett. **68**, 3092 (1992).

[39] A. K. Zvezdin and A. F. Popkov, JETP Lett. **57**, 562 (1993).

[40] L. D. Landau and E. M. Lifshitz, *Quantum Mechanics*, 3rd. ed., Pergamon Press, Oxford (1977).

[41] O. V. Louasmaa, *Experimental Principles and Methods Below 1 K*, Academic Press, San Diego (1974).

Translated by Paul F. Schippnick