

Macroscopic quantum tunneling in small antiferromagnetic particles: Effects of a strong magnetic field

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(Received 2 April 2004; revised manuscript received 18 October 2004; published 29 December 2004)

We consider an effect of a strong magnetic field on the ground-state and macroscopic coherent tunneling in small antiferromagnetic particles with uniaxial and biaxial single-ion anisotropy. We find several tunneling regimes that depend on the direction of the magnetic field with respect to the anisotropy axes. For the case of a purely uniaxial symmetry and the field directed along the easy axis, an exact instanton solution with two different scales in imaginary time is constructed. For a rhombic anisotropy the effect of the field strongly depends on its orientation: with the field increasing, the tunneling rate increases or decreases for the field parallel to the easy or medium axis, respectively. The analytic results are complemented by numerical simulations.

DOI: 10.1103/PhysRevB.70.214430

PACS number(s): 75.45.+j, 03.65.Sq, 75.10.Hk, 75.50.Tt

During the last decade macroscopic coherent quantum tunneling between two equivalent classical ground states in macroscopic or, to be more precise, mesoscopic magnetic systems became an object of intense experimental and theoretical investigations, see for a review Refs. 1 and 2. In the physics of magnetism such systems are, for instance, small magnetic particles, magnetic clusters, and high-spin molecules. The interest in tunneling effects is associated with the possibility of using these systems as potential elements for quantum computers. Initially, the calculations of tunneling effects were carried out for ferromagnets.^{3,4} However, it happened that the tunneling effects were experimentally observed by the resonant absorption of electromagnetic waves for antiferromagnetic ferritin particles.⁵ The effect of the magnetic field on the tunneling probability in ferritin particles have been experimentally studied, see Ref. 6. According to theoretical estimations,^{7,8} the level splitting in antiferromagnets is stronger than in ferromagnets and the effects can be observed at higher temperatures. Antiferromagnets are more convenient for experimental investigations of tunneling effects.

The interest in tunneling in compensated antiferromagnets has been renewed after the synthesis of high-spin molecules with antiferromagnetic coupling of spins, so-called ferric wheels, such as Fe₆, Fe₁₀, Cr₈, see, for instance, Refs. 9–11. The interference effects caused by the gyroscopic terms in the σ -model approximation were of main interest.¹² For compensated antiferromagnets such effects can arise due to an external magnetic field^{12,13} or a certain kind of the Dzyaloshinskii-Moriya interaction.¹⁴ Chioleri and Loss¹³ have also found a type of interference effect for such antiferromagnets placed in a strong magnetic field directed perpendicular to the easy axis of the antiferromagnetic particle. This effect caused by the imaginary part of the fluctuation determinant together with the direct contribution of the field to the imaginary part of the Euclidean action produces the periodic dependence of the level splitting on the field strength. Then Hu *et al.*¹⁵ have investigated these effects for a more general case of antiferromagnets with rhombic anisotropy. They have considered the case of the field directed along the hard axis

and found the behavior common to that in Ref. 13. They also suggested that such properties are universal for a wide class of antiferromagnets subject to a strong magnetic field.

In this paper we consider the effects of coherent quantum tunneling for a compensated antiferromagnetic particle with rhombic anisotropy placed in the magnetic field. In Sec. I we describe the model used for the evaluation of tunnel splitting and analyze the static energy of the particle for three different orientations of the magnetic field along the crystalline axes. Section II is devoted to the calculation of the level splitting of the ground states in the instanton approximation for the orientations of the field along the easy, middle, and hard axis separately and for relevant ranges of the field strength. Section III contains a qualitative comparison of analytical results with data of numerical calculations for level splitting and a direct comparison for the Euclidean action. The final section resumes obtained results and gives a short survey of experiments where the predicted effects could be observed. We use the standard semiclassical analysis based on the instanton technique applied to the nonlinear σ -model, as well as the direct numerical diagonalization of the corresponding quantum spin Hamiltonian. We demonstrate that the behavior of the level splitting is highly sensitive to the orientation of the magnetic field. In fact, three different scenarios for the orientation of the field along the symmetry axes are found.

I. NONLINEAR σ MODEL FOR ANTIFERROMAGNETS IN A MAGNETIC FIELD

We start from the Hamiltonian describing a magnetic particle with an even number N of magnetic ions with the spin S and coupled with the nearest-neighbor antiferromagnetic exchange interaction J . We also assume that a single-ion anisotropy with rhombic symmetry and the magnetic field \mathbf{H} are present. The macroscopic Hamiltonian of the system can be written as

$$\mathcal{H} = J \sum_{\langle \alpha\beta \rangle} S_\alpha S_\beta + B_u \sum_\alpha [(S_\alpha^x)^2 + (S_\alpha^y)^2] + B_p \sum_\alpha (S_\alpha^z)^2 - g\mu_B \sum_\alpha \mathbf{H} \cdot S_\alpha. \quad (1)$$

Here, S_α is the spin at the α th site, $g \approx 2$ is the Landé factor, and μ_B is the Bohr magneton. The first term describes the isotropic exchange interaction $J > 0$ and the summation in this term is extended over the pairs of nearest neighbors. The second and third terms give the simplest form of a rhombic single-ion anisotropy; B_u and B_p are the constants of the uniaxial anisotropy and the anisotropy in the basal plane xy , respectively. We assume that $B_u > 0$ and $B_p > 0$. Thus, z is the easy axis and x, y are the medium and hard axes, respectively. The last term corresponds to the coupling spins with the magnetic field.

We will analyze the system under the assumption that all spin pairs are equivalent and have the same coordination number z . It is true for spin dimers ($z=1$), spin wheels ($z=2$), and can be a good approximation for mesoscopic three-dimensional crystalline particles with $N \gg 1$, in which the surface variation of parameters can be neglected. Assume that the Zeeman energy $g\mu_B H S$, the anisotropy energy $B_p S^2$, and $B_u S^2$ are much smaller than the exchange energy $J_z S^2$. The classical approximation gives that the spins in each sublattice are parallel. This assumption is a necessary condition for using the σ model as a model for describing both classic and quantum dynamics of antiferromagnets.¹³ For the semiclassical dynamics, especially for tunneling, it is convenient to use the Euclidean formulation, which is based on the introduction of imaginary time $\tau = it$.

The Euclidean action for the σ model that corresponds to the microscopic Hamiltonian (1) can be written as

$$\begin{aligned} \mathcal{A}_E[\mathbf{I}(\tau)] &= \int_{-\infty}^{+\infty} \mathcal{L}_E[\theta(\tau), \phi(\tau)] d\tau \\ &= N \int_{-\infty}^{+\infty} d\tau \left\{ \frac{\hbar^2}{4J_z} [\dot{\mathbf{I}}^2 - 2i\gamma \mathbf{H} \cdot (\mathbf{I} \times \dot{\mathbf{I}})] + w_a(\mathbf{I}) \right\}, \end{aligned} \quad (2)$$

where $\gamma = g\mu_B/\hbar$ is the gyromagnetic ratio, N is the number of magnetic ions in both sublattices, and the overdot denotes the derivative with respect to the imaginary time τ .

In this approach the total spin S_{tot} of the particle becomes a slave variable, and it is determined through the unit Néel vector \mathbf{I} and its derivative $\dot{\mathbf{I}}$ with respect to τ :

$$S_{\text{tot}} = \frac{\hbar N}{J_z} \{ \gamma [\mathbf{H} - \mathbf{I}(\mathbf{H} \cdot \mathbf{I})] + i(\mathbf{I} \times \dot{\mathbf{I}}) \}. \quad (3)$$

The function $w_a(\mathbf{I})$ is an effective energy of anisotropy per spin with renormalization caused by the external field

$$w_a(\mathbf{I}) = B_u S^2 (I_x^2 + I_y^2) + B_p S^2 I_y^2 + \frac{(g\mu_B)^2}{4J_z} (\mathbf{H} \cdot \mathbf{I})^2. \quad (4)$$

In accordance with the general rules of the semiclassical approximation formulated in the instanton language the amplitude of the tunnel transition from the state $\mathbf{l} = \mathbf{l}^-$ to $\mathbf{l} = \mathbf{l}^+$ is

proportional to $\exp(-\mathcal{A}_E/\hbar)$, where the value of \mathcal{A}_E is calculated from appropriate equations of motion under the boundary conditions: $\mathbf{I}(\tau) \rightarrow \mathbf{l}^-$ at $\tau \rightarrow -\infty$ and $\mathbf{I}(\tau) \rightarrow \mathbf{l}^+$ at $\tau \rightarrow +\infty$. Tunnel splitting in the so-called dilute instanton gas approximation is determined by the modulus of the sum of such amplitudes calculated along all instanton trajectories with a minimal value of $\text{Re } \mathcal{A}_E$.

It is convenient to introduce a polar parametrization for the unit vector \mathbf{l}

$$l_z = \cos \theta, \quad l_x = \sin \theta \cos \phi, \quad l_y = \sin \theta \sin \phi. \quad (5)$$

The Lagrangian from Eq. (2) in such a parametrization takes the form

$$\begin{aligned} \mathcal{L}_E[\theta(\tau), \phi(\tau)] &= N w_a(\theta, \phi) + \frac{\hbar^2 N}{4J_z} (\dot{\theta}^2 + \dot{\phi}^2 \sin^2 \theta \\ &\quad - 2i\gamma (\dot{\theta}(H_y \cos \phi - H_x \sin \phi) + \dot{\phi}[H_z \sin^2 \theta \\ &\quad - (H_x \cos \phi + H_y \sin \phi) \sin \theta \cos \theta]). \end{aligned} \quad (6)$$

Here we omitted the term with a full derivative proportional to $N S \dot{\phi}$ because it does not create the interference effects for the tunneling between opposite points of the unit sphere for a fully compensated antiferromagnet.

At zero magnetic field instanton solutions that give minima of the action (2) with the boundary conditions $\mathbf{I}(\pm\infty) \rightarrow \pm \hat{\mathbf{e}}_z$ can be easily found. The above-mentioned conditions take the form $\theta(-\infty) = 0, \theta(+\infty) = \pi$ with arbitrary values for $\phi(\pm\infty)$. The instanton solutions correspond to the rotation of \mathbf{l} in the symmetry planes of the system, i.e., $\phi = \pi k/2$, where k is integer, and

$$\cos \theta = \tanh(\bar{\omega}\tau), \quad \sin \theta = \frac{\sigma}{\cosh(\bar{\omega}\tau)}, \quad (7)$$

where $\sigma = \pm 1$ is the polarization of the instanton. The ‘‘frequency’’ $\bar{\omega}$ is $2S\sqrt{J_z B_u}/\hbar$ for the instanton paths going through the medium axis $\phi = 0, \pi$ and $\bar{\omega} = 2S\sqrt{J_z(B_u + B_p)}/\hbar$ for the paths going through the hard axis $\phi = \pm \pi/2$. The Euclidean action calculated on these instantons is simply $\mathcal{A}_E = \hbar N (\hbar \bar{\omega} / J_z)$. Following the saddle-point approximation that corresponds to the dilute instanton gas we take into account only paths with a minimal real part of the Euclidean action, i.e., with the smallest $\bar{\omega} = 2S\sqrt{J_z B_u}/\hbar$. Thus, at zero magnetic field the only instanton pair with $\sigma = \pm 1$ and the rotation of \mathbf{l} in the plane xz contributes to the tunneling. The presence of the magnetic field drastically changes the features of the spin tunneling between two different classical ground states in the antiferromagnetic particle.

First of all we consider the influence of the magnetic field on the static classical properties of the antiferromagnetic particle. We will use the expression (4) and restrict ourselves to the cases when the field is parallel to one of the symmetry axes x, y , or z . In these cases the field simply renormalizes the value of the appropriate anisotropy constants.

The influence of the field on the ground state is essential when the field is directed along the easy axis z . In this case the renormalized constant of the uniaxial anisotropy $B_u(H)$ can change its sign, $B_u(H) = B_u(1 - H^2/H_u^2)$, where

$$H_u = \frac{2S\sqrt{JzB_u}}{g\mu_B} \quad (8)$$

is the field of the familiar spin-flop phase transition in the classical theory of antiferromagnetism. When $H > H_u$ the states with $\mathbf{l} \parallel \pm \hat{e}_z$ become unstable and the ground states have \mathbf{l} parallel to $\pm \hat{e}_x$.

When \mathbf{H} is parallel to the medium axis x , the constant B_u increases, $B_u(H) = B_u + (g\mu_B H)^2 / (4JzS^2)$ and B_p decreases as $B_p(H) = B_p - (g\mu_B H)^2 / (4JzS^2)$, with the growth of the field. Thus, the field does not effect on the ground state, but a strong enough field can change the type of the axes x and y in the basal plane. When $H > H_p$, where

$$H_p = \frac{2S\sqrt{JzB_p}}{g\mu_B}, \quad (9)$$

the axis y becomes an easy direction in the basal plane (medium axis) and the axis x is a hard one.

Finally, if the magnetic field is directed along the hard axis y , both anisotropy constants increase as $B_{u,p}(H) = B_{u,p} + (g\mu_B H)^2 / (4JzS^2)$. In this case, the field does not change the ground state, as well as the type of the axes.

The naive substitution of the renormalized constants into the expressions (7) for an instanton and the Euclidean action calculated on it leads to a wrong prediction that when the field is directed along the easy axis $\mathcal{A}_E \rightarrow 0$ at $H \rightarrow H_u$, and for the field along the medium axis the values of \mathcal{A}_E on the two classes of trajectories [$\phi = \pi k$ and $\phi = (2k+1)\pi/2$] become equal at $H = H_p$. As we will show below, both suggestions are wrong.

In addition to the abovementioned renormalizations of the anisotropy constants, the field changes the dynamics of the vector \mathbf{l} and leads to the appearance of a gyroscopic term linear in $d\mathbf{l}/d\tau$ in the Lagrangian. The role of gyroscopic terms for tunneling in antiferromagnets has been discussed in Refs. 12 and 13. The authors of these papers have shown that the gyroscopic term caused by the magnetic field can create the imaginary part of the Euclidean action \mathcal{A}_E that is a linear function of the magnetic field. As we have shown, similar effects can also arise due to the Dzyaloshinskii-Moriya interaction.¹⁴ The imaginary part of the Euclidean action \mathcal{A}_E can lead to the interference effects and to the oscillations of the tunnel splitting as a function of the magnetic field. We will show below that the gyroscopic term produces a dynamic renormalization of the real part of \mathcal{A}_E , which is quadratic in the magnetic field. It can completely suppress the static contribution to \mathcal{A}_E coming from the renormalization of the anisotropy constants.

II. INSTANTON SOLUTIONS

In order to describe macroscopic quantum tunneling between two classical states $\mathbf{l} = \hat{e}_z$ and $\mathbf{l} = -\hat{e}_z$ in the saddle-point approximation, it is necessary to find instanton solutions of the two Euler-Lagrange equations for the Euclidean action (2) for the independent variables $\theta(\tau)$ and $\phi(\tau)$. Using the identities $(\delta/\delta\phi) \int d\tau \mathbf{H} \cdot (\mathbf{l} \times \dot{\mathbf{l}}) = -2\dot{\phi}(\mathbf{H}\mathbf{l}) \sin \theta$ and

$(\delta/\delta\theta) \int d\tau \mathbf{H} \cdot (\mathbf{l} \times \dot{\mathbf{l}}) = 2\dot{\theta}(\mathbf{H}\mathbf{l}) \sin \theta$ that can be obtained through the variation of the Lagrangian (6), it is convenient to write down the Euler-Lagrange equations with the magnetic field \mathbf{H} directed along an arbitrary symmetry axis

$$\begin{aligned} \ddot{\theta} - \{\omega_u^2 + \dot{\phi}^2 + \gamma^2(H_x^2 - H_z^2) \\ + [\omega_p^2 + \gamma^2(H_y^2 - H_x^2)] \sin^2 \phi\} \sin \theta \cos \theta \\ = -2i\dot{\phi}\gamma(\mathbf{H}\mathbf{l}) \sin \theta, \end{aligned} \quad (10a)$$

$$\begin{aligned} \ddot{\phi} \sin^2 \theta + 2\dot{\phi}\dot{\theta} \sin \theta \cos \theta \\ - [\omega_p^2 + \gamma^2(H_y^2 - H_x^2)] \sin^2 \theta \sin \phi \cos \phi \\ = 2i\dot{\theta}\gamma(\mathbf{H}\mathbf{l}) \sin \theta. \end{aligned} \quad (10b)$$

Here $\omega_u = \gamma H_u = 2S\sqrt{2JzB_u}/\hbar$ and $\omega_p = \gamma H_p = 2S\sqrt{2JzB_p}/\hbar$. The terms in the right-hand side are responsible for the gyroscopic dynamics of the vector \mathbf{l} . Note that we consider the field directed along one of the crystalline axes only, so only one of the components \mathbf{H} in the system (10) is nonzero.

It is important to note that in the case $\mathbf{H} \neq 0$ simple planar solutions such as $\phi = \pi k/2$ may not exist. If such solutions are absent, the full system (10) is equivalent to the Lagrange equations for a mechanical system with two degrees of freedom. To integrate such a system, the existence of two independent integrals of motion is necessary. For the general case $\omega_u \neq 0$ and $\omega_p \neq 0$ only one first integral is known

$$\mathcal{E} = \dot{\theta}^2 + \dot{\phi}^2 \sin^2 \theta - \omega_u^2 \sin^2 \theta - \omega_p^2 \sin^2 \theta \sin^2 \phi, \quad (11)$$

with the value $\mathcal{E} = 0$ for separatrix solutions we are interested in. For this reason the system (10) cannot be solved analytically. However, approximate solutions can be constructed for all cases of interest; see a detailed consideration in the following sections.

A. Field parallel to the easy axis

We will start from the case of the field parallel to the easy axis, for which the gyroscopic terms in the right-hand side of Eqs. (10) are independent of ϕ . The field does not violate the rotational symmetry around the easy axis, and the model with an isotropic basal plane ($\omega_p = 0$) has a physical meaning for this case. Its analysis leads to instructive results and we give it completely.

If $\omega_p = 0$, the system (10) has one more integral of motion, which can be written as

$$\Omega = (\dot{\phi} - i\gamma H) \sin^2 \theta = \text{const.} \quad (12)$$

Using the two integrals of motion (11) and (12), we can simplify the system to the ordinary differential equation for $\theta(\tau)$ only

$$\dot{\theta}^2 - \omega_u^2 \sin^2 \theta - \frac{\Omega^2}{\sin^2 \theta} = \mathcal{E} \quad (13)$$

and integrate it exactly. The instanton solutions with appropriate boundary conditions $\theta \rightarrow 0, \pi$, and $\dot{\theta} \rightarrow 0$ at $\tau \rightarrow \pm\infty$ correspond to the values of the integrals $\Omega = 0, \mathcal{E} = 0$. Thus,

the solution is $\phi = i\gamma H(\tau - \tau_1)$, where τ_1 is an arbitrary constant. Equation (13) simplifies to $\dot{\theta}^2 = \omega_u^2 \sin^2 \theta$, and its solution is described by the formula (7) with $\bar{\omega} = \omega_u$. In terms of the vector \mathbf{l} , the instanton solution for \mathbf{H} parallel to the easy axis and $\omega_p = 0$ takes the form

$$l_x = \sigma \frac{\cosh[\gamma H(\tau - \tau_1)]}{\cosh[\bar{\omega}_0(\tau - \tau_0)]}, \quad (14a)$$

$$l_y = i\sigma \frac{\sinh[\gamma H(\tau - \tau_1)]}{\cosh[\bar{\omega}_0(\tau - \tau_0)]}, \quad (14b)$$

$$l_z = \tanh[\bar{\omega}_0(\tau - \tau_0)]. \quad (14c)$$

This solution has correct instanton asymptotics $l_{x,y} \rightarrow 0$ and $l_z \rightarrow \pm 1$ at $\tau \rightarrow \pm\infty$ for all H in the range $\gamma H < \omega_u$, which corresponds to $H < H_u$. It is valid in the full region of stability of the states $\mathbf{l} = \pm \hat{e}_z$. Though the constant of effective anisotropy $B_u(H)$ changes from B_u at $H=0$ to 0 at $H=H_u$, the Euclidean action calculated on the solution (14) does not depend on H , its real part is the same as at $H=0$ and the imaginary part is zero. Thus, in the uniaxial case $B_p=0$ and $H < H_u$ the action is

$$\mathcal{A}_E = 2\hbar NS \sqrt{\frac{B_u}{J_z}}. \quad (15)$$

This simple example shows that the gyroscopic term can essentially suppress the static renormalization of the anisotropy constant. For the case $\omega_p = 0$ the renormalization is completely compensated by the gyroscopic term and \mathcal{A}_E is real and does not depend on the field.

For the case $\omega_p = 0$ an exact solution of the problem of small fluctuations around the classical instanton solution is possible to construct, and the preexponential factor can be explicitly written. To do so, we introduce small perturbations ϑ and μ as

$$\theta(\tau) = \theta_0(\tau) + \vartheta, \quad (16a)$$

$$\phi(\tau) = \phi_0(\tau) + \frac{\mu}{\sin \theta_0(\tau)}, \quad (16b)$$

where $\theta_0(\tau)$ and $\phi_0(\tau)$ correspond to the instanton solution given by Eqs. (14). The variational part of the action is $\mathcal{A}_E - \mathcal{A}_E^{(0)}$, where $\mathcal{A}_E^{(0)}$ is calculated on the unperturbed instanton solution. It can be written as a sum of two independent terms $\mathcal{A}_E - \mathcal{A}_E^{(0)} = \delta^2 \mathcal{A}_E^{(\vartheta)} + \delta^2 \mathcal{A}_E^{(\mu)}$ with the decoupled degrees of freedom ϑ and μ . Both terms have the same form

$$\delta^2 \mathcal{A}_E^{(\alpha)} = \frac{N\hbar^2 \bar{\omega}}{4J_z} \int_{-\infty}^{+\infty} d\xi (f^{(\alpha)}, \hat{M} f^{(\alpha)}), \quad (17)$$

where $\xi = \bar{\omega}\tau$, $f^{(\alpha)}$ is ϑ or μ for $\delta^2 \mathcal{A}_E^{(\vartheta)}$ or $\delta^2 \mathcal{A}_E^{(\mu)}$, respectively, and \hat{M} is the linear operator

$$\hat{M} = -\frac{d^2}{d\xi^2} + 1 - \frac{2}{\cosh^2 \xi}. \quad (18)$$

This operator frequently appears in scattering problems associated with soliton theory, so its properties are well stud-

ied. The full set of its eigenvalues and normalized eigenfunctions is

$$\hat{M} f_0 = 0, \quad f_0 = \frac{1}{\sqrt{2} \cosh \xi}, \quad (19a)$$

$$\hat{M} f_k = (1 + k^2) f_k, \quad f_k = \frac{(\tanh \xi - ik) e^{-ik\xi}}{\sqrt{L(1 + k^2)}}. \quad (19b)$$

The mode f_0 is a localized eigenfunction and the modes with $f = f_k$ form a continuous spectrum.

It is worthwhile to note that in contrast to other problems of macroscopic quantum tunneling, for the case of antiferromagnets with $\omega_p = 0$ the zero mode f_0 is present for *both* kinds of fluctuations. The second zero mode is caused by the exact rotational symmetry around the axis z . The full preexponential factor for this problem is a *square* of the usual fluctuation determinant $D = \sqrt{\mathcal{A}_E^{(0)}/(2\pi\hbar)}$, and the tunnel splitting of the lowest levels Δ takes the form

$$\Delta = C\hbar\omega_u \left(\frac{\mathcal{A}_E^{(0)}}{2\pi\hbar} \right) \exp(-\mathcal{A}_E^{(0)}/\hbar), \quad (20)$$

where C is a numerical constant of order of unity. The additional large factor $\sqrt{\mathcal{A}_E^{(0)}/(2\pi\hbar)}$ is a consequence that the instanton solution (14) in the case $\omega_p = 0$ contains two (not one, as usually) continuous parameters τ_1 and τ_0 . Note that the level splitting Δ does not depend on the magnetic field even if the fluctuation determinant is taken into account. This result can be explained using exact quantum-mechanical arguments. For uniaxial system the Hamiltonian of the system commutes with the z projection of the total spin \hat{S}_z , and the eigenstates of the problem can be characterized by definite values $S_z = 0, \pm 1, \pm 2, \dots$. The two lowest levels form a doublet with $S_z = 0$ and the magnetic field does not influence on them. For this model the spin-flop transition at the field $H = H_u$ corresponds to the change of the value of S_z for the lowest level from $S_z = 0$ to the value $S_z = 1$ or higher. Then, the growth of the magnetic field leads to the growth of S_z and a sawlike dependence $\Delta(H)$ appears. Since these effects are not associated with tunneling, we will not consider them in the following, and restrict ourselves only to the region $H < H_u$.

Now we consider a more general case $\omega_p \neq 0$. It is clear that at finite nonzero ω_p the additional factor $(\mathcal{A}_E/2\pi\hbar)^{1/2}$ is absent, and the preexponential factor is smaller than that for the uniaxial case $\omega_p = 0$. The levels cannot be characterized by the quantum number $S_z = 0, \pm 1, \pm 2, \dots$, and their splitting becomes dependent on the magnetic field.

The instanton solutions with $\theta = \theta(\tau)$, $\phi = 0$ exist at $H = 0$. It can be expected that at $\gamma H \ll \omega_u$ the value of ϕ or, more accurately, the appropriate projection of the vector \mathbf{l} , $l_y \approx \sin \phi \sin \theta_0$ is small, and the function $\theta(\tau)$ can be given by the solution similar to Eq. (14). If $\omega_p \gg \omega_u$, the out-of-plane components of \mathbf{l} are small, i.e. $|l_y| \ll 1$. Assuming that $|l_y| \ll 1$ for all values of parameters, the variational technique can be applied for evaluating the action. Choosing as a trial

function $\cos \theta = \tanh[\bar{\omega}(\tau - \tau_1)]$ with some parameter $\bar{\omega}$ that have to be found later, we rewrite Eq. (10b) in the linear approximation in l_y as

$$(\hat{M} + \epsilon)l_y(\xi) = \frac{2i\gamma H \sinh \xi}{\bar{\omega} \cosh^2 \xi}. \quad (21)$$

Here $\epsilon = (\omega_p/\bar{\omega})^2$, $\xi = \bar{\omega}\tau$, and \hat{M} is the linear operator (18) introduced above with non-negative eigenvalues. Since $\epsilon > 0$, the operator $\hat{M} + \epsilon$ is positively defined. It has an inverse operator which can be written through the ordinary bra and ket notation of eigenfunctions as

$$\frac{1}{\hat{M} + \epsilon} = \frac{|f_0\rangle\langle f_0|}{\epsilon} + \sum_k \frac{|f_k\rangle\langle f_k|}{1 + \epsilon + k^2}, \quad (22)$$

where the summation is extended over the continuous spectrum. Since $\langle \sinh \xi / \cosh^2 \xi, f_0 \rangle = 0$, the formal solution of Eq. (21) does not contain a term with $1/\epsilon$. The solution $l_y(\xi)$ is determined by the summation over the states of the continuous spectrum f_k only. After simple calculations the Euclidean action as a function of the trial parameter $\bar{\omega}$ takes the form

$$\mathcal{A}_E(\bar{\omega}) = \frac{\hbar^2 N}{4Jz} \left[2 \left(\frac{\omega_u^2}{\bar{\omega}} + \bar{\omega} \right) - \frac{\pi(\gamma H)^2 \omega_p^2}{2\bar{\omega}^3} \int_{-\infty}^{+\infty} \frac{dk}{(1 + k^2 + \omega_p^2/\bar{\omega}^2) \cosh^2(\pi k/2)} \right]. \quad (23)$$

If $\omega_p = 0$, the minimum of the action (23) is reached at $\bar{\omega} = \omega_u$ and we return to Eq. (15) again. Thus, in agreement with the previous analysis the Euclidean action depends on the field for $\omega_p \neq 0$ only. If the ratio ω_p/ω_u is small, this dependence is weak, and for the case of the field directed along the easy axis the Euclidean action is

$$\mathcal{A}_E(EA) = 2\hbar NS \sqrt{\frac{B_u}{Jz}} \left[1 - \frac{\pi(\gamma H)^2 \omega_p^2}{2\omega_u^4} + \dots \right]. \quad (24)$$

Thus, for $\omega_p \ll \omega_u$ the dependence of the Euclidean action \mathcal{A}_E on the field is weaker than it can be obtained from the naive consideration (see Fig. 1). The problem also can be solved in the limit case $\omega_p \gg \omega_u$, when the approximate solution of Eq. (21) can be written as

$$l_y \simeq \frac{2i\gamma H \bar{\omega} \sinh \xi}{\omega_p^2 \cosh^2 \xi} \ll 1. \quad (25)$$

The field dependence of the Euclidean action can be evaluated as $\mathcal{A}_E \sim B_u(H) = \sqrt{1 - (H/H_u)^2}$. Thus, the naive consideration of the magnetic field through the renormalization of the anisotropy is recovered only in the limit case of high planar anisotropy $\omega_p/\omega_u \rightarrow \infty$.

B. Field perpendicular to the easy axis

The case of the field directed along the hard axis is the simplest one. The plane xz remains preferable for the rotation

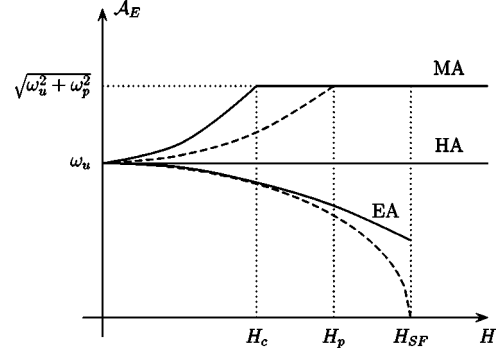


FIG. 1. The real part of the Euclidean action in units of $\hbar^2 N/2Jz$ as a function of the magnetic field (schematically). Data for the field directed along the easy, medium, and hard axes are marked as EA, MA, and HA, respectively, close to the appropriate curves. Solid lines represent the calculations of \mathcal{A}_E on the base of the full system (10), dashed lines are results of the naive consideration.

of the vector l for all values of the field. The right-hand sides of the system (10) are proportional to $\sin^2 \theta \sin \phi$, and the exact solution of the system (10) is $\phi = \phi_0 = \pi k$, $\bar{\omega} = \omega_u$. It corresponds to the rotation in the most preferable plane xz . The real part of the Euclidean action is independent of the field, but the imaginary parts have opposite signs for the two equivalent instanton trajectories with $\phi = 0$ and $\phi = \pi$,

$$\mathcal{A}_E(HA) = 2\hbar NS \sqrt{\frac{B_u}{Jz}} \pm i \frac{\pi \hbar g \mu_B H N}{2Jz}. \quad (26)$$

The nonzero imaginary part of the action (26) leads to interference of the instanton trajectories and an oscillating dependence of the transition probability on the field.^{12,13} In this geometry the most interesting effect arises due to the fluctuation determinant.^{13,15} The equations for small fluctuations ϑ and μ around the instanton solution are uncoupled again, but a complex-valued potential appears in the equation for ϑ . The eigenvalues and the fluctuation determinant are also complex. Thus, the presence of D_θ changes not only the tunneling amplitude for the instanton solution with a given value of σ , but also a phase shift for two instanton paths with $\sigma = \pm 1$. At low fields D_θ is almost real. It creates a decrease of the tunnel amplitude with the growth of the field. When the field increases, the factor D_θ produces the phase shift of oscillations of $\Delta(H)$ caused by the interference. The authors of Ref. 15 suggest that such a complicated behavior of $\Delta(H)$ caused by the fluctuation determinant D_θ and the interference of instanton trajectories with $\text{Im} \mathcal{A}_E \neq 0$ can be a universal feature for a wide class of antiferromagnets. As we saw earlier, for the field directed along the easy axis the dependence $\Delta(H)$ is strongly different from the scheme proposed by the authors of Ref. 15. For the field directed along the medium axis the behavior is also essentially different.

The case of the field parallel to the medium axis is the most complicated one, but it is interesting. The right-hand sides of Eqs. (10) are proportional to $\sin^2 \theta \cos \phi$. One type of exact solutions with the rotation of l in the plane xy containing the hard axis y (a planar solution) can be written as

$\phi = \phi_0 = \pi(k + 1/2)$ and $\cos \theta = \tanh(\bar{\omega}\tau)$ with $\bar{\omega} = \sqrt{\omega_u^2 + \omega_p^2}$. For this planar solution the structure of the Euclidean action $\mathcal{A}_E^{(p)}(MA)$ is the same as for the case of the field directed along the hard axis. Particularly, the real part of $\mathcal{A}_E^{(p)}(MA)$ does not depend on the field and the imaginary part is proportional to the field

$$\mathcal{A}_E^{(p)}(MA) = 2\hbar NS \sqrt{\frac{B_u + B_p}{Jz}} \pm i \frac{\pi \hbar g \mu_B H N}{2Jz}. \quad (27)$$

For such instanton trajectories the fluctuation determinant is similar to the factor obtained for the case of the field directed along the hard axis. It contains an imaginary part which leads to an extra contribution to the interference effects. But in contrast to the case of the field directed along the hard axis here the plane $\phi = \pi(k + 1/2)$ is not the plane passing through the medium axis. The solution with $\phi = \phi_0 = \pi(k + 1/2)$ and $\bar{\omega} = \sqrt{\omega_u^2 + \omega_p^2}$ satisfies the system (10) exactly, but the real part of the action is not minimal, at least at small magnetic fields $H < H_p$. In order to explain this fact it is sufficient to consider the value $H = 0$, when we have an exact solution with the rotation in the plane zx $\phi = \pi k$ and a smaller real part of the action. Thus, only for high fields the planar solution can be relevant. For the naive consideration of the field the real part of the Euclidean action for the solution with $\phi = \pi k$ and $\theta = \theta(\tau)$ has to be proportional to $\sqrt{\omega_u^2 + (\gamma H)^2}$, and $\text{Re } \mathcal{A}_E^{(p)}(MA)$ is equal to $\text{Re } \mathcal{A}_E^{(p)}(HA)$ at the point $H = H_p$. But as we will see below, the situation is actually more complicated.

Due to nonzero gyroscopic terms in the system (10) an exact solution with $\phi = \pi k$ does not exist for the case $H_x \neq 0$ and an appropriate approximate instanton solution is nonplanar. Following the treatment of the preceding section, we write the solution as $\cos \theta = \tanh(\bar{\omega}\tau)$, $l_y = \sin \phi \sin \theta \ll 1$ and determine l_y from the linear equation

$$(\hat{M} + \epsilon)l_y(\xi) = \frac{2i\gamma H}{\bar{\omega}} \frac{1}{\cosh^2 \xi}, \quad (28)$$

where $\epsilon = [\omega_p^2 - (\gamma H)^2]/\bar{\omega}^2$, $\xi = \bar{\omega}\tau$, and \hat{M} is defined by Eq. (18). In contrast to the similar equation (21) the right-hand side of Eq. (28) is symmetric with respect to ξ and a contribution from the localized eigenfunction (19a) is present. This contribution is proportional to $1/\epsilon$ and is mostly important for the case γH , $\omega_p \ll \omega_u$. As we will see below, such an instanton is important at low field and the abovementioned restriction is irrelevant. The Euclidean action for this nonplanar instanton as a function of $\bar{\omega}$ at $\omega_p < \omega_u$ can be written as

$$\mathcal{A}_E(\bar{\omega}) = \frac{\hbar^2 N}{4Jz} \left[2 \frac{\omega_u^2 + \gamma^2 H^2}{\bar{\omega}} + 2\bar{\omega} + \frac{(\pi\gamma H)^2 \bar{\omega}}{2(\omega_p^2 - \gamma^2 H^2)} \right] + \Delta \mathcal{A}_E, \quad (29)$$

where $\Delta \mathcal{A}_E$ is a contribution from the continuous spectrum that is determined through an integral over k with a structure that is similar to Eq. (23). This term contains the factor $(\gamma H/\bar{\omega})^2$ and it can be omitted for the case of interest $\gamma H \sim \omega_p \ll \bar{\omega}$. After minimizing over $\bar{\omega}$ the action for a nonplanar instanton for the case $\omega_p < \omega_u$ takes the form

$$\mathcal{A}_E^{(NP)}(MA) = 2\hbar NS \sqrt{\frac{B_u(1 + H^2/H_u^2)}{Jz}} \sqrt{1 + \frac{\eta H^2}{H_p^2 - H^2}}, \quad (30)$$

where numerical constant $\eta = \pi^2/4$. The problem can be solved in the opposite limit case, namely, at $\omega_u < \omega_p$. For this case one can replace the operator $\hat{M} + \epsilon$ by ϵ , and the approximate solution reads $l_y(\xi) = 2i\gamma H/(\epsilon \bar{\omega} \cosh^2 \xi)$. Then, for the Euclidean action we arrive to the same equation (30), but with another numerical constant $\eta = 8/3 \approx 2.667$. Comparing this value found for $\omega_u < \omega_p$ with $\pi^2/4 \approx 2.467$ for $\omega_p < \omega_u$, we can tell that for the two opposite limit cases the Euclidean action $\mathcal{A}_E^{(NP)}(MA)$ is approximately described by the same equation. Thus, we can suggest the Eq. (30) is a good approximation for any relation between ω_p and ω_u , that is in line with numerical data, see Sec. III.

Thus, when the field is parallel to the medium axis, the Euclidean action is real for a nonplanar instanton, and due to Eq. (30) it increases faster than it can be expected from the static renormalization of the anisotropy constant. It is possible to show that both fluctuation determinants D_θ and D_ϕ are also real. The interference effects are absent, and the tunnel splitting monotonically decreases with the growth of the field at small $H < H_c$, where H_c is a critical field, at which the values of the Euclidean action for the nonplanar instanton becomes equal to the real part of $\mathcal{A}_E^{(p)}(MA)$ for planar instantons. For small anisotropy in the basal plane, $\omega_p \ll \omega_u$, the value of H_c is small, and

$$H_c = H_p \frac{B_p}{B_p + (\pi/2)^2 B_u} \ll H_p. \quad (31)$$

But it is smaller that H_p even in the opposite limit case $\omega_u \ll \omega_p$,

$$H_c = H_p \sqrt{\frac{1}{1 + \sqrt{\eta}}} \approx 0.6163 H_p. \quad (32)$$

Thus, at low magnetic fields $H < H_c$ the Euclidean action for the nonplanar instanton solution $\mathcal{A}_E^{(NP)}(MA)$ is lower than for the planar solution. Its value reaches $\mathcal{A}_E^{(p)}(MA)$ at $H = H_c$, and the scenario of tunneling is changed to the planar one, common to that is present for the field along the hard axis with the tunneling exponent independent of the magnetic field and with interference effects caused by imaginary parts of both \mathcal{A}_E and fluctuation determinant.

III. NUMERICAL DATA

The semiclassical analysis of the coherent quantum tunneling between the classically degenerated ground states demonstrates that the level splitting is highly sensitive to the orientation of the magnetic field. Among the considered field orientations along the axes of rhombic symmetry the cases of the easy and medium axis are the most interesting ones. In both cases the Euclidean action has a zero imaginary part, and the corresponding interference effects are absent. We also have shown that for these two cases the preexponential

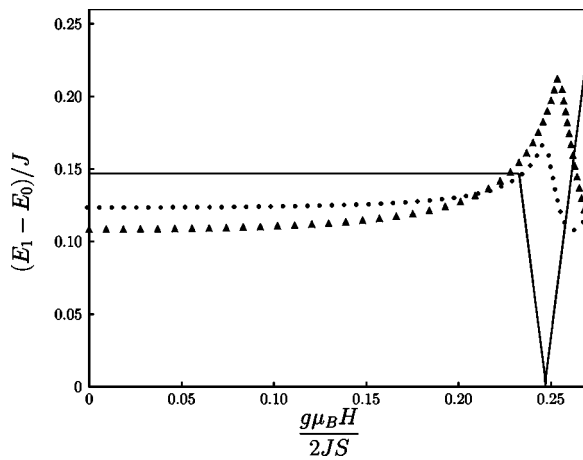


FIG. 2. Splitting of the lowest level for the quantum model with Hamiltonian (1), spin $S=5$, and $B_u/J=0.1$ for three values of the ratio $B_p/B_u=0.0$ (solid lines), 0.2 (circles), 0.4 (triangles). The magnetic field is directed along the easy axis and normalized to the exchange field $H_{ex}=JzSN/(g\mu_B)$ with $z=1$ and $N=2$. The cusps on the curves correspond to the change of the ground state that is a quantum counterpart of the spin-flop transition.

factor, which could be a source of interference, is real. Thus, for these orientations of the magnetic field interference effects does not appear, and the level splitting is mainly determined by the dependence of the real part of the Euclidean action on the magnetic field H . The character of this function is determined by the anisotropy B_p in the basal plane. The dependence is absent for the case $B_p=0$ and the field parallel to the easy axis only. The exponential factor $\exp(-\mathcal{A}_E/\hbar)$ is an increasing function of H for the field parallel to easy axis, and a decreasing function of H for the field parallel to the medium axis. This behavior strongly differs from that is present for the field directed parallel to the hard axis.

In order to check the semiclassical results found in assumption of some inequalities such as $B_p \ll B_u$ or $B_u \ll B_p$ and to estimate the role of the fluctuation determinant, which was not investigated here, we diagonalize numerically³⁰ the Hamiltonian (1) for the two-spin quantum model with high enough values of the spin (up to $S=100$), a small uniaxial anisotropy $B_u/J=0.01-0.1$ that guarantees reasonable values of the level splitting, and for several values of B_p/B_u , see figures below in this section. The numerical results show a satisfactory agreement with the instanton approximation even for moderate values of the spin $S=10-20$ even without taking into account the preexponential factor. But some discrepancies, which cannot be attributed to the approximations used in the analytical consideration, are also seen.

For the case of the field directed along the easy axis at $B_p=0$ the tunnel splitting is independent of the field up to the point of the spin-flop transition. This behavior is exactly reproduced by means of a numerical diagonalization, see Fig. 2. Due to the results of the instanton approach, for $B_p \neq 0$ the Euclidean action decreases, and the level splitting increases with the growth of the field H . The effect becomes more pronounced at large B_p . This behavior well corresponds to the numerical data represented in Fig. 2. The only difference

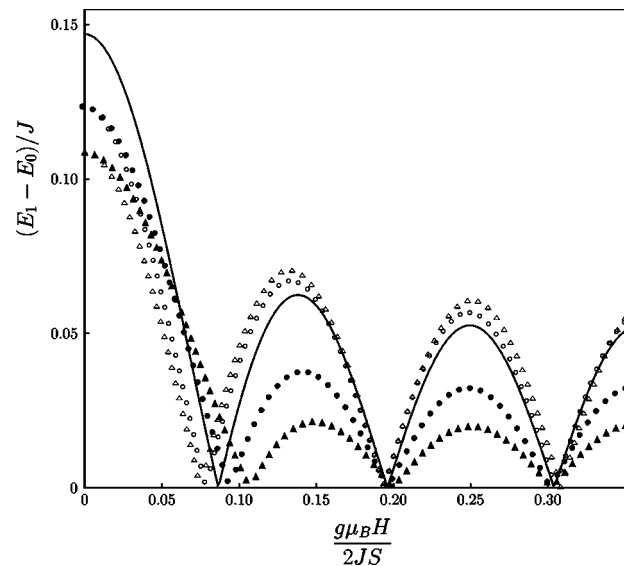


FIG. 3. Splitting of the lowest level for the quantum model with Hamiltonian (1), spin $S=5$, and $B_u/J=0.1$ for three values of the ratio $B_p/B_u=0.0$ (solid lines), 0.2 (circles), 0.4 (triangles), and the magnetic field directed along the hard axis (open symbols) or medium axis (full symbols). The field is normalized to the exchange value.

between the numerical data for the level splitting E_1-E_0 and the semiclassical results is that the field of the spin-flop transition identified as a field of the cusp on curves in Fig. 2 slowly depends on the ratio B_p/B_u for the quantum model. This dependence is completely absent in the semiclassical approximation, in particular, in the instanton approach. Probably, close to the point of the spin-flop transition, where the sharp decrease of the Euclidean action is present, the value of \mathcal{A}_E/\hbar becomes comparable with unity even for large S , and the quantum fluctuations treated beyond the semiclassical approximation become important.

In the case of the field directed along the hard axis as well as in the case of high fields, $H > H_c$, along the medium axis the instanton approach predicts that the tunneling occurs through the planar instanton paths. For these cases the level splitting $\Delta(H)$ oscillates as a function of the field with a constant period. If the field is directed along the hard axis, the behavior of $\Delta(H)$ coincides with the results of Refs. 13 and 15. The amplitude of oscillations is determined by the preexponential factor only, and it weakly depends on B_p . This kind of behavior is clearly seen in Fig. 3. In the case of high fields, $H > H_c$, directed along the medium axis the tunneling is also determined by planar instantons with a nonzero imaginary part of the action and interference effects appear. But when the ratio B_p/B_u increases, the main exponential factor drastically decreases, and the amplitude of oscillations $\Delta(H)$ decreases also. This feature is in a good agreement with the numerical calculations depicted in Fig. 3.

At low fields, $H < H_c$, parallel to the medium axis the tunneling is determined by nonplanar instantons. The real part of the Euclidean action $\text{Re}\mathcal{A}_E^{(NP)}(MA)$ monotonically increases, and the instanton approach predicts a strong monotonic decrease of $\Delta(H)$ up to $H=H_c$. For any values of B_p/B_u

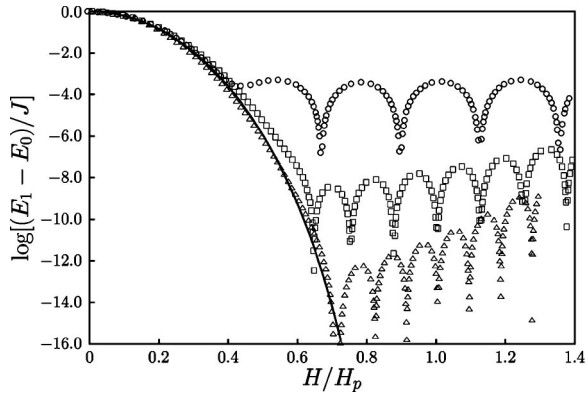


FIG. 4. Numerical data for the logarithm of the level splitting normalized by the value at $H=0$ versus the magnetic field directed along the medium axis for the spin $S=10$ and $B_u/J=0.1$ and three values of the anisotropy constant in the basal plane: $B_p/J=0.05$ (circles), 0.15 (squares), 0.25 (triangles). The magnetic field is normalized to the reorientation field H_p . The solid line describes the theoretical dependence (30) for a nonplanar instanton.

at $H < H_c$ the instanton approach also predicts that oscillations caused by interference are absent. The effect of the axes reorientation in the basal plane at $H=H_p$ does not clearly manifest itself as the spin-flop transition in Fig. 2, but it can be understood as a shift of a point, where oscillations start, to the region of high fields. Both factors, as well as the growth of the characteristic field of transition to the high-field tunneling picture H_c , are in a qualitative agreement with data of numerical calculations presented in Fig. 3.

In order to give a more detail comparison of the analytical and numerical data for small fields, $H < H_c$, parallel to the medium axis, we investigated numerically the value of spin $S=10$, for which the role of the field dependence of the fluctuation determinant is expected to be less important. The data together with the simple theoretical estimate of the level splitting, see Eq. (30), in the form $\Delta(H)/\Delta(0) = \exp\{-[\mathcal{A}(H) - \mathcal{A}(0)]/\hbar\}$, where $\mathcal{A}(H)$ is the value of the Euclidean action for nonplanar instantons, are present in Fig. 4. Here we can say about at least a semiquantitative agreement.

In order to check quantitatively the analytical expressions (24) and (30) for the real part of the Euclidean action, we propose the method of extracting the value of \mathcal{A}_E from the level splitting data without using an explicit expression for the preexponential factor. This method is based on the general theoretical formula for the level splitting

$$\Delta = DS^\beta \exp(-SA), \quad (33)$$

where D , β , and A are functions independent of the spin with normalized values of the magnetic field H/H_p or H/H_u . In the semiclassical approximation the function A means the real part of the action per unit of spin, D is the preexponential factor and β is associated with the number of zero modes: $\beta=2$ for the case of the purely uniaxial symmetry, that is $B_p=0$ and the magnetic field parallel to the easy axis,

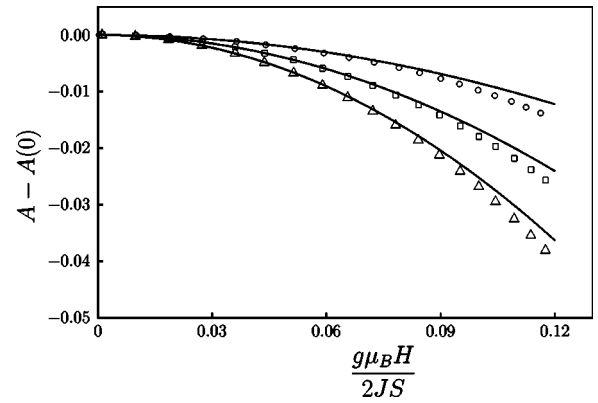


FIG. 5. Real part of the Euclidean action per unit spin obtained from the quantum model with Hamiltonian (1), $B_u/J=0.1$ for three values of the ratio $B_p/J=0.02$ (circles), 0.04 (squares), 0.08 (triangles), and the magnetic field directed along the easy axis. Solid lines are analytical predictions, Eq. (24). The field is normalized to the exchange value.

see also Eq. (20) and discussion there; $\beta=3/2$ for all rest models with biaxial symmetry.

The parameter β is also chosen floating in order to absorb corrections to the σ model and its semiclassical treatment. This method could, in principle, be applied for any semiclassical problem, but it is mostly useful for problems, where the imaginary part of the action, as well as the preexponential factor, are zero, and interference effects with oscillations are absent. For the tunneling in the external field this is just the problem we are interested in. This condition is realized at $H < H_u$ for the field directed along the easy axis and $H < H_p$ for the case of the field parallel to the medium one.

In order to obtain the values of interest, a numerical calculation of splitting of the lowest level is performed for few (at least, three) large values of spin. The real part of the action can be easily extracted from the numerical data such as qualitatively presented in Figs. 5 and 6. The functions $\mathcal{A}_E(H)$ in these figures are obtained by fitting the numerical data for the spin $10 < S < 20$ and each value of the field using Eq. (33).

For the simplest uniaxial case $B_p=0$ and the field directed along the easy axis we found that the expected value $\beta=2$ appears, and the Euclidean action is independent of the field up to the point of the spin-flop transition. Then, with growth of B_p we found a very sharp transition to the regime with one zero mode, for which the value of $\beta=3/2$ is reproduced. Thus, checking that β does not differs significantly from $3/2$ (the range 1.5 ± 0.1 is taken) side by side with more trivial conditions that the obtained values of D , β , and A are stable with respect to varying S and the exponent SA is large enough, we select sets of the parameters for the quantum model (1).

The action A obtained by fitting of Eq. (33) with the fixed value $\beta=1.5$ is plotted as functions $A(H)$ in Figs. 5 and 6 for the fields directed along the easy and medium axes. Note that the action $A(0)$ at zero field is subtracted from the functions $A(H)$. The analytical theory gives the value $A(0)=4\sqrt{B_u/J}$ independent on B_p , but numerical calculations demonstrate a

weak dependence of $A(0)$ on B_p . In details, $A_{th}(0)=1.26$ for $B_u/J=0.1$ and $A_{num}(0)$ is 1.22, 1.19 and 1.14 for the same value $B_u/J=0.1$ and B_p/J equal to 0.05, 0.10, 0.20, respectively.

The only way to describe the numerical data is to consider the fields H_u and H_p in Eqs. (24) and (30) as phenomenological parameters that predicted by the classical expressions (8) and (31). For simplicity, we rescale the fields in Fig. 6 as $H \rightarrow H/H_p$. The numerical data are fitted by Eqs. (24) and (30) using these fields in the range $H < 0.1H_u$ and $H < 0.2H_p$. Obtained values for a trial parameter $\tilde{H}_p/H_p = 1.164, 1.144, 1.145$ for $B_p/J=0.05, 0.10, 0.20$ are in a good agreement with the classical result, where obviously $\tilde{H}_p = H_p$. Appropriate analytical curves are plotted as solid lines in Figs. 5 and 6. In the case of the medium axis the theory predicts the shape of the curves obtained from the quantum model with a good accuracy up to fields close to H_c . For the easy axis the action decreases more significantly that it would be expected from the perturbative treatment of the semiclassical model. In both cases we can pretend on the quantitative agreement of the proposed theory and numerical data.

It is important to note one more discrepancy between the developed analytical theory and the presented numerical data. The analytical expression for the level splitting in the field directed along the hard axis does not contain any dependency on B_p , but in Fig. 4, as well as in the numerical data of Refs. 13 and 15, this dependency is present. It is more important for higher values of B_p/B_u . To explain it, as well as the observed dependence of the parameters H_p and H_u , we note that the σ model treats antiferromagnets in the first approximation over small ratios of the anisotropy constants or the magnetic field to the exchange integral J . Here, the values of these ratios was taken in the range 0.1–0.2, and the deviation of the σ model results that is of order of 10% from the numerical calculations are not surprising.

IV. CONCLUDING REMARKS

In conclusion, the antiferromagnetic particles can show a reach variety of tunneling behaviors that depend on the direction of the magnetic field. In addition to the oscillation behavior for the field directed along the hard axis,^{13,15} we found the growth of the tunnel splitting Δ for the field directed parallel to the easy axis and a steep decrease of Δ for the field along the medium axis. Both mentioned behaviors are connected to the tunnel exponent dependence on the field. It is important to note that such effects cannot be directly associated with the decrease or increase of the tunnel barrier, respectively, that governed by the static renormalization of the anisotropy energy.

Let us briefly discuss the possibility for experimental investigations of the tunneling effects predicted in the paper. The main point consists in kinds of antiferromagnets that could be used for experiments. The traditional antiferromagnetic samples such as small ferritin particles have unpaired spins and behave as noncompensated antiferromagnets. For this reason the destructive interference for them is mainly

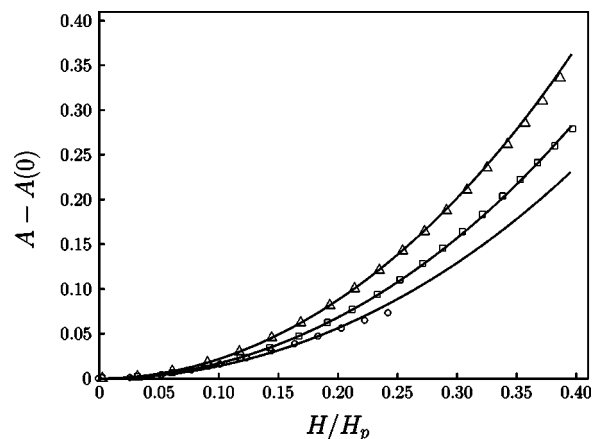


FIG. 6. Real part of the Euclidean action per unit spin obtained from the quantum model with Hamiltonian (1), $B_u/J=0.1$ for three values of the ratio $B_p/J=0.05$ (circles), 0.1 (squares), 0.2 (triangles), and the magnetic field directed along the medium axis. The magnetic field is normalized to H_p for each value of B_p . Solid lines are analytical predictions, Eq. (30), with H_p as a trial parameter.

dictated by the excess spin in the way common to ferromagnets.^{16,17} Moreover, the presence of a nonzero total magnetic moment drastically changes the structure of the ground state. It is enough to say that the degeneracy is absent except some fixed directions of the field with respect to the crystalline axis.^{12,18} We proposed a way to overcome this problem,¹⁹ but limitations caused by noncompensated spins seams to be more serious. Note that the same problem appears for ferromagnetic particles where the effects of the barrier reduction²⁰ and the oscillation behavior of the ground-state tunnel splitting^{21,22} was predicted many years ago, but observed only recently.²³

The key point in this important experimental success is based on the synthesis of high-spin molecules packed in the well-oriented monocrystals. Up to our understanding, the first possibility to investigate purely antiferromagnetic features is to use high-spin molecules with a well-defined spin structure. The molecules with ferromagnetic and antiferromagnetic couplings, uniaxial and rhombic anisotropies have been synthesized in the recent years.²⁴ For known ferromagnetic molecules such as Fe_8 the splitting is small, but the technique developed by Wernsdorfer and Sessoli²³ allows one to measure a very small tunnel splitting of order of 10^{-8} K. The first possibility discussed by many authors consists in using spin rings with antiferromagnetic coupling. For well-known antiferromagnetic molecular magnets such as Fe_{10} , Fe_6 , V_8 the problem is opposite to that for ferromagnetic molecules: the anisotropy is too small, and the barrier is too low to see clear semiclassical effects such as MQT.^{25,26} On the other hand, antiferromagnetic rings of eight chromium ions with a high anisotropy have been recently synthesized.²⁷ One more possibility is to use spin dimers containing two coupled high-spin molecules (molecular magnets) with a ferromagnetic coupling inside the molecule and an antiferromagnetic intermolecular coupling. For instance, the observation of the well-structured dimers of high-

spin molecules Mn_4 (spin $S=9/2$) with the antiferromagnetic coupling between two Mn_4 molecules has been recently reported.²⁸ Quantum tunneling in the Mn_4 dimers was investigated experimentally.²⁹ Such dimers of high-spin molecules such as Fe_8 with the macroscopic spin $S=10$ and well pronounced rhombic anisotropy could be a good candidates for observation of the effects considered in our paper.³¹ It is worth to note also that the predicted possibility of enlarging the value of the Euclidean action (to suppress the tunneling)

by means of the magnetic field directed parallel to the medium axis can be useful for following investigations.

ACKNOWLEDGMENTS

The authors thank A. K. Kolezhuk for fruitful discussions and help, and H.-J. Mikeska for useful discussions. This work was supported by Volkswagen-Stiftung, Grant No. I/75895. The authors also thank Institute of Theoretical Physics (University of Hanover) for kind hospitality.

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³⁰The implicitly restarted Arnoldi method implemented in the ARPACK package of numerical routines is used for eigenvalue calculations of large scale sparse matrices.

³¹Surely, the exchange interaction between these two molecules is antiferromagnetic, as it takes place in the case of Mn_4 ; we were not able to find the corresponding data in the literature.