PHYSICAL REVIEW B **83**, 014405 (2011)

Acoustic Faraday effect and the circular magnetic dichroism effect in single molecule magnets

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(Received 9 June 2010; revised manuscript received 25 October 2010; published 10 January 2011)

We report a theoretical study on propagation of an acoustic wave in a field-cooled magnetized crystal of molecular magnets. A transverse wave propagating in the direction of crystal magnetization is considered along with the acoustic Faraday effect and the circular magnetic dichroism effect arising by such propagation. Numerical estimations of these effects are presented, demonstrating that they can easily be observed experimentally.

DOI: 10.1103/PhysRevB.83.014405 PACS number(s): 75.50.Xx, 72.55.+s

I. INTRODUCTION

Magnetoacoustic phenomena, specifically the interaction between the acoustic waves and paramagnetic atoms, have always attracted the attention of theorists and experimenters involved in magnetism research. A vivid manifestation of this interaction is a well-known acoustic Faraday effect (AFE) in paramagnetics that is essentially a nonreciprocal effect of rotation of the polarization plane of a transverse acoustic wave propagating in a paramagnetic parallel to the magnetic field (see, for example, Ref. 1 and references therein). The AFE was for the first time theoretically considered for ferromagnetic substances[2] and for paramagnetics.[3] In dissipative media the AFE occurs along with the circular magnetic dichroism effect (MDE) that manifests itself through ellipticity of the acoustic wave polarization.

Of great research interest currently are magnetoacoustic phenomena in single molecule magnets (SMMs)[4–7] which are in fact paramagnetics.[8] A distinctive feature of SMM, which is also their advantage over regular paramagnetics, is that they can be prepared in a variety of magnetic states even in the absence of a magnetic field. Another distinction is that interaction of the SMM spin system with long-wave deformation induced by propagation of the transverse acoustic wave is independent of the magnetoelastic coupling constants (see Ref. 9 and references therein). These peculiarities of SMMs show up through magnetoacoustic effects. Therefore, we find theoretical consideration of AFE and MDE in SMMs interesting and important. This paper addresses these phenomena.

II. SMM IN A LONGITUDINAL CONSTANT MAGNETIC FIELD

We consider a simple model of SMM, making appropriate corrections where necessary. A simple model of SMM implies that a crystal of molecular magnets is composed of noninteracting magnetic molecules.[10–13] Each molecule is characterized by a strong crystal-field anisotropy (longitudinal anisotropy). For certainty, we denote this direction by the z axis. In addition, the molecule has a transverse anisotropy that can be both very weak compared to the longitudinal anisotropy (like that of the Mn_{12}-Ac and Ni_4 crystals) and comparable with it (as in Fe_8 crystal, for example).

We focus only on crystals with a weak transverse anisotropy. In the absence of magnetic fields and regardless of the transverse anisotropy, the Hamiltonian of a magnetic molecule reads

$$\widehat{H}_0 = -D\widehat{S}_z^2, \tag{1}$$

where D is the longitudinal anisotropy energy constant and $\widehat{S}_z$ is the operator of spin projection on the z axis. The energy spectrum of the molecule $E_m^{(0)} = -Dm^2$ $(m = S, S-1, \ldots, -S)$ is represented by $S + 1/2$ degenerate doublets if S is a half integer, and by S degenerate doublets and one nondegenerate level if S is an integer. The eigenfunctions ψ_m of operator $\widehat{S}_z$ are the eigenfunctions of the Hamiltonian (1). Thus, each level can be characterized by the magnetic quantum number m, which is the eigenvalue of operator $\widehat{S}_z$ $(\widehat{S}_z\psi_m = m\psi_m)$; it is seen that the energies of states ψ_m and ψ_{-m} are equal. Due to the relatively low temperatures, it would be sufficient to consider just two lower doublets of the magnetic molecule: the fundamental doublet $m = \pm S$, and the first excited doublet $m = \pm(S-1)$. In the following, the subscript 1 marks the fundamental doublet and the subscript 2 marks the first excited doublet (see Fig. 1).

If the applied dc magnetic field is directed along the z axis, the molecule Hamiltonian is

$$\widehat{H}_{\mathrm{mag}} = \widehat{H}_0 + g\mu_B H_{0z}\widehat{S}_z, \tag{2}$$

where g and μ_B are the Lande factor and the Bohr magneton, respectively.

Since operator $\widehat{S}_z$ commutes with Hamiltonian (2) (as it does with $\widehat{H}_0$), they have common eigenfunctions. It is easily seen that, as in the absence of the magnetic field, functions ψ_m are eigenfunctions of Hamiltonian (2), only now states ψ_m and ψ_{-m} correspond to different energies. According to Eq. (2), the energy of the state with the specified value of m is $E_m = -Dm^2 + g\mu_B H_{0z}m$, so, the value of splitting for each doublet is equal to $E_m - E_{-m} = 2g\mu_B H_{0z}m$. For each split doublet, the energy of the state with a positive m is higher than the energy of the state with a negative m (for definiteness, we set $H_{0z} > 0$, see Fig. 2).

In the absence of the magnetic field, the frequency of transition $S - 1 \rightarrow S$ is equal to that of transition

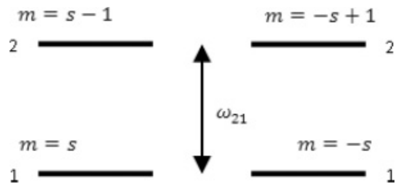

FIG. 1. Two lower doublets of a magnetic molecule.

$-S+1 \to -S$. We denote this frequency by ω_{21} (see Fig. 1):

$$\omega_{21} = \frac{E_{S-1}^{(0)} - E_S^{(0)}}{\hbar} = D\frac{2S-1}{\hbar}. \quad (3)$$

Under the dc magnetic field H_{0z}, the molecule spectrum changes: the state with $m = \pm S$ corresponds to the energy $E_{\pm S} = -DS^2 \pm g\mu_B H_{0z} S$, and the state with $m = \pm(S-1)$ corresponds to the energy $E_{\pm(S-1)} = -D(S-1)^2 \pm g\mu_B H_{0z}(S-1)$ (see Fig. 2). As a result, the frequencies of transitions between the states of the fundamental and the first excited doublets depend on sign m:

$$\omega^+ = \omega_{21} - \omega_H,$$
$$\omega^- = \omega_{21} + \omega_H, \quad (4)$$
$$\omega_H = \frac{g\mu_B H_{0z}}{\hbar}.$$

Here and following, the superscripts $(+,-)$ mark the quantities relating to the molecule states $m > 0$ and $m < 0$, respectively.

We study the case of relatively low temperatures:

$$k_B T \ll \hbar\omega^+, \hbar\omega^-. \quad (5)$$

So, under thermodynamic equilibrium, only the fundamental doublet population is largely different from zero. Note that at low temperatures the probability of thermoactivated processes which cause a change of sign of the spin projection is vanishingly small.

We also assume that the magnetic field H_{0z} is such that the energy differences $E_S - E_{-S}$, $E_{S-1} - E_{-S+1}$, and $E_{-S+1} - E_S$ are sufficient for neglecting the processes of quantum tunneling $S \rightleftarrows -S$, $S-1 \rightleftarrows -S+1$, and $-S+1 \rightleftarrows S$, respectively.

Thus, the entire ensemble of magnetic molecules may be considered as two practically independent subsystems in which the molecules differ by sign m. The respective molecule concentrations in these subsystems are designated as N^+ and N^-. In consequence of Eq. (5), in the state of thermodynamic equilibrium, N^+ is the concentration of molecules in the state ψ_S and N^- is the concentration of molecules in the state ψ_{-S}.

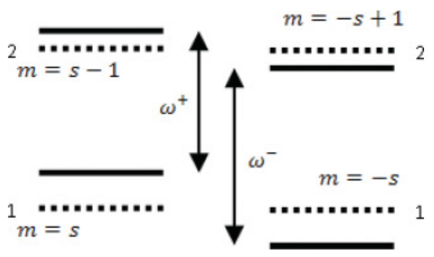

FIG. 2. Magnetic molecule spectrum (two lower doublets) in the presence of a dc magnetic field $H_{0z} > 0$ directed along the z axis. The dotted lines correspond to the nonperturbed molecule spectrum. The molecule spectrum in the presence of the dc magnetic field H_{0z} is shown by the solid lines.

The values N^+ and N^- depend on the specific mode in which the sample cooling and the H_{0z} field application were carried out. In this paper we consider the AFE and MDE when SMM is entirely polarized ($N^+ = 0$, $N^- = N$; i.e., N is the total concentration of magnetic molecules in SMMs). It is in a fully polarized (magnetized to saturation) sample that the AFE and MDE in question are most strongly manifested. This state can be obtained, for example, through field-cooled (FC) magnetization (see, for example, Refs. 11 and 14). If we then set an acceptable (fairly small) value for H_{0z}, the entirely polarized state with $N^+ = 0$, $N^- = N$ will be retained long enough; that is, all of the magnetic molecules will belong to the subsystem with $m < 0$.

Note here that to theoretically estimate an acceptable value of H_{0z} we have to extend the limits of the simple SMM model, in particular, by including the dipole interaction between the molecular spins and the interaction between the electronic and nuclear spins (hyperfine interaction).[11,15] One can just as well use the experimental data for estimations. In Ref. 14, for example, it is demonstrated that the SMM state (for Mn_{12}-Ac) resulting from FC magnetization is quite stable at $T < 1$ K and $H_{0z} \lesssim 10^3$ Θe (the magnetization value does not vary for hours; see also Refs. 15 and 16). So, since we consider Mn_{12}-Ac, we assume $T < 1$ K and $H_{0z} \lesssim 10^3$ Θe (which corresponds to $\omega_H/2\pi \lesssim 3 \times 10^9\ \text{s}^{-1}$).

Investigation of the acoustic wave propagation along the z axis is carried out in the continuous medium approximation. Therefore, we introduce (via Heisenberg representation) the operators of spin density $\widehat{\vec{S}}^-(z;t)$, relating to the subsystem with $m < 0$. These operators satisfy the well-known commutation relations expressed here in the traditional notations:

$$[\widehat{S}_i^-(z;t)\widehat{S}_j^-(z';t)] = i\varepsilon_{ijk}\widehat{S}_k^-\delta(\vec{r} - \vec{r}\,'), \quad (6)$$

where $\vec{r} = x\vec{e}_x + y\vec{e}_y + z\vec{e}_z$. Here and elsewhere, $\vec{e}_{x,y,z}$ are the unit vectors of the Cartesian system of coordinates.

Next, we use these operators to introduce, following Eqs. (1) and (2), the magnetic Hamiltonian density:

$$\widehat{\mathcal{H}}_{\text{mag}}(z;t) = -\frac{D}{N}(\widehat{S}_z^-(z;t))^2 + g\mu_B H_{0z}\widehat{S}_z^-(z;t). \quad (7)$$

III. INTERACTION OF THE ACOUSTIC WAVE WITH THE SMM SPIN SYSTEM IN THE ABSENCE OF DAMPING

In this section we regard an acoustic wave as a quantum object but solely for the simplicity of the applicable body of mathematics rather than fundamentally (see, for example, Ref. 2). Also, no relaxation (dissipative) processes that contribute to the acoustic wave damping are taken into account.

First, we examine an acoustic wave propagating without interaction with the magnetic system of the crystal. For a transverse wave propagating along the z axis, we have

$$\widehat{\vec{U}}(z;t) = \widehat{U}_x(z;t)\vec{e}_x + \widehat{U}_y(z;t)\vec{e}_y, \quad (8)$$

where $\widehat{\vec{U}}(z;t)$ is the operator of the displacement vector of a point in the medium from its original position. Then the elastic

Hamiltonian density has an ordinary form:

$$\widehat{\mathcal{H}}_{\mathrm{el}}(z;t) = \frac{\widehat{P}_x^2(z;t)}{2\rho} + \frac{\widehat{P}_y^2(z;t)}{2\rho} + \frac{K}{2}\left[\left(\frac{\partial \widehat{U}_x(z;t)}{\partial z}\right)^2 + \left(\frac{\partial \widehat{U}_y(z;t)}{\partial z}\right)^2\right], \tag{9}$$

where $\widehat{P}_{x,y}(z;t)$ is the operator of the momentum density, conjugate to operator $\widehat{U}_{x,y}(z;t)$; ρ is the density; and K is the elastic constant.

The commutation relations are

$$[\widehat{U}_i(z;t)\widehat{P}_j(z';t)] = i\hbar\delta_{ij}\delta(\vec{r} - \vec{r}\,'). \tag{10}$$

Note that, in the absence of interaction between the acoustic wave and the SMM spin system, the wave velocity $v_t = \sqrt{K/\rho}$.

We now consider the acoustic wave interaction with the SMM spin system, following the approaches developed in Ref. 9: this interaction is determined by the effect of the acoustic wave-induced rotation of each local volume of the crystal (and, hence, every magnetic molecule) as a whole. According to Ref. 9, the operator of such magnetoelastic interaction for a single molecule up to the linear terms in $\delta\widehat{\vec{\Phi}} = \nabla\widehat{\vec{U}}(z;t)/2$ (operator of the molecule anisotropy axis rotation angle) is

$$\widehat{H}_{\mathrm{mag,el}} = -D\{\widehat{S}_x^-\widehat{S}_z^-\}\delta\widehat{\Phi}_y + D\{\widehat{S}_y^-\widehat{S}_z^-\}\delta\widehat{\Phi}_x, \tag{11}$$

where the curly brackets designate an anticommutator. The matrix elements of operator $\widehat{H}_{\mathrm{mag,el}}$ differ from zero only for the transitions $m \rightleftarrows m \pm 1$.

For the acoustic transverse wave under study, $\delta\widehat{\Phi}_x = -\frac{1}{2}\frac{\partial \widehat{U}_y(z;t)}{\partial z}$, $\delta\widehat{\Phi}_y = \frac{1}{2}\frac{\partial \widehat{U}_x(z;t)}{\partial z}$; therefore, using Eq. (11), the density of the magnetoelastic Hamiltonian is expressed as

$$\widehat{\mathcal{H}}_{\mathrm{mag,el}}(z;t) = -\frac{D}{2N}\left(\{\widehat{S}_x^-(z;t)\widehat{S}_z^-(z;t)\}\frac{\partial \widehat{U}_x(z;t)}{\partial z} + \{\widehat{S}_y^-(z;t)\widehat{S}_z^-(z;t)\}\frac{\partial \widehat{U}_y(z;t)}{\partial z}\right). \tag{12}$$

In further treatment we use the following simplifying conditions. We deal with the case of relatively low temperatures (see Sec. II). So, taking into account the fact that only transitions $m \rightleftarrows m \pm 1$ differ from zero, it would be sufficient to include just a couple of states ψ_{-S} and ψ_{-S+1} for the subsystem with $m < 0$. We introduce pseudospin operators (in Schrödinger representation) for a single molecule from this subsystem, using $\varphi_1 = \psi_{-S}$, $\varphi_2 = \psi_{-S+1}$, as the basis:

$$\widehat{R}_{\mathrm{I}}^{-\mathrm{Sch}} = \frac{1}{2}\begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad \widehat{R}_{\mathrm{II}}^{-\mathrm{Sch}} = \frac{1}{2}\begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}, \quad \widehat{R}_{\mathrm{III}}^{-\mathrm{Sch}} = \frac{1}{2}\begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}. \tag{13}$$

Note that in the state of thermodynamic equilibrium at rather low temperatures only the level corresponding to $\varphi_1 = \psi_{-S}$ is populated substantially; therefore, we assume

$$\langle \widehat{R}_{\mathrm{III}}^{-\mathrm{Sch}} \rangle = \tfrac{1}{2}. \tag{14}$$

Here and elsewhere the angle brackets designate the thermodynamic average in the equilibrium state. Using Eqs. (13), it is possible to write the operators for a single molecule (in Schrödinger representation) in the following form: for $\widehat{H}_{\mathrm{mag}}$ [see Eq. (2)],

$$\widehat{H}_{\mathrm{mag}}^{\mathrm{Sch}} = -\hbar\omega^-\widehat{R}_{\mathrm{III}}^{-\mathrm{Sch}}, \tag{15}$$

and for the anticommutators in Eq. (11),

$$\left\{\widehat{S}_x^{-\mathrm{Sch}}\widehat{S}_z^{-\mathrm{Sch}}\right\} = -\sqrt{2S}(2S-1)\widehat{R}_{\mathrm{I}}^{-\mathrm{Sch}}, \tag{16}$$

$$\left\{\widehat{S}_y^{-\mathrm{Sch}}\widehat{S}_z^{-\mathrm{Sch}}\right\} = \sqrt{2S}(2S-1)\widehat{R}_{\mathrm{II}}^{-\mathrm{Sch}}. \tag{17}$$

Now, using Eqs. (15)–(17), we change the operators $\widehat{R}_i^{-\mathrm{Sch}}$ for the Heisenberg operators of pseudospin density $\widehat{R}_i^-(z;t)$ ($i = \mathrm{I, II, III}$); then the magnetic Hamiltonian density $\widehat{\mathcal{H}}_{\mathrm{mag}}(z;t)$ [Eq. (7)] can be written as

$$\widehat{\mathcal{H}}_{\mathrm{mag}}(z;t) = -\hbar\omega^-\widehat{R}_{\mathrm{III}}^-(z;t), \tag{18}$$

and the magnetoelastic Hamiltonian density $\widehat{\mathcal{H}}_{\mathrm{mag,el}}(z;t)$ [Eq. (12)] as

$$\widehat{\mathcal{H}}_{\mathrm{mag,el}}(z;t) = D\sqrt{\frac{S}{2}}(2S-1) \times \left(\widehat{R}_{\mathrm{I}}^-(z;t)\frac{\partial \widehat{U}_x(z;t)}{\partial z} - \widehat{R}_{\mathrm{II}}^-(z;t)\frac{\partial \widehat{U}_y(z;t)}{\partial z}\right). \tag{19}$$

The commutation relations for the operators $\widehat{R}_i^-(z;t)$ have the form

$$[\widehat{R}_i^-(z;t)\widehat{R}_j^-(z';t)] = i\varepsilon_{ijk}\widehat{R}_k^-(z;t)\delta(\vec{r} - \vec{r}\,'). \tag{20}$$

The total Hamiltonian density $\widehat{\mathcal{H}}_{\mathrm{tot}}$ is obtained by combining Eqs. (9), (18), and (19):

$$\widehat{\mathcal{H}}_{\mathrm{tot}}(z;t) = \widehat{\mathcal{H}}_{\mathrm{el}}(z;t) + \widehat{\mathcal{H}}_{\mathrm{mag}}(z;t) + \widehat{\mathcal{H}}_{\mathrm{mag,el}}(z;t). \tag{21}$$

Thus, the total Hamiltonian reads

$$\widehat{H}_{\mathrm{tot}} = \int \widehat{\mathcal{H}}_{\mathrm{tot}}(z;t)\,d\vec{r}. \tag{22}$$

Here the integral is taken over the crystal volume.

Using Eq. (22) and the commutation relations for Heisenberg operators, one can obtain the quantum equations of motion for these operators:

$$\dot{\widehat{U}}_{x(y)}(z;t) = \frac{i}{\hbar}[\widehat{H}_{\mathrm{tot}}\widehat{U}_{x(y)}(z;t)] = \frac{i}{\hbar}\int[\widehat{\mathcal{H}}_{\mathrm{el}}(z';t)\widehat{U}_{x(y)}(z;t)]d\vec{r}\,' = \frac{1}{\rho}\,\widehat{P}_{x(y)}(z;t), \tag{23}$$

$$\begin{aligned}\dot{\widehat{P}}_{x(y)}(z;t) &= \frac{i}{\hbar}[\widehat{H}_{\mathrm{tot}}\widehat{P}_{x(y)}(z;t)] \\ &= \frac{i}{\hbar}\int[(\widehat{\mathcal{H}}_{\mathrm{el}}(z';t) + \widehat{\mathcal{H}}_{\mathrm{mag,el}}(z';t))\widehat{P}_{x(y)}(z;t)]d\vec{r}\,' \\ &= K\frac{\partial^2 U_{x(y)}(z;t)}{\partial z^2} \underset{(-)}{+} D\sqrt{\frac{S}{2}}\,(2S-1)\frac{\partial \widehat{R}_{\mathrm{I(II)}}^-(z;t)}{\partial z},\end{aligned} \tag{24}$$

$$\begin{aligned}\dot{\widehat{R}}{}^{-}_{\mathrm{I(II)}}(z;t) &= \frac{i}{\hbar}[\widehat{H}_{\mathrm{tot}}\widehat{R}^{-}_{\mathrm{I(II)}}(z;t)] \\ &= \frac{i}{\hbar}\int[(\widehat{\mathcal{H}}_{\mathrm{mag}}(z';t)+\widehat{\mathcal{H}}_{\mathrm{mag,el}}(z';t)) \\ &\quad\times \widehat{R}^{-}_{\mathrm{I(II)}}(z;t)]\,d\vec{r}\,' \\ &= \underset{(-)}{+}\omega^{-}\widehat{R}^{-}_{\mathrm{II(I)}}(z;t) - D\sqrt{\frac{S}{2}}(2S-1) \\ &\quad\times\hbar^{-1}\widehat{R}^{-}_{\mathrm{III}}(z;t)\frac{\partial\widehat{U}_{y(x)}(z;t)}{\partial z},\end{aligned} \tag{25}$$

$$\begin{aligned}\dot{\widehat{R}}_{\mathrm{III}}3^{-}(z;t) &= \frac{i}{\hbar}[\widehat{H}_{\mathrm{tot}}\widehat{R}^{-}_{\mathrm{III}}(z;t)] \\ &= \frac{i}{\hbar}\int[\widehat{\mathcal{H}}_{\mathrm{mag,el}}(z';t)\widehat{R}^{-}_{\mathrm{III}}(z;t)]d\vec{r}\,' \\ &= D\sqrt{\frac{S}{2}}(2S-1)\hbar^{-1} \\ &\quad\times\left(\widehat{R}^{-}_{\mathrm{II}}(z;t)\frac{\partial\widehat{U}_{x}(z;t)}{\partial z}+\widehat{R}^{-}_{\mathrm{I}}(z;t)\frac{\partial\widehat{U}_{y}(z;t)}{\partial z}\right).\end{aligned} \tag{26}$$

It is seen from Eqs. (25) and (26) that $\dot{\widehat{R}}{}^{-}_{\mathrm{I(II)}}(z;t)$ are different from zero in the first order and $\dot{\widehat{R}}{}^{-}_{\mathrm{III}}(z;t)$ in the second order of interaction of the acoustic wave with the magnetic system of SMMs. Assuming this interaction weak, we may set $\dot{\widehat{R}}{}^{-}_{\mathrm{III}}(z;t)=0$; then Eqs. (23)–(26) yield the equations for the corresponding averages:

$$\rho\langle\ddot{\widehat{U}}_{x}(z;t)\rangle = K\frac{\partial^{2}\langle\widehat{U}_{x}(z;t)\rangle}{\partial z^{2}} + D\sqrt{\frac{S}{2}}(2S-1)\frac{\partial\langle\widehat{R}^{-}_{\mathrm{I}}(z;t)\rangle}{\partial z}, \tag{27}$$

$$\rho\langle\ddot{\widehat{U}}_{y}(z;t)\rangle = K\frac{\partial^{2}\langle\widehat{U}_{y}(z;t)\rangle}{\partial z^{2}} - D\sqrt{\frac{S}{2}}(2S-1)\frac{\partial\langle\widehat{R}^{-}_{\mathrm{II}}(z;t)\rangle}{\partial z}, \tag{28}$$

$$\begin{aligned}\langle\dot{\widehat{R}}{}^{-}_{\mathrm{I}}(z;t)\rangle &= \omega^{-}\langle\widehat{R}^{-}_{\mathrm{II}}(z;t)\rangle - D\sqrt{\frac{S}{2}}(2S-1)\hbar^{-1} \\ &\quad\times\left\langle\widehat{R}^{-}_{\mathrm{III}}(z;t)\frac{\partial\widehat{U}_{y}(z;t)}{\partial z}\right\rangle,\end{aligned} \tag{29}$$

$$\begin{aligned}\langle\dot{\widehat{R}}{}^{-}_{\mathrm{II}}(z;t)\rangle &= -\omega^{-}\langle\widehat{R}^{-}_{\mathrm{I}}(z;t)\rangle - D\sqrt{\frac{S}{2}}(2S-1)\hbar^{-1} \\ &\quad\times\left\langle\widehat{R}^{-}_{\mathrm{III}}(z;t)\frac{\partial\widehat{U}_{x}(z;t)}{\partial z}\right\rangle.\end{aligned} \tag{30}$$

In accordance with our approach, the averaging is performed via the density matrix which is independent of time in the Heisenberg representation. We assume this density matrix to correspond (at $t=0$) to the initial thermodynamic equilibrium state. Since $\dot{\widehat{R}}{}^{-}_{\mathrm{III}}(z;t)=0$, it is possible to assume in Eqs. (29) and (30) that

$$\widehat{R}^{-}_{\mathrm{III}}(z;t) = \widehat{R}^{-}_{\mathrm{III}}(z;0) = N\widehat{R}^{-\mathrm{Sch}}_{\mathrm{III}}. \tag{31}$$

Equality (31) implies that *in the first order* of interaction of the acoustic wave with the SMM magnetic system the populations of the magnetic molecule levels do not change and, hence, neither does the SMM magnetization projection on the z axis. So, the set of Eqs. (27)–(30) corresponds to small oscillations of magnetization near the equilibrium state.

Considering also Eq. (14), we can perform the following uncoupling in Eqs. (29) and (30):

$$\left\langle\widehat{R}^{-}_{\mathrm{III}}(z;t)\frac{\partial\widehat{U}_{x,y}(z;t)}{\partial z}\right\rangle = \frac{1}{2}N\frac{\partial\langle\widehat{U}_{x,y}(z;t)\rangle}{\partial z}. \tag{32}$$

Using Eq. (32) and neglecting, for simplicity, the symbols for the operators and the averaging, from Eqs. (27)–(30) we obtain

$$\rho\ddot{U}_{x}(z;t) = K\frac{\partial^{2}U_{x}(z;t)}{\partial z^{2}} + D\sqrt{\frac{S}{2}}(2S-1)\frac{\partial R^{-}_{\mathrm{I}}(z;t)}{\partial z}, \tag{33}$$

$$\rho\ddot{U}_{y}(z;t) = K\frac{\partial^{2}U_{y}(z;t)}{\partial z^{2}} - D\sqrt{\frac{S}{2}}(2S-1)\frac{\partial R^{-}_{\mathrm{II}}(z;t)}{\partial z}, \tag{34}$$

$$\dot{R}^{-}_{\mathrm{I}}(z;t) = \omega^{-}R^{-}_{\mathrm{II}}(z;t) - D\sqrt{\frac{S}{2}}(2S-1)(2\hbar)^{-1}N\frac{\partial U_{y}(z;t)}{\partial z}, \tag{35}$$

$$\dot{R}^{-}_{\mathrm{II}}(z;t) = -\omega^{-}R^{-}_{\mathrm{I}}(z;t) - D\sqrt{\frac{S}{2}}(2S-1)(2\hbar)^{-1}N\frac{\partial U_{x}(z;t)}{\partial z}. \tag{36}$$

It should be noted here that the description of the interaction between the SMM spin system and the acoustic wave, Eqs. (33)–(36), can be readily interpreted. Indeed, quantities $R^{-}_{\mathrm{I,II}}(z;t)$, N can be thought of as the components of some effective classical "magnetic moment" $\vec{M}_{\mathrm{ef}}$ with components ($R^{-}_{\mathrm{I}}\to(M_{\mathrm{ef}})_{x}$; $R^{-}_{\mathrm{II}}\to(M_{\mathrm{ef}})_{y}$; $N\to(M_{\mathrm{ef}})_{z}$). It is seen from Eqs. (35) and (36) that this effective classical "magnetic moment" moves in the effective "magnetic field" $\vec{H}_{\mathrm{ef}}$ with the components

$$\begin{gathered}(H_{\mathrm{ef}})_{x} = -D\sqrt{\frac{S}{2}}(2S-1)(2\hbar)^{-1}\frac{\partial U_{x}(z;t)}{\partial z}, \\ (H_{\mathrm{ef}})_{y} = D\sqrt{\frac{S}{2}}(2S-1)(2\hbar)^{-1}\frac{\partial U_{y}(z;t)}{\partial z}, \\ (H_{\mathrm{ef}})_{z} = \omega^{-},\end{gathered} \tag{37}$$

in accordance with the equation

$$\dot{\vec{M}}_{\mathrm{ef}} = [\vec{M}_{\mathrm{ef}}\times\vec{H}_{\mathrm{ef}}]. \tag{38}$$

This approach is valid under the condition

$$(M_{\mathrm{ef}})_{x,y} \ll (M_{\mathrm{ef}})_{z} = N = \mathrm{const}. \tag{39}$$

We would like to point out the obvious fact that the effective "gyromagnetic ratio" corresponding to such effective "moment" and "field" equals 1. Also note that ω^{-} is essentially the frequency of the uniform precessing of the effective "moment" (in the absence of the acoustic wave).

IV. ACOUSTIC WAVE IN SMM UNDER THE RELAXATION PROCESSES IN THE SMM SPIN SYSTEM: AFE AND MDE

We assume that the relaxation processes are relevant only in the SMM spin system and neglect them in the acoustic wave proper (see, for example, Ref. 2). These processes are included in the equations of motion for the "effective magnetic moment" via the well-known Landau-Lifshitz approach.[17,18] This approach, taking into account Eqs. (37)–(39), leads to the following changes in the equations of motion for $R_{\rm I,II}^-(z;t)$, Eqs. (35) and (36):

$$\begin{aligned}\dot{R}_{\rm I}^-(z;t) &= \omega^- R_{\rm II}^-(z;t) - D\sqrt{\frac{S}{2}}(2S-1)(2\hbar)^{-1}N\frac{\partial U_y(z;t)}{\partial z}\\ &\quad -\gamma\left(R_{\rm I}^-(z;t) + D\sqrt{\frac{S}{2}}(2S-1)(2\hbar\omega^-)^{-1}N\frac{\partial U_x(z;t)}{\partial z}\right),\end{aligned} \tag{40}$$

$$\begin{aligned}\dot{R}_{\rm II}^-(z;t) &= -\omega^- R_{\rm I}^-(z;t) - D\sqrt{\frac{S}{2}}(2S-1)(2\hbar)^{-1}N\frac{\partial U_x(z;t)}{\partial z}\\ &\quad -\gamma\left(R_{\rm II}^-(z;t) - D\sqrt{\frac{S}{2}}(2S-1)(2\hbar\omega^-)^{-1}N\frac{\partial U_y(z;t)}{\partial z}\right),\end{aligned} \tag{41}$$

where γ^{-1} is the characteristic time of the spin system relaxation for the transition at frequencies $\omega \sim \omega^- \simeq \omega_{21}$. Note that for Eq. (39) these equations can be derived within the approach involving a modified form of the Bloch equation[19,20] (assuming $\gamma^{-1} \equiv T_2$). The thing in common for both methods, which is an important assumption, too, is that the dissipation rate at each moment is proportional to the difference between the value of the "magnetic moment" and its magnitude that would set in if the momentary value of the effective "magnetic field" were "frozen."

In this way we obtain a set of equations (33), (34), (40), and (41). Assuming that all variables in this system have time and spatial dependences

$$U_{x,y}(z;t) = \tfrac{1}{2}U_{x,y}e^{i(\omega t - kz)} + \text{c.c.},$$
$$R_{\rm I,II}^-(z;t) = \tfrac{1}{2}R_{\rm I,II}^- e^{i(\omega t - kz)} + \text{c.c.},$$

and excluding variables $R_{\rm I,II}^-(z;t)$, we derive a system for variables $U_{x,y}$:

$$\begin{aligned}&\left\{\omega^2 - v_t^2k^2 - v_t^2k^2\mathbb{C}\frac{(\omega^-)^2 + i\gamma(\omega - i\gamma)}{(\omega - i\gamma)^2 - (\omega^-)^2}\right\}U_x\\ &\quad - \left\{v_t^2k^2\mathbb{C}\frac{i\omega\omega^-}{(\omega - i\gamma)^2 - (\omega^-)^2}\right\}U_y = 0,\\ &\left\{v_t^2k^2\mathbb{C}\frac{i\omega\omega^-}{(\omega - i\gamma)^2 - (\omega^-)^2}\right\}U_x\\ &\quad + \left\{\omega^2 - v_t^2k^2 - v_t^2k^2\mathbb{C}\frac{(\omega^-)^2 + i\gamma(\omega - i\gamma)}{(\omega - i\gamma)^2 - (\omega^-)^2}\right\}U_y = 0.\end{aligned} \tag{42}$$

Here

$$\mathbb{C} = \frac{\hbar\omega_{21}SN}{2\rho v_t^2} \tag{43}$$

is the quantity that should be regarded as the parameter of coupling between the acoustic wave and the SMM spin system. Note that this parameter includes just the elasticity constant (due to the relation $v_t = \sqrt{K/\rho}$) and the anisotropy constant (due to the relation $\omega_{21} = D(2S-1)\hbar^{-1}$), but it lacks magnetoelastic coupling constants (that are present in a ferromagnet[17]), which is a direct result of relation (11). We use the characteristic value $\mathbb{C} \sim 10^{-4}$ in the numerical calculations. To estimate $\mathbb{C}$, we take the following parameters for Mn$_{12}$-Ac: $\omega_{21}/2\pi \sim 3\times10^{11}$ s^{-1}, $S = 10$, $N \sim 2.5\times 10^{20}$ cm^{-3}, $\rho \sim 2g$ cm^{-3}, $v_t \sim (1.5\text{–}2)\times10^5$ cm/s (see, for example, Refs. 21 and 22).

We would like to note the following circumstance with respect to the value of the characteristic time of the spin system relaxation γ^{-1} (and, hence, the linewidth γ). This value in SMMs is largely determined by both the dipole interaction between the spins [the typical value of the dipole field is of order of $\sim$100 Θe for Mn$_{12}$-Ac (Refs. 11,23)] and the hyperfine interaction [the hyperfine field is about $\sim$250 Θe for Mn$_{12}$-Ac (Refs. 11,15; see also Ref. 24 and references therein)]. Therefore, the linewidth, which depends on random distribution of the above fields (the effect of inhomogeneous line broadening[24]), can be evaluated as 10^9 s^{-1}. Indeed, the measurements taken in Ref. 24 for Mn$_{12}$-Ac at $T = 2.33$ K yielded the linewidth value $\sim 3\times10^9$ s^{-1} for the transitions at frequencies $\omega \sim \omega_{21}$. It is also known that at $T < 1$ K the linewidth is expected to be smaller.[25] Therefore, we assume $\gamma^{-1} = 10^{-9}$ s for the numerical estimations.

Further, the dispersion equations for Eqs. (42) have the following form:

$$\omega^2 - v_t^2k^2\left[1 - \mathbb{C}\frac{(\omega^-)^2 + \gamma^2 + i\omega\gamma \pm \omega\omega^-}{(\omega^-)^2 - (\omega - i\gamma)^2}\right] = 0. \tag{44}$$

From Eq. (44) we obtain the dispersion law for an acoustic wave polarized over the right-hand circle ($U_x = -iU_y$):

$$k_- = \frac{\omega}{v_t}\left[1 - \mathbb{C}\frac{(\omega^-)^2 + \gamma^2 + i\omega\gamma - \omega\omega^-}{(\omega^-)^2 - (\omega - i\gamma)^2}\right]^{-1/2}. \tag{45}$$

Equation (45) corresponds to the nonresonance interaction of the acoustic wave with the SMM spin system. For the acoustic wave polarized over the left-hand circle ($U_x = iU_y$), we have

$$k_+ = \frac{\omega}{v_t}\left[1 - \mathbb{C}\frac{(\omega^-)^2 + \gamma^2 + i\omega\gamma + \omega\omega^-}{(\omega^-)^2 - (\omega - i\gamma)^2}\right]^{-1/2}. \tag{46}$$

Equation (46) corresponds to the resonance interaction of the acoustic wave with the SMM spin system.

Note that we assume ω to be a real quantity, which complies with the treatment of steady-state waves. In this case the wave number is generally a complex value:

$$k_\pm = \mathrm{Re}\,k_\pm + i\,\mathrm{Im}\,k_\pm.$$

The quantities $\mathrm{Im}\,k_\pm$ define the spatial damping of waves: $\sim e^{(\mathrm{Im}\,k_\pm)z}$ ($\mathrm{Im}\,k_\pm < 0$; $z > 0$). Generally speaking, the right- and left-hand polarized waves ($\mathrm{Im}\,k_+ \neq \mathrm{Im}\,k_-$) feature an MDE which forms ellipticity of polarization of the displacement vector oscillations. Thus, if at $z = 0$ the displacement vector oscillations are linearly polarized, at $z \neq 0$ they are elliptically polarized. The minor-to-major semiaxes length ratio, b/a (the ellipticity parameter), can then be written as follows:

$$\frac{b}{a} = \tanh\frac{(\mathrm{Im}\,k_- - \mathrm{Im}\,k_+)z}{2}. \qquad (47)$$

The polarization plane rotation (Faraday rotation angle) reads as follows:

$$\varphi = \frac{(\mathrm{Im}\,k_- - \mathrm{Im}\,k_+)z}{2}. \qquad (48)$$

Using Eqs. (45), (46), and (48), we find the value for the specific Faraday rotation (Faraday rotation angle per unit length):

$$\frac{\varphi}{z} = \frac{\omega}{2v_t}\,\mathrm{Re}\left\{\left[1-\mathbb{C}\frac{(\omega^-)^2+\gamma^2+i\omega\gamma-\omega\omega^-}{(\omega^-)^2-(\omega-i\gamma)^2}\right]^{-1/2} - \left[1-\mathbb{C}\frac{(\omega^-)^2+\gamma^2+i\omega\gamma+\omega\omega^-}{(\omega^-)^2-(\omega-i\gamma)^2}\right]^{-1/2}\right\}. \qquad (49)$$

Equations (47) and (49) are used in numerical calculations. Note that the effects in question are appreciable only in the zone where the acoustic frequency ω comes close to ω_{21} ($\omega^- \approx \omega_{21}$ as $\omega_H \ll \omega_{21}$). Since the typical SMM values of frequency $\omega_{21}/2\pi \sim 3 \times 10^{11}\ \mathrm{s}^{-1}$, our study is relevant for the hypersound.[26] The authors of Ref. 27 were apparently the first to emphasize the importance of research into acoustic waves with frequencies up to 100 GHz in SMMs.

There is one point to make before we start numerical estimations. Consider the value of specific Faraday rotation when it is possible to neglect relaxation in the SMM spin system; that is, in Eq. (49) we may assume $\gamma \to 0$. In addition, assume that the frequency ω is not too close to the frequency ω^-; that is, in Eq. (49), $\mathbb{C}\omega^- \ll (\omega^- - \omega)$. Then, from Eq. (49) we derive

$$\frac{\varphi}{z} \approx \frac{\mathbb{C}}{2v_t}\frac{\omega^2\omega^-}{\omega^2-(\omega^-)^2}. \qquad (50)$$

Interestingly, expression (50) coincides within designations with the expression for the specific Faraday rotation of the magnetoelastic wave in a ferromagnet in the range of long waves,[17] that is, when $\alpha k^2 \ll 1$ (where α is the exchange constant). In particular, for ferromagnets (in the long-wave limit), instead of ω^- in Eq. (50) the corresponding formula in Ref. 17 contains $\omega_S(0)$, which is the frequency of the uniform precession of ferromagnetic magnetization, and instead of $\mathbb{C}$ it has ξ, that is, the parameter of coupling between the acoustic wave and the ferromagnetic spin system. We want to stress one important circumstance, though: $\mathbb{C}$ in contrast to ξ does not contain the magnetoelastic coupling constants.[17] The condition $\alpha k^2 \ll 1$ cannot be realized in ferromagnetics for the hypersound; otherwise it would be possible to neglect the exchange interaction between the spins in a ferromagnet. Formally, this exchange interaction being neglected, a ferromagnet is similar to a SMM, which explains the analogy we found worth mentioning in our analysis.

V. DISCUSSIONS AND ESTIMATIONS

For estimations we assume in formulas (45)–(47) and (49) that $\omega^- \approx \omega_{21}$ since, as mentioned above, $\omega_H \ll \omega_{21}$. Our calculations are made for Mn$_{12}$-Ac (i.e., $\omega_{21}/2\pi \simeq 3 \times 10^{11}\ \mathrm{s}^{-1}$) in the frequency interval $0.9 \times 10^{11} < \omega/2\pi < 4.5 \times 10^{11}\ \mathrm{s}^{-1}$. The appropriateness of the interval choice becomes obvious in further consideration. Here we just note that at lower frequencies the effect of Faraday rotation is very small and at higher frequencies the damping is significant for both the left- and the right-hand waves.

First of all, consider the ω dependences of the quantities $\mathrm{Im}\,k_+$ and $\mathrm{Im}\,k_-$, which determine the damping of the right- and left-hand polarized waves, respectively. Figure 3 is a graph of the dependence of $\mathrm{Im}\,k_-$ on ω [Eq. (45)] for the entire frequency interval ($0.9 \times 10^{11} < \omega/2\pi < 4.5 \times 10^{11}\ \mathrm{s}^{-1}$). It is clearly seen that this dependence is practically linear and the value of $\mathrm{Im}\,k_-$ is such ($-10^{-2} > \mathrm{Im}\,k_- \gtrsim -10^{-1}\ \mathrm{cm}^{-1}$) that on a scale of several centimeters the damping of the corresponding right-hand polarized wave is very weak.

As for the value of $\mathrm{Im}\,k_+$, here it makes sense to distinguish between two parts in the considered frequency interval: the low-frequency range ($0.9 \times 10^{11} < \omega/2\pi < 1.5 \times 10^{11}\ \mathrm{s}^{-1}$) and the high-frequency range ($1.5 \times 10^{11} < \omega/2\pi < 4.5 \times 10^{11}\ s^{-1}$). Figure 4 is a graphical representation of the $\mathrm{Im}\,k_+$ dependence on ω [Eq. (46)] for the low-frequency part of the frequency interval. It is evident that in this region the value of $\mathrm{Im}\,k_+$ is such that the corresponding left-hand polarized wave damps on scales $\lesssim 1$ cm.

It follows from the data in Figs. 3 and 4 that in the low-frequency part of the interval, on scales $\lesssim 1$ cm, the damping of both the left- and the right-hand polarized waves is very weak, which makes it possible to observe the AFE in this

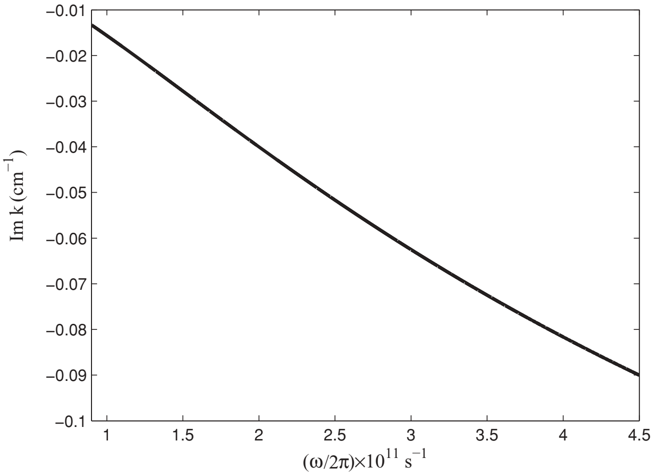

FIG. 3. Dependence of $\mathrm{Im}\,k_-$ on frequency ω ($0.9 \times 10^{11} < \omega/2\pi < 4.5 \times 10^{11}\ \mathrm{s}^{-1}$) for Mn$_{12}$-Ac.

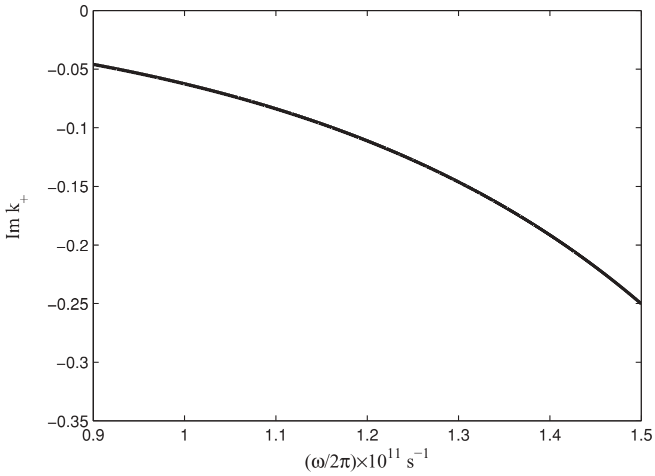

FIG. 4. Dependence of $\mathrm{Im}\,k_+$ on frequency ω ($0.9 \times 10^{11} < \omega/2\pi < 1.5 \times 10^{11}\ \mathrm{s}^{-1}$) for Mn_{12}-Ac.

zone: Figure 5 demonstrates the frequency dependence of the specific Faraday rotation [Eq. (49)]. It is seen that the average value of the specific Faraday rotation is of order 10^2 rad/cm and that the value of the effect becomes very small at lower frequencies. In consideration of this fact, we specified the lower limit of the frequency interval in our analysis.

Now let us turn to the high-frequency part of the interval. Figure 6 shows the graph of the ω dependence of $\mathrm{Im}\,k_+/\mathrm{Im}\,k_-$ (in a logarithmic scale) in this region. Comparing Figs. 3 and 6, one can see that $|\mathrm{Im}\,k_+| \gg |\mathrm{Im}\,k_-|$. That is, only the right-hand polarized wave propagates without noticeable damping in the high-frequency part of the interval. At higher frequencies its damping becomes quite appreciable.

Finally, we present the graph of the ellipticity parameter b/a versus ω [Eq. (47)] in Fig. 7. It is obvious that this graph is in full conformity with the results shown in Figs. 3–6, which is only natural.

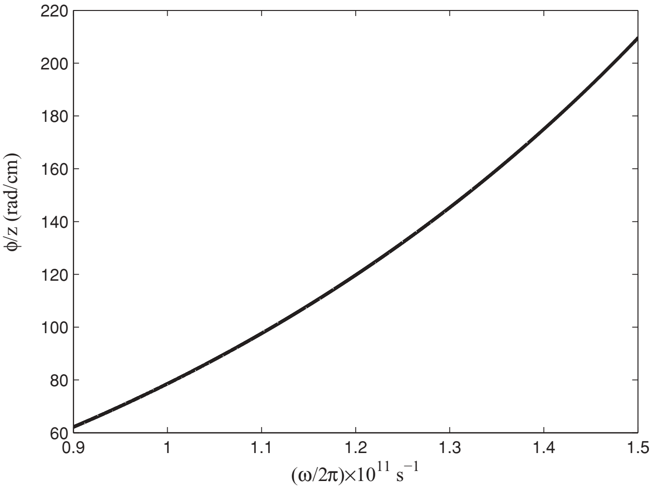

FIG. 5. Dependence of the specific Faraday rotation on frequency ω ($0.9 \times 10^{11} < \omega/2\pi < 1.5 \times 10^{11}\ \mathrm{s}^{-1}$) for Mn_{12}-Ac.

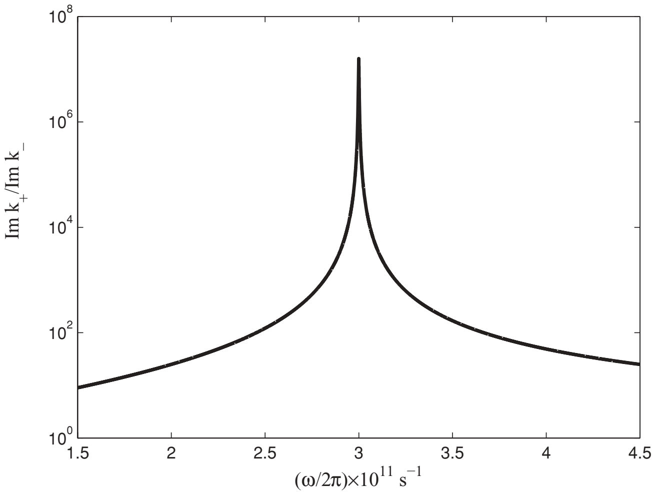

FIG. 6. Dependence of $\mathrm{Im}\,k_+/\mathrm{Im}\,k_-$ (in a logarithmic scale) on frequency ω ($1.5 \times 10^{11} < \omega/2\pi < 4.5 \times 10^{11}\ \mathrm{s}^{-1}$) for Mn_{12}-Ac.

It would be interesting to see how the results of our theoretical study would change if the magnetization state of a SMM is different from that analyzed in this work.

(i) Thus, for a fully polarized SMM state ($N^- = N$) at temperatures T lower (but not much lower) than $\hbar\omega_{21}k_B^{-1}$ (for Mn_{12}-Ac, $\hbar\omega_{21}k_B^{-1} \simeq 14$ K), we have to take into account the populations of at least two levels (states $\varphi_1 = \psi_{-S}$, $\varphi_2 = \psi_{-S+1}$). It can be done by replacing $N \Rightarrow N \tanh[\hbar\omega_{21}/(k_B T)]$ in the expression for parameter $\mathbb{C}$ [Eq. (43)], which is readily understood as the difference in populations of the levels with states $\varphi_1 = \psi_{-S}$ and $\varphi_2 = \psi_{-S+1}$ is proportional to $\tanh[\hbar\omega_{21}/(k_B T)]$.

(ii) If the SMM state is not fully polarized ($N^- \neq N$), this can obviously be taken into account for low temperatures [Eq. (5)] and weak fields ($\omega_H \ll \omega_{21}$) (in the zero order on ω_H/ω_{21}) by replacing $N \Rightarrow N^- - N^+$ in the expression for parameter $\mathbb{C}$ [Eq. (43)].

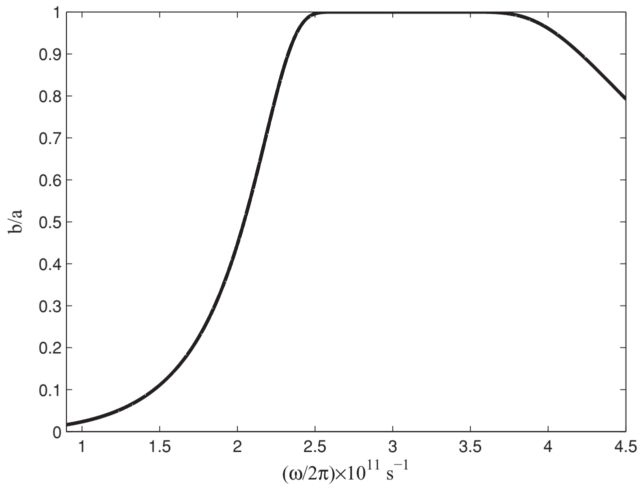

FIG. 7. Dependence of ellipticity parameter b/a on frequency ω ($0.9 \times 10^{11} < \omega/2\pi < 4.5 \times 10^{11}\ \mathrm{s}^{-1}$) for Mn_{12}-Ac.

VI. CONCLUSION

We have demonstrated a possibility of observing the acoustic Faraday effect and the magnetic circular dichroism effect in a crystal of molecular magnets. Our investigation focuses on the case when a crystal of molecular magnets is magnetized through field-cooled magnetization and cooled down to temperatures $\lesssim$1 K. In this state the effects in question manifest themselves in the hypersonic frequency range, their values reaching magnitudes by far too great for the effects to be observed experimentally.

ACKNOWLEDGMENTS

This work is supported by the Ministry of Education (Grant No. RNP. 2.1.1.2686) and the Russian Foundation for Basic Research (Grants No. 09-02-97062 and No. 09-02-97041-p).

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