Parametric interaction of two acoustic waves in a crystal of molecular magnets in the presence of a strong ac magnetic field

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We have shown that, in the presence of a strong ac magnetic field, the parametric interaction of two weak acoustic waves occurs in a crystal of molecular magnets. Any magnetic molecule is subject to a dc magnetic field perpendicular to its easy anisotropy axis. The ac magnetic field is parallel to this axis. The frequencies of the ac magnetic field and acoustic waves are close to the transition frequency of the fundamental doublet. The acoustic waves propagate in opposite directions. We have derived equations for the amplitudes of the two coupled acoustic waves. Due to the parametric interaction, one acoustic wave input into the sample excites the second acoustic wave.

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I. INTRODUCTION

Lately, tunneling of a large spin in magnetic molecules has attracted considerable attention.[1,2] Crystals of molecular magnets are expected to be a powerful source of coherent electromagnetic radiation.[3] Therefore, it is important to thoroughly study interactions of magnetic molecules with electromagnetic and acoustic waves.

Experiments on electron spin resonance[4–8] have demonstrated noticeable resonant absorption of electromagnetic radiation by molecular magnets. The imaginary part of the high-frequency susceptibility, connected with transitions between the states of the fundamental doublet of a high-spin molecule (or a rare-earth metal ion), was studied in linear and nonlinear regimes.[9–11] The dependence of the imaginary part of the susceptibility on temperature, measured in Ref. 11, was discussed in Ref. 12. The nonlinear susceptibility of high-spin molecules subjected to a bifrequency ac magnetic field was theoretically investigated in Ref. 13. The absorption of an acoustic wave by rare-earth metal magnetic ions was theoretically studied in Ref. 14.

The works cited above dealt with stationary processes (i.e., the processes at times much larger than the relaxation times generic for transitions between the states of the fundamental doublet). Nonstationary processes (i.e., processes at times much shorter than the relaxation times) were studied as well. Experiments on tunneling-state echoes in dysprosium-doped glasses were carried out in Ref. 15, while experiments on phonon echoes and saturation of the attenuation of an acoustic wave in rare-earth-ion-doped glasses were carried out in Refs. 16 and 17. The nonstationary behavior of a magnetic molecule (or a rare-earth metal ion) subject to a bifrequency ac magnetic field[18] or to an acoustic wave and an ac magnetic field[19] was investigated theoretically.

The purpose of the present paper is to study the parametric interaction of two weak acoustic waves in a crystal of molecular nanomagnets in the presence of a strong ac magnetic field. We consider a stationary process. Any molecule is subject to a dc magnetic field perpendicular to the easy anisotropy axis of the molecule. The energy levels of any molecule form doublets that are split due to the dc field or due to the dc field and the transverse anisotropy simultaneously. The ac magnetic field is parallel to the easy anisotropy axis. The frequencies of the ac magnetic field and acoustic waves are close to the transition frequency of the fundamental doublet. The acoustic waves propagate in opposite directions. We suppose the sample temperature to be low enough so that in equilibrium just the levels of the fundamental doublet are occupied. Thus, just the levels of the fundamental doublet are involved in the processes we discuss, and we can consider any magnetic molecule as a two-level system.

The energy levels in crystals of molecular magnets are distributed due to dipolar fields,[20] nuclear spins,[20,21] and crystal defects.[22,23] However, we restrict ourselves to the case in which the doublet splitting of individual molecules is dominated by the strong dc magnetic field or by the strong dc magnetic field and the noticeable transverse anisotropy simultaneously. In this case the distribution of the doublet splittings can be neglected.

We assume the magnetic molecules to be noninteracting. It is convenient to describe the statistical properties of such a system by using the one-particle density matrix depending on time and coordinates (in our case, on one coordinate; see Sec. III). The coordinates play the role of parameters. Such a coordinate dependence means that one has carried out an averaging over a small (but containing many molecules) volume.

II. FORMULATION OF THE PROBLEM

We consider a crystal of noninteracting molecular magnets (for example, Mn_{12} acetate or Fe_8) subject to dc and ac magnetic fields and two acoustic waves simultaneously. The ac magnetic field is uniform in the crystal (i.e., the length of the electromagnetic wave is much larger than the crystal size), whereas the lengths of the acoustic waves are much

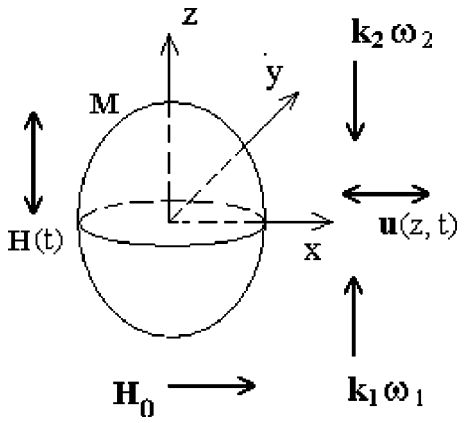

FIG. 1. A magnetic molecule (M) with the easy anisotropy axis z in a dc magnetic field $\mathbf{H}_0$ parallel to the x axis. The vertical arrow with two arrowheads depicts an ac magnetic field $\mathbf{H}(t)=\mathbf{H}\sin\omega t$. The horizontal arrow with two arrowheads depicts the lattice displacement $\mathbf{u}(z,t)$ parallel to the x axis. The vertical arrows are the wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ of two transverse contrariwise propagating acoustic waves whose frequencies are equal to ω_1 and ω_2, respectively.

shorter than the crystal size. First of all, we analyze the behavior of one magnetic molecule. The dc magnetic field is perpendicular to the easy anisotropy axis of the molecule. The ac magnetic field is parallel to this axis (see Fig. 1). The molecule Hamiltonian reads

$$\hat{H} = -D\hat{S}_z^2 + \hat{H}_{tr} - g\mu_B\hat{S}_x H_0 - g\mu_B\hat{S}_z H\sin\omega t + \hat{H}_{me}, \quad (1)$$

where z is the easy anisotropy axis; $\hat{S}_x$, $\hat{S}_y$, $\hat{S}_z$ are the x, y, and z projections of the spin operator; $\hat{H}_{tr}$ is the operator of the transverse anisotropy energy; D, g, and μ_B are the longitudinal anisotropy energy constant, the Landé factor, and the Bohr magneton, respectively; $\mathbf{H}_0$ is the dc magnetic field (we have chosen the x axis to coincide with the direction of this field); H and ω are the amplitude and angular frequency of the ac magnetic field; $\hat{H}_{me}$ is the magnetoelastic coupling.

In magnetic molecules the transverse anisotropy is weaker than the longitudinal one. In Mn_{12} acetate the transverse anisotropy, in practice, is negligible,[4] while in Fe_8 it is more essential.[24] Independently of the form of the transverse anisotropy, the molecule levels form doublets split due to the dc magnetic field and the transverse anisotropy. The two lowest levels of the molecule are of interest to us. We denote them as ε_0 and ε_1 ($\varepsilon_0<\varepsilon_1$). The eigenfunctions corresponding to ε_0 and ε_1 are denoted as φ_0 and φ_1, respectively. Below, we will find the eigenvalues and eigenfunctions numerically in the representation of operator $\hat{S}_z$. If the transverse anisotropy is characterized by two axes in the xy plane (medium and hard axes), we will suppose for definiteness that the dc magnetic field is parallel to the medium axis. In this case (as well as in the absence of the transverse anisotropy) φ_0 is a symmetric function of the z projection of the molecule spin, whereas φ_1 is an antisymmetric function. We suppose that the ac magnetic field frequency ω is close to the transition frequency $\omega_{10}=(\varepsilon_1-\varepsilon_0)/\hbar$, i.e., $|\omega-\omega_{10}|/\omega_{10}\ll 1$.

The magnetoelastic coupling in Mn_{12} acetate reads[22,25,26]

$$\hat{H}_{me} = D(\varepsilon_{xx}-\varepsilon_{yy})(\hat{S}_x^2-\hat{S}_y^2) + D\varepsilon_{xy}\{\hat{S}_x,\hat{S}_y\} + D(\varepsilon_{xz}\{\hat{S}_x,\hat{S}_z\} + \varepsilon_{yz}\{\hat{S}_y,\hat{S}_z\}) + D(\nu_{xz}\{\hat{S}_x,\hat{S}_z\} + \nu_{yz}\{\hat{S}_y,\hat{S}_z\}), \quad (2)$$

where

$$\varepsilon_{\alpha\beta} = \frac{1}{2}\left(\frac{\partial u_\alpha}{\partial x_\beta} + \frac{\partial u_\beta}{\partial x_\alpha}\right), \quad \nu_{\alpha\beta} = \frac{1}{2}\left(\frac{\partial u_\alpha}{\partial x_\beta} - \frac{\partial u_\beta}{\partial x_\alpha}\right), \quad (3)$$

are linear strain tensors at the point where the molecule is located; $\mathbf{u}$ is the lattice displacement; $\alpha,\beta=x,y,z$; $\{\hat{S}_\alpha,\hat{S}_\beta\}$ is the anticommutator. For simplicity we do not describe acoustic waves rigorously, but assume that one longitudinal and two transverse modes of the acoustic waves exist.[27,28] Below, we suppose that two transverse monochromatic acoustic waves polarized along the x axis propagate along the z axis in opposite directions (see Fig. 1), i.e., $\mathbf{u}=(u_x,0,0)$, where

$$u_x = (1/2)u_1(z)\exp[i(k_1 z-\omega_1 t)] + (1/2)u_2(z)\exp[-i(k_2 z+\omega_2 t)] + \text{c.c.}, \quad (4)$$

$u_{1,2}(z)$, $\omega_{1,2}$, $k_{1,2}=\omega_{1,2}/v$, and v are the amplitudes, angular frequencies, wave numbers, and velocity of the acoustic waves. The frequencies $\omega_{1,2}$ are close to the transition frequency $\omega_{10}(|\omega_{1,2}-\omega_{10}|/\omega_{10}\ll 1)$. For the lattice displacement (4) the magnetoelastic coupling described by Eq. (2) reduces to

$$\hat{H}'_{me} = D(\partial u_x/\partial z)\{\hat{S}_x,\hat{S}_z\}. \quad (5)$$

In the case in which the transverse anisotropy is essential (for example, for molecules Fe_8), the exact form of the magnetoelastic coupling between a magnetic molecule and inhomogeneous deformations has not been established so far. We presuppose that the interaction of the magnetic molecule with the acoustic waves of the form (4) is described by the operator

$$\hat{H}'_{me} = F(\partial u_x/\partial z)\{\hat{S}_x,\hat{S}_z\}, \quad (6)$$

similar to the operator (5) for Mn_{12} acetate. The coefficient F in Eq. (6) is of the order of D because the transverse anisotropy is weaker than the longitudinal one.

Since the frequency of the ac magnetic field and the frequencies of the acoustic waves are comparatively close to the transition frequency ω_{10}, at low temperatures when the magnetic molecules occupy just the two lowest levels, we can consider a magnetic molecule as a two-level system and describe it by two master equations[29]

$$\frac{d\rho_{01}}{dt} - i\omega_{10}\rho_{01} + \frac{i}{\hbar}[\hat{H}_{int},\hat{\rho}]_{01} = -\frac{1}{\tau_{10}}\rho_{01}, \quad (7)$$

$$\frac{d\rho_{00}}{dt} + \frac{i}{\hbar}[\hat{H}_{int},\hat{\rho}]_{00} = w_{10}\rho_{11} - w_{01}\rho_{00}. \quad (8)$$

Here, ρ_{00}, ρ_{01}, and ρ_{11} are elements of the density matrix $\hat{\rho}$ for one molecule whose coordinate is z, and

$$\hat{H}_{int} = -g\mu_B \hat{S}_z H \sin \omega t + \hat{H}'_{me} \tag{9}$$

is the operator of interaction of the molecule with the ac magnetic field and the acoustic waves; τ_{10} is the relaxation time of the phase of the matrix elements ρ_{01} and ρ_{10}; w_{10} $[w_{01}]$ is the rate of transition from the state φ_1 $[\varphi_0]$ to the state φ_0 $[\varphi_1]$. We do not give equations for $d\rho_{11}/dt$ and $d\rho_{10}/dt$ because $\rho_{00}+\rho_{11}=1$ and $\rho_{10}=\rho_{01}^*$ (the asterisk means the operation of complex conjugation).

A strong coherent driving field (in our case the ac magnetic field) can modify the relaxation processes in atoms (see Refs. 30–35 and references therein). However, for two-level atoms in free space, these effects are typically weak,[34,35] and we will neglect the influence of the ac magnetic field on the relaxation processes.

It is usual to introduce the time T_{10} of relaxation of the populations of the levels ε_0 and ε_1

$$w_{10} = \rho_{00}^{(0)}/T_{10}, \quad w_{01} = \rho_{11}^{(0)}/T_{10},$$

where $\rho_{00}^{(0)}$ and $\rho_{11}^{(0)}$ are diagonal elements of the equilibrium density matrix, $\rho_{00}^{(0)}+\rho_{11}^{(0)}=1$. Using the relaxation time T_{10}, we rewrite Eq. (8) in the form

$$\frac{d\rho_{00}}{dt} + \frac{i}{\hbar}[\hat{H}_{int}, \hat{\rho}]_{00} = -\frac{1}{T_{10}}(\rho_{00} - \rho_{00}^{(0)}). \tag{10}$$

The stationary solution of Eqs. (7) and (10) is of interest to us. We will find it in the following section.

III. DENSITY MATRIX

Since we assume that the ac magnetic field is strong while the acoustic waves are weak, we can seek the density matrix in the form $\rho_{kj}=\tilde{\rho}_{kj}+\delta\tilde{\rho}_{kj}$ $(k,j=0,1)$, where $\tilde{\rho}_{kj}$ is the solution of Eqs. (7) and (10) in the absence of the acoustic waves and $\delta\tilde{\rho}_{kj}$ is the correction of the first order in the amplitudes of the acoustic waves. The matrix $\tilde{\rho}_{kj}$ obeys the normalization condition $\tilde{\rho}_{00}+\tilde{\rho}_{11}=1$, while the matrix $\delta\tilde{\rho}_{kj}$ satisfies the condition $\delta\tilde{\rho}_{00}+\delta\tilde{\rho}_{11}=0$.

First of all, let us find the matrix $\tilde{\rho}_{kj}$. Due to the symmetry properties of the wave functions φ_0 and φ_1, the matrix elements $(\hat{S}_z)_{00}$ and $(\hat{S}_z)_{11}$ are equal to zero. Therefore, in the absence of acoustic waves Eqs. (7) and (10) reduce to

$$(d/dt - i\omega_{10} + \tau_{10}^{-1})\tilde{\rho}_{01} - \frac{i}{\hbar} g\mu_B H (\hat{S}_z)_{01}(1-2\tilde{\rho}_{00})\sin \omega t = 0, \tag{11}$$

$$\frac{d\tilde{\rho}_{00}}{dt} - \frac{i}{\hbar} g\mu_B H (\hat{S}_z)_{01}(\tilde{\rho}_{01}^* - \tilde{\rho}_{01})\sin \omega t = -\frac{1}{T_{10}}(\tilde{\rho}_{00} - \rho_{00}^{(0)}), \tag{12}$$

where the matrix element $(\hat{S}_z)_{01}=\langle\varphi_0|\hat{S}_z|\varphi_1\rangle$ is real. Since the frequency ω is close to the transition frequency ω_{10}, we use the rotating wave approximation[36] (analogous to the resonant perturbation theory for the Schrödinger equation[37]) and seek the stationary solution of Eqs. (11) and (12) in the form $\tilde{\rho}_{00}=\text{const}$, $\tilde{\rho}_{01}=R_{01}\exp(i\omega t)$, where $R_{01}=\text{const}$. Adding to the system (11) and (12) the equation complex conjugate to Eq. (11), and solving the system of linear equations for $\tilde{\rho}_{00}$, R_{01}, and R_{01}^*, one can obtain

$$\tilde{\rho}_{00} = \frac{\rho_{00}^{(0)}(1+\Omega^2\tau_{10}^2) + 2\Omega_R^2\tau_{10}T_{10}}{1+\Omega^2\tau_{10}^2 + 4\Omega_R^2\tau_{10}T_{10}}, \tag{13}$$

$$R_{01} = -\frac{(2\rho_{00}^{(0)}-1)\Omega_R\tau_{10}(1-i\Omega\tau_{10})}{1+\Omega^2\tau_{10}^2 + 4\Omega_R^2\tau_{10}T_{10}}, \tag{14}$$

where $\Omega=\omega-\omega_{10}$ is the deviation of the ac magnetic field frequency from the transition frequency, and

$$\Omega_R = g\mu_B H|(\hat{S}_z)_{01}|/(2\hbar) \tag{15}$$

is the Rabi frequency. Equations (13) and (14) are quite similar to the expressions for the density matrix of a two-level atom in an ac electric field.[29]

Now let us turn to the matrix $\delta\tilde{\rho}_{kj}$. For brevity we start from the case in which only one acoustic wave propagates in the crystal [for example, $u_2(z)\equiv 0$ in Eq. (4)]. The corresponding matrix $\delta\tilde{\rho}_{kj}$ can be easily extended to the case in which $u_2(z)\neq 0$, because $\delta\tilde{\rho}_{kj}$ is linear in the amplitudes of acoustic waves. It follows from Eqs. (7) and (10)–(12) that this matrix obeys the equations

$$(d/dt - i\omega_{10} + \tau_{10}^{-1})\delta\tilde{\rho}_{01} + 2\Omega_R(e^{i\omega t} - e^{-i\omega t})\delta\tilde{\rho}_{00} = \frac{Fk_1}{2\hbar}\{\hat{S}_x, \hat{S}_z\}_{10}(u_1 e^{ik_1 z - i\omega_1 t} - u_1^* e^{-ik_1 z + i\omega_1 t})(1-2\tilde{\rho}_{00}), \tag{16}$$

$$(d/dt + T_{10}^{-1})\delta\tilde{\rho}_{00} - \Omega_R(e^{i\omega t} - e^{-i\omega t})(\delta\tilde{\rho}_{01}^* - \delta\tilde{\rho}_{01}) = \frac{Fk_1}{2\hbar}\{\hat{S}_x, \hat{S}_z\}_{10}(u_1 e^{ik_1 z - i\omega_1 t} - u_1^* e^{-ik_1 z + i\omega_1 t})(\tilde{\rho}_{01}^* - \tilde{\rho}_{01}), \tag{17}$$

where the matrix element

$$\{\hat{S}_x, \hat{S}_z\}_{10} = \langle\varphi_1|\{\hat{S}_x, \hat{S}_z\}|\varphi_0\rangle \tag{18}$$

is real. When deriving these equations, we have taken into account that, due to the symmetry properties of the wave functions φ_0 and φ_1, the matrix elements $\{\hat{S}_x, \hat{S}_z\}_{00}$ and $\{\hat{S}_x, \hat{S}_z\}_{11}$ are equal to zero.

We make use of the rotating wave approximation once again and seek the stationary solution of Eqs. (16) and (17) in the form

$$\delta\tilde{\rho}_{00} = (1/2)R_1 e^{i(\omega_1-\omega)t} + \text{c.c.}, \tag{19}$$

$$\delta\tilde{\rho}_{01} = U_1 e^{i\omega_1 t} + V_1 e^{i(2\omega-\omega_1)t}, \tag{20}$$

where R_1, U_1, and V_1 do not depend on time. Equation (19) takes into account that diagonal elements of the density matrix are real. The second term in the right-hand side of Eq. (20) describes oscillations of the matrix element $\delta\tilde{\rho}_{01}$ at the combinative frequency $2\omega-\omega_1$.[38] The amplitude of these oscillations vanishes if the amplitude of the ac magnetic field tends to zero. Substituting expressions (19) and (20) into

Eqs. (16) and (17), one can obtain a linear system for the amplitudes R_1, U_1, and V_1^*. This system yields

$$U_1 = \frac{Fk_1}{2\hbar}\{\hat{S}_x,\hat{S}_z\}_{10}\tau_{10}u_1^*(z)e^{-ik_1z}\Bigg((2\tilde{\rho}_{00}-1)\Bigg[1-i(\Omega-\Omega_1)T_{10} + \frac{2\Omega_R^2\tau_{10}T_{10}}{1-i(2\Omega-\Omega_1)\tau_{10}}\Bigg] + 2\Omega_R T_{10}R_{01}^*\Bigg)\Bigg((1+i\Omega_1\tau_{10})\Bigg[1 - i(\Omega-\Omega_1)T_{10} + \frac{2\Omega_R^2\tau_{10}T_{10}}{1-i(2\Omega-\Omega_1)\tau_{10}}\Bigg] + 2\Omega_R^2\tau_{10}T_{10}\Bigg)^{-1}, \tag{21}$$

$$V_1 = \frac{Fk_1}{2\hbar}\{\hat{S}_x,\hat{S}_z\}_{10}\tau_{10}u_1(z)e^{ik_1z}\frac{1}{1+i(2\Omega-\Omega_1)\tau_{10}} \times(-(2\tilde{\rho}_{00}-1)2\Omega_R^2\tau_{10}T_{10} + 2\Omega_R T_{10}(1-i\Omega_1\tau_{10})R_{01}) \times\Bigg((1-i\Omega_1\tau_{10})\Bigg[1+i(\Omega-\Omega_1)T_{10} + \frac{2\Omega_R^2\tau_{10}T_{10}}{1+i(2\Omega-\Omega_1)\tau_{10}}\Bigg] + 2\Omega_R^2\tau_{10}T_{10}\Bigg)^{-1}, \tag{22}$$

where $\Omega_1=\omega_1-\omega_{10}$.

One can see that the first term in the right-hand side of Eq. (20) is proportional to $\exp(i\omega_1 t-ik_1z)$. This term describes a disturbance propagating in the positive-z direction and having the frequency ω_1 and the wave number k_1. The second term in the right-hand side of Eq. (20) is proportional to $\exp[i(2\omega-\omega_1)t+ik_1z]$. This term describes a disturbance propagating in the negative-z direction and having the frequency $2\omega-\omega_1$. As we will show below, this disturbance is responsible for the excitation of another acoustic wave with the frequency $2\omega-\omega_1$. Thus, we have to assume that the frequency of the second acoustic wave in Eq. (4) is equal to $\omega_2=2\omega-\omega_1$ [and, as a consequence, $k_2=(2\omega-\omega_1)/v$].

The relation

$$\omega_1+\omega_2=2\omega \tag{23}$$

can be interpreted as follows. Due to the influence of the strong ac magnetic field on molecular magnets, the parametric interaction of two acoustic waves occurs. In this process two electromagnetic quanta, $2\hbar\omega$, are converted into two phonons, $\hbar\omega_1$ and $\hbar\omega_2$.

The full expression for the matrix element $\delta\tilde{\rho}_{01}$, taking into account both acoustic waves in Eq. (4), is

$$\delta\tilde{\rho}_{01} = U_1e^{i\omega_1 t} + V_1e^{i(2\omega-\omega_1)t} + U_2e^{i\omega_2 t} + V_2e^{i(2\omega-\omega_2)t}, \tag{24}$$

where U_2 and V_2 are described by expressions (21) and (22) in which ω_1, k_1, and $u_1(z)$ have to be replaced by ω_2, $-k_2$, and $u_2(z)$, respectively.

IV. EQUATIONS FOR THE AMPLITUDES OF THE ACOUSTIC WAVES

After we have found the density matrix, we can derive equations for the amplitudes $u_{1,2}(z)$ of the acoustic waves. We focus on the interaction of the acoustic waves with magnetic molecules and do not consider other mechanisms of decay of the acoustic waves. We start from the density of the elastic Lagrangian

$$\mathcal{L} = \frac{1}{2}\rho\left(\frac{\partial u_x}{\partial t}\right)^2 - \frac{1}{2}\rho v^2\left(\frac{\partial u_x}{\partial z}\right)^2 - N\,\mathrm{tr}(\hat{\rho}\hat{H}'_{me}), \tag{25}$$

where ρ is the crystal density, N is the concentration of magnetic molecules, $\mathrm{tr}(\hat{A})$ means the trace of the matrix $\hat{A}$. Taking into account that

$$\mathrm{tr}(\hat{\rho}\hat{H}'_{me}) = F\frac{\partial u_x}{\partial z}(\rho_{01}+\rho_{01}^*)\{\hat{S}_x,\hat{S}_z\}_{10}, \tag{26}$$

we come to the Lagrange equation

$$\frac{\partial^2 u_x}{\partial t^2} - v^2\frac{\partial^2 u_x}{\partial z^2} = \frac{NF}{\rho}\{\hat{S}_x,\hat{S}_z\}_{10}\frac{\partial}{\partial z}(\rho_{01}+\rho_{01}^*). \tag{27}$$

In the preceding section, we have represented the matrix element ρ_{01} as the sum of $\tilde{\rho}_{01}$ and $\delta\tilde{\rho}_{01}$, where $\tilde{\rho}_{01}$ describes a magnetic molecule in the presence of the ac magnetic field; $\delta\tilde{\rho}_{01}$ is the correction linear in the amplitudes of the acoustic waves. Only the term $\delta\tilde{\rho}_{01}$ depends on z; therefore, in Eq. (27) we can replace ρ_{01} by $\delta\tilde{\rho}_{01}$. Let us substitute Eqs. (4) and (24) into Eq. (27) . We presuppose that the interaction of the acoustic waves with magnetic molecules is comparatively weak and, as a consequence, the amplitudes $u_{1,2}(z)$ are slow functions of z, namely $|du_{1,2}/dz| \ll k_{1,2}|u_{1,2}|$. In this case, we can neglect the second derivatives of functions $u_{1,2}(z)$ in the left-hand side of Eq. (27) and the first derivatives of these functions in the right-hand side of Eq. (27). We assume the frequencies ω and ω_1 to be so close to the transition frequency that $|k_2-k_1|/k_1=2|\omega-\omega_1|/\omega_1 \ll 1$. Equating the terms with identical exponential factors [$\exp(ik_1z-i\omega_1t)$ or $\exp(-ik_2z-i\omega_2t)$] in the left-hand and right-hand sides of Eq. (27) , we obtain the desired equations

$$\frac{du_1}{dz} = -k_1\alpha_1u_1 - (k_2^2/k_1)\beta_1e^{i(k_2-k_1)z}u_2^*, \tag{28}$$

$$\frac{du_2}{dz} = k_2\alpha_2u_2 + (k_1^2/k_2)\beta_2e^{i(k_2-k_1)z}u_1^*. \tag{29}$$

Here

$$\alpha_1 = \frac{NF^2}{2\hbar\rho v^2}\{\hat{S}_x,\hat{S}_z\}_{10}^2\frac{\tau_{10}(2\rho_{00}^{(0)}-1)}{1+\Omega^2\tau_{10}^2+4\Omega_R^2\tau_{10}T_{10}} \times\Bigg((1+\Omega^2\tau_{10}^2)\Bigg[1+i(\Omega-\Omega_1)T_{10}+\frac{2\Omega_R^2\tau_{10}T_{10}}{1+i(2\Omega-\Omega_1)\tau_{10}}\Bigg] - 2\Omega_R^2\tau_{10}T_{10}(1-i\Omega\tau_{10})\Bigg) \times\Bigg((1-i\Omega_1\tau_{10})\Bigg[1+i(\Omega-\Omega_1)T_{10} + \frac{2\Omega_R^2\tau_{10}T_{10}}{1+i(2\Omega-\Omega_1)\tau_{10}}\Bigg] + 2\Omega_R^2\tau_{10}T_{10}\Bigg)^{-1}, \tag{30}$$

$$\beta_1 = \frac{NF^2}{2\hbar\rho v^2}\{\hat{S}_x,\hat{S}_z\}_{10}^2\frac{\tau_{10}(2\rho_{00}^{(0)}-1)}{1+\Omega^2\tau_{10}^2+4\Omega_R^2\tau_{10}T_{10}} \times\frac{4\Omega_R^2\tau_{10}T_{10}(1+i\Omega\tau_{10})(1-i(\Omega-\Omega_2)\tau_{10}/2)}{1-i(2\Omega-\Omega_2)\tau_{10}} \times\Bigg((1+i\Omega_2\tau_{10})\Bigg[1-i(\Omega-\Omega_2)T_{10} + \frac{2\Omega_R^2\tau_{10}T_{10}}{1-i(2\Omega-\Omega_2)\tau_{10}}\Bigg]+2\Omega_R^2\tau_{10}T_{10}\Bigg)^{-1} \quad (31)$$

are dimensionless coefficients, $\Omega_2=\omega_2-\omega_{10}$. The coefficients α_2 and β_2 can be found if one replaces Ω_1, Ω_2 by Ω_2, Ω_1 in Eqs. (30) and (31), respectively. Equations (28) and (29) are valid if the functions $u_{1,2}(z)$ are really slow, i.e.,

$$|\alpha_{1,2}| \ll 1, \quad |\beta_{1,2}| \ll 1. \quad (32)$$

One can see from Eqs. (30) and (31) that the coefficients of the equations for the amplitudes of the acoustic waves contain the transition frequency ω_{10} only as a cofactor of the relaxation time τ_{10}. Therefore, the distribution of the doublet splittings is unessential if the width of this distribution, Δ, is much smaller than $\hbar/\tau_{10}$, i.e., $\Delta \ll \hbar/\tau_{10}$.

According to Eqs. (30) and (31), the important parameter controlling the behavior of the molecular magnets is $4\Omega_R^2\tau_{10}T_{10}$.

V. DISCUSSION AND NUMERICAL RESULTS

A. Weak ac magnetic field

If the ac magnetic field is weak, namely

$$4\Omega_R^2\tau_{10}T_{10} \ll 1, \quad (33)$$

then $|\beta_1/\alpha_1| \ll 1$, $|\beta_2/\alpha_2| \ll 1$, and the system of equations (28) and (29) decomposes into two independent equations

$$\frac{du_1}{dz} = -k_1\alpha_1 u_1, \quad (34)$$

$$\frac{du_2}{dz} = k_2\alpha_2 u_2, \quad (35)$$

where

$$\alpha_{1,2} = \frac{NF^2}{2\hbar\rho v^2}\{\hat{S}_x,\hat{S}_z\}_{10}^2\frac{\tau_{10}(2\rho_{00}^{(0)}-1)}{1-i\Omega_{1,2}\tau_{10}}. \quad (36)$$

The first [second] of these equations describes the decrease of the amplitude and a small change of the wave number of the acoustic wave propagating in the positive [negative] z direction and interacting with magnetic molecules. Let only one acoustic wave (whose frequency, for example, is ω_1) propagate in the crystal occupying the region $z \geqslant 0$. According to Eq. (34), we have

$$u_1(z) = u_1(0)e^{-k_1\alpha_1 z}, \quad (37)$$

where $u_1(0)$ is the amplitude in the plane $z=0$. The mean energy flux of the acoustic wave equals

$$\Pi_z(z) = (1/2)\rho v\omega_1^2|u_1(z)|^2; \quad (38)$$

therefore, the mean acoustic power absorbed by magnetic molecules in a unit volume is

$$P(z) = -\frac{\partial\Pi_z(z)}{\partial z} = P_0(z)(1+\Omega_1^2\tau_{10}^2)^{-1}, \quad (39)$$

where

$$P_0(z) = \frac{NF^2\omega_1^3}{2\hbar v^2}\{\hat{S}_x,\hat{S}_z\}_{10}^2\tau_{10}(2\rho_{00}^{(0)}-1)|u_1(z)|^2. \quad (40)$$

Here, we can replace ω_1^3 by ω_{10}^3 because we suppose that $|\omega_1-\omega_{10}|\tau_{10}$ does not exceed several units, whereas the master equations used above are valid under the condition $\omega_{10}\tau_{10} \gg 1$. After such a replacement the quantity $P_0(z)$ is independent of the frequency, and Eq. (39) describes a Lorentzian peak of absorption.

B. Strong ac magnetic field

If the ac magnetic field is strong enough ($4\Omega_R^2\tau_{10}T_{10} \gtrsim 1$), the parametric interaction of the acoustic waves may become noticeable. We focus on the case in which a crystal of molecular magnets occupies the region $0 \leqslant z \leqslant L$ and only one acoustic wave is input into the sample, for example, at the edge $z=0$. Supposing that this wave is not reflected at the edge $z=L$, we can assume that the boundary condition at this edge is $u_2(L)=0$. We also suppose that the second acoustic wave excited due to the parametric process and propagating in the negative-z direction is not reflected at the edge $z=0$; then, we can assume that at this edge the amplitude of the input wave, $u_1(0)$, is given (we concentrate on the parametric interaction and do not discuss how to match the acoustic impedances of the molecular magnets crystal and an acoustic wave guide). The solution of Eqs. (28) and (29) with the boundary conditions mentioned above reads

$$u_1(z) = u_1(0)\exp[(i\Delta k - k_1\alpha_1 + k_2\alpha_2^*)z/2] \times\sinh[k_1\gamma(L-z)+\xi]/\sinh(k_1\gamma L+\xi), \quad (41)$$

$$u_2^*(z) = -u_1(0)(k_1/k_2)^{3/2}(\beta_2^*/\beta_1)^{1/2}\exp[(-i\Delta k - k_1\alpha_1 + k_2\alpha_2^*)z/2]\sinh[k_1\gamma(L-z)]/\sinh(k_1\gamma L+\xi), \quad (42)$$

where $\Delta k = k_2 - k_1$,

$$\gamma = (\nu^2 - \beta_1\beta_2^* k_2/k_1)^{1/2}, \quad \nu = (\alpha_1 + k_2\alpha_2^*/k_1 + i\Delta k/k_1)/2, \quad (43)$$

$$\xi = \frac{1}{2}\ln\left(\frac{\nu+\gamma}{\nu-\gamma}\right). \quad (44)$$

First, we analyze the obtained solution in the case in which $k_1=k_2$, i.e., the frequencies of both acoustic waves are equal and coincide with the frequency of the ac magnetic field ($\omega_1=\omega_2=\omega$). As we will show, in this case the parametric interaction of two acoustic waves is more efficient.

1. The frequencies of the acoustic waves are equal

One can see that in this case $\Delta k=0$,

$$\alpha_1=\alpha_2=\frac{NF^2}{2\hbar\rho v^2}\{\hat{S}_x,\hat{S}_z\}_{10}^2\frac{\tau_{10}(2\rho_{00}^{(0)}-1)(1+\Omega^2\tau_{10}^2)(1+i\Omega\tau_{10})}{(1+\Omega^2\tau_{10}^2+4\Omega_R^2\tau_{10}T_{10})^2}, \tag{45}$$

$$\beta_1=\beta_2=\alpha_1 4\Omega_R^2\tau_{10}T_{10}/(1+\Omega^2\tau_{10}^2), \tag{46}$$

and Eqs. (41) and (42) reduce to

$$u_1(z)=u_1(0)e^{-ik_1 z\ \mathrm{Im}\ \alpha_1}\sinh[k_1\gamma(L-z)+\xi]/\sinh(k_1\gamma L+\xi), \tag{47}$$

$$u_2^*(z)=-u_1(0)e^{-ik_1 z\ \mathrm{Im}\ \alpha_1}(\beta_1^*/|\beta_1|)\times\sinh[k_1\gamma(L-z)]/\sinh(k_1\gamma L+\xi), \tag{48}$$

with

$$\gamma=\sqrt{(\mathrm{Re}\ \alpha_1)^2-|\beta_1|^2},\quad \xi=\frac{1}{2}\ln\left(\frac{\mathrm{Re}\ \alpha_1+\gamma}{\mathrm{Re}\ \alpha_1-\gamma}\right). \tag{49}$$

The behavior of the amplitudes $u_1(z)$ and $u_2(z)$ depends on the ratio of the coefficient characterizing the parametric coupling of the acoustic waves, $k_1|\beta_1|$, to the absorption constant of each acoustic wave in the presence of the ac magnetic field, $k_1\mathrm{Re}\ \alpha_1$,

$$\eta=|\beta_1|/\mathrm{Re}\ \alpha_1=4\Omega_R^2\tau_{10}T_{10}/\sqrt{1+\Omega^2\tau_{10}^2}. \tag{50}$$

Again, we take into account only the acoustic wave absorption caused by the interaction of the acoustic waves with magnetic molecules. Other mechanisms of the acoustic wave absorption increase the quantity $\mathrm{Re}\ \alpha_1$. Therefore, the effects we discuss below may occur at larger values of the amplitude of the ac magnetic field than we predict. Unfortunately, we are not aware of experimental works in which the absorption constant of an acoustic wave in a crystal of molecular magnets has been measured and the mechanisms of absorption of the acoustic wave have been found out in detail.

If the ratio (50) is smaller than unity, the quantity γ is real and, as a consequence, the amplitudes $|u_1(z)|$ and $|u_2(z)|$ are monotonically decreasing functions of the z coordinate. However, if the ratio (50) exceeds unity, the quantity γ is pure imaginary and Eqs. (47) and (48) reduce to

$$u_1(z)=u_1(0)e^{-ik_1 z\ \mathrm{Im}\ \alpha_1}\sin[k_1|\gamma|(L-z)+\tilde{\xi}]/\sin(k_1|\gamma|L+\tilde{\xi}), \tag{51}$$

$$u_2^*(z)=-u_1(0)e^{-ik_1 z\ \mathrm{Im}\ \alpha_1}(\beta_1^*/|\beta_1|)\times\sin[k_1|\gamma|(L-z)]/\sin(k_1|\gamma|L+\tilde{\xi}), \tag{52}$$

where $\tilde{\xi}=\arctan\sqrt{\eta^2-1}$. Therefore, for sufficiently large L, the amplitudes $|u_1(z)|$ and $|u_2(z)|$ are oscillating functions of the z coordinate. According to Eq. (50) , the greater the deviation of the ac magnetic field frequency from the transition frequency, the larger the amplitude of the ac magnetic field required to satisfy the condition $\eta>1$.

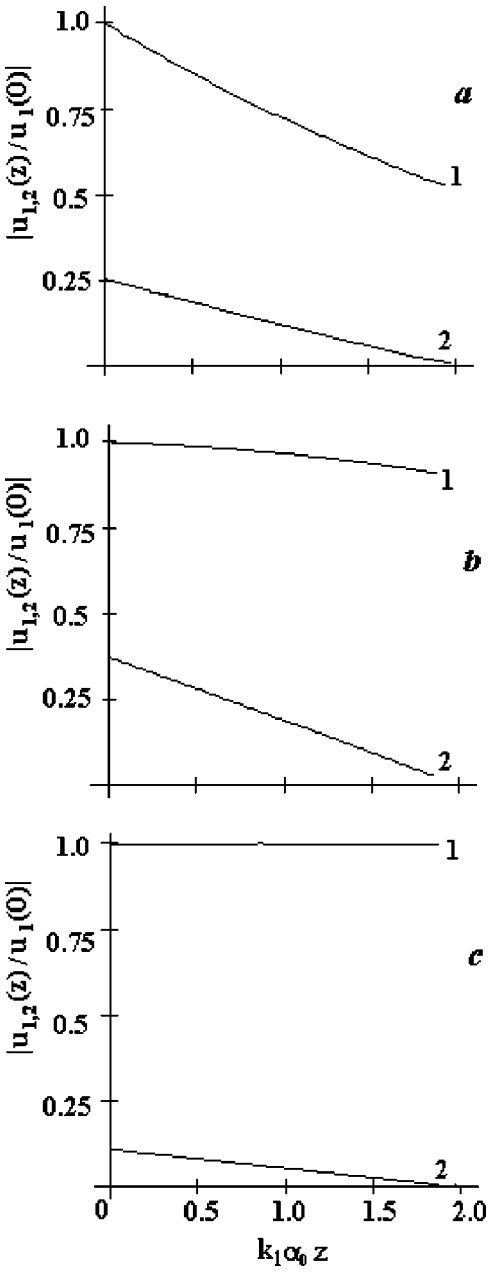

FIG. 2. Dependences of the amplitudes $|u_1(z)/u_1(0)|$ (curves 1) and $|u_2(z)/u_1(0)|$ (curves 2) on the z coordinate for three values of the amplitude of the ac magnetic field [$2\Omega_R\sqrt{\tau_{10}T_{10}}=0.8$ (a), 1.5 (b), and 4 (c)]. The frequencies of both acoustic waves and of the ac magnetic field coincide with the transition frequency. The dimensionless length of the sample is $k_1\alpha_0 L=2$, where $k_1\alpha_0$ is the imaginary part of the wave number of the acoustic wave in the absence of the ac magnetic field.

The coordinate dependences of the amplitudes of the acoustic waves, described by Eqs. (47) and (48), are plotted in Fig. 2 for several values of the amplitude of the ac magnetic field. We have chosen the frequency of the ac magnetic field to be equal to the transition frequency. We use the dimensionless coordinate $k_1\alpha_0 z$, where $\alpha_0=\alpha_1(\Omega=\Omega_R=0)$, and suppose that $k_1\alpha_0 L=2$. It is easy to see that $k_1\alpha_0$ is the imaginary part of the wave number of the acoustic wave (whose frequency is equal to the transition frequency) in the linear

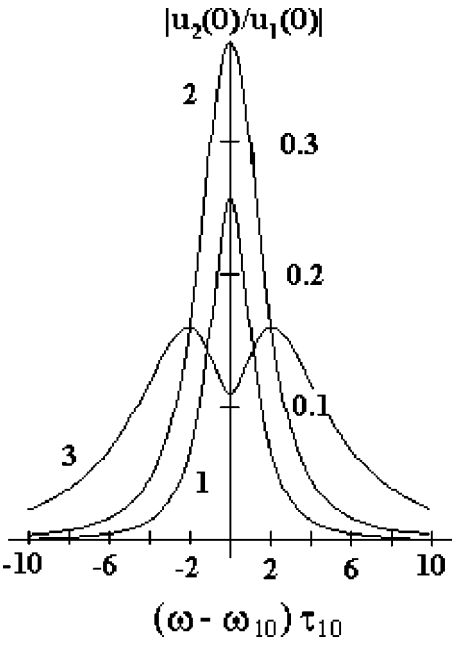

FIG. 3. The ratio of the amplitudes of the parametrically excited and input acoustic waves at the edge $z=0$ of the sample, $|u_2(0)/u_1(0)|$, as a function of the frequency of the acoustic waves for three values of the amplitude of the ac magnetic field $[2\Omega_R\sqrt{\tau_{10}T_{10}}=0.8$ (curve 1), 1.5 (2), and 4 (3)]. The dimensionless length of the sample is $k_1\alpha_0 L=2$.

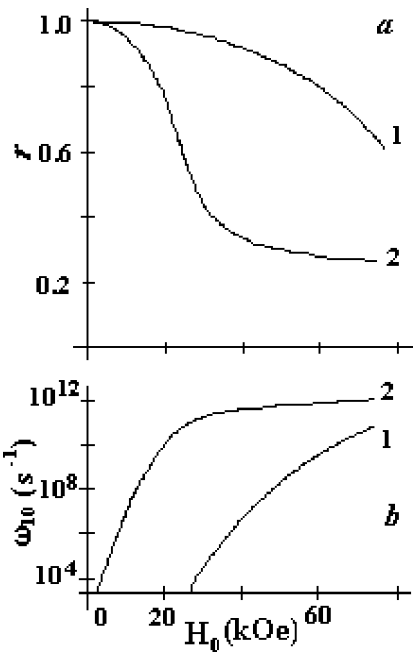

FIG. 4. Dependences of the normalized matrix element (a) and the transition frequency (b) on the dc magnetic field perpendicular to the easy anisotropy axis for Mn_{12} (curves 1) and Fe_8 (curves 2) clusters.

regime, i.e., in the absence of the ac magnetic field. One can see that at the edge $z=0$ the amplitude of the acoustic wave excited due to the parametric process can be of the order of the amplitude of the acoustic wave input into the sample.

Along the sample, the arguments of the sinusoidal functions in Eqs. (51) and (52) change by

$$k_1|\gamma|L = k_1\alpha_0 L\frac{\sqrt{(4\Omega_R^2\tau_{10}T_{10})^2-1-\Omega^2\tau_{10}^2}\sqrt{1+\Omega^2\tau_{10}^2}}{(1+\Omega^2\tau_{10}^2+4\Omega_R^2\tau_{10}T_{10})^2}. \tag{53}$$

This quantity as a function of parameter $2\Omega_R\sqrt{\tau_{10}T_{10}}$ has a maximum and is sufficiently small for large values of $2\Omega_R\sqrt{\tau_{10}T_{10}}$. In particular, for $\Omega=0$ the quantity (53) reaches its maximum value, $\sqrt{3}k_1\alpha_0 L/9$, at $2\Omega_R\sqrt{\tau_{10}T_{10}}=\sqrt{2}$. That is why the amplitudes $|u_1(z)|$ and $|u_2(z)|$ in Fig. 2(c) change less than in Fig. 2(b).

The ratio of the amplitude of the parametrically excited acoustic wave to the amplitude of the input acoustic wave in the plane $z=0$, $|u_2(0)/u_1(0)|$, as a function of frequency is depicted in Fig. 3 for the same values of parameter $2\Omega_R\sqrt{\tau_{10}T_{10}}$ as in Fig. 2. If this parameter is not very large, the ratio $|u_2(0)/u_1(0)|$ reaches a maximum value at the frequency equal to the transition frequency (curves 1 and 2 in Fig. 3). However, if the parameter $2\Omega_R\sqrt{\tau_{10}T_{10}}$ is sufficiently large, the frequency dependence of the ratio $|u_2(0)/u_1(0)|$ has two maxima (curve 3 in Fig. 3).

It should be noted that the denominator in Eqs. (51) and (52) vanishes if the sample length obeys the condition

$$k_1|\gamma|L+\tilde{\xi}=\pi n \quad (n=1,2,3,\ldots). \tag{54}$$

Under this condition there exists a nontrivial solution of Eqs. (28) and (29) with boundary conditions $u_1(0)=0$, $u_2(L)=0$, i.e., in the case in which no acoustic wave is input into the sample. The corresponding amplitudes of the acoustic waves are of the form

$$|u_1(z)|=C|\sin(k_1|\gamma|z)|, \quad |u_2(z)|=C|\sin[k_1|\gamma|(L-z)]|,$$

where C is a constant. One can see that the amplitudes satisfy the natural symmetry condition $|u_2(z)|=|u_1(L-z)|$. The existence of the nontrivial solution of Eqs. (28) and (29) in the absence of an input acoustic wave means that, under the condition (54), parametric generation of acoustic waves occurs. However, to describe the process of wave generation, a nonlinear theory is required. Our approach is linear in the amplitudes of acoustic waves; therefore, we cannot describe the parametric generation of such waves and suppose that the condition (54) is not satisfied.

To estimate the amplitude of the ac magnetic field, required for manifestation of the parametric process, we use the inequality $2\Omega_R\sqrt{\tau_{10}T_{10}}\geqslant 1$ in the form

$$H \gtrsim \hbar(g\mu_B S\sqrt{\tau_{10}T_{10}}r)^{-1}, \tag{55}$$

where $r=|(\hat{S}_z)_{01}|/S$ is a dimensionless coefficient (a normalized matrix element) of the order of unity. We have found this coefficient numerically for Mn_{12} and Fe_8 clusters (see Fig. 4). According to Refs. 4, 5, and 11, for Mn_{12} clusters $S=10$, $D=0.68$ *K*, $g=1.9$, $\hat{H}_{tr}=-B(\hat{S}_+^4+\hat{S}_-^4)$ with $B=6\times10^{-5}$ *K*, $\hat{S}_\pm=\hat{S}_x\pm i\hat{S}_y$. We have chosen these parameters, except for coefficient B. We have obtained better agreement

between our calculations and the experimental data[11] ($\omega_{10} \simeq 2\pi \times 680$ MHz at $H_0 \simeq 60.6$ kOe) for $B \simeq 3 \times 10^{-6}$ *K*, and this value has been used in our calculations. For Fe_8 clusters we have supposed that $S=10$, $D=0.23$ *K*, $g=2$, $\hat{H}_{tr}=K\hat{S}_y^2$, with $K/D=0.4$ (Refs. 6 and 24) (the chosen form of the transverse anisotropy implies that the medium anisotropy axis coincides with the *x* axis; we recall that the dc magnetic field is parallel to the medium anisotropy axis). For convenience, in Fig. 4 we have also depicted the transition frequency as a function of magnetic field. For example, if the value of dc magnetic field is $H_0=60.6$ kOe, for Mn_{12} acetate we find $r \simeq 0.793$ and $\omega_{10} \simeq 4.276 \times 10^9 s^{-1}$. Supposing $\sqrt{\tau_{10}T_{10}} \sim 10^{-8}$s, we obtain from Eq. (55) that $H \gtrsim 0.76$ Oe. Similarly, for Fe_8 clusters at $H_0=20$ kOe we have $r \simeq 0.762$ and $\omega_{10} \simeq 9.76 \times 10^9 s^{-1}$. If $\sqrt{\tau_{10}T_{10}} \sim 10^{-8}$s, we obtain $H \gtrsim 0.75$ Oe. Thus, the amplitude of the ac magnetic field, required for observation of the parametric interaction of two acoustic waves, is comparatively small ($\gtrsim 1$ Oe). It should be noted that another nonlinear effect connected with transitions between the levels of the fundamental doublet (the saturation of absorption of an intense ac magnetic field) also occurs at sufficiently small amplitudes and was observed experimentally for rare-earth ions Dy^{3+} ($H \gtrsim 2$ Oe) and magnetic molecules $CrNi_6$ ($H \gtrsim 10$ Oe) (see Refs. 9 and 10, respectively).

2. The frequencies of the acoustic waves are different

By using Eqs. (41) and (42), we have numerically investigated the ratio of the amplitudes of the parametrically excited and input acoustic waves at the edge $z=0$, $|u_2(0)/u_1(0)|$, as a function of the frequency of the input acoustic wave for a number of fixed values of the frequency and amplitude of the ac magnetic field. Typical dependences are represented in Fig. 5. As compared to the case in which the frequencies of both acoustic waves coincide, in the case of different frequencies it is necessary to give three additional parameters, namely T_{10}/τ_{10}, $\omega_{10}\tau_{10}$, and $\alpha_0=\alpha_1(\Omega=\Omega_R=0)$. In our calculations we have used $T_{10}/\tau_{10}=1$, $\omega_{10}\tau_{10} \simeq 42.8$ (such a choice corresponds to Mn_{12} acetate with $\sqrt{\tau_{10}T_{10}}=10^{-8}$s, subject to a dc magnetic field $H_0=60.6$ kOe), and $\alpha_0=10^{-2}$. One can see that (i) at fixed values of the frequency and amplitude of the ac magnetic field, the larger the difference between the frequencies of the input acoustic wave and ac magnetic field, the smaller the output amplitude of the parametrically excited acoustic wave; (ii) the larger the difference between the transition frequency and the frequency of the ac magnetic field, the smaller the maximal attainable value of the output amplitude of the parametrically excited acoustic wave. Thus, the case of equal frequencies of the acoustic waves, considered in the preceding section, is more preferable for observation of the parametric interaction of these waves.

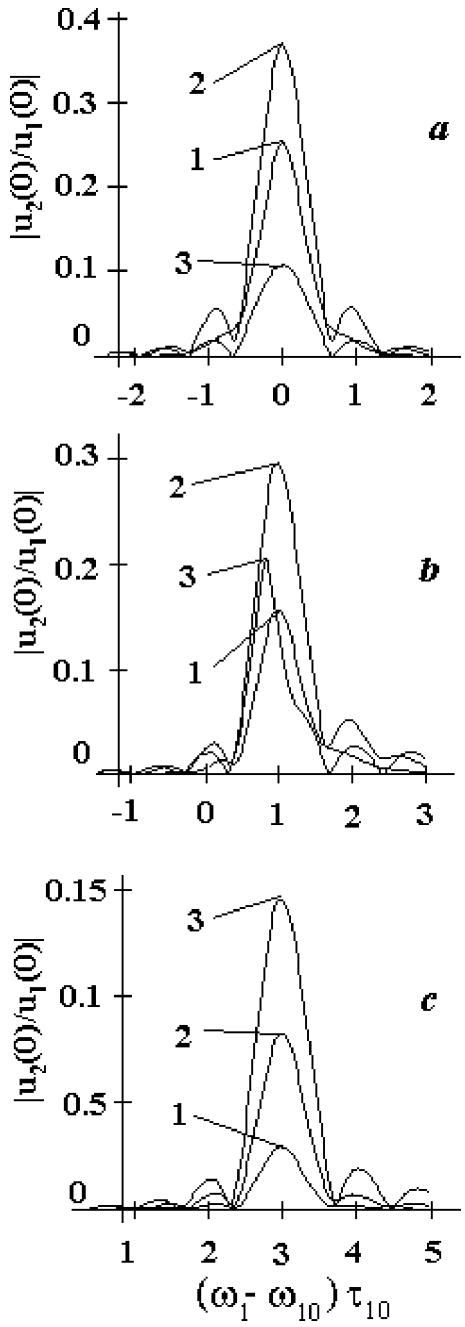

FIG. 5. The ratio of the amplitudes of the parametrically excited and input acoustic waves at the edge $z=0$ of the sample, $|u_2(0)/u_1(0)|$, as a function of the frequency of the input acoustic wave for fixed values of the frequency $[(\omega-\omega_{10})\tau_{10}=0$ (a), 1.5 (b), and 3 (c)] and amplitude $[2\Omega_R\sqrt{\tau_{10}T_{10}}=0.8$ (curves 1), 1.5 (2), and 4 (3)] of the ac magnetic field. The dimensionless length of the sample is $k_1\alpha_0 L=2$. Other parameters used are $T_{10}/\tau_{10}=1$, $\omega_{10}\tau_{10} \simeq 42.8$, and $\alpha_0=10^{-2}$.

VI. CONCLUSIONS

We have shown that two acoustic waves propagating contrarily in a crystal of molecular magnets experience parametric interaction in the presence of a strong ac magnetic field. The latter is supposed to be resonant for transitions of magnetic molecules between the states of the fundamental doublet. The frequencies of both acoustic waves are also close to the transition frequency. The parametric process can be interpreted as conversion of two electromagnetic quanta into

two phonons. If even only one acoustic wave is input into the sample and this wave is not reflected at the end of the sample, the second acoustic wave is excited due to the parametric interaction. To observe noticeable values of the output amplitude of the parametrically excited acoustic wave, it is more preferable to input into the sample the acoustic wave whose frequency coincides with the frequency of the ac magnetic field.

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