POSSIBILITY OF GENERATION AND AMPLIFICATION OF HYPERSOUND IN PARAMAGNETIC CRYSTALS

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The interaction between a nonequilibrium paramagnetic spin system and the crystal lattice is studied theoretically. Conditions under which spin-lattice interaction leads to the excitation or amplification of hypersound in crystals are studied. Parameters are introduced characterizing the efficiency of transformation of the energy of magnetic ions into the energy of hypersound. Specific calculations show that in a number of cases it is much easier to create conditions for generation of phonons than for generation of photons at the expense of the spin system energy. It is noted that nonequilibrium spin systems may be used to detect weak acoustic and electromagnetic signals.

It is well known that the methods of high-frequency acoustic spectroscopy represent a promising tool for the investigations of the properties of solids and irreversible processes taking place in them. One of the difficulties in the way of these methods is the production, transmission, and detection of acoustic vibrations with a frequency of the order of kilomegacycles and even higher. It is therefore natural to seek new means for generating hypersound directly in those materials in which its action is studied. Taking into consideration the analogy which exists between the process of photon production and phonon production at the expense of the energy of magnetic ions in crystals, it is useful to attempt to apply the theoretical and experimental material available from research on paramagnetic quantum photon generators to the development of paramagnetic hypersonic generators. The present research is devoted to the study of this possibility.

According to the theory of Al'tshuler,[1] which is supported by experimental data,[2] the creation and annihilation of phonons take place in paramagnetic crystals under the action of hypersound of frequency $\nu_{ba} = (E_b - E_a)/h \sim 10^{10}$ cps at the expense of the annihilation and creation of magnons—quanta of the Zeeman and Stark energies of the magnetic ions in a static magnetic field and in the crystalline electric field.

Let Q_t, Q_{ext} and Q_A be the figures of merit of the crystalline acoustic resonator, characterizing respectively the ordinary damping of sound in the crystal, damping by coupling with the surrounding medium, and damping as the result of the creation of the magnons. It is known that the configuration of the energy levels E_α of magnetic ions can be chosen in such a fashion that upon saturation of magnetic resonance for a certain pair (E_α, E_β), the difference in the populations Δn_{ab} for the pair (E_a, E_b) is negative. It is easy to see that in this case Q_A becomes negative and an amplification of the hypersound will take place in the crystal, described by the formula

$$G_{ba} = \frac{P_{\text{ref}}}{P_{\text{inc}}} = \left[\frac{Q_{\text{ext}}^{-1} - Q_p^{-1} - Q_A^{-1}}{Q_{\text{ext}}^{-1} + Q_p^{-1} + Q_A^{-1}}\right], \qquad (1)$$

where the subscripts "ref" and "inc" refer to the reflected and incident intensities of the hypersound. If the transition $E_a \longleftrightarrow E_b$ is forbidden for magnetic dipole transitions, and the condition

$$-Q_A^{-1} > Q_p^{-1} + Q_{\text{ext}}^{-1} \qquad (2)$$

is satisfied, the crystal will generate hypersound of frequency ν_{ba} under the action of lattice vibrations even in the absence of an external acoustic field. That is, the energy of the variable magnetic field expended in the production of $\Delta n_{ba} < 0$ is converted to the energy of hypersound.

Let

$$\mathcal{H}_1 = \cos(\omega_{ba} t)\, R \sum_{i=1}^{N} [\langle a | \mathcal{P}^i | b\rangle + \langle b | \mathcal{P}^i | a\rangle] \qquad (3)$$

be the operator describing the interaction of the magnetic ions i with the variable magnetic (R = H) and acoustic (R = A) fields, where t is the time, R is the amplitude, $R\langle a | \mathcal{P}^i | b\rangle$ is the matrix element of transition of the ion i between the states $|a\rangle$ and $|b\rangle$ under the action of the perturbation $\mathcal{H}_1$. We introduce the imaginary susceptibility and the corresponding Q by the relations

$$\chi''_R = (2\hbar V)^{-1}\,\Delta n_{ab}\,|\langle a|\mathcal{P}^i|b\rangle|^2\,g(\nu_{ba}),$$
$$Q_H = (4\pi\chi''_H\eta)^{-1}, \qquad Q_A = \omega_{ba}^{-1}Fc = (2\chi''_A\eta)^{-1}\,\rho\omega_{ba}^2, \qquad (4)$$

where η is the filling factor, F is the coefficient of sound absorption,[1] ρ is the density of the crystal, c is the velocity of sound in the crystal, $g(\nu_{ba})$ is the normalized form factor of the absorption curve of magnetic or acoustic energy of the spin system, and V is the volume of the crystal.

Let us estimate the value of Q_A for the ion Ni^{2+} in $NiSiF_6\cdot 6H_2O$ for a static field H_0 perpendicular to the symmetry axis of the crystal z. Using data on the change of the constant D of the axial crystalline field as a consequence of an applied static field X_{zz} along the z axis,[3] we obtain

$$Q_A \sim T(2S+1)\,k\rho\Delta c_z^2[2\pi\nu_{ba}N^*\alpha_{33}^2|\langle a|S_z^2|b\rangle|^2]^{-1}(\partial D/\partial X_{zz})^{-2} = 6.38\cdot 10^{11}\,T/\nu_{ba}, \qquad (5)$$

where k is Boltzmann's constant, N* is the number of Ni^{2+} ions in 1 cm^3, S is the spin, Δ is the line width of magnetic resonance, α_{33} is the elastic constant, and T is the temperature which describes the difference of populations in the levels (E_a, E_b). The following constants are used:[3]

$$c_z^2 = 2.5\cdot 10^{11}\ (\text{cm/sec})^2,\quad N^* = 4\cdot 10^{21},\quad \rho = 2.08\ \text{g/cm}^3,$$
$$\alpha_{33} = 0.5\cdot 10^{12}\ \text{dynes/cm}^2, \qquad \Delta = 3.2\cdot 10^9\ \text{cps},$$
$$\partial D/\partial X_{zz} = -3.37\cdot 10^{-26}\ \text{erg-cm}^2/\text{dyne},$$
$$|\langle a|S_z^2|b\rangle|^2 = 10^{-1};$$

The distance Ni-Ni is taken to be equal to 6.27 A. For $T = 1°$K and $\nu_{ba} = 10^{10}$ cps, we get $Q_A \sim 63.8$.

For Cr ions in $NH_4[Al_{0.99} + Cr_{0.01}]\cdot(SO_4)_2\cdot 12H_2O$, the following values are obtained:[3] $\partial D/\partial L = 8.71\times 10^{-22}$ erg-cm^2/dyne, $H_0 \parallel (111)$; in the case of the crystal $K_3[Co_{0.99} + Cr_{0.01}]\cdot(CN)_6$, one obtains $\partial D/\partial L = 1.01\times 10^{-22}$ erg-cm^2/dyne, $\partial E/\partial L = 1.22\times 10^{-23}$ erg-cm^2/dyne, $\partial g_y\beta H_0/\partial L = 1.22\times 10^{-22}\,\beta H_0$ erg-cm^2/dyne, where the y axis is parallel to the c axis of the crystal. Here L is the hydrostatic pressure on the surface of the crystal, E is the constant of a rhombic crystalline field, and β is the Bohr magneton. Substitution of these data in (5) shows that the Cr ions yield a significantly smaller value of Q_A than the Ni ions.

If the acoustic resonator is disconnected from the acoustic wave guide, then the radiation of sound in air is characterized by $Q_{ext} \sim 3\times 10^4$ (see [4]) and $Q_p \sim 5.5\times 10^4$ for $\nu = 10^9$ cps.[2] The numerical estimates that have been carried out show that the condition (2) is satisfied, and paramagnetic crystals can be used as sources of hypersound of frequency $10^{11} - 10^{12}$ cps.

As an illustration, we consider several possible schemes of operation of a generator or amplifier of hypersound, using nickel fluorosilicate with H_0 parallel to the trigonal axis z of the crystal. The spin Hamiltonian for Ni^{2+} has the form[5]

$$\mathcal{H} = \beta(g_zH_zS_z + g_xH_xS_x + g_yH_yS_y) + D(S_z^2 - \tfrac{2}{3}),$$

where S = 1, g_α are the spectroscopic splitting factors along the α axis, $D = -0.12$ cm^{-1}, and $g_z = 2.29$ at 14°K. For $H_0 \parallel z$ the position of the energy levels (E_1, E_2, E_3), corresponding to the states $(|-1\rangle, |0\rangle, |+1\rangle)$, is described by the formulas

$$E_1 = \tfrac{1}{3}D - g_z\beta H_z,\ E_2 = -\tfrac{2}{3}D,\ E_3 = \tfrac{1}{3}D + g_z\beta H_z.$$

Since $\langle 1|S_x|-1\rangle = 0$, it is convenient to use the pair of levels (E_1, E_2) for generation and amplification of hypersound. The negative difference in populations between these levels can be obtained by different means. Let $E_1 < E_3 < E_2$, and the pulsed variable magnetic field of frequency $\nu_{kl} = h^{-1}(E_l - E_k)$ and amplitude H be circularly polarized in the xy plane, where the duration of the pulse $\Delta t \ll T_1, T_2$ satisfies the condition $2g_x\beta H\hbar^{-1}\Delta t = \pi$, where T_1 and T_2 are the longitudinal and transverse relaxation times of the spin-system magnetization. To obtain a maximum difference in population between the levels (E_1, E_3), alternation of two pulses with carrier frequencies ν_{21} and ν_{23}, respectively, is necessary. If $E_1 < E_2 < E_3$, three pulses are required with carrier frequencies ν_{21}, ν_{32}, and ν_{21}. Operation of the generator in the continuous state is possible if the difference of populations of the pair of levels (E_1, E_3) becomes negative in saturation of the transitions $|1\rangle \longleftrightarrow |0\rangle$ or $|-1\rangle \longleftrightarrow |0\rangle$.

Let us consider interference effects brought about by interaction of the fields H and A for $\Delta n_{ab} < 0$. Let the relation obtained from (2) by replacing Q_A by Q_H be satisfied for the field H in the resonator. If sound vibrations are excited in a crystal which fills the resonator, then additional losses appear which are brought about by creation of phonons at the expense of the annihilation of magnons, and it is necessary to replace Q_H^{-1} by $Q_H^{-1} - Q_{HA}^{-1}$, which leads to a violation of the condition for cascade production of photons at the expense of $\Delta n_{ab} < 0$, and to a sharp change in the amplitude H. Conversely, cascade production of phonons for $\Delta n_{ab} < 0$ can be stopped as a result of creation of photons from magnons, wherein one must replace Q_A^{-1} in (2) by $Q_A^{-1} - Q_{AH}^{-1}$. Here

$$Q_{HA} = [4\pi\chi''_A A^2]^{-1}H^2, \qquad Q_{AH} = [2\chi''_H H^2]^{-1}\rho\omega_{ba}^2 A^2.$$

The relations that have been set down show that the method of double magnetic hypersonic resonance in the presence of a strong variable magnetic field (which brings about inversion of the populations, $\Delta n_{ab} < 0$) can be used for detection of a weak amplitude A (or H) without A (or H) going beyond the limits of the crystal.

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