Self-action of a hypersonic wave in acoustic paramagnetic resonance

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The behavior of a paramagnetic system formed by impurity centers in the APR region is studied over a broad range of hypersonic intensities at a frequency of 9.4 GHz. The dependence of the resonance absorption coefficient and the velocity dispersion of the hypersound on the intensity are measured for impurity paramagnetic ions possessing electron-phonon coupling in corundum. It is found that the saturation of APR for Fe^{2+} and Ni^{3+} ions with large electron-phonon coupling results in considerable deformation of the resonance curve and in the appearance of negative resonance absorption in the $H < H_R$ region. The effects are considered from the viewpoint of the nonlinear theory of propagation of a hypersonic-wave beam in a resonant medium. It is shown that the negative absorption is the result of resonant self-channeling of the hypersonic wave, which arises as a consequence of the nonlinear dispersion of the velocity of sound in acoustic paramagnetic resonance.

INTRODUCTION

The electron-phonon interaction of an impurity paramagnetic atom implanted in the lattice of a diamagnetic crystal materially affects the resonance properties of this crystal in electromagnetic and acoustic fields and has been the object of numerous investigations over the course of the years. The definite progress in the employment of hypersonic waves in problems of solid-state physics that has been achieved in recent years allows us to use for the study of this interaction a direct method that is based on the measurement of acoustic paramagnetic resonance. As a consequence of the fact that the absorption of hypersound in acoustic paramagnetic resonance (APR) is a measure of the coupling of a phonon with a definite wave vector and frequency with the electron system of the impurity center, the APR method makes it possible to obtain direct and detailed information on the electron-phonon interaction, and also on the structure of the electron energy spectrum, with which the interaction is connected.

In a number of researches that have been completed in this direction, linear APR has been studied at a very low level of hypersound, when it is possible to neglect the change of state of the paramagnetic system. On the other hand, a hypersonic wave of even comparatively low intensity can interact significantly with the system under resonance conditions for the case of strong electron-phonon coupling. The change in the state of the paramagnetic system that arises here turns out to have the opposite effect on the conditions of propagation of the same hypersonic wave which produced these changes (the self-action effect).

The aim of the present work was to study the behavior of an electron paramagnetic system in the field of a strong hypersonic wave in the case of APR. As was shown in [1,2], the electron-phonon interaction should lead to a dependence of the resonance absorption and sound dispersion on the intensity of the hypersound. This effect was discovered and studied in work on paramagnetic centers which possess weak and strong electron-phonon coupling. The effect of the (not previously predicted) resonance self-action of a narrow beam of a strong hypersonic wave in crystals containing paramagnetic centers with large electron-phonon coupling was also discovered.

OBJECTS AND FEATURES OF THE METHOD OF MEASUREMENT

As objects for the study of APR in a strong hypersonic field, we used crystals of corundum into which paramagnetic atoms possessing various electron-phonon couplings had been introduced isomorphically. As a weakly coupled ion, we chose Cr^{3+}, which was substituted for Al in the corundum (pink ruby). Detailed measurements of the linear APR were carried out for this ion,[3] and used to determine the complete set of components of the spin-phonon interaction tensor G_{ik}. According to these data, the modulus of the difference between the components responsible for absorption of a longitudinal wave along the second-order symmetry axis is $|G_{11}-G_{12}| = (6.2 \pm 0.2)\ cm^{-1}$, and the spin-lattice relaxation time at the temperature $T = 1.8\ ^\circ K$, is equal to $T_1 = (0.5 \pm 0.1)$ sec.

To measure the interaction of hypersound with the strongly coupled impurity centers, we selected corundum crystals which contained the ions Fe^{2+} and Ni^{3+}. The large value of the electron-phonon coupling for these ions is due to the features of the structure of the energy spectrum. The Fe^{2+} ion, whose APR was discovered and studied by us previously,[4] has the following structure of energy levels. The ground level in the free state ^5D in the case of implantation in the corundum splits, in a crystalline field of symmetry O_h, into a doublet and a lower orbital triplet. Subsequent removal of the degeneracy of the levels is determined by the spin-orbital interaction and by the axial component of the crystalline field.

As measurements of the APR spectrum have shown, the axial component is weaker than the spin-orbital interaction. As a result, it turns out that the ground level, with account of the perturbations mentioned, is a singlet, and above it, at a separation of 4.2 cm^{-1}, there are doublet levels. The levels of the doublet split in a magnetic field, and the transitions between them lead to resonance absorption of the hypersound. The states of these levels depend not only on the spin quantum numbers m_S, but also on the orbital m_l. By virtue of this, the spin-phonon interaction, which is directly connected with the orbital motion, takes place even in first-order perturbation theory and appreciably exceeds the interaction for ions in which the ground level in the cubic field is a singlet and the orbital motion is frozen, as, for example, in the case of Cr^{3+} in corundum. Estimation of the components of the tensor G_{ik} from the APR data for Fe^{2+} in corundum gives $|G_{11}-G_{12}| \approx 5\times10^2$ cm^{-1}.

The electron structure of the Jahn-Teller ion Ni^{3+} in corundum guarantees an even stronger electron-phonon interaction than in the case of Fe^{2+}. The fact is that the ground state in the cubic field of the oxygen octahedron of the corundum lattice for Ni^{3+} is the orbital doublet ^2E, which does not split in the first approximation, either under spin-orbital interaction or in a field of axial symmetry. At the same time, the levels of the doublet are very sensitive to the lower-symmetry deformations of the lattice, for example, in compression along an axis of symmetry of second order. The states which refer to levels split in a magnetic field depend, just as in the case of Fe^{2+} on the projections of the spin and orbital angular momenta, and are therefore strongly coupled to the deformations of the crystal lattice, lowering its symmetry. In particular, as APR measurements have shown, the amount of coupling for the longitudinal wave along the axis of symmetry of second order is $\geq 10^3$ cm^{-1}.

Three types of samples of corundum were used for the study of APR: samples containing Cr^{3+}, Fe^{2+}, and Ni^{3+}, respectively. In the samples with Cr^{3+} (ruby), the concentration of these ions amounted to $3.5\times10^{-2}\%$. In corundum with the iron impurity, which amounted in all to $\sim10^{-2}\%$, most of the impurity atoms replaced aluminum isomorphically in the lattice of the crystal and was in the state Fe^{3+} according to EPR data. The Fe^{2+} ions composed only a small part of the impurity atoms of iron (1–5%). In the corundum samples with nickel, the impurity atoms were also in two states: Ni^{2+} and Ni^{3+}. The concentration of Ni^{3+} from the EPR data was equal to $2\times10^{-4}\%$. All the samples of the crystals had the form of rods of circular cross section, with a diameter of 2.6 mm and length 15 mm. The crystallographic axis of symmetry of second order was oriented along the geometric axis of the rod. The ends were made optically flat and parallel (the roughness was $\sim0.1\ \mu$, departure from parallelism, 2"). For the purpose of introducing a high-intensity hypersonic wave into the corundum crystal, we used a previously developed effective method of excitation of hypersound in the form of a thin, hypersonic needle beam [4,5], the intensity of which could be varied over wide limits. For this purpose, textured films of ZnO were vacuum-evaporated on the ends of the crystals, with a sublayer of aluminum. The uhf electric field necessary for the excitation of the hypersound was concentrated with the help of a coaxial element onto a very small region of the film in the shape of a disk of diameter $2a = 0.5$ mm and height $d = 0.5\ \mu$, which played the role of an antenna in the radiation and detection of the hypersound. Since $a\lambda^{-1} \approx 2\times10^2$, the hypersonic wave excited by this transducer propagated in the crystal in the form of a slender, weakly diverging beam, which underwent multiple reflections from the ends of the crystal. The excitation of the hypersound was maintained in a pulsed regime (pulse length 1μ, repetition frequency 10 kHz) at a frequency of 9.4 GHz. The pulsed intensity I of the hypersound was determined by the pulsed uhf power fed to the transducer and by the losses on a single transduction. Since the losses in such a transducer are comparatively small (~20 dB) I can be varied over wide limits, from 50 $\mu W/cm^2$ to 10 W/cm^2. The lower limit was determined by the sensitivity of the pulse detector, and the upper limit by the uhf breakdown in the ZnO film.

We used a pulsed hypersonic spectrometer for recording the resonance absorption [5]. For the purpose of preventing nonlinear distortions of the echo signal in the detector on a change of I at its input, a constant level of the echo was established by means of an attenuator. The dispersion of the phase velocity of the hypersound in APR was measured by the interference method.[6] Hypersonic waves excited by the same uhf electromagnetic field source were propagated in two channels: the measuring crystal with paramagnetic centers and an identical reference crystal without paramagnetic impurities. The dispersion of the hypersonic velocity was determined by measuring the phase difference of the reference and the measured echo-signals in the resonance region. Significant difficulties arise in such a method, because of the comparatively narrow resonance line (20–30 Oe); these difficulties are connected with the necessity of establishing the constant magnetic field H with great accuracy at many points of the resonance region.

In this connection, we used automatic recording on an x-y recorder to obtain the field dependence of the interferometer signal with various initial (when the field H is outside the resonance region) echo-signal phase differences set up. The signals $\Phi_{\pm}(H) = \alpha \pm \Delta\varphi$ correspond to phase differences of $\pm\pi/2$ (α is the resonance absorption of the hypersonic wave, $\Delta\varphi$ the resonance phase change). It is easy to determine the resonance change in the phase velocity from the curves $\Phi_{\pm}(H)$:

$$\delta v = \frac{\Delta v}{v} = \frac{1}{2}(\Phi_+ - \Phi_-)\frac{1}{kL};$$

here k is the value of the wave vector, v the sound velocity, L the acoustic length. The sensitivity of the method, in addition to the effectiveness of the hypersonic excitation, is determined by the fluctuations of the phase difference of the reference and measured echo signals during the measurement. Since both crystals were in a helium cryostat under the same temperature conditions, these fluctuations did not exceed 0.1°, which permitted us to record changes of $\sim10^{-9}$ in the phase velocity at resonance.

EXPERIMENTAL RESULTS

The nonlinear effects of self-action for Cr^{3+} appear in the resonance absorption and dispersion of the phase velocity even at comparatively small pulsed intensities of the hypersound. Thus, while for linear APR, when $\partial\alpha/\partial I = 0$, at $H = H_R$, $T = 1.8$°K, $I' \leq -20$dB, the value of $\alpha_R = \alpha_R^0 = 1.4$ dB, even for $I' = -9$dB ($I \approx 10^{-4}$ W/cm^2) we have $\alpha_R < \alpha_R^0 = 2\alpha_R(I_s)$ (Fig. 1). The value of I' in decibels is the intensity of the hypersound relative to the intensity I_s at which $\alpha_R = \frac{1}{2}\alpha_R^0$.

The decrease in α with increasing I is accompanied by a decrease in the dispersion of the phase velocity; however, the dependence $\delta v(I)$ is much weaker than $\alpha(I)$ (Fig. 2). The change in the shape and width of the resonance line at saturation, which is not observed in the recording of $\alpha(H)$, is clearly seen from the curves of $\Phi_{\pm}(H)$. While at low intensity the distance between extremal points of the phase velocity $(\delta v)_e$ amounts to 30 Oe and corresponds to the width of the resonance line in the absence of saturation, this separation is tripled at $I' = +17$ dB. It is seen from Fig. 2 that in ad-

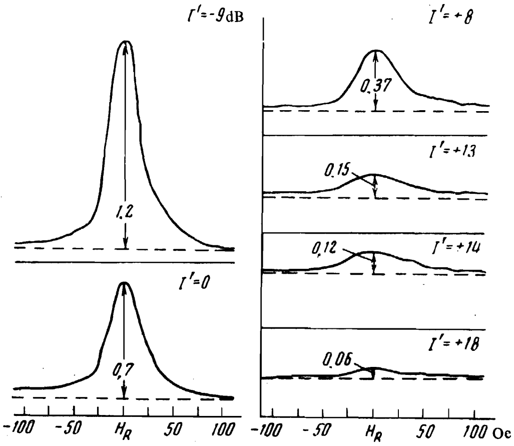


FIG. 1. APR curves α(H) of Cr^{3+} ions at different hypersonic intensities. The maximum values of the resonance absorption here and below in Figs. 3 and 5 are given in dB. I_s = 1 mW/cm², α_0 = 2.3 dB/cm, H_R = 0.45 kOe, L = 6 cm, T = 1.8°K. The figures with arrows indicate the value of the resonance absorption at the maximum of the curve, measured from the level of nonresonance absorption (the dashed line).

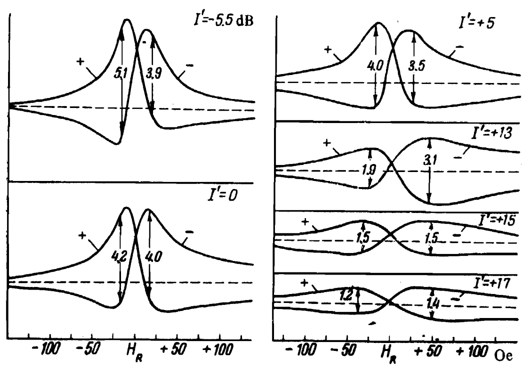


FIG. 2. $\Phi_{\pm}(H)$ resonance curves for Cr^{3+} at different hypersonic intensities. The arrows indicate the extremal values of the relative change of the phase velocity of the hypersound at resonance: $\delta v \times 10^7$; L = 6 cm.

dition to the change in the width, the resonance curve is deformed at saturation; this is manifested in the different dependences of $(\delta v)_e$ on I for $H < H_R$ and $H > H_R$.

The effect of self-action takes on a different character in the case of APR of Fe^{2+}, which possesses strong electron-phonon coupling. Upon an increase in I, just as in the case of Cr^{3+}, α decreases monotonically, reaching one-half its value at $I = I_s \approx 2 mW/cm^2$ (Fig. 3). The shape and the width of the resonance line α(H) remain unchanged here, which indicates an inhomogeneous broadening of the resonance line. However, for a hypersonic wave of high intensity, $I > I_s$, the character of the resonance absorption changes materially. With increasing I, there is at first a deformation of the resonance curve α(H). The curve is deformed to the highest degree in the region $H < H_R$ and the absorption maximum is shifted toward higher magnetic fields $H > H_R$. When I reaches I_n, α vanishes at $H < H_R$ and further increase in I leads to negative resonance absorption. The total absorption α_σ turns out to be less here than the absorption outside the resonance region—the nonresonance absorption α_0 (the absorption α_0 corresponds to the echo signal level indicated in Figs. 1–5 by the dashed lines). The effects of saturation and negative absorption were observed for hypersonic echo signals with different numbers, corresponding to one, two and three traversals of the crystal by the hypersound. Here the quantity I_s did not depend on the number of the echo signal, and the negative absorption at fixed input intensity of the hypersound was found to be proportional to it. Thus, for first, second and third echo signals, the negative absorptions amounted to 0.2 dB, 0.42 dB and 0.65 dB, respectively. It should be noted that the resonance absorption at $H = H_R$ tends monotonically to zero with increase in I.

In contrast to the resonance absorption, the change in the phase velocity in the resonance region has a monotonic character over the entire measured range of hypersonic intensities. The resonance change in the phase

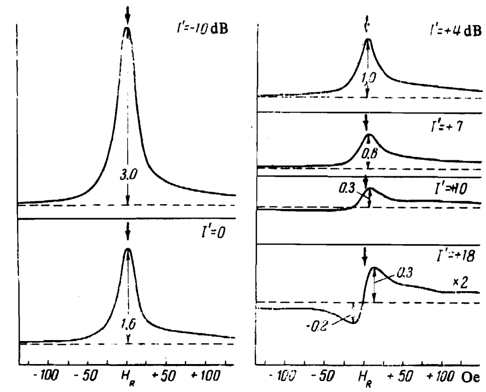


FIG. 3. APR curves α(H) of Fe^{2+} ions at various hypersonic intensities. T = 4.2°K, $I_s \approx$ 2 mW/cm², α_0 = 2.8 dB/cm², H_R = 0.98 kOe, L = 3 cm. The heavy arrows indicate the resonant positions ($H = H_R$).

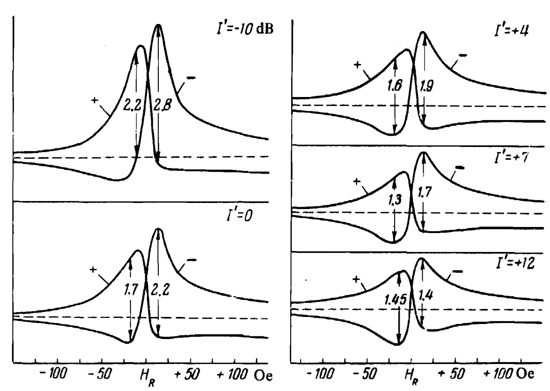
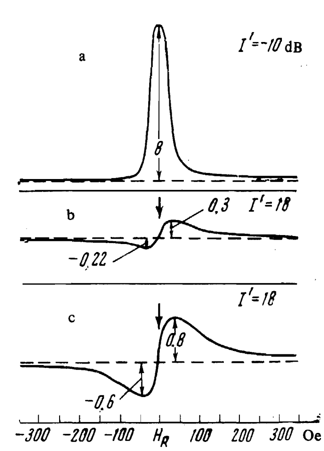


FIG. 4. Resonance curves of $\Phi_{\pm}(H)$ for Fe^{2+} at various hypersonic intensities. The arrows indicate the extremal values of the relative phase velocity change $\delta v \times 10^6$; L = 3 cm.

FIG. 5. APR curves for Ni^{3+} at various hypersonic intensities. The notation is the same as in Fig. 3. T = 4.2°K, I_s = 10 mW/cm², α_0 = 3 dB/cm⁻¹, H_R = 3.4 kOe; a, b_ first echo (L = 3 cm), c – third echo (L = 9 cm).



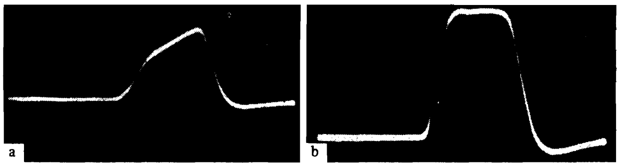


FIG. 6. Oscillogram of the echo in the propagation of hypersound in corundum with Ni^{3+} at $I = I_S$, a – $H = H_R$, b – H outside of resonance. Time scale: 1 cm 1 μsec.

velocity decreases with increasing I (Fig. 4), but this change, as in the case of Cr^{3+}, is much less than the change in α_R.

Saturation and negative absorption were also observed for the Ni^{3+} ion. The character of the saturation of the resonance line for this ion is the same as for Fe^{2+}. A decrease in α is observed on an increase in I, as well as a subsequent deformation of the resonance curve. Only the values of I_S and I_n at which nonlinear effects appear are different. The negative absorption, which appears at $I > I_n$, reaches 0.22 dB for the first echo signal (Fig. 5). Just as for the Fe^{2+} ion, this absorption increases with the number of the echo. A change in the shape of the echo at saturation has also been observed for Ni^{3+}. When $I = I_S$, the change is greatest. Here the leading edge of the echo is absorbed much more strongly than the trailing edge (Fig. 6). The distortion of the shape of the pulse increases with the number of the echo.

The pulse method of APR measurement also allows us to estimate the spin-lattice relaxation times T_1 for ions with strong electron-phonon coupling from the dependence of I_S on the repetition frequency of the pulses. These times turned out to be $\leq 10^{-6}$ sec (4.2 °K) and $\sim 2\times 10^{-5}$ sec (4.2 °K) for Fe^{2+} and Ni^{3+}, respectively.

DISCUSSION OF THE RESULTS OF THE MEASUREMENTS. THE EFFECT OF SELF-CHANNELING OF THE HYPERSONIC BEAM

The monotonic decrease in the resonance absorption and velocity dispersion with increase in the intensity of the hypersonic wave, which is observed for the Cr^{3+} ion with weak spin-phonon coupling, is in qualitative agreement with the nonlinear theory of APR saturation.[1] According to this theory, the saturation of resonance absorption in the case of inhomogeneous broadening of the resonance line with increase in I should take place much more rapidly than the saturation of the resonance dispersion of the phase velocity, which is also observed experimentally.

A different situation exists for ions with strong electron-phonon coupling, Fe^{2+} and Ni^{3+}. The effects of deformation of the resonance curve and negative absorption in the range $H < H_R$ discovered here do not follow from this theory. It is impossible to regard the negative absorption as amplification of the hypersound, since in this case the states of the levels of the resonance transition are not inverted, because of the absence of an additional-illumination source of short duration. Nor, as a consequence of the very short time of the spin-lattice relaxation, can inversion arise on slow crossing of the resonance by the magnetic field during the measurement. This, and also the fact that the character of the negative absorption—its value and its position to the left of the resonance magnetic field, $H < H_R$—is the same for the ions Fe^{2+} and Ni^{3+}, which are essentially different in their energy spectra and in their values of G_{ik} and T_1, leads to the idea that the negative absorption is connected with changes, during the growth of I, not only in the state of the paramagnetic system, but also in the structure of the hypersonic field in the paramagnetic crystal, as a result of which the losses in the propagation of the hypersonic wave decreases. Inasmuch as the interaction with a paramagnetic system of only a one-dimensional plane wave in an isotropic medium has been considered in the nonlinear theory of saturation of APR, these effects have naturally escaped from the field of view.

In view of this, we consider the nonlinear resonance self-actions, to which the spatial boundedness of the hypersonic wave in the direction perpendicular to its propagation leads. Let a beam of longitudinal hypersound be propagated in the x direction in a resonant medium. The beam is excited on the boundary of the medium by a radiator whose radius a satisfies the inequality

$$\lambda \ll a \ll L_0,\ M_0, \tag{1}$$

where λ is the wavelength of the hypersound and L_0 and M_0 are the dimensions of the sample along and perpendicular to the direction of propagation of the wave.

For simplicity, we shall not take the anisotropy of the medium into account, and we shall take the effective spin for the paramagnetic atom to be $S' = \frac{1}{2}$. The hypersound interacts with the component $\langle S_x \rangle$—the mean value of the spin projection operator on the x axis. In this case, as is well known, the relaxation processes can be described satisfactorily by means of the two characteristic times of longitudinal and transverse relaxation, T_1 and T_2. The system of joint equations for the propagation of hypersound in a resonant medium contains the equation of elasticity and the equation for $\langle S_j \rangle$ that is connected with it. This equation describes the states of the impurity atom in the sound field:

$$\frac{\partial^2 u}{\partial t^2} = v^2 \nabla^2 u + \frac{N}{\rho_0} \int \frac{\partial F(\langle S_j' \rangle)}{\partial x} g(\omega_0)\, d\omega_0, \tag{2}$$

$$\frac{d}{dt} \langle S_j' \rangle = -\frac{i}{\hbar} \langle [S_j', H_e'] \rangle + R(\langle S_j' \rangle); \tag{3}$$

Here

$$H_e' = \hbar \Delta\omega S_z + H_s, \quad R(\langle S_z \rangle) = (\langle S_z \rangle - S_z^0) T_1^{-1},$$
$$R(\langle S'_{x,y} \rangle) = \langle S'_{x,y} \rangle T_2^{-1}, \quad j = x, y, z.$$

ρ_0 is the density of the medium, u the deformation in the elastic wave, F the force connected with the electron-phonon interaction for an ion with resonance frequency ω_0, N the concentration of paramagnetic ions, and $g(\omega_0)$ the shape function of the resonance line, S′ denotes the fact that the spin projection operator is considered in a system of coordinates which is rotating with frequency ω (ω is the frequency of oscillation in the sound field) and moving with the wave along the x axis, H_e' is the effective Hamiltonian in this system of coordinates, $S_z^0 = \frac{1}{2} \tanh[\hbar\omega_0 / 2k_B T]$, H_S is the Hamiltonian of the electron-phonon interaction, $\Delta\omega = \omega_0 - \omega$, k_B is Boltzmann's constant, and S_z^0 the mean value of the operator S_z at thermodynamic equilibrium. The integral in (2) is taken over the resonance region.

In the study of the propagation of a hypersonic beam in a resonant medium, we shall follow the quasi-optical method employed in [7] for analysis of self-focusing of light in a nonlinear medium. We write down the deformation in the sound field in the form

$$u = \mathrm{Re}\, R(\mu x, \mu^{1/2} r, \mu t) \exp\{i[\omega t - kx - kS(\mu x, \mu^{1/2} r, \mu t)]\}. \quad (4)$$

The amplitude of the wave R and the eikonal S change slightly over the period of oscillation and over a distance of the order of the sound wavelength; μ is a small parameter, by means of which we take into account the different dependences of these functions on the time and the coordinates, along the beam x and in the transverse direction r.

Since $S' = 1/2$, F is a linear function of $\langle S_j' \rangle$. Substituting (4) in the system (2), (3), and neglecting terms of the order $O(\mu^2)$, we obtain equations which describe the propagation of the hypersonic beam in the resonant medium:

$$\frac{1}{v}\frac{\partial R}{\partial t} + \frac{\partial R}{\partial x} + \frac{1}{a^2}\frac{\partial R}{\partial r'}\frac{\partial S}{\partial r'} + \frac{R}{2a^2}\left(\frac{1}{r'}\frac{\partial S}{\partial r'} + \frac{\partial^2 S}{\partial r'^2}\right) + I_R(R) = 0, \quad r = ar', \quad (5)$$

$$\frac{1}{v}\frac{\partial S}{\partial t} + \frac{\partial S}{\partial x} + \frac{1}{2a^2}\left(\frac{\partial S}{\partial r'}\right)^2 = \frac{1}{2Rk^2a^2}\left(\frac{1}{r'}\frac{\partial R}{\partial r'} + \frac{\partial^2 R}{\partial r'^2}\right) + I_s(R), \quad (6)$$

where $I_R(R)$ and $I_S(R)$ are nonlinear functions of R which are expressed in terms of the characteristics of the paramagnetic system:[2]

$$I_R(R) = -\frac{GNk}{2\rho_0 v^2}\int (\langle S_x' \rangle \cos kS - \langle S_y' \rangle \sin kS)\, g(\omega_0)\, d\omega_0, \quad (7)$$

$$I_s(R) = +\frac{GN}{2R\rho_0 v^2}\int (\langle S_y' \rangle \cos kS + \langle S_x' \rangle \sin kS)\, g(\omega_0)\, d\omega_0. \quad (8)$$

Here G is the spin-phonon coupling constant.

Under stationary conditions, we find for the case of a Lorentzian shape of the inhomogeneously broadened resonance line from (3), (7) and (8)

$$\begin{gathered} g^{-1}(\omega_0) = \pi\delta_L(\varepsilon^2+1), \quad I_R(R) = RQkF_R(z)\delta_L^{-1}, \quad I_s(R) = QF_s(z)\delta_L^{-1}, \\ z = T_1T_2G^2k^2R^2\hbar^{-2}, \quad \gamma = (T_2\delta_L)^{-1}, \quad Q = S_z^0 NG^2(4\rho_0 v^2\hbar)^{-1}, \\ F_R(z) = [1+\gamma(1+z)^{1/2}](1+z)^{1/2}\{\varepsilon^2 + [1+\gamma(1+z)^{1/2}]^2\}^{-1}, \\ F_s(z) = \varepsilon\{\varepsilon^2 + [1+\gamma(1+z)^{1/2}]^2\}^{-1}, \quad \varepsilon = \Delta\omega\delta_L^{-1}, \ \gamma \ll 1, \end{gathered} \quad (9)$$

here δ_L is the inhomogeneous line width. When the sound intensity is small ($z \ll 1$), the nonlinear dependence of $I_R(R)$ and $I_S(R)$ on R can be neglected. Equations (5), (6) describe the linear effects of resonance absorption and the dispersion of the phase velocity, in which no limitation appears on the wave in the transverse direction.

We consider the case of high intensity, when $z \gg 1$, but $\gamma z^{1/2} < 1$. The solution of the nonlinear equations (5) and (6) under the conditions of stationarity will be sought in the quasi-optical approximation in the form of a beam with variable radius of curvature:[7]

$$S = \frac{1}{2}a^2 r'^2 \beta(x) + \varphi(x), \quad R^2 = \frac{R_0^2}{f^2(x)}\exp\left\{-\frac{2r'^2}{f^2(x)} - \alpha x\right\} \quad (10)$$

The boundary conditions at the entrance of the beam to the crystal have the form

$$\varphi(0) = 0, \quad f(0) = 1, \quad R = R_0 \exp(-r'^2), \quad (11)$$

where $\beta^{-1}(x)$ is the radius of curvature, $\varphi(x)$ the contribution to the eikonal due to change in the velocity, f(x) the dimensionless beam width, and R_0 the amplitude of the wave on the beam axis at $x = 0$.

Substitution of (10) in (5), (6), and (9) leads to the following equations for the functions $\beta(x)$, $\varphi(x)$, f(x), and α in the paraxial region of the beam ($r' \ll 1$):

$$\begin{gathered} \beta = f'f^{-1}, \quad \alpha = 2I_R(R)R^{-1}, \\ \varphi' = -\frac{1}{k^2a^2}\left(\frac{4}{f^2} + \frac{x}{r'}\frac{\partial\alpha}{\partial r'} + \frac{\partial^2\alpha}{\partial r'^2}\frac{x}{2}\right) + I_s, \end{gathered} \quad (12)$$

$$f''f^{-1} = \frac{1}{k^2a^4}\left[\frac{4}{f^2} + \frac{x^2}{4r'^2}\left(\frac{\partial\alpha}{\partial r'}\right)^2 + \frac{2x}{f^2 r'}\frac{\partial\alpha}{\partial r'}\right] + \frac{1}{a^2}\frac{\partial^2 I_s(R)}{\partial r'^2}. \quad (13)$$

The first term in the square brackets of (13) describes the diffraction spreading of the beam; the second and third terms, which are connected with the resonance absorption, also increase the beam width. The role of the term $a^{-2}\partial^2 I_S(R)/\partial r'^2$, to which the resonance dispersion of the phase velocity of the hypersound leads is different. Since $I_S(R) \sim \Delta\omega$, the velocity term vanishes at resonance and the beam diverges because of diffraction and resonance absorption. However, it is significant that the sign of this term changes on the transition through resonance. Therefore, on one side of resonance, for $\Delta\omega > 0$, it increases the divergence, but on the other side, $\Delta\omega < 0$, the sign of this term is negative, which leads to a decrease in the divergence of the beam.

We now estimate the value of the parameter $z = z_C$, at which one can in this case completely compensate the divergence of the beam and obtain the effect of self-channeling of the hypersonic wave. When $z \gg 1$, $\gamma z^{1/2} < 1$, $|\Delta\omega| = \delta_L$, it follows from (9) that the second and third terms in (13), which are associated with resonance absorption, are proportional to z^{-1} and $z^{-1/2}$, respectively, and the velocity term $\sim z^{1/2}$ and appreciably exceeds these terms. Using this fact, and neglecting small terms, we obtain the first integral of Eq. (13) in the form

$$f'^2 = \frac{2}{a^2}\left(1 - \frac{1}{f}\right)\left[\left(\frac{1}{f} + 1\right)\frac{1}{(ka)^2} - \gamma\frac{\Delta v}{v}(z')^{1/2}\right] \quad (14)$$

where $z' = z|_{R=R_0}$, $\Delta v/v = I_S(R_0)$ and the boundary conditions (11) are taken into account. We then find

$$z_{c'} = \frac{4}{(ka)^4\gamma^2(\Delta v/v)^2}. \quad (15)$$

If $z' > z_C'$, then the focusing action of the velocity dispersion exceeds the effect of diffraction and the beam decreases in cross-section—the effect of self-focusing. It must be noted that, in contrast to the effects of self-channeling and self-focusing of light in a nonresonant, nonlinear medium, the power of the beam that corresponds to z_C depends on a.

If the sound is propagated in a resonant medium in the form of a short pulse, then the appearance of self-channeling is complicated by transient processes. In this case, the replacement of t by the new variable $\zeta = t - xv^{-1}$ leads to a system of equations which has the same form as (5) and (6) in the stationary regime.[7] However, the equations for $\langle S_j' \rangle$, which describe the transient process in a spin system under the effect of a hypersonic pulse, change in this case. As analysis shows, in this case, the character of the process depends on the relation of the times T_1, T_2, τ_p and τ (τ_p and τ are respectively the pulse length and the time of its propagation in the crystal).

When τ and $\tau_p \gg T_2$ and T_1, the transient processes settle within the time τ_p and the self-channeling is the same as in the stationary regime. This case occurs for the Fe^{2+} ion and, consequently, the stationary solution obtained above is applicable to it. An increase in the sound intensity at resonance ($\Delta\omega = 0$) leads to a monotonic decrease of α to zero, since the velocity term in the right side of (13) is absent. Upon deviation from resonance due to this term, the resonance absorption decreases when $H < H_R$ and increases when $H > H_R$. This produces deformation of the resonance curve, shifting the absorption maximum in the direction $H > H_R$. For $z' \geq z_C'$, self-channeling develops, being manifested in the disappearance of diffraction losses and the corresponding decrease in the total nonresonance absorption, which is registered in the form of negative absorption.

For Fe^{2+}, the estimate gives: at $G = 5\times10^2$ cm^{-1}, $T_1 = T_2 = 10^{-7}$ sec, $\gamma = 0.2$, $\Delta v/v = 1.4\times10^{-6}$, $a = 2.5\times10^{-2}$ cm, and $k = 5.4\times10^4$ cm^{-1}, we have $z_c' \approx 30$, $I_c = \frac{1}{2}\rho_0 v^3 z_c' \times (T_1T_2G^2)^{-1} h2 \approx 0.1$ W/cm^2. The value of I_c agrees in order of magnitude with the measured value, and the value of the negative absorption corresponds to the diffraction losses in the propagation of the sound.[8]

For Ni^{3+}, in spite of the fact that $\tau \approx T_1$, the character of the saturation process and the self-channeling are the same as in the case of Fe^{2+}, since the intense hypersonic pulse manages to change the state of the paramagnetic system appreciably in the time τ_p because of the large value of the electron-phonon coupling. By virtue of the fact that $\tau_p < T_1$, saturation of APR is accompanied by a transient process, in which the leading front of the pulse, being absorbed, saturates the paramagnetic system. As a result, the shape of the echo is distorted.

In the case with the Cr^{3+} ion, which possesses weak spin-phonon interaction and a long time T_1, the saturation of the paramagnetic system is essentially different. The process of multiple weak actions of short pulses on a system with a long time T_1 is decisive. Since $\tau_p \ll T_1$, the change in the population of the levels for propagation of the pulse in the crystal is determined by the energy of the pulse $I\tau_p$ [9]. In our case, even for $I = I_c$ (I_c for Fe^{2+}), the energy of the pulse $I\tau_p \ll E_s$ (E_s is the saturation energy [9]). Therefore a single passage of the pulse of hypersound through the crystal is accompanied by a very small change in the state of the paramagnetic system and cannot lead to self-channeling. The observed decrease of α and δv with increasing I is due to the high frequency of repetition of the pulses. As a consequence of the fact that the repetition frequency $\nu_p \gg T_1^{-1}$, the small changes in the state of the paramagnetic system are accumulated and lead to a decrease in the resonance absorption and dispersion of the hypersound velocity.

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