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Interference nuclear acoustic resonance in crystals with a cooperative Jahn-Teller effect

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The absorption by nuclear spins of an ultrasonic wave whose direction of propagation and polarization correspond to the cooperative Jahn-Teller macrodeformation effect is investigated in crystals of the $\mathrm{TmVO_4}$ type. A dispersion equation that describes the coupling between electron-phonon modes and nuclear spins due to the hyperfine interaction of the dipole type is obtained by the Green's function method. It is shown that resonance absorption occurs in the case $T < T_c$, with an intensity proportional to $H_x^2 H_z^2$ (H_z is the projection of the magnetic field vector on the crystallographic axis c). The resonance is of an interference type. Interference of the absorption contributions of the quasi-nuclear and quasi-phonon spectrum branches produces an asymmetry of the resonance curve.

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The method of acoustic nuclear resonance (ANR) was successfully applied to the study of the dynamic interaction of atomic nuclei with internal fields in crystals.[1–3] One of the forms of such interaction is the dipole interaction of the magnetic moment of the nucleus with the local magnetic field of a paramagnetic ion, modulated by lattice vibrations. In crystals with the cooperative Jahn–Teller effect (CJTE),[4,5] the indicated hyperfine interaction, together with the electron–phonon interaction, leads to the coupling of the subsystem of nuclear spins with the electron and phonon subsystems. Since the interaction of the electron states with the lattice is appreciable in Jahn–Teller crystals as a consequence of orbital degeneracy of the ground state of the ions, the nuclear spin–phonon coupling caused by the electrons can be very effective. For this reason, it turns out to be possible to obtain information on the dynamic electron–phonon coupling by ultrasonic studies of the subsystem of nuclear spins in crystals with CJTE, as well as by changes in the state of the electron subsystem due to structural phase transitions that are characteristic of Jahn–Teller crystals. Moreover, crystals with a fundamental non-Kramers doublet of Jahn–Teller ions are characterized by a peculiar combination of elastic and magnetic properties, as a consequence of the mutual suppression of distortion of the crystal lattice and the magnetic moment, caused by the external (or internal) magnetic field.[6,7] The mutual suppression of the structural and magnetic orderings leads to singularities of the temperature and field dependence of the nuclear absorption of ultrasound, caused by the hyperfine interaction.

So far, CJTE has been most widely investigated in the rare-earth vanadates, arsenates and phosphates.[5] In the present paper we consider sound absorption by nuclei in crystals of the $\mathrm{TmVO_4}$ type (local symmetry D_{4h}, the ground state of the Tm^{3+} ion is a non-Kramers doublet, and the nuclear spin of Tm^{169} is $I=\frac{1}{2}$).

1. The interaction operator of the electrons with the lattice vibrations in such crystals is of the form

$$\mathscr{H}' = \sum_{m\varkappa} V_{m\varkappa}(b_\varkappa^+ + b_\varkappa)\sigma_z^m,$$

where σ_z^m is the electron operator on functions of the fundamental doublet, b is the operator of the phonon field, m enumerates the sites, and $\varkappa$ is the wave vector and branch of phonons. Since only the z component of the orbital momentum is conserved in systems with symmetry D_{4h}, the hyperfine dipole interaction of the magnetic moments of electrons and nuclei at a single site can be written in the form

$$\mathscr{H}'' = -B\sum_{m} I_z^m \sigma_y^m.$$

For this same reason, only one term ($\sim H_z$) is retained in the Hamiltonian $\mathscr{H}'''$ of the electron Zeeman interaction:

$$\mathscr{H}'''=-\frac{1}{2}g_e\beta_e H_z\sum_m \sigma_y^m.$$

Assuming that the y component of the magnetic field $\mathbf{H}$ is zero, i.e., $\mathbf{H}=(H_x, 0, H_z)$, the Hamiltonian of the crystal can be written in the following form:

$$\mathscr{H}=\mathscr{H}'+\mathscr{H}''+\mathscr{H}'''+\frac{1}{2}g_{\text{я}}\beta_{\text{я}}\sum_m (H_xI_x^m+H_zI_z^m)+\sum_\varkappa \hbar\omega_\varkappa\left(b_\varkappa^+b_\varkappa+\frac{1}{2}\right)$$
$$+\frac{1}{2}c\Omega U_z^2-PU_z-g_0(c\Omega/N)^{1/2}U_z\sum_m \sigma_z^m, \qquad (1)$$

where I_α are operators of the components of the nuclear angular momentum $I=\frac{1}{2}$. The last three terms of the Hamiltonian (1) describe the energy of homogeneous strain U_z of the crystal and the interactions of this strain with the external uniaxial pressure and the electrons, respectively.

For further calculations, it is convenient to perform a shift transformation on the Hamiltonian (1)[6]:

$$\tilde{\mathscr{H}}=\exp(iR)\mathscr{H}\exp(-iR), \quad R=\sum_m f_m\sigma_z^m,$$
$$f_m=i\sum_\varkappa V_{m\varkappa}(b_\varkappa^+-b_\varkappa)(\hbar\omega_\varkappa)^{-1},$$

as a result of which it takes the form

$$\tilde{\mathscr{H}}=-\frac{1}{2}g_{\text{я}}\beta_{\text{я}}\sum_m (H_xI_x^m+H_zI_z^m)+\frac{1}{2}c\Omega U_z^2-g_0(c\Omega/N)^{1/2}U_z\sum_m \sigma_z^m$$
$$-PU_z+\sum_\varkappa \hbar\omega_\varkappa\left(b_\varkappa^+b_\varkappa+\frac{1}{2}\right)-\sum_{mn\varkappa} V_{m\varkappa}V_{n\varkappa}(\hbar\omega_\varkappa)^{-1}\sigma_z^m\sigma_z^n$$
$$-\sum_m (\sigma_y^m\cos 2f_m+\sigma_x^m\sin 2f_m)\left(BI_z^m+\frac{1}{2}g_e\beta_eH_z\right). \qquad (2)$$

It follows from (2) that allowance for the hyperfine interaction leads to additional coupling (besides that due to the magnetic field H_z) between the electron and phonon subsystems. At $H_z=0$, the parameter of dynamic electron–phonon coupling is proportional to the constant of hyperfine interaction B. Moreover, this interaction, together with the electron–phonon interaction, is responsible for the nuclear–phonon coupling; therefore sound absorption by the nuclei does not occur in its absence. It is important to note that the last term in the Hamiltonian, which describes the coupling of the nuclear spins with the electron–photon subsystem, commutes with the operator of the Zeeman nuclear interaction at $H_x=0$ and, consequently, sound absorption by the nuclei does not take place here.

We consider the propagation, in the crystal, of a transverse ultrasonic wave whose direction of propagation and polarization correspond to the crystal macrodeformation observed in the structural phase transition. The absorption of the sound wave in the case of a linear response is determined by the imaginary part of the elastic susceptibility tensor

$$\chi(\omega)=-2\pi\sum_q G_q^z(\omega)|V_q|^2,$$

where $G_q^z(\omega)$ is the Fourier transform of the equal-time Green's function G_{mn}^α in the representation of the wave vector q ($G_{mn}^\alpha=\langle\langle\sigma_\alpha^m(t)|\sigma_z^n(0)\rangle\rangle$). The equation for the Green's function G_{mn}^α can be written in the form

$$i\frac{dG_{mm'}^x}{dt}=-2i\delta_{mm'}\delta(t)\bar{\sigma}_y+2i\Delta_0G_{mm'}^y$$
$$+4i\bar{\sigma}_y\sum_{n\varkappa} V_{m\varkappa}V_{n\varkappa}(\hbar\omega_\varkappa)^{-1}G_{nm'}^z-iC\gamma G_{mm'}^z-i\bar{\sigma}_zC\Gamma_{mm'}^c-2iB\gamma R_{mm'}, \qquad (3)$$
$$i\frac{dG_{mm'}^y}{dt}=-2i\Delta_0G_{mm'}^x+i\bar{\sigma}_zC\Gamma_{mm'}^s,$$
$$i\frac{dG_{mm'}^z}{dt}=i\gamma CG_{mm'}^x-i\bar{\sigma}_yC\Gamma_{mm'}^s,$$

where $R_{mm'}^z=\langle\langle I_z^m|\sigma_z^{m'}\rangle\rangle$ is the nuclear Green's function, and $\Delta_0=A\bar{\sigma}_z+g_0P(c\Omega N)^{-1/2}$,

$$A={\sum_n}' V_{m\varkappa}V_{n\varkappa}(\hbar\omega_\varkappa)^{-1}+g_0^2, \quad C=g_e\beta_eH_z+2B\bar{I}_z,$$
$$\Gamma_{mm'}^s=\langle\langle\sin 2f_m|\sigma_z^{m'}\rangle\rangle, \quad \Gamma_{mm'}^c=\langle\langle\cos 2f_m|\sigma_z^{m'}\rangle\rangle.$$

In Eqs. (3), we used, besides the splittings

$$\langle\langle\sigma_\alpha^mF_m(b_\varkappa^+, b_\varkappa)|\sigma_z^{m'}\rangle\rangle\to\bar{\sigma}_\alpha\langle\langle F_m(b_\varkappa^+, b_\varkappa)|\sigma_z^{m'}\rangle\rangle+\bar{F}G_{mm'}^\alpha,$$
$$\langle\langle\sigma_\alpha^m\sigma_\beta^n|\sigma_z^{m'}\rangle\rangle\to\bar{\sigma}_\alpha G_{nm'}^\beta+\bar{\sigma}_\beta G_{mm'}^\alpha,$$

given in Ref. 9, the analogous splitting of the type

$$\langle\langle\sigma_\alpha^mF_m(b_\varkappa^+, b_\varkappa)I_\beta^m|\sigma_z^{m'}\rangle\rangle\to\bar{\sigma}_\alpha\bar{F}R_{mm'}^\beta+\bar{\sigma}_\alpha I_\beta\langle\langle F_m(b_\varkappa^+, b_\varkappa)|\sigma_z^{m'}\rangle\rangle+\bar{F}\bar{I}_\beta G_{mm'}^\beta,$$

which are valid at $\hbar\omega_D\gg g\beta\gamma H_z$, $B\gamma\bar{I}_z$, and which take into account the smallness of the energy E_n of the nuclear excitations of the crystal in comparison with the energy E_e of the electronic excitation (ω_D is the Debye frequency and $\gamma=\langle\cos 2f\rangle$ is the vibronic suppression factor). Using the very same splittings in the equations for the functions

$$\Gamma_{mm'}^s, \quad \Gamma_{mm'}^c \text{ and } \langle\langle b_\varkappa|\sigma_z^{m'}\rangle\rangle, \quad \langle\langle b_\varkappa^+|\sigma_z^{m'}\rangle\rangle,$$

and changing to the q, E representation, we obtain

$$\Gamma_q^s(E)=-\frac{4C\gamma^2\Sigma_j|V_{jq}|^2E(\hbar\omega_{jq})^{-1}(E^2-\hbar^2\omega_{jq}^2)^{-1}}{E-4C\gamma\bar{\sigma}_y\Sigma_j|V_{jq}|^2E(\hbar\omega_{jq})^{-1}(E^2-\hbar^2\omega_{jq}^2)^{-1}}G_q^x(E), \qquad (4)$$
$$\Gamma_q^c(E)=0, \quad (E\neq0), \quad \varkappa\equiv jq.$$

Finally, in this same approximation, we obtain for the "nuclear" Green's functions $R_{mm'}^\alpha$

$$i\frac{dR_{mm'}^z}{dt}=ig_{\text{я}}\beta_{\text{я}}H_xR_{mm'}^y, \quad i\frac{dR_{mm'}^x}{dt}=-iDR_{mm'}^y,$$
$$i\frac{dR_{mm'}^y}{dt}=-ig_{\text{я}}\beta_{\text{я}}H_xR_{mm'}^z+iDR_{mm'}^x+2B\gamma\bar{I}_xG_{mm'}^y+2iB\bar{\sigma}_y\Gamma_{mm'}, \qquad (5)$$
$$D=g_{\text{я}}\beta_{\text{я}}H_z+2B\gamma\bar{\sigma}_y.$$

The set of equations (3) and (5), along with (4), is a closed one and describes the dynamic behavior of the interacting nuclear, electron and phonon subsystems. For the q-transform of the sought Green's function we find, in the E representation,

$$G_q^z(E)=\pi^{-1}(E^2-E_n^2)\{(E^2-E_f^2)C\gamma\bar{\sigma}_y+4|V_q|^2\gamma^2\bar{\sigma}_y^2C^2(\hbar\omega_q)^{-1}\}$$
$$\times\{(E^2-E_n^2)[(E^2-E_e^2)(E^2-E_f^2)-4|V_q|^2E_e^2\gamma\bar{\sigma}_yC(\hbar\omega_q)^{-1}]$$
$$-4B^2\gamma^2\bar{\sigma}_z\bar{I}_xg_{\text{я}}\beta_{\text{я}}H_x[2\Delta_0(E^2-E_f^2)+8|V_q|^2\gamma^2\bar{\sigma}_zC(\hbar\omega_q)^{-1}]\}^{-1}. \qquad (6)$$

The "bare" (neglecting the dynamical coupling of the subsystems) energies of the elementary excitations in the electron, phonon and nuclear subsystems are given by the expressions

$$E_e=\{4\Delta_0^2+C\gamma[C\gamma-2A(q)\bar{\sigma}_y]\}^{1/2},\quad E_n=(g_{я}^2\beta_{я}^2H_x^2+D^2)^{1/2},$$
$$E_f=[\hbar^2\omega_q^2+4|V_q|^2\gamma\bar{\sigma}_yC(\hbar\omega_q)^{-1}]^{1/2}, \tag{7}$$

where $A(q)$ is the Fourier transform of the quantity A. It is easy to see that when the electron–nuclear interaction is "turned off" ($B=0$) expression (6) is transformed into the corresponding expression obtained in Ref. 8 for the Green's function. We note that in the derivation of Eq. (6), we retained in the sums over the phonon branches (see Eq. (4)) only the one acoustic branch of interest to us, which made the largest contribution.[4] The energy spectrum of the elementary excitations of the crystal, which is determined by the poles of $G_q^z(E)$, is found from the equation

$$(E^2-E_n^2)\{(E^2-E_e^2)(E^2-E_f^2)-4|V_q|^2C\gamma\bar{\sigma}_yE_c^2(\hbar\omega_q)^{-1}\}$$
$$=4B^2\gamma^2\bar{\sigma}_z\bar{I}_xg_{я}\beta_{я}H_x[2\Delta_0(E^2-E_f^2)+8|V_q|^2C\gamma^2\bar{\sigma}_z(\hbar\omega_q)^{-1}]. \tag{8}$$

Equation (8) describes the coupled electron–nuclear–phonon oscillations.

In the absence of electron–nuclear interaction (at $B=0$), Eq. (8) describes the free nuclear subsystem and the coupled electron–phonon modes which were considered previously in detail.[8] In the other limiting case, when the electron–phonon interaction is absent ($V_q=0$), we obtain

$$[(E^2-E_n^2)(E^2-E_e^2)-8B^2\gamma^2\bar{\sigma}_z\bar{I}_xg_n\beta_nH_x\Delta_0](E^2-\hbar^2\omega_q^2)=0 \tag{8a}$$

(in the following, we set the external pressure P equal to zero). Equation (8a) describes the free phonons and bound electron–nuclear oscillations. The latter are described by an equation similar to the equation for the bound electron–nuclear modes in ferromagnets (in our case, the electronic order parameter is $\bar{\sigma}_z$); such equations were studied in detail in the monograph of Ref. 9. As is seen from Eq. (8), the coupling of the electron and phonon subsystems is caused by the magnetic field H_z, whereas the coupling of the nuclear subsystem with the electron–phonon subsystem occurs only at $H_x\neq 0$. It also follows from Eq. (8) that the intermingling of the nuclear branch of oscillations with the electron and phonon branches occurs only at a temperature which is below the temperature of the structural phase transition T_c. The equilibrium deformation of the crystal, which is the order parameter in structural phase transitions, is determined in self-consistent fashion by the state of the electron subsystem and is proportional to $\bar{\sigma}_z$. In the high-temperature phase of the crystal $\bar{\sigma}_z=0$, and the free nuclear oscillations are separated from the bound electron–phonon modes. It follows then that there is no nuclear absorption of the sound above the phase-transition temperature. This indicates the possibility of revealing the structural phase transition by the nuclear absorption of the acoustic oscillations. It is clear from this that we shall henceforth be interested in the temperature region $T\leqslant T_c$. Recognizing that $\bar{\sigma}_y=\frac{1}{2}C\gamma A^{-1}$, we rewrite Eq. (8) in the form

$$(E^2-E_n^2)[(E^2-\bar{E}_e^2)(E^2-\hbar^2\omega_q^2)-4|V_q|^2C\gamma\bar{\sigma}_y\hbar\omega_q]$$
$$=8B^2\gamma^2\bar{\sigma}_z\bar{I}_xg_{я}\beta_{я}H_x\Delta_0(E^2-\hbar^2\omega_q^2), \tag{8b}$$

where

$$\bar{E}_e=[E_e^2+4|V_q|^2C\gamma\bar{\sigma}_y(\hbar\omega_q)^{-1}]^{1/2}. \tag{9}$$

Using Eq. (8b), we can show that the quantity $\bar{E}_e$ determines the energy of the quasi-electron branch of the energy spectrum of the crystal in the range of values of the wave numbers q that are much smaller than the value q_e at which the electron and phonon branches intersect. Since nuclear sound absorption usually occurs at frequencies much lower than the frequency of the elementary excitations in the electron subsystem, we shall be interested in the solution of Eq. (8b) at $E\sim E_f$, $E_n\ll\bar{E}_e$. Neglecting $E^2/\bar{E}_e^2$ in comparison with unity, Eq. (8b) can be reduced to the form

$$(E^2-\bar{E}_n^2)(E^2-\hbar^2\bar{\omega}_q^2)=8|V_q|^2(\hbar\omega_q)^{-1}\zeta, \tag{10}$$

where

$$\bar{E}_n=[E_n^2-8B^2\gamma^2\bar{\sigma}_z\bar{I}_xg_{я}\beta_{я}H_x\Delta_0\bar{E}_e^{-2}]^{1/2}\quad \bar{\omega}_q=\omega_qE_e\bar{E}_e^{-1},$$
$$\zeta=4\hbar^2\omega_q^2C\gamma^3\bar{\sigma}_yB^2\bar{\sigma}_z\bar{I}_xg_{я}\beta_{я}H_x\Delta_0\bar{E}_e^{-4}. \tag{11}$$

Equation (10) describes coupled nuclear-phonon oscillations. The dependence of the nuclear spin-phonon coupling on the parameter of electron ordering $\bar{\sigma}_z$ and on the nuclear magnetization $\bar{I}_x$ is a consequence of the electron-nuclear coupling. We emphasize the possibility of variation of the value of the dynamic nuclear spin-phonon coupling by the magnetic field perpendicular to the crystalline c axis of the crystal ($H_x\perp c$). We also note that Eqs. (8b) and (10) can be used to examine the effect of the electron-nuclear coupling on the velocity of sound propagation in a crystal with CJTE. As is seen from Eq. (10), the presence of dynamic nuclear spin-phonon coupling leads to the dependence of the sound velocity on the magnetic field H_x which is characteristic of electron-nuclear interactions.

2. Denoting the solutions of Eq. (8) by E_1^2, E_2^2, and E_3^2, we represent the function $G_q^z(E)$ in the following form:

$$G_q^z(E)=2\pi^{-1}C\gamma\bar{\sigma}_y[E_1^4(E_3^2-E_2^2)+E_2^4(E_1^2-E_3^2)+E_3^4(E_2^2-E_1^2)]^{-1}$$
$$\times[(E_3^2-E_2^2)(E_1^2-E_n^2)(E_1^2-\hbar^2\omega_q^2)(E^2-E_1^2)^{-1}+$$
$$+(E_1^2-E_3^2)(E_2^2-E_n^2)(E_2^2-\hbar^2\omega_q^2)(E^2-E_2^2)^{-1}+$$
$$+(E_2^2-E_1^2)(E_3^2-E_n^2)(E_3^2-\hbar^2\omega_q^2)(E^2-E_3^2)^{-1}]. \tag{12}$$

Starting out from Eq. (12), and with the help of the relation

$$\lim_{\varepsilon\to 0}(x\pm i\varepsilon)^{-1}=P\frac{1}{x}\mp i\pi\delta(x)$$

we can obtain an expression for the imaginary part of the Green's function that determines the sound absorption coefficient at arbitrary ω and q. However, since we are interested only in that part of the energy spectrum for which the condition $E\sim E_n$, $E_f\ll E_e$ is satisfied, assuming that the root E_3 corresponds to the electron-like branch and neglecting terms of the order $E_{1/2}^2/E_3^2$, we obtain

$$\operatorname{Im}G_q^z(E)={}^1\!/_2C\gamma\bar{\sigma}_yE_3^{-1}\delta(E-E_3)+\operatorname{Im}G_{q(n-f)}^z(E), \tag{13}$$

where the imaginary part of the nuclear phonon Green's function $G^{z}_{q(n-f)}(E)$ is given by the expression

$$\operatorname{Im} G_{q(n-f)}(E)=-\frac{1}{2}C\gamma\bar{\sigma}_yE_3^{-2}\sum_{i=1,2}\frac{(E_i^2-E_n^2)(E_i^2-\hbar^2\omega_q^2)}{(2E_i^2-\bar{E}_n^2-\hbar^2\bar{\omega}_q^2)E_i}\delta(E-E_i). \quad (14)$$

The quantities E_i in Eq. (14) are the roots of the dispersion equation (10) for the coupled nuclear-phonon oscillations, obtained in the same approximation as Eq. (14). The first term in (13) describes sound absorption associated with the excitation of the quasi-electron branch E_3 of elementary excitations, and since $E_3 \cong \overline{E_e}$ in the considered range of small wave numbers, it can be written in the form

$$\operatorname{Im} G_{q(e)}(E)={}^1/{}_2C\gamma\bar{\sigma}_y\bar{E}_e^{-1}\delta(E-\bar{E}_e). \quad (15)$$

It is seen from (15) that the presence of electron—nuclear interaction leads to a certain increase in the sound absorption. In the absence of this interaction at $B=0$, Eq. (15) takes the form previously obtained[8] for small q.

We now consider in detail the absorption connected with the excitation of nuclear-phonon oscillations.

a) The case of small q ($\bar{E}_n \gg \hbar\bar{\omega}_q$, $q \ll q_n$; q_n is the value of the wave vector at which the bare phonon ($\hbar\bar{\omega}_q$) and nuclear ($\bar{E}_n$) excitation branches intersect). Assuming the inequality $8|V_q|^2(\hbar\omega_q)^{-1}\zeta \ll \bar{E}_n^2$ to be valid (weak dynamic nuclear spin–phonon coupling), we obtain for the energy of quasi-nuclear E_+ and quasi-phonon E_- branches of the spectrum, from Eq. (10):

$$\begin{aligned} E_+^2&=\bar{E}_n^2+8|V_q|^2\zeta(\bar{E}_n^2\hbar\omega_q)^{-1},\\ E_-^2&=\hbar^2\bar{\omega}_q^2-8|V_q|^2\zeta(\bar{E}_n^2\hbar\omega_q)^{-1}. \end{aligned} \quad (16)$$

Expression (14) takes in this case the following form

$$\begin{aligned}\operatorname{Im} G^{z}_{q(n-f)}(E)={}^1/{}_2C\gamma\bar{\sigma}_y\bar{E}_e^{-2}\{&[8B^2\gamma^2\bar{\sigma}_z\bar{I}_xg_n\beta_nH_x\Delta_0\bar{E}_e^{-2}\\ &-8|V_q|^2\zeta(\hbar\omega_q\bar{E}_n)^{-2}](1/E_+)\delta(E-E_+)\\ &+4|V_q|^2(\hbar\omega_q)^{-1}[\hbar\bar{\omega}_qE_e^{-2}C\gamma\bar{\sigma}_y+\zeta(\bar{E}_n^2E_-)^{-1}]\delta(E-E_-)\}.\end{aligned} \quad (17)$$

As is seen from Eq. (17), each of the absorption coefficients, due to the excitation of quasi-nuclear and quasi-phonon branches of the spectrum, consists of two terms. The first is due to the coupling of the nuclear or phonon subsystem with the electrons, and the second to nuclear-phonon coupling. In absorption by the quasi-nuclear branch, the first term is much greater than the second because of satisfaction of the condition

$$|V_q|^2\hbar\omega_qC\gamma\bar{\sigma}_y(\bar{E}_n\bar{E}_e)^{-2}\ll 1.$$

The relative contribution of the two indicated coupling channels to the ultrasonic absorption coefficient $\alpha_-(q)$ of the phonon subsystem is determined by the relation between the quantities H_x and H_z, and also by the value of the hyperfine interaction constant B and the temperature of the crystal. However, in an arbitrary case, the term due to electron-phonon coupling is the larger. This follows from the inequality

$$B^2\gamma^2\bar{\sigma}_z\bar{I}_xg_n\beta_nH_x\Delta_0(\bar{E}_n\bar{E}_e)^{-2}\ll 1. \quad (18)$$

As is seen from Eq. (11), an inequality that is the inverse of (18) cannot be satisfied, since this would lead to violation of the condition $\bar{E}_n > 0$ (still more dangerous in this sense is the region of intermingling of the branches). Thus, inequality (18) can be regarded as the upper bound of the fields. We note that in the absence of electron–nuclear coupling ($B=0$), the absorption coefficient α_- of the phonon subsystem takes the form obtained in Ref. 8. The field dependence of $\alpha_-(q)$ is of the form

$$\alpha_-(q)\sim H_z^2[a(q)H_z^2+b(q)H_x^2],$$

from which it is seen that turning on the magnetic field H_x increases the absorption coefficient in the given region of values of q. This contribution, which is dependent on H_x, is due to the hyperfine interaction, and increases linearly with increasing wave number ($b(q)\sim q$). It is easy to see that at a sufficiently small ratio H_z/H_x, determined by the condition

$$|V_q|^2C\bar{\sigma}_yE_n\ll B^2\gamma\bar{\sigma}_z\Delta_0g_n\beta_nH_x\bar{I}_x, \quad (19)$$

the absorption of ultrasonic energy by the nuclear subsystem is much greater than by the phonon system.

b) The case of large q ($E_n, \bar{E}_n \ll \hbar\bar{\omega}_q$). We obtain for the quasi-phonon (E_+) and quasi-nuclear (E_-) energies

$$\begin{aligned} E_+^2&=\hbar^2\bar{\omega}_q^2+8|V_q|^2\zeta(\hbar\omega_q)^{-1}(\hbar\bar{\omega}_q)^{-2},\\ E_-^2&=\bar{E}_n^2-8|V_q|^2\zeta(\hbar\omega_q)^{-1}(\hbar\bar{\omega}_q)^{-2}. \end{aligned} \quad (20)$$

The imaginary part of the Green's function can be written in this case in the following fashion:

$$\begin{aligned}\operatorname{Im} G^{z}_{q(n-f)}(E)={}^1/{}_2C\gamma\bar{\sigma}_y\bar{E}_e^{-2}\{&4|V_q|^2(\hbar\omega_q)^{-1}[\hbar\bar{\omega}_q\bar{E}_e^{-2}C\gamma\bar{\sigma}_y\\ &-\zeta(\hbar^2\omega_q^2E_+)^{-1}]\delta(E-E_+)+[8B^2\gamma^2\bar{\sigma}_z\bar{I}_xg_n\beta_nH_x\Delta_0\bar{E}_e^{-2}\\ &+8|V_q|^2(\hbar\omega_q)^{-1}\zeta(\hbar^2\bar{\omega}_q^2)^{-1}]\bar{E}_e^2(E_eE_-)^{-1}\delta(E-E_-)\}.\end{aligned} \quad (21)$$

The relations between the values of the terms in the coefficient of ultrasound absorption by the phonon and nuclear subsystems, and the coefficients themselves are analogous to those considered in case a). The field dependence of the coefficient $\alpha_+(q)$ (phonon branch) can be represented as

$$\alpha_+(q)\sim H_z^2[a(q)H_z^2-c(q)H_x^2],$$

i.e., at large values of q the field H_x leads to a decrease in the absorption coefficient due to the hyperfine interaction. As is easy to show, the increment quadratic in the field H_x is inversely proportional to the wave number q ($c(q)\sim q^{-1}$).

In the case $q\approx q_n$, $\bar{E}_n\approx\hbar\bar{\omega}_q$ (i.e., in the intersection region) the ultrasonic absorption coefficients associated with the excitation of each of the considered branches of the spectrum are of the same order of magnitude.

3. The analysis that has been set forth shows that, when the frequency of the external ultrasonic source approaches the region of intersection of the nuclear and phonon branches of the spectrum, the contribution of the nuclear spin-phonon coupling to the absorption coefficient increases linearly with frequency, reaching a max-

imum value near the characteristic frequency of the nuclear subsystem of the crystal. Upon further increase in the frequency, this contribution decreases in inverse proportion to the frequency, i.e., nuclear acoustic resonance takes place. As is seen from the equations, the contribution due to the dynamic nuclear spin-phonon coupling enters into the absorption coefficient has opposite signs on the left and right of the resonance region. It follows therefore that the shape of the resonance curve is asymmetric in crystals of the considered type. This asymmetry is due to the interference character of the ANR in the considered systems. The theory of interference resonance was developed by Fano[10] in investigations of optical absorption spectra of neutral atoms. Similar resonances were also considered in nuclear physics by Breit and Wigner.[11] It was shown in their work that if a discrete level appears in the continuous energy spectrum (band) of a subsystem that absorbs the energy of the external source, and if this level is excited by the same source through an independent channel, then the shape of the curve of the absorption coefficient of the first subsystem near the frequency corresponding to the energy of the discrete level, turns out to be asymmetric. The reason is that in this situation we have not a simple superposition (additivity) of the Lorentz peak on the continuous absorption background, but rather an interference. The latter is not difficult to understand if we recognize that to obtain the total scattering intensity it is in fact necessary first to combine the scattering amplitudes due to two independent channels, and only then to square the result. As is known from scattering theory,[12] in terms of which an adequate analysis of absorption effects is possible, the scattering amplitude of radiation of frequency ω by a discrete level of energy E_0 experiences a 180° phase shift on going from $\omega < E_0$ to $\omega > E_n$, and it is this which causes the effect. Interference takes place even in the absence of a dynamic interaction between the discrete level and the band. Allowance for the latter, which leads to mutual repulsion of the levels, further increases the asymmetry of the resonance curve.

In the considered problem of ANR in crystals with CJTE, the energy of the quasi-nuclear branch of the elementary excitations corresponds to a discrete level in the continuous spectrum of the quasi-phonon subsystem. Both nuclear spins and phonons can absorb the ultrasound, thanks to the coupling with electrons, as a consequence of which, two independent absorption channels are realized, the interferences of which give the sought-after form of the resonance curve. It is interesting to note that, in terms of the Green's function, the interference resonance, as was to be expected, manifests itself in the fact that the nuclear-phonon Green's function, near the characteristic nuclear frequencies as can be seen from Eq. (6), is represented in the form of the product of the "nuclear" and "phonon" Green's functions $G_{(n-f)}(E) = G_n(E) G_f(E)$. The imaginary part $G_{(n-f)}(E)$ which determines the absorption coefficient is $\mathrm{Im}G_{(n-f)}(E) = \mathrm{Im}G_f(E)\,\mathrm{Re}G_n(E) + \mathrm{Im}G_n(E)\,\mathrm{Re}G_f(E)$, from which the asymmetry of the resonance follows.

In conclusion, we note that the interference character of the ANR is due to the dipole mechanism of nuclear spin–phonon coupling, in which the direct interaction of sound with the nuclear spin is absent. The presence of direct coupling of nuclear spins with the lattice (for example, the quadrupole interaction for ions with spin $I > \frac{1}{2}$) likewise leads to the appearance of the usual resonance absorption. In this case, the contribution of the interference term can be conveniently observed by saturating the ANR signal (a similar situation occurs in NMR in ferromagnets[13]).

Nuclear sound absorption can be revealed not only by the frequency dependence but also by the characteristic field dependence. As has already been noted, nuclei do not absorb ultrasound at $H_x = 0$. In a nonzero magnetic field H_x, the coefficient of nuclear absorption increases both with increasing H_z and with increasing H_x. The increase in the field component H_z at a fixed field H_x leads to an increase in the total absorption coefficient simultaneously with a decrease in the relative contribution of the dipole nuclear spin–phonon coupling. Consequently observation of nuclear acoustic resonance is difficult under these conditions. In the opposite case of small fixed fields H_z, when the field component H_x is increased the total absorption coefficient does not increase as much, but the relative share of the nuclear spin–phonon coupling increases significantly. It is natural that the latter case is the more favorable one experimentally.

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