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SOME FEATURES OF ULTRASOUND PROPAGATION IN NONCONDUCTING CRYSTALS WITH IMPURITIES

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Some features of the propagation of elastic waves, due to the presence of quasilocal states in the low frequency range of the spectrum, are considered for the case of a disordered solid substitution solution with a concentration $c \ll 1$. Propagation of the elastic waves is investigated for an arbitrary substitution impurity by application of the averaged equations of the dynamics of a crystalline lattice obtained under the assumption that the wavelength is much greater than the mean distance between the impurities. It is shown that it is possible to write down the averaged equations in terms of the scattering amplitude. It is further shown that the introduction of sufficiently heavy impurity centers results in the appearance of an "almost forbidden" broad frequency range in which the state density is proportional to $c \ll 1$.

1. INTRODUCTION

At sufficiently low temperatures, the principal mechanism of sound absorption in a real crystal is the scattering of elastic waves by the static defects of the lattice. Recently, a series of theoretical and experimental researches[1-7] has appeared, in which it is shown that the presence of point defects can lead to the appearance of states of an almost localized type (quasilocal states) with frequencies lying within the range of the continuous spectrum of an ideal crystal. Quasilocal states have a significant role in the thermodynamic and kinetic properties of the crystal. Since the quasilocal frequencies lie near the limits of the continuous spectrum, sound absorption with frequencies close to the quasilocal should have a sharply expressed resonant character. In the present research some features of the propagation of sound waves produced by the presence of quasilocal states in the low frequency part of the spectrum are investigated for the case of a disordered solid substitution solution with concentration $c \ll 1$.[1)] It is not difficult to generalize the results obtained to the case of injected impurities.

For all real frequencies of ultrasonics and not too low concentrations of the impurities the inequality $\lambda \gg l \sim ac^{-1/3}$ holds, where λ is the wavelength of the sound, l is the mean distance between impurities, and a is the atomic separation. This inequality allows us to describe the ultrasonic absorption in terms of the average characteristics of the field of atomic displacements of the crystalline lattice.

In the second section, we shall consider the derivation of the averaged equations of the dynamics of a crystalline lattice distorted by the introduction of substitution impurities. These equations are investigated for arbitrary impurity atoms, which differ from the atoms of the ideal lattice both in mass and in the character of the elastic interactions. By using the general relations for the amplitude of the scattering of a plane wave by a point defect, one can show that the ultrasonic absorption coefficient in the absence of quasilocal states has a Rayleigh character.

In the third section, the resultant equations are used for the study of the resonant absorption of the ultrasound at a frequency close to the quasilocal. Considerable attention is paid in this section to the case in which the mass of the impurity substitution is so large that the mass density of the atoms of the substitution solution is greater or of the order of magnitude of the mass density of the atoms in the host lattice. It is shown that the introduction of solid impurity centers leads to a material rearrangement of the spectrum, that is, to the appearance of an "almost forbidden" broad frequency range in which the state density is propor-

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[1)] A similar question was studied in the work of one of the authors,[7] but the formula for the ultrasonic absorption coefficient used in that work is inapplicable for the problem considered here, as will be shown below.

tional to a small quantity—the concentration. In this entire range of frequencies the ultrasonic absorption is anomalously large, although it no longer has a resonant character. It should be noted that the appearance in the spectrum of a broad band of frequencies with low state density must lead to significant singularities both in the kinetic and thermodynamic properties of crystals with impurities. (This problem is the subject of a separate research.)

2. DERIVATION OF THE AVERAGED EQUATIONS

The absorption of ultrasound is determined, with an accuracy to within unimportant quantum fluctuations, by the classical scattering of elastic waves by point defects. In the case of a simple monatomic lattice, the classical equations of dynamics have the following form:

$$m_{\mathbf{n}}\ddot{u}_{\mathbf{n}}^{i} = \sum_{j,\mathbf{n}'} L^{ij}_{\mathbf{n}-\mathbf{n}'} u_{\mathbf{n}'}^{j} + \sum_{\mathbf{n}_\alpha, j, \mathbf{n}'} b^{ij}_{\mathbf{n}-\mathbf{n}_\alpha, \mathbf{n}'-\mathbf{n}_\alpha} u_{\mathbf{n}'}^{j} + \sum_{\mathbf{m}} F_{\mathbf{m}}^{i} e^{i\omega t}. \tag{1}$$

where $u_{\mathbf{n}}^{i}$ is the displacement of the **n**-th atom in the i-th direction, $m_{\mathbf{n}}$ is the mass of the **n**-th atom, equal to m if a basic atom of the lattice is in the **n**-th site, or to m_1 if an impurity substitution atom is found at this site; the matrix $\hat{L}$ characterizes the elastic coupling of the ideal crystal; the matrix $\hat{b}_\alpha$ characterizes the perturbation which is brought about by the impurity lying in the site $\mathbf{n}_\alpha$; $F_{\mathbf{m}}^{i} e^{i\omega t}$ is the external periodic force of frequency ω acting on the **m**-th atom. The matrices $\hat{L}$ and $\hat{b}_\alpha$ are symmetric in the indices i, j and **n**, **n'**; their elements satisfy the well known relation:

$$\sum_{\mathbf{n}'} L^{ij}_{\mathbf{n}-\mathbf{n}'} = 0, \quad \sum_{\mathbf{n}'} b^{ij}_{\mathbf{n}-\mathbf{n}_\alpha, \mathbf{n}'-\mathbf{n}_\alpha} = 0, \tag{2}$$

which follows from the condition of the vanishing of the forces in the displacement of the crystal as a whole.

The equations of the dynamics of the crystalline lattice with substitution impurities can be represented in such a fashion that the perturbation brought about by the α impurity is determined by the unit matrix

$$(\hat{\Lambda}^\alpha) \equiv \Lambda^{ij}(\mathbf{n}-\mathbf{n}_\alpha, \mathbf{n}'-\mathbf{n}_\alpha),$$

which takes into account both the change in the mass of the impurity atom and the change in the elastic coupling in the vicinity of the point $\mathbf{n}_\alpha$. Introducing in place of the variables $u_{\mathbf{n}}^{i}$ new variables $v_{\mathbf{n}}^{i}$ connected with $u_{\mathbf{n}}^{i}$ by the relations

$$v = \Big(\hat{L} + \sum_\alpha \hat{b}_\alpha\Big)^{1/2} u,$$

we find that in the v-representation the equations of dynamics take the following form:

$$\omega^2 v = \hat{L}v + \sum_\alpha \hat{\Lambda}^\alpha v + \mathcal{F}. \tag{3}$$

Limiting ourselves to the first approximation in the concentration, we can write the operator $\hat{\Lambda}^\alpha$ in the form:

$$\hat{\Lambda}^\alpha = \hat{b}_\alpha + (\hat{L} + \hat{b}_\alpha)^{1/2}\, \varepsilon_\alpha\, (\hat{L} + \hat{b}_\alpha)^{1/2}, \quad \varepsilon_\alpha = \frac{m - m_1}{m_1}\delta_{\mathbf{n}\mathbf{n}_\alpha}\delta_{\mathbf{n}'\mathbf{n}_\alpha}. \tag{3a}$$

As follows from Eq. (3a), the operator $\hat{\Lambda}$ has the following property:

$$\sum_{\mathbf{n}'} \Lambda^{ij}_{\mathbf{n}-\mathbf{n}_\alpha, \mathbf{n}_\alpha-\mathbf{n}'} = 0. \tag{4}$$

For the derivation of the equation which defines the average characteristics of the field $v_{\mathbf{n}}^{i}$, we write down Eq. (3) in integral form,[2)] representing the displacement field $v_{\mathbf{n}}^{i}$ in the form of a sum of the unperturbed wave $v_{\mathbf{n}}^{0}$ and waves scattered by the impurity atoms:

$$v = v^0 - \sum_\alpha \hat{G}\hat{\Lambda}^\alpha v. \tag{5}$$

Here $G^{ij}(\mathbf{n}-\mathbf{n}_1)$ is the Green's function of the operator $\hat{L} - \omega^2$:

$$G^{ij}(\mathbf{n}-\mathbf{n}_1) = \frac{\Delta_0}{(2\pi)^3}\sum_\nu \int \frac{e_\nu^{i}(\mathbf{k})\, e_\nu^{*j}(\mathbf{k}) \exp\{i\mathbf{k}(\mathbf{n}-\mathbf{n}_1)\}}{\omega_\nu^2(\mathbf{k}) - \omega^2 - i\gamma}, \tag{6}$$

$\gamma \to +0$; Δ_0 is the volume of the unit cell; $e_\nu^{i}(\mathbf{k})$ is a unit polarization vector; the squares of the unperturbed frequencies are the eigenvalues of the operator $\hat{L}$; integration in (6) is performed over the volume of the unit cell of the reciprocal lattice. In the case of small concentrations, the integral equation (5) permits us to obtain a solution of the problem in terms of the theory of scattering of the plane wave by a single impurity center.

We shall carry out the derivation of the averaged equations for a simplified model in which all the polarizations are independent of each other. This simplification does not change the structure of the basic formulas and at the same time allows us to avoid the considerable calculations associated with the presence of the tensor $e_\nu^{i}(\mathbf{k})e_\nu^{*j}(\mathbf{k})$ in the expression for the Green's function (6). The results obtained in the given model can be generalized to the case of "entangled" polarizations (see below).

[2)] The direct averaging of the finite-difference equation involves considerable difficulties in view of the fact that the matrix elements of the perturbation operator have the property (4).

For the subsequent investigation, we introduce a function χ_n^α which is defined as the sum of the unperturbed wave v_n^0 and the wave produced by the "radiation" of all the impurity atoms except the α-th, that is,

$$\chi_{\mathbf{n}}{}^{\alpha} = v_{\mathbf{n}}{}^{0} - \sum_{\alpha' \neq \alpha} G(\mathbf{n} - \mathbf{n}_1)\, \Lambda(\mathbf{n}_1 - \mathbf{n}_{\alpha'}, \mathbf{n}_\alpha - \mathbf{n}_{\alpha'})\, v_{\mathbf{n}_\alpha}. \tag{7}$$

The actual field of the displacements v_n is connected with χ_n^α in the following way:

$$v = (1 + \hat{G}\hat{\Lambda}^\alpha)^{-1}\chi^\alpha. \tag{7a}$$

Substituting (7a) in Eq. (7) we find

$$\chi_{\mathbf{n}}{}^{\alpha} = v_{\mathbf{n}}{}^{0} - \sum_{\alpha' \neq \alpha} \hat{G}\hat{T}^{\alpha'}\chi^{\alpha'}. \tag{8}$$

Here $\hat{T}^\alpha(\omega^2) = \hat{\Lambda}^\alpha(1 + \hat{G}\hat{\Lambda}^\alpha)^{-1}$ is the so-called scattering operator[8,10] of the field χ_n^α by the α impurity; using Eq. (4) for the operator $\hat{\Lambda}$, it is not difficult to obtain a similar property for the matrix $\hat{T}$:

$$\sum_{\mathbf{n}_1} T_{\mathbf{n}_1\mathbf{n}_2} = \sum_{\mathbf{n}_2} T_{\mathbf{n}_1\mathbf{n}_2} = 0. \tag{9}$$

In the case of low concentrations, the average distance l between impurity centers is much greater than the characteristic dimensions of the region in which the elements $T(n_1 - n_\alpha, n_2 - n_\alpha)$ are different from zero (these dimensions are $\sim$ a). Therefore, the chief contribution to the sum in α' of Eq. (8) is made by the values of $\chi_n^{\alpha'}$ corresponding to $|n_2 - n_{\alpha'}| \sim a$. Since for most impurities χ_n^α changes smoothly at atomic distances, the functions χ^α with account of the discrete argument of $\mathbf{n}$ can be represented in the form

$$\chi_{\mathbf{n}}{}^{\alpha} = \chi^{\alpha}_{\mathbf{n}_\alpha} + (\mathbf{n}^i - \mathbf{n}_\alpha{}^i)\, \psi_i{}^\alpha,$$

$$\psi_i{}^\alpha = (\chi^\alpha_{\mathbf{n}_\alpha + a_i} - \chi^\alpha_{\mathbf{n}_\alpha})/a_i \equiv \hat{\Delta}_i{}^\alpha\chi^\alpha, \tag{10}$$

where a_i is the i-th vector of the elementary lattice, which we shall assume for simplicity to be rectangular. Applying the finite-difference operator Δ_i^α to Eq. (8) and taking into account the property (9) of the operator T^α, we can get (after some transformations) an equation for ψ_i^α:

$$\psi_i{}^\alpha = \tilde{v}^0_{i,\alpha} - \frac{\Delta_0}{(2\pi)^3} \sum_{\alpha' \neq \alpha} \omega^2 \int \frac{e^{i\mathbf{k}(\mathbf{n}_\alpha - \mathbf{n}_{\alpha'})} R_{im}(\mathbf{k})\, d\mathbf{k}}{\omega^2(\mathbf{k}) - \omega^2 - i\gamma} \lambda_{mj}\psi_j{}^{\alpha'},$$

$$\lambda_{mj} = \sum_{\mathbf{n}_1, \mathbf{n}_2} n_1{}^m T_{\mathbf{n}_1\mathbf{n}_2} n_2{}^j, \quad \tilde{v}^0 = \hat{\Delta}_i{}^\alpha v_0,$$

$$R_{im}(\mathbf{k}) = \frac{1}{\omega^2(\mathbf{k})} \frac{e^{i\mathbf{k}\mathbf{a}_i} - 1}{a_i} \frac{e^{i\mathbf{k}\mathbf{a}_m} - 1}{a_m}. \tag{11}$$

Equation (11) is written in first approximation in c and in first non-vanishing approximation in a/λ. The meaning of the transformations carried out is that we have reduced the number of variables by going from the field of displacements v_n, which is specified at each point of the crystal, to the field ψ_α specified at the points n_α. A similar method was used by I. Lifshitz,[9] who investigated the features of the electron spectrum for the special case in which $\hat{\Lambda}^\alpha$ is a degenerate operator of first rank. Using Eq. (11), it is not difficult to generalize the results obtained in[9] to the case of an arbitrary operator $\hat{\Lambda}$.

In the long-wave case considered here, the principal contribution to ψ_i^α is made by impurities located relative to the α impurity center at distances much larger than l. The contribution of these impurities is greater by at least $(\lambda/l)^2 \gg 1$ than the contribution from the nearest neighbors. Carrying out the averaging in Eq. (11) over a volume containing a large number of impurities (the linear dimensions of this volume are much greater than λ) we obtain, with the desired degree of accuracy,

$$\bar{\psi}_i(\mathbf{r}) = \tilde{v}_i{}^0(\mathbf{r}) - c\sum_{\mathbf{r}'} F_{im}(\omega^2, \mathbf{r} - \mathbf{r}')\, \lambda_{mj}\bar{\psi}_j(\mathbf{r}'),$$

$$F_{im}(\omega^2, \mathbf{r} - \mathbf{r}') = \frac{\Delta_0}{(2\pi)^3}\, \omega^2 \int \frac{R_{im}(\mathbf{k})\, e^{i\mathbf{k}(\mathbf{r} - \mathbf{r}')}\, d\mathbf{k}}{\omega^2(\mathbf{k}) - \omega^2 - i\gamma} \tag{12}$$

Here $\psi_i(\mathbf{r})$ is the average value of $\psi_i^\alpha(\mathbf{r})$; $\mathbf{r}$ takes on all possible integral values, and summation is carried out over all sites of the lattice. Equation (12) can be represented in an equivalent form:

$$(\omega^2 - \hat{L})\, \bar{\psi}_i(\mathbf{r}) - c\omega^2 \sum_{\mathbf{r}'} R_{im}(\mathbf{r} - \mathbf{r}')\, \lambda_{mj}\bar{\psi}_j(\mathbf{r}')$$

$$= \hat{\Delta}_i{}^{\mathbf{r}}\mathcal{F}(\mathbf{r}), \quad R_{im}(\mathbf{r}) = \frac{\Delta_0}{(2\pi)^3} \int e^{i\mathbf{k}\mathbf{r}} R_{im}(\mathbf{k})\, d\mathbf{k}. \tag{12a}$$

For a wave propagating in the x direction, the following dispersion relation follows from Eq. (12a):

$$\omega^2 - \omega^2(\mathbf{k}) - c\Phi(\mathbf{k}_0, \omega) = 0, \quad \mathbf{k} = \{k_x, 0, 0\},$$

$$\omega^2(\mathbf{k}_0) = \omega^2, \quad \mathbf{k}_0 = \{k_{0x}, 0, 0\}. \tag{13}$$

Here $\Phi(\mathbf{k}_0, \omega)$ represents, with accuracy up to quantities of the order $(a/\lambda)^3$, the scattering amplitude at zero angle:

$$\Phi(\mathbf{k}_0, \omega) = \langle \mathbf{k}_0 | \hat{T} | \mathbf{k}_0 \rangle = \sum_{\mathbf{n}_1, \mathbf{n}_2} e^{-i\mathbf{k}_0\mathbf{n}_1} T_{\mathbf{n}_1\mathbf{n}_2} e^{i\mathbf{k}_0\mathbf{n}_2}. \tag{13a}$$

(We note that the quantity $\Phi(\mathbf{k}_0, \omega)$ has the same value in the v and u representations.) In writing down Eq. (13), we have taken into account the definition of λ_{mj} (11), the property (9) of the operator T and also the fact that in the case of long waves

$k_i k_m/\omega^2(\mathbf{k})$

$$= \delta_{ix}\,\delta_{mx}/s_{xx}{}^2\left(\mathbf{k} = \{k_x, 0, 0\},\ \omega^2(\mathbf{k}) = \sum_{p,\,q=1}^{3} s_{pq}{}^2 k_p k_q\right).$$

In the case of "entangled" polarizations, calculations completely analogous to those given above yield the following generalized equations:

$$\omega_{\nu_0}{}^2(\mathbf{k}) - \omega^2 + c\Phi_{\nu_0}(\mathbf{k}_0, \omega) = 0, \quad \omega_{\nu_0}{}^2(\mathbf{k}_0) = \omega^2,$$
$$\mathbf{k}_0 = \{k_{0x}, 0, 0\}, \tag{14}$$

$$\Phi_{\nu_0}(\mathbf{k}_0, \omega) = \langle \mathbf{k}_0\nu_0 \,|\, \hat{T} \,|\, \mathbf{k}_0\nu_0 \rangle$$
$$= \sum_{i,\,j,\,\mathbf{n}_1,\,\mathbf{n}_2} e_{\nu_0}{}^{*i}(\mathbf{k}_0) \exp(-i\mathbf{k}_0\mathbf{n}_1)\, T^{ij}_{\mathbf{n}_1\mathbf{n}_2} e_{\nu_0}{}^{j}(\mathbf{k}_0) \exp(i\mathbf{k}_0\mathbf{n}_2).$$

Here $T^{ij}_{\mathbf{n}_1\mathbf{n}_2}$ is the scattering operator. Equation (14) determines the damping of the wave propagating in the direction of the x axis and having a polarization ν_0.

The absorption coefficient of ultrasound Γ is determined by the negative part of the quantity k_x: $\Gamma = |\operatorname{Im} k_x|$. In the absence of resonance absorption, we have, in first approximation in c,

$$\Gamma = c\,|\operatorname{Im}\Phi(\mathbf{k}_0, \omega)|\,/\,2s_{xx}\omega. \tag{15}$$

For calculation of the imaginary part of $\Phi(\mathbf{k}_0, \omega)$, we make use of the well known "optical" theorem according to which

$$\operatorname{Im}\Phi(\mathbf{k}_0, \omega) = -\pi\Delta_0 \int d\mathbf{k}'\, |\Phi(\mathbf{k}', \mathbf{k}_0)|^2\, \delta(\omega^2(\mathbf{k}') - \omega^2),$$

$$\Phi(\mathbf{k}', \mathbf{k}_0) = \sum_{\mathbf{n}_1,\,\mathbf{n}_2} e^{-i\mathbf{k}'\mathbf{n}_1} T_{\mathbf{n}_1\mathbf{n}_2} e^{i\mathbf{k}_0\mathbf{n}_2} = \lambda_{im} k_i' k_{0m} + O\,[(a/\lambda)^3]. \tag{16}$$

It follows from Eq. (16) and the definition of the operator T that in the absence of resonance scattering one can represent the scattering amplitude $\Phi(\mathbf{k}, \mathbf{k}_0)$ in the form

$$\Phi(\mathbf{k}, \mathbf{k}_0) = \frac{\omega^2\Lambda_0 a^2}{s^2}\varphi(\varkappa, \varkappa_0), \qquad \varkappa = \frac{\mathbf{k}}{|\mathbf{k}|}, \qquad \omega^2(\mathbf{k}_0) = \omega^2, \tag{17}$$

where Λ_0 is the characteristic value of the matrix elements of the perturbation operator, s is the characteristic sound velocity, and φ is a nondimensional quantity of the order of unity. We find from Eqs. (15), (16), and (17) that

$$\Gamma = c\frac{\Lambda_0{}^2 a^4 \Delta_0}{s^7}\beta(\varkappa_0)\,\omega^4, \qquad \beta(\varkappa_0) = \frac{\pi}{2}\int dO\,|\varphi(\varkappa, \varkappa_0)|^2 \sim 1.$$

Thus, for an arbitrary impurity substitution, the scattering of sound in the absence of resonance has the ordinary Rayleigh character ($\Gamma \sim \omega^4$), which is determined by property (4) of the matrix elements of the perturbation operator $\hat{\Lambda}$.

3. RESONANCE SCATTERING

To study the resonance scattering of sound, we write the amplitude $\Phi(\mathbf{k}_0, \omega)$ in explicit form:

$$\Phi(\mathbf{k}_0, \omega) = (\omega/s)^2 \sum_{\mathbf{n}',\,\mathbf{n}_1,\,\mathbf{n}_2} (\varkappa_0\mathbf{n}_1)(\varkappa_0\mathbf{n}_2)\,\Lambda_{\mathbf{n}'\mathbf{n}_2} M_{\mathbf{n}_1\mathbf{n}'}(\omega^2)/D(\omega^2),$$
$$D(\omega^2) \equiv \operatorname{Det}\|1 + \hat{G}(\omega^2)\hat{\Lambda}\|. \tag{18}$$

Here $M_{\mathbf{n},\mathbf{n}'}(\omega^2)$ is the corresponding minor of the matrix $1 + \hat{G}(\omega^2)\hat{\Lambda}$. It follows from Eq. (4) in the case of low frequencies $\omega \ll \omega_D$ (ω_D is the Debye frequency) the following relation holds for the elements of the matrix $\hat{G}\hat{\Lambda}$:

$$\operatorname{Im}(\hat{G}\hat{\Lambda})_{\mathbf{n}_1\mathbf{n}} = -\frac{\pi\Delta_0}{(2\pi)^3}\sum_{\mathbf{n}'}\oint \frac{(\mathbf{k}, \mathbf{n}_1 - \mathbf{n}')^2\, d\sigma}{|\partial\omega^2(\mathbf{k})/\partial\mathbf{k}|}\Lambda_{\mathbf{n}'\mathbf{n}_1} + O(\omega^4). \tag{19}$$

(integration is carried out over the "isoenergetic" surface $\omega^2(\mathbf{k}) = \omega^2$).

Using Eqs. (18), (19), the quantity $\Phi(\mathbf{k}_0, \omega)$ for for $\omega \ll \omega_D$ can be represented in the following form:

$$\Phi(\mathbf{k}_0, \omega) = \omega^2\frac{\omega_1{}^2}{\Omega_0{}^2 - \omega^2 + i\omega_2{}^2(\omega/\omega_0)^3}$$
$$\Omega_0{}^2 = D(0)\Big/\frac{d}{d\omega^2}D(\omega^2)\Big|_{\omega^2=0}, \qquad \omega_0{}^3 = \frac{s_{xx}{}^5}{\Lambda_0\Delta_0 a^2},$$
$$\frac{\omega_1{}^2}{\Omega_0{}^2} = \frac{1}{D(0)\,s_{xx}{}^2}\sum_{\mathbf{n}_1,\,\mathbf{n}_2,\,\mathbf{n}'}(\varkappa_0\mathbf{n}_1)(\varkappa_0\mathbf{n}_2)\,\Lambda_{\mathbf{n}_2\mathbf{n}'} M_{\mathbf{n}'\mathbf{n}_1}(0), \tag{20}$$

where the actual quantities Ω_0, ω_1, ω_2 do not depend on the frequency and are parameters which completely determine the scattering amplitude at low frequencies for an arbitrary impurity substitution. If the impurity atom is an isotope, then the parameters Ω_0, ω_1, ω_2 are determined in explicit fashion [see below, Eq. (29)]. The quantity Ω_0 has the meaning of a quasilocal frequency under the condition $\Omega_0 \ll \omega_D$. The lifetime of the quasilocal state with frequency Ω_0 is inversely proportional to the imaginary part of the denominator of Eq. (20) equal to $\omega_2^2(\omega/\omega_0)^3 \ll \omega_2^2$; it follows from the "optical" theorem that $\omega_2 \sim \omega_1$.

The character of the sound absorption for $\Omega_0 \ll \omega_0$ depends appreciably on the genesis of the quasilocal state. In what follows, we shall distinguish two cases.[3]

A. Quasilocal states which arise when the mass of the substitute atom $m_i \gg m$ (the mass effect, first predicted by Kagan and Iosilevskiĭ[1], and also independently by Brout and Vissher[2] and by Lehman and Dewames[3]); here

$$\Omega_0{}^2 \sim \omega_D{}^2 m/m_1, \quad \omega_1{}^2 \sim \omega_D{}^2, \quad \Omega_0{}^2 \lll \omega_1{}^2, \quad \omega_2{}^2 \sim \omega_D{}^2. \tag{21a}$$

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[3] Other possible situations are reduced qualitatively to one of the cases considered, A or B.

B. Quasilocal states which arise from impurity atoms which are weakly coupled with the atoms of the host lattice. It is not difficult to show that for impurities of this type, the following relations hold:

$$\Omega_0^2 \sim \omega_1^2 \ll \omega_D^2, \quad \omega_1^2 \sim \omega_2^2. \tag{21b}$$

Let us first consider the mass effect. In the given situation, most interest attaches to the case of the "strong" effect of the heavy mass when the mass of the impurity substitute is so great that

$$cm_1 \gtrsim m \quad (c\omega_1^2 \gtrsim \Omega_0^2). \tag{22}$$

The meaning of the inequality (22) is the following. For low frequency vibrations $\omega \ll \omega_D$, the deformations which arise in the vicinity of the heavy impurity exceed the deformations arising at other points of the crystal by a factor of $m_1/m \gg 1$. It then follows that the effective deformation of a crystal with impurities is $\epsilon_{\text{eff}} \sim \epsilon_0(1+cm_1/m)$, where ϵ_0 is the deformation of an ideal crystal. For $cm_1/m \gtrsim 1$, we have $(\epsilon_{\text{eff}}-\epsilon_0)/\epsilon_0 \gtrsim 1$, that is, in the field of deformations, the greatest effect is produced by the "superstructure" of the heavy substitute impurities. The latter circumstance leads to rearrangement of all the low-frequency portions of the spectrum.

Taking account of the relation (22), we investigate the dispersion equation (13), which determines simultaneously the renormalized sound velocity and the absorption coefficient of sound in the crystal. We find from Eqs. (13) and (20) that the dependence of k_x on ω has the following form:

$$k_x = \frac{\omega}{s_{xx}}\left(1+\frac{c\omega_1^2}{\Omega_0^2-\omega^2+i\omega_0^2(\omega/\omega_0)^3}\right)^{1/2}. \tag{23}$$

It follows from the inequalities (21a) and (22) that the term proportional to the concentration in the radicand of (23) is greater than or of the order of unity in a wide band of frequencies $\omega_2 \lesssim c^{1/2}\omega_1 \gg \Omega_0$. The nonlinear dependence of the renormalized wavelength $\bar{\lambda}$ on c then follows directly. The dependence of $\bar{\lambda}$ and Γ on ω also has a number of peculiarities.

If ω lies to the left of the resonant frequency Ω_0 and if the difference $\delta\omega = \Omega_0 - \omega \sim \Omega_0$, then $\operatorname{Im}\Phi(\mathbf{k}_0,\omega) \ll \operatorname{Re}\Phi(\mathbf{k}_0,\omega)$. In this case, the quantity $\bar{\lambda}(\omega)$ and the absorption coefficient Γ are determined by the formulas

$$\bar{\lambda}(\omega) = s_{xx}\Big/\omega\left(1+\frac{c\omega_1^2}{\Omega_0^2-\omega^2}\right)^{1/2}$$

$$\Gamma = |\operatorname{Im} k_x| = \frac{c}{\bar{\lambda}(\omega)}\frac{\omega_1^2\omega_2^2}{(\Omega_0^2-\omega^2)^2}\left(\frac{\omega}{\omega_0}\right)^3 \tag{24}$$

As is seen from Eqs. (24), in the limiting case as $\omega \to 0$, the sound absorption has a Rayleigh character ($\Gamma \sim \omega^4$), and the wavelength $\bar{\lambda} = \bar{s}_{xx}/\omega$, and the renormalized sound velocity $\bar{s}_{xx}$ differs significantly from the "bare" one s_{xx}:

$$\bar{s}_{xx} = s_{xx}/(1+c\omega_1^2/\Omega_0^2)^{1/2}. \tag{25}$$

Equation (24) is in agreement with the qualitative considerations given above.

With decrease of $\delta\omega$, the wavelength $\bar{\lambda}$ falls off, and when

$$\delta\omega \sim \delta\omega_1 = c^{1/3}(\omega_1/\omega_D)^2\Omega_0 \ll \Omega_0$$

it becomes less than the mean distance between the impurities. In this region of frequencies, the method of averaging used in the derivation of Eq. (13) is inapplicable, and the formula (25) is valid only in order of magnitude. Near resonance, at very small

$$\delta\omega \sim \delta\omega_2 = \omega_2^2\Omega_0^2/\omega_0^3 \lesssim c^{1/2}\Omega_0 \ll \delta\omega_1$$

the absorption coefficient $\Gamma = |\operatorname{Im} kx|$ [Eq. (23)] increases sharply:

$$\Gamma = \Omega_0 c^{1/2}(\omega_0/\Omega_0)^{3/2}/s_{xx},$$

and the characteristic damping length for the sound $1/\Gamma$ becomes $\sim\bar{\lambda}$. Thus the resonant scattering of long waves with frequencies lying in the range $\delta\omega_2$ is so large that the sound width $|\Omega_0-\omega| \le \delta\omega_2$ is not propagated in the crystal.

It follows from the inequality (22) that the region of frequencies for which anomalous sound absorption takes place is actually much greater than $\delta\omega_2$. The latter statement follows from the fact that the expression under the radical in the first of the formulas (24) is negative in the range of frequencies $[\Omega_0, \Omega_1]$, where $\Omega_1^2 = \Omega_0^2 + c\omega_1^2$, $\Omega_1 \gtrsim \Omega_0$. For these frequencies, $\operatorname{Re}\bar{\lambda}(\omega) = 0$, which determines also the impossibility of propagation in a crystal with heavy impurities of sound waves with wavelength $\bar{\lambda} \gtrsim l = ac^{-1/3}$ and a frequency belonging to the interval given above.[4] (We note that according to (22) and (21a), in an ideal crystal the frequencies $\omega \lesssim \Omega_1$ correspond to wavelengths $\lambda \gg l$.) For $\omega > \Omega_1$, the quantity $\lambda(\omega)$ again becomes real, whence $\lambda(\omega) \to \infty$, when $(\omega-\Omega_1) \to +0$, that is, the lattice vibrations with frequency Ω_1 are very close to optical.

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[4] A similar situation must take place also for heavy interstitial impurities. The quasilocal states in the interstitial impurities were considered by Kosevich, [6] however in view of an error in [6] the strong heavy-mass effect pointed out above was not observed there. The forbidden range of frequencies in this work is obtained as the direct consequence of scattering and is a narrow range of frequencies $\Delta\omega \sim c\Omega_0 \ll \delta\omega_2$ located close to Ω_0.

The character of the impurity distribution does not have a significant effect on the qualitative features of the propagation of long waves ($\lambda \gg l$). Therefore, the mechanism of the appearance of the forbidden band $[\Omega_0, \Omega_1]$ is most simply studied in the example of a crystal with heavy impurities, which form an ordered "superstructure" whose symmetry is identical with the symmetry of the host lattice. In such a completely ordered crystal there are $N_0 = [1/c] \gg 1$ "basic" atoms and one heavy impurity atom in a single unit cell, whose colume is equal to $\Delta_0/c \gg \Delta_0$.

With the aim of simplifying the calculations, we shall assume that the impurity differs from the atom of the host lattice only in mass. Writing an integral equation in the u-representation analogous to (5), we find

$$u_{\mathbf{r}_i} = \frac{m_1}{m}\omega^2 \sum_j F(\mathbf{r}_i - \mathbf{r}_j, \omega^2) u_{\mathbf{r}_j},$$

$$F(\mathbf{r}, \omega^2) = \sum_{\mathbf{k}} \frac{e^{i\mathbf{k}\mathbf{r}}}{\omega^2(\mathbf{k}) - \omega^2}, \tag{26}$$

$m_1/m \gg 1$. Here u_{r_i} is the displacement of an atom in the $\mathbf{r}_i$ site of the "superlattice," and summation is carried out over all sites. Noting that the stationary states of the ordered lattice are plane waves $u_{r_i} = e^{i\mathbf{k}\cdot\mathbf{r}_i}$ we find from Eq. (26) the equation for the determination of the spectrum of plane waves:

$$1 = c\frac{m_1}{m}\omega^2 \frac{1}{\omega^2(\mathbf{k}) - \omega^2} + c\frac{m_1}{m}\omega^2 \sum_{\mathbf{b}_m \neq 0} \frac{1}{\omega^2(\mathbf{k} + \mathbf{b}_m) - \omega^2}, \tag{27}$$

where the vector $\mathbf{k}$ lies inside the elementary cell of the reciprocal "superlattice" (the volume of the elementary cell is equal to $c/\Delta_0 \ll 1/\Delta_0$), $\mathbf{b}$ are the vectors of the sites of the reciprocal "superlattice," summation in (27) extends over all the sites $\mathbf{b}_m \neq 0$ situated inside the "ordinary" elementary lattice, corresponding to the ideal crystal. (The number of such sites is equal to $N_0 \gg 1$.) For $\omega \ll \omega(1/l)$ and $k \ll 1/l$, one can replace the sum over $\mathbf{b}_m$ by the corresponding integral and Eq. (27) in first approximation in c takes on the following form:

$$1 - \frac{m_1}{m}\omega^2 F_0 = \frac{c(m_1/m)\omega^2}{\omega^2(\mathbf{k}) - \omega^2}, \quad F_0 = \frac{\Delta_0}{(2\pi)^3}\int \frac{d\mathbf{k}}{\omega^2(\mathbf{k})}. \tag{28}$$

(Integration in F_0 is carried out over the volume $1/\Delta_0$.)

Equation (28) is identical with the dispersion equation (13) in all ranges of frequency $\omega \ll \omega(1/l)$ for $|\omega - \Omega_0| \gg \delta\omega_2$, when one can neglect $\operatorname{Im}\Phi(\mathbf{k}_0, \omega)$ in comparison with $\operatorname{Re}\Phi(\mathbf{k}_0, \omega)$. It is not difficult to establish the correctness of the last statement by noting that in the case in which the impurity atom is an isotope, the scattering amplitude is determined by the formula

$$\Phi(\mathbf{k}_0, \omega) = \frac{m_1}{m}\omega^2 \left[1 - \frac{m_1}{m}\omega^2 \int \frac{d\mathbf{k}}{\omega^2(\mathbf{k}) - \omega^2 - i\gamma}\right]^{-1}. \tag{29}$$

As analysis of (27) and (28) shows, the region of frequencies $[\Omega_0, \Omega_1]$ for the ordered location of impurities is forbidden for all wavelengths $\lambda \gtrsim l$, that is, in the integral $[\Omega_0, \Omega_1]$ the spectral density $\nu(\omega) = 0$. The quantity $\Delta\Omega = \Omega_1 - \Omega_0$ represents a gap separating the two branches of vibration of the ordered crystal. One of these branches determines the acoustic vibrations of the lattice as $\omega \to 0$ (Ω_0 is approximately the end-point frequency of this branch). The other branch, which begins with $\omega = \Omega_1$, is the first optical branch.

A disordered distribution of impurities leads to the result that the band of frequencies $[\Omega_0, \Omega_1]$ is no longer forbidden; however, the density of states inside this region is very low. To calculate the function $\nu(\omega)$ in the interval $[\Omega_0, \Omega_1]$, we make use of the well known formula:[9]

$$\nu(\omega, c) = \frac{1}{\pi N}\langle \operatorname{Im} \operatorname{Sp} \hat{G}\rangle, \quad \hat{G} = (\omega^2 - \hat{H} - i0)^{-1},$$

$$\operatorname{Sp} \hat{G} = \sum_n \langle G_{nn}(\omega)\rangle = \sum_k \langle G_{kk}(\omega)\rangle. \tag{29a}$$

Here N is the number of sites in the crystal, G_{nn} and G_{kk} are the matrix elements of the operator G in the coordinate and k-representations, respectively, $\langle \ldots \rangle$ denotes averaging over the different configurations of the impurities.

Noting that G_{nn_0} is a Green's function of Eq. (1), and taking into account (12a), (13), and the relation (29a) in first approximation in c, one can obtain the following equality:

$$\langle G_{kk}(\omega)\rangle = [\omega^2(\mathbf{k}) - \omega^2 + c\Phi(\omega^2)]^{-1}, \tag{30}$$

where $\Phi(\omega)$ is determined by Eq. (29). Substituting (30) in (29a), we find

$$\nu(\omega, c) = \frac{\Delta_0 c \operatorname{Im}\Phi(\omega)}{(2\pi)^3\pi}\int \frac{d\mathbf{k}}{[\omega^2(\mathbf{k}) - \omega^2 + c\operatorname{Re}\Phi(\omega)]^2}$$

$$\sim \frac{1}{\omega_D^2}\frac{c}{(Ac-1)^{1/2}}\left(\frac{m_1}{m}\right)^2\left(\frac{\omega}{\omega_D}\right)^4,$$

$$A = \omega_1^2/(\Omega_0^2 - \omega^2). \tag{31}$$

Formula (31) is valid for frequencies lying inside the interval $[\Omega_0, \Omega_1]$ sufficiently far from its ends; for these frequencies, the denominator of the integrand vanishes nowhere. It follows from Eq. (31) that $\nu(\omega, c) \ll \nu_0(\omega)$ ($\nu_0(\omega)$ is the spectral density of the ideal lattice, and the dependence of

$\nu(\omega, c)$ on concentration has a nonlinear character.

In the case in which the "weak" mass effect takes place:

$$c \ll m/m_1 \qquad (c\omega_1^2 \ll \Omega_0^2),\ \text{but}\ (\Omega_0/\omega_0)^3 \ll c, \qquad (32)$$

the analysis carried about above for very heavy masses remains valid; in particular, as follows from (32), the interval of forbidden frequencies, $\Delta\Omega > \delta\omega_2$. However, now $\Delta\Omega \sim c\omega_1/\Omega_0$, that is, $\Delta\Omega$ is much less than the quantity Ω_0. For ω lying to the left of Ω_0, at distances $\delta\omega > \delta\omega_2$, the renormalized wavelength and the absorption coefficient Γ are determined as before by the formulas (23a), but in view of the inequality (32), the renormalized sound velocity differs from the bare value by a small correction:

$$\bar{s}_{xx} = s_{xx}(1 - {}^1\!/_2 c\omega_1^2/\Omega_0^2).$$

For very low concentrations, when

$$c \ll (\Omega_0/\omega_0)^3,$$

the quantity $c\Phi(\omega)$ in the dispersion equation (13) is always much less than ω^2 and the anomalously strong sound absorption is absent in all regions of frequencies. The absorption coefficient in this case is equal to

$$\Gamma = \frac{\omega}{s_{xx}} \frac{c(\omega_1\omega_2)^2(\omega/\omega_0)^3}{(\Omega_0^2 - \omega^2)^2 + \omega_2^4(\omega/\omega_0)^6}. \qquad (33)$$

It follows from Eq. (33) that the sound absorption curve for ω close to Ω_0 has a Lorentzian character.

If quasilocal states appear for impurity atoms weakly bound with the atoms of the basic lattice, then, with account of the relation (21b), one can note that the condition $c\omega_1^2 < \Omega_0^2$ is always satisfied. Absorption of ultrasound in this case has the same character as in the case of the "weak" mass effect.

In concluding the present section, we note that the new quasilocal states with resonant frequency Ω^* can arise in pairs close to the impurity locations (the distance between impurities is of the order $a \ll l$)[11]. Such pairs can be considered as new impurity centers, the concentration of which is equal to c^2. Account of such pairs and the dispersion necessary in the narrow range of frequencies close to Ω^*:

$$\omega^2(\mathbf{k}) - \omega^2 + c\Phi(\omega) + c^2\Phi_2(\omega) = 0,$$

where $\Phi(\omega)$ is the scattering amplitude for a single impurity, $\Phi_2(\omega)$ is the scattering amplitude for a pair having a resonant character near Ω^*. The resonant character of the scattering by the pair can be reduced to $c\Phi(\Omega^*) \ll c^2\Phi_2(\Omega^*)$; here the absorption coefficient is proportional to c^2.

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198