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LOCAL OSCILLATIONS IN AN IONIC CRYSTAL IN THE PRESENCE OF A LONG-RANGE DEFECT

V. V. BRYKSIN and Yu. A. FIRSOV

Institute of Semiconductors, USSR Academy of Sciences

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Differential equations are obtained, the eigenvalues of which determine the spectrum of local oscillations in the phonon spectrum in the presence of long-range (charge) defects in an ionic crystal. The method proposed is similar to the effective-mass method for the calculation of shallow impurity donor and acceptor levels. The equations are used for the estimate of the number of local-oscillation levels that drop out of the longitudinal optical phonon branch in the case of an isotropic space. For certain values of the parameters of the theory, the defect may be a scattering center for long-wave phonons, the cross section of which is much greater than the square of the lattice constant. After a simple modification of the equations (terms containing the electric field are excluded) they are also valid for the case of defects in homopolar crystals in which oscillations of the type considered may appear in the phonon spectrum as the result of anharmonism forces.

1. INTRODUCTION

THE method of calculation of local oscillations was first formulated about twenty years ago by I. Lifshitz.[1] Later, Montroll and co-workers[2] gave this method the mathematical formulation which is now regarded as canonical (the apparatus of the Green's function). The method of I. Lifshitz leads to a set of linear equations, the order of which increases rapidly along with account of interaction of the defect with groups of ever more distant neighbors. The determinant of this set also gives the eigenfrequencies of the local oscillations, due to the given defect. The larger the number of neighbors taken into consideration, the higher the order of the determinant and the larger the number of local levels separated out from the continuous phonon spectrum. As a rule, the interaction with consecutive neighbors becomes weaker and the additional local levels, which arise when account is taken of these corrections, turn out to be shallower. Generally speaking, account of interaction with non-nearest neighbors in this method is accompanied by great difficulties. In this case, if the radius of action of the defect R is much greater than the lattice constant a, this method becomes ineffective.

For clarity, we consider the following example. In the KCl lattice, let one of the K^+ ions be replaced, say, by an Sm^{++} ion (we locate it at the origin of the coordinates), and let the compensating negative charge be located at a distance that is larger than the effective radius of action of the center (generally, it is assumed that they are arranged in a row). The presence of such a defect leads to the displacement of the equilibrium position of a large number of neighboring K^+ and Cl^- ions, while the value of these displacements depends on the distance of the ion from the defect (i.e., Sm^{++}). Such a nonuniform displacement of the ions of the lattice leads to a change in the equilibrium distance between them, as a result of which the force constants change, due to the anharmonism of the ion-ion interaction. In turn, this change in the force constants leads to the appearance of local (or "quasi-local," i.e., not separating out from the continuous spectrum) frequencies. In view of the large radius of the Coulomb field of the defect, the change in the force constants of the interionic interaction varies slightly from ion to ion, depending on the distance from the defect. This situation strongly recalls the separation of the shallow levels from the electron spectrum in the presence of long-range action of the defect, when the calculation is made by means of the effective-mass method.

Another example is a polaron of small radius in an ionic crystal. In the region of high temperatures, when the polaron (excess charge, polarizing the region) is located at the ground site for a long time in comparison with the period of oscillation of the lattice, one can regard it simply as an impurity ion of a different charge (but with the same mass). It is true that in this case there still exists a specific mechanism of interaction, which leads to a separation of an additional series of local levels,[3] but, as a rule, these levels are sufficiently deep, and therefore they are easily distinguished from those of which we shall speak below.

Thus the purpose of the present research is the development of a method of calculation of a large number of shallow local phonon levels, which are generally connected with a long-range defect. Inasmuch as the radius of the center is much greater than the lattice constant, the transition to the continuum approximation is a natural one. Formally, such a transition should be similar to the effective-mass method used for the determination of shallow acceptor and donor levels in the electron spectrum. In the simplest case of a nondegenerate band, according to this method, it is necessary to find the form of the spectrum $\epsilon(\mathbf{k}) = D_{\alpha\beta}k_\alpha k_\beta$ (here $D_{\alpha\beta}$ is a tensor of second rank, proportional to the reciprocal of the effective-mass tensor), and then to make the substitution $\mathbf{k} \to i\nabla$. The term $\epsilon(i\nabla)$ will play the role of the kinetic operator in the Schrödinger equation. Here the form of the potential energy changes insignificantly—the inertia-free dielectric permittivity ϵ_∞ is introduced: $e^2/r \to e^2/\epsilon_\infty r$. In this case, when the effective radius R is much greater than a, such a change is almost obvious.

In the case of n-fold degenerate bands at the extremum point, the problem reduces to the solution of the set of n differential equations of second order, i.e., the order of the Schrödinger differential equation is raised.

Thus, it would appear that, in analogy with the effective-mass method, it is necessary to obtain a differential equation (or a set of differential equations) the eigenvalues of which determine the spectrum of local oscillations.1) In the case of an ionic lattice of the type of NaCl or CsCl, there are two degenerate transverse branches $\omega_t^{(1)}(q)$ and $\omega_t^{(2)}(q)$ and one nondegenerate longitudinal branch $\omega_l(q)$, but the dispersion laws in this case have a completely different form[5,6] than for the electronic spectra. This is explained by the fact that the physical cause which leads to a splitting of the triply degenerate (at q = 0) state is different for the phonon spectrum than for the electronic spectrum.[6] In the first case, this cause is the long-range electric field, which accompanies the longitudinal optical oscillations, and in the second it is the spin-orbit interaction. Now, if one were to succeed in greatly lessening the effect of the electric field **E**, then the longitudinal and transverse phonon branches would coalesce at q = 0.2) According to [6], we have

$$\omega_l^2(\mathbf{q})=\omega_l^2\left\{1+mq^2a^2+na^2\frac{q_x^4+q_y^4+q_z^4}{q^2}\right\}, \tag{1a}$$

$$[\omega_t^{1,2}(\mathbf{q})]^2=\omega_t^2\left\{1-lq^2a^2+sa^2\left[\frac{1}{2}\frac{q_x^4+q_y^4+q_z^4}{q^2}\right.\right.$$
$$\left.\left.\pm\frac{1}{q^2}(q_x^4q_y^4+q_x^4q_z^4+q_y^4q_z^4-q^2q_x^2q_y^2q_z^2)^{1/2}\right]\right\}; \tag{1b}$$

here m, n, *l*, s are dimensionless constants, the analytic expression for which was found in [6];3) ω_l is the limiting value of the frequency of the longitudinal waves and the transverse optical oscillations.

It is evident that even in the nondegenerate case for $\omega_l(q)$ the prescription of the substitution $q \rightarrow i\nabla$ is ambiguous. Additional difficulties arise also in finding the form of the potential energy, which is also reflected in the order of the differential equation. Actually, all these difficulties are connected with the presence of the macroscopic electric field in the equations of motion.[6] Therefore, the transition to the continuum approximation in the case considered cannot be achieved by simply copying of the effective-mass method. We now turn to the description of the correct procedure for transition to the continuum approximation.

2. TRANSITION TO THE CONTINUUM APPROXIMATION

We introduce the following notation: **P**—the macroscopic dipole moment; **v**—the vector characterizing the displacement of the + and − ions in the n-th cell relative to one another: $\mathbf{v}(n) = \mathbf{u}\binom{n}{+} - \mathbf{u}\binom{n}{-}$;

$$\tilde{\mathbf{w}}(n)=\frac{1}{2}\left[\mathbf{u}\binom{n}{+}+\mathbf{u}\binom{n}{-}\right],$$

$$\mathbf{w}(n)=\frac{1}{M_++M_-}\left[M_+\mathbf{u}\binom{n}{+}+M_-\mathbf{u}\binom{n}{-}\right],$$

$M = \frac{M_+M_-}{M_+ + M_-}$—the reduced mass; where $\tilde{\mathbf{w}}(n)$ is a vector characterizing the displacement of the center of mass of the cell; **w**(n) is the displacement of the geometric center of the cell; **E** is the macroscopic electric field which describes the force exerted on the given cell by the other cells through the electric long-range interaction. In the absence of a defect and with neglect of spatial dispersion, the equations of motion in the optical branch have the form[9]

$$-\frac{d^2v_\alpha(\mathbf{r})}{dt^2}=\omega_0^2v_\alpha(\mathbf{r})-\left(\frac{\varepsilon_0-\varepsilon_\infty}{4\pi}\frac{v_a}{M}\right)^{1/2}\omega_0E_\alpha(\mathbf{r}),$$
$$\operatorname{div}(\mathbf{E}+4\pi\mathbf{P})=0, \tag{2}$$
$$P_\alpha(\mathbf{r})=\left(\frac{\varepsilon_0-\varepsilon_\infty}{4\pi}\frac{M}{v_a}\right)^{1/2}\omega_0v_\alpha(\mathbf{r})+\frac{\varepsilon_\infty-1}{4\pi}E_\alpha(\mathbf{r}),$$

where v_a is the volume of the elementary cell, ϵ_0 the static dielectric permittivity, and ω_0 the limiting frequency of the transverse phonons.

In the (2), no account is taken of corrections with respect to $(qa)^2$, which lead to the dispersion laws (1) and which determine the form of the kinetic operator. When they are taken into account, the form of Eqs. (2) becomes more complicated[6] (in (3) we change over to the simplest model of nondeformed ions, i.e., we set $\epsilon_\infty = 1$[6]):

$$-\frac{d^2v_\alpha(\mathbf{r})}{dt^2}=\omega_0^2v_\alpha(\mathbf{r})-\frac{e}{M}E_\alpha(\mathbf{r})-\sum_\beta\Pi_{\alpha\beta}(i\nabla)v_\beta(\mathbf{r}),$$
$$\operatorname{div}(\mathbf{E}+4\pi\mathbf{P})=0,\quad P_\alpha(\mathbf{r})=\frac{e}{v_a}v_\alpha(\mathbf{r}). \tag{3}$$

Here and below we shall mean by e the effective ionic charge:

$$e=\omega_0[Mv_a(\varepsilon_0-1)/4\pi]^{1/2}. \tag{4}$$

The matrix $\Pi_{\alpha\beta}(q)$ has the structure[6]

$$\Pi_{\alpha\beta}(\mathbf{q})=\omega_0^2a^2\{rq_\alpha q_\beta+\delta_{\alpha\beta}[(l-{}^1\!/_2s)q^2+sq_\alpha^2]\}. \tag{5}$$

Transforming to the q-representation in these equations, i.e., putting $\Pi_{\alpha\beta}(q)$ in the form (5) and assuming

$$E_\alpha=-\frac{4\pi}{q^2}\sum_\beta q_\alpha q_\beta P_\beta,$$

and then eliminating **E** and **P** from (3), we get a set of equations for $v_\alpha(q)$, from which the dispersion laws (1) are obtained.

We now proceed to an elucidation of the form of the potential energy operator. Account of a defect possess-

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1) The possibility of the use of a method similar to the effective-mass method in calculations of the phonon spectrum was pointed out in [4]; however, no specific results were given there.

2) In [7], one of the authors, together with V. L. Gurevich and A. I. Larkin, showed that by increasing the concentration of conduction electrons in ionic crystals, it is possible to screen the field **E**, and the branches ω_l and ω_t coalesce at q = 0 in this case. This fact was confirmed in neutronoscopic investigations of the ionic spectrum of PbTe and in experiments on the Raman scattering of the laser beam of GaAs.

3) K. B. Tolpygo called our attention to the fact that similar expressions were obtained for ω_l (q) and ω_t (q) in [5] for the case of a lattice of the NaCl type. The parameters m, n, *l* and s were given there in numerical form. To obtain them, a method was used in [5] that was developed by K. B. Tolpygo in [6] that differs from the traditional approach [9] with the use of the Szigeti relation. Furthermore, the equations of motion in the long-wave region (see (3)), which are necessary for us in the present work, were not obtained. The case of nondeformed ions, considered in the present work, leads to identical results both for the dispersion laws and for the constants m, n, *l*, and s by both methods. [5,6]

ing the additional charge δe, for the second of the Eqs. (3), is obvious in the continuum approximation:

$$\operatorname{div}(\mathbf{E}+4\pi\mathbf{P})=4\pi\delta e\delta(\mathbf{r}). \qquad (6)$$

The additional charge polarizes the medium, i.e., the + and − ions of each cell are displaced from their original equilibrium positions. We shall denote the new equilibrium values of v, w, and $\tilde{\mathrm{w}}$ by the index (0) and call them the polarization displacements (in the absence of the defect, they are equal to zero). The quantities $\mathrm{v}^{(0)}$, $\mathrm{w}^{(0)}$ and $\tilde{\mathrm{w}}^{(0)}$ are decreasing functions of the vector number of the cell l. For not too large an |l| we have

$$a \gg |\mathbf{v}^{(0)}(l)|,\ |\mathbf{w}^{(0)}(l)|,\ |\tilde{\mathbf{w}}^{(0)}(l)| > A=\begin{cases}(\hbar/M\omega_0)^{1/2} & \text{for}\quad \hbar\omega_0>kT\\ (\hbar^2/MkT)^{1/2} & \text{for}\quad \hbar\omega_0<kT\end{cases}.$$

Here A is the amplitude of the thermal oscillations of the ions in an ideal lattice.

The large values of the amplitudes of the equilibrium displacements can require account of the anharmonic terms in the equations of motion. The expressions for the harmonic and anharmonic parts of the force acting on the ion $\binom{l}{k}$ (k is the type of ion in the lattice, and k = + or −) have the form

$$F_\alpha^{\mathrm{r}}\binom{l}{k}=-\sum_{l',k',\beta}\Theta_{\alpha\beta\gamma}\begin{pmatrix}l & -l'\\ k & k'\end{pmatrix}\left[u_\beta\binom{l}{k}-u_\beta\binom{l'}{k'}\right], \qquad (7)$$

$$F_\alpha^{\mathrm{a}}\binom{l}{k}=\frac{1}{2}\sum_{l',k',\beta,\gamma}\Theta_{\alpha\beta\gamma}\begin{pmatrix}l & -l'\\ k & k'\end{pmatrix}\left[u_\beta\binom{l}{k}-u_\beta\binom{l'}{k'}\right]\left[u_\gamma\binom{l}{k}-u_\gamma\binom{l'}{k'}\right]. \qquad (8)$$

Here we limit ourselves to the model of pair and central forces, i.e.,

$$\Theta_{\alpha\beta}\begin{pmatrix}l & -l'\\ k & k'\end{pmatrix}=\frac{\partial^2\Psi_{kk'}(r^2)}{\partial r_\alpha\,\partial r_\beta},\qquad \Theta_{\alpha\beta\gamma}\binom{l}{kk'}=\frac{\partial^3\Psi_{kk'}(r^2)}{\partial r_\alpha\,\partial r_\beta\,\partial r_\gamma} \qquad (9)$$

with r equal to r$\begin{pmatrix}l & -l'\\ \mathrm{k} & \mathrm{k}'\end{pmatrix}$, a vector drawn from the equilibrium position of the ion $\binom{l'}{\mathrm{k}'}$ to the equilibrium position of the ion $\binom{l}{\mathrm{k}}$. Usually, in the case of short-range interaction, the ratio of the anharmonic contribution to the harmonic one is of the order of $\gamma\mathrm{A}/\mathrm{a} \ll 1$, where γ is a numerical factor that depends on the specific form of the interaction potential, usually reaching a value 5–10. Upon substitution of $\mathrm{v}^{(0)}$, $\mathrm{w}^{(0)}$ or $\tilde{\mathrm{w}}^{(0)}$ in (8) in place of one of the square brackets, we get a force that is similar in form to the anharmonic force. The ratio of the corresponding contribution to the purely harmonic contribution will be of the order of $\gamma\mathrm{v}_0/\mathrm{a} < 1$, but it can be equated to the "kinetic" term in the differential equation used by us to determine the local oscillations. We restrict ourselves in the following to the cubic anharmonic term, since account of higher order terms leads in any case to corrections in the parameter $(\mathrm{a}/\mathrm{R})^2$ (R is the radius of the considered localized phonon state).

We divide the anharmonic forces into long-range, connected with the macroscopic electric field, and the short-range. This procedure can be rigorously established with the help of the method of Ewald. The corresponding proof is given in [10]. In the present work, we carry our this division, being guided by intuitive but lucid considerations. The expression (8) is used only to estimate the coefficients stemming from the short-range contribution. Using (8) and (9) and setting v in place of u$\binom{l}{+}$ − u$\binom{l}{-}$ we expand the displacement u$\binom{l'}{\mathrm{k}'}$ in a series in r$\begin{pmatrix}l & -l'\\ \mathrm{k} & \mathrm{k}'\end{pmatrix}$ for $l \neq l'$. As a result we obtain4)

$$\tilde{F}_\alpha^{\mathrm{a}}\binom{r}{\mathrm{opt}}=M\sum_{\beta,\gamma}v_\gamma\Theta_{\alpha\beta\gamma}(i\nabla)\,w_\beta, \qquad (10)$$

where $\tilde{\mathrm{F}}^{\mathrm{a}}\binom{\mathrm{r}}{\mathrm{opt}}$ is the short-range part of the anharmonic force acting in the optical branch of oscillations,

$$\mathbf{F}\binom{r}{\mathrm{opt}}=\frac{1}{M_+ + M_-}\left\{M_-\mathbf{F}\binom{r}{+}-M_+\mathbf{F}\binom{r}{-}\right\}, \qquad (11)$$

and

$$\Theta_{\alpha\beta\gamma}(\mathbf{q})=i\lambda_1(q_\alpha\delta_{\beta\gamma}+q_\beta\delta_{\alpha\gamma}+q_\gamma\delta_{\alpha\beta})+i\lambda_2 q_\alpha\delta_{\alpha\beta}\delta_{\beta\gamma}. \qquad (12)$$

To obtain (10) it was taken into account that

$$\sum_{l'}\Theta_{\alpha\beta\gamma}\begin{pmatrix}l & -l'\\ k & k'\end{pmatrix}=0,$$

after which we transformed to the continuum approximation, i.e., we formally replaced the discrete variable $\mathbf{r}(l)$ by the continuous coordinate r. The constants λ_1 and λ_2 introduced in (12) are determined in terms of the derivatives of the function $\Psi_{\mathrm{kk}'}(\mathrm{r}^2)$ with respect to the coordinate. In [10], which is devoted to the microscopic basis of the proposed method of calculation, the quantities λ are expressed in terms of microscopic parameters.

The long-range contribution to the anharmonic force acting in the optical branch of oscillations, is sought, in the model of undeformed ions, in the form

$$\frac{e}{M}\frac{1}{M_+ + M_-}\left\{E_\alpha\left[\mathbf{r}+\mathbf{u}\binom{r}{+}\right]M_- - E_\alpha\left[\mathbf{r}+\mathbf{u}\binom{r}{-}\right]M_+\right\}$$
$$\approx\frac{e}{M}\sum_\beta\frac{\partial E_\alpha(\mathbf{r})}{\partial r_\beta}\left[\tilde{w}_\beta(\mathbf{r})+\frac{1}{2}\frac{M_- - M_+}{M_- + M_+}v_\beta(\mathbf{r})\right]. \qquad (13)$$

Adding (10) and (13) to the right side of the first of Eqs. (3), we get

$$\frac{d^2v_\alpha}{dt^2}=-\omega^2 v_\alpha+\frac{e}{M}E_\alpha+\sum_\beta\Pi_{\alpha\beta}(i\nabla)v_\beta$$
$$+\frac{e}{M}\sum_\beta\frac{\partial E_\alpha}{\partial r_\beta}\left[\tilde{w}_\beta+\frac{1}{2}\frac{M_- - M_+}{M_- + M_+}v_\beta\right]+\sum_{\beta,\gamma}v_\gamma\Theta_{\alpha\beta\gamma}(i\nabla)\,w_\beta. \qquad (14)$$

Further, it is necessary to generalize the third of Eqs. (3) to the anharmonic case. The simplest method consists of the following. We find **P** from the equation

$$\Delta\varphi=4\pi\operatorname{div}\mathbf{P}, \qquad (15)$$

where φ is the potential at the point **r**, due to the electric field of the ionic charges. In the framework of the continuum approximation, we have

$$\varphi(\mathbf{r})=\frac{e}{v_a}\int_V d^3r'\left\{\left|\mathbf{r}'+\mathbf{u}\binom{r'}{+}-\mathbf{r}\right|^{-1}-\left|\mathbf{r}'+\mathbf{u}\binom{r'}{-}-\mathbf{r}\right|^{-1}\right\}. \qquad (16)$$

Expanding this expression in a series in u with accuracy to quadratic terms, we obtain

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4)We note that in such a procedure terms of the type w∇v, v∇v and w∇w fall out. We neglect contributions containing products of higher order.

$$\varphi(\mathbf{r})=\frac{e}{v_a}\int_V d^3r'\left\{-\sum_\alpha\frac{\partial}{\partial r_\alpha}\frac{v_\alpha(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}+\sum_{\alpha,\beta}\frac{\partial^2}{\partial r_\alpha\partial r_\beta}\frac{v_\alpha(\mathbf{r}')\tilde{w}_\beta(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}\right\}. \tag{17}$$

Applying the Laplace operator to (17), we can write

$$\Delta\varphi=4\pi\,\mathrm{div}\frac{e}{v_a}\left\{\mathbf{v}-\sum_\beta\frac{\partial}{\partial r_\beta}(v_\beta\tilde{\mathbf{w}})\right\}.$$

Equating this expression with (15), we have

$$P_\alpha=\frac{e}{v_a}v_\alpha-\frac{e}{v_a}\sum_\beta\frac{\partial}{\partial r_\beta}(v_\beta\tilde{w}_\alpha). \tag{18}$$

We note that the value of **P** obtained in this fashion is valid with accuracy to within the curl of an arbitrary vector, in particular, up to $(e/v_\alpha)\,\mathrm{curl}\,(\tilde{\mathbf{w}}\times\mathbf{v})$. However, inasmuch as only div **P** is under study in what follows, such an ambiguity is unimportant.

The further procedure of solution of Eqs. (6), (14), and (18) consists in the splitting of v and w into static and dynamic parts, in which the first is found from setting the force equal to zero in the absence of thermal oscillations (the condition of equilibrium of the lattice with a defect). In the equations for the dynamic displacements (we indicate them with a prime index), one can omit terms proportional to w′.

However, Eqs. (6), (14), and (18) are clearly insufficient; it is necessary to supplement them with an equation for $\mathbf{w}^{(0)}$(or $\tilde{\mathbf{w}}^{(0)}$). As is shown in the Appendix, such an equation (the stability equation) for lattices of the type in question, under the condition that one can restrict oneself in the interaction to nearest neighbors only, is

$$\begin{gathered}(c_{12}+c_{44})\frac{\partial}{\partial r_\alpha}\mathrm{div}\,\tilde{\mathbf{w}}^{(0)}+c_{44}\,\Delta\tilde{w}_\alpha^{(0)}\\+(c_{11}-c_{12}-2c_{44})\frac{\partial^2\tilde{w}_\alpha^{(0)}}{\partial r_\alpha^2}=-U_0\frac{\partial}{\partial r_\alpha}\delta(\mathbf{r}).\end{gathered} \tag{19}$$

Here c_{ij} are the usual elastic constants, and U_0 is a quantity having the dimension of energy, the expression for which can be obtained only with the help of microscopic theory.[10] The right side of (19) is the additional force due to the presence of a change in the short-range force of interaction because of the defect. It is natural that in the case of short-range interaction, the corresponding change in the potential energy in the continuum approximation can be described by an expression of the form $U_0 v_a\delta(\mathbf{r})$ and the corresponding force is $-U_0 v_a\partial\delta(\mathbf{r})/\partial r_\alpha$. We note that the problem of the determination of the displacement in the presence of a point defect in simple crystals was considered in [11], and the equation obtained there differed from (19) only in that the displacement of the atom from the equilibrium position appeared on the left side in it, and not $\mathbf{w}^{(0)}$. This difference was due to the fact that one must have not one atom but two per elementary cell for the crystals considered here.

It is of interest to note that even in ordinary homopolar crystals, the expression (19) preserves its form; this can lead to the result that, besides the deep local levels obtained by the method of I. Lifshitz, a series of shallow local levels can appear when account is taken of the anharmonic contribution to the force. It is convenient to treat these levels in the continuum approximation (the corresponding potential energy will fall off very slowly).

Thus the set of equations for the determination of the spectrum of local oscillations takes the form

$$\begin{gathered}(\omega^2-\omega_0^2)v_\alpha'=-\frac{e}{M}E_\alpha'-\sum_\beta\Pi_{\alpha\beta}(i\nabla)v_\beta'\\-\frac{e}{M}\frac{M_--M_+}{M_++M_-}\sum_\beta\frac{\partial E_\alpha^{(0)}}{\partial r_\beta}v_\beta'-\frac{e}{M}\sum_\beta\frac{\partial E_\alpha'}{\partial r_\beta}\left[\tilde{w}_\beta^{(0)}+\frac{1}{2}\frac{M_--M_+}{M_-+M_+}v_\beta^{(0)}\right]\\-\sum_{\beta,\gamma}v_\gamma'\Theta_{\alpha\beta\gamma}(i\nabla)w_\beta^{(0)},\end{gathered} \tag{20a}$$

$$\mathrm{div}\,(\mathbf{E}'+4\pi\mathbf{P}')=0, \tag{20b}$$

$$P_\alpha'=\frac{e}{v}\left\{v_\alpha'-\sum_\beta\frac{\partial}{\partial r_\beta}\left(v_\beta'\tilde{w}_\alpha^{(0)}+\frac{M_--M_+}{2(M_++M_-)}v_\beta^{(0)}v_\alpha'\right)\right\} \tag{20c}$$

The quantities $\mathbf{w}^{(0)}$ and $\tilde{\mathbf{w}}^{(0)}$ which appear here are determined from (19) and with the help of the connection between v, w, and $\tilde{\mathbf{w}}$:

$$\tilde{\mathbf{w}}=\mathbf{w}+\frac{1}{2}\frac{M_--M_+}{M_-+M_+}\mathbf{v}. \tag{21}$$

In the lowest order in $(a/r)^2$ the quantity $\mathbf{v}^{(0)}$ is determined from the equations

$$\begin{gathered}0=-\omega_0^2v_\alpha^{(0)}+\frac{e}{M}E_\alpha^{(0)},\qquad P_\alpha^{(0)}=\frac{e}{v_a}v_\alpha^{(0)},\\\mathrm{div}\,(\mathbf{E}^{(0)}+4\pi\mathbf{P}^{(0)})=4\pi\delta e\delta(r).\end{gathered} \tag{22}$$

The solutions of these equations are

$$E_\alpha^{(0)}=\frac{\delta e}{\varepsilon_0}\frac{r_\alpha}{r^3},\qquad v_\alpha^{(0)}=\frac{e}{M\omega_0^2}\frac{\delta e}{\varepsilon_0}\frac{r_\alpha}{r^3}. \tag{23}$$

The solution of Eq. (19) can be expressed by means of the quadrature:[12]

$$\begin{gathered}\tilde{w}_\alpha^{(0)}=\frac{U_0}{(2\pi)^3}\frac{\partial}{\partial r_\alpha}\int d^3q\frac{\exp(-i\mathbf{q}\mathbf{r})}{c_{44}q^2+(c_{11}-3c_{44})q_\alpha^2}\frac{1}{1+f(\mathbf{q})},\\f(\mathbf{q})=\sum_\alpha\frac{2c_{44}q_\alpha^2}{c_{44}q^2+(c_{11}-3c_{44})q_\alpha^2}.\end{gathered} \tag{24}$$

Here we have used the Cauchy relation $c_{12}=c_{44}$.

We emphasize that Eqs. (20) were obtained under the assumption that the impurity ion itself does not participate in the oscillations described by this set of equations. This approximation should not affect the law of oscillation of ions located a distance of several lattice constants from the impurity ion. For the correct description of the oscillations of the ions in the immediate vicinity of the impurity center, which corresponds to deep levels of local oscillations, the continuum approximation may be completely inapplicable.

The third, fourth, and fifth terms of the right side of (20a) play the role of the potential energy and the second term the role of the kinetic energy. The presence in (20) of a macroscopic electric field would increase the order of the set of differential equations in comparison with what would have been the case for the electron spectrum.

3. TRANSFORMATION OF THE SET OF EQUATIONS OF MOTION

Simplification of the form of Eqs. (20) can be achieved by means eliminating from them the macroscopic electric field. For this purpose, we divide the vector v′ into irrotational and solenoidal parts:

$$\mathbf{v}'=\mathrm{grad}\,\Psi+\mathrm{rot}\,\mathbf{A}, \tag{25}$$

and apply to **A** the condition

$$\mathrm{div}\,\mathbf{A}=0. \tag{25a}$$

Taking it into account that $\mathbf{E} = -\operatorname{grad}\Psi$, we get from (20b) and (20a):

$$\Delta\varphi = \frac{4\pi e}{v_a}\Big\{\Delta\Psi - \operatorname{div}\Big[\operatorname{div}\Big(\mathbf{w}^{(0)} + \frac{M_- - M_+}{M_- + M_+}\mathbf{v}^{(0)}\Big)\operatorname{grad}\Psi\Big] - \sum_{\alpha,\beta}\frac{\partial}{\partial r_\beta}\Big(w_\alpha^{(0)} + \frac{M_- - M_+}{M_- + M_+}v_\alpha^{(0)}\Big)\frac{\partial^2\Psi}{\partial r_\alpha\,\partial r_\beta} - \sum_{\alpha,\beta}\frac{\partial}{\partial r_\beta}\Big[\frac{\partial}{\partial r_\alpha}\Big(w_\alpha^{(0)} + \frac{M_- - M_+}{M_- + M_+}v_\alpha^{(0)}\Big)\operatorname{rot}_\alpha\mathbf{A}\Big]\Big\}. \quad (26)$$

We now apply the divergence operation to both parts of Eq. (20a). Here we take into account the circumstance that the transverse and longitudinal branches of the oscillations are split, i.e., to find the shallow levels (less than $\omega_l - \omega_t$), one can omit all terms proportional to **A**. Using (26), we obtain

$$(\omega^2 - \varepsilon_0\omega_0^2)\,\Delta\Psi = -\sum_{\alpha,\beta}\frac{\partial^2}{\partial r_\alpha\,\partial r_\beta}\Pi_{\alpha\beta}(i\nabla)\Psi - e\frac{M_- - M_+}{M_- M_+}\sum_{\alpha,\beta}\frac{\partial E_\alpha^{(0)}}{\partial r_\beta}\frac{\partial^2\Psi}{\partial r_\alpha\,\partial r_\beta} - \omega_0^2(\varepsilon_0 - 1)\operatorname{div}\{(\operatorname{div}\mathbf{w}^{(0)})\operatorname{grad}\Psi\} - \sum_{\alpha,\beta,\gamma}\frac{\partial}{\partial r_\alpha}\Big\{\frac{\partial\Psi}{\partial r_\gamma}\Theta_{\alpha\beta\gamma}(i\nabla)\,w_\beta^{(0)}\Big\}. \quad (27)$$

We substitute in (27) the explicit expressions (5) and (12) for $\Pi_{\alpha\beta}(i\nabla)$ and $\Theta_{\alpha\beta\gamma}(i\nabla)$:

$$(\omega^2 - \omega_l^2)\,\Delta\Psi = -\omega_l^2a^2\Big(m\Delta + n\sum_\alpha\frac{\partial^4}{\partial r_\alpha^4}\Big)\Psi + \lambda_1\Delta\Psi\operatorname{div}\mathbf{w}^{(0)} - \omega_l^2a^3\sum_{\alpha,\beta}\frac{\partial^2\Psi}{\partial r_\alpha\,\partial r_\beta}\frac{\partial}{\partial r_\beta}\Big[\frac{r_\alpha}{r^3}\rho_\alpha(\mathbf{r})\Big] - \omega_0^2(\varepsilon_0 - 1)\operatorname{div}\{(\operatorname{div}\mathbf{w}^{(0)})\operatorname{grad}\Psi\} + \lambda_2\sum_{\alpha,\beta}\frac{\partial^2\Psi}{\partial r_\alpha^2}\frac{\partial w_\alpha^{(0)}}{\partial r_\alpha} + \Big(\lambda_2 - \lambda_1\frac{c_{11} - 3c_{44}}{c_{44}}\Big)\sum_\alpha\frac{\partial\Psi}{\partial r_\alpha}\frac{\partial^2 w_\alpha^{(0)}}{\partial r_\alpha^2}, \quad (28)$$

where

$$\rho_\alpha = \frac{1}{\omega_l^2a^3}\Big\{\frac{e\delta e}{\varepsilon_0}\frac{M_- - M_+}{M_+M_-} - 2\lambda_1\frac{r^3}{r_\alpha}w_\alpha^{(0)}\Big\},$$

$$m = -\frac{r + l - s/2}{\varepsilon_0}, \qquad n = -\frac{s}{\varepsilon_0}. \quad (29)$$

In (28) it was taken into account that the following equalities hold in all points of space except r = 0:

$$2\frac{\partial}{\partial r_\alpha}\operatorname{div}\tilde{\mathbf{w}}^{(0)} + \Delta\tilde{w}_\alpha^{(0)} = -\frac{c_{11} - 3c_{44}}{c_{44}}\frac{\partial^2\tilde{w}_\alpha^{(0)}}{\partial r_\alpha^2}$$

$$\operatorname{div}\mathbf{v}^{(0)} = \operatorname{div}\mathbf{E}^{(0)} = 0, \qquad \operatorname{div}\tilde{\mathbf{w}}^{(0)} = \operatorname{div}\tilde{\mathbf{w}}^{(0)}.$$

At the point **r** = 0, the obtained equation (28) is not valid, since it was assumed in all the calculations that the region $r > a$ is considered, while the region $r < a$ is unimportant. From the sense of this conclusion, Eq. (28) can be extended in the vicinity of the point r = 0, which is a problem of the microtheory (see also below).

Now it is necessary to find four boundary conditions for the differential equation (28). It is obvious that such conditions follow from the boundedness of the vector $\mathbf{v}'$ as $r \to \infty$. We thus find: 1) ψ increases as $r \to \infty$ no more rapidly than the linear function r; 2) $|\partial\psi/\partial r|$ is bounded as $r \to \infty$; 3) $|\partial^2\psi/\partial r^2|_{r\to\infty} = 0$; 4) $|\partial^3\psi/\partial r^3|_{r\to\infty} = 0$.

Similarly, one can obtain equations for the transverse branches of oscillations by applying the curl operation to both parts of (20a). We shall not write down this expression here because of its complexity. In the next section we shall write it down for the simplest case of isotropic propagation.

The expression (28) is very involved, and its exact solution is best sought by means of a high-speed computer. However, in the case of isotropic space, Eq. (28) becomes greatly simplified. The study of this case allows us to understand under what conditions the continuum approximation is applicable and when a large number of levels can be separated from the spectrum.

4. SOLUTION OF THE EQUATION OF MOTION AND THE DETERMINATION OF THE SPECTRUM OF LOCAL OSCILLATIONS FOR THE CASE OF ISOTROPIC PROPAGATION

In the first place, we shall assume that the space is elastically isotropic, i.e., $c_{11} = 3c_{44}$. Then the expression for $\tilde{w}_\alpha^{(0)}$ is easily obtained from (24):

$$\tilde{w}_\alpha^{(0)} = -\frac{U_0}{4\pi c_{11}}\frac{r_\alpha}{r^3}. \quad (30)$$

Correspondingly, the expression for ρ_α (29) is greatly simplified:

$$\rho_\alpha(\mathbf{r}) \equiv \rho = \frac{e\delta e}{\omega_l^2Ma^3}\Big\{\frac{1}{\varepsilon_0}\frac{M_- - M_+}{M_+ + M_-} + \lambda_1\Big[\frac{U_0M}{2\pi c_{11}e\delta e} + \frac{M_- - M_+}{M_- + M_+}\frac{1}{\omega_l^2}\Big]\Big\}. \quad (31)$$

Moreover, in the case of an elastically isotropic space, $\operatorname{div}\mathbf{w}^{(0)} = 0$.

Second, we shall assume that the anharmonic forces in the crystal are also isotropic, i.e., $\lambda_2 = 0$.

Finally, in the third place, we shall assume the spectrum of optical phonons to be isotropic: n = 0.

Of course, in real crystals, these relations are not satisfied. Thus, for example, in real crystals, n is never much less than the quantity m and for crystals of the NaCl type, to the contrary, $|n| \gg |m|$. As a rule, alkali halide crystals do not form an elastically isotropic medium. So far as the relation between λ_1 and λ_2 is concerned, $|\lambda_2| \gg |\lambda_1|$ in crystals of the NaCl type and $|\lambda_1| \sim |\lambda_2|$ for the CsCl type. Therefore, by the parameters m, λ_1 and ρ we must understand certain angle-averaged quantities. Thus, in the case of uniform averaging over the angles, we must take m, say, to mean the quantity $m + \frac{3}{5}n$.

After such simplifications, Eq. (28) takes the form

$$(\omega^2 - \omega_l^2)\,\Delta\Psi + a^2\omega_l^2m\Delta^2\Psi + \rho a^3\omega_l^2\frac{1}{r^3}\Big(\Delta\Psi - 3\frac{((\mathbf{r}\nabla)\nabla\Psi)\mathbf{r}}{r^2}\Big) = 0. \quad (32)$$

Similarly, one can obtain equations in the transverse branch of oscillations:

$$(\omega^2 - \omega_0^2)\,\Delta\mathbf{A} + a^2\omega_0^2l\Delta^2\mathbf{A} + \rho a^3\omega_l^2\frac{1}{r^3}\Big(\Delta\mathbf{A} - 3\frac{((\mathbf{r}\nabla)\operatorname{rot}\mathbf{A})\mathbf{r}}{r^2}\Big) = 0. \quad (33)$$

In what follows, we shall consider only Eq. (32), i.e., local oscillations that drop out of the longitudinal branch of the spectrum. For convenience, we introduce the nondimensional quantities

$$\varepsilon = (\omega^2 - \omega_l^2)\,/\,m\omega_l^2, \quad \rho' = -\rho\,/\,m, \quad z = r\,/\,a. \quad (34)$$

Then (32) takes the form

$$\varepsilon\Delta\Psi + \Delta^2\Psi + \frac{\rho'}{z^3}\Big[\Delta\Psi - 3\frac{\partial^2\Psi}{\partial z^2}\Big] = 0. \quad (35)$$

We solve this equation by the substitution $\Psi = R(z)\,O$, where O depends only on the angular coordinates and is an eigenfunction of the angular part of the Laplacian. The equation for R(z) has the form

$$R^{\mathrm{IV}}+\frac{4}{z}R'''+\left[\frac{2l(l+1)}{z^2}+\frac{2\rho'}{z^3}+\varepsilon\right]R''-2\left(\frac{\rho'}{z^4}-\frac{\varepsilon}{z}\right)R' + \frac{l(l+1)}{z^2}\left[\frac{l(l+1)+2}{z^2}-\frac{\rho'}{z^3}+\varepsilon\right]R=0, \qquad (36)$$

where $l = 0, 1, 2, \ldots$.

Finding the spectrum from (36) is a problem that is extremely complicated, since this equation is of fourth order, not belonging to the Fuchs class. Therefore, it is necessary to solve even such a problem with a high-speed computer. But for the purpose of illustration, we consider several special cases which allow us to make judgments on the general picture of the location of the levels of local oscillations. Inasmuch as the angular dependence of the considered equations was strongly deformed in the transition to the isotropic space, (36) quite reliably describes the case of spherically symmetric state, i.e., the state with $l = 0$. In the case $l = 0$, Eq. (36) can be appreciably simplified. By introducing the new variable $y = R'$, we obtain

$$y'''+\frac{4}{z}y''+\left(\frac{2\rho'}{z^3}+\varepsilon\right)y'-2\left(\frac{\rho'}{z^4}-\frac{\varepsilon}{z}\right)y=0. \qquad (37)$$

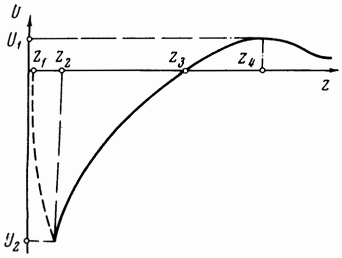
We represent (37) in the form

$$\frac{1}{z^2}\frac{d}{dz}\left\{z^2\left[y''+\frac{2}{z}y'+\left(\frac{2\rho'}{z^3}-\frac{2}{z^2}+\varepsilon\right)y\right]\right\}=0. \qquad (38)$$

Carrying out the integration, and assuming the integration constant to be equal to zero (by virtue of the boundary conditions), we obtain a differential equation of second order which, by the substitution $y = \chi/z$, is converted to the normal form:

$$\chi''+\left(\frac{2\rho'}{z^3}-\frac{2}{z^2}+\varepsilon\right)\chi=0. \qquad (39)$$

Inasmuch as the effective potential energy as $z \to 0$ is proportional to z^{-3}, the solution of Eq. (39) corresponds to the falling of the particle onto the center. However, from the sense of the derivation of (39), it retains its form only if $z > 1$, and its form in the region $z \lesssim 1$ can be obtained only from the microtheory. Having the purpose only of illustrating the fact that there can be many levels of the local oscillations, while χ falls off when $z \gg 1$ (when $|\epsilon| < 1$), we use the following method. In the region $0 < z < \gamma \sim 1$, we formally choose the shape of the potential energy so that it does not correspond to the falling of the particle onto the center. In what follows it will be seen that the final result does not depend on the quantity γ, if $\gamma \lesssim 1$.



Dependence of the "potential energy" $U = 9/4z^2 - 2\rho'/z^3$ on z. The dashed curve shows the path of U for $z < \gamma$ with account of "cut-off"; $U_1 = (81/128)\rho'$, $U_2 = 9/4\gamma^2 - 2\rho'/\gamma^3$, $z_1 = (9\gamma^3/8\rho')^{1/2}$, $z_2 = \gamma$, $z_3 = (8/9)\rho'$, $z_4 = (4/3)\rho'$.

It is necessary to distinguish two cases: $\rho' > 0$ and $\rho' < 0$. In the first case, (39) describes a localized state, and in the second, the scattering of the "particle," i.e., the defect is a scattering center for the long-wave phonons. We consider the case $\rho' > 0$, i.e., the formation of local oscillations in the presence of the long-range defect.

To estimate the number of levels of local oscillations, we use the WKB method. The radial part of the momentum p_r is, according to (39)

$$p_r=\left(\frac{2\rho'}{z^3}-\frac{9}{4z^2}+\varepsilon\right)^{1/2}, \quad z > \gamma \sim 1. \qquad (40a)$$

In the region $0 < z < \gamma \sim 1$, in correspondence with what was shown above, we set

$$p_r=\left(\frac{2\rho'}{\gamma^3}-\frac{9}{4z^2}+\varepsilon\right)^{1/2}, \quad 0 < z < \gamma \sim 1. \qquad (40b)$$

The form of the potential energy (see the drawing) indicates that in the region $0 < \epsilon$ $(81/128)\rho'$ levels of "quasi-local" oscillations can exist, i.e., not coming from the continuous spectrum.

We compute the number of levels of the local oscillations. According to the WKB method, the frequency of the local oscillations can be found from the relation

$$\int_{z_1}^{z_2} p_r\,dz=\pi\left(n+\frac{1}{2}\right), \qquad (41)$$

where z_1 and z_2 are the "turning points," and n is an integer. The number of levels of the local oscillations n_0', located in the range

$$\frac{9}{4\gamma^2}-\frac{2\rho'}{\gamma^3}<\varepsilon<0,$$

can be determined from (41) by setting $\epsilon = 0$ in p_r. Carrying out the integration over z, we have

$$n_0'=(2/\pi)\sqrt{2\rho'/\gamma}. \qquad (42)$$

In (42), we have limited ourselves to the lowest approximation in the parameter ρ'; here the contribution from the lower limit of integration can be neglected.

It is seen from the drawing that the dimensionless radius of the local state (in units of a) is of the order of ρ', i.e., the condition for the applicability of the theory that has been considered is the criterion

$$\rho' \gg 1. \qquad (43)$$

In real situations, the condition (43) is satisfied because of two factors. First, the number m characterizing the width of the dispersion of the phonon band is usually small (of the order of 0.1). Second, the value of the parameter entering into the determination of ρ' (34) depends on the quantity $a^2\varphi'''/\varphi''$, where φ is the potential energy of the interionic interaction, the prime denotes differentiation with respect to the square of the lattice constant, while the value of this parameter is of the order of 10. Thus, one must expect that the value of ρ' in alkali-halide crystals is of the order of 100.

The value of n_0' (42) depends on γ. However, the levels calculated from (42), include not only shallow levels but also some rather deep ones, for which ϵ

$\sim \rho'$. Such levels of local oscillations can obviously not be found by means of the method developed, since their depth is of the order of ω_0 and consequently all phonon bands participate in their formation, and a method analogous to the effective-mass method is not applicable for their calculation. Furthermore, the radius of their state is of the order of unity, for which reason the continuum approximation is inapplicable. Therefore, for the correct calculation of the levels of local oscillations under consideration, it is necessary to subtract from (42) the number of these deep-lying levels (we denote them by n_0''), which lie below the level $\epsilon = \epsilon_0$, and $\epsilon_0 \sim 1$. We denote the right-hand turning point (in the region $z > \gamma$) by $z'(\epsilon_0)$. In lowest order in ρ', we have $z' = (-2\rho'/\epsilon_0)^{1/3}$. The number of local oscillations lying in the energy range $\epsilon_0 > \epsilon > 9/4\gamma^2 - 2\rho'/\gamma^3$ can be found from (41) while, in integration over z in the expression for p_r (40a) in the lowest order in ρ', one can omit the term $9/4z^2$ in the radicand. As a result of such integration, we get

$$n_0'' = \frac{2}{\pi}\sqrt{\frac{2\rho'}{\gamma}} - \frac{2}{\pi}\sqrt{\frac{2\rho'}{z'}}. \tag{44}$$

Subtracting the expression (44) for n_0'' from (42), we obtain the number of levels of local oscillations contained in the energy range $0 > \epsilon > \epsilon_0$:

$$n_0 = n_0' - n_0'' = \frac{2}{\pi}\sqrt{\frac{2\rho'}{z'}} = \frac{2}{\pi}(-\varepsilon)^{1/6}(2\rho')^{1/3}. \tag{45}$$

Thus n_0 does not depend on γ and is proportional to the cube root of the large parameter ρ'. We note that (45) is a minimal estimate for n_0, since for each n there are still several values of the orbital numbers l. However, in contrast with the hydrogen atom, there is no degeneracy in l here.

5. DISCUSSION OF THE RESULTS

Thus, with the help of the continuum approximation, we have obtained the differential equations (20), the eigenvalues of which determine the spectrum of local oscillations of the crystal lattice in the presence of a long-range defect. Principal attention has been given in the present research to the case of a charged defect in an ionic crystal. However, the method put forth allows us, in principle, to find the shallow local levels both in the case of an electrically neutral defect in the ionic crystal and also in the case of a defect in homopolar crystals. So far as we know, such a problem has never been proposed by anyone. In the case of a neutral defect in an ionic crystal, one needs to set $\delta_e = 0$ in (6), which leads to the equation $v^{(0)} = E^{(0)} = 0$. In the case of a homopolar crystal, it is necessary to omit all terms in (20a) connected with the electric field (i.e., the first, third, fifth terms of the right-hand side). Only the last term of the right side of (20a) is responsible for the presence of such local oscillations in homopolar crystals. Equations (20b) and (20c) need not be considered in this case. The appearance of local oscillations of large radius, due to a defect of small radius (of the order of the lattice constant) is connected with the fact that, because of anharmonic forces, this defect can interact, through the nearest neighbors, with an atom of the lattice located at an appreciable distance from it. However, in the case of cubic homopolar crystals, it is impossible to decouple the three differential equations, since in the absence of an electric field in the crystal, the optical phonon bands are degenerate at the point $\mathbf{q} = 0$.

In the case of ionic crystals, these equations can be decoupled into longitudinal and transverse, using a method similar to the effective-mass method. However, the resultant equation for the longitudinal branch is more complicated than in the case of the calculation of electronic spectra by the effective-mass method for a nondegenerate band. In particular, a fourth-order equation is obtained (see (28)), while in the case of electronic spectra, the corresponding equation is of second order. Similar complications occur also for the doubly degenerate transverse branches, where a set of two equations of fourth order is obtained, and not second order as in the case of electronic spectra. The reason for such a complication is the presence of an electric field in the ionic crystal (we recall that because of the presence of the electric field, the spectrum of phonons is more involved in the region of small wave numbers[5,6]).

The criterion for the applicability of the developed method of calculation is the inequality (43), which holds for sufficiently narrow width of the dispersion of the phonon band and for sufficiently strong anharmonic force in the ion-ion interaction. The sign of the quantity ρ' determines where the defect is "attractive," i.e., forming a local state or "repulsive," i.e., a scattering center for long-wave phonons. In the first case, this defect should affect the optical properties of the crystal and in the second, its thermal conductivity. We note that in the second case, the defect can be a very effective scattering center for long-wave phonons, since its cross section for long-wave phonons can be very large, much larger than the square of the lattice constant.

APPENDIX

DERIVATION OF THE STABILITY EQUATION (19)

In correspondence with the ideas of transition to the continuum approximation, we write down the harmonic force (7), replacing the discrete variable $r(l)$ by the continuous coordinate $\mathbf{r}$:

$$F_\alpha{}^h\binom{\mathbf{r}}{} = -\frac{1}{v_a}\sum_{k',\beta}\int_V d^3r'\left[u_\beta\binom{\mathbf{r}}{k} - u_\beta\binom{\mathbf{r}'}{k'}\right]\frac{\partial^2}{\partial r_\alpha\,\partial r_\beta}\Psi_{kk'}(|\mathbf{r}-\mathbf{r}'|^2). \tag{A.1}$$

We now expand $u\binom{\mathbf{r}}{\mathbf{k}'}$ in (A.1) in a series in $\mathbf{r}'$ around $\mathbf{r}' = \mathbf{r}$:

$$u_\alpha\binom{\mathbf{r}'}{k'} \approx u_\alpha\binom{\mathbf{r}}{k'} + \sum_\beta \frac{\partial}{\partial r_\beta}u_\alpha\binom{\mathbf{r}}{k'}(\mathbf{r}'-\mathbf{r})_\beta + \frac{1}{2}\sum_{\beta,\gamma}\frac{\partial^2}{\partial r_\beta\,\partial r_\gamma}u_\alpha\binom{\mathbf{r}}{k'}(\mathbf{r}-\mathbf{r}')_\beta(\mathbf{r}-\mathbf{r}')_\gamma + \ldots \tag{A.2}$$

Substituting the expansion (A.2) in (A.1), we find the force acting in the acoustic branch of oscillations:

$$F\binom{r}{ak} = F\binom{r}{+} + F\binom{r}{-},$$

where

$$F_\alpha\begin{pmatrix}\mathbf{r}\\ ak\end{pmatrix}=\frac{1}{v_a}\sum_{\alpha,\gamma,\nu}\Bigg\{\frac{\partial^2\tilde{w}_\beta(\mathbf{r})}{\partial r_\gamma\,\partial r_\nu}\int_V d^3r'\,r_\gamma'r_\nu'\,\frac{\partial^2}{\partial r_\alpha'\,\partial r_\beta'}\Psi_{+-}(r'^2)$$

$$+\frac{\partial^2}{\partial r_\gamma\,\partial r_\nu}u_\beta\begin{pmatrix}\mathbf{r}\\ +\end{pmatrix}\int_V d^3r'\,r_\gamma'r_\nu'\,\frac{\partial^2}{\partial r_\alpha'\,\partial r_\beta'}\Psi_{--}(r'^2)$$

$$+\frac{\partial^2}{\partial r_\gamma\,\partial r_\nu}u_\beta\begin{pmatrix}\mathbf{r}\\ -\end{pmatrix}\int_V d^3r'\,r_\gamma'r_\nu'\,\frac{\partial^2}{\partial r_\alpha'\,\partial r_\beta'}\Psi_{++}(r'^2)\Bigg\}. \quad (A.3)$$

Generally speaking, $\Psi_{++} \neq \Psi_{--}$. However, in this case, when it suffices in the short-range case to restrict ourselves to the interaction only with nearest neighbors, i.e., $\Psi_{++} = \Psi_{--} = 0$, (A.3) is simplified:

$$F_\alpha\begin{pmatrix}\mathbf{r}\\ ak\end{pmatrix}=-(M_+ + M_-)\sum_\beta L_{\alpha\beta}(i\nabla)\tilde{w}_\beta(\mathbf{r}). \quad (A.4)$$

According to elasticity theory for cubic crystals, we have

$$L_{\alpha\beta}(\mathbf{q})=\rho^{-1}\{q_\alpha q_\beta(c_{12}+c_{44})$$
$$+\delta_{\alpha\beta}[q^2c_{44}+q_\alpha^2(c_{11}-c_{12}-2c_{44})]\}, \quad (A.5)$$

where ρ is the density of the crystal. Formulas (A.4) and (A.5) also show the validity of Eq. (19).

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