Vibrational spectrum of a crystal with impurities in the presence of localized states near the band edge

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The density of vibrational states is studied in a crystal with impurities that result in the appearance of localized states near the edge of the unperturbed spectrum ω_m. A group expansion of the one-particle Green's function and the density of states with respect to complexes of interacting impurity centers is obtained. In the case of small impurity concentrations $c \ll c_0$ (c_0 is a characteristic concentration), the group expansions converge in a broad region in the vicinity of ω_m, excluding the region of concentration broadening of the local oscillations. The density of states is found through the region of convergence of the group expansions, including the transition region between states which can be described approximately by plane waves and fluctuation states caused by pairs of impurity atoms located at distances which are smaller than the mean distance. The density of states is also considered in the case of sufficiently high concentrations $c \gg c_0$ (but $c \ll 1$). The results are compared with those of the coherent-potential technique.

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Localized vibrational states of a crystalline lattice which contains isolated impurity atoms have by now been well studied.[1,2] It is of interest to consider the behavior of the vibrational spectrum of a crystal for sufficiently high impurity concentrations c, when the effects of interaction between them begin to play a part (however, $c \ll 1$, so that the interaction is essentially via exchange of virtual phonons and we can neglect the direct interaction between the impurities). Qualitative and, in certain cases, quantitative consideration of such effects has been carried out by a number of authors,[3–6] An extensive range of problems devoted to the structure of the energy spectrum has been studied by I. Lifshitz[3] (broadening of the local level, shift of the band edge, structure of the spectrum near its actual boundary, and so on). An equivalent approach has been developed by Pokrovskiĭ and co-workers[5] for the electron spectrum in the presence of localized states near the bottom of the band.

In the present research, we have obtained a group expansion of the one-particle Green's function of the oscillations of a crystal with respect to complexes of interacting impurity atoms (similar to the way in which the group expansion of the sum over states is obtained in the Maier theory of a nonideal gas). Such an approach, in contrast to the previous cases, allows us to calculate, in a unique systematic way, the density of states over the entire range of frequencies considered, including the transition region between the fluctuation states and the states that are described approximately by plane waves.

Impurity centers are considered which lead to the appearance of localized states near the edge of the unperturbed spectrum, so that the characteristic concentration

$$c_0 = \left| \frac{\omega_0{}^2 - \omega_m{}^2}{\Omega^2} \right|^{3/2} \ll 1$$

(ω_0 is the frequency of the localized excitation of the isolated defect; the dispersion law near the edge of the spectrum is of the form $\omega_k^2 = \omega_m^2 - \Omega^2 (ak)^2$, a is the lattice constant, and the wave vector **k** is measured from the extremal points of the spectrum).

At low concentrations ($c \ll c_0$), the obtained spectral density describes successively states having the character of plane waves, then states intermediate between plane waves and fluctuations and, finally, states due to pairs of neighboring impurities. The state of relatively high concentrations, $c \gg c_0$ (but $c \ll 1$), is also considered.

In the present work, we have, for simplicity, considered a single-atom cubic crystal, containing isotopic substitution impurities. To be able to consider independently all three branches of the vibrational spectrum, a model of the crystal is used with equal central and non-central interaction forces between nearest neighbors. However, the results remain in force also for more realistic models, and after corresponding transformations they can also be used to describe impurities in electron, exciton and other spectra of elementary excitations of nonideal crystals.

THE GREEN'S FUNCTION AND THE DENSITY OF VIBRATIONAL STATES IN A CRYSTAL WITH IMPURITIES

In the harmonic approximation, the density of vibrational states of a crystal can be represented in the form

$$g(\omega^2) = \frac{1}{3\pi N} \operatorname*{Im}_{\delta\to 0} \sum_{l,\alpha} M_l \langle\langle u_{l,\alpha} | u_{l,\alpha} \rangle\rangle^{\omega - i\delta}, \tag{1}$$

where $M_l = M$, M' is the mass of the matrix atom and the impurity atom, $u_{l\alpha}$ is the displacement of the l-th atom in the direction α, N is the number of sites in the crystal, and the normalization condition is

$$\int g(\omega^2)\, d\omega^2 = 1.$$

The advanced Green's function is defined as usual:

$$\langle\langle A | B \rangle\rangle^{\omega - i\delta} = i \int_{-\infty}^{0} \langle [A(t), B(0)] \rangle e^{i\omega t + \delta t}\, dt$$

(the index $\omega - i\delta$ will be omitted below).

We expand the displacements in normal vibrations of unperturbed crystal:

$$u_{l,\alpha}=\sum_{\mathbf{q},j} e_{\alpha\mathbf{q}j}\exp(i\mathbf{q}\mathbf{R}_l)B_{\mathbf{q}j},$$

$$B_{\mathbf{q}j}=(2MN\omega_{\mathbf{q}j})^{-1/2}(a_{\mathbf{q}j}+\overset{+}{a}_{-\mathbf{q}j}),\qquad [a_{\mathbf{q}j},\overset{+}{a}_{\mathbf{q}'j'}]=\delta_{\mathbf{q}\mathbf{q}',jj'};$$

here $\omega_{\mathbf{q}j}$ and $e_{\mathbf{q}j}$ are the frequency and the polarization vector of the phonon of the j-th branch with the wave vector $\mathbf{q}$, and $\mathbf{R}_l$ is the radius vector of the l-th site. The density of states is then divided into two parts:

$$g(\omega^2)=g_1(\omega^2)+g_2(\omega^2),$$

$$g_1(\omega^2)=\frac{M}{3\pi}\operatorname{Im}\sum_{\mathbf{q},j}\langle\langle B_{\mathbf{q}j}|B_{-\mathbf{q}j}\rangle\rangle, \tag{2}$$

$$g_2(\omega^2)=\frac{\varepsilon M}{3\pi N}\operatorname{Im}\sum_{s,\alpha}\langle\langle u_{s,\alpha}|u_{s,\alpha}\rangle\rangle;$$

$\varepsilon=(M-M')/M$, and the index s runs only over the sites occupied by the impurity.

The Hamiltonian of the crystal has in the harmonic approximation the form

$$\mathscr{H}=\mathscr{H}_0+\mathscr{H}_1,\qquad \mathscr{H}_0=\sum_{\mathbf{q}j}\omega_{\mathbf{q}j}\overset{+}{a}_{\mathbf{q}j}a_{\mathbf{q}j},$$

$$\mathscr{H}_1=\frac{\varepsilon}{\varepsilon-1}\frac{1}{4N}\sum_{\mathbf{q}j\mathbf{q}'j'}\sum_{s,\alpha,\alpha'}e_{\alpha\mathbf{q}j}e_{\alpha'\mathbf{q}'j'}\exp[i(\mathbf{q}+\mathbf{q}')\mathbf{R}_s]$$
$$\times(\omega_{\mathbf{q}j}\omega_{\mathbf{q}'j'})^{1/2}(a_{\mathbf{q}j}-\overset{+}{a}_{-\mathbf{q}j})(a_{\mathbf{q}'j'}-\overset{+}{a}_{-\mathbf{q}'j'}). \tag{3}$$

Writing down the chain of equations for the function $\langle\langle B_{\mathbf{q}j}|B_{-\mathbf{q}j}\rangle\rangle$, and expanding the polarization operator in the groups of interacting impurities, similar to what was done in[7], we get

$$\langle\langle B_{\mathbf{q}j}|B_{-\mathbf{q}j}\rangle\rangle=(MN)^{-1}(\omega^2-\omega_{\mathbf{q}j}^2-R_{\mathbf{q}j})^{-1}, \tag{4}$$

where

$$R_{\mathbf{q}j}=\frac{c\varepsilon\omega^2}{D}\left(1+c\sum_{l\neq0}\frac{A_{0l}e^{i\mathbf{q}\mathbf{R}_l}+A_{0l}A_{l0}}{1-A_{0l}A_{l0}}+\ldots\right),$$

$$D=1-\frac{\varepsilon\omega^2}{3N}\sum_{\mathbf{k}\neq-\mathbf{q},j'}(\omega^2-\omega_{\mathbf{k}j'}^2-R_{\mathbf{k}j'})^{-1},$$

$$A_{0l}=\frac{\varepsilon\omega^2}{3ND}\sum_{\mathbf{k}\neq-\mathbf{q},j'}e^{i\mathbf{k}\mathbf{R}_l}(\omega^2-\omega_{\mathbf{k}j'}^2-R_{\mathbf{k}j'})^{-1}; \tag{5}$$

c is the impurity concentration, and the summation in the expressions $A_{0l}A_{l0}$ is taken over the non-coinciding wave vectors. It should be noted that the diagonal of the Green's function $\langle\langle B_{\mathbf{q}j}|B_{-\mathbf{q}j}\rangle\rangle$ and the polarization operator $R_{\mathbf{q}j}$ are self-averaging quantities; therefore, we can average directly the quantity $R_{\mathbf{q}j}$ over the random distribution of impurities. The second term in the brackets in the first of Eqs. (5) determines the contribution from all possible pairs of interacting impurities and the omitted components correspond to groups of three and more impurities. As is seen from (4) and (5), the expressions for D and A_{0l} contain the polarization operator $R_{\mathbf{q}j}$, and therefore the completely renormalized representation (4), (5) forms a self-consistent set of equations for the determination of $R_{\mathbf{q}j}$.

By modifying the procedure of obtaining the equations for the Green's functions $\langle\langle B_{\mathbf{q}j}|B_{-\mathbf{q}j}\rangle\rangle$, we can obtain its other representation:

$$\langle\langle B_{\mathbf{q}j}|B_{-\mathbf{q}j}\rangle\rangle=(MN)^{-1}(\omega^2-\omega_{\mathbf{q}j}^2)^{-1}(1+\tilde{R}_{\mathbf{q}j}(\omega^2-\omega_{\mathbf{q}j}^2)^{-1}), \tag{6}$$

where

$$\tilde{R}_{\mathbf{q}j}=\frac{c\varepsilon\omega^2}{D_0}\left(1+c\sum_{l\neq0}\frac{\tilde{A}_{0l}e^{i\mathbf{q}\mathbf{R}_l}+\tilde{A}_{0l}\tilde{A}_{l0}}{1-\tilde{A}_{0l}\tilde{A}_{l0}}+\ldots\right),$$

$$D_0=1-\frac{\varepsilon\omega^2}{3N}\sum_{\mathbf{k},j'}(\omega^2-\omega_{\mathbf{k}j'}^2)^{-1},$$

$$\tilde{A}_{0l}=\frac{\varepsilon\omega^2}{3ND_0}\sum_{\mathbf{k},j'}e^{i\mathbf{k}\mathbf{R}_l}(\omega^2-\omega_{\mathbf{k}j'}^2)^{-1}; \tag{7}$$

the expressions for D_0, $\tilde{A}_{0l}$, and $\tilde{A}_{0l}\tilde{A}_{l0}$ do not contain limitations in the summation over $\mathbf{k}$. The first two terms of the renormalized representation obtained here for $R_{\mathbf{q}j}$ (7) are identical in form with the corresponding expression (5) for $R_{\mathbf{q}j}$; however, the next terms will be different. In the derivation of expressions (4)–(7) it was taken into account that, within the framework of the chosen model wherein forces of the central and noncentral interaction are equal, three branches of the vibrational spectrum of the nonideal crystal remain independent and are not entangled with one another. One representation or the other will be used in what follows, depending on the considered branch of frequencies. In similar fashion, we can find the expression for $g_2(\omega^2)$; however, it turns out that for $c\ll1$ and $c_0\ll1$ the contribution $g_2(\omega^2)$ can be neglected everywhere in comparison with $g_1(\omega^2)$.

DENSITY OF STATES FOR LOW IMPURITY CONCENTRATIONS

1. Let the frequency ω_0 of the local oscillation (LO) of the impurity center, which is determined by the equation

$$\operatorname{Re}D_0(\omega_0^2)=0, \tag{8}$$

lie outside the continuous spectrum, close to its edge, so that the characteristic concentration $c_0\ll1$,

$$c_0=\left|\frac{\omega_0^2-\omega_m^2}{\Omega^2}\right|^{3/2}. \tag{9}$$

It is assumed here that the dispersion law, close to the band edge, is of the form

$$\omega_{\mathbf{k}j}^2=\omega_{\mathbf{k}}^2=\omega_m^2-\Omega^2(ak)^2 \tag{10}$$

for all three branches of the unperturbed spectrum. We shall call small such concentrations for which $c\ll c_0$. Here the mean distance between the nearest impurity centers $\bar{r}\approx ac^{-1/3}$ is much greater than the radius $r_0\sim ac_0^{-1/3}$ of the spatial distribution of the LO.

Local oscillations arise in the system when the solution of Eq. (8) lies outside the continuous spectrum of the unperturbed crystal ($\omega_0^2>\omega_m^2$). Here, one or another of the group expansions (4) and (6) for the Green's function is convergent throughout the entire range of frequencies with the exception of an immediate vicinity of the LO frequency ω_0, of width $\Delta_0/2\omega_0$ ($\Delta_0\sim c_0^{2/3}\Omega^2\times\exp[-\vartheta(c_0/c)^{1/3}]$, $\vartheta\sim1$), and also the region of the actual boundary of the spectrum of the considered crystal. In the region Δ_0 of concentration broadening of the

LO frequency, the density of states is determined by a large group of impurity centers located at distances that are greater than or of the order of $\bar{r}$, and near the actual boundary it is necessary to take into account the fluctuations of the cluster of impurity centers; this question was studied in detail in[3].

Outside of the region Δ_0, but still very close to ω_0, the renormalized representation (6) and (7) converges better (in terms of the parameter c/c_0). Substituting (6) and (7) in (2), transforming in $R_{\mathbf{k}j}$, from summation over l to integration, and recognizing that in the region $0<\omega^2-\omega_m^2\ll\Omega^2$, $|\omega^2-\omega_0^2|\gg\Delta_0$, we have

$$D_0=\frac{\varepsilon\omega^2}{4\pi\Omega^2}\left[\left(\frac{\omega^2-\omega_m^2}{\Omega^2}\right)^{1/2}-c_0^{1/3}\right],$$

$$A_{0l}=\frac{\varepsilon\omega^2 a}{4\pi\Omega^2 D_0 R_l}\exp\left(-\varkappa\frac{R_l}{a}\right),\quad \varkappa=\left(\frac{\omega^2-\omega_m^2}{\Omega^2}\right)^{1/2}, \tag{11}$$

we obtain the following expression for the density of states in the given range of frequencies:

$$g(\omega^2)=g_1(\omega^2)=\frac{\pi c^2R_\omega^3[1+\operatorname{sign}(\omega^2-\omega_0^2)\exp(-\varkappa R_\omega/a)]}{\Omega^2a^3\varkappa|\varkappa-c_0^{1/3}|(1+\varkappa R_\omega/a)}. \tag{12}$$

The principal contribution to the density of states for the given frequency is due here to the pairs of impurities found at such a distance $R_l=R_\omega$ that $|\tilde{A}_{0l}|=1$:

$$R_\omega=a|\varkappa-c_0^{1/3}|^{-1}\exp(-\varkappa R_\omega/a). \tag{13}$$

Since $a\ll R_\omega\ll ac^{-1/3}$ here and, in addition, it is assumed that the broadening of the discrete levels corresponding to different l is larger than the distances between the neighboring levels, one can actually replace summation over l by integration.

Near the boundary of the continuous spectrum (for $\varkappa\ll c_0^{1/3}$), as is seen from (13), $R_\omega\approx ac_0^{-1/3}$, and

$$g(\omega^2)=\pi c^2/c_0^{5/3}\Omega^2. \tag{14}$$

As $\varkappa\to 0$, the unrenormalized representation (6) and (7) ceases to converge, while the completely renormalized representation converges in the range $\omega^2<\omega_0^2-c^{1/2}c_0^{1/6}\Omega^2$. We note that when $\omega_m^2<\omega^2<\omega_0^2-c^{1/2}c_0^{1/6}\Omega^2$, both employed representations converge and lead to identical expressions for the density of states.

As has already been noted, when finding the density of states $g_1(\omega^2)$ with the help of the renormalized group expansion it is necessary to solve the self-consistent set of equations (4) and (5). For this purpose, we use the following method: We represent $g_1(\omega^2)$ in the form

$$g_1(\omega^2)=g_3(\omega^2)+g_4(\omega^2), \tag{15}$$

$$g_3(\omega^2)=\frac{1}{\pi N}\operatorname{Im}\sum_{\mathbf{k}}(\omega^2-\omega_k^2-R_{\mathbf{k}=0})^{-1}, \tag{16}$$

$$g_4(\omega^2)=\frac{1}{\pi N}\operatorname{Im}\sum_{\mathbf{k}}\left(\frac{1}{\omega^2-\omega_{\mathbf{k}}^2-R_{\mathbf{k}}}-\frac{1}{\omega^2-\omega_{\mathbf{k}}^2-R_{\mathbf{k}=0}}\right). \tag{17}$$

In the polarization operator $R_{\mathbf{k}}$, we separate the part that depends on **k**:

$$R_{\mathbf{k}}=R_1+R_{2,\mathbf{k}},\qquad R_1=c\varepsilon\omega^2/D,$$

$$R_{2,\mathbf{k}}=\frac{c\varepsilon\omega^2}{D}\sum_{l\neq 0}\frac{A_{0l}e^{i\mathbf{k}\mathbf{R}_l}+A_{0l}A_{l0}}{1-A_{0l}A_{l0}}. \tag{18}$$

It is not difficult to see that $R_{2,\mathbf{k}}$ differs appreciably from $R_{2,\mathbf{k}=0}$ for $k\gtrsim R_\omega^{-1}\sim c_0^{1/2}$ and just such **k** gives the basic contribution to $g_4(\omega^2)$. Taking it into account that here also $\omega^2-\omega_{\mathbf{k}}^2-R_1\gg R_{2,\mathbf{k}}$, we get

$$g_4(\omega^2)=\frac{1}{\pi N}\operatorname{Im}\sum_{\mathbf{k}}\frac{R_{2,\mathbf{k}}-R_{2,\mathbf{k}=0}}{(\omega^2-\omega_{\mathbf{k}}^2-R_1)^2}. \tag{19}$$

Apart from insignificant corrections, the calculation of $g_4(\omega^2)$ leads to the expression (12) in the region of convergence, if we substitute

$$\omega_m^2\to\tilde{\omega}_m^2\equiv\omega_m^2+\operatorname{Re}R_1=\omega_m^2-4\pi cc_0^{-1/3}\Omega^2$$

everywhere in it.

Using the dispersion law (10) for the calculation of the integral in (16), we transform the self-consistent equation (16) for $g_3(\omega^2)$ into the following algebraic equation:

$$g_3^4-C_1Lg_3^2-{}^1/_4C_1^2M^2=0, \tag{20}$$

$$L=\frac{\omega_m^2-\omega^2}{\Omega^2}+\frac{c\varepsilon\omega^2}{\Omega^2}\frac{\operatorname{Re}D}{(\operatorname{Re}D)^2+[\pi\varepsilon\omega^2(g_3+g_4)]^2}$$

$$M=\frac{\pi c(\varepsilon\omega^2)^2}{\Omega^2}\frac{g_3+g_4}{(\operatorname{Re}D)^2+[\pi\varepsilon\omega^2(g_3+g_4)]^2}+\frac{\operatorname{Im}R_{2,\mathbf{k}=0}}{\Omega^2}$$

$$C_1=(4\pi^2\Omega^2)^{-2}.$$

In the entire range of frequencies, only one of the roots of Eq. (20) is real and positive. Far from the renormalized "edge" of the spectrum $\tilde{\omega}_m$, when the condition

$$\omega^2-\tilde{\omega}_m^2\gg\Delta_1,\quad \Delta_1=4\pi^2c^2c_0^{-4/3}\ \Omega^2,$$

is satisfied, $g_3(\omega^2)$ has the order of magnitude $c^3/c_0^{7/3}\tilde{\varkappa}$, where $\tilde{\varkappa}^2=(\omega^2-\tilde{\omega}_m^2)/\Omega^2$, and in this region, the principal contribution to $g(\omega^2)$ is made by $g_4(\omega^2)$. As the transition region $|\omega^2-\tilde{\omega}_m^2|\sim\Delta$ is approached, the density of states begins to increase sharply, and in the limits of this region, as well as wherever $\omega^2<\tilde{\omega}_m^2$, one must use, generally speaking, the expression (15), where $g_3(\omega^2)$ is determined from Eq. (20). The general form of the density of states in the case of LO is represented in Fig. 1.

If $c\ll c_0/2\pi^2$, then, at $\omega^2<\tilde{\omega}_m^2+\Delta_1$, we can neglect the quantity $g_4(\omega^2)$ in (15), and then

$$g(\omega^2)\approx g_3(\omega^2)=\frac{1}{4\pi^2\Omega^2}\left[\frac{\omega_m^2-\omega^2+\Delta_1}{\Omega^2}-\frac{cc_0^{1/3}}{c_0^{2/3}+\varkappa^2}\right]^{1/2}. \tag{21}$$

We have discarded here terms that make a contribution proportional to c^3 to $g_3(\omega^2)$.

As we move away from the transition region, the expression (21) approaches the density of states of the unperturbed crystal asymptotically. In the case in which $\tilde{\omega}_m^2-\omega^2\gg\Delta_1$, the vibrational states can be described approximately by the plane waves[3,5] with frequency

$$\omega(\mathbf{k})=\left[\omega_{\mathbf{k}}^2-\frac{4\pi cc_0^{1/3}\Omega^2}{c_0^{2/3}+(\omega_m^2-\omega_{\mathbf{k}}^2)/\Omega^2}\right]^{1/2} \tag{22}$$

and with a damping

$$\gamma_{\mathbf{k}}=\frac{\operatorname{Im}R_{\mathbf{k}}}{2\omega_{\mathbf{k}}}=\frac{8\pi^3c\Omega^4g_3(\omega_{\mathbf{k}}^2)}{\omega_{\mathbf{k}}[c_0^{2/3}+(\omega_m^2-\omega_{\mathbf{k}}^2)/\Omega^2]}. \tag{23}$$

It is seen from (22), (23) that when $\omega_m^2-\omega_{\mathbf{k}}^2\gg\Delta_1$, the condition

$$k\frac{d\omega(\mathbf{k})}{dk}\gg\gamma_{\mathbf{k}} \tag{24}$$

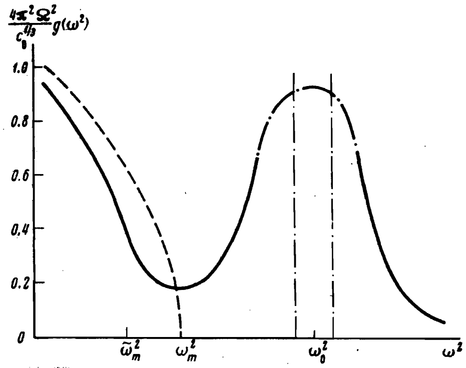

FIG. 1. Density of states in the case of the development of local oscillations near the edge of the unperturbed spectrum for $c/c_0=1/10\pi$. The dashed curves give the density of states of the unperturbed crystal, and the dot-dash curve the region of the concentration broadening of the LO.

is also satisfied, i.e., the lifetime of such excitations is large in comparison with the time of their propagation over a distance of the order of a wavelength.

$$g_3(\omega^2)=c/2\pi c_0^{2/3}\,\Omega^2 \text{ when } \tilde{\omega}_m^2-\omega^2=0.$$

At the same time, the expressions (19) and (20) remain valid and enable us to describe in continuous fashion the transition from a state of plane-wave type to fluctuation states that are due to pairs of impurities.

2. We now proceed to the consideration of such impurity centers which lead to the generation of resonance states close to the edge of the unrenormalized spectrum. The expression for D_0 can be represented in this case in the form

$$D_0=\frac{\varepsilon\omega^2}{4\pi\Omega^2}[c_0^{1/3}+|\varkappa|(\theta(\omega^2-\omega_m^2)+i\theta(\omega_m^2-\omega^2))]. \tag{25}$$

Here, as before, we can assume that the characteristic concentration is $c_0 \ll 1$ and $c \ll c_0$. The shift in the "boundary" of the spectrum turns out to be equal to $\tilde{\omega}_m^2-\omega_m^2=4\pi c c_0^{-1/3}\Omega^2$ and differs only in sign from the case of LO. In the region $\tilde{\omega}_m^2-\omega^2 \gg \Delta_1$, the expression for the density of states, and also the frequencies and damping of the excitations, are obtained from the corresponding formulas (21)–(23), if we reverse the sign of the second term under the radical in (21) and (22). Near the transition frequency, when $\tilde{\varkappa}^2 \ll c_0^{2/3}$, the quantity $\operatorname{Im}R_{2,\mathbf{k}}$ due to pairs of impurities is determined by the expression

$$\operatorname{Im}R_{2,\mathbf{k}}=\frac{2\pi^2c^2r_\omega^3}{|D|a^3}\left[1+\operatorname{sign}(\omega^2-\omega_0^2)\frac{\sin kr_\omega}{kr_\omega}\right],$$
$$r_\omega \approx ac_0^{-1/3}. \tag{26}$$

It is seen from (26) that $\operatorname{Im}R_{2,\mathbf{k}=0}=0$ in the transition region for the LO, and for the resonance states, $\operatorname{Im}R_{2,\mathbf{k}=0}=16\pi^3c^3c_0^{-4/3}\Omega^2$ and plays an important role in Eq. (20). As a result, the resonance states $g(\omega^2)$ in the region $|\omega^2-\tilde{\omega}_m^2| \ll c_0^{-2/3}\Omega^2$ can be described with the help of a standard function that does not depend on the parameters of the problem:

$$g_3(\omega^2)=\frac{c}{2\pi c_0^{2/3}\Omega^2}f\left(\frac{\omega^2-\tilde{\omega}_m^2}{\Delta_1}\right), \quad \Delta_1=\frac{4\pi^2c^2}{c_0^{4/3}}\Omega^2, \tag{27}$$

where $f(x)$ is determined by the solution of the problem

$$f^4-xf^2-(f+2\pi)^2=0 \tag{28}$$

and its plot is shown in Fig. 2. When $x \gg 1$, the function $f(x)\approx\sqrt{x}$, the expression (27) here is identical with the earlier determination of the density of states in the region $\tilde{\omega}_m^2-\omega^2 \gg \Delta_1$. When $-x \gg 1$ we have $f(x)\approx 2\pi/(\sqrt{|x|}-1)$, and $g_3(\omega^2)$ is equal to

$$g_3(\omega^2)=\frac{2\pi c^2}{c_0^{4/3}\Omega^2(\tilde{\varkappa}-2\pi c/c_0^{2/3})}, \quad \tilde{\varkappa}^2=\frac{\omega^2-\tilde{\omega}_m^2}{\Omega^2} \gg \frac{\Delta_1}{\Omega^2}. \tag{29}$$

If $|\omega^2-\omega_m^2| \ll c_0^{2/3}\Omega^2$, then $g_4(\omega^2) \ll g_3(\omega^2)$. As ω^2 increases, the function $g_3(\omega^2)$ falls off, and for $\omega^2 \sim \omega_m^2+c_0^{2/3}\Omega^2$, it is equal to $g_4(\omega^2)$. In the region $\omega^2 \gtrsim \omega_m^2+c_0^{2/3}\Omega^2$, the density of states is determined by the expression (12). It must be noted that whereas for the LO we have near ω_0 a region Δ_0 where the group expansion ceases to converge, for the resonance states the group expansion enables us to calculate the density of states everywhere up to the region in which the fine structure of the spectrum, due to the discreteness of the spatial distribution of the impurity centers begins to appear.

HIGH IMPURITY CONCENTRATIONS

We shall define as "high" such impurity concentrations for which the condition $c_0 \ll c \ll 1$ is satisfied (where, as before, c_0 is defined in (9)). The region where the group expansions (4), (5), and (6), (7) cease to converge broadens in this case to

$$|\omega^2-\omega_m^2|\sim\Delta_2, \quad \Delta_2=(2\pi c)^{2/3}\Omega^2. \tag{30}$$

In the present work, we shall consider the range of frequencies $|\omega^2-\omega_m^2| \gg \Delta_2$. Since $\Delta_2 \gg |\omega_0^2-\omega_m^2| \equiv c_0^{2/3}\Omega^2$, the qualitative results for the LO in the resonance states are identical for $c \gg c_0$.

We first consider the density of states inside the unperturbed spectrum: $\omega_m^2-\omega^2\Delta_2$. Taking into account in this region the corrections to the density of states that are linear and quadratic in the impurity concentration, we obtain

$$g(\omega^2)=\frac{1}{4\pi^2\Omega^2}\left(|\varkappa|\mp\frac{2\pi cc_0^{1/3}}{|\varkappa|^3}-\frac{8\pi^2\beta c^2}{|\varkappa|^5}\right)$$

$$\beta=\int_0^\infty\frac{x^3\cos x\cos 3x+x^2\sin 2x+x\cos^2 x}{x^4+2x^2\cos 2x+1}dx-\frac{1}{4}\approx 0.178.$$

$$\varkappa^2=(\omega^2-\omega_m^2)/\Omega^2. \tag{31}$$

The minus and plus signs correspond to LO and resonance states, respectively. In the vicinity of the "forbidden" region Δ_2, the principal correction to the density of states is negative and is determined by the last component in (31). When $-\varkappa^2 \gg 4\pi\beta cc_0^{-1/3} \gg \Delta_2/\Omega^2$, the principal correction is determined by the second component in (31) and, if we recognize that $\omega_m^2-\omega^2 \gg c_0^{-1/3}\Omega^2$, this correction is identical here with the corresponding correction to the density of states at small impurity concentrations in (21).

In the region of frequencies $\omega_m^2-\omega^2 \gg \Delta_2$ that we have considered, the condition (24) is satisfied, and the states can be decribed approximately by plane waves.

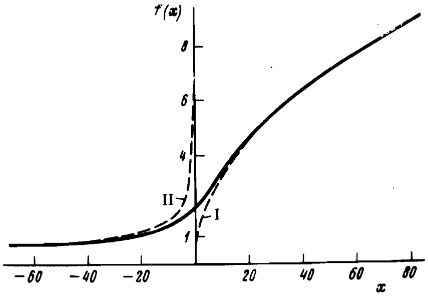

FIG. 2. Universal function $f(x)$ for the density of states in the case of resonance states. The dashed curves give the asymptotes I—$\sqrt{x}$, $x \gg 1$ and II—$2\pi/(|x|^{1/2}-1)$, $-x \gg 1$.

The principal term in the damping is determined by the same formula (23) as for small concentrations. It follows from (24) that at high concentrations, the achieved values of the wave vector is $k > (2\pi c)^{1/3} a^{-1}$, i.e., the wavelength should be less than the mean distance between the impurities. Longer-wavelength excitations are already damped out at distances of the order of the wavelength. At low concentrations, it follows from the conditions $\omega_m^2 - \omega^2 \gg \Delta_1$ and (24) that the region of achievable values of the wave vector is much wider: $k > 4\pi c/ac_0^{2/3}$. The latter circumstance can be understood by assuming the scattering cross section for one impurity to be equal to $a^2 c_0^{-2/3}$. Then the mean free path of the excitation turns out to be $ac_0^{2/3}/4\pi c$, which is much greater than the mean distance between the impurities $\sim ac^{-1/3}$.

We find also the density of states for high concentrations in the range $1 \gg (\omega^2 - \omega_m^2)/\Omega^2 \gg \Delta_2/\Omega^2$, where the pair fluctuations of the impurity atoms are decisive:

$$g(\omega^2) = \pi c^2 \xi^3/\Omega^2 \varkappa^5, \tag{32}$$

and ξ is determined from the condition $\xi = e^{\xi}$, $\xi \approx 0.57$.

At still higher concentrations, when $c \sim 1$, it is necessary to take into account the direct interaction between the impurities, since this interaction becomes more important than the exchange of virtual phonons. Moreover, in this case it is no longer possible to restrict oneself to consideration of only the first terms in the group expansions (4), (5) and (6), (7).

CONCLUSION

The group expansions obtained in the present work and the estimates of their region of convergence enables us not only to obtain the specific results discussed above, but also to estimate the region of applicability of the different approximation methods in the theory of nonideal crystals, for example, the coherent-potential method.[8,9]

In the polarization operator R_k of (5), we keep only the first term in the brackets. As a result, we discard all terms that describe the interaction between the impurities; however, R_k is determined as usual in self-consistent fashion, since this same polarization operator enters in the definition of the denominator D. Such an approximation corresponds just to the coherent-potential method for not too high concentrations of the impurities. If we carry out such a calculation systematically for the case of LO at $c \ll c_0$, then an impurity band arises near the frequency $[\omega_0^2 - \Delta, \omega_0^2 + \Delta]$ with a high density of states. The width of this band, $2\Delta = 4\sqrt{2} c^{1/2} c_0 \Omega^2$, is much less than the distance from ω_0^2 to the edge of the fundamental band $\tilde{\omega}_m^2$, and the density of states in the interval between the bands is equal to zero. Moreover, it turns out that in the interval $[\omega_0^2, \omega_0^2 + 8\pi c \Omega^2/c_0^{1/3}]$ the states have the character of plane waves. All these results do not coincide with those of the present research, the reason being that such an approximation is inapplicable in the given regions, since the discarded terms of the group expansion turn out to be of the same order as those retained. At the same time, for $\tilde{\omega}_m^2 - \omega^2 \gg \Delta_1$, the method of coherent potential correctly describes the density of states and the shift of the band edge.

For $c \gg c_0$, the width of the region of dispersion of the impurity band is of the order of Δ_2; therefore in this case we cannot speak of a dispersion law for the impurity band. A different situation can arise if a new small parameter appears, of the type $\varepsilon\omega^2/4\pi\Omega^2 \ll 1$.

It should also be pointed out that the group expansions obtained here can be applicable also to the spectra of elementary excitations of other types in the presence of nearby localized states, in particular, for impurities in the electronic subsystem. If we replace ω^2 by E and ω_m^2 by E_m, $c_0^{1/3}$ by $a\alpha$ and Ω^2 by $1/2ma^2$, then in the region $\tilde{\omega}_m^2 - \omega^2 \gg \Delta_1$, the expressions obtained here in the linear approximation in the concentration agree with the results of[5]. Inside the fluctuation region, but far from the transition region not considered in[5], the expressions (12), (29), (32) have qualitatively the same form as the corresponding results obtained in[5] with the use of certain qualitative considerations from[4], but differ by a constant factor, equal to $\frac{1}{2}$.

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