

Phonons in an impurity metal

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The applicability of the adiabatic approximation to the calculation of the phonon spectrum of a metal with impurities is analyzed. A successive procedure is presented for determining the adiabatic phonons in such a system, and the necessary nonadiabatic corrections are considered. The latter are determined in the long-wave limit by a large number of interference processes in which electron-phonon and electron-impurity interactions participate. When their diverging contributions cancel one another, these processes result in a weak damping of the adiabatic phonons and in a renormalization of the speed of sound. In the case of optical phonons the contribution of the electron interaction to the damping in "pure" metals ($c < \Omega/\varepsilon_F$, where c is the defect density, Ω is the optical frequency, and ε_F is the Fermi energy) is found to be stronger than in "dirty" metals ($c > \Omega/\varepsilon_F$). Anharmonic processes in impurity metals are considered.

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1. INTRODUCTION

The Born-Oppenheimer adiabatic approximation is the basis of the microscopic analysis of the properties of such multiparticle systems as molecules and solids. Because of the large difference between the masses of the electrons and nuclei ($m/M < 1/2000$) it is possible to distinguish between two types of motion, namely rapid motion of the electrons, which manage to attune themselves to the slow motion of the nuclei, while the latter move in the self-consistent field of the electrons. The parameter for the applicability of the adiabatic approximation is not the ratio m/M itself, but the ratio of the characteristic frequency Θ of the nuclear motion to the energy spacing $\Delta\varepsilon$ between the levels of the electron subsystem.

Usually the parameter $\Theta/\Delta\varepsilon \sim (m/M)^{1/2} \ll 1$. Exceptions are systems with degenerate electronic terms. The most important example of such systems are metals. Since the electronic states fill densely the energy region near the Fermi energy ε_F , the parameter $\Theta/\Delta\varepsilon$ is not small regardless of the value of Θ . Migdal¹ has shown that this is the cause of the strong normalization of the spectrum of the electronic excitations near ε_F . This violation of the adiabatic approximation, however, is local and does not pertain to electrons located far from the Fermi surface. For this reason, in those cases when one deals with the metal properties determined by all the electrons, e.g., with the phonon spectrum, the adiabatic approximation is somehow still valid accurate to the small parameter Θ/ε_F . For the case of a pure metal, the corresponding microscopic analysis was carried out by Brovman and Kagan² (see also their review³ and the article by Maksimov⁴).

In principle, the qualitative considerations advanced above can be directly made applicable also to a metal with impurities. However, whereas the applicability of the adiabatic approximation to relatively short-wave phonons is not subject to doubt, the situation is not so obvious for long-wave phonons.⁵ If the phonon wavelength exceeds the electron mean free path, the diffusion decrease of the electron velocity comes into play. This can give the impression that the distinction between two subsystems in disordered metals, fast (electrons)

and slow (nuclei), is now no longer justified, and the adiabatic approximation no longer holds.⁶

A detailed analysis of this question is particularly important because its results play a fundamental role in the calculation of the kinetic coefficients or the critical superconducting transition temperature in a disordered metal. A number of recent studies⁶⁻⁸ resulted in a negative coefficient of the temperature dependent part of the resistivity because of singularities in the electron-phonon interaction in impurity metals. The results of these studies patently contradict the results of Ref. 9, where it was shown that in the low temperature limit ($T < c\Theta$, where c is the impurity density) the contributions of the different electron-phonon interaction processes to the resistivity practically cancel one another. At $T > c\Theta$, two terms are significant in the phonon part of the resistivity, namely $\propto T^5$ (the Bloch-Grüneisen law) and $\propto cT^2$ (Ref. 10).

The results of Refs. 6–8 are in all probability due to the explicit⁶ or implicit^{7,8} erroneous assumption that the adiabatic approximation is violated in the models considered by the authors of the cited papers. However, Pippard¹¹ has shown phenomenologically long ago that ultrasound attenuates weakly in such systems (the microscopic analysis is contained in a paper by Schmid¹²). As will be shown below, this circumstance is closely connected with the question of applicability of adiabatic approximation. The same can be stated also concerning the cancellation of the various contributions to the resistivity at $T < c\Theta$ (Ref. 9).

In a disordered metal, many electron-phonon interaction processes contribute to the formation of the phonon spectrum. Many of these processes, taken separately, could cause the appearance of instability of the phonons in the long-wave limit. Taken jointly, however they cancel each other, and it is possible as a result to devise a consistent procedure for constructing the phonon spectrum in the adiabatic approximation and find subsequently the small nonadiabatic corrections to the spectrum. (We encounter a similar cancellation also in the analysis of anharmonic interaction of phonons). This cancellation of diverging contributions has a simple physical meaning. The divergences themselves re-

sult from the already mentioned diffusion slowdown of the electrons, which "lag" the nuclei as a result. This produces immediately strong electric fields that cause the electrons to "catch up" with the nuclei. The competition between these two opposing mechanisms determines the small (to the extent that the nonadiabaticity parameter is small) renormalizations and the damping of the long-wave phonons.

2. ADIABATIC PHONONS OF AN IMPURITY METAL

The Hamiltonian of a metal with impurities is of the form

$$H = \frac{1}{2} \sum_i M_i \dot{u}_i^2 + \sum_{im} V_{ii,im}(\mathbf{R}_i - \mathbf{R}_m) + \sum_{ik\mathbf{k}'} V_{ei,i}(\mathbf{k} - \mathbf{k}') \times \exp\{-i(\mathbf{k} - \mathbf{k}') \cdot \mathbf{R}_i\} a_{\mathbf{k}'}^+ a_{\mathbf{k}} + H_{ee}. \quad (1)$$

Here M_i is the mass of the ion located at the lattice site with coordinate \mathbf{R}_i ; $V_{ii,im}(\mathbf{R})$ and $V_{ei,i}(\mathbf{r})$ are respectively the potentials of the ion-ion and electron-ion interactions; $a_{\mathbf{k}}^+$ and $a_{\mathbf{k}}$ are the electron creation and annihilation operators. To find the equilibrium positions $\{\mathbf{R}_i^e\}$ of the ions and the phonon spectrum it is necessary to calculate the total static (with the ions immobile) energy of the system as a function of $\{\mathbf{R}_i\}$. The equilibrium positions of the ions are obtained by equating to zero the first derivatives of this energy with respect to \mathbf{R}_i , and by investigating the small harmonic oscillations of the ions about $\{\mathbf{R}_i^e\}$ we obtain the phonon spectrum.

It is known that the purely ion-ion contribution to the total static energy cannot serve even as the zeroth approximation, since the contribution of the electron subsystem turns out to be of the same order, and must be taken into account from the very beginning. This can be easily done with the aid of a formal procedure proposed by Kozlov (cf. also Refs. 4 and 13). We calculate the contributions of the electron sub-system $E_{ei}(\{\mathbf{R}_i\})$ by perturbation theory with respect to the operator of the electron-ion interaction H_{ei} . We add the energy $E_{ei}(\{\mathbf{R}_i\})$ to the ion-ion part of the Hamiltonian (1) and subtract it, respectively, from the electron-ion interaction operator. The result can be written in the form

$$H = \frac{1}{2} \sum_r M_r \dot{u}_r^2 + \frac{1}{2} \sum_{im} V_{ii,im}(\mathbf{R}_i - \mathbf{R}_m) + E_{ei}(\{\mathbf{R}_i\}) + \hat{d}H_{ei} + H_{ee}. \quad (2)$$

The symbol \hat{d} in front of the electron-ion interaction operator indicates that the latter is purely dynamic. This means that in the calculation of the energy corrections due to the electron-ion interaction it is necessary to retain only the dynamic contributions ($\omega \neq 0$), subtracting the static contributions already taken into account in $E_{ei}(\{\mathbf{R}_i\})$. In the case of a pure metal these contributions are always small to the extent that the non-adiabaticity parameter is small.²

If we confine ourselves to the harmonic approximation, allowance for the contributions of order higher than second in H_{ei} to $E_{ei}(\{\mathbf{R}_i\})$ reduces in the case of a regular metal to a renormalization of the electron spectrum, i. e., to replacement of the plane waves by Bloch waves and to a corresponding distortion of the spherical Fermi surface. Therefore if the true elec-

tron spectrum and the wave functions of the electrons are used directly, there is no need, in the harmonic approximation, to take into account in the electronic part of the energy of the regular crystal the terms of order higher than second in H_{ei} (cf. Refs. 2 and 3). It arises only in the analysis of the anharmonic interaction or dynamic corrections in an impurity metal. For simplicity in the calculation of the total energy of the metal

$$\frac{1}{2} \sum_{im} V_{ii,im}(\mathbf{R}_i - \mathbf{R}_m) + E_{ei}(\{\mathbf{R}_i\}) = \frac{1}{2} \sum_{im} V_{im}^{eff}(\mathbf{R}_i, \mathbf{R}_m), \quad (3)$$

however, we shall describe the electronic subsystem using plane waves and a spherical Fermi surface, as well as leave out everywhere the summation over the reciprocal-lattice vectors. This simplification, as can be easily verified, does not influence qualitatively the singularities of interest to us in the behavior of the long-wave phonons.

We consider now adiabatic phonons [corresponding to the effective interaction (3) in the static approximation] in a metal with impurities. Using the standard procedure of expanding in small ion displacements u_i about their equilibrium positions \mathbf{R}_i^e , which we assume to be known, we obtain after a Fourier transformation

$$\begin{aligned} \omega^2 u_{\mathbf{k}}^\alpha - \sum_{\beta, \mathbf{k}'} D_{\alpha\beta}(\mathbf{k}, \mathbf{k}') u_{\mathbf{k}'}^\beta &= 0; \\ u_{\mathbf{k}}^\alpha &= \frac{1}{N} \sum_i M_i^{1/2} u_i^\alpha \exp(i\mathbf{k} \cdot \mathbf{R}_i), \end{aligned} \quad (4)$$

and the dynamic matrix $D_{\alpha\beta}(\mathbf{k}, \mathbf{k}')$ is connected with the effective ion-ion interaction (3) by the relation (see, e. g., Ref. 14)

$$D_{\alpha\beta}(\mathbf{k}, \mathbf{k}') = \frac{1}{2N^2} \sum_{im} \frac{1}{(M_i M_m)^{1/2}} \frac{\partial^2 V_{im}^{eff}(\mathbf{R}_i, \mathbf{R}_m)}{\partial \mathbf{R}_i^\alpha \partial \mathbf{R}_m^\beta} \exp\{-i\mathbf{k} \cdot \mathbf{R}_i + i\mathbf{k}' \cdot \mathbf{R}_m\}. \quad (5)$$

We assume the defect concentration to be small and use in the calculation of the lattice sums the set of vectors $\{\mathbf{R}_i\}$ corresponding to equilibrium positions of the ions in the regular lattice, neglecting by the same token the dilatation effects. N is the number of unit cells in the lattice and coincides in this case with the number of ions. Generalization to the case of a polyatomic unit cell entails no difficulty.

After determining the eigenvalues of (4), we can in principle obtain the phonon spectrum of an impurity metal. In this case, of course, the quantity \mathbf{k} cannot be interpreted as the phonon quasimomentum, especially in the case of short-wave phonons. On the other hand, for long-wave excitations, a good zeroth approximation can be the phonons of a pure metal, which are weakly scattered by the defects, and the corresponding quasimomentum characterizes sufficiently well the phonons of the impurity metal. We shall assume below that we know the proper phonon frequencies $\omega_s(\mathbf{k})$ and polarization vectors $e_s(\mathbf{k})$ (the normalized eigenvectors $u_{\mathbf{k}}$ of the pure metal, which are connected with the dynamic matrix $D_{\alpha\beta}^{(0)}(\mathbf{k}) \delta_{\mathbf{k}\mathbf{k}'}$).

Introduction of a small number of impurities in a pure metal causes corrections to appear in the dynamic

matrix (5), due to the change in the energy of the ion-ion interaction proper

$$\delta D_{ii,ab}(\mathbf{k}, \mathbf{k}') = \frac{1}{2M^{(0)}} \int \frac{d^2q}{(2\pi)^2} \delta V_{ii}(\mathbf{q}; \mathbf{k}, \mathbf{k}') q^a q^b - \frac{1}{2M^{(0)2}} \int \frac{d^2q}{(2\pi)^2} V_{ii}^{(0)}(\mathbf{q}) [\delta M(\mathbf{q}-\mathbf{k}) \delta_{\mathbf{k}\mathbf{k}'} + \delta M(\mathbf{q}-\mathbf{k}') \delta_{\mathbf{k}\mathbf{k}}] q^a q^b, \quad (6)$$

where

$$\delta V_{ii}(\mathbf{q}; \mathbf{k}, \mathbf{k}') = \frac{1}{N^2} \sum_{im} \delta V_{ii,im}(\mathbf{q}) \exp\{i(\mathbf{q}-\mathbf{k})R_i - i(\mathbf{q}-\mathbf{k}')R_m\}, \\ \delta V_{ii,im}(\mathbf{q}) = \int d^3R \delta V_{ii,im}(\mathbf{R}) e^{-i\mathbf{q}\mathbf{R}}, \quad \delta V_{ii,im}(\mathbf{R}) = V_{ii,im}(\mathbf{R}) - V_{ii}^{(0)}(\mathbf{R}), \\ \delta M(\mathbf{q}) = \frac{1}{N} \sum_i \delta M_i e^{-i\mathbf{q}\mathbf{R}_i}, \quad \delta M_i = M_i - M^{(0)}.$$

On the other hand, in second-order perturbation theory in the electron-ion interaction operator H_{ei} , we obtain for the total electron energy the expression

$$E_{ei}^{(2)}(\{\mathbf{R}_i\}) = \frac{i}{2N^2} \sum_{im} \int \frac{d^2q}{(2\pi)^2} e^{i\mathbf{q}(\mathbf{R}_i - \mathbf{R}_m)} V_{ei,i}(\mathbf{q}) V_{ei,m}(\mathbf{q}) \\ \times \int_{-\infty}^{\infty} dt \sum_{\mathbf{k}\mathbf{k}'} \langle T a_{\mathbf{k}+\mathbf{q}}^+(t) a_{\mathbf{k}}(t) a_{\mathbf{k}'}^+(0) a_{\mathbf{k}'+\mathbf{q}}(0) \rangle. \quad (7)$$

Here $\langle T \dots \rangle$ denotes T -ordered averaging over the ground state of the unperturbed electron subsystem.

The expression for the impurity correction to the electronic part of the dynamic matrix is of the form

$$\delta D_{ei,ab}(\mathbf{k}, \mathbf{k}') = \frac{1}{2M^{(0)}} \int \frac{d^2q}{(2\pi)^2} q^a q^b \delta [V_{ei}(\mathbf{q}; \mathbf{k}-\mathbf{q}) V_{ei}(\mathbf{q}; \mathbf{q}-\mathbf{k}')] \\ \times \sum_{\mathbf{k}_i, \mathbf{k}_i'} \Pi(\mathbf{k}_i, \mathbf{k}_i'; \mathbf{q}) - \frac{1}{2M^{(0)2}} \int \frac{d^2q}{(2\pi)^2} q^a q^b [\delta M(\mathbf{k}-\mathbf{q}) \delta_{\mathbf{k}\mathbf{k}'} \\ + \delta M(\mathbf{k}'-\mathbf{q}) \delta_{\mathbf{k}\mathbf{k}'}] V_{ei}^{(0)}(\mathbf{q}) \sum_{\mathbf{k}_i, \mathbf{k}_i'} \Pi(\mathbf{k}_i, \mathbf{k}_i'; \mathbf{q}), \quad (8)$$

where

$$V_{ei}(\mathbf{q}; \mathbf{k}-\mathbf{q}) = V_{ei}^{(0)}(\mathbf{k}) \delta_{\mathbf{k}\mathbf{k}'} + \delta V_{ei}(\mathbf{q}; \mathbf{k}-\mathbf{q}), \\ \delta V_{ei}(\mathbf{q}; \mathbf{k}-\mathbf{q}) = \frac{1}{N} \sum_i \exp\{-i(\mathbf{k}-\mathbf{q})\mathbf{R}_i\} \delta V_{ei,i}(\mathbf{q}).$$

$\Pi(\mathbf{k}_i, \mathbf{k}_i'; \mathbf{q})$ is the static limit of the mean value of the four-fermion operator in expression (7).

Assuming the impurities to be a small perturbation of the phonon system of the pure metal, we obtain in first order in the impurity density the following formula for the corrections to the phonon frequencies and polarization vectors, averaged over the impurity configuration,

$$\delta \omega_s^2(\mathbf{k}) = \frac{1}{M^{(0)}} [\Delta \mathcal{V}(\mathbf{k}) |(\mathbf{k}, \mathbf{e}_s(\mathbf{k}))|^2 + \overline{\Delta M} \omega_s^2(\mathbf{k})], \\ \delta \mathbf{e}_s(\mathbf{k}) = \frac{(\mathbf{k}, \mathbf{e}_s(\mathbf{k}))}{M^{(0)}} \sum_{j \neq s} \frac{\Delta \mathcal{V}(\mathbf{k})}{\omega_s^2(\mathbf{k}) - \omega_j^2(\mathbf{k})} (\mathbf{e}_j^*(\mathbf{k}), \mathbf{k}) \mathbf{e}_j(\mathbf{k}). \quad (9)$$

Here

$$\Delta \mathcal{V}(\mathbf{k}) = \overline{\delta V_{ei}}(\mathbf{k}) + \frac{\overline{\delta V_{ei}}(\mathbf{k}) V_{ei}^{(0)}(\mathbf{k}) \pi(\mathbf{k})}{1 - V_{ee}(\mathbf{k}) \pi(\mathbf{k})}, \\ \overline{\delta V_{ei}}(\mathbf{k}) = \sum_j c_j \delta V_{ei,j}(\mathbf{k}), \quad \overline{\delta V_{ee}}(\mathbf{k}) = \sum_j c_j \delta V_{ee,j}(\mathbf{k}), \\ \overline{\Delta M} = \sum_j c_j \delta M_j,$$

where c_j is the density of the impurities of species j ,

$\delta V_{ii,j}$ and $\delta V_{ei,j}$ are the changes of the potentials of the ion-ion and electron-ion interaction following introduction of the impurity of species j , and $\pi(\mathbf{k})$ is the static limit of the polarization operator that is irreducible with respect to the Coulomb interaction. In the long-wave limit ($\mathbf{k} \rightarrow 0$), when we can use the Coulomb asymptotic form of the potentials

$$\delta V_{ii,j} = 4\pi e^2 \delta Z_j Z^{(0)}/k^2, \quad \delta V_{ei,j} = -4\pi \delta Z_j e^2/k^2 + \delta b_j,$$

where $\delta Z_j = Z_j - Z^{(0)}$, and Z_j and $Z^{(0)}$ are respectively the valences of the j th impurity and of the matrix ion, we obtain

$$\delta \omega_s^2(\mathbf{k}) = \frac{1}{M^{(0)}} \left[\left(\frac{4\pi e^2}{\kappa^2} Z^{(0)} \overline{\Delta Z} + \overline{\Delta Z} b^{(0)} + \overline{\Delta b} Z^{(0)} \right) |(\mathbf{k}, \mathbf{e}_s(\mathbf{k}))|^2 - \overline{\Delta M} \omega_s^2(\mathbf{k}) \right], \\ \delta \mathbf{e}_s(\mathbf{k}) = \left(\frac{4\pi e^2}{\kappa^2} Z^{(0)} \overline{\Delta Z} + \overline{\Delta Z} b^{(0)} + Z^{(0)} \overline{\Delta b} \right) \sum_{j \neq s} \frac{(\mathbf{k}, \mathbf{e}_s(\mathbf{k})) (\mathbf{k}, \mathbf{e}_j^*(\mathbf{k}))}{\omega_s^2(\mathbf{k}) - \omega_j^2(\mathbf{k})} \mathbf{e}_j(\mathbf{k}), \\ \overline{\Delta Z} = \sum_j c_j \delta Z_j, \quad \overline{\Delta b} = \sum_j c_j \delta b_j. \quad (10)$$

Here b is the limit of the pseudopotential of the electron-ion interaction as $\mathbf{k} \rightarrow 0$ (Ref. 3). The result (10) is physically quite clear: In the long-wave limit the phonon spectrum of an impurity metal takes the same form as the phonon spectrum of a certain fictitious regular crystal (customarily called virtual in the literature, see, e.g., Ref. 15) with a potential that is somewhat modified compared with the pure metal ($Z^{(0)} \rightarrow Z^{(0)} + \overline{\Delta Z}$, $b^{(0)} \rightarrow b^{(0)} + \overline{\Delta b}$) and with an ion mass $M^{(0)} \rightarrow M^{(0)} + \overline{\Delta M}$. It is interesting that we can always choose the impurity compositions such as to make $\overline{\Delta Z} = \overline{\Delta b} = \overline{\Delta M} = 0$. In this case the phonon spectrum of the impurity metal does not differ at all from the spectrum of the pure metal, since the corrections (10) vanish. In order for the polarization vectors to remain unchanged, it suffices to have $\overline{\Delta Z} = \overline{\Delta b} = 0$. [The purely longitudinal modes of the oscillations do not change at all in the approximation (10) when impurities are introduced, even if $\Delta \bar{V} \neq 0$.] We note that in this case the effects connected with the change of the volume upon introduction of impurities with different ΔZ_j and δb_j have likewise apparently a tendency to cancel each other.

Expression (10) corresponds to the Born approximation for the scattering of phonons by impurities. Going outside the framework of this approximation, we can obtain the phonon-spectrum contributions that are linear in the density and are proportional to $\delta \bar{Z}^2$, $\delta \bar{M}^2$, etc. These contributions are usually significant if the vibrational spectrum of the crystal contains the so-called local or quasilocal modes. Their investigation is the subject of an extensive literature, and they will not be discussed here. We imply by the same token that we are dealing throughout with frequencies far from those of these modes.

Among the corrections proportional to $\overline{\delta Z}^2$, we discuss here only the contributions to the energy of the electron subsystems, shown in Fig. 1a. The dashed line shows here the procedure of averaging over the configurations of the impurities (see, e.g., Ref. 16). Each wavy line (which is transformed after quantization of the field of the vibrations into a phonon propagator) corresponds in the site representation to differentia-



FIG. 1.

tion $\partial/\partial R_i^\alpha$. The shaded loop denotes a polarization operator reducible with respect to Coulomb interaction, and the electron-phonon vertex with the heavy dot corresponds to the quantity $(k^\alpha - k'^\alpha)\delta V_{ei,j}(k-k')$.

It is easy to verify that if the corrections shown in Fig. 1a were taken into account in the dynamic matrix (5), this would lead to the appearance of an unphysical gap in the spectrum of the acoustic phonons. To eliminate this gap and obtain the correct limiting behavior of the acoustic phonons as $k \rightarrow 0$ it is necessary to take into account also the diagram shown in Fig. 1b. Here the vertex with the heavy dot without wavy lines corresponds to scattering of an electron by a static impurity $\delta V_{ei,j}(k-k')$ and the vertex with two wavy lines corresponds to scattering of an electron by an impurity with emission (absorption) of two phonons: $(k^\alpha - k'^\alpha)(k^\beta - k'^\beta)\delta V_{ei,j}(k-k')$.

Comparing diagrams a and b of Fig. 1, we can verify directly that the corresponding expressions are equal in absolute value and opposite in sign. Thus, the static parts of the diagrams a and b of Fig. 1 cancel each other and do not take part in the formation of the adiabatic phonons. In the dynamic regime, however, there is no such cancellation and as a result these processes play the principal role in the formation of the low-temperature limit of the phonon part of the resistivity of impurity metals^{9,10} (the same pertains also to other kinetic coefficients), and it will be shown below that they must be taken into account in the analysis of the nonadiabatic corrections to the phonon spectrum and in the check on the validity of the adiabatic approximation in impurity metals. We note that cancellation of these contributions corresponds in the static approximation to the requirement that the total force acting on any ion at equilibrium be zero.

3. NONADIABATIC CORRECTIONS TO THE PHONON SPECTRUM OF AN IMPURITY METAL

The nonadiabatic (dynamic) corrections to the spectrum of adiabatic phonons in a metal are due to the lag of the electrons in the vibrational motion of the ions, as a result of which transitions between different states of the electron subsystem of the metal become possible. In quasiparticle language this corresponds to the scattering of electrons by phonons. In the Hamiltonian (2) this process is described by the "dynamic" electron-ion interaction operator. Since we have already taken into account, in the calculation of the spectrum of the adiabatic phonons, the static energy (3) of the electron subsystem, we must now, when calculating the dynamic corrections, use this effective Hamiltonian (2). This means that, gathering together the contributions made to the polarization operator $\Pi(q, \omega)$ at finite frequencies ($\omega \neq 0$), we must then subtract from it the already accounted for static part $\Pi(q, 0)$, i. e., find the difference

$$\Delta\Pi(q, \omega) = \Pi(q, \omega) - \Pi(q, 0). \quad (11)$$

Since the problem of nonadiabatic corrections to the phonon spectrum in regular metals has already been analyzed in detail earlier,²⁻⁵ we shall dwell here only on the impurity contributions. It is necessary first of all to consider the corrections corresponding to diagrams a and b of Fig. 1. Diagram b of Fig. 1, by virtue of its structure, is independent of the frequency ω , so that it suffices to consider diagram a of Fig. 1. Then, confining ourselves for simplicity to the random-phase approximation, we obtain with the aid of (11)

$$\delta\omega_s(k) = -\frac{i}{2} \frac{m}{M^{(0)}} \frac{1}{\tau_{ir}}, \quad (12)$$

where

$$\frac{1}{\tau_{ir}} = N(0) \int \frac{d\Omega_p d\Omega_{p'}}{(4\pi)^2} \overline{\delta V^2(\mathbf{p}-\mathbf{p}')} (1 - \cos \angle(\mathbf{p}\mathbf{p}') [1 - V_{ee}(\mathbf{p}-\mathbf{p}') \pi(\mathbf{p}-\mathbf{p}')])^{-2}.$$

The integration with respect to the angles of the vectors \mathbf{p} and \mathbf{p}' is carried out over the Fermi surface, m is the electron mass, and $N(0)$ is the density of the electronic states on the Fermi surface. The correction (12) is small to the extent that the ratio of the electron and ion masses is small ($m/M^{(0)} = \nu^2 \ll 1$), but it does not vanish as $k \rightarrow 0$ when the frequency of the acoustic phonons vanishes, $\omega_s(k) \rightarrow 0$. In the long-wave limit

$$\tau kv \ll 1 \quad (13)$$

(v is the Fermi velocity) this correction can therefore become equal to or even exceed, in absolute value, the frequency of the corresponding adiabatic acoustic phonon.

Thus, if we were to confine ourselves to the approximation (12), we would obtain in an impurity metal strongly damped phonons, i. e., unstable long-wave phonons. Inelastic scattering of electrons by defects (Fig. 1a), which gives rise to the correction (12) to the spectrum of the adiabatic phonons, leads also to the appearance of a term proportional to cT^2 in the temperature-dependent part of the resistivity of the pure metal.¹⁰ From the foregoing arguments it follows that the temperature region of applicability of this law is bounded from below, since at $T \rightarrow 0$ the main contribution is made by processes in which long-wave phonons participate ($k \sim T/s$, s is speed of sound), and these processes are themselves poorly defined in this case.

Indeed, as shown in Refs. 9 and 17, in the long-wave limit, ($\lambda > \tau v$) an important role is assumed by various interference processes with participation of the following elementary interactions: scattering of electrons by "pure" phonons and by vibrating and static impurities. All these processes lead jointly to cancellation of the term cT^2 in the resistivity of the impurity metal.⁹ We shall show below that these processes ensure a small damping of the long-wave acoustic phonons and, by the same token, the applicability of the adiabatic approximation in an impurity metal.

The interaction of electrons with long-wave phonons is quite complicated: During the interaction with the phonon wave the electron manages to be scattered many

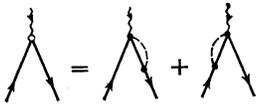


FIG. 2.

times from static fields. As a result, the amplitude of the interaction of the electron with a pure phonon is diffusely amplified, and besides the usual "bare" vertex of the electron-ion interaction there appears a vertex "clothed" by a ladder of impurity dashed lines¹⁷ (see also Ref. 12). In the bare vertex we have already taken into account some impurity corrections, using Z in place of $Z^{(0)}$, i. e., the pure phonons were taken to mean the phonons of the virtual crystal. When account is taken of the renormalization of the polarization operator we obtain for the effective vertex of the electron-phonon interaction

$$-ik \frac{pv}{3} \frac{\xi}{1-\xi+i\tau\omega\xi} \theta\left(\frac{1}{2}|\omega|-\epsilon\right), \quad (14)$$

where

$$\xi = t + il, \quad t = [\text{arctg}(\tau kv + \tau\omega) + \text{arctg}(\tau kv - \tau\omega)] / 2\tau kv, \\ l = \frac{1}{4\tau kv} \ln \left[\frac{1 + (\tau kv + \tau\omega)^2}{1 + (\tau kv - \tau\omega)^2} \right],$$

$\theta(x)$ is the Heaviside function, and $1/\tau$ is determined by the same integral as $1/\tau_{tr}$ but without the factor $1 - \cos \star(\mathbf{pp}')$ in the integrand. To simplify the deviation we assumed a contact electron-impurity interaction potential, and $1/\tau = 1/\tau_{tr}$.

Thus, at $\tau kv, \tau\omega \ll 1$ and $|\epsilon| < |\omega|/2$ the pure electron-phonon vertex is anomalously enhanced, and this by itself could lead to the appearance of instability of the phonon spectrum in the long-wave limit. However, besides the processes (14)–(16) considered in Ref. 17, it is necessary also to take into account the impurity vertex (Fig. 2)^{9,12}

$$-\frac{p}{\tau} \theta\left(\frac{1}{2}|\omega|-\epsilon\right) - \frac{k}{\tau}, \quad (15)$$

which also undergoes a diffuse enhancement of the type (14) and leads to an effective interaction

$$ikpv / (\tau kv)^2. \quad (16)$$

In the derivation of (16) we took into account only the first term in (15). In addition, we have retained in (14) and (16) only the leading terms in the small parameter k^2/κ^2 ($1/\kappa$ is the Debye screening length).

Figure 3 shows various contributions made to the electron energy with participation of the processes (14)

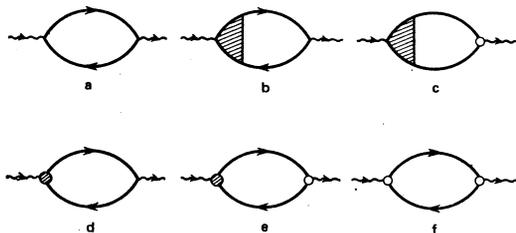


FIG. 3.

and (16). The shaded triangular vertex corresponds to the interaction (14), while the vertex with the shaded circle corresponds to the interaction (16). Diagrams a–e of Fig. 3 correspond jointly to the nonadiabatic correction to the phonon spectrum of the impurity metal

$$\delta\omega_s(\mathbf{k}) = -\frac{i}{2} |\langle \mathbf{e}_s(\mathbf{k}), \mathbf{k} \rangle|^2 \frac{v^2 m}{3M} \tau \left\{ \xi - \frac{6}{(\tau kv)^2} [1 - (1 - i\tau\omega_s(\mathbf{k}))\xi] \right. \\ \left. + \frac{9}{(\tau kv)^2} [1 - (1 - i\tau\omega_s(\mathbf{k}))\xi]^2 \right\} [1 - (1 - i\tau\omega_s(\mathbf{k}))\xi]^{-1}, \quad (17)$$

which causes no instabilities whatever in the entire range of values of the parameters τkv and $\tau\omega$, and consequently cannot cancel out the singular contribution (12). All that we have for its cancellation is the single diagram f of Fig. 3, in which there is not a single transverse dashed line. (Strictly speaking, to obtain the correct results in the case of a general-form electron-impurity potential it is necessary in this case to renormalize diagram f of Fig. 3 by ladders of impurity dashed lines. It can be verified⁹ that in this case it will be necessary to retain, in the limit $\tau kv \gg 1$, only the contribution of the p -amplitude of the electron-impurity interaction, which will lead in a number of places to replacement of $1/\tau$ by $1/\tau_{tr}$.) The expression corresponding to diagram of Fig. 3 yields, jointly with (12),

$$\delta\omega_s(\mathbf{k}) = -\frac{i}{2} \frac{m}{M} \frac{1}{\tau} (1-\xi). \quad (18)$$

In the upshot we obtain from (17) and (18) the nonadiabatic corrections to the phonon spectrum, in the form

$$\delta\omega_s(\mathbf{k}) = -\frac{i}{2\tau} \frac{m}{M^{(0)}} \left[f_0(k, \omega_s(\mathbf{k})) + \frac{(\tau kv)^2}{3} f_1(k, \omega_s(\mathbf{k})) \cos^2 \star(\mathbf{e}_s(\mathbf{k}), \mathbf{k}) \right], \quad (19)$$

from which follows, in the limits $\tau kv \ll 1$ and $\tau kv \gg 1$, Pippard's phenomenological formula¹¹ for the damping of longitudinal ultrasound oscillations. At $\tau kv \ll 1$, the decisive term in (19) is the first. In this case

$$\delta\omega_s(\mathbf{k}) = -\frac{i}{2\pi} \frac{m}{M^{(0)}} \left[\frac{(\tau kv)^2}{3} - i\tau\omega_s(\mathbf{k}) \right], \quad (20)$$

and the nonadiabatic processes lead not only to damping of the phonons, but also to a renormalization of the speed of sound $\delta s = -ms^{(0)}/2M$, which depends neither on the phonon wavelength nor on the defect density. These dependences manifest themselves in the fact that the wavelength region in which this renormalization takes place is bounded by the condition $\lambda > \tau v$. Such a specific concentration and frequency dependence can help separate the quantity δs when an attempt is made to observe it experimentally against the background of the renormalizations connected with other mechanisms. The latter vary as a rule linearly with the defect density, and are practically independent of frequency. The value of $\delta s/s$ for most metals is of the order of 10^{-4} – 10^{-5} . If it is recognized, however, that the electron mass in δs should in fact be taken to be the effective mass, suitably averaged over the Fermi surface, one can hope to find among the light elements metals for which the ratio $\delta s/s$ is noticeably larger; thus, for beryllium we have apparently $\delta s/s \sim 10^{-2}$.

We note also that Eqs. (18) and (20) contain no dependence on the phonon polarization vector, and con-

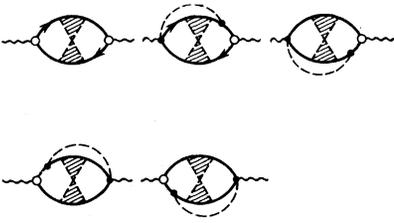


FIG. 4.

sequently take the same form for longitudinal as well as transverse modes. At $\lambda < \tau v^2/3s$ we can put everywhere $\omega_s(\mathbf{k})=0$, so that the angular dependence in (19) can be due only to the second term, which is proportional to $\cos^2 \star(\mathbf{e}(\mathbf{k}), \mathbf{k})$, and vanishes for purely transverse modes. In a real crystal, separation of the purely longitudinal and transverse modes at an arbitrary direction of the vector \mathbf{k} is impossible and the angle between the vectors $\mathbf{e}(\mathbf{k})$ and \mathbf{k} changes when \mathbf{k} is rotated. The second term in (19) becomes of the same order as the first only if $\lambda \sim \tau v$, and exceeds it at $\lambda < \tau v$. In this region, the senior term of the expansion (19) in $\lambda/\tau v$ corresponds to phonon damping in a pure metal.

Besides the diagrams considered above with impurity renormalizations in the "diffusion" channel, there is also a class of diagrams where a singular behavior appears as a result of impurity renormalizations in the "Cooper" channel. These singularities are essential, e.g., in the frequency dependence of the residual resistivity of metals,¹⁵ and become enhanced in quasi-two-dimensional and quasi-one-dimensional systems. Figure 4 shows several diagrams of this type, whose individual contributions to the dynamic matrix are proportional in the three dimensional case to $\omega^{3/2}$. In the sum however, they cancel each other in such a way that the pole responsible for the appearance of the singularity is eliminated from the integrand. The same can be stated also concerning other diagrams not presented here. Thus, processes with singularities in the "Cooper" channel must be completely excluded from consideration when the phonon spectrum is calculated. We emphasize that this statement is valid for systems of any dimensionality. This makes it possible to extend the results of the present paper to two-dimensional and one-dimensional systems, in which an essential role is played by localization effects (see the discussion at the end of the article).

It remains for us to consider the contribution due to the second term in the vertex (15). Leaving out the description of the rather cumbersome calculations, we present only the final result. Although this term is similar in structure to the pure electron-phonon vertex, it makes no contribution to any "static" diagrams. Participating, however, in the "dynamic" diagrams, this vertex can undergo diffuse enhancement of the type (14) and lead in turn to an instability of the long-wave phonons. Since this vertex already contains the factor $1/\tau$, cancellation of its contribution calls for the use of diagrams of type (15), but with one more impurity dashed line added. As a result we obtain phonon-spectrum corrections similar to (17), but containing an ad-

ditional smallness in the defect density c . In this case the contribution due to the vertex proportional to $k/\tau(\tau c_F)$ is not cancelled out. To cancel it we must already resort to processes of third order in c . Thus, we are dealing with an hierarchy of cancellations, in which to cancel out terms of order n in c it is necessary to invoke part of the processes of order $n+1$, the remaining processes of order $n+1$ are cancelled by part of the processes of order $n+2$, etc.

Since operations with impurity diagrams of high order are extremely complicated, owing to the need for taking into account corrections that eliminate the multiple occupation of one and the same site (see, e.g., the review by Elliott *et al.*¹⁵ and the references therein), it is possible to track this cancellation in each order in c . This cancellation follows of necessity, however, from the lack of divergences in the final result, which in turn stems from the general properties of closed diagrams averaged over impurity configurations. We can use here the analysis used in Ref. 9 to prove the cancellation of phonon contributions to the resistivity of an impurity metal. It suffices only to replace the function $\bar{R}(\mathbf{k}, \mathbf{k}')$, in Ref. 9 by the expression for the total energy of the electron-ion system. By repeating the reasoning of Ref. 9 we can verify that the dynamic corrections are proportional to $\tau\omega^2$ or to $\omega(\tau kv)^2$.

4. ANHARMONIC INTERACTION OF PHONONS IN AN IMPURITY METAL

Just as in the harmonic case, for a correct description of the anharmonic interaction in a metal it is necessary to take into account the contribution due to the energy of the electronic subsystem. In this case all the "static" diagrams must be taken into account even in the zeroth approximation when formulating the adiabatic part of the anharmonic interaction, after which the "dynamic" diagrams should lead to small nonadiabatic corrections. In the analysis of the anharmonic interaction in impurity metals we encounter the same problems with the long-wave phonons as in the harmonic case. Here we consider this problem in general form and indicate an algorithm for eliminating the "dangerous" diagrams.

The static contribution of n th order in the electron-ion interaction to the total energy of the impurity metal

$$E_{st}^{(n)} = \frac{1}{N^n (n-1)!} \sum_{\{\mathbf{R}_i\}} \int \dots \int \frac{d^3 q_1}{(2\pi)^3} \dots \frac{d^3 q_n}{(2\pi)^3} \int_{-\infty}^{\infty} \dots \int_{-\infty}^{\infty} dt_1 \dots \dots dt_{(n-1)} V_{et,1}(\mathbf{q}_1) \dots V_{et,n}(\mathbf{q}_n) \quad (21)$$

$$\times \sum_{\{\mathbf{k}_i\}} \langle T a_{\mathbf{k}_1+\mathbf{q}_1}^\dagger(t_1) a_{\mathbf{k}_1}(t_1) \dots a_{\mathbf{k}_n+\mathbf{q}_n}^\dagger(0) a_{\mathbf{k}_n}(0) \rangle \exp \left\{ -i \sum_{i=1}^n \mathbf{q}_i \mathbf{R}_{i,t_i} \right\} \delta \left(\sum_{i=1}^n \mathbf{q}_i \right)$$

can be represented in the form of a closed diagram with n electron-ion interaction free vertices, with the factor $1/(n-1)!$ canceled out. We separate the impurity parts $\delta V_{et,i}^i(\mathbf{q}_i)$ of a certain number of vertices and take the j th variation of the energy (21) with respect to the displacements of the ions relative to the equilibrium position, after which we average over the impurity configurations. Pure equilibrium vertices renormalize the electron spectrum, transforming plane waves into

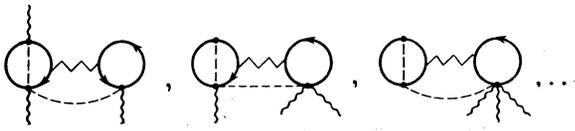


FIG. 5.

Bloch waves, while independently averaged equilibrium impurity vertices, renormalized the energy of the ground state of the system (see, e.g., Ref. 16). For this reason we need retain in the skeleton diagrams only the vertices from which at least one line emerges—an impurity dashed line or a phonon line.

Any diagram containing a vertex with one wavy and one dashed line, corresponding to electron scattering by vibrating impurities, leads to the appearance of a gap in the spectrum of the acoustic phonons as $k \rightarrow 0$, since, in contrast to a pure vertex, it does not contain the small factor k . In fact, however, there are always several diagrams that add up to zero, thereby preventing the appearance of a gap. The prescription for finding such canceling diagrams is clear from the example considered in Sec. 3 (Fig. 1): It is necessary to consider all the diagrams that differ only in permutation of the phonon tails. An example for third-order anharmonicity is shown in Fig. 5, where the sawtooth denotes Coulomb interaction.

We shall show that in the general case the sum of such “dangerous” diagrams vanishes in the statistical limit. Consider a diagram containing j pure and m impurity vertices. Such a diagram occurs when anharmonicity of order not lower than j is considered. In the case of anharmonicity of order j , all j phonon lines are connected to pure vertices: one line only to each vertex. To obtain the corresponding contribution to the anharmonicity of order $j+1$, it is necessary to connect one more phonon line in turn to all the vertices, both pure and impurity, of this diagram. It is easy to verify that the aggregate of diagrams obtained in this case adds up to zero, inasmuch as in the coordinate representation this procedure corresponds to taking the derivative in (21) with respect to all $\partial/\partial R_i$ in succession, which leads to the appearance of the factor $\sum_{i=1}^{j+m} q_i = 0$.

Thus, when calculating the energy (21) we must retain only the diagrams in which the impurity vertices, connected by dashed lines, must of necessity be in equilibrium, while one and only one phonon line enters in a pure vertex. The pure vertices must be taken to mean in the virtual-crystal approximation. As a result, the only diagrams preserved are those whose contribution to the anharmonicity of order j is proportional to $\Pi_{i=1}^j k_i$, where k_i are the external phonon momenta, thereby ensuring “correct” behavior of the acoustic phonons in the long-wave limit.

The situation with the nonadiabatic corrections to the anharmonic processes is perfectly analogous to the harmonic case. Here, too, we encounter a large number of diagrams that diverge in the long-wave limit and in final analysis are mutually cancelled out. Since the concrete calculation of the nonadiabatic corrections to

the anharmonic processes is in itself of no interest, it suffices for us to verify that they are always small and cannot cause any unpleasantness whatever. Even by virtue of their definition, which is analogous to (11), they contain the small factor ω/ε_F , and the absence of any divergences from the final result follows from the analysis given at the end of the preceding section (see also Ref. 9).

5. OPTICAL PHONONS

We consider now impurity renormalizations of optical phonons, assuming that, despite the introduction of impurities, the polyatomic structure of the unit cell of the crystal lattice on the whole is preserved. The foregoing analysis can be directly transferred also to this case—it suffices to take into account in Eqs. (5)–(8) the fact that the unit cell contains many atoms and that additional branches of the vibrational spectrum appear. On the whole this does not change the system of statements made above, and the differences are due only to the fact that the frequency of the optical phonons tends as $k \rightarrow 0$ not to zero but to a finite optical frequency Ω close in value to the Debye frequency Θ .

In the case of optical phonons, just as in the case of acoustic phonons, diagrams a and b of Fig. 1 cancel each other thereby excluding the corresponding renormalization of the optical frequency Ω . Real renormalizations can be obtained in the virtual-crystal approximation or in more complicated approximations (see, e.g., Ref. 15).

Of greatest interest in this case are the nonadiabatic renormalizations of the spectrum. They are determined by the same equations (17)–(20) as the renormalizations of the spectrum of the acoustic phonons. However, since the optical frequency is finite, the picture is different. Two cases can be singled out.

We consider first the case of relatively pure metals, in which the defect density satisfies the condition $c < \Omega/\varepsilon_F$ ($\tau\Omega > 1$). In the long-wave limit $\tau kv < \tau\Omega$, i.e., $\lambda > 2a\varepsilon_F/\Omega$, where a is the lattice constant, we have then

$$|\xi| \approx \left| \frac{1}{1+(\tau\Omega)^2} + \frac{i(\tau kv)^{2/3}}{1+(\tau\Omega)^2} \right| \ll 1, \quad (22)$$

and the renormalizations (17) are small, in which case the decisive renormalization is (12). It corresponds to finite damping of the optical phonons $\gamma_{im}^{(1)} = m/\tau M$. The principal role in the damping of shorter-wavelength phonons ($\lambda < 2a\varepsilon_F/\Omega$) is played by processes of pure electronic interaction

$$\gamma_{ph} = \frac{\pi |e_i(\mathbf{k})\mathbf{k}|^2 m v}{6 k M}$$

[the limit of Eq. (17) at $\tau kv > 1$].

We turn now to the case of dirty metals. Since the ratio Ω/ε_F is small, satisfaction of the condition $c > \Omega/\varepsilon_F$ ($\tau\Omega < 1$) can still be reached at relatively low defect densities. An important role is then assumed in the wavelength region $\lambda > \tau v$, besides the scattering of the electrons by the vibrating impurities (12), also

by the entire aggregate of the processes considered in Sec. 3. The phonon renormalizations are obtained from (20), i. e., the damping of the optical phonons in this region takes the form

$$\gamma_{im}^{(2)} = \frac{m}{M\tau} \frac{(\tau kv)^2}{3},$$

and the renormalization of the optical frequency is $\delta\Omega = \Omega m/M$. This form is preserved down to wavelengths $\lambda \sim \tau v$. For shorter wavelength phonons the damping is determined by the pure electron-phonon interaction γ_{ph} , and the renormalization of the optical frequency vanishes. A substantial contribution to the damping of the optical phonons is made also by the anharmonic interaction. It is characterized, however, by an entirely different concentration behavior, and it is this which raises hopes of separating these processes in experiment.

We emphasized that in dirty metals the contribution of the electron-phonon interaction to the damping of long-wave phonons is smaller than in pure ones ($\gamma_{im}^{(1)} > \gamma_{im}^{(2)}$)! This result, paradoxical at first glance, is in fact easy to understand physically. The cancellation of the different contributions to the renormalization of the phonons takes place when they are scattered not by individual defects independently, but coherently by a large number of impurities. In this case we are dealing in fact with motion of a wave of oscillations in a certain "averaged" crystal. For the scattering to be coherent it is necessary to satisfy simultaneously two conditions: The wavelength and the period of the phonon oscillations must be larger than the mean free path and the lifetime of the electron, respectively. In the case of acoustic phonons, whose oscillation period is connected with the wavelength, both conditions are satisfied simultaneously at $\lambda > \tau v$. For optical phonons there is no such connection, and simultaneous satisfaction of the two coherence conditions can be reached only in sufficiently dirty metals ($c > \Omega/\varepsilon_F$), in which the lifetime of the electron becomes less than the period of the optical oscillations.

6. CONCLUDING REMARKS

The analysis performed in this study enabled us to verify that the adiabatic approximation is applicable with very good accuracy to an impurity metal. It is ensured by a subtle balance between individually diverging contributions of a large number of different elementary processes. In the adiabatic approximation all these give a strictly zero contribution, but they play a fundamental role in different nonadiabatic phenomena, some of which were considered above. In fact, this is a consequence of a simple physical requirement: The only low-frequency excitations permissible in an electron-ion system are those which do not violate strongly the local electroneutrality. What actually take place are only small (since the nonadiabaticity parameter is small) violations of the electroneutrality, which cause

a renormalization of the speed of sound and damping of the long-wave phonons.

It must be emphasized that the main statements of the present paper are qualitatively valid in all orders in the defect density, i. e., the region of applicability of the presented analysis coincides with the region of applicability of perturbation theory in the impurity density. Its parameter is the quantity $1/\tau\varepsilon_F$, and consequently, a qualitatively new situation can arise only if $\tau\varepsilon_F \sim 1$. It is known that in the latter case the ground state is altered by the Anderson localization and a fundamentally different approach is required, not connected with averaging over the impurity configurations.

In the investigation of the resistivity of two-dimensional and one-dimensional metals, the approach to the metal-insulator transition from the metal side can be realized by using the procedure proposed in Ref. 18. However, as noted in Sec. 3 of the present paper, the diagrams that lead to the appearance of singularities in the resistivity¹⁸ do not participate in the formation of the phonon spectrum. This seems to indicate that the adiabatic approximation remains most readily valid also in the case of localization, although the final answer to this question calls for further analysis.

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