Two-Phonon Deformation Potential Coupling: Free-Carrier Absorption in InSb

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We calculate the contribution of the free-carrier absorption due to scattering of electrons by two nonpolar optical phonons. An effective Hamiltonian is formulated for the interaction between the electrons and two phonons. Using the previously calculated values of the two-phonon effective deformation potential, we find that significant contributions to the absorption coefficient occur because of two-phonon processes. The absorption coefficient due to two nonpolar optical phonons varies with wavelength λ of the incident radiation as $\lambda^{1.5}$. The resultant calculated absorption coefficient varies as $\lambda^{2.2}$ while the experimental value varies as λ^2. Thus the agreement between theory and experiment is improved if two-phonon processes are taken into account. The variation of the absorption coefficient with temperature is also calculated.

I. INTRODUCTION

Studies of free-carrier optical absorption in semiconductors have proved to be useful to gain understanding of the carrier interactions. Free-carrier optical absorption can only occur through scatterings by impurities or lattice vibrations. Lattice scatterings make both the conservation of energy and momentum possible. Detailed measurements have been performed on many semiconductors. Theoretical calculations taking into account various forms of scattering have also been provided. For some well-characterized semiconductors, such as InSb, there have been considerable efforts to achieve accurate quantitative calculations of the free-carrier absorption near $\lambda = 9$ μm. These calculations[1-3] considered intraband electron scattering by optical phonons. The results do not agree with experiment either in the wavelength λ dependence or in the magnitude of the absorption coefficient α. Experiments show that α is proportional to λ^2, whereas theoretically $\alpha = c\lambda^{2.4}$. To improve agreement with experiment, Haga and Kimura[1] included a contribution to α due to scattering by acoustic phonons. However, they have to assume a very large value for the acoustic-phonon deformation potential $D = 30$ eV, much larger than the accepted value of $D = 7$ eV. Demidenko[3] included the effects of band nonparabolicity but did not find any significant contribution in the frequency range (8–15 μm) of interest. Jensen[2] included both nonparabolicity and contributions from the intermediate states in other bands. He found an increase in the absorption coefficient α. Nevertheless, α was still 30% smaller than the experimental value and further, the λ dependence of α was poorer, $\alpha \propto \lambda^3$. These calculations quoted above have exhausted the usual carrier-scattering mechanisms, and the outstanding disagreement that exists between theory and experiment is still puzzling.

Recently, Ngai and Johnson,[4] henceforth referred to as I, have shown that the effective two-optical-phonon deformation potential in InSb is very large. In I, it was demonstrated by a resonant carrier-phonon coupling experiment that two nonpolar-optical- (NPO or TO) phonon scatterings cause splittings in the magneto-optical spectrum of comparable size to that due to polar-optical (LO) phonons. It is natural, in view of this new information, to consider the contribution of 2NPO-phonon scattering to free-carrier optical absorption in InSb. In this work we show that good agreement with experiment is obtained if contribution to α from 2NPO- and LO-phonon scatterings are both included.

In Sec. II we derive the effective carrier–2NPO-phonon deformation-potential-interaction Hamiltonian. In Sec. III we calculate the frequency-dependent conductivity due to two-phonon processes by extending the Green's-function theory of Gurevich, Firsov, and Lang[5] for one-phonon processes. The magnitude of the 2NPO deformation potential used in the present calculation is consistent with the value determined in I.

II. TWO-PHONON DEFORMATION POTENTIAL

The Hamiltonian H of the system is given by

$$H = H_0 + H_{eR} + H_{ep} \quad . \tag{1}$$

The unperturbed Hamiltonian H_0 is the sum of the following three terms:

$$\begin{aligned} H_e &= \sum_{\lambda} E_\lambda c_\lambda^\dagger c_\lambda , \\ H_R &= \sum_{\vec{\chi},\vec{\epsilon}} \hbar\omega_{\vec{\chi},\vec{\epsilon}} (A_{\vec{\chi},\vec{\epsilon}}^\dagger A_{\vec{\chi},\vec{\epsilon}} + \tfrac{1}{2}), \\ H_L &= \sum_{\vec{q},nj} \hbar\omega_{\vec{q},nj} (a_{\vec{q},nj}^\dagger a_{\vec{q},nj} + \tfrac{1}{2}). \end{aligned} \tag{2}$$

$c_\lambda^\dagger$ (c_λ) is creation (annihilation) operator for electrons in state λ. We will assume isotropic parabolic bands for electrons with energy $E_{c\vec{k}} = \hbar^2 k^2/$

$2m^*$, where m^* is the effective mass of the electrons in band c and $\vec{k}$ is the quasimomentum. $\omega_{\vec{\chi},\vec{\epsilon}}$ is the frequency of photons with wave vector $\vec{\chi}$ and polarization $\vec{\epsilon}$. $\omega_{\vec{q},nj}$ denotes the frequency of phonons with wave vector $\vec{q}$, branch index n, and polarization j. $A^\dagger$ (A) and $a^\dagger$ (a) are the creation (annihilation) operators for photons and phonons, respectively.

H_{eR} is the electron-photon interaction given by

$$H_{eR} = -\frac{e}{m^*}\left(\frac{2\pi\hbar}{\Omega\epsilon_\infty\omega_{\vec{\chi}}}\right)^{1/2}\langle\lambda'|\vec{\epsilon}\cdot\vec{p}\,e^{i\vec{\chi}\cdot\vec{r}}|\lambda\rangle \times c^\dagger_{\lambda'}c_\lambda A^\dagger_{\vec{\chi},\vec{\epsilon}} + \text{c.c.}, \tag{3}$$

where ϵ_∞ is the high-frequency dielectric constant, Ω is the volume of the crystal, e is the electronic charge, $\vec{p}$ is the momentum operator of the electron, and $|\lambda\rangle$ is the electron wave function in state λ.

Electron-phonon interaction H_{ep} is usually given as an expansion in its atomic displacements $u_{n\alpha}$ of the αth atom in the nth cell:

$$H_{ep} = \sum_{n,\alpha}\vec{\nabla}V\cdot\vec{u}_{n\alpha} + \frac{1}{2}\sum_{\substack{n,\alpha\\ n',\alpha'}}\vec{u}_{n'\alpha'}\cdot\vec{\nabla}\vec{\nabla}V\cdot\vec{u}_{n\alpha}+\cdots = H_1+H_2+\cdots. \tag{4}$$

As was indicated in I, an electron interacts with two phonons either by H_2 or by a repeated application of H_1 through intermediate states. The latter process is formally written as $H_1(1/E-H)H_1$, where H is the entire Hamiltonian for the electrons and the lattice. Interference of matrix elements between H_2 and $H_1(1/E-H)H_1$ may result in some cancellation and weaken the resultant electron–two-phonon interaction. For the case of long-wavelength acoustic-phonon modes, the results of interference and cancellation are dramatic as shown by Holstein.[6] In the limit of long wavelength, an acoustic phonon is equivalent to translation of the lattice. The detailed cancellation in this special case reflects the translational invariance of the crystalline lattice. However, for two NPO phonons, in spite of interference, reasons have been advanced to show that cancellation is believed not to be serious. It was argued in I, that, for near-zone-edge optical phonons with large momentum transfer, translational invariance of the crystal no longer enforces detailed cancellation of H_2 and $H_1(1/E-H)H_1$ as happens for the case of long-wavelength acoustic phonons. However, the assumption made in I that only H_2 gives rise to the observed resonant electron–2NPO-phonon coupling is not necessary. In general, finite contributions to $H_1(1/E-H)H_1$ exist with all possible intermediate states from all bands. The results and conclusions of I remain unchanged if the deformation potential determined therein is taken pertaining to the effective Hamiltonian H_2^{eff} as defined immediately below. This persistent inference of matrix elements is most conveniently handled by defining an effective carrier–two-phonon interaction Hamiltonian H_2^{eff}. The procedure is illustrated diagrammatically in Fig. 1 and corresponds to the following expression:

$$H_2^{\text{eff}} = \sum_{\lambda\lambda'}\sum_{\vec{q}nj}\sum_{\vec{q}'n'j'} V_{\vec{q}nj,\vec{q}'n'j'} \times M_{\lambda'\lambda}(\vec{Q})\,c^\dagger_{\lambda'}c_\lambda\Phi_{\vec{q}nj}\,\Phi_{\vec{q}'n'j'}, \tag{5}$$

$$V_{\vec{q}nj,\vec{q}'n'j'} = \hbar\mathfrak{D}(\vec{q}nj,\vec{q}'n'j')/\rho\Omega a^2(\omega_{\vec{q}nj}\,\omega_{\vec{q}'n'j'})^{1/2}, \tag{6}$$

$$M_{\lambda'\lambda}(\vec{Q}) = \langle F_{\lambda'}|e^{i\vec{Q}\cdot\vec{r}}|F_\lambda\rangle, \tag{7}$$

where $|F_\lambda\rangle$ is the envelope function of electron states.

$\Phi_{\vec{q}nj} = a_{\vec{q}nj} + a^\dagger_{-\vec{q}nj}$ is the field operator for phonons of branch n and polarization j, and $\vec{Q} = \vec{q}+\vec{q}'$. ρ is the mass density and a is the lattice constant of the crystal. $\mathfrak{D}$ is the two-phonon effective deformation potential and is a function of $\vec{q}nj$, $\vec{q}'n'j'$. It depends also on the electronic quantum numbers λ, λ' because it must involve transitions between these states and various intermediate states. Since in this work, we consider only the case when both states belong to the same band, this dependence is not important. Hence we have suppressed the labels λ and λ' in $\mathfrak{D}$. It is expected to be smaller than the value if determined from the $H_{ep}^{(2)}$ term alone.

It will be found convenient to rewrite H_2^{eff} in a form that resembles ordinary electron deformation-potential interaction with one phonon. This is accomplished by first replacing the sums over $\vec{q}$ and $\vec{q}'$ by sums over $\vec{Q}$ and $\vec{q}$. For a fixed $\vec{Q}$, let the set of critical points in the joint energy density of states for all combinations of two phonons $nj\,\vec{q}$ and $n'j'\vec{q}'$ with the sum of their wave vectors $\vec{q}+\vec{q}' = \vec{Q}$ be indexed by Ξ. Further, denote the energy of these two-phonon critical points by $\hbar\omega_\Xi(\vec{Q})$. Second, the summation over $\vec{q}$ is transformed to integration over the variable ω_s, which depends

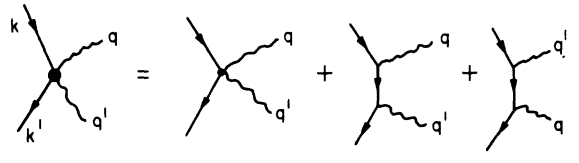

FIG. 1. Diagram for the effective electron–two-phonon-interaction vertex. The solid and the wavy lines represent, respectively, electron and phonon propagators.

on the parameters $njn'j'$, and is the sum $\omega_{nj\vec{q}} + \omega_{n'j'\vec{q}'}$. Let

$$f(\vec{q}; nj, n'j', \vec{Q}) = \mathfrak{D}(\vec{q}nj, \vec{q}'n'j')/(\omega_{\vec{q}nj}\omega_{\vec{q}'n'j'})^{1/2}. \quad (8)$$

Then

$$\sum_{nj,n'j'}\sum_{\vec{q}} f(\vec{q}; nj, n'j', \vec{Q}) = \frac{\Omega}{(2\pi)^3}\sum_{nj,n'j'}\int d\vec{q} f(\vec{q}; nj, n'j', \vec{Q}) = \frac{\Omega}{(2\pi)^3}\sum_{nj,n'j'}\int N(\omega_s) f(q(\omega_s); nj, n'j', \vec{Q}) d\omega_s. \quad (9)$$

$N(\omega_s)$ in Eq. (9) is the joint density of states of phonon pairs $nj\vec{q}$ and $n'j'\vec{q}'$ with $\vec{q}+\vec{q}' = \vec{Q}$. We replace $\sum_{nj,n'j'} N(\omega_s)$ by

$$\sum_{nj,n'j'} N(\omega_s) = [(2\pi)^3/\Omega_c]\sum_{\Xi} p_{\Xi}(\vec{Q})\,\delta(\omega_s - \omega_{\Xi}(\vec{Q})). \quad (10)$$

Here Ω_c = volume of the primitive cell. For a crystal with zinc-blende structure and lattice constant a such as InSb, $\Omega_c = \frac{1}{4}a^3$. Note that the indices nj and $n'j'$ are subsumed in Ξ. Hence Ξ can be defined as an index for the critical points of the combinations of two phonons $|nj\vec{q}\rangle$ $|n'j'\vec{Q}-\vec{q}\rangle$. From Eq. (10) we see that the quantity $p_{\Xi}(\vec{Q})$ has the meaning of the fraction of the Brillouin-zone (BZ) volume that constitute the critical point Ξ for a fixed $\vec{Q}$. This meaning can be brought out clearly by considering a hypothetical case of a lattice with two totally dispersionless phonons $n_h j_h$ and $n'_h j'_h$ with constant frequencies ω_h and ω'_h, respectively. Let Ξ_h be the critical point made up by combining these two-phonon branches with critical-point energy $\hbar\omega_{\Xi} = \hbar\omega_h + \hbar\omega'_h$. Then from Eq. (10), we must have $p_{\Xi_h}(\vec{Q}) = 1$ for any $\vec{Q}$.

Continuing from Eq. (10), we substitute into Eq. (9),

$$\sum_{nj,n'j'}\sum_{\vec{q}} f(\vec{q}; nj, n'j', \vec{Q}) = \frac{\Omega}{\Omega_c}\sum_{\Xi} p_{\Xi}(\vec{Q}) f(\omega_{\Xi}(\vec{Q}); nj, n'j', \vec{Q}). \quad (11)$$

The quantity $f(\omega_{\Xi}(\vec{Q}); nj, nj, \vec{Q})$ is an average of $f(\vec{q}, nj, nj, \vec{Q})$ over all sets of phonon pairs $\{|nj\vec{q}\rangle, |n'j', \vec{q}')\}$ that constitute Ξ. If we define $\bar{\omega}_{\Xi}$ by $\bar{\omega}_{\Xi}^2 = \mathrm{Av}\{\omega_{\vec{q}nj}\omega_{\vec{q}n'j}\}$ as the average over Ξ of the product $\omega_{\vec{q}nj}\omega_{\vec{q}n'j}$, and assume that $\mathfrak{D}(\vec{q}nj, \vec{q}nj)$ depends on Ξ and $\vec{Q}$ only, then

$$\sum_{nj,n'j'}\sum_{\vec{q}} f(\vec{q}; nj, n'j', \vec{Q}) = (\Omega/\Omega_c)\sum_{\Xi} p_{\Xi}(\vec{Q})\mathfrak{D}_{\Xi}(\vec{Q})/\bar{\omega}_{\Xi}. \quad (12)$$

Substituting back into Eq. (5) gives,

$$H_2^{eff} = \sum_{\lambda',\lambda,\vec{Q},\Xi} (\Omega/\Omega_c)^{1/2}(\hbar/\rho\Omega a^2\bar{\omega}_{\Xi})^{1/2}(\mathfrak{D}p^{1/2})_{Q,\Xi} \times M_{\lambda'\lambda}(\vec{Q})\, c^{\dagger}_{\lambda'} c_{\lambda}\, \Phi_{\{\vec{q}\},\Xi}\Phi_{\{-\vec{q}+\vec{Q}\},\Xi}. \quad (13)$$

We have rewritten the product of $p_{\Xi}(\vec{Q})\,\mathfrak{D}_{\Xi}(\vec{Q})$ as $(\mathfrak{D}p^{1/2})_{Q,\Xi}$. The product $\Phi_{\{\vec{q}\},\Xi}\Phi_{\{-\vec{q}+\vec{Q}\},\Xi}$ symbolically stands for the totality of all pairs of phonons that constitute Ξ. Since $\Omega_c = \frac{1}{4}a^3$ for InSb, we have an alternative expression for H_2^{eff} as

$$H_2^{eff} = \sum_{\lambda',\lambda,\vec{Q},\Xi} (\hbar/\rho\Omega a^2\bar{\omega}_{\Xi})^{1/2} d_{\Xi}(\vec{Q}) \times M_{\lambda'\lambda}(\vec{Q})\, c^{\dagger}_{\lambda'} c_{\lambda}\, \Phi_{\{\vec{q}\},\Xi}\Phi_{\{-\vec{q}+\vec{Q}\},\Xi}, \quad (14)$$

with

$$d_{\Xi}(\vec{Q}) = (4\hbar/\rho a^5\bar{\omega}_{\Xi})^{1/2}(\mathfrak{D}p^{1/2})_{Q,\Xi}.$$

This last form of H_2^{eff} resembles a one-phonon deformation-potential interaction with deformation potential $d_{\Xi}(\vec{Q})$. In the present formulation of carrier–two-phonon interaction, properties of the lattice normal vibrational modes that are necessary to describe the interaction are the ensemble $\{\Xi, \omega_{\Xi}(\vec{Q}), p_{\Xi}(\vec{Q})\}$. The first two quantities, Ξ and $\omega_{\Xi}(\vec{Q})$, can often be estimated by inspection of the phonon dispersion curves. On the other hand, $p_{\Xi}(\vec{Q})$ can only, in principle, be obtained if we know the phonon spectrum over the entire BZ. This information is generally not readily available from present day lattice-dynamics experiments or calculations. One advantage of the effective-Hamiltonian formulation is that this information is absorbed into the observable quantity $(\mathfrak{D}p^{1/2})_{Q,\Xi}$ of $d_{\Xi}(\vec{Q})$ which are directly related to experimental observations.

III. TWO-PHONON CONDUCTIVITY TENSOR

We derive the expression for the frequency-dependent conductivity tensor $\sigma_{ij}(\omega)$ by extending the formalism of Gurevich *et al.*[5] to the 2NPO processes. The absorption coefficient α is related to the conductivity by

$$\alpha = 4\pi\,\mathrm{Re}\sigma/cn_r,$$

where n_r is the refractive index and c is the velocity of light in vacuum. Hence in the following we calculate only the real part of $\sigma(\omega)$. Gurevich *et al.*[5] have shown that the general expression for $\mathrm{Re}\sigma(\omega)$ can be written

$$\mathrm{Re}\sigma_{ij}(\omega) = 2(e^2\hbar^2/\Omega m^{*2})\sum_{\vec{k},\vec{k}'}(k_i k'_j/\omega)\,\mathrm{Im}Q^R_{\vec{k},\vec{k}'}(\omega), \quad (15)$$

where $Q^R_{\vec{k},\vec{k}'}(\omega)$ is the Fourier transform of the retarded Green's time function

$$Q^R_{\vec{k},\vec{k}'}(\omega) = \int_{-\infty}^{+\infty} e^{i\omega t}\, Q^R_{\vec{k},\vec{k}'}(t)\,dt,$$

$$Q^R_{\vec{k},\vec{k}'}(t) = \frac{i}{\hbar}\,\langle e^{iH't/\hbar}\, c^{\dagger}_{\vec{k}'} c_{\vec{k}'}\, e^{-iH't/\hbar}\, c^{\dagger}_{\vec{k}} c_{\vec{k}}$$

$$-c_{\vec{k}}^{\dagger}c_{\vec{k}}e^{iH't/\hbar}c_{\vec{k}'}^{\dagger}c_{\vec{k}'}e^{-iH't/\hbar}\rangle, \quad t>0$$
$$=0, \quad t<0. \tag{16}$$

Here $H'=H-\zeta N$, ζ is the chemical potential. The symbol $\langle\cdots\rangle$ denotes thermal average. Equation (15) can be expanded in powers of the electron-phonon interaction.[7] In Ref. 5 it is shown that the zero-order term does not contribute to $\mathrm{Re}\sigma_{ij}$ and the contribution to $\mathrm{Re}\sigma_{ij}$ is calculated from the next nonzero term involving electron scattering by one LO phonon

$$\mathrm{Re}\sigma_{xx}^{(1)}=\frac{2^{3/2}n_0e^4}{3\pi m^*\omega_l\hbar}\left(\frac{m^*}{2\hbar\omega_l}\right)^{1/2}\left(\frac{1}{\epsilon_\infty}-\frac{1}{\epsilon_0}\right)\left(\frac{\omega_l}{\omega}\right)^{5/2}$$
$$\times\frac{\sinh y}{\sinh z}\left[|y-z|K_1(|y-z|)+(y+z)K_1(|y+z|)\right], \tag{17}$$

where K_1 is the modified Bessel function of the first order, n is electron density, $y=\hbar\omega/2k_BT$, $z=\hbar\omega_l/2k_BT$, and ω_l is the frequency of LO phonons.

In the Appendix we calculate $\mathrm{Re}\sigma_{ij}^{(2)}$, the contribution to $\mathrm{Re}\sigma_{ij}$ due to electron scattering by two nonpolar optical phonons. It is shown that

$$\mathrm{Re}\sigma_{xx}^{(2)}(\omega)=\frac{e^2}{2m^{*2}\hbar\omega^3}\sum_{\substack{\vec{q},\vec{q}'\\ j,j'}}\left\{|V_{\vec{q}\vec{q}'}|^2\frac{(q_x+q_x')^2\sinh(\hbar\omega/2k_BT)}{\sinh(\hbar\omega_{q'}/2k_BT)\sinh(\hbar\omega_q/2k_BT)}\right.$$
$$\left.\times\sum_{l,m=-1,1}\left[\sinh\left(\frac{\hbar(\omega+l\omega_q+m\omega_{q'})}{2k_BT}\right)\right]^{-1}\mathrm{Im}\Pi_{\vec{q},\vec{q}'}(\omega+l\omega_q+m\omega_{q'})\right\}. \tag{18}$$

Here j stands for both branch index and polarization of phonons and

$$\Pi_{\vec{q},\vec{q}'}(\omega)=\frac{1}{\Omega}\sum_{\vec{k}}\frac{n_{\vec{k}+\vec{q}+\vec{q}'}-n_{\vec{k}}}{\omega+(E_{\vec{k}}-E_{\vec{k}+\vec{q}+\vec{q}'})/\hbar+iS}, \tag{19}$$

where $n_{\vec{k}}$ is the Fermi function of $E_{\vec{k}}$ and $S=0^+$. $V_{\vec{q},\vec{q}'}=\hbar\mathfrak{D}/\rho\Omega a^2(\omega_q\omega_{q'})^{1/2}$ characterizes the two-phonon deformation potential as defined in Eq. (6).

After changing the variables $\vec{q}$ and $\vec{q}'$ in Eq. (18) to $\vec{Q}=\vec{q}+\vec{q}'$ and $\vec{q}$, we get

$$\mathrm{Re}\sigma_{xx}^{(2)}\frac{e^2\hbar\mathfrak{D}^2}{2m^{*2}\omega^3\rho^2\Omega^2a^4}\sum_{\vec{Q},\vec{q},j,j'}\left\{\frac{Q_x^2}{\omega_{\vec{q}}\omega_{\vec{Q}-\vec{q}}}\frac{\sinh(\hbar\omega/2k_BT)}{\sinh(\hbar\omega_q/2k_BT)\sinh(\hbar\omega_{Q-q}/2k_BT)}\right.$$
$$\left.\times\sum_{l,m=-1,1}\left[\sinh\left(\frac{\hbar(\omega+l\omega_q+m\omega_{Q-q})}{2k_BT}\right)\right]^{-1}\mathrm{Im}\Pi(\omega+l\omega_q+m\omega_{Q-q})\right\}. \tag{20}$$

As shown in Eq. (8)–(11) we can write

$$\frac{1}{\Omega}\sum_{\vec{q},jj'}=\sum_{jj'}\frac{1}{(2\pi)^3}\int d^3\vec{q}=\frac{N}{\Omega}\int g(\omega_s)d\omega_s,$$

where N is the number of unit cells and $g(\omega_s)$ is the density of phonon states. Assuming that

$$g(\omega_s)=\sum_{\alpha}p(\omega_\alpha)\,\delta(\omega_s-\omega_\alpha),$$

where ω_α are the frequencies at critical points, the following expression for $\sigma^{(2)}$ is obtained:

$$\mathrm{Re}\sigma_{xx}^{(2)}=\frac{e^2}{2m^{*2}\omega^3}\sum_{\alpha}\frac{\hbar(\mathfrak{D}p_\alpha^{1/2}/\Omega_c^{1/2})^2}{\rho^2a^4\omega_\alpha^2}\frac{\sinh(\hbar\omega/2k_BT)}{[\sinh(\hbar\omega_\alpha/2k_BT)]^2}$$
$$\times\frac{1}{(2\pi)^3}\int d^3Q\,Q^2\sin^2\theta\cos^2\phi\left(\sum_{l,m=-1,1}\frac{\Pi_Q(\omega+l\omega_\alpha+m\omega_\alpha)}{\sinh[(\hbar\omega+l\omega_\alpha+m\omega_\alpha)/2k_BT]}\right). \tag{21}$$

The total conductivity σ_{xx} is the sum of $\sigma_{xx}^{(1)}$ and $\sigma_{xx}^{(2)}$. We next calculate $\sigma_{xx}^{(2)}$ for Boltzmann and Fermi statistics.

Boltzmann Statistics

In the case of Boltzmann statistics,

$$n_{\vec{k}}=n_0(2\pi\hbar^2/m^*k_BT)^{3/2}\exp(-\hbar^2k^2/2m^*k_BT),$$

where n_0 is the concentration of electrons. Substituting this in Eq. (18) we get

$$\mathrm{Im}\Pi_Q(\omega)=n_0\left(\frac{\pi m^*}{2k_BT}\right)^{1/2}Q^{-1}(1-e^{-\hbar\omega/k_BT})$$
$$\times\exp\left(\frac{\hbar\omega}{2k_BT}-\frac{m^*\omega^2}{2k_BTQ^2}-\frac{\hbar^2Q^2}{8m^*k_BT}\right). \tag{22}$$

From Eqs. (21) and (22) after performing integration over $\vec{Q}$ and using the relation[8]

$$\int_0^\infty d\eta\,\eta e^{-\eta-z^2/4\eta} = \tfrac{1}{2}z^2K_2(|z|)$$
$$= (\tfrac{1}{8}\pi|z|^2)^{1/2}e^{-z}, \quad z \gg 1$$
$$= 1 \qquad z \ll 1, \qquad (23)$$

where K_2 is a modified Bessel function of order 2, we obtain

$$\mathrm{Re}\sigma_{xx}^{(2)} \sum_{\mathbf{x}} \frac{4n_0e^2m^{*1/2}}{3\pi^{3/2}\omega_{\mathbf{x}}^2} \frac{(\mathfrak{D}p_{\mathbf{x}}^{1/2}/\Omega_c^{1/2})^2}{\rho^2a^4(\hbar\omega)^{3/2}}$$
$$\times \frac{\sinh z}{\sinh y_1 \sinh y_2} \mathfrak{F}\left(\frac{\hbar\omega}{2k_BT}, \frac{\hbar\omega_{\mathbf{x}}}{2k_BT}, \frac{\hbar\omega_{\mathbf{x}}}{2k_BT}\right), \qquad (24)$$

with

$$\mathfrak{F}(z, y_1, y_2) = z^{-3/2}\,[\tfrac{1}{2}(z-y_1-y_2)^2K_2(|z-y_1-y_2|)$$
$$+\tfrac{1}{2}(z-y_1+y_2)^2K_2(|z-y_1+y_2|)$$
$$+\tfrac{1}{2}(z+y_1-y_2)^2K_2(|z+y_1-y_2|)$$
$$+\tfrac{1}{2}(z+y_1+y_2)^2K_2(|z+y_1+y_2|)]. \qquad (25)$$

In order to compare the frequency and temperature dependence of $\sigma^{(1)}$ and $\sigma^{(2)}$, the contributions to σ due to one LO phonon and 2 NPO phonons, respectively, we first consider the limiting cases as in Ref. 5 at very-low temperatures ($\hbar\omega_{\mathbf{x}} \gg k_BT$) when the optical phonons are not excited and electron scattering takes place through phonon emission.

(i) When $\hbar\omega \gg 2\hbar\omega_{\mathbf{x}}$, Eq. (14) gives

$$\mathrm{Re}\sigma_{xx}^{(2)} = \sum_{\mathbf{x}} \frac{2^{3/2}}{3\pi} \frac{n_0e^2m^{*1/2}\mathfrak{D}^2p_{\mathbf{x}}}{\rho^2a^4\omega_{\mathbf{x}}^2\Omega_c}$$
$$\times (\hbar\omega)^{-3/2}\{1-2\omega_{\mathbf{x}}/\omega\}^{3/2}. \qquad (26)$$

This shows that $\sigma \sim \lambda^{1.5}$ as in the case of scattering by acoustic phonons.[1] σ varies as $\lambda^{2.5}$ in case of one LO phonon. The total conductivity σ_{xx} is the sum of $\sigma_{xx}^{(1)}$ and $\sigma_{xx}^{(2)}$. With the two-phonon deformation potential determined in Sec. II, the resultant σ is found to vary roughly as $\lambda^{2.15}$.

(ii) If $\hbar\omega \ll 2\hbar\omega_{\mathbf{x}}$, then

$$\mathrm{Re}\sigma_{xx}^{(2)} = \sum_{\mathbf{x}} \frac{2^{3/2}}{3\pi} \frac{n_0e^2m^{*1/2}\mathfrak{D}^2p_{\mathbf{x}}}{\rho^2a^4\Omega_c\omega_{\mathbf{x}}^2} (\hbar\omega)^{-3/2}$$
$$\times (2\omega_{\mathbf{x}}/\omega - 1)^{3/2}e^{-2\hbar\omega_{\mathbf{x}}/k_BT}. \qquad (27)$$

(iii) If $|\hbar(\omega - 2\omega_{\mathbf{x}})| \ll k_BT$, then

$$\mathrm{Re}\sigma_{xx}^{(2)} = \sum_{\mathbf{x}} \frac{32}{3\pi} \frac{n_0e^2m^{*1/2}\mathfrak{D}^2p_{\mathbf{x}}}{(2\pi)^{1/2}\rho^2a^4\Omega_c\omega_{\mathbf{x}}^2} \left(\frac{k_BT}{\hbar^2\omega^2}\right)^{3/2}. \qquad (28)$$

Here $\sigma^{(2)}$ varies as $T^{3/2}$, whereas in the case of one LO phonons it varies as $T^{1/2}$. From Eqs. (27) and (28) it is seen that at sufficiently low temperatures, $\mathrm{Re}\sigma^{(2)}(\omega)$ shows an abrupt increase in the frequency range where $\hbar(\omega - 2\omega_{\mathbf{x}}) \sim k_BT$.

The results of detailed computation from Eqs. (24) and (17) are shown in Figs. 2 and 3 for InSb. We assume that the optical-phonon frequencies are independent of branch indices. Then the sums over branch indices give a factor of 4 for the two TO branches. In Figs. 2(a) and 2(b), the conductivities σ, $\sigma^{(1)}$, and $\sigma^{(2)}$ at room temperature are plotted as function of wavelength λ of the incident radiation. From magneto-optical data, Ngai and Johnonson[4] estimated the two-phonon deformation potential $\mathfrak{D}p_{\mathbf{x}}^{1/2}$ to be in the range of 10^4–1.5×10^4 eV. We show conductivities for two values of $\mathfrak{D}p_{\mathbf{x}}^{1/2} = 10^4$ and 1.5×10^4 eV. All other relevant material parameters at room temperature are taken from Ref. 2, e.g., $\hbar\omega_{\mathbf{x}} = 265\,^\circ$K, $m^* = 0.0116m_e$, $\epsilon_0 = 18.7$, $\epsilon_\infty = 15.7$, refractive index $n_r = 3.3$ at $\lambda = 9$ μm. Our calculations in the wavelength region 8–30 μm show that

$$\sigma \propto \lambda^{2.15} \quad \text{for } \mathfrak{D}p_{\mathbf{x}}^{1/2} = 1.5\times10^4 \text{ eV}$$
$$\propto \lambda^{2.25} \quad \text{for } \mathfrak{D}p_{\mathbf{x}}^{1/2} = 10^4 \text{ eV}.$$

This frequency dependence is in better agreement with experiment[9] ($\sigma \propto \lambda^2$) than the previous calculations.[1-3] The absorption coefficient as calculated by Jensen[2] for $\lambda = 9$-μm and $T = 298\,^\circ$K using only one-phonon scattering is smaller than experimental value. For this wavelength and temperature we find that the ratio $\sigma^{(2)}/\sigma^{(1)}$ varies between 0.8 and 1.8 depending on the value of $\mathfrak{D}p_{\mathbf{x}}^{1/2}$. Thus both the magnitude and the frequency dependence of σ show that the two-phonon processes are significant for free-carrier absorption in InSb.

Figures 3(a) and 3(b) are plots of σ, $\sigma^{(1)}$, and $\sigma^{(2)}$ as functions of temperature for incident radiation of wavelength $\lambda = 9$-μm. In this calculation the temperature variation of the material parameters are neglected and the values at room temperature are used. At low temperatures $\sigma^{(1)}$ dominates, but as the temperature increases $\sigma^{(2)}$ increases in strength and ultimately becomes larger than $\sigma^{(1)}$. The crossover temperature depends on the two-phonon deformation potential. For $\mathfrak{D}p_{\mathbf{x}}^{1/2} = 10^4$ eV, the crossover temperature is 350 °K. Further experimental studies of the temperature dependence of the absorption coefficient will serve to confirm the importance of 2NPO scattering in free-carrier absorptions.

Fermi Statistics at $T = 0$

In this case with $n_{\mathbf{k}} = 1$ for $|k| \le k_F$ (Fermi momentum) and $n_{\mathbf{k}} = 0$ for $|k| > k_F$, we find from Eq. (18) that

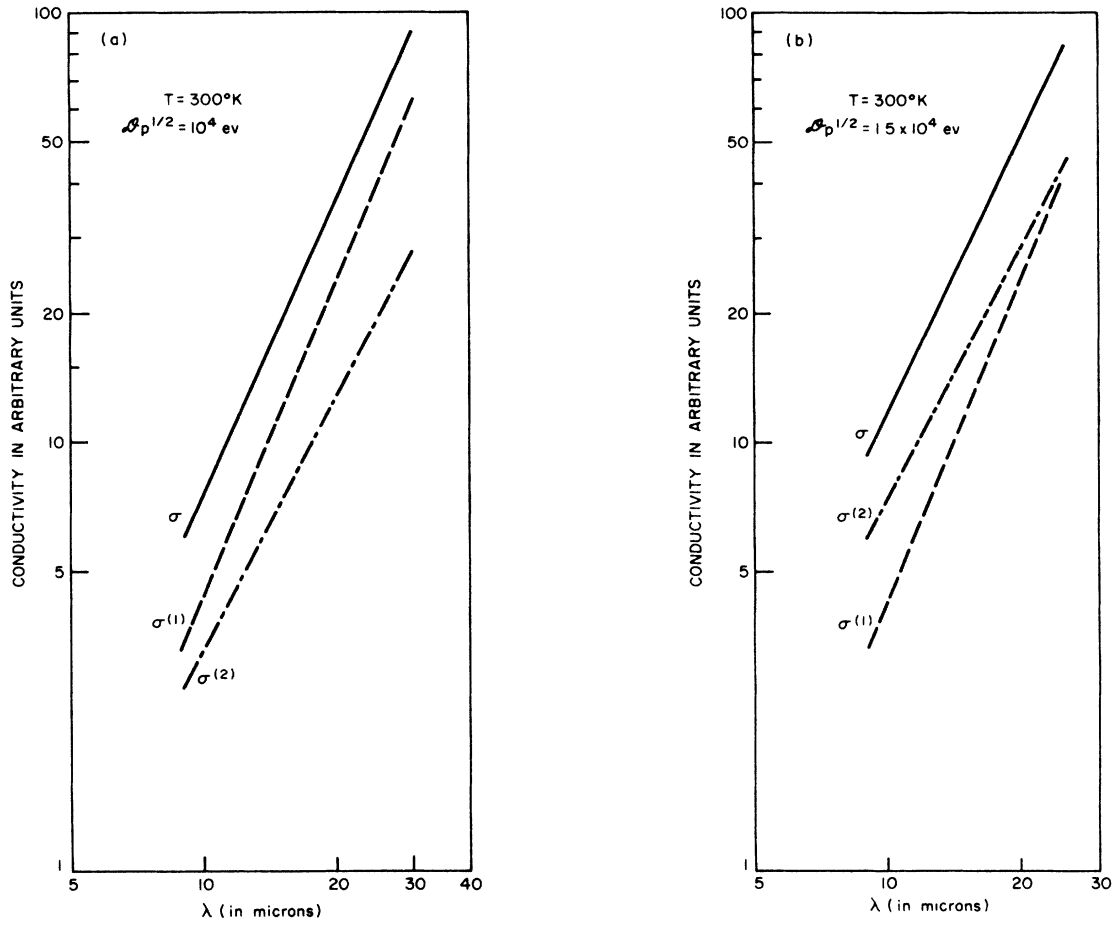

FIG. 2. (a) Conductivity versus wavelength of incident light at $T = 300\,°\mathrm{K}$ and $\mathfrak{D}\,p^{1/2} = 10^4$ eV. σ is the total conductivity. $\sigma^{(1)}$ and $\sigma^{(2)}$ denote, respectively, the contributions from one-phonon and two-phonon scattering. (b) Conductivity versus wavelength at $T = 300\,°\mathrm{K}$ and $\mathfrak{D}\,p^{1/2} = 1.5 \times 10^4$ eV.

$$\mathrm{Im}\Pi_{\vec{Q}}(\omega) = \frac{m^*}{4\pi\hbar Q}\left[\left(k_F^2 - \frac{m^{*2}\omega^2}{\hbar^2 Q^2} - \frac{Q^2}{4} + \frac{m^*\omega}{\hbar}\right)\theta(Q - 2^{1/2}k_F\{(1+\tfrac{1}{2}\xi) - (1+\xi)^{1/2}\}^{1/2})\right.$$
$$\times\theta(2^{1/2}k_F\{(1+\tfrac{1}{2}\xi) + (1+\xi)^{1/2}\}^{1/2} - Q) - \left(k_F^2 - \frac{m^{*2}\omega^2}{\hbar^2 Q^2} - \frac{Q^2}{4} - \frac{m^*\omega}{\hbar}\right)$$
$$\left.\times\theta(Q - 2^{1/2}k_F\{(1-\tfrac{1}{2}\xi) - (1-\xi)^{1/2}\}^{1/2})\theta(2^{1/2}k_F\{(1-\tfrac{1}{2}\xi) + (1-\xi)^{1/2}\}^{1/2} - Q)\right], \tag{29}$$

where

$$\xi = \frac{\hbar\omega}{E_F} = \frac{2m^*\omega}{\hbar k_F^2}$$

and θ is the step function

$$\theta(x) = 1, \quad x > 0$$
$$= 0, \quad x < 0.$$

In the second term inside the square bracket of Eq. (29) $\xi < 1$. Using Eq. (29) in (24), we get the following for $\sigma^{(2)}(\omega)$:

$$\mathrm{Re}\sigma_{xx}^{(2)}(\omega) = \sum_{\vec{q}} \frac{8m^{*2}e^2\mathfrak{D}^2 p_{\vec{q}}}{9\pi^3\hbar^3\rho^2 a^4\Omega_c} \times\left(\frac{E_F}{\hbar\omega}\right)^{3/2}\frac{1}{\omega_{\vec{q}}^2}\, f\left(\frac{\hbar\omega - 2\hbar\omega_{\vec{q}}}{E_F}\right), \tag{30}$$

where

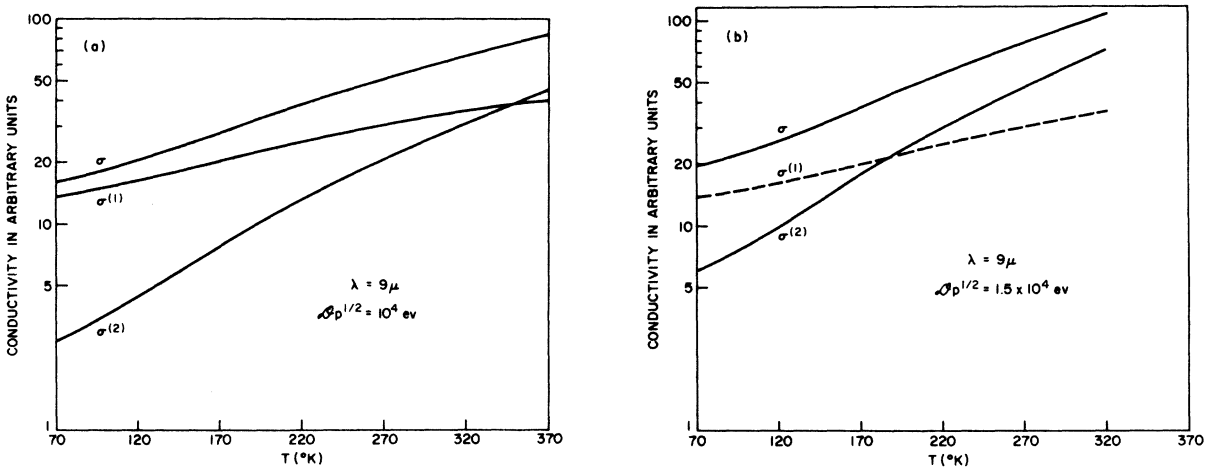

FIG. 3. (a) Conductivity as a function of temperature at $\lambda = 9$ μm for $\mathfrak{D}\, p^{1/2} = 10^4$ eV. (b) Conductivity versus T at $\lambda = 9$ μm and $\mathfrak{D}\, p^{1/2} = 1.5 \times 10^4$ eV.

$$f(\xi) = 0, \qquad \xi < 0$$
$$= (1+\xi)^{3/2} - (1-\xi)^{3/2}, \qquad 0 \le \xi \le 1$$
$$= (1+\xi)^{3/2}, \qquad \xi > 1. \tag{31}$$

From Eq. (31) when $\hbar\omega/E_F \gg 2\hbar\omega_{\vec{k}}/E_F$, we have

$$\mathrm{Re}\sigma_{xx}^{(2)}(\omega) = \sum_{\vec{k}} \frac{2^{3/2} n_0 e^2 m^{*1/2} \mathfrak{D}^2 p_{\vec{k}}}{3\pi\rho^2 a^4 \Omega_c \omega_{\vec{k}}^2} \times \left(\frac{1}{\hbar\omega}\right)^{3/2} \left(1 - \frac{2\omega_{\vec{k}}}{\omega}\right)^{3/2}. \tag{32}$$

This agrees with Eq. (26) obtained from Boltzmann statistics for the case $\hbar(\omega - 2\omega_{\vec{k}})/k_B T \gg 1$.

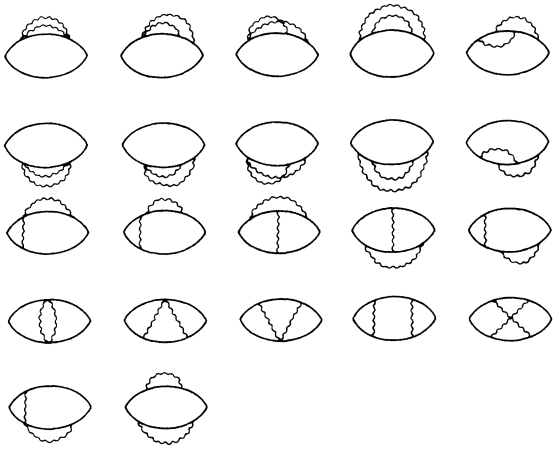

FIG. 4. Sketch of topologically distinct diagrams of all two-phonon processes that contribute to the conductivity. The solid and wavy lines are, respectively, the electron and phonon propagator. The phonon vertices where two wavy lines appear represent H_2 and the vertices with one wavy line represent H_1.

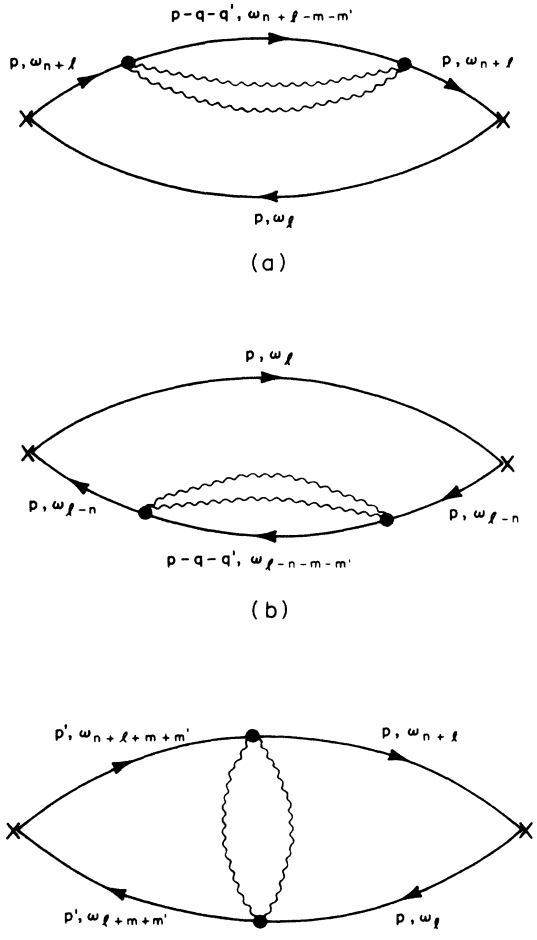

FIG. 5. Diagrams for computing $\tilde{Q}_{\vec{k}\vec{k}'}(\omega_n)$.

IV. CONCLUSIONS

In summary, we find that 2NPO scatterings occur with significant strength in a weakly polar material such as InSb. Their contributions to the free-carrier absorption coefficient are comparable to that caused by polar interaction. The comparison between theory and experiment is improved when both polar-optical and 2NPO-phonon scatterings are included. We have used the 2NPO deformation potential as determined earlier by a magneto-optical-resonance experiment.[4] The agreement achieved here gives further evidence for the published size of the 2NPO deformation potential. We have also calculated the temperature dependence of the absorption coefficient. At higher temperatures (~ room temperature) where 2NPO phonon scatterings dominate, the predicted temperature dependence is noticeably different from what is expected for LO-polar scatterings only. We suggest this to be checked experimentally. The results and conclusions of this work for InSb should also be applicable to either nonpolar or weakly polar semiconductors with a simple band structure such as Ge.

APPENDIX

$Q^R_{\vec{k}\vec{k}'}(\omega)$ is the analytic continuation of the thermodynamic Green's function on the upper half-plane, i.e.,

$$Q^R_{\vec{k}\vec{k}'}(\omega)=\tilde{Q}_{\vec{k}\vec{k}'}(-i\omega+s), \tag{A1}$$

where

$$\tilde{Q}_{\vec{k}\vec{k}'}(\omega_n)=\tfrac{1}{2}\int_{-1/k_BT}^{1/k_BT} e^{i\hbar\omega_n\tau}\,\tilde{Q}_{\vec{k}\vec{k}'}(\tau)\,d\tau,$$

$$\tilde{Q}_{\vec{k}\vec{k}'}(\tau)=\langle T_\tau(e^{H'\tau}c^\dagger_{\vec{k}'}c_{\vec{k}}e^{-H'\tau}c^\dagger_{\vec{k}}c_{\vec{k}})\rangle.$$

T_τ is the ordering operator with respect to the variable τ.

We used the diagrammatic technique of many-body theory to evaluate contributions to $\tilde{Q}_{\vec{k}\vec{k}'}(\omega_n)$ from two-phonon processes. The totality of these contributions consists of the sum of the diagrams shown in Fig. 4. Only topologically distinct diagrams are displayed in this figure. The effective electron–two-phonon-interaction vertex as defined in Fig. 1 permits partial summations of these diagrams. The diagrams that appear in the first row in Fig. 4 when summed give rise a single diagram, diagram A in Fig. 5. The electron–two-phonon-interaction vertex depicted as a heavy dot in the figure corresponds now to the effective Hamiltonian of Eq. (14). In exactly the same manner, summing the diagrams in the second (fourth) row in Fig. 4, we obtain diagram B (C) in Fig. 5. The remaining diagrams of Fig. 4 are not expressible entirely in terms of H_2^{eff}. They have the common characteristics that in the process, which any one of these diagrams describes, either the electron or the hole have to scatter with large momentum change only. Contributions from these classes of diagrams to $\tilde{Q}_{\vec{k}\vec{k}'}(\omega_n)$ can be estimated individually. They are found to be negligible compared with those of Fig. 5 for InSb, and hence will be dropped.

The contributions from the three diagrams are, respectively,

$$\tilde{Q}^{\mathrm{I}}_{\vec{k}\vec{k}'}(\omega_n)=(-k_BT)^3\sum_{\substack{l,m,m'\\ \vec{q},\vec{q}'}}\delta_{\vec{k}',\vec{k}+\vec{q}+\vec{q}'}\,|V_{\vec{q}\vec{q}'}|^2 g^{(0)}_{\vec{k}'}(\omega_{n+l+m+m'})g^{(0)}_{\vec{k}}(\omega_{l+n})$$
$$\times g^{(0)}_{\vec{k}}(\omega_l)g^{(0)}_{\vec{k}'}(\omega_{l+m+m'})d^{(0)}_{\vec{q}}(\omega_m)d^{(0)}_{\vec{q}'}(\omega_{m'}), \tag{A2}$$

$$\tilde{Q}^{\mathrm{II}}_{\vec{k}\vec{k}'}(\omega_n)=(-k_BT)^3\sum_{\substack{l,m,m'\\ \vec{q},\vec{q}'}}\delta_{\vec{k},\vec{k}'}\,|V_{\vec{q}\vec{q}'}|^2[g^{(0)}_{\vec{k}}(\omega_{n+l})]^2 g^{(0)}_{\vec{k}-\vec{q}-\vec{q}'}(\omega_{n+l-m-m'})g^{(0)}_{\vec{k}}(\omega_l)d^{(0)}_{\vec{q}}(\omega_m)d^{(0)}_{\vec{q}'}(\omega_{m'}), \tag{A3}$$

and

$$\tilde{Q}^{\mathrm{III}}_{\vec{k}\vec{k}'}(\omega_n)=\tilde{Q}^{\mathrm{II}}_{\vec{k}\vec{k}'}(-\omega_n), \tag{A4}$$

where $g^{(0)}_{\vec{k}}(\omega_l)$ is the Fourier transform of the zero-order finite-temperature Green's function given by

$$g^{(0)}_{\vec{k}}(\omega_l)=1/(i\hbar\omega_l-E_{\vec{k}}+\zeta),\quad \hbar\omega_l=(2l+1)\pi k_BT$$
$$=0,\qquad\qquad \hbar\omega_l=2l\pi k_BT, \tag{A5}$$

and $d^{(0)}_{\vec{q}}(\omega_m)$ is the same for phonons:

$$d^{(0)}_{\vec{q}}(\omega_m)=-2\hbar\omega_{\vec{q}}/[(\hbar\omega_{\vec{q}})^2+(\hbar\omega_m)^2],\quad \hbar\omega_m=2\pi mk_BT$$
$$=0,\qquad\qquad \hbar\omega_m=(2m+1)\pi k_BT. \tag{A6}$$

Substituting (A5) and (A6) in (A2)–(A4) and after performing fractional decomposition of the product of four functions $g^{(0)}$, the sum over l is done using the identity

$$\sum_{l} 1/(i\hbar\omega_l - E_{\vec{k}}) = n_{\vec{k}}/k_B T. \tag{A7}$$

Thus we obtain

$$\tilde{Q}^{\mathrm{I}}_{\vec{k}\vec{k}'}(\omega_n) = \frac{(k_B T)^2}{(\hbar\omega_n)^2} \sum_{\substack{m,m' \\ \vec{q},\vec{q}'}} |V_{\vec{q}\vec{q}'}|^2 \delta_{\vec{k}',\vec{k}+\vec{q}+\vec{q}'} (n_{\vec{k}} - n_{\vec{k}'}) d^{(0)}_{\vec{q}}(\omega_m) d^{(0)}_{\vec{q}'}(\omega_{m'})$$
$$\times \left\{ \frac{1}{i\hbar\omega_{n+m+m'} + E_{\vec{k}} - E_{\vec{k}'}} + \frac{1}{i\hbar\omega_{m+m'-n} + E_{\vec{k}} - E_{\vec{k}'}} - \frac{2}{i\hbar\omega_{m+m'} + E_{\vec{k}} - E_{\vec{k}'}} \right\} \tag{A8}$$

and

$$\tilde{Q}^{\mathrm{II}}_{\vec{k}\vec{k}'}(\omega_n) + \tilde{Q}^{\mathrm{III}}_{\vec{k}\vec{k}'}(\omega_n) = \frac{(k_B T)^2}{(\hbar\omega_n)^2} \delta_{\vec{k},\vec{k}'} \sum_{\substack{m,m' \\ \vec{q},\vec{q}'}} |V_{\vec{q}\vec{q}'}|^2 (n_{\vec{k}-\vec{q}-\vec{q}'} - n_{\vec{k}}) d^{(0)}_{\vec{q}}(\omega_m) d^{(0)}_{\vec{q}'}(\omega_{m'})$$
$$\times \left\{ \frac{2}{i\hbar\omega_{m+m'} + E_{\vec{k}-\vec{q}-\vec{q}'} - E_{\vec{k}}} - \frac{1}{i\hbar\omega_{m+m'-n} + E_{\vec{k}-\vec{q}-\vec{q}'} - E_{\vec{k}}} - \frac{1}{i\hbar\omega_{m+m'+n} + E_{\vec{k}-\vec{q}-\vec{q}'} - E_{\vec{k}'}} \right\} . \tag{A9}$$

Hence

$$\sum_{\vec{k},\vec{k}'} k_x k'_x \tilde{Q}_{\vec{k},\vec{k}'}(\omega_n) = \left(\frac{k_B T}{\hbar\omega_n}\right)^2 \sum_{\vec{k},\vec{q},\vec{q}'} \left\{ \sum_{m,m'} d^{(0)}_{\vec{q}}(\omega_m) d^{(0)}_{\vec{q}'}(\omega_{m'}) \right.$$
$$\times \left[k_x (k_x + q_x + q'_x) \left(\frac{n_{\vec{k}} - n_{\vec{k}+\vec{q}+\vec{q}'}}{i\hbar\omega_{m+m'+n} + E_{\vec{k}} - E_{\vec{k}+\vec{q}+\vec{q}'}} + \frac{n_{\vec{k}} - n_{\vec{k}+\vec{q}+\vec{q}'}}{i\hbar\omega_{m+m'-n} + E_{\vec{k}} - E_{\vec{k}+\vec{q}+\vec{q}'}} \right) \right.$$
$$- k_x^2 \left(\frac{n_{\vec{k}-\vec{q}-\vec{q}'} - n_{\vec{k}}}{i\hbar\omega_{m+m'+n} + E_{\vec{k}-\vec{q}-\vec{q}'} - E_{\vec{k}}} + \frac{n_{\vec{k}-\vec{q}-\vec{q}'} - n_{\vec{k}}}{i\hbar\omega_{m+m'-n} + E_{\vec{k}-\vec{q}-\vec{q}'} - E_{\vec{k}}} \right)$$
$$\left. \left. + 2k_x (k_x + q_x + q'_x) \frac{n_{\vec{k}+\vec{q}+\vec{q}'} - n_{\vec{k}}}{i\hbar\omega_{m+m'} + E_{\vec{k}} - E_{\vec{k}+\vec{q}+\vec{q}'}} - 2k_x^2 \frac{n_{\vec{k}} - n_{\vec{k}-\vec{q}-\vec{q}'}}{i\hbar\omega_{m+m'} + E_{\vec{k}-\vec{q}-\vec{q}'} - E_{\vec{k}}} \right] \right\} . \tag{A10}$$

In Eq. (A10) make the following replacements: (i) in the third and sixth terms $\vec{k} \to \vec{k} + \vec{q} + \vec{q}'$; (ii) in the second term $\vec{k} \to \vec{k} - \vec{q} - \vec{q}'$, and then $\vec{q} \to -\vec{q}$, $\vec{q}' \to -\vec{q}'$, $m \to -m$, $m' \to -m'$; and (iii) in the fourth term $\vec{q} \to -\vec{q}$, $\vec{q}' \to -\vec{q}'$, $m \to -m$, $m' \to -m'$. Then we have

$$\sum_{\vec{k},\vec{k}'} k_x k'_x \tilde{Q}_{\vec{k}\vec{k}'}(\omega_n) = \sum_{\vec{k},\vec{q},\vec{q}'} |V_{\vec{q}\vec{q}'}|^2 \left(\frac{k_B T}{\hbar\omega_n}\right)^2 (q_x + q'_x)^2 (n_{\vec{k}+\vec{q}+\vec{q}'} - n_{\vec{k}})$$
$$\times \sum_{m,m'} \frac{d^{(0)}_{\vec{q}}(\omega_m) d^{(0)}_{\vec{q}'}(\omega_{m'})}{i\hbar\omega_{m+m'+n} + E_{\vec{k}} - E_{\vec{k}+\vec{q}+\vec{q}'}} + (\text{a real term}). \tag{A11}$$

Since in the calculation of $\mathrm{Re}\sigma_{xx}(\omega)$ we need $\mathrm{Im}\tilde{Q}_{\vec{k}\vec{k}'}(\omega)$, the real term in Eq. (A11) will be ignored. The sums over m and m' in Eq. (A11) can be converted into integrals over E and E' using the relation[5]

$$\sum_{m'} \frac{d^{(0)}_{\vec{q}'}(\omega_{m'})}{i\hbar\omega_{n+m+m'} + E_{\vec{k}} - E_{\vec{k}+\vec{q}+\vec{q}'}} = -\frac{1}{2\pi k_B T} \int_{-\infty}^{+\infty} dE' \coth \frac{E'}{2k_B T}$$
$$\times \left[(E' + i\hbar\omega_{n+m} + E_{\vec{k}} - E_{\vec{k}+\vec{q}+\vec{q}'})^{-1} \mathrm{Im} \left(\frac{1}{\hbar\omega_{\vec{q}'} + E' + is\hbar} + \frac{1}{\hbar\omega_{\vec{q}'} - E' - is\hbar} \right) \right.$$
$$\left. + \left(\frac{1}{\hbar\omega_{q'} - E' + i\hbar\omega_{n+m}} + \frac{1}{\hbar\omega_{q'} + E' - i\hbar\omega_{n+m}} \right) \mathrm{Im} \left(\frac{1}{E' + E_{\vec{k}} - E_{\vec{k}+\vec{q}+\vec{q}'} + is\hbar} \right) \right] . \tag{A12}$$

The integrals over E and E' can be easily done using the relation $\mathrm{Im}[(x+is)^{-1}] = -\pi\delta(x)$. From Eqs. (A12) and (A11) we finally obtain (after making the analytic continuation, i.e., replacing ω_n by $-i\omega + s$)

$$\mathrm{Im}\sum_{m,m'} d^{(0)}_{\vec{q}}(\omega_m)\, d^{(0)}_{\vec{q}'}(\omega_{m'})/(i\hbar\omega_{m+m'+n}+E_{\vec{k}}-E_{\vec{k}+\vec{q}+\vec{q}'})$$

$$= (2k_BT)^{-2}\sinh(\hbar\omega/2k_BT)\,[\sinh(\hbar\omega_{\vec{q}}/2k_BT)\sinh(\hbar\omega_{\vec{q}'}/2k_BT)]^{-1}$$

$$\times\Bigg[\sinh\left(\frac{\hbar\omega+\hbar\omega_q+\hbar\omega_{q'}}{2k_BT}\right)^{-1}\mathrm{Im}(\hbar\omega+\hbar\omega_q+\hbar\omega_{q'}+E_{\vec{k}}-E_{\vec{k}+\vec{q}+\vec{q}'}+is\hbar)^{-1}$$

$$+\sinh\left(\frac{\hbar\omega+\hbar\omega_q-\hbar\omega_{q'}}{2k_BT}\right)^{-1}\mathrm{Im}(\hbar\omega+\hbar\omega_q-\hbar\omega_{q'}+E_{\vec{k}}-E_{\vec{k}+\vec{q}+\vec{q}'}+is\hbar)^{-1}$$

$$+\sinh\left(\frac{\hbar\omega-\hbar\omega_q+\hbar\omega_{q'}}{2k_BT}\right)^{-1}\mathrm{Im}(\hbar\omega-\hbar\omega_q+\hbar\omega_{q'}+E_{\vec{k}}-E_{\vec{k}+\vec{q}+\vec{q}'}+is\hbar)^{-1}$$

$$+\sinh\left(\frac{\hbar\omega-\hbar\omega_q-\hbar\omega_{q'}}{2k_BT}\right)^{-1}\mathrm{Im}(\hbar\omega-\hbar\omega_q-\hbar\omega_{q'}+E_{\vec{k}}-E_{\vec{k}+\vec{q}+\vec{q}'}+is\hbar)^{-1}\Bigg]. \quad \text{(A13)}$$

Equation (18) then follows from Eqs. (A13), (A11), and (15).

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