V. CONCLUSION

For solids with filled and low-lying valence bands, the HF band structure can be calculated in a simple and a nearly exact way, using the APW scheme. The assumption of a negligible contribution of the region outside the APW spheres to exchange has been justified. The method, applied to neon and argon, gives results which are in good agreement with previous calculations of Lipari and Fowler on argon.

The HF gap is larger than the experimental optical gap by several eV, suggesting a very strong influence of the correlations. A satisfactory treatment of these correlations is obtained in a semiempirical manner: The Mott-Littleton approximation gives an evaluation of the interaction of one charge in one cell with all the other cells; the correlation energy for the valence hole is calculated using atomic results. It is shown that a polarization potential is adequate to take into account the correlation energy of the free electron in the same cell.

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Quantum Theory of a Basic Light-Matter Interaction

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Quantum field-theoretical methods are applied to the problem of determining how the exciton-lattice interaction affects the dispersion of an electromagnetic field associated with the exciton-radiation interaction. An exact solution for the retarded Green's function of the radiation field is calculated for a quantum model consisting of three interacting boson fields—photon, exciton, and phonon. The classical Green's function of a damped-harmonic-oscillator model of a dielectric is shown to be a special case of this quantum Green's function. Two sets of dispersion relations are derived; one set has well-defined energy, the other has well-defined momentum. Results of the theory clearly suggest that the exciton-lattice interaction is capable of literally damping out the "polariton" effects associated with the exciton-radiation interaction in the field solutions with well-defined energy. A Poynting theorem based on the classical model is also derived which includes effects of both spatial dispersion and damping.

I. INTRODUCTION

The quantum theory of light-matter interactions in semiconductors and dielectrics has been approached from two different directions. One group[1] has studied the electron-lattice interaction and has shown how it produces damping of excited electronic states (excitons). Another group[2] has studied the electron-radiation interaction and has shown how it produces dispersion of the radiation field. Neither of these groups has dealt rigorously with both the electron-lattice (H_{eL}) and electron-radiation (H_{eR}) interactions simultaneously.

These interactions will be given equal attention in the present paper. Quantum-mechanical results are derived which have a form closely resembling results based on a classical damped-harmonic-oscillator model of a dielectric.

A brief discussion of this classical model is given in Sec. II. A generalization of Poynting's theorem is derived which includes effects of both spatial dispersion and damping. From this an energy velocity is defined which determines the speed at which energy (electromagneticlike and/or matterlike) propagates through a classical dielectric.

A quantum field-theoretical model consisting of

three boson fields—photon, exciton, and phonon—is studied in detail in Sec. III. Formal solutions for the causal Green's functions of the photon and exciton fields are calculated following closely the method of quantum field theory described, for example, in Abrikosov *et al.*[3] The retarded Green's function of the electromagnetic field is then obtained in terms of functions which have poles in the lower-half frequency plane. Connection with the classical model is then made.

Dispersion relations are derived in Sec. IV for two physically interesting solutions which have well-defined momentum and energy, respectively. Results of the theory are described in Sec. V.

II. CLASSICAL MODEL

Much of the theory of electromagnetic energy propagation in semiconductors and dielectrics is based on a classical damped-harmonic-oscillator model.[4] This classical model is based on the incorporation of the following equation into Maxwell's equations:

$$\ddot{\vec{P}} + \Gamma\dot{\vec{P}} + \omega_0^2\vec{P} = (\Omega_P^2/4\pi)\vec{E} \ . \tag{2.1}$$

Here $\vec{P}$ represents the polarization field associated with the oscillators; ω_0 is the oscillator resonance frequency; and Ω_P, which has the dimension of a frequency, gives a measure of the strength of coupling between the oscillators and the electric field $\vec{E}$. The damping is assumed to be proportional to $\dot{\vec{P}}$ and Γ gives a measure of the strength of this damping.

This model is general enough to describe a number of interesting optical properties of solids. However, it possesses several potentially important omissions. Included among these omissions are (a) frequency-dependent damping[5-7] and (b) theoretical justification from first principles. In regard to point (a), one can assume Γ depends on frequency, but it is not obvious how this dependence arises. In regard to point (b), a theoretical basis for the classical model will be established in Sec. III.

Another important omission will now be discussed. The above model assumes that the oscillators are uncoupled. Direct interaction between the oscillators permits energy propagation that is not electromagnetic in origin. This produces a nonlocal spatial connection between $\vec{P}$ and $\vec{E}$ which has often been called spatial dispersion.[8] Spatial dispersion can be particularly important in semiconductors which possess highly mobile excitons.

Interaction between the oscillators can also be incorporated into the classical theory by introducing the term $-v^2\nabla^2\vec{P}$ into Eq. (2.1)[9]:

$$\ddot{\vec{P}} + \Gamma\dot{\vec{P}} + \omega_0^2\vec{P} - v^2\nabla^2\vec{P} = (\Omega_P^2/4\pi)\vec{E} \ . \tag{2.2}$$

Here v is a disposable parameter which has the dimension of a velocity. A classical theory can be based on Maxwell's equation plus Eq. (2.2) which includes Eq. (2.1) as a special case: $v^2 = 0$.

At this point one usually looks for plane-wave solutions of the form $e^{i(\vec{k}\cdot\vec{x}-\omega t)}$. However, to make connection with the derivation in Sec. III, it is more convenient here to calculate the classical retarded Green's function of the electromagnetic field, D_R.[10] Making use of Maxwell's equations and Eq. (2.2), one obtains

$$D_R(\vec{k}, \omega) = 4\pi c/[\omega^2\epsilon(\vec{k}, \omega) - c^2k^2] \ . \tag{2.3}$$

The dielectric constant $\epsilon(\vec{k}, \omega)$ is given by

$$\epsilon(\vec{k}, \omega) = 1 + \Omega_P^2/(\omega_0^2 + v^2k^2 - \omega^2 - i\omega\Gamma) \ . \tag{2.4}$$

A. Poynting's Theorem

It is possible to derive a Poynting's theorem based on Eq. (2.2) in a manner similar to that employed by Loudon[11] who recently derived a Poynting's theorem based on Eq. (2.1). Making use of Maxwell's equations, one has

$$\frac{c}{4\pi}\int_\sigma (\vec{E}\times\vec{H})\cdot d\vec{\sigma} + \frac{1}{4\pi}\int_\tau (\vec{E}\cdot\dot{\vec{E}} + \vec{H}\cdot\dot{\vec{H}} + 4\pi\vec{E}\cdot\dot{\vec{P}})\, d\tau = 0 \ . \tag{2.5}$$

Taking the scalar product of $\dot{\vec{P}}$ with respect to both sides of Eq. (2.2) and making use of some standard vector calculus identities, one finds that

$$\vec{E}\cdot\dot{\vec{P}} = \frac{4\pi}{\Omega_P^2}[\ddot{\vec{P}}\cdot\dot{\vec{P}} + \Gamma\dot{P}^2 + \omega_0^2\vec{P}\cdot\dot{\vec{P}} - v^2(\nabla^2\vec{P})\cdot\dot{\vec{P}}] = \frac{\partial W_1}{\partial t} + \vec{\nabla}\cdot\vec{S}_1 + \frac{4\pi\Gamma}{\Omega_P^2}\dot{P}^2 \ , \tag{2.6}$$

where

$$\vec{S}_1 = -(4\pi v^2/\Omega_P^2)[\dot{\vec{P}}\times(\vec{\nabla}\times\vec{P}) + \dot{\vec{P}}(\vec{\nabla}\cdot\vec{P})] \tag{2.7}$$

and

$$W_1 = (2\pi/\Omega_P^2)[\dot{P}^2 + \omega_0^2P^2 + v^2(\vec{\nabla}\times\vec{P})^2 + v^2(\vec{\nabla}\cdot\vec{P})^2] \ . \tag{2.8}$$

Substituting Eq. (2.6) for $\vec{E}\cdot\dot{\vec{P}}$ into Eq. (2.5), one obtains Poynting's theorem in its usual form:

$$\frac{c}{4\pi}\int_\sigma \vec{S}\cdot d\vec{\sigma} + \int_\tau d\tau\, \dot{W} + \frac{4\pi\Gamma}{\Omega_P^2}\int_\tau d\tau\, \dot{P}^2 = 0 \ . \tag{2.9}$$

Here, the Poynting vector $\vec{S}$ has been redefined to include the $\vec{S}_1$ term:

$$\vec{S} = (c/4\pi)\vec{E}\times\vec{H} + \vec{S}_1 \ . \tag{2.10}$$

The energy density W is the sum of the electromagnetic energy density $(1/8\pi)(E^2 + H^2)$ and the mechanical energy density W_1 associated with the oscilla-

tors:

$$W=(1/8\pi)(E^2+H^2)+W_1\ . \tag{2.11}$$

The third term in Eq. (2.9) represents the rate at which the oscillators dissipate energy due to the damping mechanism.

Two types of waves are of physical interest in isotropic solids: transverse waves $\vec{\nabla}\cdot\vec{P}=0$, and longitudinal waves $\vec{\nabla}\times\vec{P}=0$. The standard Poynting vector $(c/4\pi)\vec{E}\times\vec{H}$, and $-(4\pi v^2/\Omega_P^2)\vec{P}\times(\vec{\nabla}\times\vec{P})$, will be nonzero when the solutions are transverse, $\vec{k}\perp\vec{E}$. The vector $-(4\pi v^2/\Omega_P^2)\vec{P}(\vec{\nabla}\cdot\vec{P})$ defines the energy flux for a longitudinal wave, $\vec{k}\parallel\vec{E}$. As is evident, longitudinal solutions are very mechanicallike in character. The transverse solutions contribute both electromagnetic and mechanical components to the energy flux. This mixed character of the transverse solutions is more obvious in the present treatment because of the introduction of spatial dispersion: $v^2\neq 0$. These solutions have been called polaritons in the open literature because of their unusual dispersive character.[12] The term polariton has also been applied to optical phonons in which spatial dispersion is usually negligible: $v^2\approx 0$.[13] The energy flux when $v^2=0$ is carried by only the standard Poynting vector, but this is merely a formality. The energy density W even when $v^2=0$, has both electromagnetic and mechanical components.

Following Brillouin,[14] one may define an energy propagation velocity as follows:

$$v_E=\bar{S}/\bar{W}\ , \tag{2.12}$$

where $\bar{S}$ and $\bar{W}$ denote values of S and W averaged over a period. Two different average values are defined in Sec. IV. The functional form obtained for v_E will be more complicated when spatial dispersion is included, but as Loudon has recently stressed it should be a useful concept even when effects of damping are important. When damping is neglected, it can be shown that $v_E=d\omega/dk$, the group velocity.

III. QUANTUM FIELD THEORY

Many theoretical papers have attacked the problem of light-matter interactions in semiconductors.[15,16] The problem is concisely stated by Osaka *et al.* in Ref. 6 who elaborate on the method of quantum field theory described in Ref. 3. However, the treatment in Ref. 6 applies only in the extremely weak-damping limit and is difficult to generalize. The "polariton" field operators which were introduced to deal with the exciton-radiation problem turn out to be more of a hindrance than a help in solving the problem when damping is included.[2,6,7] The present treatment will be more straightforward; the problem will be worked out in terms of Green's functions of the fields without making any canonical transformations.

A quantum field-theoretical model is treated consisting of three boson fields—photon, exciton, and phonon—representing the electromagnetic, polarization, and damping fields, respectively. Units will be assumed in which $\hbar=1$ so that energy and frequency can be used interchangeably. A quantum of energy of a bare photon, exciton, and phonon is given by ck, $\omega_E(k)$, and $\omega_P(k)$, respectively. Annihilation operators for the photons, excitons, and phonons are denoted by $a(k)$, $b(k)$, and $c(k)$, respectively.

The exciton-radiation interaction is assumed to be given by[2]

$$H_{eR}=\left[\sum_{\vec{k}}\ \Omega_P\left(\frac{\omega_E(\vec{k})}{4ck}\right)^{1/2}\gamma(\vec{k})\varphi^\dagger(\vec{k})+\frac{\Omega_P^2}{4ck}\ \varphi^\dagger(\vec{k})\varphi(\vec{k})\right]\ , \tag{3.1}$$

where

$$\varphi(\vec{k})=a(\vec{k})+a^\dagger(-\vec{k})\ , \tag{3.2}$$

$$\gamma(\vec{k})=i[b(\vec{k})-b^\dagger(-\vec{k})]\ . \tag{3.3}$$

The exciton-lattice interaction is assumed to be given by[1]

$$H_{eL}=V^{-1/2}\sum_{\vec{q},\vec{k}}\Gamma_0(\vec{q})b^\dagger(\vec{k}+\vec{q})b(\vec{k})\psi(\vec{q})\ , \tag{3.4}$$

where

$$\psi(\vec{q})=i[c(\vec{q})-c^\dagger(-\vec{q})]\ . \tag{3.5}$$

Here V equals the volume of the medium over which periodic boundary conditions are applied. Eventually V will be allowed to go to infinity. The coupling parameter $\Gamma_0(\vec{q})$ in any realistic model will have a complicated functional dependence on $\vec{q}$. (For example, see Appendix B where the deformation-potential interaction is considered.)

In the present treatment the interaction H_{lR} between the photons and phonons is neglected. It will have a form similar to H_{eR} and make an analogous contribution to photon dispersion as does H_{eR}.[17] However, H_{lR} is not included because it will not introduce any new physics unless the energies $\omega_E(\vec{k})$ and $\omega_P(\vec{k})$ are approximately equal. It will be assumed throughout this paper that $\omega_E(\vec{k})\gg\omega_P(\vec{k})$.

The properties of the electromagnetic field are determined by the quantum analog of the classical retarded Green's function mentioned in Sec. II. It is defined by[3]

$$D_R(\vec{k},\omega)=-i2\pi k^{-1}\int_0^\infty e^{i\omega t}\,dt\,S_P \times\{e^{\beta\Omega-\beta H}[\tilde{\varphi}(\vec{k},t),\ \varphi^\dagger(\vec{k})]\}\ , \tag{3.6}$$

where $\beta=1/k_BT$ and

$$\tilde{\varphi}(\vec{k},t)=e^{iHt}\varphi(\vec{k})e^{-iHt}\ , \tag{3.7}$$

$$H=\sum_{\vec{k}}[ckA^{\dagger}(\vec{k})A(\vec{k})+\omega_E(\vec{k})B^{\dagger}(\vec{k})B(\vec{k}) + \omega_P(\vec{k})C^{\dagger}(\vec{k})C(\vec{k})] + H_{eL} + H_{eR} . \quad (3.8)$$

It is very difficult to calculate $D_R(\vec{k}, \omega)$ directly from Eq. (3.6). Therefore, causal "Matsubara" Green's functions will be introduced because these are simpler to calculate. Calculation of $D_R(\vec{k}, \omega)$ will be carried out later following the procedure outlined in Ref. 3.

The approach will be as follows: The exciton-phonon interaction H_{eL} will be treated as a perturbation in the Dyson sense on

$$H_0=\sum_{\vec{k}}[ckA^{\dagger}(\vec{k})A(\vec{k})+\omega_E(\vec{k})B^{\dagger}(\vec{k})B(\vec{k}) + \omega_P(\vec{k})C^{\dagger}(\vec{k})C(\vec{k})] + H_{eR} . \quad (3.9)$$

The following simplifying notation is adopted: $a_1(\vec{k})\equiv A(\vec{k})$, $a_2(\vec{k})\equiv B(\vec{k})$, $a_3(\vec{k})\equiv A^{\dagger}(-\vec{k})$, $a_4(\vec{k})\equiv B^{\dagger}(-\vec{k})$. The causal Green's functions are defined as follows:

$$\mathcal{G}_{\mu\nu}(\vec{k}, \tau)=-\langle T_\tau \tilde{a}_\mu(\vec{k}, \tau)\bar{a}_\nu(\vec{k}, 0)\rangle , \quad (3.10)$$

$$\mathfrak{D}(\vec{q}, \tau)=-\langle T_\tau \tilde{\psi}(\vec{q}, \tau)\bar{\psi}(\vec{q}, 0)\rangle , \quad (3.11)$$

where for a "Schrödinger" operator $\mathcal{F}(k)$,

$$\tilde{\mathcal{F}}(\vec{k}, \tau)=e^{H\tau}\mathcal{F}(\vec{k})e^{-H\tau} , \quad (3.12)$$

$$\bar{\tilde{\mathcal{F}}}(\vec{k}, \tau)=e^{H\tau}\mathcal{F}^{\dagger}(\vec{k})e^{-H\tau} , \quad (3.13)$$

$$\langle T_\tau \tilde{\mathcal{F}}(\vec{k}, \tau)\bar{\tilde{\mathcal{F}}}(\vec{k}, 0)\rangle = S_P\{e^{\beta\Omega-\beta H}T_\tau \tilde{\mathcal{F}}(\vec{k}, \tau)\bar{\tilde{\mathcal{F}}}(\vec{k}, 0)\} . \quad (3.14)$$

Here as in Eqs. (3.10) and (3.11), T_τ is the "imaginary" time-ordering operator which for Bosons has the following meaning:

$$\langle T_\tau \mathcal{F}(\vec{k}, \tau)\bar{\mathcal{F}}(\vec{k}, 0)\rangle = \begin{cases}\langle \tilde{\mathcal{F}}(\vec{k}, \tau)\bar{\tilde{\mathcal{F}}}(\vec{k}, 0)\rangle , & \tau>0\\ \langle \bar{\tilde{\mathcal{F}}}(\vec{k}, 0)\tilde{\mathcal{F}}(\vec{k}, \tau)\rangle , & \tau<0 .\end{cases} \quad (3.15)$$

Applying the operation $\partial/\partial\tau$ to $\mathcal{G}_{\mu\nu}(\vec{k}, \tau)$, one obtains after a straightforward calculation (summation over repeated indices is implied)

$$\frac{\partial}{\partial\tau}\mathcal{G}_{\mu\nu}(\vec{k}, \tau)=-C_{\mu\alpha}(\vec{k})\mathcal{G}_{\alpha\nu}(\vec{k}, \tau)-\bar{\delta}_{\mu\nu}\delta'(\tau) + V^{-1/2}\delta_{\mu 2}\sum_{\vec{q}}\Gamma_{(0)}(\vec{q})\langle T_\tau \tilde{a}_2(\vec{k}-\vec{q}, \tau)\tilde{\psi}(\vec{q}, \tau)\bar{\tilde{a}}_\nu(\vec{k}, 0)\rangle - V^{-1/2}\delta_{\mu 4}\sum_{\vec{q}}\Gamma_{(0)}(\vec{q})\langle T_\tau \tilde{a}_4(\vec{k}-\vec{q}, \tau)\tilde{\psi}(\vec{q}, \tau)\bar{\tilde{a}}_\nu(\vec{k}, 0)\rangle , \quad (3.16)$$

where

$$C_{\mu\nu}(\vec{k})=\begin{pmatrix} ck+A & iB & A & -iB\\ -iB & \omega_E(\vec{k}) & -iB & 0\\ -A & -iB & -ck-A & iB\\ -iB & 0 & -iB & -\omega_E(\vec{k})\end{pmatrix} , \quad (3.17a)$$

$$A=\Omega_P^2/2ck , \quad B=\Omega_P(\omega_E(\vec{k})/4ck)^{1/2} , \quad (3.17b)$$

$$\delta'(\tau)=\delta(\tau)+\delta(\tau\pm\beta)+\delta(\tau\pm 2\beta)+\cdots , \quad (3.18a)$$

$$\bar{\delta}_{\mu\nu}=\begin{pmatrix}1&0&0&0\\0&1&0&0\\0&0&-1&0\\0&0&0&-1\end{pmatrix} . \quad (3.18b)$$

One may recognize the first part of Eq. (3.16) as the equation of motion of an undamped polariton. The rest of Eq. (3.16) is responsible for the damping.

It is convenient to work in the four-dimensional momentum space:

$$(k_m)\equiv(\vec{k}, \omega_m) , \quad \omega_m=2\pi m k_B T \ \ (m \text{ is an integer}) .$$

Multiplying both sides of Eq. (3.16) by $e^{i\omega_m\tau}$ and integrating τ from 0 to β, one obtains Dyson's equation (see Appendix A for details):

$$[i\omega_m\delta_{\mu\alpha}-\tilde{C}_{\mu\alpha}(k_m)]\mathcal{G}_{\alpha\nu}(k_m)=\bar{\delta}_{\mu\nu} , \quad (3.19)$$

where

$$\tilde{C}_{\mu\nu}(k_m)=C_{\mu\nu}(\vec{k})+\Sigma_{\mu\nu}(k_m) , \quad (3.20a)$$

$$\mathcal{G}_{\mu\nu}(k_m)=\int_0^\beta d\tau\,\mathcal{G}_{\mu\nu}(\vec{k}, \tau)e^{i\omega_m\tau} , \quad (3.20b)$$

$$\mathfrak{D}(q_n)=\int_0^\beta d\tau\,\mathfrak{D}(\vec{q}, \tau)e^{i\omega_m\tau} . \quad (3.20c)$$

In Eq. (3.20a) $\Sigma_{\mu\nu}(k_m)$ is the "self-energy" tensor and is given by

$$\Sigma_{\mu\nu}(k_m)=\begin{cases}\Sigma_{rs}(k_m) , & r \text{ and } s=2, 4\\ 0 & \text{otherwise} ,\end{cases} \quad (3.21a)$$

where

$$\Sigma_{rs}(k_m)=\mp k_B T\sum_n\sum_{r'=2,4}\int\frac{d\vec{q}}{(2\pi)^3}\,\Gamma_0(\vec{q})\mathfrak{D}(q_n) \times\mathcal{G}_{rr'}(k_m-q_n)\Gamma_{r's}(-q_n, k_m) . \quad (3.21b)$$

The minus sign applies for $r=2$; the plus sign for $r=4$. The "vertex" tensor Γ_{rs} is given by the infinite heirarchy of Feynman diagrams shown in Appendix A.

Equations (3.19)–(3.21) are completely general.

However, explicit calculation of the self-energy tensor is possible only in some very special cases. (One of these cases is discussed in Appendix B.) Nevertheless, a great deal can be learned from the above equations.

Only four of the 16 tensor components of $\Sigma_{\mu\nu}$ are nonzero. The remaining four are related in the following way:

$$\Sigma_{22}(\vec{k}, \omega_m) = -\Sigma_{44}(\vec{k}, -\omega_m) \equiv \gamma_1(\vec{k}, i\omega_m) ,$$
$$\Sigma_{24}(\vec{k}, \omega_m) = -\Sigma_{42}(\vec{k}, -\omega_m) \equiv \gamma_2(\vec{k}, i\omega_m) . \quad (3.22)$$

These relations can be proven by inspection of Eqs. (3.10) and (3.21b) and the diagrams defining the vertex tensor in Appendix A. Analytic continuation away from real values of ω_m is being anticipated by writing these as functions of $\vec{k}$ and $i\omega_m$. Letting

$$A_{\mu\nu}(\vec{k}, i\omega_m) \equiv i\omega_m \delta_{\mu\nu} - \tilde{C}_{\mu\nu}(\vec{k}, \omega_m) , \quad (3.23)$$

one obtains

$$\mathcal{G}_{\mu\nu}(\vec{k}, \omega_m) = A^{-1}_{\mu\alpha}(\vec{k}, i\omega_m)\bar{\delta}_{\alpha\nu} . \quad (3.24)$$

One can now formally calculate $D_R(\vec{k}, \omega)$ defined by Eq. (3.6). Letting

$$G^{(R)}_{\mu\nu}(\vec{k}, \omega) = -i\int_0^\infty dt\, e^{i\omega t} S_P \{e^{\beta\Omega - \beta H}[\tilde{a}_\mu(\vec{k}, t), a^\dagger_\nu(\vec{k})]\} \quad (3.25)$$

and using the analytic properties of the Green's functions (see, for example, Ref. 3, p. 145), one can show that

$$G^{(R)}_{\mu\nu}(\vec{k}, i\omega_m) = A^{-1}_{\mu\alpha}(\vec{k}, i\omega_m)\bar{\delta}_{\alpha\nu}, \quad \omega_m > 0 . \quad (3.26)$$

If $A^{-1}_{\mu\alpha}(\vec{k}, \omega)$ is an analytic function of ω in the upper-half frequency plane (one has to examine this assumption for each exciton-lattice interaction studied), it then follows from uniqueness theorems about analytic functions that

$$G^{(R)}_{\mu\nu}(\vec{k}, \omega) \equiv A^{-1}_{\mu\alpha}(\vec{k}, \omega)\bar{\delta}_{\alpha\nu} . \quad (3.27)$$

Referring to Eqs. (3.6), (3.25), and (3.27), one obtains after a tedious but straightforward calculation:

$$D_R(\vec{k}, \omega) = 2\pi k^{-1}[A^{-1}_{11}(\vec{k}, \omega) - A^{-1}_{13}(\vec{k}, \omega) + A^{-1}_{31}(\vec{k}, \omega) - A^{-1}_{33}(\vec{k}, \omega)]$$
$$= 4\pi c/[\omega^2 \tilde{\epsilon}(\vec{k}, \omega) - c^2k^2] , \quad (3.28)$$

$$\tilde{\epsilon}(k, \omega) = 1 + \frac{\tilde{\Omega}^2_P(\vec{k}, \omega)}{\omega^2_E(\vec{k}) - \omega^2 - i\omega\tilde{\Gamma}(\vec{k}, \omega)} , \quad (3.29)$$

where

$$\omega^2\tilde{\Omega}^2_P(\vec{k}, \omega) = \Omega^2_P\{\omega^2 - \omega[\gamma_1(\vec{k}, \omega) - \gamma_1(\vec{k}, -\omega)]$$
$$- \tfrac{1}{2}\omega_E(\vec{k})[\gamma_1(\vec{k}, \omega) + \gamma_1(\vec{k}, -\omega) - \gamma_2(\vec{k}, \omega) - \gamma_2(\vec{k}, -\omega)]$$
$$- \gamma_1(\vec{k}, \omega)\gamma_1(\vec{k}, -\omega) + \gamma_2(\vec{k}, \omega)\gamma_2(\vec{k}, -\omega)\} , \quad (3.30)$$

$$-i\omega\tilde{\Gamma}(\vec{k}, \omega) = \omega[\gamma_1(\vec{k}, \omega) - \gamma_1(\vec{k}, -\omega)] + \omega_E(\vec{k})[\gamma_1(\vec{k}, \omega) + \gamma_1(\vec{k}, -\omega)] + \gamma_1(\vec{k}, \omega)\gamma_1(\vec{k}, -\omega) - \gamma_2(\vec{k}, \omega)\gamma_2(\vec{k}, -\omega) . \quad (3.31)$$

The retarded Green's function has been written in this form in order to compare it with the classical Green's function given in Eq. (2.3). Here $\tilde{\epsilon}(\vec{k}, \omega)$, is the quantum analog of the classical dielectric constant $\epsilon(\vec{k}, \omega)$, $\tilde{\Omega}_P(\vec{k}, \omega)$ is the quantum analog of Ω_p, and the $\mathrm{Re}\tilde{\Gamma}(\vec{k}, \omega)$ is the quantum analog of the damping parameter Γ. In addition one may incorporate the rest of the term, $-i\omega\tilde{\Gamma}(\vec{k}, \omega)$, into $\omega^2_E(\vec{k})$ by letting

$$\tilde{\omega}^2_E(\vec{k}, \omega) = \omega^2_E(\vec{k}) + \omega\,\mathrm{Im}\,\tilde{\Gamma}(\vec{k}, \omega) . \quad (3.32)$$

One can see that a close connection exists between the quantum and classical theories.

This connection between the classical and quantum theories should be very useful. Although $\tilde{\omega}_E(\vec{k}, \omega)$, $\tilde{\Omega}_P(\vec{k}, \omega)$, and $\mathrm{Re}\tilde{\Gamma}(\vec{k}, \omega)$ can be calculated from first principles in only special cases, even less can be said, for that matter, about their classical counterparts. Valuable information about all three of these functions can possibly be learned by substituting trial functions for them into Eq. (3.29) and then calculating optical parameters such as indices of refraction, extinction coefficients, etc. One can then compare these theoretical results with actual experimental data.

The classical theory is a special case of the quantum theory. It is given by

$$\tilde{\omega}^2_E(\vec{k}, \omega) = \omega^2_0 + v^2k^2 ,$$
$$\mathrm{Re}\tilde{\Gamma}(\vec{k}, \omega) = \Gamma , \quad \tilde{\Omega}_P(\vec{k}, \omega) = \Omega_P . \quad (3.33)$$

This special case is nontrivial and will be discussed further in Sec. V.

IV. BASIC SOLUTIONS OF PROBLEM

Several papers have talked about dispersion relations which include spatial dispersion and damping, but it is not clear what physical situation was being depicted in each case.[18,19] An attempt is now made to clear up this problem by deriving dispersion relations for two clearly defined physical situations.

In an isotropic medium the vector components of the Fourier transforms of the vector potential, retarded Green's function, and "externally" impressed current density are related by

$$A_i(\vec{k},\omega)=D_R(\vec{k},\omega)\left(\delta_{il}-\frac{c^2k_ik_l}{\omega^2\epsilon(\vec{k},\omega)}\right)\times j_l^{\rm imp}(\vec{k},\omega)\ . \tag{4.1}$$

Here a gauge is employed where the scalar potential vanishes.[20] This equation is very useful, for it determines the vector potential in terms of impressed current sources which are completely arbitrary. Two different current distributions will be considered. One has a well-defined momentum; the other has a well-defined energy.

A. Quasiparticle Solutions

A quasiparticle is a quantum of excitation which is introduced into the system at some time $t=0$ with momentum $\vec{k}$ and is allowed to exist without further influence from any impressed currents. This physical situation is created by the following current distribution:

$$\vec{j}_{\rm imp}(\vec{x},t)\propto\delta(t)e^{i\vec{k}\cdot\vec{x}}\ ,\qquad \vec{j}_{\rm imp}(\vec{k}',\omega)\propto\delta(\vec{k}'-\vec{k})\ . \tag{4.2}$$

Here $\vec{k}$ is an arbitrary wave vector which defines the momentum of the quasiparticle. Two cases will be treated: $\vec{k}\perp\vec{j}_{\rm imp}$ (transverse case) and $\vec{k}\parallel\vec{j}_{\rm imp}$ (longitudinal case). Substituting Eq. (4.2) into (4.1), one obtains in the transverse case

$$\vec{A}_T(\vec{x},t)\propto e^{i\vec{k}\cdot\vec{x}}\int_{-\infty}^{\infty}d\omega\,e^{-i\omega t}D_R(\vec{k},\omega)\ . \tag{4.3}$$

For times $t>0$, one may close the counter by adding the half-circle in the lower-half ω plane without affecting the value of the above integral (see Fig. 1). One thus obtains

$$\vec{A}_T(\vec{x},t)=e^{i\vec{k}\cdot\vec{x}}\sum_\tau\vec{A}_\tau(\vec{k})\,e^{-i\tilde{\omega}_\tau(\vec{k})t}\ , \tag{4.4}$$

where

$$\vec{A}_\tau(\vec{k})\propto[R_{es}D_R(\vec{k},\omega)]_{\omega=\tilde{\omega}_\tau(k)}\ . \tag{4.5}$$

Here $R_{es}D_R(\vec{k},\omega)$ denotes the residue of $D_R(\vec{k},\omega)$ at one of its poles in the lower half of the frequency plane, $\tilde{\omega}_\tau(\vec{k})=\omega_\tau'(\vec{k})-i\omega_\tau''(\vec{k})$. The wave vector $\vec{k}$ is completely arbitrary. This establishes the following dispersion relation:

$$\tilde{\omega}_\tau^2(\vec{k})\tilde{\epsilon}[\vec{k},\tilde{\omega}_\tau(\vec{k})]=c^2k^2\ . \tag{4.6}$$

In the longitudinal case ($\vec{k}\parallel\vec{j}_{\rm imp}$), one obtains from Eqs. (4.1) and (4.2)

$$\vec{A}_L(\vec{x},t)\propto e^{i\vec{k}\cdot\vec{x}}\int_{-\infty+i\delta}^{\infty+i\delta}d\omega\,\frac{e^{-i\omega t}}{\omega^2\tilde{\epsilon}(\vec{k},\omega)}\ , \tag{4.7}$$

where $\delta\to0^+$. This contour can be closed in the same manner so that one obtains

$$\vec{A}_L(\vec{x},t)=e^{i\vec{k}\cdot\vec{x}}\sum_\lambda A_\lambda(\vec{k})e^{-i\tilde{\omega}_\lambda(\vec{k})t}\ , \tag{4.8}$$

where

$$\vec{A}_\lambda(\vec{k})\propto\left[R_{es}\left(\frac{1}{\omega^2\tilde{\epsilon}(\vec{k},\omega)}\right)\right]_{\omega=\tilde{\omega}_\lambda(\vec{k})}\ . \tag{4.9}$$

Here $\tilde{\omega}_\lambda(\vec{k})=\omega_\lambda'(\vec{k})-i\omega_\lambda''(\vec{k})$ is the value of ω at a pole in $[\omega^{-2}\tilde{\epsilon}^{-1}(\vec{k},\omega)]$. This establishes the dispersion relation

$$\tilde{\epsilon}[\vec{k},\tilde{\omega}_\lambda(\vec{k})]=0\ . \tag{4.10}$$

B. Forced-Harmonic Solutions

Another useful impressed current distribution is that given by

$$\vec{j}_{\rm imp}(\vec{x},t)\propto\delta(x_1)e^{-i\omega t}\ ,\qquad j_{\rm imp}(\vec{k},\omega')\propto\delta(\omega-\omega')\delta(k_2)\delta(k_3)\ . \tag{4.11}$$

This corresponds to the case in which the medium is forced to oscillate at an arbitrary frequency, ω.

From Eq. (4.1), a transverse solution satisfies ($\vec{k}_1\perp j_{\rm imp}$)

$$\vec{A}_T(\vec{x},t)\propto e^{-i\omega t}\int_{-\infty}^{\infty}dk_1\,e^{ik_1x_1}D_R(\vec{k}_1,\omega)\ ,\qquad \vec{k}_1=\hat{e}_1k_1\ . \tag{4.12}$$

If $x_1>0$, one can close this contour by adding the semicircle in the upper half k_1 (see Fig. 2), obtain-

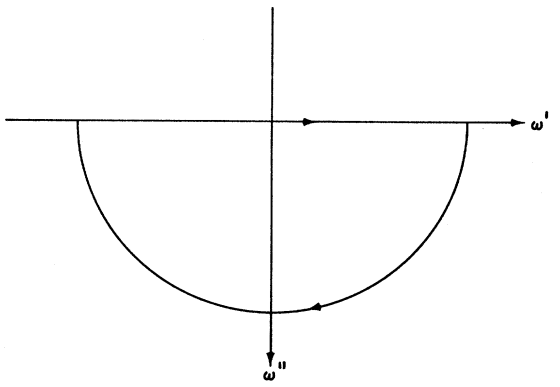

FIG. 1. Contour integral applicable to quasiparticle solutions.

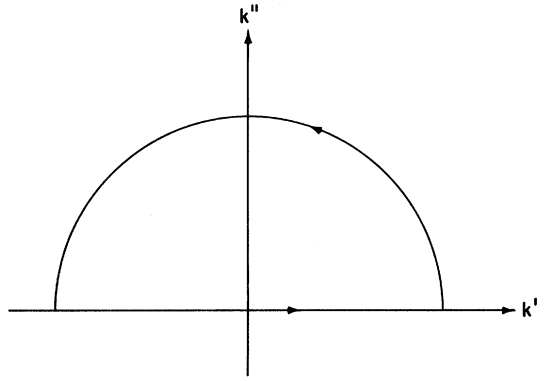

FIG. 2. Contour integral applicable to forced-harmonic solutions.

ing

$$\vec{A}_T(\vec{x}, t) = e^{-i\omega t} \sum_\tau \vec{A}_\tau(\omega) e^{i\tilde{k}_\tau(\omega) x_1} , \quad (4.13)$$

where

$$\vec{A}_\tau(\omega) \propto [R_{es} D_R(\vec{k}_1, \omega)]_{k_1 = \tilde{k}_\tau(\omega)} . \quad (4.14)$$

Here $\tilde{k}_\tau(\omega) = k'_\tau(\omega) + ik''_\tau(\omega)$ is a solution of the dispersion relation

$$\omega^2 \tilde{\epsilon}[\tilde{k}_\tau(\omega), \omega] = c^2 \tilde{k}_\tau^2(\omega) . \quad (4.15)$$

A corresponding longitudinal solution satisfies

$$\vec{A}_L(x, t) = e^{-i\omega t} \sum_\lambda \vec{A}_\lambda(\omega) e^{i\tilde{k}_\lambda(\omega) x_1} , \quad (4.16)$$

$$\vec{A}_\lambda(\omega) \propto (1/\omega^2) [R_{es} \tilde{\epsilon}^{-1}(k, \omega)]_{k = \tilde{k}_\lambda(\omega)} . \quad (4.17)$$

Here $\tilde{k}_\lambda(\pi) = k'_\lambda(\omega) + ik''_\lambda(\omega)$ satisfies the dispersion relation

$$\tilde{\epsilon}[\tilde{k}_\lambda(\omega), \omega] = 0 . \quad (4.18)$$

C. Physical Meaning of Different Solutions

A quasiparticle solution has a well-defined momentum $\vec{k}$. The real part of the frequencies in Eqs. (4.4) and (4.8) ($\omega'_\tau(\vec{k})$, $\tau = \tau, \lambda$) equals the quasiparticle energy. These solutions damp out expononentially in time; the square of the vector-potential amplitude has a reciprocal lifetime given by

$$T_\tau^{-1}(\vec{k}) = 2\omega''_\tau(\vec{k}) , \quad \tau = \tau, \lambda \quad (4.19)$$

where $\omega''_\tau(\vec{k})$ is the imaginary part of the energy. An optical-phonon polariton created in a Raman scattering experiment is a quasiparticle (see Sec. V).

The forced-harmonic solutions have a well-defined energy. As is evident from Eqs. (4.13) and (4.16), these solutions damp out exponentially in space; the square of the vector-potential amplitude has an effective absorption coefficient given by

$$\alpha_\tau(\omega) = 2k''_\tau(\omega) , \quad \tau = \tau, \lambda . \quad (4.20)$$

Here $k''_\tau(\omega)$ is the imaginary part of the wave number in Eq. (4.15) or (4.18). A monochromatic light wave entering a solid with frequency ω will propagate through the solid as a superposition of one or more forced-harmonic solutions with complex wave vectors and the same real frequency ω.

D. Field Vectors

The Fourier transforms of the electromagnetic and polarization field vectors in the quantum theory are related to the Fourier transform of the vector potential in the same manner as in the classical theory[21]:

$$\vec{E}(\vec{k}, \omega) = i\omega c^{-1} \vec{A}(\vec{k}, \omega) , \quad (4.21)$$

$$\vec{H}(\vec{k}, \omega) = i\vec{k} \times \vec{A}(\vec{k}, \omega) , \quad (4.22)$$

$$4\pi \vec{P}(\vec{k}, \omega) = [\tilde{\epsilon}(\vec{k}, \omega) - 1] \vec{E}(\vec{k}, \omega) , \quad (4.23)$$

where $\vec{A}(\vec{k}, \omega)$ is given by Eq. (4.1). The electromagnetic and polarization field vectors $\vec{E}(\vec{x}, t)$ and $\vec{P}(\vec{x}, t)$ can be obtained by taking inverse Fourier transforms, closing the contours in the $\vec{k}$ and ω planes in the same way as was done earlier in computing $\vec{A}(\vec{x}, t)$.

For the field vectors of transverse quasiparticle solution, one obtains

$$\vec{E}_\tau(\vec{x}, t) = i\omega_\tau(\vec{k}) c^{-1} \vec{A}_\tau(\vec{x}, t) , \quad (4.24)$$

$$\vec{H}_\tau(\vec{x}, t) = i\vec{k} \times \vec{A}_\tau(\vec{x}, t) , \quad (4.25)$$

$$4\pi \vec{P}_\tau(\vec{x}, t) = \{\tilde{\epsilon}[\vec{k}, \tilde{\omega}_\tau(\vec{k})] - 1\} \vec{E}_\tau(\vec{x}, t) , \quad (4.26)$$

where

$$\vec{A}_\tau(\vec{x}, t) = \vec{A}_\tau(\vec{k}) e^{i[\vec{k} \cdot \vec{x} - \tilde{\omega}_\tau(\vec{k}) t]} . \quad (4.27)$$

The field vectors of a corresponding longitudinal solution are given by replacing τ by λ and $\tilde{\epsilon}$ by 0 in the above expressions.

The field vectors of the forced-harmonic solutions can be obtained from the above solutions by replacing $\tilde{\omega}_\tau(k)$ by ω and k by $\tilde{k}_\tau(\omega)$, $\tau = \tau, \lambda$.

E. Energy Velocity

One can determine how the electromagnetic and polarization fields propagate through a solid by constructing wave packets from linear combinations of the above solutions.[16] However, one would have to employ numerical methods before anything meaningful could be obtained when damping and spatial dispersion were included. Analytical expressions can be obtained by employing the energy-velocity concept derived in Sec. II.

Many light-matter experiments involve only a small range of energies and $\vec{k}$ vectors near the center of the fundamental Brillouin zone. Here the classical model should be approximately satisfied:

$$\tilde{\omega}^2(k, \omega) \approx \omega_0^2 + v^2 k^2 , \quad \tilde{\Omega}_P(k, \omega) \approx \Omega_P ,$$

$$\mathrm{Re}\tilde{\Gamma}(k,\omega)\approx\Gamma\ .$$

Probably the poorest of these approximations is that of neglecting frequency and/or wave-vector dependence in the $\mathrm{Re}\tilde{\Gamma}(k,\omega)$.[5-7]

The energy velocity defined in Eq. (2.12) depends only implicitly on Γ through a dispersion relation. Poynting's theorem does not appear to be affected by including frequency or wave-vector dependence in Γ. With Γ assumed to be an arbitrary function of k and ω, some potentially useful expressions will now be derived that relate v_E to ω_0, v, Ω_P, and Γ.

The energy velocity equals the ratio $\bar{S}/\bar{W}$ where the bars denote average values. In the quasiparticle case it is natural to define this average in space by integrating the instantaneous functions $S(x,t)$ and $W(x,t)$ given by Eqs. (2.10) and (2.11) over a period $\lambda=2\pi/k$ in x:

$$\bar{S}(t)=\frac{1}{\lambda}\int_{x_1}^{x_1+\lambda}S(x,t)\,dx_1\ ,$$
$$\bar{W}(t)=\frac{1}{\lambda}\int_{x_1}^{x_1+\lambda}W(x,t)\,dx_1\ . \tag{4.28}$$

Here x_1 is along the direction which the wave vector is directed. In the forced-harmonic case it is natural to define this average with respect to time t:

$$\bar{S}(x)=\frac{1}{T}\int_{t}^{t+T}S(x,t)\,dt\ ,$$
$$\bar{W}(x)=\frac{1}{T}\int_{t}^{t+T}W(x,t)\,dt\ , \tag{4.29}$$

where $T=2\pi/\omega$. The energy velocity of each of the above solutions will be independent of x and t; it will be a function only of k in the quasiparticle case and ω in the forced-harmonic case.

The calculation of the energy velocity is now straightforward to carry out so that only the final results will be presented here. The results below are for the quasiparticle and forced-harmonic solutions derived earlier. The quasiparticle energy velocities are denoted by $v_\tau(k)$; the forced-harmonic energy velocities are denoted by $v_\tau(\omega)$, $\tau=\tau,\ \lambda$:

$$v_\tau(k)=k^{-1}\Gamma^{-1}\left[\Gamma\omega_\tau'-2\omega_\tau'\omega_\tau''+2v^2k^2\omega_\tau'\omega_\tau''(\omega_\tau'^2+\omega_\tau''^2)^{-1}\right]\ , \tag{4.30}$$

$$v_\lambda(k)=v^2k\,(\Omega_P^2+\omega_0^2+v^2k^2-\tfrac{1}{4}\Gamma^2)^{1/2}\,(\Omega_P^2+\omega_0^2+v^2k^2)^{-1}\ , \tag{4.31}$$

$$v_\tau(\omega)=c\Gamma[n_\tau\Gamma+2\kappa_\tau\omega+2\beta^2\omega\kappa_\tau(\kappa_\tau^2-n_\tau^2)]^{-1}\ , \tag{4.32}$$

$$v_\lambda(\omega)=v^2k_\lambda'\omega(\Omega_P^2+\omega_0^2+v^2k_\lambda'^2)^{-1}\ , \tag{4.33a}$$

$$2v^2k_\lambda'=\omega^2-\omega_0^2-\Omega_P^2+[(\omega^2-\omega_0^2-\Omega_P^2)^2+\Gamma^2\omega^2]^{1/2}\ , \tag{4.33b}$$

where

$$n_\tau=ck_\tau'/\omega\ ,\quad \kappa_\tau=ck_\tau''/\omega\ ,\quad \beta=v/c\ . \tag{4.34}$$

The above expressions for the transverse energy velocities cannot be simplified any further. The frequencies ω_τ' and ω_τ'' and wave numbers k_τ' and k_τ'' have to be determined numerically from Eqs. (4.6) and (4.15), respectively. When β^2 is set equal to zero, Eq. (4.32) reduces to the result recently derived by Loudon.[11] Numerical results based on Eqs. (4.30) and (4.32) will be given in Sec. V.

V. RESULTS

A. Dispersion Relations

The dispersion relations plotted in Figs. 3 and 4 are for the optical model in which the following assumptions have been made: $\tilde{\Omega}_P(k,\omega)=\Omega_P$, $\tilde{\omega}_E^2(k,\omega)=\omega_0^2+v^2k^2$, and $\mathrm{Re}\tilde{\Gamma}(k,\omega)=\Gamma$. The values of ω_0, v, and Ω_P are listed in the figure caption. These results demonstrate the significant difference between the quasiparticle and forced-harmonic solutions when damping is included.

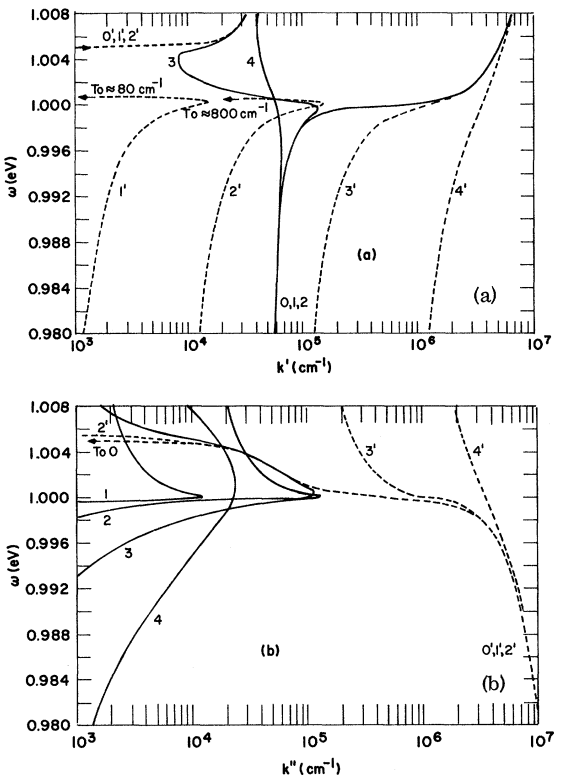

FIG. 3. Transverse forced-harmonic solutions of the classical model with $\Omega_P=0.1$ eV, $\omega_0=1.0$ eV, $\beta=v/c=10^{-3}$: (0) and (0′), $\Gamma=0$ eV; (1) and (1′), $\Gamma=10^{-5}$ eV; (2) and (2′), $\Gamma=10^{-4}$ eV; (3) and (3′), $\Gamma=10^{-3}$ eV; (4) and (4′), $\Gamma=10^{-2}$ eV.

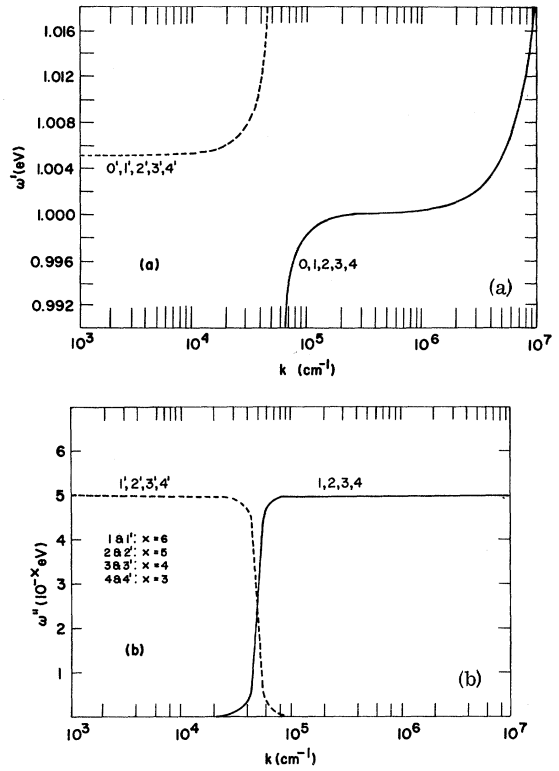

FIG. 4. Transverse quasiparticle solutions of the classical model with same values assigned to the parameters as in Fig. 3.

The forced-harmonic solutions shown in Fig. 3 change character dramatically as Γ passes through the following value:

$$\Gamma = 2(v/c)\Omega_P \ . \tag{5.1}$$

When Γ is smaller than $2(v/c)\Omega_P$, the dispersion relations are similar in character to the form taken when damping is neglected. "Polariton" effects will have their usual meaning here. However, when Γ exceeds $2(v/c)\Omega_P$, the dispersion curves continue to "repel" each other, but take on a more "classical" character. With increasing Γ, one solution becomes more and more photonlike while the other becomes more and more excitonlike. As Γ continues to grow, the absorption coefficient $2k''(\omega)$ of the excitonlike solution continues to increase but the absorption coefficient of the photonlike solution starts decreasing.

These results are not unreasonable. If the exciton field is strongly damped out by phonons, the effect of the exciton-photon coupling could conceivably become less important so that the normal polariton effects are not apparent. Hopfield suggested this possibility in qualitative terms several years ago.[22] Equation (5.1) puts this in quantitative terms. The energy velocities plotted against ω in

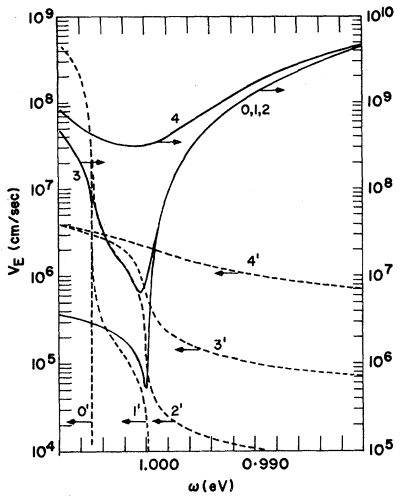

FIG. 5. Energy velocities for forced-harmonic solutions shown in Fig. 3.

Fig. 5 give a vivid indication of this.

Damping has appreciably less effect on the quasiparticle solutions as can be seen in Fig. 4. Here the dispersion relations retain their undamped character for all values of Γ. The energy velocities plotted against k in Fig. 6 illustrate the photonlike and matterlike character of the quasiparticle solutions.

Figure 7 illustrates another model which could be of some importance in semiconductors especially near liquid-helium temperature.[6,7] For the same set of parameters ω_0, v, and Ω_P, some frequency dependence in Γ has been added. Only the forced-

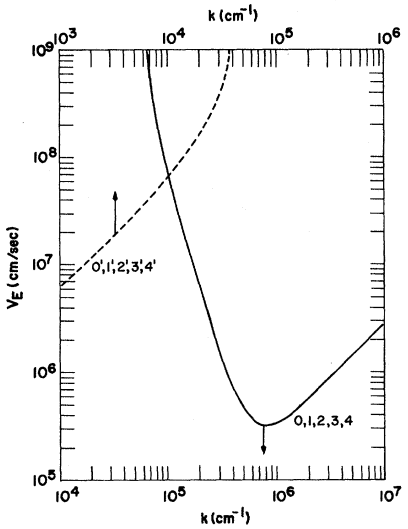

FIG. 6. Energy velocities for quasiparticle solutions shown in Fig. 4.

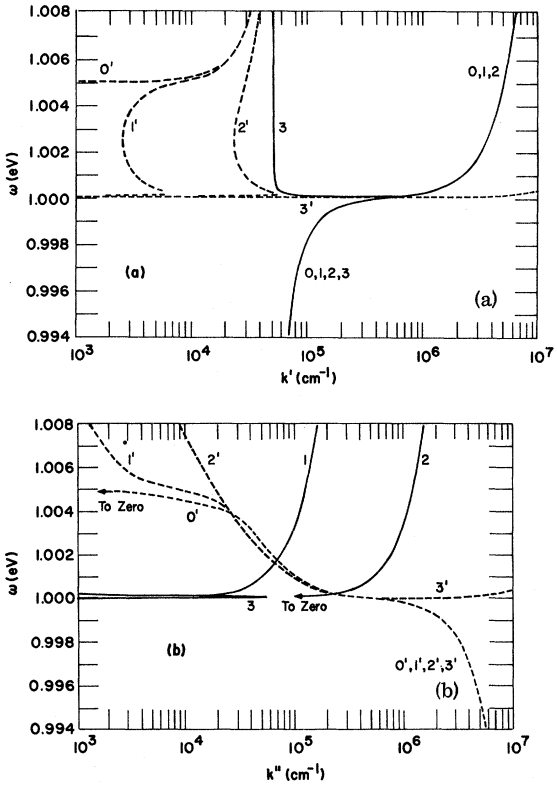

FIG. 7. Transverse forced-harmonic solutions of the model described in text with $\Omega_p = 0.1$ eV, $\omega_0 = 1.0$ eV, $\beta = v/c = 10^{-3}$: (0) and (0′), $\alpha = 0$ eV; (1) and (1′), $\alpha = 10^{-1}$ eV; (2) and (2′), $\alpha = 1$ eV; (3) and (3′), $\alpha = 300$ eV.

harmonic case is considered, it being assumed that

$$\Gamma(k,\omega) = \begin{cases} \gamma(\omega/\omega_0 - 1), & \omega > \omega_0 \\ 0, & \omega \leq \omega_0 \end{cases} . \tag{5.2}$$

The right-hand side of Eq. (5.2) has a form similar to that derived using the deformation-potential interaction at $T = 0\,^\circ$K (see Appendix B). The solutions in this model are similar in character to the earlier model. However, dispersion curves 3 and 3′ are observed to cross, which is unusual. "Polariton" effects are clearly evident in solutions 0, 1, and 2 where effects of damping are "small." The solutions take on a more classical character when γ exceeds the value

$$\gamma = 2(c/v)\Omega_P , \tag{5.3}$$

as can be seen from an inspection of solutions 3 and 3′ in Fig. 7.

B. Phonon Polaritons

Benson and Mills[23] have recently raised a question as to why Raman scattering data from optical phonons[13] fits a theoretical dispersion relation which neglects damping, but does not fit one which includes damping.[24] This experiment is shown diagrammatically in Fig. 8. A light beam indicated by the dashed line is scattered with the simultaneous creation of a TO phonon indicated by the solid line. (The TO phonon is the "polariton" in this experiment, i.e., it is analogous to the exciton. The anharmonic phonon-phonon coupling is responsible for the damping in a manner analogous to H_{eL}.[17,23]) The crystal is transparent at the energies of the incident and scattered photon, but the TO phonon is strongly damped out by phonon-phonon interactions.

Puthoff *et al.*[24] applied a dispersion relation equivalent to Eq. (4.15) which is satisfied by an energy conserving phonon-polariton solution. They assumed energy of the created phonon polariton was real and that its momentum was complex. However, damping in this experiment gives rise to a nonvanishing linewidth in the observed energy of the scattered photon caused by the finite lifetime of the created phonon due to phonon-phonon coupling. Thus energy is not conserved! However, it is reasonable to expect this light scattering experiment describes a momentum-conserving event within the crystal at the moment the photon is scattered and the phonon is created. If momentum is conserved,

$$\vec{k} = \vec{k}_I - \vec{k}_S , \tag{5.4}$$

where $\vec{k}_I$ and $\vec{k}_S$ are the momenta of the incident and scattered photons, respectively, inside the crystal and $\vec{k}$ is the momentum of the created phonon. In this case the phonon is expected to be a quasiparticle solution of Eq. (4.6) where $\vec{k}$ is real and given by Eq. (5.4) and ω is complex and equal to $\omega_\tau'(\vec{k}) - i\omega_\tau''(\vec{k})$. This solution decays exponentially in time with an effective lifetime T_τ satisfying Eq. (4.19). If T_τ^{-1} is small compared to $\omega_\tau'(k)$, then the linewidth of the scattered photon will be small compared to $\omega_\tau'(\vec{k})$ and the energies, ω_I and ω_S, of the incoming and outgoing photons should be related to $\omega_\tau'(\vec{k})$ by

$$\omega_\tau'(\vec{k}) = \omega_I - \omega_S . \tag{5.5}$$

Here $\omega_\tau'(k)$ is also related to k, theoretically, through the dispersion relation [Eq. (4.6)].

The experimental parameters measured in the

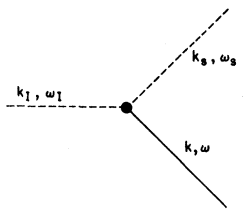

FIG. 8. Phonon-polariton scattering geometry: (k_I, ω_I) and (k_S, ω_S) denote momentum four vectors of the incident and scattered photon and (k, ω) denotes momentum four vector of the created phonon.

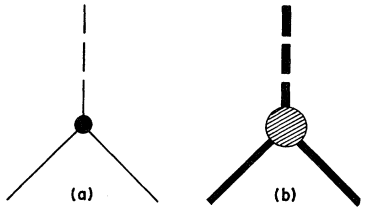

FIG. 9. Feynman diagrams associated with (a) Eq. (A6) and (b) Eq. (A7).

experiment are k_I, ω_I, k_S, and ω_S. Thus, from Eqs. (5.4) and (5.5) one can determine $\vec{k}$ and $\omega_\tau'(\vec{k})$ from experiment and compare them with theory. The experimental results have been found in all cases to date to be in close agreement with theoretical results which neglect damping.[23] The quasiparticle dispersion relations are in close agreement with these as long as $\Omega_P > 2\Gamma$. This latter condition is well satisfied in $LiNbO_3$. From Ref. 24, $\Omega_P \approx 10^4$ cm^{-1} and $\Gamma \approx 34$ cm^{-1} for the 628-cm^{-1} line in $LiNbO_3$.

VI. CONCLUSION

The quantum theory of a basic light-matter interaction has been treated quantum field theoretically. The classical theory has been shown to be a special case of the quantum theory. Where this special case applies, an energy velocity has been defined and calculated without recourse to wave packets.

Two basic field solutions are defined, called quasiparticle and forced-harmonic solutions, which have a well-defined momentum and energy, respectively. They coincide in dispersive character when damping is neglected, but possess significantly different dispersive characteristics when damping is included.

ACKNOWLEDGMENTS

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APPENDIX A: DERIVATION OF EQ. (3.19)

The type of term difficult to treat in deriving Eq. (3.19) is that given by

$$P_{r\nu}(\vec{k},\vec{q};\omega_m) = \int_0^\beta d\tau \langle T_\tau \tilde{a}_r(\vec{k}-\vec{q},\tau) \times \tilde{\psi}(\vec{q},\tau)\bar{\tilde{a}}_\nu(\vec{k},0)\rangle e^{i\omega_m\tau} . \quad (A1)$$

This term is essentially equivalent to that encountered in the electron-phonon problem (see, for example, Ref. 3, p. 136. The only difference is that the exciton is a boson instead of a fermion. To evaluate the above expression, one, therefore, introduces the interaction representation defined by

$$\mathcal{F}(\tau) = e^{H_0\tau}\mathcal{F}e^{-H_0\tau} \quad (A2)$$

and the theorem (see Ref. 3, p. 104)

$$\langle T_\tau \tilde{a}_r(\vec{k}-\vec{q},\tau)\tilde{\psi}(\vec{q},\tau)\bar{\tilde{a}}_\nu(\vec{k},0)\rangle = \langle T_\tau a_r(\vec{k}-\vec{q},\tau)\psi(\vec{q},\tau)\bar{a}_\nu(\vec{k},0)S(\beta)\rangle_0 \langle S(\beta)\rangle_0^{-1} . \quad (A3)$$

Here

$$S(\beta) = T_\tau \exp[-\int_0^\beta H_{eL}(\tau')\,d\tau'] \quad (A4)$$

and

$$\langle \cdots \rangle_0 \equiv S_P\{\exp[\beta(\Omega_0 - H_0)]\cdots\} . \quad (A5)$$

Expanding the numerator and denominator on the right-hand side of Eq. (A3) in a power series in $H_{eL}(\tau')$, one can associate with the corresponding expansions in Eq. (A1) the same set of Feynman diagrams as is done in the electron-phonon problem. The simplest of these occurs in first order and is shown in Fig. 9(a). It corresponds to the term

$$P_{r\nu}(\vec{k},\vec{q};\omega_m) = \Gamma_0^*(\vec{q})k_BT\sum_{\omega_n}\sum_{s=2,4}\mathcal{G}_{rs}^{(0)}(k_m-q_n) \times \mathcal{G}_{s\nu}^{(0)}(k_m)\mathfrak{D}^{(0)}(q_n) , \quad (A6)$$

where $\mathcal{G}_{\mu\nu}^{(0)}(k_m)$ and $\mathfrak{D}^{(0)}(q_n)$ are Green's functions satisfying, respectively, Eqs. (3.20b) and (3.20c) with $H_{eL}=0$. To all orders in $H_{eL}(\tau')$ one can show that Fig. 9(b) is associated with

$$P_{r\nu}(k,q;\omega_m) = k_BT\sum_{\omega_n}\sum_{s,t=2,4}\mathcal{G}_{rs}(k_m-q_n) \times \Gamma_{st}(q_n;k_m)\mathcal{G}_{t\nu}(k_m)\mathfrak{D}(q_n) , \quad (A7)$$

where the $\mathcal{G}$'s and $\mathfrak{D}$ are the exact Green's functions and $\Gamma_{st}(q_n;k_m)$ is called the vertex tensor. This vertex tensor is associated with the infinite sum of connected Feynman diagrams that can be constructed having two external-exciton lines and one external-phonon line (see Fig. 10). The two lowest-order terms of the vertex tensor are given:

$$\Gamma_{st}(q_n;k_m) = \Gamma_0^*(q)\left(\delta_{st} - k_BT\sum_{\omega_1}\sum_{u=2,4}\int\frac{d\vec{q}_1}{(2\pi)^3}|\Gamma_0(\vec{q}_1)|^2 \times \mathcal{G}_{su}^{(0)}(k_m-q_n-q_1)\,\mathcal{G}_{ut}^{(0)}(k_m-q_1)\mathfrak{D}^{(0)}(q_1)\right) , \quad (A8)$$

where $q_1 = (\vec{q}_1, \omega_1)$.

APPENDIX B: DEFORMATION-POTENTIAL INTERACTION

Migdal[25] has shown that the diagrammatic expan-

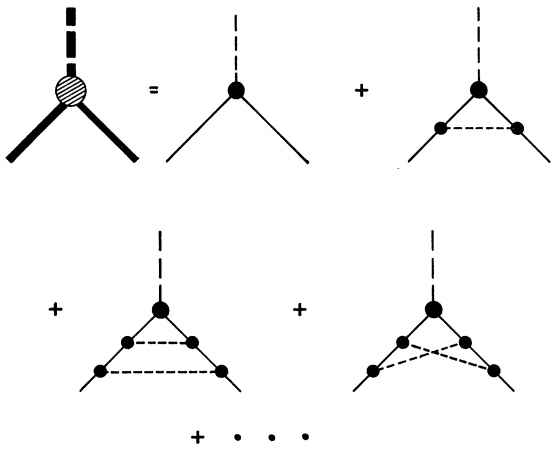

FIG. 10. Feynman diagram defining the vertex tensor.

sion for the vertex tensor in the case of the electron–acoustical-phonon interaction converges quickly at very low temperature and is given by

$$\Gamma_{\alpha\beta}(q) \approx \Gamma_0^*(q)\delta_{\alpha\beta}[1+O((m_0/m)^{1/2})] . \tag{B1}$$

Here m_0 equals electron mass, m equals the nuclear mass, and α and β are the electron-spin quantum numbers. It will be shown that a similar result is obtained for the exciton–acoustical-phonon interaction.

The coupling strength in this case is given by[1]

$$\Gamma_0(q) = (q/2\rho u)^{1/2} D\mathfrak{F}(q) , \tag{B2}$$

where D equals the deformation potential, ρ equals the density of the material, u equals the velocity of sound, and $\mathfrak{F}(q)$ is a dimensionless function of q which has a magnitude less than or equal to unity with $\mathfrak{F}(0) = 1$. Making use of Eq. (A8), one obtains consistent to terms of third order in Γ_0

$$\Gamma_{rs}(q_n; k_m) = \Gamma_0^*(q)[\delta_{rs} + \Gamma_{rs}^{(1)}(q_n; k_m)] , \tag{B3}$$

where

$$\Gamma_{rs}^{(1)}(q_n; k_m) = -(2\pi)^{-3} k_B T \sum_{\omega_1} \sum_{u=2,4} \int d\vec{q}_1 \, |\Gamma_0(\vec{q}_1)|^2 \, \mathcal{G}_{ru}^{(0)}(k_m - q_n - q_1) \, \mathcal{G}_{us}^{(0)}(k_m - q_1) \mathfrak{D}^{(0)}(q_1) . \tag{B4}$$

Here

$$q_1 \equiv (\vec{q}_1, \omega_1) , \quad k_m \equiv (\vec{k}, \omega_m) , \quad q_n \equiv (\vec{q}, \omega_n) .$$

The Green's function $\mathfrak{D}^{(0)}(q_1)$ is given by

$$\mathfrak{D}^{(0)}(\vec{q}_1, \omega_1) = -2uq_1(\omega_1^2 + u^2 q_1^2)^{-1} . \tag{B5}$$

The Green's function $\mathcal{G}_{rs}^{(0)}(k_m)$ is a solution of Eq. (3.19) with $\Sigma_{rs} \equiv 0$. When k is large in magnitude compared to ω_m/c, it can be shown that $\mathcal{G}_{rs}^{(0)}(\vec{k}, \omega_m)$ is diagonal and equal to the "bare"-exciton Green's function given by

$$\begin{aligned} &\mathcal{G}_{rs}^{(0)}(k_m) = \delta_{rs}\mathcal{G}_r^{(0)}(k_m) , \\ &\mathcal{G}_2^{(0)}(k_m) = [i\omega_m - \omega_E(\vec{k})]^{-1} , \\ &\mathcal{G}_4^{(0)}(k_m) = -[i\omega_m + \omega_E(\vec{k})]^{-1} . \end{aligned} \tag{B6}$$

As will be now shown, this latter property makes the calculation of $\Gamma_{rs}^{(1)}$ similar to that carried out by Migdal. We, too, restrict the proof to low temperatures where

$$k_B T \sum_{\omega_1} \approx (2\pi)^{-1} \int_{-\infty}^{\infty} d\omega_1 .$$

In the proof it will be assumed that $i\omega_m = \pm\omega_E(\vec{k})$; these are the dispersion relations satisfied by a bare undamped exciton. It will be further assumed that

$$\omega_E(\vec{k}) = \omega_0 + k^2/2M , \tag{B7}$$

where M represents the effective mass of the exciton.

In estimating the magnitude of $\Gamma_{rs}^{(1)}$ it is convenient to integrate with respect to q_1 first and then carry out the integration with respect to ω_1 last. The following term in Eq. (B4) has the following form:

$$-|\Gamma_0(q_1)|^2 \mathfrak{D}^{(0)}(q_1) = \begin{cases} \lesssim g^2 , & \omega_1 < uq_1 \\ \ll g^2 , & \omega_1 \gg \omega_D \end{cases} \tag{B8}$$

where $g^2 = D^2\rho^{-1}u^{-2}$ and ω_D equals the Debye energy. Thus, to a good approximation

$$\Gamma_{rs}^{(1)}(q_n; k_m) \approx (2\pi)^{-4} g^2 \sum_{u=2,4} \int_{-\omega_D}^{\omega_D} d\omega_1 \int d\vec{q}_1 \, \mathcal{G}_{ru}^{(0)}(k_m - q_n - q_1) \, \mathcal{G}_{us}^{(0)}(k_m - q_1) . \tag{B9}$$

In this expression one expects that the region near the poles in $\mathcal{G}_{ru}^{(0)}$ and $\mathcal{G}_{us}^{(0)}$ will give the most significant contribution to the integration with respect to q_1. One also expects that this contribution will be largest when these poles nearly coincide at as large a value of q_1 as possible where the density of states

is correspondingly large. As can be seen from an inspection of Eqs. (B6), (B7), and (B9), the above conditions will be met when q_1, $k-q_1$, and $k-q_1-q$ are all large compared to ω_0/c and are nearly equal in magnitude. In estimating the magnitude of $\Gamma_{rs}^{(1)}$, one is thus justified in approximating the Green's functions in Eq. (B9) by the bare-exciton Green's functions given by Eq. (B7). From this point on, the calculation essentially duplicates that of Migdal. One obtains

$$\Gamma_{rs}^{(1)}(q_n;k)=\delta_{rs}\Gamma_r^{(1)}(\vec{q},\vec{k};\ \omega_n)\ , \tag{B10}$$

where

$$\Gamma_2^{(1)}(\vec{q},\vec{k};\omega_n)=\Gamma_4^{(1)}(\vec{q},\vec{k};-\omega_n)$$

$$\approx(2\pi)^{-3}g^2Mk\int\frac{d\Omega\, i\omega_n}{i\omega_n-\vec{v}\cdot\vec{q}}$$

$$\lesssim\frac{g^2kM}{2\pi^2\hbar^2}\ . \tag{B11}$$

Here $\vec{v}=\vec{k}/M$ and $\hbar$ has been introduced so that practical units can be employed. Substituting the values $D=1$ eV, $\rho=2$ g/cm^3, $u=10^5$ cm/sec, and $M=m_0$, into Eq. (B11), one obtains $\Gamma_2^{(1)}\lesssim 10^{-8}k$. This is small compared to unity near the center of the Brillouin zone, $k\lesssim 10^7$ cm^{-1}. At larger values of k, $vq=kq/M$ will be large compared to uq so that here too $\Gamma_2^{(1)}$ will be small in the physically interesting case: $i\omega_n\approx uq$. Therefore, to a good approximation

$$\Gamma_{rs}(\vec{q})\approx\delta_{rs}\Gamma_0^*(\vec{q})\ . \tag{B12}$$

This result greatly simplifies the problem of calculating the self-energy tensor.

Substituting Eq. (B12) into Eq. (3.21), one gets in the low-temperature limit ($T\approx 0$ °K):

$$\Sigma_{rs}(k_m)\approx\mp(2\pi)^{-4}\int d\omega_n d\vec{q}\,|\Gamma_0(q)|^2$$

$$\times\,\mathfrak{D}^{(0)}(q_n)\mathcal{G}_{rs}^{(0)}(k_m-q_n)\ , \tag{B13}$$

where the $-$ sign applies when $r=2$; the $+$ sign when $r=4$. The Green's functions $\mathcal{G}_{rs}$ have been approximated by the undamped functions because the imaginary parts of Σ_{rs}, which will now be calculated, are shown to be small compared to ω_0 and can be treated as infinitesimals in Eq. (B13). The real parts of Σ_{rs} are also small compared to ω_0 and can be incorporated into $\omega_E(k)$ by renormalizing the exciton mass. The self-energy of the phonon Green's function is given by

$$\pi(q_n)\approx(2\pi)^{-4}\,|\Gamma_0(\vec{q})|^2\int d\omega_m d\vec{k}\sum_{r,s=2,4}\mathcal{G}_{rs}^{(0)}(k_m)$$

$$\times\,\mathcal{G}_{sr}^{(0)}(q_n-k_m)\ . \tag{B14}$$

The real part of π is not negligible compared to uq. However, without loss of generality the real part of π can be included in $\mathfrak{D}^0(q_n)$ by renormalizing the velocity of sound u. The imaginary part of π due to H_{eL} is zero because none of the diagrams associated with the right-hand side of Eq. (B14) conserve energy as long as $\omega_E(\vec{k})\gg\omega_P(\vec{q})$.

As was the case in evaluating $\Gamma_{rs}^{(1)}$, the most significant contribution to the integration with respect to q in Eq. (B13) comes when $\vec{q}-\vec{k}$ is large in magnitude compared to ω_0/c. One is therefore justified in approximating $\mathcal{G}_{rs}^{(0)}$ by the bare-exciton Green's functions given in Eqs. (B6). Integrating with respect to ω_n first, one obtains after making use of Eqs. (B5) and (B6)

$$\Sigma_{rs}(k_m)\approx\delta_{rs}\Sigma_r(k_m)\ ,$$

$$\Sigma_2(\vec{k},\omega_m)=-\Sigma_4(\vec{k},-\omega_m)=(2\pi)^{-3}\frac{D^2}{2\rho u}\tag{B15}$$

$$\times\int\frac{d\vec{p}\,|\vec{k}-\vec{p}|\,\mathcal{F}(\vec{k}-\vec{p})}{i\omega_m-u|\vec{k}-\vec{p}|-\omega_E(\vec{p})}\ .$$

The right-hand side of this final expression can be analytically continued into complex values of ω_m. Since the damping is assumed to be a small perturbation, one is primarily interested in the case when $i\omega_m=\omega+i\delta$, where ω is a real number and $\delta\to 0^+$. One therefore obtains after making use of the fact that $(x-i\delta)^{-1}=1/x+i\pi\delta(x)$ [refer also to Eq. (3.22)]:

$$\gamma_1(\vec{k},\omega)=\Delta_1(\vec{k},\omega)-i\delta_1(\vec{k},\omega)\ , \tag{B16}$$

$$\Delta_1(\vec{k},\omega)=-\frac{D^2}{2\rho u}(2\pi)^{-3}\,\mathcal{P}\int\frac{d\vec{p}\,|\vec{k}-\vec{p}|\,\mathcal{F}(\vec{k}-\vec{p})}{\omega_E(p)+u|\vec{k}-\vec{p}|-\omega}\ , \tag{B17}$$

$$\delta_1(\vec{k},\omega)=\pi\frac{D^2}{2\rho u}(2\pi)^{-3}\int d\Omega\,P_1^{\,2}|\vec{k}-\vec{p}_1|$$

$$\times\,\mathcal{F}(\vec{k}-\vec{p}_1)v^{-1}(p_1)\ , \tag{B18}$$

$$\gamma_2(\vec{k},\omega)=0\ , \tag{B19}$$

where $v(p)=d\omega_E(p)/dp$ and $\vec{p}_1$ is given by

$$\omega_E(\vec{p}_1)+u|\vec{k}-\vec{p}_1|=\omega\ . \tag{B20}$$

One cannot obtain analytical expressions for Δ_1 and δ_1 except in very special cases. When $\mathcal{F}(\vec{k}-\vec{p}_1)$ is approximated by $\mathcal{F}(0)=1$ and $u|\vec{k}-\vec{p}_1|$ is neglected, one can obtain an analytic expression for δ_1 in two interesting cases: $k\ll p_1$ and $k=p_1$. In these two cases, one obtains when $\omega>\omega_0$

$$\delta_1(\vec{k},\omega)\approx\begin{cases}a(\omega-\omega_0)\ , & k\ll p_1\\ \frac{2}{3}a(\omega-\omega_0)\ , & k=p_1\end{cases} \tag{B21}$$

where $a=M^2D^2/\pi\rho\hbar^3$. For the previous values used in calculating $\Gamma_{rs}^{(1)}$, $a\approx 0.5\times10^{-2}$. Therefore, the damping contribution $\delta_1(k,\omega)$ from the deformation-potential interaction is very small compared to ω_0.

From Eqs. (3.31), (B16), and (B19) one has that

$$\Gamma = \mathrm{Re}\tilde{\Gamma}(\vec{k}, \omega) \approx \delta_1(\vec{k}, \omega)\left(1 + \frac{\omega_E(\vec{k})}{\omega}\right) - \delta_1(\vec{k}, -\omega)\left(1 - \frac{\omega_E(\vec{k})}{\omega}\right) . \quad \text{(B22)}$$

When $\omega \approx \pm \omega_E(\vec{k})$, this reduces to

$$\Gamma \approx \pm 2\delta_1(\vec{k}, \pm\omega) . \quad \text{(B23)}$$

Since $\delta_1(\vec{k}, \omega)$ is positive definite, Eq. (B23) assures that the poles in $D_R(\vec{k}, \omega)$ are in the lower half of the ω plane.

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