is considered. The notation used is that of Koster,[13] and the chlorine ion is taken to be at the origin of the unit cell. In the table, n_j is the number of times the jth irreducible representation occurs. The figure in brackets after a representation label is the degeneracy of the representation, and if no figure is given the representation is singly degenerate.

*Work supported by the National Research Council of Canada.

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PHYSICAL REVIEW B VOLUME 3, NUMBER 8 15 APRIL 1971

Dynamics of Interacting Photon-Phonon Fields in Dielectric Crystals*

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(Received 8 October 1970)

The dynamic interaction between electromagnetic waves and transverse optical phonons in dielectric crystals has been studied by means of the Green's-function method. Emphasis has been given to the line shapes of the absorption bands at finite temperatures. The spectral functions for the photon and phonon fields are found to consist of the superposition of symmetric and asymmetric Lorentzian lines even if the frequency dependence of the energy shift and damping functions is neglected. The source of the asymmetry is the anharmonic coupling between the transverse photons and transverse optical phonons. General expressions for the energy shift and the damping functions are derived. The possible mechanisms that may occur in the physical process of Raman scattering are examined in detail, and expressions for the corresponding scattering amplitudes are developed. In the limiting case of absence of dispersion of the electromagnetic waves in the medium, the bare excitation spectra for both fields are also discussed.

I. INTRODUCTION

The excitation spectrum of interacting polaritons in dielectric crystals has been recently studied[1] by means of the Green's-function method. In a dielectric medium and for certain values of wave vectors of the electromagnetic field, the polariton spectrum arises from the interaction between transverse photons and transverse optical (TO) phonons. The polarization operator for the interacting polariton system has been calculated in successive approximations and the excitation spectrum has been discussed in detail. The polariton spectral function arising from polariton-polariton interactions is found to have a Lorentzian line shape, while asymmetric broadening will arise only when the frequency variation of the damping function is taken into consideration. We refer to I for details as well as for polariton literature.

Benson and Mills[2] have recently developed a theory of light scattering from polaritons in the presence of lattice damping. Their calculation is based on the assumption that the lattice anharmonicity is the dominant factor in the damping process and the anharmonic coupling between the electromagnetic field and TO phonons has been completely ignored. For the process of Raman scattering, they found that the spectral functions for the photon and phonon fields are described by Lorentzian lines. Asymmetric broadening results only when the frequency dependence and the variation of the linewidth with respect to the scattering angle are taken into account.[2] Barker[3] studied the Raman scattering spectrum of TO phonons in GaP. He found that the TO phonon mode showed considerable asymmetric broadening.[3] The purpose of this study is to show that the line shape of the spectrum arising from photon-phonon[4] interactions is always asymmetric. The source of the asymmetry is the anharmonic coupling between the photon and phonon fields. Whenever the lattice anharmonicity makes an important contribution to the damping process, the contribution arising from the anharmonic photon-phonon coupling is also substantial and results in the asymmetric broadening of the spectral line.

This paper is arranged as follows. In Sec. II we make use of the Hamiltonian obtained in I to derive the Dyson equation for the coupled photon-phonon Green's functions. General expressions for the photon and phonon Green's functions are developed and compared with the results derived from previous studies. The excitation spectrum is discussed in Sec. III in successive approximations. The spectral functions for the photon and phonon fields are found to have an asymmetric line shape. The asymmetry is caused by the photon-phonon anharmonic coupling. As an example, the possible mechanisms that may occur in the physical process of Raman scattering are discussed in Sec. IV, and the scattering amplitudes as well as the components of the polarization operator that contribute to the processes in question are calculated in the appendices. Finally, the excitation spectrum is examined in Sec. V for the special case where the electromagnetic waves in the medium suffer no dispersion but only scattering.

II. DERIVATION OF PHOTON AND PHONON GREEN'S FUNCTIONS

The Hamiltonian for a dielectric crystal, consisting of N unit cells in volume V with r atoms per unit cell, can be taken in the form of[1]

$$\mathcal{H} = \mathcal{H}_l + \mathcal{H}_r + \mathcal{H}_{lr}, \tag{1}$$

where $\mathcal{H}_l$, $\mathcal{H}_r$, and $\mathcal{H}_{lr}$ are the Hamiltonians for the lattice, the free electromagnetic field, and the interaction between them, respectively. The lattice Hamiltonian is known to be given by[1]

$$\mathcal{H}_l = \frac{1}{4}\sum_{\vec{k},j} \omega^0_{\vec{k},j}(A^\dagger_{\vec{k}j}A_{\vec{k}j} + B^\dagger_{\vec{k}j}B_{\vec{k}j}) + \sum_{n>2}\sum_{\vec{k}_1 j_1,\ldots,\vec{k}_n j_n} V_n(\vec{k}_1 j_1, \vec{k}_2 j_2, \ldots, \vec{k}_n j_n) \times A_{\vec{k}_1 j_1}A_{\vec{k}_2 j_2}\cdots A_{\vec{k}_n j_n}, \tag{2}$$

where the first term describes the free phonon field, while the second is the anharmonic interaction between them. $\omega^0_{\vec{k}j}$ is the bare phonon energy of the harmonic phonon field with wave vector $\vec{k}$ and polarization j. The operators are $A_{\vec{k}j} = a_{\vec{k}j} + a^\dagger_{\vec{k}j}$ and $B_{\vec{k}j} = a_{\vec{k}j} - a^\dagger_{\vec{k}j}$, where $a^\dagger_{\vec{k}j}$ and $a_{\vec{k}j}$ are the phonon creation and annihilation operators, respectively, which satisfy Bose statistics. The Hamiltonian for the free transverse electromagnetic field is

$$\mathcal{H}_r = \sum_{\vec{k}\lambda} ck\, b^\dagger_{\vec{k}\lambda} b_{\vec{k}\lambda}, \tag{3}$$

where $b^\dagger_{\vec{k}\lambda}$ and $b_{\vec{k}\lambda}$ are the creation and annihilation operators, respectively, for a photon with wave vector $\vec{k}$ and polarization λ (= 1, 2). ck is the energy of the radiation field, and we have assumed a system of units where $\hbar = 1$.

The photon-phonon interactions are described by the interaction Hamiltonian[1]

$$\mathcal{H}_{lr} = -\frac{i}{2}\sum_{\vec{k},\lambda,j}\left(\frac{\omega^0_{\vec{k}j}}{ck}\right)^{1/2}\kappa_j(\vec{k}\lambda)A_{\vec{k}\lambda}B_{\vec{k}j} + \frac{\omega_p^2}{4}\sum_{k,\lambda}\frac{1}{ck}A^\dagger_{\vec{k}\lambda}A_{\vec{k}\lambda} - \sum_{n>2}\sum_{\substack{k_1\lambda_1,\vec{k}_2 j_2,\\ \ldots,\vec{k}_n j_{n-1}}}\phi_n(\vec{k}_1\lambda_1, \vec{k}_2 j_2, \vec{k}_3 j_3, \ldots, \vec{k}_n j_{n-1})A_{\vec{k}_1\lambda_1}B_{\vec{k}_2 j_2}A_{\vec{k}_3 j_3}\cdots A_{\vec{k}_n j_{n-1}} + \sum_{n>2}\sum_{\substack{\vec{k}_1\lambda_1,\vec{k}_2\lambda_2,\\ \vec{k}_3 j_3,\ldots,\vec{k}_n j_{n-2}}} g_n(\vec{k}_1\lambda_1, \vec{k}_2\lambda_2, \vec{k}_3 j_3, \ldots, \vec{k}_n j_{n-2})A_{\vec{k}_1\lambda_1}A_{\vec{k}_2\lambda_2}A_{\vec{k}_3 j_3}\cdots A_{\vec{k}_n j_{n-2}}, \tag{4}$$

where $A_{\vec{k}\lambda} = b_{\vec{k}\lambda} + b^\dagger_{\vec{k}\lambda}$, and $\kappa_j(\vec{k}\lambda)$ is the coupling constant defined by Eq. (8) of I and satisfies the relation

$$\sum_j |\kappa_j(\vec{k}\lambda)|^2 = \omega_p^2, \tag{5}$$

where ω_p is the plasma frequency. The first term in (4) describes the dispersion of the electromagnetic waves in the medium, while the functions

$$\phi_n(\vec{k}_1\lambda_1, \vec{k}_2 j_2, \ldots, \vec{k}_n j_{n-1})$$

and

$$g_n(\vec{k}_1\lambda_1, \vec{k}_2\lambda_2, \vec{k}_3 j_3, \ldots, \vec{k}_n j_{n-2})$$

are the anharmonic coupling functions describing photon-phonon scattering processes and their explicit expressions are given in I.

To diagonalize the Hamiltonian (1), we introduce the retarded double-time Green's function in the matrix form[5]

$$G(\vec{k}, t-t') \equiv \langle\langle Y_{\vec{k}j\lambda}; Y^\dagger_{\vec{k}j\lambda}\rangle\rangle = -i\theta(t-t')\langle[Y_{\vec{k}j\lambda}(t), Y^\dagger_{\vec{k}j\lambda}(t')]_-\rangle, \tag{6a}$$

where $Y^\dagger_{\vec{k}j\lambda}$ is the row vector

$$Y^{\dagger}_{\vec{k}j\lambda}(t) = (A^{\dagger}_{\vec{k}\lambda}(t) \;\; B^{\dagger}_{\vec{k}\lambda}(t) \;\; A^{\dagger}_{\vec{k}j}(t) \;\; B^{\dagger}_{\vec{k}j}(t)) \tag{6b}$$

and the angular brackets in (6a) denote the average over the canonical ensemble appropriate to the total Hamiltonian $\mathcal{H}$. $\theta(t)$ is the usual step function and the operators $Y_{\vec{k}j\lambda}$ and $Y^{\dagger}_{\vec{k}j\lambda}$ are in the Heisenberg representation. In what follows, the time arguments of the operators will be suppressed for convenience. The Fourier transform of the Green's function (6a) with respect to the argument t satisfies the equation of motion

$$\omega G(\vec{k};\omega) = (1/2\pi)\langle [Y_{\vec{k}j\lambda}, Y^{\dagger}_{\vec{k}j\lambda}]_-\rangle_{t=t'} + \langle\langle [Y_{\vec{k}j\lambda}, \mathcal{H}]_-; Y^{\dagger}_{\vec{k}j\lambda}\rangle\rangle . \tag{7}$$

Using Eqs. (2)–(4) and (7), we derive the equation of motion for the Green's function $G(\vec{k};\omega)$ as

$$G^{-1}_{00}(\vec{k};\omega)G(\vec{k};\omega) = \hat{I} + \langle\langle F_{\vec{k}j\lambda}; Y^{\dagger}_{\vec{k}j\lambda}\rangle\rangle , \tag{8}$$

where the unperturbed Green's function $G_{00}(\vec{k};\omega)$ is given by

$$G^{-1}_{00}(k;\omega) = \begin{pmatrix} \omega & -ck & 0 & 0 \\ -ck-\omega_p^2/ck & \omega & 0 & i\sum_j \kappa_j(\vec{k},\lambda)(\omega^0_{\vec{k}j}/ck)^{1/2} \\ -i\sum_\lambda \kappa_j(\vec{k},\lambda)(\omega^0_{\vec{k}j}/ck)^{1/2} & 0 & \omega & -\omega^0_{\vec{k}j} \\ 0 & 0 & -\omega^0_{\vec{k}j} & \omega \end{pmatrix} \tag{9}$$

and

$$\hat{I} = \frac{1}{\pi}\begin{pmatrix} 0 & 1 & 0 & 0 \\ 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 0 & 1 & 0 \end{pmatrix} .$$

The function $F_{\vec{k}j\lambda}$ is defined as

$$F_{\vec{k}j\lambda} \equiv \begin{pmatrix} 0 \\ F_1(\vec{k}\lambda) \\ F_2(\vec{k}j) \\ F_3(\vec{k}j) \end{pmatrix} , \tag{10a}$$

where

$$F_1(\vec{k}\lambda) = -2\sum_{n>2}\sum \phi_n(\vec{k}_1 j_1, \vec{k}_2 j_2, \ldots, \vec{k}_{n-1} j_{n-1}, -\vec{k}\lambda) B_{\vec{k}_1 j_1} A_{\vec{k}_2 j_2} \cdots A_{\vec{k}_{n-1} j_{n-1}} + 4\sum_{n>2}\sum g_n(\vec{k}_1\lambda_1, \vec{k}_2 j_2, \ldots, \vec{k}_{n-1} j_{n-2}, -\vec{k}\lambda) A_{\vec{k}_1\lambda_1} A_{\vec{k}_2 j_2} \cdots A_{\vec{k}_{n-1} j_{n-2}}, \tag{10b}$$

$$F_2(\vec{k}j) = 2\sum_{n>2}\sum \phi_n(\vec{k}_1\lambda_1, \vec{k}_2 j_2, \ldots, \vec{k}_{n-1} j_{n-2}, -\vec{k}j) A_{\vec{k}_1\lambda_1} A_{\vec{k}_2 j_2} \cdots A_{\vec{k}_{n-1} j_{n-2}} , \tag{10c}$$

$$F_3(\vec{k}j) = 2\sum_{n>2}\sum [nV_n(\vec{k}_1 j_1, \vec{k}_2 j_2, \ldots, \vec{k}_{n-1} j_{n-1}, -\vec{k}j) A_{\vec{k}_1 j_1} A_{\vec{k}_2 j_2} \cdots A_{\vec{k}_{n-1} j_{n-1}} + (n-2) g_n(\vec{k}_1\lambda_1, \vec{k}_2\lambda_2, \vec{k}_3 j_3, \ldots, \vec{k}_{n-1} j_{n-3} - \vec{k}j) A_{\vec{k}_1\lambda_1} A_{\vec{k}_2\lambda_2} A_{\vec{k}_3 j_3} \cdots A_{\vec{k}_{n-1} j_{n-3}} - (n-2)\phi_n(\vec{k}_1\lambda_1, \vec{k}_2 j_2, \ldots, \vec{k}_{n-1} j_{n-2}, -\vec{k}j) A_{\vec{k}_1\lambda_1} B_{\vec{k}_2 j_2} A_{\vec{k}_3 j_3} \cdots A_{\vec{k}_{n-1} j_{n-2}}] . \tag{10d}$$

Considering the equation of motion for the Green's function $\langle\langle F_{\vec{k}j\lambda}; Y^{\dagger}_{\vec{k}j\lambda}\rangle\rangle$ with respect to the argument t', we find

$$\langle\langle F_{\vec{k}j\lambda}; Y^{\dagger}_{\vec{k}j\lambda}\rangle\rangle = \{(1/2\pi)\langle [F_{\vec{k}j\lambda}, Y^{\dagger}_{\vec{k}j\lambda}]_-\rangle_{t=t'} + \langle\langle F_{\vec{k}j\lambda}; F^{\dagger}_{\vec{k}j\lambda}\rangle\rangle\} G_{00}(\vec{k};\omega) . \tag{11}$$

Substituting expression (11) into (8), we derive the Dyson equation

$$[G^{-1}_{00}(\vec{k},\omega) - \Pi_{j\lambda}(\vec{k},\omega)]G(\vec{k};\omega) = \hat{I} , \tag{12}$$

where the polarization operator $\Pi_{j\lambda}(\vec{k},\omega)$ is defined by

$$\Pi_{j\lambda}(\vec{k},\omega) = P(\vec{k};\omega)[\hat{I} + G_{00}(\vec{k},\omega)P(\vec{k},\omega)]^{-1} , \tag{13}$$

and the function $P(\vec{k},\omega)$ is

$$P(\vec{k},\omega) = (1/2\pi)\langle [F_{\vec{k}j\lambda}, Y^{\dagger}_{\vec{k}j\lambda}]_-\rangle_{t=t'} + \langle\langle F_{\vec{k}j\lambda}; F^{\dagger}_{\vec{k}j\lambda}\rangle\rangle . \tag{14}$$

For frequencies ω far from the zeros of the denominator in (13), we may expand the denominator into power series, and retaining only the first term, we have

$$\Pi_{j\lambda}(\vec{k},\omega) \approx P(\vec{k},\omega)\hat{I}^{-1} . \tag{15}$$

Substituting Eq. (15) into Eq. (12), we obtain

$$[G^{-1}_{00}(\vec{k};\omega) - P(\vec{k},\omega)\hat{I}^{-1}]G(\vec{k};\omega) = \hat{I} . \tag{16}$$

The system of equations (16) shall be used to obtain the components of the Green's function $G(\vec{k};\omega)$ describing the excitation spectrum arising from photon-phonon interactions. In deriving (16) we have confined ourselves only to the diagonal part of the Green's function with respect to the phonon polarization index j. To include nondiagonal contributions we refer to I and Eq. (42) of I.

Taking the matrix elements of (16), we have

$$\langle\langle A_{\vec{k}\lambda};A^\dagger_{\vec{k}\lambda}\rangle\rangle=\frac{ck}{\pi}D_{\vec{k}j}(\omega)\{[\omega^2-c^2k^2-\omega_p^2-ckP_{21}(\vec{k},\omega)]D_{\vec{k}j}(\omega)-\sum_{j\lambda}X_j^2(\vec{k}\lambda)\omega^0_{\vec{k}j}[\omega^0_{\vec{k}j}+P_{43}(\vec{k},\omega)]+ck\Lambda_{\vec{k}}(\omega)\}^{-1}$$

$$=\frac{ck}{\pi}[\omega^2\epsilon(\vec{k},\omega)-c^2k^2]^{-1}, \quad (17)$$

$$\langle\langle B_{\vec{k}\lambda};B^\dagger_{\vec{k}\lambda}\rangle\rangle=-\frac{1}{ck\pi}+\left(\frac{\omega}{ck}\right)^2\langle\langle A_{\vec{k}\lambda};A^\dagger_{\vec{k}\lambda}\rangle\rangle, \quad (18)$$

$$\langle\langle A_{\vec{k}j};A^\dagger_{\vec{k}j}\rangle\rangle=\frac{1}{\pi}\{[\omega^0_{\vec{k}j}+P_{34}(\vec{k},\omega)][\omega^2-c^2k^2-\omega_p^2-ckP_{21}(\vec{k},\omega)]+\sum_{j\lambda}X_j^2(\vec{k}\lambda)\omega^0_{\vec{k}j}\}[\omega^2\epsilon(\vec{k},\omega)-c^2k^2]^{-1}D^{-1}_{\vec{k}j}(\omega), \quad (19a)$$

$$\langle\langle B_{\vec{k}j};B^\dagger_{\vec{k}j}\rangle\rangle=\frac{1}{\pi}\{[\omega^0_{\vec{k}j}+P_{43}(\vec{k},\omega)][\omega^2-c^2k^2-\omega_p^2-ckP_{21}(\vec{k},\omega)]+ckP_{23}(\vec{k},\omega)P_{41}(\vec{k},\omega)\}[\omega^2\epsilon(\vec{k},\omega)-c^2k^2]^{-1}D^{-1}_{\vec{k}j}(\omega). \quad (19b)$$

In Eqs. (17)–(19), $\epsilon(\vec{k},\omega)$ may be defined as the frequency- and wave-vector-dependent dielectric function

$$\epsilon(\vec{k},\omega)=1-\frac{\omega_p^2}{\omega^2}-\frac{1}{\omega^2}\sum_{j,\lambda}X_j^2(\vec{k}\lambda)\omega^0_{\vec{k}j}\times[\omega^0_{\vec{k}j}+P_{43}(\vec{k},\omega)]D^{-1}_{\vec{k}j}(\omega)-\frac{ck}{\omega^2}P_{21}(\vec{k},\omega)+\frac{ck}{\omega^2}\Lambda_{\vec{k}}(\omega)D^{-1}_{\vec{k}j}(\omega), \quad (20)$$

and the function $D_{\vec{k}j}(\omega)$ is given by

$$D_{\vec{k}j}(\omega)=[\omega-P_{33}(\vec{k},\omega)][\omega-P_{44}(\vec{k},\omega)]-[\omega^0_{\vec{k}j}+P_{43}(\vec{k},\omega)][\omega^0_{\vec{k}j}+P_{34}(\vec{k},\omega)]. \quad (21)$$

The components of the function $P(\vec{k},\omega)$ appearing in expressions (17)–(21) have the following form:

$$P_{21}(\vec{k},\omega)=P^0_{21}+\pi\langle\langle F_1(\vec{k}\lambda);F_1^\dagger(\vec{k}\lambda)\rangle\rangle, \quad (22a)$$

$$P_{33}(\vec{k},\omega)=P^0_{33}+\pi\langle\langle F_2(\vec{k}j);F_3^\dagger(\vec{k}j)\rangle\rangle, \quad (22b)$$

$$P_{44}(\vec{k},\omega)=P^0_{44}+\pi\langle\langle F_3(\vec{k}j);F_2^\dagger(\vec{k}j)\rangle\rangle, \quad (22c)$$

$$P_{34}(\vec{k},\omega)=\pi\langle\langle F_2(\vec{k}j);F_2^\dagger(\vec{k}j)\rangle\rangle, \quad (22d)$$

$$P_{43}(\vec{k},\omega)=P^0_{43}+\pi\langle\langle F_3(\vec{k}j);F_3^\dagger(\vec{k}j)\rangle\rangle; \quad (22e)$$

$$P_{23}(\vec{k},\omega)=P^0_{23}+\pi\langle\langle F_1(\vec{k}\lambda);F_3^\dagger(\vec{k}j)\rangle\rangle, \quad (23a)$$

$$P_{41}(\vec{k},\omega)=P^0_{41}+\pi\langle\langle F_3(\vec{k}j);F_1^\dagger(\vec{k}\lambda)\rangle\rangle, \quad (23b)$$

$$X_j(\vec{k}\lambda)=\kappa_j(\vec{k}\lambda)-i(ck/\omega^0_{\vec{k}j})^{1/2}P^0_{31}, \quad (23c)$$

where

$$P^0_{21}=\langle[F_1(\vec{k}\lambda),B^\dagger_{\vec{k}}]_-\rangle_{t=t'}=8\sum_{n>2}\sum g_n(\vec{k}_1j_1,\vec{k}_2j_2,\ldots,\vec{k}_{n-2},j_{n-2},\vec{k}\lambda,-\vec{k}\lambda)\langle A_{\vec{k}_1j_1}A_{\vec{k}_2j_2}\cdots A_{\vec{k}_{n-2}j_{n-2}}\rangle, \quad (24a)$$

$$P^0_{33}=\langle[F_2(\vec{k}j),B^\dagger_{\vec{k}j}]_-\rangle_{t=t'}=-P^0_{44}=-\langle[F_3(\vec{k}j),A^\dagger_{\vec{k}j}]_-\rangle_{t=t'}$$
$$=4\sum_{n>2}\sum(n-2)\phi_n(\vec{k}_1\lambda_1,\vec{k}_2j_2,\ldots,\vec{k}_{n-2}j_{n-3},\vec{k}j,-\vec{k}j)\langle A_{\vec{k}_1\lambda_1}A_{\vec{k}_2j_2}\cdots A_{\vec{k}_{n-2}j_{n-3}}\rangle, \quad (24b)$$

$$P^0_{43}=\langle[F_3(\vec{k}j),B^\dagger_{\vec{k}j}]_-\rangle_{t=t'}=4\sum_{n>2}\sum[n(n-1)V_n(\vec{k}_1j_1,\vec{k}_2j_2,\ldots,\vec{k}_{n-2}j_{n-2},\vec{k}j,-\vec{k}j)\langle A_{\vec{k}_1j_1}A_{\vec{k}_2j_2}\cdots A_{\vec{k}_{n-2}j_{n-2}}\rangle$$
$$+(n-2)(n-3)g_n(\vec{k}_1\lambda_1,\vec{k}_2\lambda_2,\vec{k}_3j_3,\ldots,\vec{k}_{n-2}j_{n-4},\vec{k}j,-\vec{k}j)\langle A_{\vec{k}_1\lambda_1}A_{\vec{k}_2\lambda_2}A_{\vec{k}_3j_3}\cdots A_{\vec{k}_{n-2}j_{n-4}}\rangle$$
$$-(n-2)(n-3)\phi_n(\vec{k}_1\lambda_1,\vec{k}_2j_2,\ldots,\vec{k}_{n-2}j_{n-3},\vec{k}j,-\vec{k}j)\langle A_{\vec{k}_1\lambda_1}B_{\vec{k}_2j_2}A_{\vec{k}_3j_3}\cdots A_{\vec{k}_{n-2}j_{n-3}}\rangle], \quad (24c)$$

$$P^0_{23}=\langle[F_1(\vec{k}\lambda),B^\dagger_{\vec{k}j}]_-\rangle_{t=t'}=P^0_{41}=\langle[F_3(\vec{k}j),B^\dagger_{\vec{k}\lambda}]_-\rangle_{t=t'}$$
$$=-4\sum_{n>2}\sum\phi_n(\vec{k}_1j_1,\vec{k}_2j_2,\ldots,\vec{k}_{n-2}j_{n-2},\vec{k}j,-\vec{k}\lambda)\langle B_{\vec{k}_1j_1}A_{\vec{k}_2j_2}\cdots A_{\vec{k}_{n-2}j_{n-2}}\rangle$$
$$+8\sum_{n>2}\sum(n-2)g_n(\vec{k}_1\lambda_1,\vec{k}_2j_2,\ldots,\vec{k}_{n-2}j_{n-3},\vec{k}j,-\vec{k}\lambda)\langle A_{\vec{k}_1\lambda_1}A_{\vec{k}_2j_2}\cdots A_{\vec{k}_{n-1}j_{n-3}}\rangle, \quad (24d)$$

$$P^0_{31}=\langle[F_2(\vec{k}j),B^\dagger_{\vec{k}\lambda}]_-\rangle_{t=t'}=-\langle[F_1(\vec{k}\lambda),A^\dagger_{\vec{k}j}]_-\rangle_{t=t'}=4\sum_{n>2}\sum\phi_n(\vec{k}_1j_1,\vec{k}_2j_2,\ldots,\vec{k}_{n-2}j_{n-2},\vec{k}\lambda,-\vec{k}j)\langle A_{\vec{k}_1j_1}A_{\vec{k}_2j_2}\cdots A_{\vec{k}_{n-2}j_{n-2}}\rangle. \quad (24e)$$

The expression (20) for the dielectric function $\epsilon(\vec{k}, \omega)$ describes the physical process arising from photon-phonon interactions in a dielectric medium. For example, the third term in (20) describes coupled excitations of the polariton type. Its importance depends on the magnitude of the coupling function $\kappa_j(\vec{k}\lambda)$ or the renormalized one $X_j(\vec{k}\lambda)$ given by Eq. (23c). The same term in (20) includes anharmonic effects to all orders for the coupled photon-phonon system. The fourth term in (20) represents direct scattering processes arising from the anharmonic coupling between electromagnetic waves and phonons of the medium. It is an entirely nonlinear effect caused by the absorption or scattering of the photon $\vec{k}\lambda$ by the assembly of phonons. The function $\Lambda_{\vec{k}}(\omega)$ in the last term of (20) consists of terms which are quadratic and higher order in the P's; it makes a negligibly small contribution to $\epsilon(\vec{k}, \omega)$, and, therefore, it shall be discarded.

Expressions (17)–(21) are reduced to those recently derived by Benson and Mills,[2] if all the anharmonic coupling functions between the electromagnetic and the phonon fields are neglected, i.e., when the coupling functions

$$\phi_n(\vec{k}_1\lambda_1, \vec{k}_2 j_2, \ldots) \text{ and } g_n(\vec{k}_1\lambda_1, \vec{k}_2\lambda_2, \vec{k}_3 j_3, \ldots)$$

are taken to be equal to zero and only the bare phonon anharmonic function $V_n(\vec{k}_1 j_1, \vec{k}_2 j_2, \ldots)$ is retained.

In order to make a connection between the expression for the Green's function (17) and the corresponding one for the polariton field derived in our earlier work,[1] we write the function $D_{\vec{k}j}(\omega)$ as

$$D^{-1}_{\vec{k}j}(\omega) = (\omega^2 - \omega^{02}_{\vec{k}j})^{-1}\,[1 - \Sigma_{\vec{k}j}(\omega)]^{-1}\ , \tag{25a}$$

where

$$\Sigma_{\vec{k}j}(\omega) = \frac{\omega^0_{\vec{k}j}}{\omega^2 - \omega^{02}_{\vec{k}j}} \left(P_{43}(\vec{k}, \omega) + P_{34}(\vec{k}, \omega) + \frac{\omega}{\omega^0_{\vec{k}j}}\,[P_{33}(\vec{k}, \omega) + P_{44}(\vec{k}, \omega)] \right)\ . \tag{25b}$$

In (25b) we have retained only linear terms in the P's. If we are now allowed to expand (25a) into power series and keep only the first term in the expansion, we have

$$D^{-1}_{\vec{k}j}(\omega) \approx (\omega^2 - \omega^{02}_{\vec{k}j})^{-1}\,[1 + \Sigma_{\vec{k}j}(\omega)]\ . \tag{25c}$$

Substituting (25c) into (20) and then the resulting expression for $\epsilon(\vec{k}, \omega)$ into (17), we have

$$\langle\langle A_{\vec{k}\lambda}; A^\dagger_{\vec{k}\lambda}\rangle\rangle \approx \frac{ck}{\pi} \left(\omega^2 - c^2k^2 - \omega_p^2 - ckP_{21}(\vec{k}, \omega) - \sum_{j\lambda} \frac{X_j^2(\vec{k}\lambda)\omega^0_{\vec{k}j}}{\omega^2 - \omega^{02}_{\vec{k}j}}\,[\omega^0_{\vec{k}j} + P_{43}(\vec{k}, \omega)]\,[1 + \Sigma_{\vec{k}j}(\omega)] \right)^{-1}\ . \tag{25d}$$

If we now transform all the terms in expression (25d) into the polariton representation, the derived expression is identical with the polariton Green's function given by Eq. (25) of I. Rewriting Eq. (17) in the form of (25d), we have taken all the ω-dependent terms into the denominator; that is, expression (20) for the dielectric function has been linearized through the Eq. (25c) and, therefore, we have approximated the line shape of the function (17) with that of (25d), which is obviously described by a modified Lorentzian curve. The expansion (25c) is applicable provided that the function $\Sigma_{kj}(\omega)$ is nonsingular and varies slowly with ω. In a similar fashion the phonon Green's function (19) can be transformed into the equivalent Green's function in the polariton representation. Therefore, our earlier work in I is a linearized version of the results derived in the present study.

We consider now the special case which occurs when the first term in the expression for the interaction Hamiltonian (4) is equal to zero, i.e., when the quantity $X_j(\vec{k}\lambda)$ is negligibly small and can be taken to be zero. Then the photon $\vec{k}\lambda$ suffers no dispersion in the medium but only scattering, which is caused by the third and fourth nonvanishing terms in the Hamiltonian (4). In this case, the photon and phonon Green's functions (17) and (19) are no longer coupled and they are given by

$$\langle\langle A_{\vec{k}\lambda}; A^\dagger_{\vec{k}\lambda}\rangle\rangle_b = (ck/\pi)[\omega^2 - c^2k^2 - \omega_p^2 - ckP_{21}(\vec{k}, \omega)]^{-1}\ , \tag{26}$$

$$\langle\langle A_{\vec{k}j}; A^\dagger_{\vec{k}j}\rangle\rangle_b = (1/\pi)[\omega^0_{\vec{k}j} + P_{34}(\vec{k}, \omega)]D^{-1}_{\vec{k}j}(\omega)\ . \tag{27}$$

The expressions (26) and (27) represent different excitation spectra. In view of (22a) and (10b), expression (26) describes the physical processes where the absorbed photon $\vec{k}\lambda$ is converted into phonons, as well as photon-photon scattering processes with the emission or absorption of phonons. The excitation spectrum of (27) arises from phonon-phonon interactions and phonon-phonon scattering processes through the emission or absorption of photons.

III. EXCITATION SPECTRUM

We shall now discuss the excitation spectrum arising from photon-phonon interactions in successive approximations by using the general expressions for the photon and phonon Green's functions derived in Sec. II.

A. Static Approximation

The excitation spectrum in the static approximation is defined when all dynamic contributions in the expression for the polarization operator are neglected, i.e., when all the Green's functions $\langle\langle F_i; F_l^\dagger\rangle\rangle$ with $i, l = 1, 2, 3$ in expressions (22) and (23) are discarded. In this approximation the Green's functions (17) and (19) take the form

$$\langle\langle A_{\vec{k}\lambda}; A^\dagger_{\vec{k}\lambda}\rangle\rangle^0 = (ck/\pi)[\omega^2\epsilon_0(\vec{k},\omega) - c^2k^2]^{-1}, \tag{28a}$$

$$\langle\langle A_{\vec{k}j}; A^\dagger_{\vec{k}j}\rangle\rangle^0 = \frac{1}{\pi}\frac{\omega^0_{\vec{k}j}}{D^0_{\vec{k}j}(\omega)} + \frac{\omega^2 - P^{02}_{33}}{\omega^0_{\vec{k}j} + P^0_{43}}\left(\frac{d[\omega^2\epsilon_0(\vec{k},\omega)]}{d\omega^2} - 1\right)[\omega^2\epsilon_0(\vec{k},\omega) - 1]^{-1}, \tag{28b}$$

$$\langle\langle B_{\vec{k}j}; B^\dagger_{\vec{k}j}\rangle\rangle^0 = \frac{1}{\pi}\frac{\omega^0_{\vec{k}j} + P^0_{43}}{D^0_{\vec{k}j}(\omega)} + \frac{1}{\pi}\left[(\omega^0_{\vec{k}j} + P^0_{43})\left(\frac{d[\omega^2\epsilon_0(\vec{k},\omega)]}{d\omega^2} - 1\right) + \frac{ckP^{02}_{23}}{D^0_{\vec{k}j}(\omega)}\right][\omega^2\epsilon_0(\vec{k},\omega) - c^2k^2]^{-1}, \tag{28c}$$

where

$$\epsilon_0(\vec{k},\omega) = 1 - \frac{\omega_p^2}{\omega^2} - \frac{1}{\omega^2}\sum_{j\lambda}\frac{X_j^2(\vec{k}\lambda)\,\omega^0_{\vec{k}j}(\omega^0_{\vec{k}j} + P^0_{43})}{D^0_{\vec{k}j}(\omega)} - \frac{ck}{\omega^2}P^0_{21}, \tag{29a}$$

$$D^0_{\vec{k}j}(\omega) = \omega^2 - \omega^{02}_{\vec{k}j} - \omega^0_{\vec{k}j}P^0_{43} - P^{02}_{33}. \tag{29b}$$

Expression (29a) for the dielectric function is a well-behaved function of ω, i.e., for $\omega \to \infty$,

$$\epsilon_0(\vec{k},\omega) \to 1 - \omega_p^2/\omega^2 - (ck/\omega^2)P^0_{21},$$

while for $\omega \to 0$ and $\vec{k} \to 0$,

$$\epsilon_0(k,\omega) = 1 - \sum|\kappa_j(\vec{k}\lambda)|^2/D^0_{\vec{k}j}(\omega). \tag{29c}$$

If the energy of excitation $\omega_{\vec{k}\rho}$ is determined from the ρth solution of the equation

$$\omega^2_{\vec{k}\rho}\epsilon_0(k,\omega_{\vec{k}\rho}) - c^2k^2 = 0, \tag{30}$$

then we may expand the denominators in (28) in power series, and retaining only the first term in the expansion, we have

$$\langle\langle A_{\vec{k}\lambda}; A^\dagger_{\vec{k}\lambda}\rangle\rangle^0 = [(ck/\pi)\lambda^0_{\vec{k}}(\omega_{\vec{k}\rho})](\omega^2 - \omega^2_{\vec{k}\rho})^{-1}, \tag{31a}$$

where

$$[\lambda^0_{\vec{k}}(\omega_{\vec{k}\rho})]^{-1} = \sum_\rho\left(\frac{d[\omega^2\epsilon_0(\vec{k},\omega)]}{d\omega^2}\right)_{\omega=\omega_{\vec{k}\rho}}. \tag{31b}$$

In this approximation, the Hamiltonian may be written as

$$\mathcal{H}^0_{p,\,\mathrm{ren}} = \mathrm{const} + \sum_{\vec{k}\rho}\omega_{\vec{k}\rho}\alpha^\dagger_{\vec{k}\rho}\alpha_{\vec{k}\rho}, \tag{32}$$

where $\alpha^\dagger_{\vec{k}\rho}$ and $\alpha_{\vec{k}\rho}$ are the polariton creation and annihilation operators with wave vector $\vec{k}$ and band index ρ, and where ren stands for renormalized. From (28), we derive the following expressions for the occupation numbers:

$$N^0_{\vec{k}\lambda}(\omega_{\vec{k}\rho}) \equiv \langle A^\dagger_{\vec{k}\lambda}A_{\vec{k}\lambda}\rangle^0 = (ck/\omega_{\vec{k}\rho})\lambda^0_{\vec{k}}(\omega_{\vec{k}\rho})\eta_{\vec{k}\rho}, \tag{33a}$$

$$N^0_{\vec{k}j}(\omega_{\vec{k}\rho}) \equiv \langle A^\dagger_{\vec{k}j}A_{\vec{k}j}\rangle^0 = (\omega^0_{\vec{k}j}/\omega_{\vec{k}\rho})\bar{\lambda}^0_{\vec{k}}(\omega_{\vec{k}\rho})\eta_{\vec{k}\rho}, \tag{33b}$$

$$\bar{N}_{\vec{k}j}(\omega_{\vec{k}\rho}) \equiv \langle B^\dagger_{\vec{k}j}B_{\vec{k}j}\rangle^0 = \left(\frac{\omega_{\vec{k}\rho}}{\omega^0_{\vec{k}j}}\right)\bar{\lambda}^0_{\vec{k}}(\omega_{\vec{k}\rho})\eta_{\vec{k}\rho}, \tag{33c}$$

where

$$\bar{\lambda}^0_{\vec{k}}(\omega_{\vec{k}\rho}) = 1 - \lambda^0_{\vec{k}}(\omega_{\vec{k}\rho}) \quad\text{and}\quad \eta_{\vec{k}\rho} = \coth\tfrac{1}{2}\beta\omega_{\vec{k}\rho}.$$

Here $\beta = (K_B T)^{-1}$, where K_B is Boltzmann's constant and T the absolute temperature. In deriving (33) we have retained only linear terms in the P^0's.

In order to evaluate the energy of excitation $\omega_{\vec{k}\rho}$ through Eq. (30) in a self-consistent manner, one has to calculate the functions P^0_{il}, which are given by expressions (24). This can be done, as has been suggested in I, by decoupling the correlation functions in (24) into products of photon, phonon, and mixed occupation numbers $N_{\vec{q}\lambda'}$, $N_{\vec{q}j'}$, $\bar{N}_{\vec{q}j'}$, and $\langle A^\dagger_{\vec{q}\lambda}A_{\vec{q}j}\rangle$, respectively, which are given by Eqs. (33). For example,

$$P^0_{31} = 4\sum_{\vec{q}j'}\phi_4(\vec{q}j', -\vec{q}j', \vec{k}\lambda, -\vec{k}j)N^0_{\vec{q}j'} + (\text{terms with } n > 4), \tag{34a}$$

$$P^0_{21} = 8\sum_{\vec{q}j'}g_4(\vec{q}j', -\vec{q}j', \vec{k}\lambda, -\vec{k}\lambda)N^0_{\vec{q}j'} + (\text{terms with } n > 4), \tag{34b}$$

$$P^0_{33} = 8\sum_{\vec{q}j'\lambda'}\phi_4(\vec{q}\lambda' - \vec{q}j', \vec{k}j, -\vec{k}j)\langle A^\dagger_{\vec{q}\lambda'}A_{\vec{q}j'}\rangle^{(0)} + (\text{terms with } n > 4), \tag{34c}$$

$$P^0_{23} = 4\sum_{\vec{q},\lambda',j'}[\phi_4(\vec{q}j' - \vec{q}\lambda', \vec{k}\lambda, -\vec{k}\lambda)\langle B^\dagger_{\vec{q}j'}A_{\vec{q}\lambda'}\rangle^{(0)} + 2g_4'(\vec{q}\lambda' - \vec{q}j', \vec{k}j - \vec{k}\lambda^\dagger)\langle A^\dagger_{\vec{q}\lambda'}A_{\vec{q}j'}\rangle^{(0)}] + (\text{terms with } n > 4), \tag{34d}$$

$$P^0_{43} = 8\sum_{\vec{q},\lambda',j'}[6V_4(\vec{q}j', -\vec{q}j', \vec{k}j, -\vec{k}j)N^0_{\vec{q}j'} + g_4(\vec{q}\lambda', -\vec{q}\lambda', \vec{k}j, -\vec{k}j)N_{\vec{q}\lambda'} + \phi_4(\vec{q}\lambda', -\vec{q}j', \vec{k}j, -\vec{k}j)\langle B^\dagger_{\vec{q}j'}A_{\vec{q}\lambda'}\rangle] + (\text{in terms with } n > 4). \tag{34e}$$

The energy of excitation $\omega_{\vec{k}\rho}$ must be derived finally from Eq. (30) by computation. In this approximation the excitation energy $\omega_{\vec{k}\rho}$, as well as the dielectric function $\epsilon_0(\vec{k},\omega_{\vec{k}\rho})$, is a real quantity and temperature dependent.

The bare photon and phonon Green's functions (26) and (27) in the static approximation become

$$\langle\langle A_{\vec{k}\lambda}; A^\dagger_{\vec{k}\lambda}\rangle\rangle^0_b = (ck/\pi)(\omega^2 - c^2k^2 - \omega_p^2 - ckP^0_{21})^{-1}, \tag{35a}$$

$$\langle\langle A_{\vec{k}j};A^{\dagger}_{\vec{k}j}\rangle\rangle^0_b=\frac{1}{\pi}\frac{\omega^0_{\vec{k}j}}{\tilde{D}^0_{\vec{k}j}(\omega)}, \tag{35b}$$

$$\langle\langle B_{\vec{k}j};B^{\dagger}_{\vec{k}j}\rangle\rangle^0_b=\frac{1}{\pi}\frac{\omega^0_{\vec{k}j}+P^0_{43}}{\tilde{D}^0_{\vec{k}j}(\omega)}, \tag{35c}$$

where

$$\tilde{D}_{\vec{k}j}(\omega)=\omega^2-\omega^0_{\vec{k}j}(\omega^0_{\vec{k}j}+P^0_{43}), \tag{36}$$

and the function P^0_{43} is given by expression (34e) with the last term taken equal to zero. In this approximation expressions (35) describe the bare renormalized photon and phonon fields, respectively, which are independent of one another. From (35) we derive the following expressions for the occupation numbers:

$$\langle A^{\dagger}_{\vec{k}\lambda}A_{\vec{k}\lambda}\rangle^0_b=(ck/\Omega_{\vec{k}\lambda})\eta_{\vec{k}\lambda} \tag{37a}$$

and

$$\langle A^{\dagger}_{\vec{k}j}A_{\vec{k}j}\rangle^0_b=(\omega^0_{\vec{k}j}/\tilde{\omega}_{\vec{k}j})\eta_{\vec{k}j},$$

$$\langle B^{\dagger}_{\vec{k}j}B_{\vec{k}j}\rangle^0_b=(\tilde{\omega}_{\vec{k}j}/\omega^0_{\vec{k}j})\eta_{\vec{k}j}, \tag{37b}$$

where $\eta_{\vec{k}\lambda}=\coth\frac{1}{2}\beta\Omega_{\vec{k}\lambda}$, and $\Omega_{\vec{k}\lambda}$ and $\tilde{\omega}_{\vec{k}j}$ are the renormalized energies of excitation for the bare photon and phonon fields determined from the solutions of the following equations, respectively:

$$\Omega^2_{\vec{k}\lambda}-c^2k^2-\omega_p^2-ck\,P^0_{21}=0, \tag{37c}$$

$$\tilde{\omega}^2_{\vec{k}j}-\omega^{02}_{\vec{k}j}-\omega^0_{\vec{k}j}P^0_{43}=0. \tag{37d}$$

In this approximation, the Hamiltonian takes the form

$$\mathcal{H}^0_{\text{ren}}=\text{const}+\sum_{\vec{k}\lambda}\Omega_{\vec{k}\lambda}\tilde{b}^{\dagger}_{\vec{k}\lambda}\tilde{b}_{\vec{k}\lambda}+\sum_{\vec{k}j}\tilde{\omega}_{\vec{k}j}\tilde{a}^{\dagger}_{\vec{k}j}a_{\vec{k}j}, \tag{37e}$$

where $\tilde{b}^{\dagger}_{\vec{k}\lambda}$, $\tilde{b}_{\vec{k}\lambda}$ and $\tilde{a}^{\dagger}_{\vec{k}\lambda}$, $\tilde{a}_{\vec{k}\lambda}$ are the new photon and phonon creation and annihilation operators describing the renormalized photon and phonon fields, respectively.

B. Photon-Phonon Line Shapes

To study the line shapes arising from photon-phonon interactions, we make use of the spectral function $J_{\vec{k}}(\omega)$ given by the relation

$$J_{\vec{k}}(\omega)=-2(e^{\beta\omega}-1)^{-1}\,\text{Im}\bar{G}(\vec{k};\omega),$$

where Im stands for the imaginary part and $\bar{G}(\vec{k};\omega)$ is the Green's function for the physical process in question.

Taking the imaginary parts of (19) and (17) for the phonon and photon Green's functions, respectively, we find

$$-\text{Im}\,\langle\langle A_{\vec{k}j};A^{\dagger}_{\vec{k}j}\rangle\rangle=\frac{1}{\pi}\left(\frac{B_{\vec{k}}(\omega)}{\text{Re}D_{\vec{k}j}(\omega)}\right)\frac{\Gamma_{\vec{k}}(\omega)-[\omega^2\text{Re}\epsilon(\vec{k},\omega)-c^2k^2]\hat{\Gamma}_{\vec{k}}(\omega)/B_{\vec{k}}(\omega)}{[\omega^2\text{Re}\epsilon(\vec{k},\omega)-c^2k^2]^2+\Gamma^2_{\vec{k}}(\omega)}, \tag{38}$$

$$-\text{Im}\,\langle\langle A_{\vec{k}\lambda};A^{\dagger}_{\vec{k}\lambda}\rangle\rangle=\left(\frac{ck}{\pi}\right)\frac{\Gamma_{\vec{k}}(\omega)-[\omega^2\text{Re}\epsilon(\vec{k},\omega)-c^2k^2]\,\text{Im}D_{\vec{k}j}(\omega)/\text{Re}D_{\vec{k}j}(\omega)}{[\omega^2\text{Re}\epsilon(\vec{k},\omega)-c^2k^2]^2+\Gamma^2_{\vec{k}}(\omega)}, \tag{39}$$

where

$$\Gamma_{\vec{k}}(\omega)=ck\,\text{Im}P_{21}(\vec{k},\omega)+[\omega^2-c^2k^2-\omega_p^2-ck\,\text{Re}P_{21}(\vec{k},\omega)]\frac{\text{Im}D_{\vec{k}j}(\omega)}{\text{Re}D_{\vec{k}j}(\omega)}+\sum_{j\lambda}X_j^2(\vec{k}\lambda)\omega^0_{\vec{k}j}\frac{\text{Im}P_{43}(\vec{k},\omega)}{\text{Re}D_{\vec{k}j}(\omega)}, \tag{40a}$$

$$B_{\vec{k}}(\omega)=[\omega^0_{\vec{k}j}+\text{Re}P_{34}(\vec{k},\omega)][\omega^2-c^2k^2-\omega_p^2-ck\,\text{Re}P_{21}(\vec{k},\omega)]+\sum_{j\lambda}X_j^2(\vec{k}\lambda)\omega^0_{\vec{k}j}+\text{Im}P_{43}(\vec{k},\omega)\,P_{21}(\vec{k},\omega), \tag{40b}$$

$$\hat{\Gamma}_{\vec{k}}(\omega)=ck[\omega^0_{\vec{k}j}+\text{Re}P_{34}(\vec{k},\omega)]\,\text{Im}P_{21}(\vec{k},\omega)-[\omega^2-c^2k^2-\omega_p^2-ck\,\text{Re}P_{21}(\vec{k},\omega)]\,\text{Im}P_{34}(\vec{k},\omega), \tag{40c}$$

$$\text{Im}D_{\vec{k}j}(\omega)=\omega^0_{\vec{k}j}\,\text{Im}\left(P_{34}(\vec{k},\omega)+P_{43}(\vec{k},\omega)+\frac{\omega}{\omega^0_{\vec{k}j}}[P_{33}(\vec{k},\omega)+P_{44}(\vec{k},\omega)]\right), \tag{40d}$$

$$\text{Re}\epsilon(\vec{k},\omega)=1-\frac{\omega_p^2}{\omega^2}-\frac{1}{\omega^2}\sum_{j\lambda}\frac{X_j^2(\vec{k}\lambda))\,\omega^0_{\vec{k}j}[\omega^0_{\vec{k}j}+\text{Re}P_{43}(\vec{k},\omega)]}{\text{Re}D_{\vec{k}j}(\omega)}-\frac{ck}{\omega^2}\text{Re}P_{21}(\vec{k},\omega). \tag{40e}$$

The phonon shape function (38) is proportional to the expression for the absorption coefficient or to the phonon scattering cross section, while the photon shape function (39) via (18) is proportional to the scattering cross section for the electro-optic effect.[2]

The first term in (38) and (39) describes a Lorentzian line if the frequency dependence of the damping function $\Gamma_{\vec{k}}(\omega)$ is neglected. This will be the case for frequencies ω^2 determined from the solutions of the equation

$$\omega^2\,\text{Re}\epsilon(\vec{k},\omega)-c^2k^2=0. \tag{41}$$

Asymmetries will arise only when the frequency dependence of the functions $\omega^2\text{Re}\epsilon(\vec{k},\omega)$, $B_{\vec{k}}(\omega)$, and $\Gamma_{\vec{k}}(\omega)$ is taken into consideration. The second term in (38) and (39) produces asymmetric broadening to the spectral lines for frequencies ω^2, which do not satisfy Eq. (41). This term disappears if the anharmonic coupling between the electromagnetic and phonon fields is neglected, i.e., when the coupling functions

$$\phi_n(\vec{k}_1\lambda_1, \vec{k}_2 j_2, \ldots) \text{ and } g_n(\vec{k}_1\lambda_1, \vec{k}_2\lambda_2, \vec{k}_3 j_3, \ldots)$$

are discarded. Therefore, expressions (38) and (39) indicate that the spectral lines in question are broad and asymmetric, and the extent of the asymmetric broadening depends not only on the frequency dependence of the functions $\omega^2 \operatorname{Re}\epsilon(\vec{k}, \omega)$, $\Gamma_{\vec{k}}(\omega)$, $\hat{\Gamma}_{\vec{k}}(\omega)$, and $B_{\vec{k}}(\omega)$ but also on the value of the quantities $\hat{\Gamma}_{\vec{k}}(\omega)/\operatorname{Re}D_{\vec{k}j}(\omega)$ and $\operatorname{Im}D_{\vec{k}j}(\omega)/\operatorname{Re}D_{\vec{k}j}(\omega)$. Considering expressions (40), we may deduce that the line shape described by (39) is more asymmetric than that of (38), since $\operatorname{Im}D_{\vec{k}j}(\omega) > \hat{\Gamma}_{\vec{k}j}(\omega)$.

Expressions (38) and (39) are applicable in the whole range of frequencies ω. If we make the expansion

$$\omega^2 \operatorname{Re}\epsilon(\vec{k}, \omega) - c^2k^2 = \lambda_{\vec{k}}^{-1}(\nu_{\vec{k}\rho})(\omega^2 - \nu_{\vec{k}\rho}^2) , \qquad (42)$$

where the frequencies $\nu_{\vec{k}\rho}$ are determined from the ρth solutions of Eq. (41) and

$$\lambda_{\vec{k}}^{-1}(\nu_{\vec{k}\rho}) = \sum_\rho \left(\frac{d[\omega^2 \operatorname{Re}\epsilon(\vec{k}, \omega)]}{d\omega^2} \right)_{\omega=\nu_{\vec{k}\rho}} ,$$

then (38) and (39) may take the approximate form

$$-\operatorname{Im}\langle\langle A_{\vec{k}j}; A_{\vec{k}j}^\dagger \rangle\rangle \approx \frac{1}{\pi}\left(\frac{B_{\vec{k}}(\omega)}{\operatorname{Re}D_{\vec{k}}(\omega)}\,\lambda_{\vec{k}}(\nu_{\vec{k}\rho})\right) \frac{\tilde{\Gamma}_{\vec{k}}(\omega) - (\omega^2 - \nu_{\vec{k}\rho}^2)\hat{\Gamma}_{\vec{k}}(\omega)/B_{\vec{k}}(\omega)}{(\omega^2 - \nu_{\vec{k}\rho}^2)^2 + \tilde{\Gamma}_{\vec{k}}^2(\omega)} , \qquad (43)$$

$$-\operatorname{Im}\langle\langle A_{\vec{k}\lambda}; A_{\vec{k}\lambda}^\dagger \rangle\rangle \approx \left(\frac{ck}{\pi}\,\lambda_{\vec{k}}(\nu_{\vec{k}\rho})\right) \frac{\tilde{\Gamma}_{\vec{k}}(\omega) - (\omega^2 - \nu_{\vec{k}\rho}^2)\operatorname{Im}D_{\vec{k}j}(\omega)/\operatorname{Re}D_{\vec{k}j}(\omega)}{(\omega^2 - \nu_{\vec{k}\rho}^2)^2 + \tilde{\Gamma}_{\vec{k}}^2(\omega)} , \qquad (44)$$

where $\tilde{\Gamma}_{\vec{k}}(\omega) = \lambda_{\vec{k}}(\nu_{\vec{k}\rho})\Gamma_{\vec{k}}(\omega)$. This approximation indicates that the center of the spectral lines is in the neighborhood of frequencies $\nu_{\vec{k}\rho}$ and that in the range of frequencies $\omega \sim \nu_{\vec{k}\rho}$ the function $\omega^2 \operatorname{Re}\epsilon(\vec{k}, \omega)$ varies smoothly with ω and results in screening the damping function $\Gamma_{\vec{k}}(\omega)$ by the factor $\lambda_{\vec{k}}(\nu_{\vec{k}\rho})$.

We now proceed to discuss how to calculate the expressions appearing in Eq. (40). A rigorous calculation of the Green's functions involved in the expressions for the components of the polarization operator $P_{il}(\vec{k}, \omega)$ can be done only by evaluating them by means of the total Hamiltonian (1). Unfortunately, this is an impossible task, as has been discussed in I; therefore, following I, we shall evaluate $P_{il}(\vec{k}, \omega)$ in successive approximations. For example, in the case of polariton-polariton scattering the Green's functions in Eqs. (22) can be transformed in the polariton representation and then the resulting polariton Green's functions can be calculated in the lowest approximation by means of the zeroth-order Hamiltonian (32). The photon and phonon operators $A_{\vec{k}\lambda}$ and $A_{\vec{k}j}$, $B_{\vec{k}j}$ can be transformed into the polariton operators by the relations

$$\mu_{\vec{q}\lambda\rho} = \left(\frac{cq}{\omega_{\vec{q}\rho}}\,\lambda_{\vec{q}}^0(\omega_{\vec{q}\rho})\right)^{1/2}, \quad A_{\vec{q}\lambda} = \mu_{\vec{q}\lambda\rho} A_{\vec{q}\rho} , \qquad (45a)$$

$$\mu_{\vec{q}j\rho} = \left(\frac{\omega_{\vec{q}j}^0}{\omega_{\vec{q}\rho}}\,\overline{\lambda}_{\vec{q}}^0(\omega_{\vec{q}\rho})\right)^{1/2}, \quad A_{\vec{q}j} = i\mu_{\vec{q}j\rho} A_{\vec{q}\rho} , \qquad (45b)$$

$$\overline{\mu}_{\vec{q}j\rho} = \left(\frac{\omega_{\vec{q}\rho}}{\omega_{\vec{q}j}^0}\,\overline{\lambda}_{\vec{q}}^0(\omega_{\vec{q}\rho})\right)^{1/2}, \quad B_{\vec{q}j} = i\overline{\mu}_{\vec{q}j\rho} B_{\vec{q}\rho} , \qquad (45c)$$

where $A_{\vec{q}\rho} = \alpha_{\vec{q}\rho} + \alpha_{-\vec{q}\rho}^\dagger$ and $B_{\vec{q}\rho} = \alpha_{\vec{q}\rho} - \alpha_{-\vec{q}\rho}^\dagger$. Since the maximum value of the transformation coefficients in (45) is unity, which occurs in the absence of dispersion for the mode with wave vector $\vec{q}$ [for the mode in question the first term in the Hamiltonian (4) is equal to zero], we conclude that the effect of dispersion is to screen the bare anharmonic coupling functions, resulting in the reduction of the absolute value of the scattering amplitudes. This is in agreement with the results of I. As an example, we shall now discuss in detail the possible mechanisms that result in the physical process of Raman scattering. The scattering amplitudes will be considered only in the lowest approximation, which is sufficient to describe the physical processes of our interest.

IV. RAMAN SCATTERING

If we take $n = 3$ in expressions (10), we have

$$F_1(\vec{k}\lambda) = -2\sum \phi_3(\vec{k}_1 j_1, \vec{k}_2 j_2, -\vec{k}\lambda)\, B_{\vec{k}_1 j_1} A_{\vec{k}_2 j_2} + 4\sum g_3(\vec{k}_1\lambda_1, \vec{k}_2 j_2, -\vec{k}\lambda)\, A_{\vec{k}_1\lambda_1} A_{\vec{k}_2 j_2} , \qquad (46)$$

$$F_2(\vec{k}j) = 2\sum \phi_3(\vec{k}_1\lambda_1, \vec{k}_2 j_2, -\vec{k}j)\, A_{\vec{k}_1\lambda_1} A_{\vec{k}_2 j_2} , \qquad (47)$$

$$F_3(\vec{k}j) = 2\sum [3V_3(\vec{k}_1 j_1, \vec{k}_2 j_2, -\vec{k}j)\, A_{\vec{k}_1 j_1} A_{\vec{k}_2 j_2} + g_3(\vec{k}_1\lambda_1, \vec{k}_2\lambda_2, -\vec{k}j)\, A_{\vec{k}_1\lambda_1} A_{\vec{k}_2\lambda_2} - \phi_3(\vec{k}_1\lambda_1, \vec{k}_2 j_2, -\vec{k}j)\, A_{\vec{k}_1\lambda_1} B_{\vec{k}_2 j_2}] . \qquad (48)$$

Expressions (46)–(48) indicate that the polariton mode $\vec{k}\rho$ may decay into two modes: one photon and one phonon, two photons and two phonons. A question arises here whether or not these modes are dressed by the electromagnetic field of the medium. If there is a strong dispersion for the polariton mode $\vec{k}\rho$, one may expect that the modes arising from the decay of the polariton mode $\vec{k}\rho$ are dressed, i.e., the polariton $\vec{k}\rho$ decays into two polaritons. In this case the photon and phonon operators in expressions (46)–(48) have to be transformed into the polariton representation via relations (45), and then the resulting two-polariton Green's functions $\langle\langle F_i; F_l^\dagger\rangle\rangle$ can be evaluated in the lowest approximation by means of the zeroth-order renormalized Hamiltonian (32). The quantities that contribute to the expressions for the damping functions $\Gamma_{\vec{k}}^{\rho}(\omega)$ and $\hat{\Gamma}_{\vec{k}}^{\rho}(\omega)$ are calculated in Appendix A. It is shown that the bare anharmonic coupling functions are now screened by the field of the scattered modes, since the larger the dispersion, the smaller the transformation coefficients μ in (45) and, therefore, the smaller the scattering amplitudes of the anharmonic coupling functions. Considering that the damping function $\Gamma_{\vec{k}}^{\rho}(\omega)$ in (43) and (44) is also screened by a factor $\lambda_{\vec{k}}^{\rho}(\nu_{\vec{k}\rho})$, one may conclude that the spectral width arising from the decay of the polariton $\vec{k}\rho$ into two polaritons will be very small and hence difficult to measure. However, in the case of weak dispersion for the mode $\vec{k}\rho$, there is a possibility of a detectable width arising from polariton-polariton scattering.

A more favorable process than the one discussed previously is the physical process where the polariton $\vec{k}\rho$ decays into another polariton $\vec{k}_1\rho_1$ through the emission or absorption of the bare phonon $\vec{k}_2(=\vec{k}-\vec{k}_1)j_2$. Physically, this process indicates that the phonon $\vec{k}_2 j_2$ suffers no dispersion; i.e., the coupling function $\kappa_{j_2}(\vec{k}_2\lambda_2)$ is very small or close to zero and can be neglected, though $\vec{k}_2=\vec{k}-\vec{k}_1$ is in the range of wave vectors in the polariton regime. In this case, the $\vec{k}_1\rho_1$ mode produces screening through (45), while for the bare phonon mode we have

$$A_{\vec{q}j}=\mu_{\vec{q}j}\tilde{A}_{\vec{q}j}, \quad B_{\vec{q}j}=\overline{\mu}_{\vec{q}j}\tilde{B}_{\vec{q}j}, \tag{49}$$

where $\mu_{\vec{q}j}=(\omega_{\vec{q}j}^0/\tilde{\omega}_{\vec{q}j})^{1/2}$, $\overline{\mu}_{\vec{q}j}=1/\mu_{\vec{q}j}$, and $\tilde{\omega}_{\vec{q}j}$ is the renormalized excitation energy for the bare phonon, which is determined from the solutions of Eq. (37d). The coefficients $\mu_{\vec{q}j}$ and $\overline{\mu}_{\vec{q}j}$ deviate from unity mainly at high temperatures. After applying the transformations (45) and (49) into the F's in (46)–(48), the resulting Green's functions $\langle\langle F_i; F_l^\dagger\rangle\rangle$ can be evaluated via the Hamiltonian

$$\mathcal{H}^0=\text{const}+\sum_{\vec{k}\rho}\omega_{\vec{k}\rho}\alpha_{\vec{k}\rho}^\dagger\alpha_{\vec{k}\rho}+\sum_{\vec{q}j}\tilde{\omega}_{\vec{q}j}\tilde{\alpha}_{\vec{q}j}^\dagger\tilde{\alpha}_{\vec{q}j}. \tag{50}$$

The scattering amplitudes for the process in question are given by the Eqs. (B1)–(B7) in Appendix B.

Finally we consider the physical process where the polariton $\vec{k}\rho$ decays into two bare particles. In this case the scattered bare phonons and photons are only renormalized by their own fields to account for small static corrections. The phonon operators are transformed according to relations (49), while for the photon operators we have

$$A_{\vec{k}\lambda}=\mu_{\vec{k}\lambda}\tilde{A}_{\vec{k}\lambda}, \quad \mu_{\vec{k}\lambda}=(ck/\Omega_{\vec{k}\lambda})^{1/2}. \tag{51}$$

Using relations (46)–(49) and (51), the Green's functions $\langle\langle F_i; F_j^\dagger\rangle\rangle$ are calculated in Appendix C by means of the Hamiltonian (37e). The expressions for $M_{\vec{k}j}(\omega)$ given by (C1) consist of three terms. In the range of frequencies, ω corresponding to the energies of the polariton mode $\vec{k}\rho$, the first and second terms in (C1) describe the conversion of the polariton $\vec{k}\rho$ into two phonons and two photons, respectively, while the third term represents the scattering of the polariton mode $\vec{k}\rho$ into one phonon and one photon. Similar processes are described by expressions (C2) and (C3). A comparison between the first term of (C1) and the first term of (C2) indicates that both terms describe similar physical processes and, therefore, are of the same order of magnitude. Considering that

$$\mathrm{Im}M_{\vec{k}j}(\omega)=\mathrm{Im}D_{\vec{k}j}(\omega),$$
$$\mathrm{Im}P_{21}(\vec{k},\omega)=\mathrm{Im}\,\langle\langle F_1(\vec{k}\lambda);\; F_1^\dagger(\vec{k}\lambda)\rangle\rangle\ ,$$

and that the expressions for $\mathrm{Im}\,\langle\langle F_1(\vec{k}\lambda); F_1^\dagger(\vec{k}\lambda)\rangle\rangle$ and $\mathrm{Im}\,\langle\langle F_2(\vec{k}j);\; F_2^\dagger(\vec{k}j)\rangle\rangle$ are responsible for the asymmetric broadening of the spectral lines described by (38) and (39), we conclude that, for the detailed study of the line shapes arising from photon-photon interactions, not only the lattice anharmonicity but also the anharmonic coupling of the electromagnetic field must be taken into consideration. Since the renormalization factors $\mu_{\vec{q}j}$ and $\mu_{\vec{q}\lambda}$ are of the order of unity, the physical process in question is the most favorable.

V. PHOTON-PHONON SCATTERING

We shall now discuss the excitation spectrum for the bare photon and phonon fields described by (26) and (27), respectively. Taking the imaginary parts of (26) and (27), we have

$$-\mathrm{Im}\,\langle\langle A_{\vec{k}\lambda}; A_{\vec{k}\lambda}^\dagger\rangle\rangle_b=\frac{ck}{\pi}\,\frac{ck\,\mathrm{Im}\,P_{21}(\vec{k},\omega)}{[\omega^2-c^2k^2-\omega_p^2-ck\,\mathrm{Re}\,P_{21}(\vec{k},\omega)]^2+[ck\,\mathrm{Im}\,P_{21}(\vec{k},\omega)]^2}\ , \tag{52}$$

$$-\mathrm{Im}\,\langle\langle A_{\vec{k}j}; A_{\vec{k}j}^\dagger\rangle\rangle_b=\frac{1}{\pi}\,\frac{[\omega_{\vec{k}j}^0+\mathrm{Re}\,P_{34}(\vec{k},\omega)]\,\mathrm{Im}\,D_{\vec{k}j}(\omega)+\mathrm{Re}\,D_{\vec{k}j}(\omega)\,\mathrm{Im}\,P_{34}(\vec{k},\omega)}{[\mathrm{Re}\,D_{\vec{k}j}(\omega)]^2+[\mathrm{Im}\,D_{\vec{k}j}(\omega)]^2}\ . \tag{53}$$

The photon spectral function (52) is roughly a Lorentzian line centered at the maximum frequencies ω, which are determined from the solutions of the equation

$$\omega^2-c^2k^2-\omega_p^2-ck\,\mathrm{Re}\,P_{21}(\vec{k},\omega)=0\ , \tag{54}$$

with a linewidth of the order of $\text{Im}\,P_{21}(\vec{k},\omega)$ in energy units, provided that $\text{Im}\,P_{21}(\vec{k},\omega) \ll ck$ and the functions $\text{Re}\,P_{21}(\vec{k},\omega)$ and $\text{Im}\,P_{21}(\vec{k},\omega)$ vary slowly with ω in the region of the maximum frequencies. In the case of Raman scattering, for $n=3$ and for frequencies ω in the neighborhood of the solutions of Eq. (54), the function

$$\text{Im}\,P_{21}(\vec{k},\omega) = \pi\,\text{Im}\langle\langle F_1(\vec{k}\lambda); F_1^\dagger(\vec{k}\lambda)\rangle\rangle$$

is given by the imaginary part of expression (C2). It consists of two terms describing the physical process where the absorbed photon decays into two phonons and into one photon and one phonon, respectively.

The phonon spectral function (53) is an asymmetric Lorentzian line even when the frequency dependence of the functions $\text{Re}\,D_{\vec{k}j}(\omega)$, $\text{Im}\,D_{\vec{k}j}(\omega)$, and $\text{Im}\,P_{34}(\vec{k},\omega)$ is neglected. The asymmetric broadening in the line shape described by function (53) occurs at frequencies ω for which $\text{Re}\,D_{\vec{k}j}(\omega) \neq 0$. It is caused by the damping function

$$\text{Im}\,P_{34}(\vec{k},\omega) = \pi\,\text{Im}\langle\langle F_2(\vec{k}j); F_2^\dagger(\vec{k}j)\rangle\rangle \quad ,$$

which arises from the anharmonic coupling between the electromagnetic field and the phonon field. Considering that the functions $\text{Re}\,D_{\vec{k}j}(\omega)$, $\text{Im}\,D_{\vec{k}j}(\omega)$, and $\text{Im}\,P_{34}(\vec{k},\omega)$ are also varying functions of ω, one should expect the line shape of the function (53) to be asymmetric. For the process of Raman scattering and for frequencies ω in the range of frequencies corresponding to the phonon mode $\vec{k}j$ the expressions for $\text{Im}\,D_{\vec{k}j}(\omega) = \text{Im}\,M_{\vec{k}j}(\omega)$ and $\text{Im}\,P_{34}(\vec{k},\omega)$ are given by the imaginary parts of (C1) and (C3), respectively. They describe processes where the phonon $\vec{k}j$ decays into two phonons or two photons as well as the scattering of the phonon $\vec{k}j$ into another phonon $\vec{k}_2 j_2$ with the emission or absorption of a photon.

VI. CONCLUSION

The present study is concerned with the line shapes arising from photon-phonon interactions in dielectric crystals. General expressions have been developed for the spectral functions of the photon and phonon fields, respectively. It is shown that the line shapes for the fields in question exhibit asymmetric broadening. The asymmetry is mainly due to the anharmonic coupling between the electromagnetic and the phonon fields. Additional asymmetries will arise when consideration is given to the frequency dependence of the energy shift and the damping functions as well as the variation of the shape function with respect to the scattering angle.[2]

Raman scattering processes have been discussed in detail. The spectral functions (38) and (39) can be used in the same spirit to study higher-order processes than for $n=3$. In the present study, no quantitative estimates have been made for the coupling functions causing the asymmetric broadening of the spectral lines. The extent of the proposed mechanism depends on the crystal structure, the anharmonicity of the lattice, the temperature, and the strength of the external electromagnetic field. Explicit expressions for the anharmonic coupling functions are given in I, but only numerical calculations made on real crystals will reveal the importance of the proposed effect. To our knowledge, there is nothing known in the literature about the anharmonic coupling functions ϕ_n and g_n. Numerical computations of the results derived in the present study and comparison with the observed data are highly desirable.

APPENDIX A

We shall calculate here the Green's functions (22) for the physical process of resonance Raman scattering where the polariton $\vec{k}\rho$ decays into two polaritons. We need to calculate the following expressions:

$$M_{\vec{k}j}(\omega) = \pi\omega^0_{\vec{k}j}\{\langle\langle F_3(\vec{k}j); F_3^\dagger(\vec{k}j)\rangle\rangle + \langle\langle F_2(\vec{k}j); F_2^\dagger(\vec{k}j)\rangle\rangle + (\omega/\omega^0_{\vec{k}j})[\langle\langle F_2(\vec{k}j); F_3^\dagger(\vec{k}j)\rangle\rangle + \langle\langle F_3(\vec{k}j); F_2^\dagger(\vec{k}j)\rangle\rangle]\}\,, \quad \text{(A1)}$$

$\langle\langle F_1(\vec{k}\lambda);\ F_1^\dagger(\vec{k}\lambda)\rangle\rangle$, $\langle\langle F_2(\vec{k}j);\ F_2^\dagger(\vec{k}j)\rangle\rangle$, and $\langle\langle F_3(\vec{k}j);\ F_3^\dagger(\vec{k}j)\rangle\rangle$. For $n=3$ the expressions for $F_1(\vec{k}\lambda)$, $F_2(\vec{k}j)$, and $F_3(\vec{k}j)$ are given by Eqs. (46)–(48). Using (45), we transform the F's into the polariton representation, and then, substituting the resulting expressions into (A1), we evaluate the two-polariton Green's functions by means of the Hamiltonian (32). We find

$$M_{\vec{k}j}(\omega) = 2\omega^0_{\vec{k}j}\sum\Big([\Delta_+(\vec{k}_1,\vec{k}_2,-\vec{k},\omega)(\omega_{\vec{k}_1\rho_1}+\omega_{\vec{k}_2\rho_2}) - 2\omega\hat{\Delta}_+(\vec{k}_1,\vec{k}_2,-\vec{k},\omega)]\,\frac{\eta_{\vec{k}_1\rho_1}+\eta_{\vec{k}_2\rho_2}}{\omega^2-(\omega_{\vec{k}_1\rho_1}+\omega_{\vec{k}_2\rho_2})^2}$$
$$+[\Delta_-(\vec{k}_1,\vec{k}_2,-\vec{k},\omega)(\omega_{\vec{k}_1\rho_1}-\omega_{\vec{k}_2\rho_2}) - 2\omega\hat{\Delta}_-(\vec{k}_1,\vec{k}_2,-\vec{k},\omega)]\,\frac{\eta_{\vec{k}_2\rho_2}-\eta_{\vec{k}_1\rho_1}}{\omega^2-(\omega_{\vec{k}_1\rho_1}-\omega_{\vec{k}_2\rho_2})^2}\Big) \quad , \quad \text{(A2)}$$

where

$$\Delta_\pm(\vec{k}_1,\vec{k}_2,-\vec{k},\omega) = |\,\phi_3(\vec{k}_1\lambda_1,\vec{k}_2j_2,-\vec{k}j)\,\mu_{\vec{k}_1\lambda_1\rho_1}|^2(\mu^2_{\vec{k}_2j_2\rho_2}+\bar{\mu}^2_{\vec{k}_2j_2\rho_2})$$

$$+\phi_3(\vec{k}_1\lambda_1,\vec{k}_2 j_2,-\vec{k}j)\,\phi_3^*(\vec{k}_2 j_2,\vec{k}_1\lambda_1,-\vec{k}j)\,\mu_{\vec{k}_1\lambda_1\rho_1}\mu_{\vec{k}_2\lambda_2\rho_2}(\mu_{\vec{k}_1 j_1\rho_1}\mu_{\vec{k}_2 j_2\rho_2}\pm\overline{\mu}_{\vec{k}_1 j_1\rho_1}\overline{\mu}_{\vec{k}_2 j_2\rho_2})$$

$$+2|\overline{V}_3(\vec{k}_1,\vec{k}_2,-\vec{k})|^2+(\omega/\omega^0_{\vec{k}j})\overline{V}_3(\vec{k}_1,\vec{k}_2,-\vec{k})[\phi_3(\vec{k}_1\lambda_1,\vec{k}_2 j_2,-\vec{k}j)\mu_{\vec{k}_1\lambda_1\rho_1}\mu_{\vec{k}_2 j_2\rho_2}+\phi_3(\vec{k}_2 j_2,\vec{k}_1\lambda_1,-\vec{k}j)\mu_{\vec{k}_2\lambda_2\rho_2}\mu_{\vec{k}_1 j_1\rho_1}], \quad \text{(A3a)}$$

$$\overline{V}_3(\vec{k}_1,\vec{k}_2,-\vec{k})=3V_3(\vec{k}_1 j_1,\vec{k}_2 j_2,-\vec{k}j)\mu_{\vec{k}_1 j_1\rho_1}\mu_{\vec{k}_2 j_2\rho_2}+g_3(\vec{k}_1\lambda_1,\vec{k}_2\lambda_2,-\vec{k}j)\mu_{\vec{k}_1\lambda_1\rho_1}\mu_{\vec{k}_2\lambda_2\rho_2}, \quad \text{(A3b)}$$

$$\hat{\Delta}_\pm(\vec{k}_1,\vec{k}_2,-\vec{k},\omega)=\overline{V}_3(\vec{k}_1,\vec{k}_2,-\vec{k})[\phi_3(\vec{k}_1\lambda_1,\vec{k}_2 j_2,-\vec{k}j)\mu_{\vec{k}_1\lambda_1\rho_1}\mu_{\vec{k}_2 j_2\rho_2}$$
$$\pm\phi_3(\vec{k}_2 j_2,\vec{k}_1\lambda_1,-\vec{k}j)\mu_{\vec{k}_2\lambda_2\rho_2}\mu_{\vec{k}_1 j_1\rho_1}]+(\omega/\omega^0_{\vec{k}j})[|\phi_3(\vec{k}_1\lambda_1,\vec{k}_2 j_2-\vec{k}j)\mu_{\vec{k}_1\lambda_1\rho_1}\mu_{\vec{k}_2 j_2\rho_2}\overline{\mu}_{\vec{k}_2 j_2\rho_2}|^2$$
$$\pm\tfrac{1}{2}\phi_3(\vec{k}_1\lambda_1,\vec{k}_2 j_2,-\vec{k}j)\phi_3^*(\vec{k}_2 j_2,\vec{k}_1\lambda_1,-\vec{k}j)\mu_{\vec{k}_1\lambda_1\rho_1}\mu_{\vec{k}_2 j_2\rho_2}(\overline{\mu}_{\vec{k}_1 j_1\rho_1}\mu_{\vec{k}_2 j_2\rho_2}+\mu_{\vec{k}_1 j_1\rho_1}\overline{\mu}_{\vec{k}_2 j_2\rho_2})]\ . \quad \text{(A3c)}$$

Similarly,

$$\pi\langle\langle F_1(\vec{k}\lambda);F_1^\dagger(\vec{k}\lambda)\rangle\rangle=2\sum\Bigg([L_+(\vec{k}_1,\vec{k}_2,-\vec{k})(\omega_{\vec{k}_1\rho_1}+\omega_{\vec{k}_2\rho_2})-2\omega\hat{L}_+(\vec{k}_1,\vec{k}_2,-\vec{k})]\frac{\eta_{\vec{k}_1\rho_1}+\eta_{\vec{k}_2\rho_2}}{\omega^2-(\omega_{\vec{k}_1\rho_1}+\omega_{\vec{k}_2\rho_2})^2}$$
$$+[L_-(\vec{k}_1,\vec{k}_2,-\vec{k})(\omega_{\vec{k}_1\rho_1}-\omega_{\vec{k}_2\rho_2})-2\omega\hat{L}_-(\vec{k}_1,\vec{k}_2,-\vec{k})]\frac{\eta_{\vec{k}_1\rho_1}-\eta_{\vec{k}_2\rho_2}}{\omega^2-(\omega_{\vec{k}_1\rho_1}-\omega_{\vec{k}_2\rho_2})^2}\Bigg), \quad \text{(A4)}$$

$$L_\pm(\vec{k}_1,\vec{k}_2,-\vec{k})=|\phi_3(\vec{k}_1 j_1,\vec{k}_2 j_2,-\vec{k}\lambda)\overline{\mu}_{\vec{k}_1 j_1\rho_1}\mu_{\vec{k}_2 j_2\rho_2}|^2+|2g_3(\vec{k}_1\lambda_1,\vec{k}_2 j_2,-\vec{k}\lambda)\mu_{\vec{k}_1\lambda_1\rho_1}\mu_{\vec{k}_2 j_2\rho_2}|^2$$
$$\pm\phi_3(\vec{k}_1 j_1,\vec{k}_2 j_2,-\vec{k}\lambda)\phi_3^*(\vec{k}_2 j_2,\vec{k}_1 j_1,-\vec{k}\lambda)\overline{\mu}_{\vec{k}_1 j_1\rho_1}\mu_{\vec{k}_1 j_1\rho_1}\overline{\mu}_{\vec{k}_2 j_2\rho_2}\mu_{\vec{k}_2 j_2\rho_2}$$
$$\pm4g_3(\vec{k}_1\lambda_1,\vec{k}_2 j_2,-\vec{k}\lambda)g_3^*(\vec{k}_2 j_2,\vec{k}_1\lambda_1,-\vec{k}\lambda)\mu_{\vec{k}_1\lambda_1\rho_1}\mu_{\vec{k}_1 j_1\rho_1}\mu_{\vec{k}_2\lambda_2\rho_2}\mu_{\vec{k}_2 j_2\rho_2}, \quad \text{(A5a)}$$

$$\hat{L}_\pm(\vec{k}_1,\vec{k}_2,-\vec{k})=2\phi_3(\vec{k}_1 j_1,\vec{k}_2 j_2,-\vec{k}\lambda)g_3^*(\vec{k}_1\lambda_1,\vec{k}_2 j_2,-\vec{k}\lambda)\overline{\mu}_{\vec{k}_1 j_1\rho_1}\mu_{\vec{k}_1\lambda_1\rho_1}\mu^2_{\vec{k}_2 j_2\rho_2}$$
$$\pm\phi_3(\vec{k}_1 j_1,\vec{k}_2 j_2,-\vec{k}\lambda)g_3^*(\vec{k}_2 j_2,\vec{k}_1\lambda_1,-\vec{k}\lambda)\mu_{\vec{k}_2 j_2\rho_2}\mu_{\vec{k}_1 j_1\rho_1}(\overline{\mu}_{\vec{k}_1 j_1\rho_1}\mu_{\vec{k}_2\lambda_2\rho_2}+\overline{\mu}_{\vec{k}_2 j_2\rho_2}\mu_{\vec{k}_1\lambda_1\rho_1}), \quad \text{(A5b)}$$

$$\pi\langle\langle F_2(\vec{k}j);F_2^\dagger(\vec{k}j)\rangle\rangle=2\sum[|\phi_3(\vec{k}_1\lambda_1,\vec{k}_2 j_2,-\vec{k}j)\mu_{\vec{k}_1\lambda_1\rho_1}\mu_{\vec{k}_2 j_2\rho_2}|^2$$
$$+\phi_3(\vec{k}_1\lambda_1,\vec{k}_2 j_2,-\vec{k}j)\phi_3^*(\vec{k}_2 j_2,\vec{k}_1\lambda_1,-\vec{k}j)\mu_{\vec{k}_1\lambda_1\rho_1}\mu_{\vec{k}_2\lambda_2\rho_2}\mu_{\vec{k}_1 j_1\rho_1}\mu_{\vec{k}_2 j_2\rho_2}]$$
$$\times\Bigg((\eta_{\vec{k}_1\rho_1}+\eta_{\vec{k}_2\rho_2})\frac{\omega_{\vec{k}_1\rho_1}+\omega_{\vec{k}_2\rho_2}}{\omega^2-(\omega_{\vec{k}_1\rho_1}+\omega_{\vec{k}_2\rho_2})^2}+(\eta_{\vec{k}_2\rho_2}-\eta_{\vec{k}_1\rho_1})\frac{\omega_{\vec{k}_1\rho_1}-\omega_{\vec{k}_2\rho_2}}{\omega^2-(\omega_{\vec{k}_1\rho_1}-\omega_{\vec{k}_2\rho_2})^2}\Bigg), \quad \text{(A6)}$$

$$\pi\langle\langle F_3(\vec{k}j);F_3^\dagger(\vec{k}j)\rangle\rangle=2\sum\Bigg([S_+(\vec{k}_1,\vec{k}_2,-\vec{k})(\omega_{\vec{k}_1\rho_1}+\omega_{\vec{k}_2\rho_2})-2\omega\hat{S}_+(\vec{k}_1,\vec{k}_2,-\vec{k})]\frac{\eta_{\vec{k}_1\rho_1}+\eta_{\vec{k}_2\rho_2}}{\omega^2-(\omega_{\vec{k}_1\rho_1}+\omega_{\vec{k}_2\rho_2})^2}$$
$$+[S_-(\vec{k}_1,\vec{k}_2,-\vec{k})(\omega_{\vec{k}_1\rho_1}-\omega_{\vec{k}_2\rho_2})-2\omega\hat{S}_-(\vec{k}_1,\vec{k}_2,-\vec{k})]\frac{\eta_{\vec{k}_1\rho_1}-\eta_{\vec{k}_2\rho_2}}{\omega^2-(\omega_{\vec{k}_1\rho_1}-\omega_{\vec{k}_2\rho_2})^2}\Bigg), \quad \text{(A7)}$$

$$S_\pm(\vec{k}_1,\vec{k}_2,-\vec{k})=2|\overline{V}_3(\vec{k}_1,\vec{k}_2,-\vec{k})|^2+|\phi_3(\vec{k}_1\lambda_1,\vec{k}_2 j_2,-\vec{k})\mu_{\vec{k}_1\lambda_1\rho_1}\overline{\mu}_{\vec{k}_2 j_2\rho_2}|^2$$
$$\pm\phi_3(\vec{k}_1\lambda_1,\vec{k}_2 j_2,-\vec{k}j)\phi_3^*(\vec{k}_2 j_2,\vec{k}_1\lambda_1,-\vec{k}j)\mu_{\vec{k}_1\lambda_1\rho_1}\mu_{\vec{k}_2\lambda_2\rho_2}\overline{\mu}_{\vec{k}_1 j_1\rho_1}\overline{\mu}_{\vec{k}_2 j_2\rho_2}, \quad \text{(A8a)}$$

$$\hat{S}_\pm(\vec{k}_1,\vec{k}_2,-\vec{k})=\overline{V}_3(\vec{k}_1,\vec{k}_2,-\vec{k})[\phi_3(\vec{k}_1\lambda_1,\vec{k}_2 j_2,-\vec{k}j)\mu_{\vec{k}_1\lambda_1\rho_1}\overline{\mu}_{\vec{k}_2 j_2\rho_2}\pm\phi_3(\vec{k}_2 j_2,\vec{k}_1\lambda_1,-\vec{k}j)\mu_{\vec{k}_2 j_2\rho_2}\overline{\mu}_{\vec{k}_1 j_1\rho_1}]\ . \quad \text{(A8b)}$$

Taking the real and imaginary parts of expressions (A2)–(A7), we have

$$\mathrm{Im}M_{\vec{k}j}(\omega)=\mathrm{Im}D_{\vec{k}j}(\omega)=\pi\omega^0_{\vec{k}j}\sum\Big(\{\Delta_+(\vec{k}_1,\vec{k}_2,-\vec{k},\omega)[\delta(\omega+\omega_{\vec{k}_1\rho_1}+\omega_{\vec{k}_2\rho_2})-\delta(\omega-\omega_{\vec{k}_1\rho_1}-\omega_{\vec{k}_2\rho_2})]$$
$$+2\hat{\Delta}_+(\vec{k}_1,\vec{k}_2,-\vec{k},\omega)[\delta(\omega+\omega_{\vec{k}_1\rho_1}+\omega_{\vec{k}_2\rho_2})+\delta(\omega-\omega_{\vec{k}_1\rho_1}-\omega_{\vec{k}_2\rho_2})]\}(\eta_{\vec{k}_1\rho_1}+\eta_{\vec{k}_2\rho_2})$$
$$+\{\Delta_-(\vec{k}_1,\vec{k}_2,-\vec{k},\omega)[\delta(\omega+\omega_{\vec{k}_1\rho_1}-\omega_{\vec{k}_2\rho_2})-\delta(\omega-\omega_{\vec{k}_1\rho_1}+\omega_{\vec{k}_2\rho_2})]$$
$$+2\hat{\Delta}_-(\vec{k}_1,\vec{k}_2,-\vec{k},\omega)[\delta(\omega+\omega_{\vec{k}_1\rho_1}-\omega_{\vec{k}_2\rho_2})+\delta(\omega-\omega_{\vec{k}_1\rho_1}+\omega_{\vec{k}_2\rho_2})]\}(\eta_{\vec{k}_2\rho_2}-\eta_{\vec{k}_1\rho_1})\Big), \quad \text{(A9)}$$

$$\mathrm{Im}P_{34}(\vec{k},\omega)=\pi\,\mathrm{Im}\langle\langle F_2(\vec{k}j);F_2^\dagger(\vec{k}j)\rangle\rangle=\sum\big[\,|\phi_3(\vec{k}_1\lambda_1,\vec{k}_2j_2,-\vec{k}j)\mu_{\vec{k}_1\lambda_1\rho_1}\mu_{\vec{k}_2j_2\rho_2}|^2+\phi_3(\vec{k}_1\lambda_1,\vec{k}_2j_2,-\vec{k}j)$$
$$\times\phi_3^*(\vec{k}_2j_2,\vec{k}_1\lambda_1,-\vec{k}j)\mu_{\vec{k}_1\lambda_1\rho_1}\mu_{\vec{k}_2\lambda_2\rho_2}\mu_{\vec{k}_1j_1\rho_1}\mu_{\vec{k}_2j_2\rho_2}\big]\{(\eta_{\vec{k}_1\rho_1}+\eta_{\vec{k}_2\rho_2})[\delta(\omega+\omega_{\vec{k}_1\rho_1}+\omega_{\vec{k}_2\rho_2})-\delta(\omega-\omega_{\vec{k}_1\rho_1}-\omega_{\vec{k}_2\rho_2})]$$
$$+(\eta_{\vec{k}_2\rho_2}-\eta_{\vec{k}_1\rho_1})[\delta(\omega+\omega_{\vec{k}_1\rho_1}-\omega_{\vec{k}_2\rho_2})-\delta(\omega-\omega_{\vec{k}_1\rho_1}+\omega_{\vec{k}_2\rho_2})]\}\ . \qquad \text{(A10)}$$

In view of (A2), (A4), and (A7), the expressions for

$$\mathrm{Im}P_{21}(\vec{k},\omega)=\pi\,\mathrm{Im}\langle\langle F_1(\vec{k}\lambda);F_1^\dagger(\vec{k}\lambda)\rangle\rangle \quad\text{and}\quad \mathrm{Im}P_{43}(\vec{k},\omega)=\pi\,\mathrm{Im}\langle\langle F_3(\vec{k}j);F_3^\dagger(\vec{k}j)\rangle\rangle$$

can be obtained from Eq. (A9) if we make the replacement of the functions $\omega^0_{\vec{k}j}\Delta_\pm(\vec{k}_1,\vec{k}_2,-\vec{k},\omega)$ and $\omega^0_{\vec{k}j}\hat{\Delta}_\pm(\vec{k}_1,\vec{k}_2,-\vec{k},\omega)$ by $L_\pm(\vec{k}_1,\vec{k}_2,-\vec{k})$, $\hat{L}_\pm(\vec{k}_1,\vec{k}_2,-\vec{k})$ and $S_\pm(\vec{k}_1,\vec{k}_2,-\vec{k})$, $\hat{S}_\pm(\vec{k}_1,\vec{k}_2,-\vec{k})$, respectively.

APPENDIX B

We consider the physical process where the polariton $\vec{k}\rho$ decays into the polariton $\vec{k}_1\rho_1$ with the emission or absorption of the phonon $\vec{k}_2j_2$. In this case expression (A9) is applicable if we replace $\omega_{\vec{k}_2\rho_2}$ and $\eta_{\vec{k}_2\rho_2}$ by $\tilde{\omega}_{\vec{k}_2j_2}$ and $\tilde{\eta}_{\vec{k}_2j_2}$, respectively, as well as the coupling functions $\Delta_\pm$ and $\hat{\Delta}_\pm$ by the expressions

$$\Delta_+=\Delta_-=|\phi_3(\vec{k}_1\lambda_1,\vec{k}_2j_2,-\vec{k}j)\mu_{\vec{k}_1\lambda_1\rho_1}|^2(\mu^2_{\vec{k}_2j_2}+\bar{\mu}^2_{\vec{k}_2j_2})+|3V_3(\vec{k}_1j_1,\vec{k}_2j_2,-\vec{k}j)\mu_{\vec{k}_1j_1}\mu_{\vec{k}_2j_2}|^2$$
$$+3(\omega/\omega^0_{\vec{k}j})V_3(\vec{k}_1j_1,\vec{k}_2j_2,-\vec{k}j)\phi_3^*(\vec{k}_1\lambda_1,\vec{k}_2j_2,-\vec{k}j)\mu_{\vec{k}_1j_1}\mu_{\vec{k}_1\lambda_1\rho_1}\mu^2_{\vec{k}_2j_2}\ , \qquad \text{(B1)}$$

$$\hat{\Delta}_+=\hat{\Delta}_-=(\omega/\omega^0_{\vec{k}j})|\phi_3(\vec{k}_1\lambda_1,\vec{k}_2j_2,-\vec{k}j)\mu_{\vec{k}_1\lambda_1\rho_1}\mu_{\vec{k}_2j_2}\bar{\mu}_{\vec{k}_2j_2}|^2$$
$$+3V_3(\vec{k}_1j_1,\vec{k}_2j_2,-\vec{k}j)\phi_3^*(\vec{k}_1\lambda_1,\vec{k}_2j_2,-\vec{k}j)\mu_{\vec{k}_1j_1}\mu_{\vec{k}_1\lambda_1j_1}\mu^2_{\vec{k}_2j_2}\ . \qquad \text{(B2)}$$

Similarly, the coupling functions $L_\pm$, $\hat{L}_\pm$, $S_\pm$, $\hat{S}_\pm$, and $\mathrm{Im}P_{34}(\vec{k},\omega)$ take the form

$$L_+=L_-=|\phi_3(\vec{k}_1j_1,\vec{k}_2j_2,-\vec{k}\lambda)\bar{\mu}_{\vec{k}_1j_1}\mu_{\vec{k}_2j_2}|^2+|2g_3(\vec{k}_1\lambda_1,\vec{k}_2j_2-\vec{k}\lambda)\mu_{\vec{k}_1\lambda_1\rho_1}\mu_{\vec{k}_2j_2}|^2\ , \qquad \text{(B3)}$$

$$\hat{L}_+=\hat{L}_-=2\phi_3(\vec{k}_1j_1,\vec{k}_2j_2,-\vec{k}\lambda)g_3^*(\vec{k}_1\lambda_1,\vec{k}_2j_2,-\vec{k}\lambda)\bar{\mu}_{\vec{k}_1j_1}\mu_{\vec{k}_1\lambda_1\rho_1}\mu^2_{\vec{k}_2j_2}\ , \qquad \text{(B4)}$$

$$S_+=S_-=|3V_3(\vec{k}_1j_1,\vec{k}_2j_2,-\vec{k}j)\mu_{\vec{k}_1j_1}\mu_{\vec{k}_2j_2}|^2+|\phi_3(\vec{k}_1\lambda_1,\vec{k}_2j_2,-\vec{k}j)\mu_{\vec{k}_1\lambda_1\rho_1}\bar{\mu}_{\vec{k}_2j_2}|^2\ , \qquad \text{(B5)}$$

$$\hat{S}_+=\hat{S}_-=3V_3(\vec{k}_1j_1,\vec{k}_2j_2,-\vec{k}j)\phi_3^*(\vec{k}_1\lambda_1,\vec{k}_2j_2,-\vec{k}j)\mu_{\vec{k}_1\lambda_1\rho_1}\mu_{\vec{k}_1j_1}\mu_{\vec{k}_2j_2}\bar{\mu}_{\vec{k}_2j_2}\ , \qquad \text{(B6)}$$

$$\mathrm{Im}P_{34}(\vec{k},\omega)=\sum|\phi_3(\vec{k}_1\lambda_1,\vec{k}_2j_2,-\vec{k}j)\mu_{\vec{k}_1\lambda_1\rho_1}\mu_{\vec{k}_2j_2}|^2$$
$$\times\{(\eta_{\vec{k}_1\rho_1}+\tilde{\eta}_{\vec{k}_2j_2})[\delta(\omega+\omega_{\vec{k}_1\rho_1}+\tilde{\omega}_{\vec{k}_2j_2})-\delta(\omega-\omega_{\vec{k}_1\rho_1}-\tilde{\omega}_{\vec{k}_2j_2})]$$
$$+(\tilde{\eta}_{\vec{k}_2j_2}-\eta_{\vec{k}_1\rho_1})[\delta(\omega+\omega_{\vec{k}_1\rho_1}-\tilde{\omega}_{\vec{k}_2j_2})-\delta(\omega-\omega_{\vec{k}_1\rho_1}+\tilde{\omega}_{\vec{k}_2j_2})]\}\ . \qquad \text{(B7)}$$

APPENDIX C

Using (49), (51), and (46)-(48), we evaluate the Green's functions that appear in the expression for $M_{\vec{k}j}(\omega)$ given by (A1) by means of the Hamiltonian (37e). We find

$$M_{\vec{k}j}(\omega)=2\pi\omega^0_{\vec{k}j}\sum\{|3V_3(\vec{k}_1j_1,\vec{k}_2j_2,-\vec{k}j)\mu_{\vec{k}_1j_1}\mu_{\vec{k}_2j_2}|^2$$
$$\times G_+(\vec{k}_1j_1,\vec{k}_2j_2,\omega)+|g_3(\vec{k}_1\lambda_1,\vec{k}_2\lambda_2,-\vec{k}j)\mu_{\vec{k}_1\lambda_1}\mu_{\vec{k}_2\lambda_2}|^2G_+(\vec{k}_1\lambda_1,\vec{k}_2\lambda_2,\omega)$$
$$+\tfrac{1}{2}|\phi_3(\vec{k}_1\lambda_1,\vec{k}_2j_2,-\vec{k}j)\mu_{\vec{k}_1\lambda_1}|^2[(\mu^2_{\vec{k}_2j_2}+\bar{\mu}^2_{\vec{k}_2j_2})G_+(\vec{k}_1\lambda_1,\vec{k}_2j_2,\omega)+2(\omega/\omega^0_{\vec{k}j})\hat{G}(\vec{k}_1\lambda_1,\vec{k}_2j_2,\omega)]\}, \qquad \text{(C1)}$$

$$\pi\langle\langle F_1(\vec{k}\lambda);F_1^\dagger(\vec{k}\lambda)\rangle\rangle=\pi\sum\{|\phi_3(\vec{k}_1j_1,\vec{k}_2j_2,-\vec{k}\lambda)|^2[\bar{\mu}^2_{\vec{k}_1j_1}\mu^2_{\vec{k}_2j_2}G_+(\vec{k}_1j_1,\vec{k}_2j_2,\omega)+G_-(\vec{k}_1j_1,\vec{k}_2j_2,\omega)]$$
$$+|2g_3(\vec{k}_1\lambda_1,\vec{k}_2j_2,-\vec{k}\lambda)\mu_{\vec{k}_2j_2}\mu_{\vec{k}_1\lambda_1}|^2G_+(\vec{k}_1\lambda_1,\vec{k}_2j_2,\omega)\}, \qquad \text{(C2)}$$

$$\pi\langle\langle F_2(\vec{k}j);F_2^\dagger(\vec{k}j)\rangle\rangle=\pi\sum|\phi_3(\vec{k}_1\lambda_1,\vec{k}_2j_2,-\vec{k}\lambda)\mu_{\vec{k}_2j_2}\mu_{\vec{k}_1\lambda_1}|^2G_+(\vec{k}_1\lambda_1,\vec{k}_2j_2,\omega), \qquad \text{(C3)}$$

where

$$G_\pm(\vec{k}_1 j_1, \vec{k}_2 j_2, \omega) = \frac{2}{\pi}(\tilde{\eta}_{\vec{k}_1 j_1} + \tilde{\eta}_{\vec{k}_2 j_2}) \frac{\tilde{\omega}_{\vec{k}_1 j_1} + \tilde{\omega}_{\vec{k}_2 j_2}}{\omega^2 - (\tilde{\omega}_{\vec{k}_1 j_1} + \tilde{\omega}_{\vec{k}_2 j_2})^2} \pm (\tilde{\eta}_{\vec{k}_2 j_2} - \tilde{\eta}_{\vec{k}_1 j_1}) \frac{\tilde{\omega}_{\vec{k}_1 j_1} - \tilde{\omega}_{\vec{k}_2 j_2}}{\omega^2 - (\tilde{\omega}_{\vec{k}_1 j_1} - \tilde{\omega}_{\vec{k}_2 j_2})^2}, \tag{C4}$$

$$\hat{G}(\vec{k}_1\lambda_1, \vec{k}_2 j_2, \omega) = \frac{-2\omega}{\pi}\left(\frac{\eta_{\vec{k}_1\lambda_1} + \tilde{\eta}_{\vec{k}_2 j_2}}{\omega^2 - (\Omega_{\vec{k}_1\lambda_1} + \omega_{\vec{k}_2 j_2})^2} + \frac{\tilde{\eta}_{\vec{k}_2 j_2} - \eta_{\vec{k}_1\lambda_1}}{\omega^2 - (\Omega_{\vec{k}_1\lambda_1} - \tilde{\omega}_{\vec{k}_2 j_2})^2}\right). \tag{C5}$$

The functions $G_+(\vec{k}_1\lambda_1, \vec{k}_2\lambda_2, \omega)$ and $G_+(\vec{k}_1\lambda_1, \vec{k}_2 j_2, \omega)$ can be derived from $G_+(\vec{k}_1 j_1, \vec{k}_2 j_2, \omega)$ given by (C4) if we replace $\tilde{\eta}_{\vec{k}_1 j_1}$, $\tilde{\eta}_{\vec{k}_2 j_2}$, $\tilde{\omega}_{\vec{k}_1 j_1}$, $\tilde{\omega}_{\vec{k}_2 j_2}$, and $\tilde{\eta}_{\vec{k}_1 j_1}$, $\tilde{\omega}_{\vec{k}_1 j_1}$ by $\eta_{\vec{k}_1\lambda_1}$, $\eta_{\vec{k}_2\lambda_2}$, $\Omega_{\vec{k}_1\lambda_1}$, $\Omega_{\vec{k}_2\lambda_2}$, and $\eta_{\vec{k}_1\lambda_1}$, $\Omega_{\vec{k}_1\lambda_1}$, respectively. The function $\pi\omega^0_{\vec{k}j}\langle\langle F_3(\vec{k}j); F_3^\dagger(\vec{k}j)\rangle\rangle$ can be obtained from (C1) if we take $\mu^2_{\vec{k}_2 j_2}$ and $\hat{G}(\vec{k}_1\lambda_1, \vec{k}_2 j_2, \omega)$ in the last term of (C1) equal to zero.

The real and imaginary parts of expressions (C1)–(C3) are obtained by replacing the functions $G_\pm(\vec{k}_1 j_1, \vec{k}_2 j_2, \omega)$, $G_+(\vec{k}_1\lambda_1, \vec{k}_2\lambda_2, \omega)$, $G_\pm(\vec{k}_1\lambda_1, \vec{k}_2 j_2, \omega)$, and $\hat{G}(\vec{k}_1\lambda_1, \vec{k}_2 j_2, \omega)$ by their real and imaginary parts, respectively. The real parts of the G's are given by the principal values of (C4) and (C5), while the imaginary parts are given by

$$\mathrm{Im}\,G_\pm(\vec{k}_1 j_1, \vec{k}_2 j_2, \omega) = (\tilde{\eta}_{\vec{k}_1 j_1} + \tilde{\eta}_{\vec{k}_2 j_2})[\delta(\omega + \tilde{\omega}_{\vec{k}_1 j_1} + \tilde{\omega}_{\vec{k}_2 j_2}) - \delta(\omega - \omega_{\vec{k}_1 j_1} - \omega_{\vec{k}_2 j_2})]$$
$$\pm (\tilde{\eta}_{\vec{k}_2 j_2} - \tilde{\eta}_{\vec{k}_1 j_1})[\delta(\omega + \omega_{\vec{k}_1 j_1} - \omega_{\vec{k}_2 j_2}) - \delta(\omega - \tilde{\omega}_{\vec{k}_1 j_1} + \tilde{\omega}_{\vec{k}_2 j_2})], \tag{C6}$$

$$\mathrm{Im}\,\hat{G}(\vec{k}_1 j_1, \vec{k}_2 j_2, \omega) = (\tilde{\eta}_{\vec{k}_1 j_1} + \tilde{\eta}_{\vec{k}_2 j_2})[\delta(\omega + \tilde{\omega}_{\vec{k}_1 j_1} + \tilde{\omega}_{\vec{k}_2 j_2}) + \delta(\omega - \tilde{\omega}_{\vec{k}_1 j_1} - \tilde{\omega}_{\vec{k}_2 j_2})]$$
$$+ (\tilde{\eta}_{\vec{k}_2 j_2} - \tilde{\eta}_{\vec{k}_1 j_1})[\delta(\omega + \tilde{\omega}_{\vec{k}_1 j_1} - \tilde{\omega}_{\vec{k}_2 j_2}) + \delta(\omega - \tilde{\omega}_{\vec{k}_1 j_1} + \tilde{\omega}_{\vec{k}_2 j_2})]. \tag{C7}$$

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*Issued as NRC Report No. 11788.

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