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Excitons, Plasmons, and Polaritons in Insulating Crystals*

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In this paper, a quantum-mechanical model of interacting radiation and nonmetallic matter is used to study the connection between excitons, plasmons, and polaritons. An explicit description of polaritons as mixed particles consisting of photons and matter oscillators is given, and it is shown that polariton states form a suitable basis for calculations of nonlinear optical effects in crystals. Plasmon-photon interactions, in addition, are shown to occur in the crystal.

INTRODUCTION

Since intense laser-light sources became available, nonlinear optical effects[1] such as Brillouin and Raman scattering, two-photon absorption, and harmonic generation have been observed, and the interaction between strong electromagnetic waves and condensed media has become a subject of great interest in solid-state physics.

It is well known in nonlinear optics that the method of small perturbations, successfully used for gases by Armstrong, Bloembergen, Ducuing, and Pershan,[2] gives rise to some difficulties when applied to crystals.[3] In solids, even the Coulomb interactions between electrons which are responsible for many-particle excitations, such as excitons and plasmons, contribute to nonlinear processes.[4] A breakdown of the method of small perturbations, in addition, takes place at resonance positions where the light frequency coincides with the excitation frequency of the crystal so that this method is limited to cases far from resonance. In semiconductors with a small energy gap, however, optical resonances may become important. It must be noted that within the framework of a perturbation approach, a discrimination between proper anharmonicities and renormalization effects – the latter ones leading to oscillations with shifted frequencies – should be made.

To largely avoid the above-mentioned difficulties, it is advantageous to remember the concept of polaritons, introduced by Hopfield,[5] Fano,[6] and Pekar,[7] and to use polariton states as a suitable basis in analyzing nonlinear optical phenomena in semiconductors and insulators by means of a perturbation approach.

In the present paper, the interaction between a nonmetallic ideal crystal and the radiation field is studied quantum mechanically. Lattice vibrations are not taken into account. In Sec. I, the Hamiltonian of the total system is written consistently in the electron-hole-pair representation. In addition, the Coulomb interactions are considered. Those parts of the Coulomb interactions which lead to exciton and plasmon excitations, respectively, are examined in Sec. II. As is shown in Sec. III, the complete Hamiltonian can be split into a *linear* and a *nonlinear* part. The linear part containing strong interactions is responsible for polariton excitations. With the help of a simplified but characteristic and exactly solvable model, the polariton problem is discussed in detail. It is worth mentioning that polaritons, in principle, imply the coupling between plasmons and transverse photons. In Sec. IV, the polariton states are shown to be applicable for the evaluation of nonlinear optical-transition amplitudes.

I. HAMILTONIAN IN ELECTRON-HOLE-PAIR REPRESENTATION

We consider a two-band crystal consisting of a periodic arrangement of N atoms, each one bearing one valence electron. Spin coordinates are neglected; thus, these N electrons in the ground state fill up the valence band, and separated by a finite gap, there may exist a higher-lying empty conduction band. If the crystal is coupled to a radiation field, and we take into account the Coulomb forces between the electrons, the Hamiltonian of the complete system reads

$$H=\frac{1}{8\pi}\int d^3r\,(\vec{E}^2+\vec{H}^2)+\int d^3r\,\Psi^\dagger(\vec{r})\left[\frac{1}{2m}\left(\vec{p}-\frac{e}{c}\vec{A}\right)^2+V(\vec{r})\right]\Psi(\vec{r})+\tfrac{1}{2}\iint' d^3r\,d^3r'\,\Psi^\dagger(\vec{r})\Psi^\dagger(\vec{r}')\,\frac{e^2}{|\vec{r}-\vec{r}'|}\,\Psi(\vec{r}')\Psi(\vec{r})\,. \tag{1.1}$$

$V(\vec{r})$ is the self-consistent one-electron potential, and the prime at the twofold integral over the electron coordinates in the last term of (1.1) means that those parts of the Coulomb interactions which are already incorporated in $V(\vec{r})$ must be subtracted.[8] We choose the Coulomb gauge $\mathrm{div}\vec{A}=0$, and expand the vector potential $\vec{A}$ as follows[9]:

$$\vec{A}(\vec{r})=\sum_{\vec{q}}\left(\frac{2\pi\hbar c^2}{\mathrm{vol}\,\omega_{\vec{q}}}\right)^{1/2}\vec{e}_{\vec{q}}\,(\xi_{\vec{q}}\,e^{i(\vec{q}\vec{r}-\omega_{\vec{q}}t)}+\xi^\dagger_{\vec{q}}\,e^{-i(\vec{q}\vec{r}-\omega_{\vec{q}}t)}), \tag{1.2}$$

where $\omega_{\vec{q}}=c|\vec{q}|$ is the frequency of the free-radiation field, $\vec{e}_{\vec{q}}$ is a unit vector describing the polarization, and vol is the volume of the crystal. The sum over $\vec{q}$ includes summation over the directions of polarization. The photon-creation and photon-annihilation operators obey the commutation relations

$$[\xi_{\vec{q}},\,\xi^\dagger_{\vec{q}'}]_-=\delta_{\vec{q}',\vec{q}}\,,\quad[\xi_{\vec{q}},\,\xi_{\vec{q}'}]_-=[\xi^\dagger_{\vec{q}},\,\xi^\dagger_{\vec{q}'}]_-=0. \tag{1.3}$$

The electron wave field $\Psi(\vec{r})$ is expanded by Bloch functions which shall be eigenstates of the self-consistent one-particle Hamiltonian

$$\Psi(\vec{r})=\textstyle\sum_{\vec{k}}a_{\vec{k}}\,\psi_{c\vec{k}}(\vec{r})+\sum_{\vec{k}}b^\dagger_{\vec{k}}\,\psi_{v\vec{k}}(\vec{r})\,. \tag{1.4}$$

The Bloch functions of wave vector $\vec{k}$ and band number μ,

$$\psi_{\mu\vec{k}}(\vec{r})=[1/(\mathrm{vol})^{1/2}]\,e^{i\vec{k}\vec{r}}u_{\mu\vec{k}}(\vec{r})\,, \tag{1.5}$$

are normalized within the total crystal volume and obey the eigenvalue equation

$$[\vec{p}^2/2m+V(\vec{r})]\,\psi_{\mu\vec{k}}(\vec{r})=\epsilon_{\mu\vec{k}}\,\psi_{\mu\vec{k}}(\vec{r})\,. \tag{1.6}$$

The operators $a^\dagger_{\vec{k}}$, $a_{\vec{k}}$ are creation and annihilation operators of electrons with wave vector $\vec{k}$ in the conduction band (denoted by c), and $b^\dagger_{\vec{k}}$, $b_{\vec{k}}$ are creation and annihilation operators of holes with wave vector $-\vec{k}$ in the valence band (denoted by v). These Fermi operators fulfill the anticommutation relations

$$[a_{\vec k}, a^\dagger_{\vec k'}]_+ = \delta_{\vec k',\vec k}, \quad [a_{\vec k}, a_{\vec k'}]_+ = [a^\dagger_{\vec k}, a^\dagger_{\vec k'}]_+ = 0;$$
$$[b_{\vec k}, b^\dagger_{\vec k'}]_+ = \delta_{\vec k',\vec k}, \quad [b_{\vec k}, b_{\vec k'}]_+ = [b^\dagger_{\vec k}, b^\dagger_{\vec k'}]_+ = 0. \tag{1.7}$$

We insert (1.2) and (1.4) into (1.1), and go to the electron-hole-pair description using the two-particle operators[10]

$$c^\dagger_{\vec k\vec k'} = a^\dagger_{\vec k} b^\dagger_{\vec k'} \quad \text{and} \quad c_{\vec k\vec k'} = b_{\vec k'} a_{\vec k}. \tag{1.8}$$

In crystals where the number of excited electrons is small compared with the total number of electrons, the electron-hole pairs can be treated approximately as bosons with

$$[c_{\vec k\vec k'}, c^\dagger_{\vec k''\vec k'''}]_- \approx \delta_{\vec k'',\vec k}\,\delta_{\vec k''',\vec k'},$$
$$[c_{\vec k\vec k'}, c_{\vec k''\vec k'''}]_- = [c^\dagger_{\vec k\vec k'}, c^\dagger_{\vec k''\vec k'''}]_- = 0. \tag{1.9}$$

Because of the Pauli principle, however, the following relation must hold:

$$(c_{\vec k\vec k'})^n = (c^\dagger_{\vec k\vec k'})^n = 0 \quad \text{for } n \geq 2. \tag{1.10}$$

An exact treatment of the electron-hole pairs requires instead of (1.9) the commutation rule

$$[c_{\vec k\vec k'}, c^\dagger_{\vec k''\vec k'''}]_- = \delta_{\vec k'',\vec k}\,\delta_{\vec k''',\vec k'} - \delta_{\vec k''',\vec k'}\, a^\dagger_{\vec k''} a_{\vec k}$$
$$- \delta_{\vec k'',\vec k}\, b^\dagger_{\vec k'''} b_{\vec k'}. \tag{1.11}$$

We suppose that in our crystal model the total number of electrons is conserved during all excitation processes of interest here, so that starting with a crystal having a filled valence band and an empty conduction band, an equal number of excited electrons and created holes is observed always. The excited states of the crystal therefore are well described by electron-hole-pair operators acting on the ground state. A representation of the Hamiltonian by means of creation and annihilation operators $c^\dagger_{\vec k\vec k'}$, $c_{\vec k\vec k'}$, which is equivalent to the original fermion Hamiltonian, is desirable now. To this end, a translation rule for scattering processes is needed in going from the fermion to the electron-hole-pair description. The way to perform this procedure is illustrated in Fig. 1, using an intraband scattering process out of the Coulomb interaction term which leads to the following operator expression:

$$a^\dagger_{\vec k+\vec q}\, a_{\vec k}\, b^\dagger_{\vec k'+\vec q}\, b_{\vec k'} \to \sum_{\vec k'',\vec k'''} a^\dagger_{\vec k+\vec q}\, b^\dagger_{\vec k''}\, b_{\vec k''}\, a_{\vec k} a^\dagger_{\vec k'''}\, b^\dagger_{\vec k'+\vec q}\, b_{\vec k'}\, a_{\vec k'''} = \sum_{\vec k'',\vec k'''} c^\dagger_{\vec k+\vec q\vec k''}\, c_{\vec k\vec k''}\, c^\dagger_{\vec k'''\vec k'+\vec q}\, c_{\vec k'''\vec k'}$$
$$= c^\dagger_{\vec k+\vec q\vec k'+\vec q}\, c_{\vec k\vec k'} + \sum_{\vec k''\vec k'''} c^\dagger_{\vec k+\vec q\vec k''}\, c^\dagger_{\vec k'''\vec k'+\vec q}\, c_{\vec k'''\vec k'}\, c_{\vec k\vec k''}. \tag{1.12}$$

In the electron-hole-pair representation, the commutation rule (1.9) must now be used instead of (1.11). The normal-ordered sum term in (1.12) provides the same results in the fermion and electron-hole-pair representation if the equivalent operator series acts on an electron-hole-pair state. The insertion of electron and hole-density operators, respectively, is no identity in the fermion picture, but is only a trick used to find the equivalent boson expression. After the density operators are inserted, we introduce the new pair operators which group together the one-particle operators. This procedure holds even if we allow many-particle pair states.

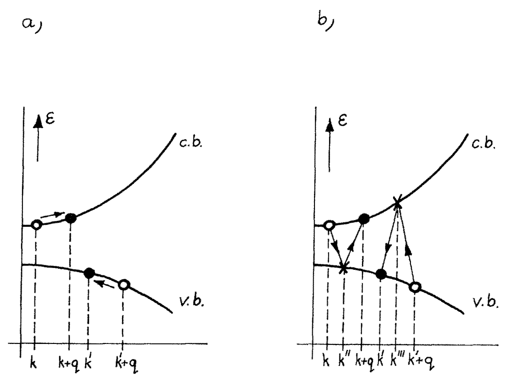

FIG. 1. The scattering process (1.12). (a) Fermion representation, (b) boson representation, and c.b. and v.b. refer to the conduction band and the valence band, respectively.

In doing that, and neglecting umklapp processes in the optical region,[11] we obtain, after straightforward calculations, the following complete Hamiltonian of our system:

$$H = H_{\text{rad}} + H_{\text{sc}} + H^{(2)}_{\text{er I}} + H^{(3)}_{\text{er I}} + H^{(2)}_{\text{er II}} + H^{(3)}_{\text{er II}}$$
$$+ H^{(4)}_{\text{er II}} + H^{(2)}_{\text{C}} + H^{(3)}_{\text{C}} + H^{(4)}_{\text{C}}. \tag{1.13}$$

The contributions to (1.13) are defined as follows. The Hamiltonian of the radiation field reads

$$H_{\mathrm{rad}}=\sum_{\vec q}\hbar\omega_{\vec q}\left(\xi^\dagger_{\vec q}\xi_{\vec q}+\tfrac{1}{2}\right). \tag{1.14}$$

The self-consistent Hamiltonian of the crystal is

$$H_{\mathrm{sc}}=\sum_{\vec q,\vec k}\hbar\Omega_{\vec k+\vec q\,\vec k}\,c^\dagger_{\vec k+\vec q\,\vec k}\,c_{\vec k+\vec q\,\vec k}+\epsilon_0, \tag{1.15}$$

where ϵ_0 is the ground-state energy of the crystal with empty conduction band, and

$$\hbar\Omega_{\vec k+\vec q\,\vec k}=\epsilon_{c\vec k+\vec q}-\epsilon_{v\vec k}$$

is the energy difference between conduction-band and valence-band levels. The interaction between the electrons and the radiation field, which is linear in the vector potential, consists of

$$H^{(2)}_{\mathrm{er\,I}}=-\sum_{\vec q,\vec k}g_{\vec q}\,D^{cv}_{\vec k+\vec q\,\vec k}\left(c^\dagger_{\vec k+\vec q\,\vec k}-c_{-(\vec k+\vec q)-\vec k}\right)\left(\xi_{\vec q}+\xi^\dagger_{-\vec q}\right), \tag{1.16}$$

$$H^{(3)}_{\mathrm{er\,I}}=-\sum_{\vec q,\vec k,\vec k'}g_{\vec q}\left(D^{cc}_{\vec k+\vec q\,\vec k}\,c^\dagger_{\vec k+\vec q\,\vec k'}\,c_{\vec k\vec k'}-D^{vv}_{\vec k+\vec q\,\vec k}\,c^\dagger_{\vec k'\vec k}\,c_{\vec k'\,\vec k+\vec q}\right)\left(\xi_{\vec q}+\xi^\dagger_{-\vec q}\right). \tag{1.17}$$

The electron-photon interaction, which is quadratic in the vector potential, consists of the following parts:

$$H^{(2)}_{\mathrm{er\,II}}=N\sum_{\vec q}f_{\vec q\vec q}\left(\xi_{\vec q}+\xi^\dagger_{-\vec q}\right)\left(\xi^\dagger_{\vec q}+\xi_{-\vec q}\right), \tag{1.18}$$

$$H^{(3)}_{\mathrm{er\,II}}=\sum_{\vec q,\vec q',\vec k}f_{\vec q\vec q'}\,v^{cv}_{\vec k+\vec q\,\vec k+\vec q'}\left(c^\dagger_{\vec k+\vec q\,\vec k+\vec q'}+c_{-(\vec k+\vec q)-(\vec k+\vec q')}\right)\times\left(\xi_{\vec q}+\xi^\dagger_{-\vec q}\right)\left(\xi^\dagger_{\vec q'}+\xi_{-\vec q'}\right), \tag{1.19}$$

$$H^{(4)}_{\mathrm{er\,II}}=\sum_{\vec q,\vec q',\vec k,\vec k'}f_{\vec q\vec q'}\left(v^{cc}_{\vec k+\vec q\,\vec k+\vec q'}\,c^\dagger_{\vec k+\vec q\,\vec k'}\,c_{\vec k+\vec q'\,\vec k'}-v^{vv}_{\vec k+\vec q\,\vec k+\vec q'}\,c^\dagger_{\vec k'\,\vec k+\vec q'}\,c_{\vec k'\,\vec k+\vec q}\right)\times\left(\xi_{\vec q}+\xi^\dagger_{-\vec q}\right)\left(\xi^\dagger_{\vec q'}+\xi_{-\vec q'}\right). \tag{1.20}$$

The components of the Coulomb interaction are

$$H^{(2)}_{\mathrm{C}}=\frac{2\pi e^2}{\mathrm{vol}}\sum_{\vec q,\vec k,\vec k'}\frac{1}{|\vec q|^2}\Big(v^{cv}_{\vec k+\vec q\,\vec k}v^{cv}_{\vec k'\,\vec k'+\vec q}\,c^\dagger_{\vec k+\vec q\,\vec k}\,c^\dagger_{\vec k'\,\vec k'+\vec q}+v^{cv*}_{\vec k'\,\vec k'+\vec q}\,v^{cv*}_{\vec k+\vec q\,\vec k}\,c_{\vec k'\,\vec k'+\vec q}\,c_{\vec k+\vec q\,\vec k}$$
$$+2\,v^{cv}_{\vec k+\vec q\,\vec k}v^{cv*}_{\vec k'+\vec q\,\vec k'}\,c^\dagger_{\vec k+\vec q\,\vec k}\,c_{\vec k'+\vec q\,\vec k'}-2v^{cc}_{\vec k+\vec q\,\vec k}v^{vv}_{\vec k'\,\vec k'+\vec q}\,c^\dagger_{\vec k+\vec q\,\vec k'+\vec q}\,c_{\vec k\vec k'}\Big), \tag{1.21}$$

$$H^{(3)}_{\mathrm{C}}=\frac{4\pi e^2}{\mathrm{vol}}\sum_{\vec q,\vec k,\vec k',\vec k''}\frac{1}{|\vec q|^2}\Big[v^{cv}_{\vec k+\vec q\,\vec k}\,c^\dagger_{\vec k+\vec q\,\vec k}\left(v^{cc}_{\vec k'\,\vec k'+\vec q}\,c^\dagger_{\vec k'\,\vec k''}\,c_{\vec k'+\vec q\,\vec k''}-v^{vv}_{\vec k'\,\vec k'+\vec q}\,c^\dagger_{\vec k''\,\vec k'+\vec q}\,c_{\vec k''\,\vec k'}\right)$$
$$+\left(v^{cc}_{\vec k'+\vec q\,\vec k'}\,c^\dagger_{\vec k'+\vec q\,\vec k''}\,c_{\vec k'\,\vec k''}-v^{vv}_{\vec k'+\vec q\,\vec k'}\,c^\dagger_{\vec k''\,\vec k'}\,c_{\vec k''\,\vec k'+\vec q}\right)v^{cv*}_{\vec k+\vec q\,\vec k}\,c_{\vec k+\vec q\,\vec k}\Big], \tag{1.22}$$

$$H^{(4)}_{\mathrm{C}}=\frac{2\pi e^2}{\mathrm{vol}}\sum_{\vec q,\vec k,\vec k',\vec k'',\vec k'''}\frac{1}{|\vec q|^2}\Big[v^{cc}_{\vec k+\vec q\,\vec k}\,c^\dagger_{\vec k+\vec q\,\vec k''}\left(v^{cc}_{\vec k'\,\vec k'+\vec q}\,c^\dagger_{\vec k'\,\vec k'''}\,c_{\vec k'+\vec q\,\vec k'''}-v^{vv}_{\vec k'\,\vec k'+\vec q}\,c^\dagger_{\vec k'''\,\vec k'+\vec q}\,c_{\vec k'''\,\vec k'}\right)c_{\vec k\vec k''}$$
$$-v^{vv}_{\vec k\,\vec k+\vec q}\,c^\dagger_{\vec k''\,\vec k+\vec q}\left(v^{cc}_{\vec k'+\vec q\,\vec k'}\,c^\dagger_{\vec k'+\vec q\,\vec k'''}\,c_{\vec k'\,\vec k'''}-v^{vv}_{\vec k'+\vec q\,\vec k'}\,c^\dagger_{\vec k'''\,\vec k'}\,c_{\vec k'''\,\vec k'+\vec q}\right)c_{\vec k''\,\vec k}\Big]. \tag{1.23}$$

Here the following notations:

$$g_{\vec q}=\frac{e}{m}\left(\frac{2\pi\hbar}{\mathrm{vol}\,\omega_{\vec q}}\right)^{1/2},\quad f_{\vec q\vec q'}=\frac{\pi e^2\hbar}{m\,\mathrm{vol}}\,\frac{\vec e_{\vec q}\cdot\vec e_{\vec q'}}{(\omega_{\vec q}\omega_{\vec q'})^{1/2}}, \tag{1.24}$$

$$D^{\mu\mu'}_{\vec k+\vec q\,\vec k}=\hbar(\vec e_{\vec q}\cdot\vec k)\,v^{\mu\mu'}_{\vec k+\vec q\,\vec k}+\vec e_{\vec q}\cdot\vec p^{\mu\mu'}_{\vec k+\vec q\,\vec k}, \tag{1.25}$$

$$\vec p^{\mu\mu'}_{\vec k\vec k'}=(N/\mathrm{vol})\int_{\mathrm{cell}}d^3r\,u^*_{\mu\vec k}(\vec r)\,\vec p\,u_{\mu'\vec k'}(\vec r), \tag{1.26}$$

$$v^{\mu\mu'}_{\vec k\vec k'}=(N/\mathrm{vol})\int_{\mathrm{cell}}d^3r\,u^*_{\mu\vec k}(\vec r)\,u_{\mu'\vec k'}(\vec r); \tag{1.27}$$

and time-reversal properties:

$$\epsilon_{\mu\,-\vec k}=\epsilon_{\mu\vec k},\quad v^{\mu\mu'*}_{-\vec k-\vec k'}=v^{\mu\mu'}_{\vec k\vec k'}, \tag{1.28}$$

$$\vec p^{\mu\mu'*}_{-\vec k-\vec k'}=-\vec p^{\mu\mu'}_{\vec k\vec k'};\quad D^{\mu\mu'*}_{-(\vec k+\vec q)-\vec k}=-D^{\mu\mu'}_{\vec k+\vec q\,\vec k}$$

were used, and in H_{C} the Fourier transform of $e^2/|\vec r-\vec r'|$ was introduced. The photon wave vector is not neglected in the matrix elements (1.25)–(1.27), and no dipole approximation is used. This fact is meaningful for plasmon-photon interactions.

II. EXCITON AND PLASMON EQUATIONS

Before discussing the interaction problem between radiation and crystal, we show first that the crystal Hamiltonian, consisting of H_{sc} and the bilinear Coulomb terms $H^{(2)}_{\mathrm{C}}$, contains an eigenvalue problem for excitons as well as for low-lying plasmons.

A. Exciton Equation

Let us neglect the virtual parts of $H^{(2)}_{\mathrm{C}}$ having two creation or annihilation operators, and take the Hamiltonian

$$H_{\mathrm{cryst}}=H_{\mathrm{sc}}+\frac{4\pi e^2}{\mathrm{vol}}\sum_{\vec q,\vec k,\vec k'}\frac{v^{cv}_{\vec k+\vec q\,\vec k}\,v^{cv*}_{\vec k'+\vec q\,\vec k'}}{|\vec q|^2}\,c^\dagger_{\vec k+\vec q\,\vec k}\,c_{\vec k'+\vec q\,\vec k'}-\frac{4\pi e^2}{\mathrm{vol}}\sum_{\vec q,\vec k,\vec k'}\frac{v^{cc}_{\vec k+\vec q\,\vec k}\,v^{vv}_{\vec k'\,\vec k'+\vec q}}{|\vec q|^2}\,c^\dagger_{\vec k+\vec q\,\vec k'+\vec q}\,c_{\vec k\vec k'}. \tag{2.1}$$

An exciton state is given by the following superposition of free eigenstates:

$$|\Phi^{ex}\rangle = \sum_{\vec q,\vec k} \alpha_{\vec k+\vec q\vec k}\, c^{\dagger}_{\vec k+\vec q\vec k}\,|0\rangle . \qquad (2.2)$$

The normalization condition reads

$$\sum_{\vec q,\vec k} |\alpha_{\vec k+\vec q\vec k}|^2 = 1\,, \qquad (2.3)$$

and we get the eigenvalue equation

$$(H_{\text{cryst}} - \epsilon_0)\,|\Phi^{ex}\rangle = \hbar\omega^{ex}\,|\Phi^{ex}\rangle . \qquad (2.4)$$

Inserting (2.2) into (2.4), and multiplying from the left by $\langle 0|c_{\vec k''+\vec q\vec k''}$, we obtain the exciton equation

$$\hbar(\omega^{ex} - \Omega_{\vec k+\vec q\vec k})\,\alpha_{\vec k+\vec q\vec k} + \frac{4\pi e^2}{\text{vol}} \sum_{\vec k'} \left(\frac{v^{cc}_{\vec k+\vec q\,\vec k'+\vec q}\, v^{vv}_{\vec k'\vec k}}{|\vec k-\vec k'|^2} - \frac{v^{cv}_{\vec k+\vec q\vec k}\, v^{cv*}_{\vec k'+\vec q\vec k'}}{|\vec q|^2} \right) \alpha_{\vec k'+\vec q\vec k'} = 0. \qquad (2.5)$$

This type of equation is known from exciton theories, and (2.5) leads to an exciton effective-mass equation. For a further treatment of (2.5) we refer to the extensive literature on exciton problems.[12,13]

B. Plasmon Equations

The interband Coulomb interactions of $H_C^{(2)}$ are responsible for low-lying plasma oscillations in insulating crystals. To show this, we follow the idea of Horie,[13] but we need only the comparatively simple Hamiltonian

$$H_{\text{cryst}} = H_{sc} + \frac{2\pi e^2}{\text{vol}} \sum_{\vec q,\vec k,\vec k'} \left(2\,\frac{v^{cv}_{\vec k+\vec q\vec k}\, v^{cv*}_{\vec k'+\vec q\vec k'}}{|\vec q|^2}\, c^{\dagger}_{\vec k+\vec q\vec k}\, c_{\vec k'+\vec q\vec k'} + \frac{v^{cv}_{\vec k+\vec q\vec k}\, v^{cv}_{\vec k',\vec k'+\vec q}}{|\vec q|^2}\, c^{\dagger}_{\vec k+\vec q\vec k}\, c^{\dagger}_{\vec k',\vec k'+\vec q} + \frac{v^{cv*}_{\vec k+\vec q\vec k}\, v^{cv*}_{\vec k',\vec k'+\vec q}}{|\vec q|^2}\, c_{\vec k',\vec k'+\vec q}\, c_{\vec k+\vec q\vec k} \right) . \qquad (2.6)$$

In (2.6), the intraband processes of $H_C^{(2)}$ have been neglected. We look for excitation energies in describing the many-body plasmon states by the superposition

$$|\Phi^{pl}\rangle = A^{\dagger}\,|\Phi^0\rangle , \qquad (2.7)$$

with

$$A^{\dagger} = \sum_{\vec q,\vec k} (\alpha_{\vec k+\vec q\vec k}\, c^{\dagger}_{\vec k+\vec q\vec k} + \beta^{*}_{\vec k\vec k+\vec q}\, c_{\vec k\vec k+\vec q}) , \qquad (2.8)$$

where $|\Phi^0\rangle$ is the ground state defined by[14]

$$A\,|\Phi^0\rangle = 0 \quad \text{and} \quad H_{\text{cryst}}|\Phi^0\rangle = E^0|\Phi^0\rangle , \qquad (2.9)$$

and the following relation must hold:

$$H_{\text{cryst}}|\Phi^{pl}\rangle = E\,|\Phi^{pl}\rangle . \qquad (2.10)$$

With regard to (1.9), the normalization condition

$$\sum_{\vec q,\vec k} (|\alpha_{\vec k+\vec q\vec k}|^2 - |\beta_{\vec k\vec k+\vec q}|^2) = 1 \qquad (2.11)$$

results in the commutation relation

$$[A, A^{\dagger}]_- = 1 . \qquad (2.12)$$

A is the adjoint operator to $A^{\dagger}$. Because of (2.9) and (2.10), the excitation energy $\hbar\omega^{pl} = E - E^0$ is determined by the equation

$$(\hbar\omega^{pl} A^{\dagger} - [H_{\text{cryst}}, A^{\dagger}]_-)\,|\Phi^0\rangle = 0 . \qquad (2.13)$$

In order that (2.13) be fulfilled, the following equations must hold:

$$\hbar(\omega^{pl} - \Omega_{\vec k+\vec q\vec k})\,\alpha_{\vec k+\vec q\vec k} - \frac{4\pi e^2}{\text{vol}\,|\vec q|^2}\, v^{cv}_{\vec k+\vec q\vec k} \sum_{\vec k'} v^{cv*}_{\vec k'+\vec q\vec k'}\,\alpha_{\vec k'+\vec q\vec k'} + \frac{4\pi e^2}{\text{vol}\,|\vec q|^2}\, v^{cv}_{\vec k+\vec q\vec k} \sum_{\vec k'} v^{cv}_{\vec k',\vec k'+\vec q}\,\beta^{*}_{\vec k',\vec k'+\vec q} = 0 , \qquad (2.14a)$$

$$\hbar(\omega^{pl} + \Omega_{\vec k\vec k+\vec q})\,\beta^{*}_{\vec k\vec k+\vec q} + \frac{4\pi e^2}{\text{vol}\,|\vec q|^2}\, v^{cv*}_{\vec k\vec k+\vec q} \sum_{\vec k'} v^{cv}_{\vec k',\vec k'+\vec q}\,\beta^{*}_{\vec k',\vec k'+\vec q} - \frac{4\pi e^2}{\text{vol}\,|\vec q|^2}\, v^{cv*}_{\vec k\vec k+\vec q} \sum_{\vec k'} v^{cv*}_{\vec k'+\vec q\vec k'}\,\alpha_{\vec k'+\vec q\vec k'} = 0 . \qquad (2.14b)$$

These are typical plasmon equations for insulating crystals of the type discussed earlier by Horie.[13] Equations (2.14a) and (2.14b) lead to the plasmon dispersion relation

$$1 - \frac{4\pi e^2}{\text{vol}\,|\vec q|^2} \sum_{\vec k} \left(\frac{|v^{cv}_{\vec k+\vec q\vec k}|^2}{\hbar(\omega^{pl} - \Omega_{\vec k+\vec q\vec k})} - \frac{|v^{cv}_{\vec k\vec k+\vec q}|^2}{\hbar(\omega^{pl} + \Omega_{\vec k\vec k+\vec q})} \right) = 0 . \qquad (2.15)$$

Having shown the influence of $H_C^{(2)}$, giving rise to exciton and plasmon excitations, we further note that the Hamiltonian resulting from (2.6) in the case of $\vec k' = \vec k$ can be diagonalized exactly by carrying out a Bogolyubov transformation.[15]

III. POLARITONS IN A SIMPLIFIED CRYSTAL MODEL

In order to investigate the interaction with the radiation field, we collect the different parts of the complete Hamiltonian (1.13) in the following way:

$$H = H_0 + H_1 + H_{nl} , \qquad (3.1)$$

where

$$H_0 = H_{rad} + H_{sc} \qquad (3.2)$$

are the free Hamiltonians of the radiation field and the crystal, respectively, and

$$H_1 = H^{(2)}_{er\,I} + H^{(2)}_{er\,II} + H^{(2)}_{C} \qquad (3.3)$$

contains all the coupling terms which are bilinear in the particle operators; also,

$$H_{nl} = H^{(3)}_{er\,I} + H^{(3)}_{er\,II} + H^{(3)}_{C} + H^{(4)}_{er\,II} + H^{(4)}_{C} \qquad (3.4)$$

are anharmonic terms of third and fourth order in the particle operators. The interactions (3.3) are called *linear processes*, and the interactions (3.4) *nonlinear processes*.

Simplifying our crystal model, we can show that the eigenstates of H_0+H_1 are polariton states, and the anharmonic terms H_{nl} lead to nonlinear optical effects. Our simplification consists in replacing the c band by a single excitation level lying over the maximum of the valence band at $\vec{k}=0$,[16] whereby a reduction of sums over wave vectors results in the following terms:

$$H_{sc}=\sum_{\vec{q}} \hbar\Omega_{0\vec{q}}c^\dagger_{0\vec{q}}c_{0\vec{q}}+\epsilon_0, \tag{3.5}$$

$$H_C^{(2)}=\frac{4\pi e^2}{\text{vol}}\sum_{\vec{q}}\frac{|v^{cv}_{0\vec{q}}|^2}{|\vec{q}|^2}c^\dagger_{0\vec{q}}c_{0\vec{q}} +\frac{2\pi e^2}{\text{vol}}\sum_{\vec{q}}\frac{|v^{cv}_{0\vec{q}}|^2}{|\vec{q}|^2}(c^\dagger_{0\vec{q}}c^\dagger_{0-\vec{q}}+c_{0-\vec{q}}c_{0\vec{q}}), \tag{3.6}$$

$$H_C^{(3)}=-\frac{4\pi e^2}{\text{vol}}\sum_{\vec{q},\vec{k}}\frac{v^{cv*}_{0\vec{q}}v^{vv}_{\vec{k}\vec{k}+\vec{q}}}{|\vec{q}|^2}c^\dagger_{0\vec{k}+\vec{q}}(c^\dagger_{0-\vec{q}}+c_{0\vec{q}})c_{0\vec{k}}, \tag{3.7}$$

$$H_C^{(4)}=\frac{2\pi e^2}{\text{vol}}\sum_{\vec{q},\vec{k},\vec{k}'}\frac{v^{vv}_{\vec{k}'+\vec{q}\vec{k}'}v^{vv}_{\vec{k}\vec{k}+\vec{q}}}{|\vec{q}|^2}c^\dagger_{0\vec{k}+\vec{q}}c^\dagger_{0\vec{k}'}c_{0\vec{k}'+\vec{q}}c_{0\vec{k}}, \tag{3.8}$$

$$H^{(2)}_{er\,I}=-\sum_{\vec{q}}g_{\vec{q}}D^{cv}_{0\vec{q}}(c^\dagger_{0\vec{q}}-c_{0-\vec{q}})(\xi^\dagger_{\vec{q}}+\xi_{-\vec{q}}), \tag{3.9}$$

$$H^{(3)}_{er\,I}=\sum_{\vec{q},\vec{k}}g_{\vec{q}}D^{vv}_{\vec{k}+\vec{q}\vec{k}}c^\dagger_{0\vec{k}}c_{0\vec{k}+\vec{q}}(\xi_{\vec{q}}+\xi^\dagger_{-\vec{q}}), \tag{3.10}$$

$$H^{(3)}_{er\,II}=\sum_{\vec{q},\vec{q}';\vec{q}'\neq\vec{q}}f_{\vec{q}\vec{q}'}v^{cv}_{0\vec{q}'-\vec{q}}(c^\dagger_{0\vec{q}'-\vec{q}}+c_{0-(\vec{q}'-\vec{q})})\times(\xi_{\vec{q}}+\xi^\dagger_{-\vec{q}})(\xi^\dagger_{\vec{q}'}+\xi_{-\vec{q}'}), \tag{3.11}$$

$$H^{(4)}_{er\,II}=-\sum_{\vec{q},\vec{q}',\vec{k};\vec{q}'\neq\vec{q}}f_{\vec{q}\vec{q}'}v^{vv}_{\vec{k}+\vec{q}\,\vec{k}+\vec{q}'}c^\dagger_{0\vec{k}+\vec{q}'}c_{0\vec{k}+\vec{q}}\times(\xi_{\vec{q}}+\xi^\dagger_{-\vec{q}})(\xi^\dagger_{\vec{q}'}+\xi_{-\vec{q}'}). \tag{3.12}$$

We notice that intraband processes as well as two-photon processes belong to the nonlinearities H_{nl}. The eigenvalue problem described by the Hamiltonian H_0+H_1 of our simplified model can be solved exactly. The corresponding eigenstates give the basis for a later perturbation treatment of the nonlinearities H_{nl}. With the aid of the unitary transformations

$$\begin{pmatrix}\eta^\dagger_{\vec{q}}\\ \eta_{-\vec{q}}\end{pmatrix}=\begin{pmatrix}\sqrt{C_+} & \sqrt{C_-}\\ \sqrt{C_-} & \sqrt{C_+}\end{pmatrix}\begin{pmatrix}\xi^\dagger_{\vec{q}}\\ \xi_{-\vec{q}}\end{pmatrix},$$

$$\begin{pmatrix}d^\dagger_{0\vec{q}}\\ d_{0-\vec{q}}\end{pmatrix}=\begin{pmatrix}\sqrt{D_+} & \sqrt{D_-}\\ \sqrt{D_-} & \sqrt{D_+}\end{pmatrix}\begin{pmatrix}c^\dagger_{0\vec{q}}\\ c_{0-\vec{q}}\end{pmatrix}, \tag{3.13a}$$

$$\begin{pmatrix}\xi^\dagger_{\vec{q}}\\ \xi_{-\vec{q}}\end{pmatrix}=\begin{pmatrix}\sqrt{C_+} & -\sqrt{C_-}\\ -\sqrt{C_-} & \sqrt{C_+}\end{pmatrix}\begin{pmatrix}\eta^\dagger_{\vec{q}}\\ \eta_{-\vec{q}}\end{pmatrix},$$

$$\begin{pmatrix}c^\dagger_{0\vec{q}}\\ c_{0-\vec{q}}\end{pmatrix}=\begin{pmatrix}\sqrt{D_+} & -\sqrt{D_-}\\ -\sqrt{D_-} & \sqrt{D_+}\end{pmatrix}\begin{pmatrix}d^\dagger_{0\vec{q}}\\ d_{0-\vec{q}}\end{pmatrix}, \tag{3.13b}$$

we introduce new quasiparticle operators, where we used the following abbreviations:

$$C_+=\frac{(\tilde{\omega}_{\vec{q}}+\omega_{\vec{q}})^2}{4\tilde{\omega}_{\vec{q}}\omega_{\vec{q}}}, \quad C_-=\frac{(\tilde{\omega}_{\vec{q}}-\omega_{\vec{q}})^2}{4\tilde{\omega}_{\vec{q}}\omega_{\vec{q}}}, \tag{3.14a}$$

$$D_+=\frac{(\tilde{\Omega}_{0\vec{q}}+\Omega_{0\vec{q}})^2}{4\tilde{\Omega}_{0\vec{q}}\Omega_{0\vec{q}}}, \quad D_-=\frac{(\tilde{\Omega}_{0\vec{q}}-\Omega_{0\vec{q}})^2}{4\tilde{\Omega}_{0\vec{q}}\Omega_{0\vec{q}}}, \tag{3.14b}$$

$$\tilde{\omega}_{\vec{q}}=(\omega^2_{\vec{q}}+\omega^2_p)^{1/2}, \tag{3.15a}$$

$$\tilde{\Omega}_{0\vec{q}}=(\Omega^2_{0\vec{q}}+2\Omega_{0\vec{q}}\lambda_{0\vec{q}})^{1/2}. \tag{3.15b}$$

The classical plasma frequency is given by

$$\omega_p=(4\pi Ne^2/m\,\text{vol})^{1/2}, \tag{3.16a}$$

and $\lambda_{0\vec{q}}$ is defined as

$$\lambda_{0\vec{q}}=4\pi e^2|v^{cv}_{0\vec{q}}|^2/\text{vol}\,\hbar|\vec{q}|^2. \tag{3.16b}$$

It is easy to see that

$$C_+-C_-=D_+-D_-=1, \tag{3.17}$$

and the new creation and annihilation operators fulfill the commutation relations

$$[\eta_{\vec{q}},\eta^\dagger_{\vec{q}'}]_-=\delta_{\vec{q}'\vec{q}}, \quad [\eta_{\vec{q}},\eta_{\vec{q}'}]_-=[\eta^\dagger_{\vec{q}},\eta^\dagger_{\vec{q}'}]_-=0, \tag{3.18a}$$

$$[d_{0\vec{q}},d^\dagger_{0\vec{q}'}]_-=\delta_{\vec{q}',\vec{q}}, \quad [d_{0\vec{q}},d_{0\vec{q}'}]_-=[d^\dagger_{0\vec{q}},d^\dagger_{0\vec{q}'}]_-=0. \tag{3.18b}$$

In terms of the operators (3.13), we can write

$$H_{ph}=H_{rad}+H^{(2)}_{er\,II}=\sum_{\vec{q}}\hbar\tilde{\omega}_{\vec{q}}(\eta^\dagger_{\vec{q}}\eta_{\vec{q}}+\tfrac{1}{2}), \tag{3.19}$$

and with respect to an energy scale where

$$\epsilon_0-\tfrac{1}{2}\sum_{\vec{q}}\hbar(\Omega_{0\vec{q}}+\lambda_{0\vec{q}})$$

is put equal to zero, we have

$$H_{cryst}=H_{sc}+H^{(2)}_C=\sum_{\vec{q}}\hbar\tilde{\Omega}_{0\vec{q}}(d^\dagger_{0\vec{q}}d_{0\vec{q}}+\tfrac{1}{2}). \tag{3.20}$$

Equations (3.19) and (3.20) are Hamiltonians of harmonic oscillators with renormalized frequencies in comparison to the frequencies of the free-radiation field H_{rad} and the self-consistent crystal field H_{sc}, respectively. The frequency shift by an amount of the classical plasma frequency in (3.15a) stems from ground-state fluctuations, and the dispersion law is shown in Fig. 2. The difference $\tilde{\omega}^2_{\vec{q}}-\omega^2_{\vec{q}}=\omega_p$ is independent of the wave vector $\vec{q}$. If we had neglected the virtual processes of $H^{(2)}_{er\,II}$, characterized by the operator products $\xi^\dagger_{-\vec{q}}\xi^\dagger_{\vec{q}}$ and $\xi_{\vec{q}}\xi_{-\vec{q}}$, a singularity in the dispersion law at $\vec{q}=0$ would appear. The ground state, $|\chi_0\rangle$ say, of the system

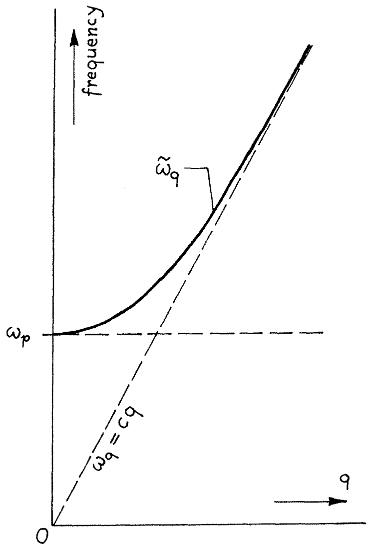

FIG. 2. Dispersion law (3.15a).

with the Hamiltonian (3.19) is defined by $\eta_{\vec{q}}|\chi_0\rangle = 0$ for all $\vec{q}$, and has the form

$$|\chi_0\rangle = \exp\left\{-\tfrac{1}{2}\sum_{\vec{q}} \frac{\tilde{\omega}_{\vec{q}} - \omega_{\vec{q}}}{\tilde{\omega}_{\vec{q}} + \omega_{\vec{q}}}\, \xi^\dagger_{\vec{q}}\xi^\dagger_{-\vec{q}}\right\}|0\rangle\, \mathrm{M}. \qquad (3.21)$$

M is a normalization factor, and $|0\rangle$ is the vacuum state, i.e., $\xi_{\vec{q}}|0\rangle = 0$ for all $\vec{q}$. $|\chi_0\rangle$ is, because of the virtual processes, no longer a vacuum state with regard to the free-photon field. The frequency shift in (3.20) or (3.15b) stems from the interband Coulomb interactions $H_C^{(2)}$. Here $\tilde{\Omega}^2_{0\vec{q}} - \Omega^2_{0\vec{q}} = 2\Omega_{0\vec{q}}\lambda_{0\vec{q}}$ is $\vec{q}$ dependent. The eigenvalue equation of the lattice functions $u_{\mu\vec{k}}(\vec{r})$, introduced in (1.5), leads to the relation

$$v^{\mu'\mu}_{\vec{k}'\vec{k}} = \frac{(\hbar/m)(\vec{k}'-\vec{k})\cdot \vec{p}^{\,\mu'\mu}_{\vec{k}'\vec{k}}}{\epsilon_{\mu'\vec{k}'} - \epsilon_{\mu\vec{k}} - (\hbar^2/2m)(\vec{k}'^2 - \vec{k}^2)} . \qquad (3.22)$$

From (3.22), we see that $\lambda_{0\vec{q}}$ has no singularity at $\vec{q} = 0$, and we can write

$$2\Omega_{0\vec{q}}\lambda_{0\vec{q}} = \omega_p^2 f^{cv}_{0\vec{q}} \qquad (3.23)$$

with the oscillator strength

$$f^{cv}_{0\vec{q}} = \frac{1}{N}\,\frac{2\cos^2\Theta\,|\vec{p}^{\,cv}_{0\vec{q}}|^2}{m\hbar(\Omega_{0\vec{q}} + \hbar\vec{q}^2/2m)^2(\Omega_{0\vec{q}})^{-1}} . \qquad (3.24)$$

Θ is the angle between $\vec{q}$ and $\vec{p}^{\,cv}_{0\vec{q}}$. In the case of small $\vec{q}$ values, (3.24) reduces to the usual form[17]

$$f^{cv}_{0\vec{q}} \approx (1/N)\, 2\cos^2\Theta\, |\vec{p}^{\,cv}_{0\vec{q}}|^2/m\hbar\Omega_{0\vec{q}} \qquad (3.24')$$

Equation (3.23) is a $\vec{q}$-dependent plasmalike contribution to the new frequency $\tilde{\Omega}_{0\vec{q}}$ which takes into account not only the Coulomb coupling between excited electron and hole, but also the collective behavior of electron-hole pairs with opposite wave vectors. We remember of the f-sum rule, and see

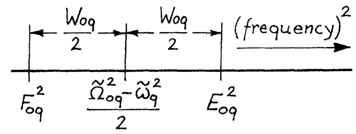

FIG. 3. Separation of the squares of the polariton eigenfrequencies.

at once that $\tilde{\omega}^2_{\vec{q}} - \omega^2_{\vec{q}} > \tilde{\Omega}^2_{0\vec{q}} - \Omega^2_{0\vec{q}}$. We can say that each "quasiphoton," described by the creation and annihilation operators $\eta^\dagger_{\vec{q}}$ and $\eta_{\vec{q}}$, is affected by the total ground-state fluctuations, whereas each "quasi-electron-hole pair," described by the creation and annihilation operators $d^\dagger_{0\vec{q}}, d_{0\vec{q}}$, is affected only by the collective motion of electron-hole pairs with relative wave vectors $\vec{q}$ and $-\vec{q}$.

In terms of the new quasiparticle creation and annihilation operators (3.13), the Hamiltonian of the linear system reads

$$\begin{aligned} H' &= H_0 + H_1 = H_{\mathrm{ph}} + H_{\mathrm{cryst}} + H^{(2)}_{\mathrm{er\,I}} , \\ &= \sum_{\vec{q}} \hbar\tilde{\omega}_{\vec{q}}(\eta^\dagger_{\vec{q}}\eta_{\vec{q}} + \tfrac{1}{2}) + \sum_{\vec{q}} \hbar\tilde{\Omega}_{0\vec{q}}(d^\dagger_{0\vec{q}}d_{0\vec{q}} + \tfrac{1}{2}) \\ &\quad - \sum_{\vec{q}} g_{\vec{q}}\tilde{D}^{cv}_{0\vec{q}}(d^\dagger_{0\vec{q}} - d_{0-\vec{q}})(\eta^\dagger_{\vec{q}} + \eta_{-\vec{q}}), \end{aligned} \qquad (3.25)$$

where

$$\tilde{D}^{cv}_{0\vec{q}} = (\omega_{\vec{q}}\tilde{\Omega}_{0\vec{q}}/\tilde{\omega}_{\vec{q}}\Omega_{0\vec{q}})^{1/2} D^{cv}_{0\vec{q}} ,$$

$$D^{cv}_{0\vec{q}} = \vec{e}_{\vec{q}}\cdot \vec{p}^{\,cv}_{0\vec{q}} , \qquad (3.26)$$

By means of a further linear canonical transformation (3.27a) and its inverse (3.27b), we introduce in the next step two types of polariton[5-7] creation and annihilation operators $\hat{d}^\dagger_{0\vec{q}}$, $\hat{d}_{0\vec{q}}$ and $\hat{\eta}_{0\vec{q}}$, $\hat{\eta}_{0\vec{q}}$, respectively, which are suitable in describing the collective behavior of quasiphotons, and quasi-electron-hole pairs in dielectric media,

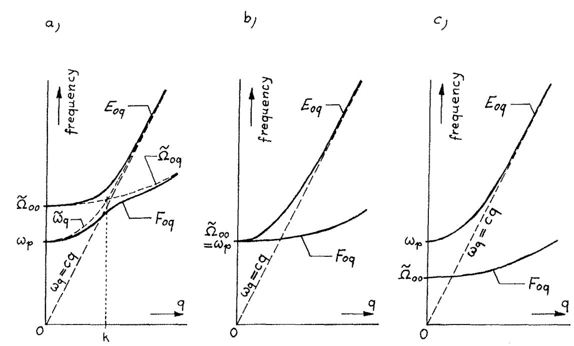

FIG. 4. Polariton dispersion law. (a) Weakly curved valence band with $\tilde{\Omega}_{00} > \omega_p$, and a resonance position at $\vec{q} = \vec{k}$, (b) degenerate case with $E_{00} = F_{00}$ and $\Omega_{00} = \omega_p$ at a resonance position $\vec{q} = 0$, and (c) weakly curved valence band where $\tilde{\omega}_{\vec{q}} > \tilde{\Omega}_{0\vec{q}}$ in the whole $\vec{q}$ space.

$$\begin{pmatrix}\hat d^\dagger_{0\vec q}\\ \hat d_{0-\vec q}\\ \hat\eta_{0\vec q}\\ \hat\eta^\dagger_{0-\vec q}\end{pmatrix}=\begin{pmatrix}2Z_{0\vec q}(A_+)^{1/2} & -2Z_{0\vec q}(A_-)^{1/2} & -Y^*_{0\vec q}(E_{0\vec q}/\tilde\Omega_{0\vec q})^{1/2}-X^*_{0\vec q}(\tilde\Omega_{0\vec q}/E_{0\vec q})^{1/2} & -Y^*_{0\vec q}(E_{0\vec q}/\tilde\Omega_{0\vec q})^{1/2}+X^*_{0\vec q}(\tilde\Omega_{0\vec q}/E_{0\vec q})^{1/2}\\ -2Z_{0\vec q}(A_-)^{1/2} & 2Z_{0\vec q}(A_+)^{1/2} & Y^*_{0\vec q}(E_{0\vec q}/\tilde\Omega_{0\vec q})^{1/2}-X^*_{0\vec q}(\tilde\Omega_{0\vec q}/E_{0\vec q})^{1/2} & Y^*_{0\vec q}(E_{0\vec q}/\tilde\Omega_{0\vec q})^{1/2}+X^*_{0\vec q}(\tilde\Omega_{0\vec q}/E_{0\vec q})^{1/2}\\ -Y_{0\vec q}(\tilde\omega_{\vec q}/F_{0\vec q})^{1/2}-X_{0\vec q}(F_{0\vec q}/\tilde\omega_{\vec q})^{1/2} & -Y_{0\vec q}(\tilde\omega_{\vec q}/F_{0\vec q})^{1/2}+X_{0\vec q}(F_{0\vec q}/\tilde\omega_{\vec q})^{1/2} & 2Z_{0\vec q}(B_+)^{1/2} & -2Z_{0\vec q}(B_-)^{1/2}\\ Y_{0\vec q}(\tilde\omega_{\vec q}/F_{0\vec q})^{1/2}-X_{0\vec q}(F_{0\vec q}/\tilde\omega_{\vec q})^{1/2} & Y_{0\vec q}(\tilde\omega_{\vec q}/F_{0\vec q})^{1/2}+X_{0\vec q}(F_{0\vec q}/\tilde\omega_{\vec q})^{1/2} & -2Z_{0\vec q}(B_-)^{1/2} & 2Z_{0\vec q}(B_+)^{1/2}\end{pmatrix}\begin{pmatrix}d^\dagger_{0\vec q}\\ d_{0-\vec q}\\ \eta_{\vec q}\\ \eta^\dagger_{-\vec q}\end{pmatrix}, \quad (3.27a)$$

$$\begin{pmatrix}d^\dagger_{0\vec q}\\ d_{0-\vec q}\\ \eta_{\vec q}\\ \eta^\dagger_{-\vec q}\end{pmatrix}=\begin{pmatrix}2Z_{0\vec q}(A_+)^{1/2} & 2Z_{0\vec q}(A_-)^{1/2} & Y^*_{0\vec q}(\tilde\omega_{\vec q}/F_{0\vec q})^{1/2}+X^*_{0\vec q}(F_{0\vec q}/\tilde\omega_{\vec q})^{1/2} & Y^*_{0\vec q}(\tilde\omega_{\vec q}/F_{0\vec q})^{1/2}-X^*_{0\vec q}(F_{0\vec q}/\tilde\omega_{\vec q})^{1/2}\\ 2Z_{0\vec q}(A_-)^{1/2} & 2Z_{0\vec q}(A_+)^{1/2} & -Y^*_{0\vec q}(\tilde\omega_{\vec q}/F_{0\vec q})^{1/2}+X^*_{0\vec q}(F_{0\vec q}/\tilde\omega_{\vec q})^{1/2} & -Y^*_{0\vec q}(\tilde\omega_{\vec q}/F_{0\vec q})^{1/2}-X^*_{0\vec q}(F_{0\vec q}/\tilde\omega_{\vec q})^{1/2}\\ Y_{0\vec q}(E_{0\vec q}/\tilde\Omega_{0\vec q})^{1/2}+X_{0\vec q}(\tilde\Omega_{0\vec q}/E_{0\vec q})^{1/2} & Y_{0\vec q}(E_{0\vec q}/\tilde\Omega_{0\vec q})^{1/2}-X_{0\vec q}(\tilde\Omega_{0\vec q}/E_{0\vec q})^{1/2} & 2Z_{0\vec q}(B_+)^{1/2} & 2Z_{0\vec q}(B_-)^{1/2}\\ -Y_{0\vec q}(E_{0\vec q}/\tilde\Omega_{0\vec q})^{1/2}+X_{0\vec q}(\tilde\Omega_{0\vec q}/E_{0\vec q})^{1/2} & -Y_{0\vec q}(E_{0\vec q}/\tilde\Omega_{0\vec q})^{1/2}-X_{0\vec q}(\tilde\Omega_{0\vec q}/E_{0\vec q})^{1/2} & 2Z_{0\vec q}(B_-)^{1/2} & 2Z_{0\vec q}(B_+)^{1/2}\end{pmatrix}\begin{pmatrix}\hat d^\dagger_{0\vec q}\\ \hat d_{0-\vec q}\\ \hat\eta_{0\vec q}\\ \hat\eta^\dagger_{0\vec q}\end{pmatrix}. \quad (3.27b)$$

In (3.27a) and (3.27b), we have used the notation

$$X_{0\vec q}=-X^*_{0-\vec q}=-\frac{D^{cv}_{0\vec q}}{|D^{cv}_{0\vec q}|}\left(\frac{\tilde\omega_{\vec q}[W_{0\vec q}-(\tilde\Omega^2_{0\vec q}-\tilde\omega^2_{\vec q})]}{8\tilde\Omega_{0\vec q}W_{0\vec q}}\right)^{1/2}, \quad (3.28)$$

$$Y_{0\vec q}=-Y^*_{0-\vec q}=\frac{D^{cv}_{0\vec q}}{|D^{cv}_{0\vec q}|}\left(\frac{\tilde\Omega_{0\vec q}[W_{0\vec q}-(\tilde\Omega^2_{0\vec q}-\tilde\omega^2_{\vec q})]}{8\tilde\omega_{\vec q}W_{0\vec q}}\right)^{1/2}, \quad (3.29)$$

$$Z_{0\vec q}=Z^*_{0\vec q}=Z_{0-\vec q}=\frac{1}{2}\left(\frac{W_{0\vec q}+(\tilde\Omega^2_{0\vec q}-\tilde\omega^2_{\vec q})}{2W_{0\vec q}}\right)^{1/2}, \quad (3.30)$$

$$W_{0\vec q}=W^*_{0\vec q}=W_{0-\vec q}=\{(\tilde\Omega^2_{0\vec q}-\tilde\omega^2_{\vec q})^2$$
$$+4\tilde\omega_{\vec q}\tilde\Omega_{0\vec q}[(2g_{\vec q}/\hbar)|\tilde D^{cv}_{0\vec q}|]^2\}^{1/2}>0, \quad (3.31)$$

$$A_+=\frac{(E_{0\vec q}+\tilde\Omega_{0\vec q})^2}{4E_{0\vec q}\tilde\Omega_{0\vec q}}, \quad A_-=\frac{(E_{0\vec q}-\tilde\Omega_{0\vec q})^2}{4E_{0\vec q}\tilde\Omega_{0\vec q}}, \quad (3.32)$$

$$B_+=\frac{(\tilde\omega_{\vec q}+F_{0\vec q})^2}{4\tilde\omega_{\vec q}F_{0\vec q}}, \quad B_-=\frac{(\tilde\omega_{\vec q}-F_{0\vec q})^2}{4\tilde\omega_{\vec q}F_{0\vec q}}, \quad (3.33)$$

$$E_{0\vec q}=[\tfrac{1}{2}(\tilde\Omega^2_{0\vec q}+\tilde\omega^2_{\vec q})+W_{0\vec q}/2]^{1/2}, \quad (3.34a)$$

$$F_{0\vec q}=[\tfrac{1}{2}(\tilde\Omega^2_{0\vec q}+\tilde\omega^2_{\vec q})-W_{0\vec q}/2]^{1/2}. \quad (3.34b)$$

In addition, we note the relations

$$E^2_{0\vec q}+F^2_{0\vec q}=\tilde\Omega^2_{0\vec q}+\tilde\omega^2_{\vec q}, \quad E^2_{0\vec q}-F^2_{0\vec q}=W_{0\vec q}, \quad (3.35)$$

$$Z^2_{0\vec q}-X_{0\vec q}Y^*_{0\vec q}=\tfrac{1}{4}, \quad (3.36)$$

$$A_+-A_-=B_+-B_-=1. \quad (3.37)$$

It is easy to see now that the transformation (3.27) is unitary, and it leaves us with the following commutation relations of the polariton-creation and polariton-annihilation operators:

$$[\hat d_{0\vec q},\hat d^\dagger_{0\vec q'}]_-=[\hat\eta_{0\vec q},\hat\eta^\dagger_{0\vec q'}]_-=\delta_{\vec q',\vec q},$$
$$[\hat d_{0\vec q},\hat d_{0\vec q'}]_-=[\hat d^\dagger_{0\vec q},\hat d^\dagger_{0\vec q'}]_-=[\hat\eta_{0\vec q},\hat\eta_{0\vec q'}]_-=[\hat\eta^\dagger_{0\vec q},\hat\eta^\dagger_{0\vec q'}]_-=0. \quad (3.38)$$

The operators $\hat d^\dagger_{0\vec q}$, $\hat d_{0\vec q}$ commute with the operators $\hat\eta^\dagger_{0\vec q}$, $\hat\eta_{0\vec q}$. In the polariton representation, the Hamiltonian (3.25) gets the very simple form of two sets of decoupled oscillators, namely,

$$H'=\sum_{\vec q}\hbar E_{0\vec q}(\hat d^\dagger_{0\vec q}\hat d_{0\vec q}+\tfrac{1}{2})+\sum_{\vec q}\hbar F_{0\vec q}(\hat\eta^\dagger_{0\vec q}\hat\eta_{0\vec q}+\tfrac{1}{2}). \quad (3.39)$$

The polariton dispersion law has two independent branches belonging to the eigenfrequencies $E_{0\vec q}$ and $F_{0\vec q}$. The separation of the squares of the frequencies for special $\vec q$ values is shown in Fig. 3. In order to discuss the polariton dispersion law, let us expand the expressions (3.34a) and (3.34b) for the following different additional assumptions: (a) As long as in a certain range of $\vec q$ values $\tilde\Omega_{0\vec q}\rangle\tilde\omega_{\vec q}$, and

$$(\tilde\Omega^2_{0\vec q}-\tilde\omega^2_{\vec q})^2>4\tilde\omega_{\vec q}\tilde\Omega_{0\vec q}[(2g_{\vec q}/\hbar)|\tilde D^{cv}_{0\vec q}|]^2, \quad (3.40)$$

then (3.31) reads approximately

$$W_{0\vec{q}} \approx (\tilde{\Omega}^2_{0\vec{q}} - \tilde{\omega}^2_{\vec{q}}) + 2\tilde{\omega}_{\vec{q}} \frac{[(2g_{\vec{q}}/\hbar)|\tilde{D}^{cv}_{0\vec{q}}|]^2}{\tilde{\Omega}_{0\vec{q}}} , \quad (3.41)$$

so that the polariton eigenfrequencies become

$$E_{0\vec{q}} \approx \tilde{\Omega}_{0\vec{q}} + \frac{\tilde{\omega}_{\vec{q}}}{\tilde{\Omega}_{0\vec{q}}} \frac{[(2g_{\vec{q}}/\hbar)|\tilde{D}^{cv}_{0\vec{q}}|]^2}{2\tilde{\Omega}_{0\vec{q}}} , \quad (3.42a)$$

$$F_{0\vec{q}} \approx \tilde{\omega}_{\vec{q}} - \frac{[(2g_{\vec{q}}/\hbar)|\tilde{D}^{cv}_{0\vec{q}}|]^2}{2\tilde{\Omega}_{0\vec{q}}} . \quad (3.42b)$$

Hence, in this region the following inequalities are satisfied:

$$E_{0\vec{q}} > \tilde{\Omega}_{0\vec{q}} > \tilde{\omega}_{\vec{q}} > F_{0\vec{q}} , \quad (3.43)$$

$$\tilde{\omega}_{\vec{q}} - F_{0\vec{q}} > E_{0\vec{q}} - \tilde{\Omega}_{0\vec{q}} . \quad (3.44)$$

(b) For a special $\vec{q}$ value, $\vec{q} = \vec{k}_0$ say, let the resonance condition $\tilde{\omega}_{\vec{k}_0} = \tilde{\Omega}_{0\vec{k}_0}$ be fulfilled. Then we have

$$(W_{0\vec{k}_0})_{res} = 4\tilde{\Omega}_{0\vec{k}_0} \tilde{\Omega}'_{0\vec{k}_0} , \quad (3.45)$$

where

$$\tilde{\Omega}'_{0\vec{k}_0} = (g_{\vec{k}_0}/\hbar)|\tilde{D}^{cv}_{0\vec{k}_0}| . \quad (3.46)$$

The polariton eigenfrequencies now are

$$E_{0\vec{k}_0} = (\tilde{\Omega}^2_{0\vec{k}_0} + 2\tilde{\Omega}_{0\vec{k}_0}\tilde{\Omega}'_{0\vec{k}_0})^{1/2} \approx \tilde{\Omega}_{0\vec{k}_0} + \tilde{\Omega}'_{0\vec{k}_0} , \quad (3.47a)$$

$$F_{0\vec{k}_0} = (\tilde{\Omega}^2_{0\vec{k}_0} - 2\tilde{\Omega}_{0\vec{k}_0}\tilde{\Omega}'_{0\vec{k}_0})^{1/2} \approx \tilde{\Omega}_{0\vec{k}_0} - \tilde{\Omega}'_{0\vec{k}_0} , \quad (3.47b)$$

and we find in the resonance case

$$E_{0\vec{k}_0} > \tilde{\Omega}_{0\vec{k}_0} = \tilde{\omega}_{\vec{k}_0} > F_{0\vec{k}_0} . \quad (3.48)$$

(c) As long as in a certain range of $\vec{q}$ values $\tilde{\Omega}_{0\vec{q}} < \omega_{\vec{q}}$, and

$$(\tilde{\omega}^2_{\vec{q}} - \tilde{\Omega}^2_{0\vec{q}})^2 > 4\tilde{\omega}_{\vec{q}}\tilde{\Omega}_{0\vec{q}}[(2g_{\vec{q}}/\hbar)|\tilde{D}^{cv}_{0\vec{q}}|]^2 , \quad (3.40)$$

then (3.31) reads approximately

$$W_{0\vec{q}} \approx (\tilde{\omega}^2_{\vec{q}} - \tilde{\Omega}^2_{0\vec{q}}) + 2\tilde{\Omega}_{0\vec{q}} \frac{[(2g_{\vec{q}}/\hbar|\tilde{D}^{cv}_{0\vec{q}}|^2]^2}{\tilde{\omega}_{\vec{q}}} \quad (3.49)$$

so that the polariton eigenfrequencies in this case become

$$E_{0\vec{q}} \approx \tilde{\omega}_{\vec{q}} + \left(\frac{\tilde{\Omega}_{0\vec{q}}}{\tilde{\omega}_{\vec{q}}}\right) \frac{[(2g_{\vec{q}}/\hbar)|\tilde{D}^{cv}_{0\vec{q}}|]^2}{2\tilde{\omega}_{\vec{q}}} , \quad (3.50a)$$

$$F_{0\vec{q}} \approx \tilde{\Omega}_{0\vec{q}} - \frac{[(2g_{\vec{q}}/\hbar)|\tilde{D}^{cv}_{0\vec{q}}|]^2}{2\tilde{\omega}_{\vec{q}}} , \quad (3.50b)$$

and we have the following inequalities:

$$E_{0\vec{q}} > \tilde{\omega}_{\vec{q}} > \tilde{\Omega}_{0\vec{q}} > F_{0\vec{q}} , \quad (3.51)$$

$$\tilde{\Omega}_{0\vec{q}} - F_{0\vec{q}} > E_{0\vec{q}} - \tilde{\omega}_{\vec{q}} . \quad (3.52)$$

Our discussion gives rise to different dispersion laws depending on specified crystal data. Some possibilities are shown qualitatively in Fig. 4 assuming that the $\vec{q}$ dependence of $\tilde{\Omega}_{0\vec{q}}$ and $\tilde{\omega}_{\vec{q}}$ is known.

Normally, the two branches $E_{0\vec{q}}$ and $F_{0\vec{q}}$ in $\vec{q}$ space have no point of contact so that $E_{0\vec{q}} > F_{0\vec{q}}$ holds continuously for the same $\vec{q}$ value. But as it is shown in Fig. 4(b), a possible pathological case exists, when at $\vec{q} = 0$ we have $E_{00} = F_{00}$. This situation takes place if

$$\lim[(g_{\vec{q}}/\hbar)|\tilde{D}^{cv}_{0\vec{q}}|] = 0 \quad \text{as} \quad \vec{q} \to 0 ,$$

and if further $\tilde{\Omega}_{00} = \omega_p$. By neglecting the terms $H^{(2)}_{er\,II}$ and $H^{(2)}_C$ in the Hamiltonian (3.25), some changes in the polariton frequencies result. Instead of the quasi-photon and quasi-electron-hole-pair frequencies $\tilde{\omega}_{\vec{q}}$ and $\tilde{\Omega}_{0\vec{q}}$, respectively, the free frequencies $\omega_{\vec{q}}$ and $\Omega_{0\vec{q}}$ occur, whereas the Hamiltonian retains its character (3.25), but written now with photon and electron-hole-pair operators. A change of energy scale, putting

$$\epsilon_0 - \tfrac{1}{2} \textstyle\sum_{\vec{q}} \hbar\Omega_{0\vec{q}}$$

equal to zero, would be appropriate. Treating this system in the same way as it was done in previous sections, the resulting polariton frequencies would be instead of (3.34)

$$E_{0\vec{q}} = \{\tfrac{1}{2}[(\Omega^2_{0\vec{q}} + \omega^2_{\vec{q}}) + W_{0\vec{q}}]\}^{1/2}, \quad (3.53a)$$

$$F_{0\vec{q}} = \{\tfrac{1}{2}[(\Omega^2_{0\vec{q}} + \omega^2_{\vec{q}}) - W_{0\vec{q}})]\}^{1/2} , \quad (3.53b)$$

where now (3.31) is replaced by

$$W_{0\vec{q}} = \{(\Omega^2_{0\vec{q}} - \omega^2_{\vec{q}})^2 + 4\omega_{\vec{q}}\Omega_{0\vec{q}}[(2g_{\vec{q}}/\hbar)|D^{cv}_{0\vec{q}}|]^2\}^{1/2} > 0 . \quad (3.54)$$

The two branches of the dispersion law (3.53), which are shown in Fig. 5, have both excitonlike and photonlike regions. In the case of $\Omega_{0\vec{q}} > \omega_{\vec{q}}$, we have $\omega_{\vec{q}} - F_{0\vec{q}} > E_{0\vec{q}} - \Omega_{0\vec{q}}$, whereas in the opposite case $\Omega_{0\vec{q}} < \omega_{\vec{q}}$, we have $\Omega_{0\vec{q}} - F_{0\vec{q}} > E_{0\vec{q}} - \omega_{\vec{q}}$. This asymmetry is due to the virtual terms

$$\textstyle\sum_{\vec{q}} g_{\vec{q}} D^{cv}_{0\vec{q}} c^\dagger_{0\vec{q}} \xi^\dagger_{\vec{q}} \quad \text{and} \quad \sum_{\vec{q}} g_{\vec{q}} D^{cv}_{0\vec{q}} c_{0\,-\vec{q}} \xi_{-\vec{q}}$$

present in $H^{(2)}_{er\,I}$. A further consequence of such virtual processes is the fact that the polariton ground state, $|\Phi_0\rangle$ say, is different from the free vacuum $|0\rangle$.

Let us return to our system (3.25). In order to get an additional information on the polariton behavior, we look for expectation values of the current operator which is defined as

$$\vec{J} = (e/m) \int d^3r\, \Psi^\dagger(\vec{r})[\vec{p} - (e/c)\vec{A}]\Psi(\vec{r}) . \quad (3.55)$$

Inserting (1.2) and (1.4) into (3.55), switching to the electron-hole-pair description, and reducing the resulting expression to the simplified crystal model of interest in this section leads to

$$\vec{J} = \vec{J}^{(2)}_c + \vec{J}^{(2)}_p + \vec{J}^{(3)}_p , \quad (3.56)$$

where

$$\vec{J}^{(2)}_C = -e \sum_{\vec{q}} \vec{v}^v_{\vec{q}} c^\dagger_{0\vec{q}} c_{0\vec{q}} , \quad (3.57)$$

$$\vec{J}_p^{(2)} = -e \sum_{\vec q} g_{\vec q}\vec e_{\vec q} v_{0\vec q}^{cv}(c_{0\vec q}^\dagger + c_{0\,-\vec q})(\xi_{\vec q}^\dagger + \xi_{-\vec q}) \;, \qquad (3.58)$$

$$\vec{J}_p^{(3)} = e \sum_{\vec q,\vec k} g_{\vec q}\vec e_{\vec q} v_{\vec k+\vec q\,\vec k}^{v\;v} c_{0\vec k}^\dagger c_{0\,\vec k+\vec q}(\xi_{\vec q} + \xi_{-\vec q}^\dagger) \;. \qquad (3.59)$$

In (3.57), the electron velocity is given by

$$\vec v_{\vec k}^{\mu} = (1/\hbar)\vec\nabla_{\vec k}\epsilon_{\mu\vec k} \;. \qquad (3.60)$$

We have neglected the nonlinear parts H_{nl} of the total Hamiltonian in the present discussion; for that reason, it is consistent to regard only the bilinear terms of the current operator, i.e., the contributions $\vec J_c^{(2)}$ and $\vec J_p^{(2)}$. The polarization current $\vec J_p^{(2)}$ has no diagonal elements in the electron-hole-pair description, whereas in the polariton representation it has diagonal as well as nondiagonal elements with respect to polariton eigenstates. The diagonal terms show that there exists a real polarizability of our medium. After having transformed (3.57) and (3.58) into the polariton representation with the aid of (3.27), we see at once that, as we have expected, there exists no ground-state current. The polariton ground state, which is different from the vacuum state $|0\rangle$, is denoted by $|\Phi_0\rangle$, and we find

$$\langle\Phi_0|\vec J_c^{(2)}|\Phi_0\rangle = \langle\Phi_0|\vec J_p^{(2)}|\Phi_0\rangle = 0 \;, \qquad (3.61)$$

In the excited one-polariton states $\hat d_{0\vec q}^\dagger|\Phi_0\rangle$ and $\hat\eta_{0\vec q}^\dagger|\Phi_0\rangle$, respectively, we get the expectation values

$$\langle\Phi_0|\hat d_{0\vec q}\vec J_c^{(2)}\hat d_{0\vec q}^\dagger|\Phi_0\rangle$$

$$= -e\vec v_{\vec q}^{\,v}\,\frac{W_{0\vec q} + (\tilde\Omega_{0\vec q}^2 - \tilde\omega_{\vec q}^2)}{2W_{0\vec q}} = -\langle\vec J_c^{(+)}(\vec q)\rangle \;, \qquad (3.62a)$$

$$\langle\Phi_0|\hat\eta_{0\vec q}\vec J_c^{(2)}\hat\eta_{0\vec q}^\dagger|\Phi_0\rangle$$

$$= e\vec v_{\vec q}^{\,v}\,\frac{W_{0\vec q} - (\tilde\Omega_{0\vec q}^2 - \tilde\omega_{\vec q}^2)}{2W_{0\vec q}} = \langle\vec J_c^{(-)}(\vec q)\rangle \;. \qquad (3.62b)$$

With respect to (3.60), we have

$$\langle\vec J_c^{(+)}(-\vec q)\rangle = -\langle\vec J_c^{(+)}(\vec q)\rangle$$

and

$$\langle\vec J_c^{(-)}(-\vec q)\rangle = -\langle\vec J_c^{(-)}(\vec q)\rangle \;,$$

so that further

$$\sum_{\vec q}\langle\vec J_c^{(+)}(\vec q)\rangle = \sum_{\vec q}\langle\vec J_c^{(-)}(\vec q)\rangle = 0 \;.$$

In the resonance case, at $\vec q = \vec k$ say, (3.62) reduces to

$$\langle\vec J_c^{(+)}(\vec k)\rangle_{res} = \langle\vec J_c^{(-)}(\vec k)\rangle_{res} = \tfrac{1}{2}e\vec v_{\vec k}^{\,v} \;.$$

A much more remarkable result is the agreement of the expectation values of the polarization current $\vec J_p^{(2)}$ in the two energetically different polariton states $\hat d_{0\vec q}^\dagger|\Phi_0\rangle$ and $\hat\eta_{0\vec q}^\dagger|\Phi_0\rangle$,

$$\langle\Phi_0|\hat d_{0\vec q}\vec J_p^{(2)}\hat d_{0\vec q}^\dagger|\Phi_0\rangle$$

$$= \langle\Phi_0|\hat\eta_{0\vec q}\vec J_p^{(2)}\hat\eta_{0\vec q}^\dagger|\Phi_0\rangle$$

$$= \frac{e g_{\vec q}\vec e_{\vec q}\omega_{\vec q}}{W_{0\vec q}}\left(\frac{2g_{\vec q}}{\hbar}D_{0\vec q}^{cv*}v_{0\vec q}^{cv} + \frac{2g_{\vec q}}{\hbar}D_{0\vec q}^{cv}v_{0\vec q}^{cv*}\right) = \langle\vec J_p(\vec q)\rangle \;. \qquad (3.63)$$

Because of (3.63), we can say that the polariton states $\hat d_{0\vec q}^\dagger|\Phi_0\rangle$ and $\hat\eta_{0\vec q}^\dagger|\Phi_0\rangle$, which belong to different energies, are *states of equal polarizability*.

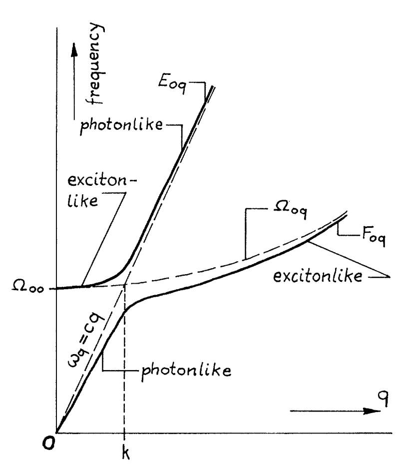

FIG. 5. Polarition dispersion law where $H_{er\,II}^{(2)}$ and $H_c^{(2)}$ are neglected. The wave vector $\vec q = \vec k$ refers to a resonance position.

IV. TRANSITION AMPLITUDES OF SOME NONLINEAR EFFECTS

We have shown that – in agreement with Hopfield[5] – the propagation of radiation in the medium is best described by polariton waves. Our model of Sec. III predicts two polariton levels with the energies $\hbar F_{0\vec q}$ and $\hbar E_{0\vec q}$, respectively, and consequently, we have *two* optical refractive indices

$$(n_{\vec q}^{\alpha})^2 = \omega_{\vec q}^2/F_{0\vec q}^2 = 2\omega_{\vec q}^2/[(\tilde\Omega_{0\vec q}^2 + \tilde\omega_{\vec q}^2) - W_{0\vec q}] \;, \qquad (4.1a)$$

$$(n_{\vec q}^{\beta})^2 = \omega_{\vec q}^2/E_{0\vec q}^2 = 2\omega_{\vec q}^2/[(\tilde\Omega_{0\vec q}^2 + \tilde\omega_{\vec q}^2) + W_{0\vec q}] \;, \qquad (4.1b)$$

where α denotes the lower, and β the higher polariton level.[18] In the frequency range larger than E_{00}, the possibility arises that for two different wave vectors $\vec k$ and $\vec k'$, say, the frequencies $E_{0\vec k}$ and $F_{0\vec k'}$ coincide. This situation is illustrated in Fig. 6.

If highly intense electromagnetic fields, as they are available from lasers, interact with the crystal, the anharmonic terms should no longer be neglected. Because of the interaction H_{nl} polaritons of different wave vectors are coupled, and H_{nl}, for instance, causes transitions between the polariton levels α and β. Treating the nonlinearities

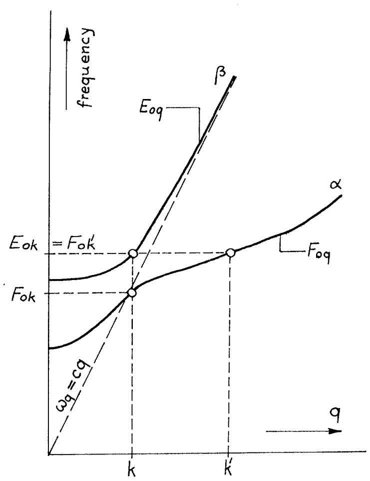

FIG. 6. Representation of the polariton levels α and β.

$$H_{\mathrm{nl}}=H_{\mathrm{nl}}^{(3)}+H_{\mathrm{nl}}^{(4)} \tag{4.2}$$

with

$$H_{\mathrm{nl}}^{(3)}=H_{\mathrm{C}}^{(3)}+H_{\mathrm{er\,I}}^{(3)}+H_{\mathrm{er\,II}}^{(3)} \tag{4.3}$$

and

$$H_{\mathrm{nl}}^{(4)}=H_{\mathrm{C}}^{(4)}+H_{\mathrm{er\,II}}^{(4)} \tag{4.4}$$

in the polariton representation (3.27) as a perturbation, we get in first-order perturbation-theory nonlinear effects of third and fourth[19] order. A third-order effect, for instance, is the fusion of two polaritons whereby a new one arises, and an example of a fourth-order process is the annihilation of three initially present polaritons whereby a new one is created. We do not give here a complete analysis of all possible nonlinear transitions which could be obtained for our model in first-order perturbation theory, though this would easily be possible within the proposed frame, but we conclude the present investigations with the results for third- and fourth-order transition amplitudes for processes where by polariton fusion a sum frequency, a second- and a third-harmonic polariton oscillation, respectively, arises.

A. Generation of Sum Frequency by Two-Polariton Fusion

A multiple occupation of polariton states in our system is possible, and we consider the case where at time $t_0=0$ two polariton states with wave vectors $\vec{k}_1$ and $\vec{k}_2$, frequencies $F_{0\vec{k}_1}$ and $F_{0\vec{k}_2}$, and occupation numbers $N_{\vec{k}_1}^{\alpha}$ and $N_{\vec{k}_2}^{\alpha}$ on the level α are excited, while the level β is empty. The normalized state describing this situation is denoted by $|0;N_{\vec{k}_1}^{\alpha},N_{\vec{k}_2}^{\alpha}\rangle$. Under the influence of H_{nl}, occasionally a polariton $\vec{k}_1$ and a polariton $\vec{k}_2$ on the level α is annihilated, and a new one on the level β with the frequency $E_{0\vec{k}_3}=F_{0\vec{k}_1}+F_{0\vec{k}_2}$ is created. In first-order perturbation theory the transition amplitude for this process is given by

$$\langle (N_{\vec{k}_2}-1)^{\alpha},(N_{\vec{k}_1}-1)^{\alpha};1_{\vec{k}_3}^{\beta}|H_{\mathrm{nl}}^{(3)}|0;N_{\vec{k}_1}^{\alpha},N_{\vec{k}_2}^{\alpha}\rangle\ . \tag{4.5}$$

Transforming $H_{\mathrm{nl}}^{(3)}$ into the polariton representation, and paying attention to the time-reversal properties of Sec. I, we find

$$\begin{aligned}
\langle (N_{\vec{k}_2}-1)^{\alpha},(N_{\vec{k}_1}-1)^{\alpha};1_{\vec{k}_3}^{\beta}|H_{\mathrm{C}}^{(3)}|0;N_{\vec{k}_1}^{\alpha},N_{\vec{k}_2}^{\alpha}\rangle = &-\delta_{\vec{k}_3,-(\vec{k}_1+\vec{k}_2)}(N_{\vec{k}_1}^{\alpha}N_{\vec{k}_2}^{\alpha})^{1/2}\,\frac{2\pi e^2 D_{0\vec{k}_1}^{cv*}D_{0\vec{k}_2}^{cv*}}{\mathrm{vol}\,|D_{0\vec{k}_1}^{cv}|\,|D_{0\vec{k}_2}^{cv}|}\\
&\times\left(\frac{W_{0\vec{k}_1}-(\tilde{\Omega}_{0\vec{k}_1}^2-\tilde{\omega}_{\vec{k}_1}^2)}{2W_{0\vec{k}_1}}\right)^{1/2}\left(\frac{W_{0\vec{k}_2}-(\tilde{\Omega}_{0\vec{k}_2}^2-\tilde{\omega}_{\vec{k}_2}^2)}{2W_{0\vec{k}_2}}\right)^{1/2}\left(\frac{W_{0\vec{k}_1+\vec{k}_2}+(\tilde{\Omega}_{0\vec{k}_1+\vec{k}_2}^2-\tilde{\omega}_{\vec{k}_1+\vec{k}_2}^2)}{2W_{0\vec{k}_1+\vec{k}_2}}\right)^{1/2}\\
&\times\left\{\frac{v_{0\vec{k}_1}^{cv}v_{\vec{k}_2\vec{k}_2+\vec{k}_1}^{vv*}}{|\vec{k}_1|^2}\left(\frac{\Omega_{0\vec{k}_1}F_{0\vec{k}_1}}{\tilde{\Omega}_{0\vec{k}_1}^2}\right)^{1/2}\left[\left(\frac{\Omega_{0\vec{k}_2}F_{0\vec{k}_2}\Omega_{0\vec{k}_1+\vec{k}_2}E_{0\vec{k}_1+\vec{k}_2}}{\tilde{\Omega}_{0\vec{k}_2}^2\tilde{\Omega}_{0\vec{k}_1+\vec{k}_2}^2}\right)^{1/2}+\left(\frac{\tilde{\Omega}_{0\vec{k}_2}^2\tilde{\Omega}_{0\vec{k}_1+\vec{k}_2}^2}{\Omega_{0\vec{k}_2}F_{0\vec{k}_2}\Omega_{0\vec{k}_1+\vec{k}_2}E_{0\vec{k}_1+\vec{k}_2}}\right)^{1/2}\right]\right.\\
&+\frac{v_{0\vec{k}_2}^{cv}v_{\vec{k}_1\vec{k}_1+\vec{k}_2}^{vv*}}{|\vec{k}_2|^2}\left(\frac{\Omega_{0\vec{k}_2}F_{0\vec{k}_2}}{\tilde{\Omega}_{0\vec{k}_2}^2}\right)^{1/2}\left[\left(\frac{\Omega_{0\vec{k}_1}F_{0\vec{k}_1}\Omega_{0\vec{k}_1+\vec{k}_2}E_{0\vec{k}_1+\vec{k}_2}}{\tilde{\Omega}_{0\vec{k}_1}^2\tilde{\Omega}_{0\vec{k}_1+\vec{k}_2}^2}\right)^{1/2}+\left(\frac{\tilde{\Omega}_{0\vec{k}_1}^2\tilde{\Omega}_{0\vec{k}_1+\vec{k}_2}^2}{\Omega_{0\vec{k}_1}F_{0\vec{k}_1}\Omega_{0\vec{k}_1+\vec{k}_2}E_{0\vec{k}_1+\vec{k}_2}}\right)^{1/2}\right]\\
&\left.+\frac{v_{0\vec{k}_1+\vec{k}_2}^{cv*}v_{-\vec{k}_1\vec{k}_2}^{vv}}{|\vec{k}_1+\vec{k}_2|^2}\left(\frac{\Omega_{0\vec{k}_1+\vec{k}_2}E_{0\vec{k}_1+\vec{k}_2}}{\tilde{\Omega}_{0\vec{k}_1+\vec{k}_2}^2}\right)^{1/2}\left[\left(\frac{\Omega_{0\vec{k}_1}F_{0\vec{k}_1}\Omega_{0\vec{k}_2}F_{0\vec{k}_2}}{\tilde{\Omega}_{0\vec{k}_1}^2\tilde{\Omega}_{0\vec{k}_2}^2}\right)^{1/2}-\left(\frac{\tilde{\Omega}_{0\vec{k}_1}^2\tilde{\Omega}_{0\vec{k}_2}^2}{\Omega_{0\vec{k}_1}F_{0\vec{k}_1}\Omega_{0\vec{k}_2}F_{0\vec{k}_2}}\right)^{1/2}\right]\right\},
\end{aligned} \tag{4.6}$$

$$\langle (N_{\vec{k}_2}-1)^{\alpha},(N_{\vec{k}_1}-1)^{\alpha};1_{\vec{k}_3}^{\beta}|H_{\mathrm{er\,I}}^{(3)}|0;N_{\vec{k}_1}^{\alpha},N_{\vec{k}_2}^{\alpha}\rangle=\delta_{\vec{k}_3,-(\vec{k}_1+\vec{k}_2)}\,\tfrac{1}{2}(N_{\vec{k}_1}^{\alpha}N_{\vec{k}_2}^{\alpha})^{1/2}$$

$$
\times\Bigg\{ g_{\vec{k}_1} D^{v}_{\vec{k}_2+\vec{k}_1 \vec{k}_2}\, \frac{D^{cv*}_{0\vec{k}_2}}{|D^{cv}_{0\vec{k}_2}|}\left(\frac{W_{0\vec{k}_1}+(\tilde{\Omega}^2_{0\vec{k}_1}-\tilde{\omega}^2_{\vec{k}_1})}{2W_{0\vec{k}_1}}\right)^{1/2}\left(\frac{W_{0\vec{k}_2}-(\tilde{\Omega}^2_{0\vec{k}_2}-\tilde{\omega}^2_{\vec{k}_2})}{2W_{0\vec{k}_2}}\right)^{1/2}\left(\frac{W_{0\vec{k}_1+\vec{k}_2}+(\tilde{\Omega}^2_{0\vec{k}_1+\vec{k}_2}-\tilde{\omega}^2_{\vec{k}_1+\vec{k}_2})}{2W_{0\vec{k}_1+\vec{k}_2}}\right)^{1/2}
$$

$$
\times\left(\frac{\omega_{\vec{k}_1}}{F_{0\vec{k}_1}}\right)^{1/2}\left[\left(\frac{\tilde{\Omega}_{0\vec{k}_2}\Omega_{0\vec{k}_1+\vec{k}_2}E_{0\vec{k}_1+\vec{k}_2}}{\Omega_{0\vec{k}_2}F_{0\vec{k}_2}\tilde{\Omega}^2_{0\vec{k}_1+\vec{k}_2}}\right)^{1/2}+\left(\frac{\Omega_{0\vec{k}_2}F_{0\vec{k}_2}\tilde{\Omega}^2_{0\vec{k}_1+\vec{k}_2}}{\tilde{\Omega}^2_{0\vec{k}_2}\Omega_{0\vec{k}_1+\vec{k}_2}E_{0\vec{k}_1+\vec{k}_2}}\right)^{1/2}\right]+g_{\vec{k}_2}D^{v}_{\vec{k}_1+\vec{k}_2\vec{k}_1}\,\frac{D^{cv*}_{0\vec{k}_1}}{|D^{cv}_{0\vec{k}_1}|}
$$

$$
\times\left(\frac{W_{0\vec{k}_1}-(\tilde{\Omega}^2_{0\vec{k}_1}-\tilde{\omega}^2_{\vec{k}_1})}{2W_{0\vec{k}_1}}\right)^{1/2}\left(\frac{W_{0\vec{k}_2}+(\tilde{\Omega}^2_{0\vec{k}_2}-\tilde{\omega}^2_{\vec{k}_2})}{2W_{0\vec{k}_2}}\right)^{1/2}\left(\frac{W_{0\vec{k}_1+\vec{k}_2}+(\tilde{\Omega}^2_{0\vec{k}_1+\vec{k}_2}-\tilde{\omega}^2_{\vec{k}_1+\vec{k}_2})}{2W_{0\vec{k}_1+\vec{k}_2}}\right)^{1/2}
$$

$$
\times\left(\frac{\omega_{\vec{k}_2}}{F_{0\vec{k}_2}}\right)^{1/2}\left[\left(\frac{\tilde{\Omega}^2_{0\vec{k}_1}\Omega_{0\vec{k}_1+\vec{k}_2}E_{0\vec{k}_1+\vec{k}_2}}{\Omega_{0\vec{k}_1}F_{0\vec{k}_1}\tilde{\Omega}^2_{0\vec{k}_1+\vec{k}_2}}\right)^{1/2}+\left(\frac{\Omega_{0\vec{k}_1}F_{0\vec{k}_1}\tilde{\Omega}^2_{0\vec{k}_1+\vec{k}_2}}{\tilde{\Omega}^2_{0\vec{k}_1}\Omega_{0\vec{k}_1+\vec{k}_2}E_{0\vec{k}_1+\vec{k}_2}}\right)^{1/2}\right]
$$

$$
+\,g_{\vec{k}_1+\vec{k}_2}D^{v}_{-\vec{k}_1\vec{k}_2}\,\frac{D^{cv*}_{0\vec{k}_1}D^{cv*}_{0\vec{k}_2}D^{cv*}_{0\vec{k}_1+\vec{k}_2}}{|D^{cv}_{0\vec{k}_1}|\,|D^{cv}_{0\vec{k}_2}|\,|D^{cv}_{0\vec{k}_1+\vec{k}_2}|}\left(\frac{W_{0\vec{k}_1}-(\tilde{\Omega}^2_{0\vec{k}_1}-\tilde{\omega}^2_{\vec{k}_1})}{2W_{0\vec{k}_1}}\right)^{1/2}\left(\frac{W_{0\vec{k}_2}-(\tilde{\Omega}^2_{0\vec{k}_2}-\tilde{\omega}^2_{\vec{k}_2})}{2W_{0\vec{k}_2}}\right)^{1/2}
$$

$$
\times\left(\frac{W_{0\vec{k}_1+\vec{k}_2}-(\tilde{\Omega}^2_{0\vec{k}_1+\vec{k}_2}-\tilde{\omega}^2_{\vec{k}_1+\vec{k}_2})}{2W_{0\vec{k}_1+\vec{k}_2}}\right)^{1/2}\left(\frac{\omega_{\vec{k}_1+\vec{k}_2}}{E_{0\vec{k}_1+\vec{k}_2}}\right)^{1/2}\left[\left(\frac{\tilde{\Omega}^2_{0\vec{k}_1}\Omega_{0\vec{k}_2}F_{0\vec{k}_2}}{\Omega_{0\vec{k}_1}F_{0\vec{k}_1}\tilde{\Omega}^2_{0\vec{k}_2}}\right)^{1/2}-\left(\frac{\Omega_{0\vec{k}_1}F_{0\vec{k}_1}\tilde{\Omega}^2_{0\vec{k}_2}}{\tilde{\Omega}^2_{0\vec{k}_1}\Omega_{0\vec{k}_2}F_{0\vec{k}_2}}\right)^{1/2}\right]\Bigg\}, \qquad (4.7)
$$

and

$$
\langle N_{\vec{k}_2}-1)^{\alpha},\,(N_{\vec{k}_1}-1)^{\alpha};\,1^{\beta}_{\vec{k}_3}\,|\,H^{(3)}_{\mathrm{er\,II}}\,|\,0;\,N^{\alpha}_{\vec{k}_1},\,N^{\alpha}_{\vec{k}_2}\rangle=\delta_{\vec{k}_3,-(\vec{k}_1+\vec{k}_2)}(N^{\alpha}_{\vec{k}_1}N^{\alpha}_{\vec{k}_2})^{1/2}\Bigg[2f_{\vec{k}_1\vec{k}_2}v^{cv*}_{0\vec{k}_1+\vec{k}_2}\left(\frac{W_{0\vec{k}_1}+(\tilde{\Omega}^2_{0\vec{k}_1}-\tilde{\omega}^2_{\vec{k}_1})}{2W_{0\vec{k}_2}}\right)^{1/2}
$$

$$
\times\left(\frac{W_{0\vec{k}_2}+(\tilde{\Omega}^2_{0\vec{k}_2}-\tilde{\omega}^2_{\vec{k}_2})}{2W_{0\vec{k}_2}}\right)^{1/2}\left(\frac{W_{0\vec{k}_1+\vec{k}_2}+(\tilde{\Omega}^2_{0\vec{k}_1+\vec{k}_2}-\tilde{\omega}^2_{\vec{k}_1+\vec{k}_2})}{2W_{0\vec{k}_1+\vec{k}_2}}\right)^{1/2}\left(\frac{\omega_{\vec{k}_1}\omega_{\vec{k}_2}\Omega_{0\vec{k}_1+\vec{k}_2}E_{0\vec{k}_1+\vec{k}_2}}{F_{0\vec{k}_1}F_{0\vec{k}_2}\tilde{\Omega}^2_{0\vec{k}_1+\vec{k}_2}}\right)^{1/2}
$$

$$
-f_{\vec{k}_2\vec{k}_2+\vec{k}_1}v^{cv}_{0\vec{k}_1}\,\frac{D^{cv}_{0\vec{k}_1}(D^{cv}_{0\vec{k}_1+\vec{k}_2}+D^{cv*}_{0\vec{k}_1+\vec{k}_2})}{|D^{cv}_{0\vec{k}_1}|\,|D^{cv}_{0\vec{k}_1+\vec{k}_2}|}\left(\frac{W_{0\vec{k}_1}-(\tilde{\Omega}^2_{0\vec{k}_1}-\tilde{\omega}^2_{\vec{k}_1})}{2W_{0\vec{k}_1}}\right)^{1/2}
$$

$$
\times\left(\frac{W_{0\vec{k}_2}+(\tilde{\Omega}^2_{0\vec{k}_2}-\tilde{\omega}^2_{\vec{k}_2})}{2W_{0\vec{k}_2}}\right)^{1/2}\left(\frac{W_{0\vec{k}_1+\vec{k}_2}-(\tilde{\Omega}^2_{0\vec{k}_1+\vec{k}_2}-\tilde{\omega}^2_{\vec{k}_1+\vec{k}_2})}{2W_{0\vec{k}_1+\vec{k}_2}}\right)^{1/2}\left(\frac{\Omega_{0\vec{k}_1}F_{0\vec{k}_1}\omega_{\vec{k}_2}\omega_{\vec{k}_1+\vec{k}_2}}{\Omega^2_{0\vec{k}_1}F_{0\vec{k}_2}E_{0\vec{k}_1+\vec{k}_2}}\right)^{1/2}
$$

$$
-f_{\vec{k}_1+\vec{k}_2\vec{k}_1}v^{cv}_{0\vec{k}_2}\,\frac{D^{cv}_{0\vec{k}_2}(D^{cv}_{0\vec{k}_1+\vec{k}_2}+D^{cv*}_{0\vec{k}_1+\vec{k}_2})}{|D^{cv}_{0\vec{k}_2}|\,|D^{cv}_{0\vec{k}_1+\vec{k}_2}|}\left(\frac{W_{0\vec{k}_1}+(\tilde{\Omega}^2_{0\vec{k}_1}-\tilde{\omega}^2_{\vec{k}_1})}{2W_{0\vec{k}_1}}\right)^{1/2}
$$

$$
\times\left(\frac{W_{0\vec{k}_2}-(\tilde{\Omega}^2_{0\vec{k}_2}-\tilde{\omega}^2_{\vec{k}_2})}{2W_{0\vec{k}_2}}\right)^{1/2}\left(\frac{W_{0\vec{k}_1+\vec{k}_2}-(\tilde{\Omega}^2_{0\vec{k}_1+\vec{k}_2}-\tilde{\omega}^2_{\vec{k}_1+\vec{k}_2})}{2W_{0\vec{k}_1+\vec{k}_2}}\right)^{1/2}\left(\frac{\omega_{\vec{k}_1}\Omega_{0\vec{k}_1}F_{0\vec{k}_2}\omega_{\vec{k}_1+\vec{k}_2}}{F_{0\vec{k}_1}\tilde{\Omega}^2_{0\vec{k}_2}E_{0\vec{k}_1+\vec{k}_2}}\right)^{1/2}\Bigg]. \qquad (4.8)
$$

The transition probability essentially depends on the occupation numbers initially present, and the created polariton with the frequency $E_{0\vec{k}_3}=F_{0\vec{k}_1}+F_{0\vec{k}_2}$ must in addition fulfill the condition $\vec{k}_1+\vec{k}_2+\vec{k}_3=0$. This wave-vector condition, of course, refers to polaritons, and not to single photons. The interaction Hamiltonian $H^{(3)}_{\mathrm{nl}}$ even causes transitions which take place only on a single polariton level. A polariton $\vec{k}_3$ with the frequency $F_{0\vec{k}_3}=F_{0\vec{k}_1}+F_{0\vec{k}_2}$, which, for instance, arises while two polaritons $(\vec{k}_1;\ F_{0\vec{k}_1})$ and $(\vec{k}_2;\ F_{0\vec{k}_2})$ on the same level are annihilated, has to obey the condition $\vec{k}_1+\vec{k}_2-\vec{k}_3=0$.

B. Generation of a Second-Harmonic Polariton

We consider the case where initially $N^{\alpha}_{\vec{k}_1}$ polaritons $(\vec{k}_1;\ F_{0\vec{k}_1})$ are excited on the level α. The transition amplitude

$$\langle (N_{\vec k_1}-2)^\alpha;\ 1^\beta_{\vec k_2}|\,H^{(3)}_{\mathrm{nl}}\,|0;\ N^\alpha_{\vec k_1}\rangle$$

for the case of a second-harmonic polariton generation ($\vec k_2;\ E_{0\vec k_2}$) on the level β during the annihilation of two polaritons ($\vec k_1;\ F_{0\vec k_1}$) consists of

$$\langle (N_{\vec k_1}-2)^\alpha;\ 1^\beta_{\vec k_2}|\,H^{(3)}_{C}\,|0;\ N^\alpha_{\vec k_1}\rangle = \delta_{\vec k_2,-2\vec k_1}[N^\alpha_{\vec k_1}(N_{\vec k_1}-1)^\alpha]^{1/2}\frac{\pi e^2(D^{cv*}_{0\vec k_1})^2}{\mathrm{vol}\,|D^{cv}_{0\vec k_1}|^2}\,\frac{W_{0\vec k_1}-(\tilde\Omega^2_{0\vec k_1}-\tilde\omega^2_{\vec k_1})}{2W_{0\vec k_1}}$$
$$\times\left(\frac{W_{02\vec k_1}+(\tilde\Omega^2_{02\vec k_1}-\tilde\omega^2_{2\vec k_1})}{2W_{02\vec k_1}}\right)^{1/2}\left\{\frac{v^{cv*}_{02\vec k_1}v^{vv}_{-\vec k_1\vec k_1}}{4|\vec k_1|^2}\left(\frac{\Omega_{02\vec k_1}E_{02\vec k_1}}{\tilde\Omega^2_{02\vec k_1}}\right)^{1/2}\left(\frac{\tilde\Omega^2_{0\vec k_1}}{\Omega_{0\vec k_1}F_{0\vec k_1}}-\frac{\Omega_{0\vec k_1}F_{0\vec k_1}}{\tilde\Omega^2_{0\vec k_1}}\right)\right.$$
$$\left.-2\frac{v^{cv}_{0\vec k_1}v^{vv*}_{\vec k_1 2\vec k_1}}{|\vec k_1|^2}\left[\left(\frac{\tilde\Omega^2_{02\vec k_1}}{\Omega_{02\vec k_1}E_{02\vec k_1}}\right)^{1/2}+\frac{\Omega_{0\vec k_1}F_{0\vec k_1}}{\tilde\Omega^2_{02\vec k_1}}\left(\frac{\Omega_{02k_1}E_{02k_1}}{\tilde\Omega^2_{02\vec k_1}}\right)^{1/2}\right]\right\}, \qquad (4.9)$$

$$\langle (N_{\vec k_1}-2)^\alpha;\ 1^\beta_{\vec k_2}|\,H^{(3)}_{\mathrm{er\,I}}\,|0;\ N^\alpha_{\vec k_1}\rangle = \delta_{\vec k_2,-2\vec k_1}[N^\alpha_{\vec k_1}(N_{\vec k_1}-1)^\alpha]^{1/2}\left\{\frac{g_{\vec k_1}D^{vv}_{\vec k_1 2\vec k_1}g_{\vec k_1}D^{cv*}_{0\vec k_1}\omega_{\vec k_1}}{\hbar W_{0\vec k_1}}\left(\frac{W_{02\vec k_1}+(\tilde\Omega_{02\vec k_1}-\tilde\omega_{2\vec k_1})}{2W_{02\vec k_1}}\right)^{1/2}\right.$$
$$\times\left[\left(\frac{\tilde\Omega^2_{02\vec k_1}}{\Omega_{02\vec k_1}E_{02\vec k_1}}\right)^{1/2}+\frac{\tilde\Omega^2_{0\vec k_1}}{\Omega_{0\vec k_1}F_{0\vec k_1}}\left(\frac{\Omega_{02\vec k_1}E_{02\vec k_1}}{\tilde\Omega^2_{02\vec k_1}}\right)^{1/2}\right]-\frac{g_{2\vec k_1}D^{vv}_{-\vec k_1\vec k_1}D^{cv*}_{02\vec k_1}(D^{cv*}_{0\vec k_1})^2}{4|D^{cv}_{02\vec k_1}|\,|D^{cv}_{0\vec k_1}|^2}$$
$$\left.\times\frac{W_{0\vec k_1}-(\tilde\Omega^2_{0\vec k_1}-\tilde\omega^2_{\vec k_1})}{2W_{0\vec k_1}}\left(\frac{W_{02\vec k_1}-(\tilde\Omega^2_{02\vec k_1}-\tilde\omega^2_{2\vec k_1})}{2W_{02\vec k_1}}\right)^{1/2}\times\left(\frac{\omega_{2\vec k_1}}{E_{02\vec k_1}}\right)^{1/2}\left(\frac{\tilde\Omega^2_{0\vec k_1}}{\Omega_{0\vec k_1}F_{0\vec k_1}}-\frac{\Omega_{0\vec k_1}F_{0\vec k_1}}{\tilde\Omega^2_{0\vec k_1}}\right)\right\}, \qquad (4.10)$$

and

$$\langle (N_{\vec k_1}-2)^\alpha;\ 1^\beta_{\vec k_2}|\,H^{(3)}_{\mathrm{er\,II}}\,|0;\ N^\alpha_{\vec k_1}\rangle = \delta_{\vec k_2,-2\vec k_1}[N^\alpha_{\vec k_1}(N_{\vec k_1}-1)^\alpha]^{1/2}$$
$$\times\left\{f_{\vec k_1\vec k_1}v^{cv*}_{02\vec k_1}\frac{W_{0\vec k_1}+(\tilde\Omega^2_{0\vec k_1}-\tilde\omega^2_{\vec k_1})}{2W_{0\vec k_1}}\left(\frac{W_{02\vec k_1}+(\tilde\Omega^2_{02\vec k_1}-\tilde\omega^2_{2\vec k_1})}{2W_{02\vec k_1}}\right)^{1/2}\frac{\omega_{\vec k_1}}{F_{0\vec k_1}}\left(\frac{\Omega_{02k_1}E_{02k_1}}{\tilde\Omega^2_{02\vec k_1}}\right)^{1/2}\right.$$
$$\left.-4f_{2\vec k_1\vec k_1}v^{cv}_{0\vec k_1}\frac{g_{\vec k_1}D^{cv}_{0\vec k_1}D^{cv*}_{02\vec k_1}\omega_{\vec k_1}}{\hbar W_{0\vec k_1}|D^{cv}_{02\vec k_1}|}\left(\frac{W_{02\vec k_1}-(\tilde\Omega^2_{02\vec k_1}-\tilde\omega^2_{2\vec k_1})}{2W_{02\vec k_1}}\right)^{1/2}\left(\frac{\omega_{2\vec k_1}}{E_{02\vec k_1}}\right)^{1/2}\right\}. \qquad (4.11)$$

The second-harmonic polariton with the frequency $E_{0\vec k_2}=2F_{0\vec k_1}$ has to obey the wave-vector condition $\vec k_2+2\vec k_1=0$. On the other hand, a second-harmonic polariton ($\vec k_2;\ F_{0\vec k_2}=2F_{0\vec k_1}$) arising during a fusion process which takes place only on the level α, should satisfy the condition $\vec k_2-2\vec k_1=0$. Remembering the transformation (3.27), we know how photons of wave vector $\vec k_2=\pm 2\vec k_1$ contribute to a second-harmonic polariton.

C. Generation of Third-Harmonic Polariton

As an example of a fourth-order effect we evaluate the transition amplitude for the generation of a third-harmonic polariton ($\vec k_2;\ E_{0\vec k_2}=3F_{0\vec k_1}$) on the level β during the annihilation of three polaritons ($\vec k_1;\ F_{0\vec k_1}$) on the level α. In first-order perturbation theory this transition is caused by the interaction $H^{(4)}_{\mathrm{nl}}$, and we obtain

$$\langle (N_{\vec k_1}-3)^\alpha;\ 1^\beta_{\vec k_2}|H_C^{(4)}|0;\ N^\alpha_{\vec k_1}\rangle = \delta_{\vec k_2,-3\vec k_1}[N^\alpha_{\vec k_1}(N_{\vec k_1}-1)^\alpha(N_{\vec k_1}-1)^\alpha]^{1/2}$$
$$\times\frac{\pi e^2 v^{vv*}_{\vec k_1 3\vec k_1}v^{vv}_{3\vec k_1,-3\vec k_1}(D^{cv*}_{0\vec k_1})^3}{8\,\mathrm{vol}\,|\vec k_1|^2|D^{cv}_{0\vec k_1}|^3}\left(\frac{W_{0\vec k_1}-(\tilde\Omega^2_{0\vec k_1}-\omega^2_{\vec k_1})}{2W_{0\vec k_1}}\right)^{1/2}\left(\frac{W_{03\vec k_1}+(\tilde\Omega^2_{03\vec k_1}-\tilde\omega^2_{3\vec k_1})}{2W_{03\vec k_1}}\right)^{1/2}$$
$$\times\left[\left(\frac{\Omega_{0\vec k_1}F_{0\vec k_1}\Omega_{03\vec k_1}E_{03\vec k_1}}{\tilde\Omega^2_{0\vec k_1}\tilde\Omega^2_{03\vec k_1}}\right)^{1/2}+\left(\frac{\tilde\Omega^2_{0\vec k_1}\tilde\Omega^2_{03\vec k_1}}{\Omega_{0\vec k_1}F_{0\vec k_1}\Omega_{03\vec k_1}E_{03\vec k_1}}\right)^{1/2}\right]\left(\frac{\tilde\Omega^2_{0k_1}}{\Omega_{0k_1}F_{0k_1}}-\frac{\Omega_{0k_1}F_{0k_1}}{\tilde\Omega^2_{0\vec k_1}}\right) \qquad (4.12)$$

and

$$\langle (N_{\vec{k}_1}-3)^{\alpha};\; 1^{\beta}_{\vec{k}_2} | H^{(4)}_{\mathrm{erII}} | 0;\, N^{\alpha}_{\vec{k}_1}\rangle = \delta_{\vec{k}_2,-3\vec{k}_1}\left[N^{\alpha}_{\vec{k}_1}(N_{\vec{k}_1}-1)^{\alpha}(N_{\vec{k}_1}-2)^{\alpha}\right]^{1/2}$$
$$\times\Bigg\{ f_{\mathrm{k}_1\mathrm{k}_1}\,\frac{v^{v\;v}_{3\vec{k}_1\vec{k}_1} D^{cv*}_{0\vec{k}_1}}{2|D^{cv}_{0\vec{k}_1}|}\;\frac{W_{0\vec{k}_1}+(\tilde{\Omega}^2_{0\vec{k}_1}-\tilde{\omega}^2_{\vec{k}_1})}{2W_{0\vec{k}_1}}\left(\frac{W_{0\vec{k}_1}-(\tilde{\Omega}^2_{0\vec{k}_1}-\tilde{\omega}^2_{\vec{k}_1})}{2W_{0\vec{k}_1}}\right)^{1/2}\left(\frac{W_{0\,3\vec{k}_1}+(\tilde{\Omega}^2_{0\,3\vec{k}_1}-\tilde{\omega}^2_{3\vec{k}_1})}{2W_{0\,3\vec{k}_1}}\right)^{1/2}\times\frac{\omega_{\vec{k}_1}}{F_{0\vec{k}_1}}$$
$$\times\left[\left(\frac{\Omega_{0\vec{k}_1}F_{0\vec{k}_1}\Omega_{0\,3\vec{k}_1}E_{0\,3\vec{k}_1}}{\tilde{\Omega}^2_{0\vec{k}_1}\tilde{\Omega}^2_{0\,3\vec{k}_1}}\right)^{1/2}+\left(\frac{\tilde{\Omega}^2_{0\vec{k}_1}\tilde{\Omega}^2_{0\,3\vec{k}_1}}{\Omega_{0\vec{k}_1}F_{0\vec{k}_1}\Omega_{0\,3\vec{k}_1}E_{0\,3\vec{k}_1}}\right)^{1/2}\right]+f_{3\vec{k}_1\vec{k}_1}\,\frac{v^{v\;v}_{-\vec{k}_1\vec{k}_1}D^{cv}_{0\,3\vec{k}_1}(D^{cv*}_{0\vec{k}_1})^2}{2|D^{cv}_{0\,3\vec{k}_1}|\,|D^{cv}_{0\vec{k}_1}|^2}\;\frac{W_{0\vec{k}_1}-(\tilde{\Omega}^2_{0\vec{k}_1}-\tilde{\omega}^2_{\vec{k}_1})}{2W_{0\vec{k}_1}}$$
$$\times\left(\frac{W_{0\vec{k}_1}+(\tilde{\Omega}^2_{0\vec{k}_1}-\tilde{\omega}^2_{\vec{k}_1})}{2W_{0\vec{k}_1}}\right)^{1/2}\times\left(\frac{W_{0\,3\vec{k}_1}-(\tilde{\Omega}^2_{0\,3\vec{k}_1}-\tilde{\omega}^2_{3\vec{k}_1})}{2W_{0\,3\vec{k}_1}}\right)^{1/2}\left(\frac{\omega_{\vec{k}_1}\omega_{3\vec{k}_1}}{F_{0\vec{k}_1}E_{0\,3\vec{k}_1}}\right)^{1/2}\left(\frac{\tilde{\Omega}^2_{0\vec{k}_1}}{\Omega_{0\vec{k}_1}F_{0\vec{k}_1}}-\frac{\Omega_{0\vec{k}_1}F_{0\vec{k}_1}}{\tilde{\Omega}^2_{0\vec{k}_1}}\right)\Bigg\}\,. \qquad (4.13)$$

In this case, the wave-vector condition $\vec{k}_2+3\vec{k}_1=0$ is necessary in order to allow the transition mentioned. A third-harmonic generation of a polariton ($\vec{k}_2;\ F_{0\vec{k}_2}=3F_{0\vec{k}_1}$), which includes only the level α, can happen if $\vec{k}_2-3\vec{k}_1=0$.

The examples reported show that nonlinear processes imply simultaneously a frequency as well as a wave-vector condition; hence, only certain wave vectors for special polariton transitions come into consideration. With respect to our model, these $\vec{q}$ values essentially depend on the curvature of the polariton levels, and of their separation in $\vec{q}$ space.

V. CONCLUSION

Starting with an electron-hole-pair model of interacting radiation and solid matter, we have described quantum mechanically collective effects in insulating crystals. Our considerations are restricted to excitons, plasmons, and polaritons. Plasmon-photon coupling in the crystal arises if we do not neglect the photon wave vector in comparison to the electron wave vector. A simplified, but exactly solvable model, which leads to two optical refractive indices, is used to derive the polariton-dispersion curves. Polaritons, in this paper, are described as coupled oscillations of photons and electron-hole pairs. A perturbation treatment of nonlinear optical transitions including Coulomb forces may show the advantage of polariton states in nonlinear optics.

The generalization of our simplified polariton model, and the coupling of the system to phonons shall be subject of further research. To take into consideration the interaction with lattice vibrations is recommended by the existence of photon-plasmon coupling. Strong electron-phonon interactions are important for near-resonance effects in solids. An increase of scattering efficiencies for TO and LO phonons was observed in resonant Raman scattering experiments on semiconductors and insulators with exciting laser energies near that of an absorption edge.[20-22] It was pointed out in recent publications[23,24] that polaritons should be used as the true electromagnetic modes of the crystal in the theory of Raman scattering. Similarly, the plasmon-photon coupling was observed in semiconductors, and intensively studied on a polariton basis.[25,26]

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$$\sum_j e^{i(\vec{k}'+\vec{q}-\vec{k})\vec{R}_j} \neq 0 \quad \text{if} \quad \begin{cases} \vec{k}'+\vec{q}-\vec{k}=0, & \text{normal processes} \\ \vec{k}'+\vec{q}-\vec{K}_j, & \text{umklapp processes} \end{cases}$$

where $\vec{R}_j$ and $\vec{K}_j$ are lattice and reduced reciprocal-lattice vectors, respectively. We neglect umklapp processes, and write

$$\sum_j e^{i(\vec{k}'+\vec{q}-\vec{k})\vec{R}_j} = N\delta_{\vec{k}'+\vec{q},\vec{k}} \;.$$

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Electron Mobility in II-VI Semiconductors

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The electron drift mobility in CdS, CdSe, CdTe, ZnS, ZnSe, and ZnTe is calculated by an iterative solution of the Boltzmann equation for lattice scattering. Piezoelectric, deformation-potential acoustic-mode, and polar-mode scattering are included. The acoustic deformation potential appropriate to acoustic-mode scattering appears to be much higher than previously expected.

I. INTRODUCTION

The electron mobility in II-VI compound semiconductors can be understood by a consideration of the scattering of conduction electrons by fundamental lattice vibrations.[1] Although impurity scattering[2,3] is also well known, this mechanism does not contribute to the lattice mobility. Its effect in commonly pure materials is negligible at temperatures above ~100 °K. The theory of electron scattering by lattice vibrations[4,5] is exceptionally accurate for isotropic direct-gap materials because of our knowledge of the conduction-band structure.[6] By an iterative solution of the Boltzmann equation,[7,8] the electron mobility follows exactly from the assumed model described below. The wurtzitelike and zincblende-like crystals CdS, CdSe, CdTe, ZnS, ZnSe, and ZnTe, being wide-gap semiconductors, are especially well suited to calculation and are the only materials discussed here. The direct-gap III-V semiconductors have been discussed previously.[8]

There are five main conclusions evident from the present work. First, the three scattering mechanisms discussed by several authors[1,9,10] are sufficient to predict the lattice mobility, i.e., polar-mode scattering,[5] acoustic mode via deformation-potential coupling,[11] and acoustic mode via piezoelectric coupling.[12] Second, Matthiessen's rule[13] (reciprocal mobility is the sum of reciprocal component mobili-