Properties of anti-Stokes scattering of coherent polariton waves in direct-gap semiconductors

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Canonical transformation of a model Hamiltonian is used to introduce the "phonoriton" elementary excitation, which arises from the restructuring of photon, exciton, and phonon excitations of a semiconductor in the presence of a polariton wave. A system of macroscopic phonoriton equations that permits analysis of the nonstationary problem is derived and analyzed. The conditions under which phonoriton splitting can be observed are indicated. It is shown that greatest experimental interest attaches to spectrum restructuring by interaction between the exciton component of the initial wave and longitudinal optical phonons. The question of supplementary boundary conditions for acoustic phonoriton waves is considered.

In Ref. 1 we considered the restructuring of polariton excitations and of longitudinal acoustic phonon of a semiconductor in the presence of an intense polariton wave **k**, and introduced an elementary excitation, the phonoriton, consisting of polariton and acoustic waves.

The primary purpose of the present paper is to derive and analyze macroscopic equations that describe all three quantized fields (excitons, photons, and longitudinal phonons) that interact with the initial coherent polariton wave. This system of macroscopic equations, derived for arbitrary anisotropic crystals, will allow us to consider, in the general case, the nonstationary problem in both the anti-Stokes and the Stokes approximations in the exciton-phonon interaction. We shall consider the need for separating the exciton and photon components of the polariton excitations in the analysis of questions connected with the phonoriton restructuring of spectra. The phonoriton elementary excitations have in this case three (photon, exciton, and phonon) components. These elementary excitations will be introduced for both longitudinal acoustic (LA) and longitudinal optic (LO) phonons by a canonical transformation of a model Hamiltonian in the approximation in which a coherent polariton **k** wave is given. After analyzing the feasibility of experimentally observing the phonoriton splitting of the spectra, we shall show that the most interesting case arises when the spectrum is restructured by interaction between the exciton component of the initial **k** wave and LO phonons. We conclude by considering the question of the supplementary boundary conditions for acoustic phonoriton waves.

We consider first the conditions under which an initial polariton wave of momentum **k** of finite amplitude can be associated with the concept of macroscopic filling of a **k** mode. The rigorous notion of macroscopic occupation of a polariton mode can be introduced, for a physically plausible case of passage of an electromagnetic wave of frequency $\omega_{\mathbf{k}}$ set by an external source and close to the frequency of the exciton transition, only if the exciton absorption coefficient is small at the specified frequency, when the wave vector of the initial polariton wave satisfies the condition $\mathrm{Re}\,\mathbf{k} \gg \mathrm{Im}\,\mathbf{k}$. On the other hand, the concept of macroscopic occupation of a polariton **k** mode is most important if the corresponding exciton component is large, i.e., the initial polariton wave is exciton-like. The reason for the last condition is that it is just through the exciton component that the polariton wave interacts with the various elementary excitations of the medium. These two conditions, that the initial polariton **k** wave have a small absorption coefficient and that this wave be exciton-like, can be satisfied simultaneously in certain cases, viz., when the polariton-wave frequency $\omega_{\mathbf{k}}$ is in the range

$$\Omega_c \gtrsim \omega_t - \omega_{\mathbf{k}} \gg (^1/_4\omega_{lt}^2 + \gamma\omega_{lt})^{1/2} - {}^1/_2\omega_{lt}, \tag{1}$$

where $\hbar\omega_t$ is the exciton-level energy reckoned from the valence band, $\hbar\omega_{lt}$ the longitudinal-transverse polariton splitting, γ the characteristic reciprocal lifetime of the **k** exciton, and the energy $\hbar\Omega_c$ characterizes the strength of the exciton-photon interaction. In fact, it is known[2–4] that the dispersion polariton equation takes, in the approximation where the exciton-photon interaction is resonant, the form

$$\begin{aligned} &(\omega - \omega_{\mathbf{k}}^{phot})(\omega - \omega_{\mathbf{k}}^{ex} + i\gamma) = {}^1/_4\Omega_c^2; \\ &\omega_{\mathbf{k}}^{ex} = \omega_t + \frac{\hbar k^2}{2M^{ex}}; \quad \omega_{\mathbf{k}}^{phot} = \frac{c}{\varepsilon_g^{1/2}}k. \end{aligned} \tag{2}$$

The variables $\omega_{\mathbf{k}}^{phot}$ and $\omega_{\mathbf{k}}^{ex}$ stand here for the respective frequencies of the photon and exciton terms in the absence of exciton-photon interaction, M^{ex} is the translational mass of the exciton, and ε_g is the crystal background dielectric constant of the exciton transition. It can be easily seen[5] from an analysis of the dispersion equation (2) that in the considered case of polariton-like propagation of the initial electromagnetic wave ($\Omega_c > \gamma$) the characteristic scale of variation of the dielectric-constant component connected with the exciton resonance is determined by the parameter ω_{lt}. The spectral width of the absorption line corresponding to the exciton resonance is then determined by the values of ω_{lt} and γ. For exciton transitions with clearly pronounced polariton properties ($\omega_{lt} \gtrsim \gamma$) the conditions $\mathrm{Re}\,\mathbf{k} \gg \mathrm{Im}\,\mathbf{k}$, defined by the right-hand side of inequality (1), will certainly be met at $\omega_t - \omega_{\mathbf{k}} \gg \omega_{lt}$, so that the lower bound of the frequency region (1) can be taken in this case to be the value of the

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longitudinal-transverse splitting. The intensity of the exciton component of a polariton wave having the frequency $\omega_{\mathbf{k}}$ of the lower polariton branch is characterized, in turn, by a weighting factor

$$\varphi^{ex}(\mathbf{k})=\frac{1}{2}\left[1-\frac{\omega_{\mathbf{k}}^{ex}-\omega_{\mathbf{k}}^{phot}}{[(\omega_{\mathbf{k}}^{ex}-\omega_{\mathbf{k}}^{phot})^2+\Omega_c^2]^{1/2}}\right]$$

$$\approx\left[1+4\frac{(\omega_t-\omega_{\mathbf{k}})^2}{\Omega_c^2}\right]^{-1}, \tag{3}$$

i.e., the characteristic frequency scale of the exciton-component is determined by the strength Ω_c of the exciton-photon interaction; this strength is connected with the oscillator strength $4\pi\beta$ of the exciton transition and with the parameter ω_{lt} by the familiar relations[3,5]

$$\Omega_c=\left(\frac{4\pi\beta}{\varepsilon_g}\right)^{1/2}\omega_t=(2\omega_{lt}\omega_t)^{1/2}. \tag{4}$$

It can be seen from the last relation that the inequality $\Omega_c\gg\omega_{lt}\gtrsim\gamma$ holds for typical direct-gap semiconductors, and it is this which justifies the introduction of the frequency range (1). For the semiconductor CdS, for example, the corresponding values are $\Omega_c\approx 100$ meV and $\omega_{lt}\approx 2$ meV (Ref. 6). The condition (1), which we regard hereafter as satisfied, means in fact that the frequency $\omega_{\mathbf{k}}$ of the initial polariton **k** wave should belong to the low-frequency "transparency region" near the exciton-absorption line. The wave vector **k** of the initial polariton wave is then connected with its frequency $\omega_{\mathbf{k}}$ by the polariton dispersion equation (2).

It appears that the following can be stated in general. In the sense indicated earlier,[1] the initial polariton **k** wave can be related to a nonequilibrium condensate of **k** excitons having a restructured polariton spectrum that constitutes elementary Bose-like excitations of the crystal. Actually, an electromagnetic wave from an external source can be used to produce a nonequilibrium Bose condensate of elementary excitations of the medium (excitons), generally speaking, in an arbitrary **k** mode determined by the frequency interval (1). The corresponding density N_0 of the **k**-mode Bose-condensate excitons is given by

$$N_0=\varphi^{ex}(\mathbf{k})\frac{I}{\hbar\omega_{\mathbf{k}}V_{\mathbf{k}}}, \tag{5}$$

where $V_{\mathbf{k}}$ is the group velocity of the initial polariton wave and I is flux of the external electromagnetic wave that penetrates into the crystal through its boundary.

The Hamiltonian that describes the system of interacting excitons, photons, and longitudinal phonons in a direct-gap semiconductor can be represented in the form

$$H=\sum_{\mathbf{p}}\hbar\left\{\omega_{\mathbf{p}}^{ex}b_{\mathbf{p}}^{+}b_{\mathbf{p}}+\omega_{\mathbf{p}}^{phot}a_{\mathbf{p}}^{+}a_{\mathbf{p}}+\Omega_{\mathbf{p}-\mathbf{k}}c_{\mathbf{p}-\mathbf{k}}^{+}c_{\mathbf{p}-\mathbf{k}}+i\frac{\Omega_c}{2}(a_{\mathbf{p}}^{+}b_{\mathbf{p}}-b_{\mathbf{p}}^{+}a_{\mathbf{p}})+\sum_{\mathbf{l}}iM(\mathbf{p}-\mathbf{l})[b_{\mathbf{p}}^{+}b_{\mathbf{l}}(c_{\mathbf{p}-\mathbf{l}}-c_{-\mathbf{p}+\mathbf{l}}^{+})]\right\}. \tag{6}$$

Here $(\omega_{\mathbf{p}}^{phot},a_{\mathbf{p}})$, $(\omega_{\mathbf{p}}^{ex},b_{\mathbf{p}})$ and $(\Omega_{\mathbf{p}-\mathbf{k}},c_{\mathbf{p}-\mathbf{k}})$ are respectively the frequency and annihilation operators of the photon, exciton, and longitudinal-phonon excitations of the semiconductor. As already indicated, Ω_c is a measure of the exciton-phonon interaction strength, and the matrix element $M(\mathbf{p}-\mathbf{l})$ determines the binding energy of excitons with longitudinal phonons. This Hamiltonian was written assuming that the exciton-phonon interaction is resonant, and can be substantially simplified when account is taken of the presence of a macroscopically filled polariton **k** mode. In the present case of a coherent polariton **k** wave of given amplitude, it is valid to make in the Hamiltonian (6) the substitution

$$\sum_{\mathbf{l}}iM(\mathbf{p}-\mathbf{l})[b_{\mathbf{p}}^{+}b_{\mathbf{l}}(c_{\mathbf{p}-\mathbf{l}}-c_{-\mathbf{p}+\mathbf{l}}^{+})]\to iM(\mathbf{p}-\mathbf{k})$$

$$\times[\alpha_{\mathbf{k}}e^{-i\omega_{\mathbf{k}}t}b_{\mathbf{p}}^{+}(c_{\mathbf{p}-\mathbf{k}}-c_{-\mathbf{p}+\mathbf{k}}^{+})-\alpha_{\mathbf{k}}^{*}e^{i\omega_{\mathbf{k}}t}b_{\mathbf{p}}(c_{\mathbf{p}-\mathbf{k}}^{+}-c_{-\mathbf{p}+\mathbf{k}})], \tag{7}$$

where the c-numbers $\alpha_{\mathbf{k}}(t)$ are determined by the following relation with the amplitude $P_0(t)$ of the exciton polarization that is connected with the initial polariton wave:

$$\alpha_{\mathbf{k}}(t)=\int\langle b_{\mathbf{k}}\rangle\frac{V}{(2\pi)^3}d^3p=\left[\frac{\varepsilon_g V}{2\hbar\omega_t\beta}\right]^{1/2}P_0(t)$$

$$=\left[\frac{\varepsilon_g V}{2\hbar\omega_t\beta}\right]^{1/2}P_0e^{-i\omega_{\mathbf{k}}t}. \tag{8}$$

The substitution proposed is tantamount to retaining in the Hamiltonian (6) only those terms of the part that describes the exciton-phonon interaction which characterize the interaction of the phonon system with the excitons of the macroscopically filled **k** mode. This substitution is permissible, roughly speaking, because one macroscopically occupied exciton **k** mode influences the dynamic properties and the kinetic behavior of the crystal excitons, photons, and longitudinal phonons to the same degree, in the sense of proportionality to the crystal volume V, as all other exciton modes. The influence of the latter is determined primarily by population of the modes $\mathbf{p}\neq\mathbf{k}$ by excitons, which we shall call scattered; this influence can be quite weak. Eliminating the explicit time dependence from the resultant Hamiltonian by using the canonical transformation

$$S_1=\exp\left[i\omega_{\mathbf{k}}t\sum_{\mathbf{p}}(b_{\mathbf{p}}^{+}b_{\mathbf{p}}+a_{\mathbf{p}}^{+}a_{\mathbf{p}})\right],$$

which shifts the photon and exciton frequencies by $\omega_{\mathbf{k}}$, we obtain the following model phonoriton Hamiltonian:

$$H_{\mathbf{k}}=\sum_{\mathbf{p}}\hbar\left\{(\omega_{\mathbf{p}}^{ex}-\omega_{\mathbf{k}})b_{\mathbf{p}}^{+}b_{\mathbf{p}}+\Omega_{\mathbf{p}-\mathbf{k}}c_{\mathbf{p}-\mathbf{k}}^{+}c_{\mathbf{p}-\mathbf{k}}+(\omega_{\mathbf{p}}^{phot}-\omega_{\mathbf{k}})a_{\mathbf{p}}^{+}a_{\mathbf{p}}+i\frac{\Omega_c}{2}(a_{\mathbf{p}}^{+}b_{\mathbf{p}}-b_{\mathbf{p}}^{+}a_{\mathbf{p}})\right\}+H_S+H_{AS}, \tag{9}$$

$$H_{AS}=\hbar\sum_{\mathbf{p}}iM(\mathbf{p}-\mathbf{k})\alpha_{\mathbf{k}}(b_{\mathbf{p}}^{+}c_{\mathbf{p}-\mathbf{k}}-b_{\mathbf{p}}c_{\mathbf{p}-\mathbf{k}}^{+}),$$

$$H_S=\hbar\sum_{\mathbf{p}}iM(\mathbf{p}-\mathbf{k})\alpha_{\mathbf{k}}(b_{\mathbf{p}}c_{-\mathbf{p}+\mathbf{k}}-b_{\mathbf{p}}^{+}c_{-\mathbf{p}+\mathbf{k}}^{+}). \tag{10}$$

The terms H_S and H_{AS} of (9) describe respectively the Stokes and anti-Stokes interactions of the excitons between the macroscopically occupied **k** mode and the longitudinal phonons. The variables $\alpha_{\mathbf{k}}=\alpha_{\mathbf{k}}(t=0)$ are assumed to be real and can always be made so by suitable choice of the

origin of time. The model phonoriton Hamiltonian (9) is a Hermitian quadratic form in the operators $a_{\mathbf{p}}, b_{\mathbf{p}}, c_{\mathbf{p}-\mathbf{k}}$ and can be diagonalized. The diagonalization introduces phonorition annihilation operators $\tilde{a}_{\mathbf{p}}, \tilde{b}_{\mathbf{p}}, \tilde{c}_{\mathbf{p}-\mathbf{k}}$, that are connected in the general case with the old operators by the relation

$$\begin{vmatrix} \tilde{b}_{\mathbf{p}} \\ \tilde{c}_{\mathbf{p}-\mathbf{k}} \\ \tilde{a}_{\mathbf{p}} \\ \tilde{b}^{+}_{2\mathbf{k}-\mathbf{p}} \\ \tilde{c}^{+}_{-\mathbf{p}+\mathbf{k}} \\ \tilde{a}^{+}_{2\mathbf{k}-\mathbf{p}} \end{vmatrix} = \left| A_{ij} \right| \begin{vmatrix} b_{\mathbf{p}} \\ c_{\mathbf{p}-\mathbf{k}} \\ a_{\mathbf{p}} \\ b^{+}_{2\mathbf{k}-\mathbf{p}} \\ c^{+}_{-\mathbf{p}+\mathbf{k}} \\ a^{+}_{2\mathbf{k}-\mathbf{p}} \end{vmatrix}, \tag{11}$$

where the normalization of the A_{ij} matrix elements is subject to the natural requirements that the commutation rules be preserved for the new phonoriton operators. With allowance for this requirement, the A_{ij} matrix elements and the general dispersion equation are obtained by considering six conditions of like form, of which the first is

$$\frac{1}{\hbar}[\tilde{a}_{\mathbf{p}}, H_{\mathbf{k}}] = E\tilde{a}_{\mathbf{p}},$$

where the variable E determines the phonoriton frequency. The dispersion equation in the variable E, which corresponds to the general form of the Hamiltonian (9), is

$$[(\nu_1\nu_3 - {}^1/_4\Omega_c^2)\nu_2\nu_5 + Q^2(\nu_2-\nu_5)\nu_3](\nu_4\nu_6 - {}^1/_4\Omega_c^2) - Q^2\nu_6(\nu_2-\nu_5)(\nu_1\nu_3 - {}^1/_4\Omega_c^2) = 0, \tag{12}$$

where the variables ν_i are defined as

$$\nu_1 = \omega_{\mathbf{p}}^{ex} - \omega_{\mathbf{k}} - E, \quad \nu_2 = \Omega_{\mathbf{p}-\mathbf{k}} - E, \quad \nu_3 = \omega_{\mathbf{p}}^{phot} - \omega_{\mathbf{k}} - E,$$

$$\nu_4 = -\omega^{ex}_{2\mathbf{k}-\mathbf{p}} + \omega_{\mathbf{k}} - E, \quad \nu_5 = -\Omega_{\mathbf{p}-\mathbf{k}} - E, \quad \nu_6 = -\omega^{phot}_{2\mathbf{k}-\mathbf{p}} + \omega_{\mathbf{k}} - E,$$

and the relation between Q and the exciton density in the macroscopically occupied $\mathbf{k}$ mode is

$$Q = \alpha_{\mathbf{k}} M(\mathbf{p}-\mathbf{k}) = (N_0 V)^{1/2} M(\mathbf{p}-\mathbf{k}). \tag{13}$$

Of greatest interest to us, however, is the anti-Stokes-interaction frequency range corresponding to the phonoritonic spectrum restructuring near the frequency $E = \Omega_{\mathbf{p}-\mathbf{k}}$. In this case we can neglect in the Hamiltonian (9) the term H_S that describes the exciton-phonon Stokes interaction. This simplifies greatly the phonoritonic transformation (11), with the new phonoriton operators $\tilde{a}_{\mathbf{p}}, \tilde{b}_{\mathbf{p}}$ and $\tilde{c}_{\mathbf{p}-\mathbf{k}}$ now determined by linear combinations only of the annihilation operators $a_{\mathbf{p}}, b_{\mathbf{p}}$ and $c_{\mathbf{p}-\mathbf{k}}$. Moreover, in this approximation, which is resonant in the anti-Stokes interaction, we can diagonalize using the explicit canonical transformation

$$S_2 = \exp\left\{\sum_{\mathbf{p}} [\delta_{\mathbf{p}}(b_{\mathbf{p}}^{+}a_{\mathbf{p}} - a_{\mathbf{p}}^{+}b_{\mathbf{p}}) + \varphi_{\mathbf{p}}(b_{\mathbf{p}}^{+}c_{\mathbf{p}-\mathbf{k}} - c^{+}_{\mathbf{p}-\mathbf{k}}b_{\mathbf{p}}) + \gamma_{\mathbf{p}}(a_{\mathbf{p}}^{+}c_{\mathbf{p}-\mathbf{k}} - c^{+}_{\mathbf{p}-\mathbf{k}}a_{\mathbf{p}})]\right\} \exp\left[i\frac{\pi}{2}\sum_{\mathbf{p}} (c^{+}_{\mathbf{p}-\mathbf{k}}c_{\mathbf{p}-\mathbf{k}} - a_{\mathbf{p}}^{+}a_{\mathbf{p}}) \right];$$

$$\tilde{a}_{\mathbf{p}} = S_2 a_{\mathbf{p}} S_2^{+}, \quad \tilde{b}_{\mathbf{p}} = S_2 b_{\mathbf{p}} S_2^{+}, \quad \tilde{c}_{\mathbf{p}-\mathbf{k}} = S_2 c_{\mathbf{p}-\mathbf{k}} S_2^{+}, \tag{14}$$

which is determined by three sets of real parameters $\delta_{\mathbf{p}}, \varphi_{\mathbf{p}}$ and $\gamma_{\mathbf{p}}$. The matrix A_{ij} that relates the set of annihilation operators $b_{\mathbf{p}}, c_{\mathbf{p}-\mathbf{k}}, a_{\mathbf{p}}$ to the new phonoriton annihilation operators $\tilde{b}_{\mathbf{p}}, \tilde{c}_{\mathbf{p}-\mathbf{k}}, \tilde{a}_{\mathbf{p}}$ takes in this case the abbreviated form

$$\begin{vmatrix} 1-2\frac{(\varphi_{\mathbf{p}}^2+\delta_{\mathbf{p}}^2)}{\Delta_{\mathbf{p}}^2}\sin^2\frac{\Delta_{\mathbf{p}}}{2}, & -\frac{\varphi_{\mathbf{p}}}{\Delta_{\mathbf{p}}}\sin\Delta_{\mathbf{p}} + 2\frac{\delta_{\mathbf{p}}\gamma_{\mathbf{p}}}{\Delta_{\mathbf{p}}^2}\sin^2\frac{\Delta_{\mathbf{p}}}{2}, & -\frac{\delta_{\mathbf{p}}}{\Delta_{\mathbf{p}}}\sin\Delta_{\mathbf{p}} - 2\frac{\varphi_{\mathbf{p}}\gamma_{\mathbf{p}}}{\Delta_{\mathbf{p}}^2}\sin^2\frac{\Delta_{\mathbf{p}}}{2} \\ -i\frac{\varphi_{\mathbf{p}}}{\Delta_{\mathbf{p}}}\sin\Delta_{\mathbf{p}} - 2i\frac{\delta_{\mathbf{p}}\gamma_{\mathbf{p}}}{\Delta_{\mathbf{p}}^2}\sin^2\frac{\Delta_{\mathbf{p}}}{2}, & -i+2i\frac{(\gamma_{\mathbf{p}}^2+\varphi_{\mathbf{p}}^2)}{\Delta_{\mathbf{p}}^2}\sin^2\frac{\Delta_{\mathbf{p}}}{2}, & -i\frac{\gamma_{\mathbf{p}}}{\Delta_{\mathbf{p}}}\sin\Delta_{\mathbf{p}} + 2i\frac{\varphi_{\mathbf{p}}\delta_{\mathbf{p}}}{\Delta_{\mathbf{p}}^2}\sin^2\frac{\Delta_{\mathbf{p}}}{2}, \\ \frac{\delta_{\mathbf{p}}}{\Delta_{\mathbf{p}}}\sin\Delta_{\mathbf{p}} - 2i\frac{\varphi_{\mathbf{p}}\gamma_{\mathbf{p}}}{\Delta_{\mathbf{p}}^2}\sin^2\frac{\Delta_{\mathbf{p}}}{2}, & -i\frac{\gamma_{\mathbf{p}}}{\Delta_{\mathbf{p}}}\sin\Delta_{\mathbf{p}} - 2i\frac{\varphi_{\mathbf{p}}\delta_{\mathbf{p}}}{\Delta_{\mathbf{p}}^2}\sin^2\frac{\Delta_{\mathbf{p}}}{2}, & i-2i\frac{(\delta_{\mathbf{p}}^2+\gamma_{\mathbf{p}}^2)}{\Delta_{\mathbf{p}}^2}\sin^2\frac{\Delta_{\mathbf{p}}}{2} \end{vmatrix}$$

$$\Delta_{\mathbf{p}}^2 = \gamma_{\mathbf{p}}^2 + \varphi_{\mathbf{p}}^2 + \delta_{\mathbf{p}}^2. \tag{15}$$

The corresponding phonoriton dispersion equation, which can also be obtained in this approximation from the more general dispersion equation (12), is cubic in the phonoriton frequency E:

$$\left(\nu_1\nu_3 - \frac{\Omega_c^2}{4}\right)\nu_2 - Q^2\nu_3 = (\omega_{\mathbf{p}}^{ex} - \omega_{\mathbf{k}} - E)(\omega_{\mathbf{p}}^{phot} - \omega_{\mathbf{k}} - E)$$

$$\times(\Omega_{\mathbf{p}-\mathbf{k}} - E) - D^2(\Omega_{\mathbf{p}-\mathbf{k}} - E) - Q^2(\omega_{\mathbf{p}}^{phot} - \omega_{\mathbf{k}} - E) = 0, \tag{16}$$

whose real roots determine three phonoriton dispersion branches. We note that within the framework of the present model Hamiltonian (9) the diagonalization procedure proposed permits an exact account to be taken also of the exciton-photon and exciton-phonon interaction. The new elementary excitations can be correctly introduced, generally speaking, just in the approximation that is resonant in the anti Stokes approximation. This statement is valid because the roots of (16) are real, whereas the analysis of the general case entails consideration of the dispersion equation (12), which can have complex roots. Complex roots are obtained, for example, in the analysis of the approximation $E \approx -\Omega_{\mathbf{p}-\mathbf{k}}$ that is resonant with respect to the Stokes interaction. This result has a clear physical meaning and is due to the instability of the Stokes polariton $\mathbf{p}$ wave and of the corresponding scattering $\mathbf{p} - \mathbf{k}$ wave when their amplitude are increased by decay of the initial polariton $\mathbf{k}$ wave.

It follows from these results that phonoriton elementary excitations have exciton, phonon, and photon components whose respective contributions x_i, y_i, and z_i to the intensities of the phonoriton excitations are given by

$$x_i = \frac{1}{R_i}\frac{\Omega_c^2}{4}(\Omega_{\mathbf{p}-\mathbf{k}} - E_i)^2, \quad y_i = \frac{1}{R_i}\frac{\Omega_c^2}{4}Q^2,$$

$$z_i = \frac{1}{R_i}[Q^2 - (\omega_{\mathbf{p}}^{ex} - \omega_{\mathbf{k}} - E_i)(\Omega_{\mathbf{p}-\mathbf{k}} - E_i)]^2, \tag{17}$$

$$R_i = \frac{\Omega_c^2}{4}(\Omega_{\mathbf{p}-\mathbf{k}} - E_i)^2 + \frac{\Omega_c^2}{4}Q^2 + [Q^2 - (\omega_{\mathbf{p}}^{ex} - \omega_{\mathbf{k}} - E_i)(\Omega_{\mathbf{p}-\mathbf{k}} - E_i)]^2,$$

where the subscript $i = 1,2,3$ labels the dispersion branches of the phonoriton excitations. It appears that in the most frequently encountered case $\Omega_c \gg Q$, where the exciton-photon interaction is stronger than the exciton-phonon interaction, the phonoriton dispersion equation (16) can be made even simpler:

$$(E-\omega_{\mathbf{p}})(E-\omega_{\mathbf{k}}-\Omega_{\mathbf{p}-\mathbf{k}})=\varphi^{ex}(\mathbf{p})Q^2. \tag{18}$$

This equation, roughly speaking, describes the restructuring of the polariton and phonon spectra and conforms to the analysis of Ref. 1.

Depending on whether the anti-Stokes interaction investigated is between optic or acoustic longitudinal phonons, we can introduce respectively optic or acoustic phonoriton excitations. In the former case the squared matrix element of the exciton-phonon Fröhlich interaction for polar direct-gap semiconductors takes the form[6,7]

$$|M(\mathbf{p}-\mathbf{k})|^2=\pi\left(\frac{\varepsilon_0}{\varepsilon_\infty}-1\right)\left(\frac{m_e-m_h}{m_e+m_h}\right)^2\frac{a_B^3}{V}\Omega_0\frac{\hbar(\mathbf{p}-\mathbf{k})^2}{2\mu}, \tag{19}$$

where Ω_0 is the optic-phonon frequency, m_e and m_h are the average masses of the electron and hole constituting the exciton, μ is the exciton reduced mass, a_B is the exciton Bohr radius, and ε_0 and ε_∞ are respectively the static and high-frequency dielectric constants of the crystal for the optical phonon. In the latter case, of acoustic phonoriton excitations, the squared exciton-phonon interaction matrix element is connected with the crystal strain potential D by the relation[4,6,8]

$$|M(\mathbf{p}-\mathbf{k})|^2=\frac{|\mathbf{p}-\mathbf{k}|D^2}{2\hbar V\rho u}, \tag{20}$$

where ρ is the crystal density and u is the average velocity of the longitudinal acoustic waves in the crystal.

Of greatest interest, as mentioned, is the case when the frequency $\omega_{\mathbf{k}}$ of the initial polariton $\mathbf{k}$ wave and the frequency $\omega_{\mathbf{k}}+\Omega_{\mathbf{p}-\mathbf{k}}$ of the corresponding anti-Stokes component are in the frequency region (1) of the lower polariton branch. The oscillator strength of the phonoriton resonance induced at the anti-Stokes frequency $\omega_{\mathbf{k}}+\Omega_{\mathbf{p}-\mathbf{k}}$ is defined in accord with (16) as

$$\left(\frac{\omega_{\mathbf{k}}+\Omega_{\mathbf{p}-\mathbf{k}}-\omega_{\mathbf{p}}^{phot}}{\omega_{\mathbf{k}}+\Omega_{\mathbf{p}-\mathbf{k}}-\omega_{\mathbf{p}}^{ex}}\right)Q^2, \tag{21}$$

and the phonon, exciton, and photon spectra restructuring corresponding to this resonance produces a characteristic gap whose value $\Delta(\mathbf{p}-\mathbf{k})$ depends on the scattering angle and on the intensity of the initial polariton $\mathbf{k}$ wave. In accord with the forms of the matrix elements (19) and (20), the spectral gap $\Delta(\mathbf{p}-\mathbf{k})$ for both acoustic and optical phonoriton excitation is a maximum in the "backscattering" geometry. Figure 1 shows, in particular, the optic phonoriton dispersion curves for the CdS crystal (the abscissa axis corresponds to scattering along the vector $\mathbf{k}$, $\omega_{\mathbf{k}}=2.74$ eV, the flux density of the external electromagnetic wave that eneters the crystal through its boundary is $I=41\ GW/cm^2$).

The complication in this approximation which results from the possibility that the $\mathbf{p}$ exciton may not be scattered into the initial macroscopically occupied $\mathbf{k}$ mode, and also from allowing for the finite lifetime of the longitudinal phonons, introduces the parameters $\gamma(\mathbf{p})$ and $\gamma_{ph}^{A}(\mathbf{p}-\mathbf{k})$, which are the respective reciprocal lifetimes of the scattered $\mathbf{p}$ exciton and of the scattering $\mathbf{p}-\mathbf{k}$ phonon. The conditions for the validity of introducing the phonoriton elementary excitations, and for the possibility of observing experimentally the phonoriton restructuring of the spectra, are

$$\Omega_c \gtrsim \Omega_{\mathbf{p}-\mathbf{k}}, \tag{22a}$$

$$[2\Delta(\mathbf{p}-\mathbf{k})\Omega_{\mathbf{p}-\mathbf{k}}]^{1/2} \gtrsim \gamma(\mathbf{p})+\gamma_{ph}^{A}(\mathbf{p}-\mathbf{k}), \tag{22b}$$

$$\Delta(\mathbf{p}-\mathbf{k}) > \gamma_{pul}(\mathbf{k}), \tag{22c}$$

where $\gamma_{pul}(\mathbf{k})$ is the spectral width of the initial polariton wave or of the corresponding pulse. Conditions (22a)–(22c) have the following physical meaning. The first condition is tantamount to requiring that the frequencies of the initial polariton $\mathbf{k}$ wave and of its anti-Stokes component be located in the range (1). The second condition (22b) is similar to the condition for the observation of polariton restructuring of the spectra and reduces, generally speaking, to the requirement that the frequencies of the phonoriton oscillations exceed the reciprocal lifetimes of the phonoriton excitations. The third condition has a clear physical meaning. Note that a high initial-polariton $\mathbf{k}$-wave intensity was used in Fig. 1 solely to be able to represent in a single scale the entire phonoritron spectrum-restructuring range. In particular, to obtain experimentally an exciton component density $N_0 = 10^{17}$ cm^{-3} in an initial polariton $\mathbf{k}$ wave in the semiconductor CdS, the maximum phonoriton gap can reach $\Delta^{max} \approx 0.3$ meV for optical phonoritons and $\Delta^{max} \approx 0.02$–$0.03$ meV for acoustic ones. For the acoustic phonoriton we have for CdS $\gamma_{ph}^{A}(\mathbf{p}-\mathbf{k}) < \gamma(\mathbf{p}) \lesssim 0.01$ meV at $T = 4.2$ K (Ref. 9) and the basic conditions (22b) can be easily met, but the phonoriton gap itself is also small in this case. For optical phonoritons, in turn, we have for CdS

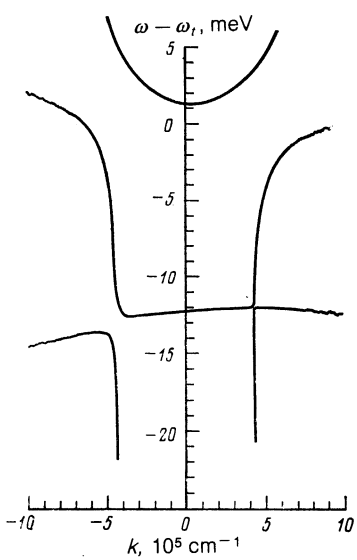

FIG. 1. Phonoriton dispersion curves for CdS: $\omega_t = 2.52$eV, $m_e = 0.2\ m_0$, $m_h = 1.35\ m_0$, $a_B = 27$ Å, $\varepsilon_0 = 8.87$, $\varepsilon_\infty = 5.10$, $\Omega_0 = 38$ meV

$\gamma(\mathbf{p}) < \gamma_{\rm ph}^{A}(\mathbf{p}-\mathbf{k}) \approx 0.1$ meV (Refs. 10–12), condition (22b) is likewise easy to satisfy, but the phononiton gap is large here. Naturally, the frequency spread of the absorption-line connected with the induced phononiton resonance can exceed substantially the corresponding phononiton gap $\Delta(\mathbf{p}-\mathbf{k})$. Note that in this approach all the dynamic phononitonic phenomena are independent of the population of the considered phonon modes.

We have analyzed so far a simplified case with a preferred direction of the dipole moment $\mathbf{d}$ of the exciton transition. There was therefore no need to consider the change of the polarization of the initial polariton $\mathbf{k}$ wave in the course of Stokes or anti-Stokes interaction. We examine now the macroscopic phononiton equations that enable us not only to investigate the changes of the spectra, but also to analyze the evolution in space and time of the phonon, exciton, and photon excitation of a semiconductor in the presence of a specified coherent polariton $\mathbf{k}$ wave.

To make the approach general, we derive the necessary system of macroscopic equations for a sufficiently arbitrary anisotropic direct-gap semiconductor. That is to say, we introduce a system of principal optical axes, in terms of which the oscillator-strength tensor $4\pi\hat{\beta}$ is diagonal with components $4\pi\beta_i$, where $i=1,2,3$. The symmetric tensor $\hat{\mathbf{M}}(\mathbf{p}-\mathbf{k})$ of the exciton-phonon interaction matrix elements can then be represented in the form $\hat{\chi}M(\mathbf{p}-\mathbf{k})$, where the elements χ_{ij} of the tensor $\hat{\chi}$ are determined by the angle between the linear-polarization vector of the initial polariton $\mathbf{k}$ wave and the scattering direction.[6] Analysis of a model Hamiltonian which generalizes the Hamiltonian (9) to include the anisotropic case enables us to obtain for the case in question the following system of phononiton equations in terms of the operators of the vector potential $\mathbf{A}(\mathbf{r},t)$, the polarization vector $\mathbf{P}(\mathbf{r},t)$, and the scalar phonon potential $\Phi(\mathbf{r},t)$:

$$\operatorname{rot}\operatorname{rot}\mathbf{A}+\frac{\varepsilon_g}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{A}=4\pi\frac{\varepsilon_g^{1/2}}{c}\frac{\partial}{\partial t}\mathbf{P}, \tag{23a}$$

$$\left[\frac{\partial^2}{\partial t^2}+2\gamma\frac{\partial}{\partial t}+\omega_t^2-\hbar\omega_t\left(\frac{1}{M_i^{ex}}\right)\Delta\right]P_i=-\frac{\omega_t^2\beta_i}{c\varepsilon_g^{1/2}}\frac{\partial}{\partial t}A_i$$
$$-\omega_t\left\{\begin{array}{c}\frac{1}{\hbar}D\\ \Omega_0\left[\pi\left(\frac{m_e-m_h}{m_e+m_h}\right)^2\left(\frac{\varepsilon_0}{\varepsilon_\infty}-1\right)\frac{a_B^{\,3}\rho}{\mu}\right]^{1/2}\end{array}\right\}$$
$$\times\left[\sum_{l=1}^{3}P_{0l}\cos(\omega_{\mathbf{k}}t-\mathbf{kr})\left(\frac{\beta_i}{\beta_l}\right)^{1/2}\chi_{li}\right]\Delta\Phi, \tag{23b}$$

$$\left[\frac{\partial^2}{\partial t^2}+2\gamma_\Phi^{A}\frac{\partial}{\partial t}+\left\{\begin{array}{c}-u^2\Delta\\ \Omega_0^2\end{array}\right\}\right]\Phi$$
$$=\frac{\varepsilon_g}{\omega_t\rho}\left\{\begin{array}{c}\frac{1}{\hbar}D\\ \Omega_0\left[\pi\left(\frac{m_e-m_h}{m_e+m_h}\right)^2\cdot\left(\frac{\varepsilon_0}{\varepsilon_\infty}-1\right)\frac{a_B^{\,3}\rho}{\mu}\right]^{1/2}\end{array}\right\}$$
$$\times\left[\sum_{i,j=1}^{3}P_{0i}\cos(\omega_{\mathbf{k}}t-\mathbf{kr})\frac{\chi_{ij}}{(\beta_i\beta_j)^{1/2}}P_j\right]+\Lambda_f(t). \tag{23c}$$

$$\mathbf{A}=(A_1,A_2,A_3);\quad \mathbf{P}=(P_1,P_2,P_3);$$

The upper and lower operators in the curly brackets pertain here to scattering by longitudinal acoustic and optic phonons, respectively. Generalization of the system of equations to include the case of propagation of a given coherent polariton pulse entails replacement of $\mathbf{P}_0\cos(\omega_{\mathbf{k}}t-\mathbf{k}\cdot\mathbf{r})$ by a specified function $\mathbf{P}_0(\mathbf{k},\mathbf{r},t)$, that defines the polarization vector of the initial polariton pulse. This system of differential equations was obtained in the Coulomb gauge

$$\operatorname{div}(\hat{\varepsilon}_{pol}\mathbf{A})=0,\qquad \mathbf{E}=-\frac{\varepsilon_g^{1/2}}{c}\frac{\partial}{\partial t}\mathbf{A}, \tag{24}$$

where $\mathbf{E}$ is the electric-field operator and $\hat{\varepsilon}_{\rm pol}$ is the polariton dielectric tensor and is diagonal with components

$$\varepsilon_{pol}^{ii}(\omega,\mathbf{p})=\varepsilon_g+\frac{4\pi\beta_i\omega_t^2}{\omega_t^2-\omega^2+(\hbar p^2/M_i^{ex})\omega_t-2i\omega\gamma}. \tag{25}$$

The $M_i^{\rm ex}$ in (23b) and (25) are the translational masses of the exciton along the principal axes. In addition, the phase of the linearly polarized initial polariton $\mathbf{k}$ wave was chosen to be zero, so that the corresponding components of the polarization vector $\mathbf{P}_0$ are real.

Let us dwell on the physical meaning of the equations derived. The first, (23a), is a Maxwell wave equation having in the right-hand side a source due to the exciton polarization. The second vector equation (23b) describes the evolution of the exciton-polarization vector $\mathbf{P}(\mathbf{r},t)$. In accordance with the form of the right-hand side of this equation, this evolution is governed both by the temporal variation of the vector-potential operator $\mathbf{A}(\mathbf{r},t)$ and by the presence of an initial polariton $\mathbf{k}$ wave in the semiconductor. The latter mechanism is connected with the possibility of creation or annihilation of an the exciton-polarization $\mathbf{P}(\mathbf{r},t)$ wave through scattering of the exciton component of the polariton $\mathbf{k}$ wave by longitudinal optic or acoustic phonons. An alternate form of this equation is a Schrödinger equation for the exciton wave function that describes the translational motion of the exciton as a whole. The last equation (23c) of the system is the phonon wave equation in terms of the operator of the scalar phonon potential $\Phi(\mathbf{r},t)$, which defines the corresponding operator of the longitudinal phonon displacement vector $\mathbf{u}(\mathbf{r})=\nabla\Phi(\mathbf{r},t)$. The first term in the right-hand side of this equation describes the source of the longitudinal phonon waves; this source can be called coherent and is connected with the change of the phonon-wave amplitude in scattering of excitons from the initial polariton $\mathbf{k}$ wave. The second term $\Lambda_f(t)$ in the right-hand side of (23c) is the thermal source of longitudinal phonons and its strength is determined by the corresponding phonon occupation numbers. For longitudinal optical phonons, in particular, this source can be altogether neglected.

The derived system of macroscopic equations is valid for the description of both Stokes and anti-Stokes scattering of the initial polariton $\mathbf{k}$ wave. If the coupling of the polarization $\mathbf{P}(\mathbf{r},t)$ wave and the polariton $\mathbf{k}$ wave by the phonons is neglected, an assumption formally equivalent to the case $M(\mathbf{p}-\mathbf{k})=0$, Eqs. (13a) and (23b) form a closed system of linear polariton equations[2,3] in terms of $\mathbf{A}(\mathbf{r},t)$ and $\mathbf{P}(\mathbf{r},t)$.

It is easy to derive from the system of macroscopic phononiton equations an expression for the corresponding

symmetric dielectric tensor of a semiconductor in the presence of an intense polariton **k** wave:

$$\varepsilon_{ij}(\Omega, \mathbf{p}-\mathbf{k}, \mathbf{k}) = \varepsilon_g + \frac{2\pi\omega_t^2}{\Omega}\left[\frac{Q_iQ_j}{(\beta_i\beta_j)^{1/2}}\frac{1}{\Omega-\omega_{\mathbf{k}}-\Omega_{\mathbf{p}-\mathbf{k}}} - (\Omega_{\mathbf{p}-\mathbf{k}}-\omega_{\mathbf{p}}^{ex})\frac{\delta_{ij}}{\beta_i}\right]^{-1}. \quad (26)$$

This expression was derived for the case when one can neglect in the system (23) the exciton and phonon damping and also the action of the thermal source $\Lambda_f(t)$. In analogy with (13), we have here

$$Q_i = \sum_{j=1}^{3}\alpha_{j\mathbf{k}}M_{ji}(\mathbf{p}-\mathbf{k}), \quad (27)$$

where the c-numbers $\alpha_{j\mathbf{k}}$ describe the macroscopic occupation of the initial **k** mode with coherent excitons polarized along the j axis. We note that the general form of the phonoriton dispersion equation that corresponds to the system (23) and to the dielectric tensor (16) is a rather complicated algebraic equation of seventh order even for the case of the resonant anti-Stokes interaction approximation. The simpler case described by Eq. (16) with the substitution $E = \Omega - \omega_{\mathbf{k}}$ can apparently occur only in certain special scattering geometries.

We note one more interesting problem encountered in the analysis of the macroscopic equations (23) for the case of scattering by longitudinal acoustic phonons, viz., two phonoriton waves having the same frequency can correspond to an external acoustic or polariton wave of a specified frequency in the range in which the phonoriton restructuring of the lower dispersion polariton wave and of the acoustic wave takes place. This raises the problem of specifying additional boundary conditions, a problem that has a direct bearing on the phonon equation (23c), in contrast to the analogous problem when account is taken of spatial dispersion for the polariton case. If the external acoustic or polariton wave is coherent (as is the electromagnetic wave that penetrates through the crystal boundary), its propagation in that region of the superconductor which is excited by the initial intense polariton **k** wave is described by the system of coherent equations (23). This system expressed in terms of the c-numbers $\mathbf{A}(\mathbf{r},t)$,$\mathbf{P}(\mathbf{r},t)$ and $\Phi(\mathbf{r},t)$ describes only coherent phonoriton excitations and does not contain the thermal source $\Lambda_f(t)$ in the right-hand side of (23c), at all since $\langle\Lambda_f(t)\rangle = 0$. In this case the additional phonon boundary conditions apparently take the form

$$\nabla\Phi(rt)|_{S=\mathbf{u}_0(t)}, \quad (28)$$

where $\mathbf{u}_0(t)$ is a given function that describes the coherent oscillations on the boundary S. In the particular case when the crystal boundary coincides with the region where the initial polariton **k** wave is excited, the corresponding function satisfies $\mathbf{u}_0(t) = 0$, meaning that there are no coherent phonon oscillations on the crystal boundary.

From an analysis of the form of the macroscopic phonoriton equations (23) it can be concluded that the premise of macroscopic occupation of the polariton **k** mode is manifested in these equations only implicitly, namely, via the specified exciton-polarization amplitude $\mathbf{P}_0$ that is connected with the initial polariton **k** wave. Moreover, to a certain degree these equations have the customary nonlinear-optics form. In many problems, however, it is just the macroscopic occupation of the polariton **k** mode which plays an exclusive role. In particular, the very description of the propagation of the polariton **k** wave calls for a special approach.[1] One encounters here, for example, a question such as the preservation of the degree of coherence of the initial polariton **k** wave in real kinetic scattering of **k** excitons from a macroscopically occupied mode, followed by a probable return of these scattered exciton to the **k** mode. A nontrivial aspect of problems of this type is that the excitons of the macroscopically occupied **k** mode are not in kinetic equilibrium with the system of scattered **P** excitons, a situation that requires a separate analysis of how the **k**-exciton Bose condensate evolves. The last problem does not arise at all when the Belyaev diagrammatic technique[13] is used to describe an equilibrium Bose-particle system with condensate in the $\mathbf{k} = 0$ mode.

We note in conclusion that a spectrum restructuring similar to that considered above can occur under much more general assumption: any anti-Stokes three-particle scattering of a sufficiently strong **k** wave by a translationally symmetric crystalline excitation can be accompanied by such restructuring. In particular, a similar effect results from three-particle interaction between the exciton component of a polariton wave and biexcitons in direct-gap semiconductors.[14,15]

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