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Polariton theory of resonant Raman scattering in polar semiconductors: local-field effects and crystal optics approximation

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Abstract. The theory of the first-order resonant Raman scattering in polar semiconductors, when the energy of the incident photons is in the region of the exciton energies, is presented. Local-field effects are taken into account. The explicit introduction of boson annihilation and creation operators for the exciton states, as well as the treatment of the phonon system within the framework of perturbation theory, are avoided. The method of Legendre transforms is used to derive the Bethe–Salpeter equation for the two-particle electron–hole Green function. In the case of the first-order Raman effect only the single-phonon line diagrams, contributing to the kernel of the Bethe–Salpeter equation, must be taken into consideration. This treatment provides an equation for obtaining the ‘dressed’ (by the interaction with phonons) polariton spectra. Knowledge of the imaginary part of the polariton frequency provides both the decay rate of the number of the quanta in the incident radiation and the Raman efficiency. On the basis of this theory the influence of local-field effects on the Raman efficiency is discussed.

1. Introduction

The interest in polariton approaches to resonant Raman scattering (RRS) and to the theoretically equivalent resonant Brillouin scattering (RBS) can be understood from the fact that those techniques have been established as the best ones for the investigation of exciton states and excitonic polariton parameters (Kotels 1982, Weisbuch and Ulbrich 1982).

Turning our attention to the theoretical situation, we find that two approaches have been used to define RRS (or RBS) cross-sections within the polariton framework. The first approach is based on the factorisation of the three sequential scattering events.

(i) The incident photon is transmitted across the interface and produces a polariton inside the crystal.

(ii) The scattering from one polariton state to another occurs due to the emission or absorption of a phonon.

(iii) The scattered polariton is transmitted out of the crystal to produce the scattered photon.

Although most theoretical efforts have been focused mainly on step (ii) (Ovander 1961, 1962, Mills and Burstein 1969, Hopfield 1969) in order to make the first approach

complete, we must include all three steps (i)–(iii), especially solid angle effects due to the discontinuous change in momentum across the boundary in step (iii) (Lax and Nelson 1975).

The second approach, originating with the works by Brenig (Brenig *et al* 1972) and Zeyher (Zeyher *et al* 1972, 1974), is to consider the Raman scattering of photons in a crystal as a single unified process involving refraction, absorption and scattering. In terms of the above method RRS is regarded as a combination of elastic surface scattering of phonons at the vacuum–crystal interface and inelastic scattering of excitonic polaritons inside the semi-infinite crystal. Surface scattering is treated by defining normal modes over the whole space, in a manner that incoming and outgoing normal modes reduce asymptotically to free photons far from the crystal, but they are excitonic polaritons inside the crystal. The inelastic phonon scattering between the normal modes inside the crystal is described by using the Born approximation.

At first sight the above mentioned approaches are two independent ways to formulate the RRS cross section. However, the second approach has been regarded as the better one, since it is free from the factorisation approximation for sequential scattering events. The equivalence between the two approaches for the RRS cross-section calculation (Matsushita and Nakayama 1984) is now well established.

Turning our attention to the details of the two equivalent approaches, we find some disadvantages as follows.

(i) The phonon system has been treated within the framework of perturbation theory, while the interaction with phonons may become substantial, especially in the presence of polar longitudinal optical phonons in non-centrosymmetric polar crystals.

(ii) With the exception of the work by Mills and Burstein (1969), almost all of the above mentioned studies are based on the complicated procedure of introducing boson annihilation and creation operators for the exciton states (see for example Steyn-Ross and Gardiner 1983).

(iii) The contribution of the local-field effects to the polariton spectra, and therefore to the RRS cross section, has not been taken into consideration.

The purpose of this paper is to carry on our recently published field theoretical approach to the theory of polaritons (Glinskii and Koinov 1986, 1987, Koinov and Glinskii 1988) in order to overcome the above mentioned disadvantages of the RRS cross section calculations.

In § 2 we outline the general viewpoint and some of the main results of our field theoretical approach to the polariton theory. We then derive an expression for the Raman efficiency which takes into account the local-field effects. In § 3 we give some explicit numerical and graphical results for the RRS cross section in copper halides in order to evaluate how large the local-field effects are compared to the older work by Mills and Burstein (1969).

2. First-order resonant Raman scattering through excitonic polaritons

2.1. Excitation spectra in zero-order approximation

In a previous publication (Koinov and Glinskii 1988), referred to as paper 1, we have pointed out that the photon $D_{\alpha\beta}(Q; i\omega_p)$, phonon

$$S_{\alpha\beta} \left(\begin{matrix} l & l' \\ \mathbf{K} & \mathbf{K}' \end{matrix} \middle| i\omega_p \right)$$

and two-particle electron-hole Green function

$$K \left(\begin{matrix} r_1\sigma_1 & r_3\sigma_3 \\ r_2\sigma_2 & r_4\sigma_4 \end{matrix} \middle| u_{21}; u_{43}; i\omega_p \right)$$

have common poles, located at the energies $\hbar\omega_\nu(Q)$, which are just the polariton modes with a band index ν and a wavevector Q (Q is a vector within the Brillouin zone). The polariton spectrum has to be computed numerically for actual crystals with appropriate values for 'bare' phonon energies $\hbar\Omega_\lambda(Q)$ using the exact one-electron states. It can be said that for a wavevector Q such that cQ is in the region of exciton energies (c is the vacuum speed of light), the branches in the high-energy part of the spectrum are mostly excitonic polaritons with a small phonon content, and the low-energy ones are mostly phonons with a small polariton contribution. The number of branches is equal to the total number of modes. In the limit when the photon-phonon interaction is vanishingly small, referred to as the zero-order approximation, we have the specific case where the excitons and photons are coupled into excitonic polaritons, while the phonon system is independent. In the zero-order approximation the photon and the two-particle electron-hole Green functions have common poles $\omega_\nu^{(0)}(Q)$, while the phonon Green function is equal to the 'bare' one. Thus, taking the limit $\chi_{\alpha\beta}(z|\xi) \rightarrow 0$ in equations (43) of paper 1, one sees that

$$D_{\alpha\beta}^{-1}(Q; i\omega_p)_{\chi \rightarrow 0} = D_{\alpha\beta}^{(e-\omega)^{-1}}(Q; i\omega_p) = D_{\alpha\beta}^{(0)^{-1}}(Q; i\omega_p) - \Pi_{\alpha\beta}^{(e-\omega)}(Q; i\omega_p) \quad (1a)$$

$$\begin{aligned} K^{-1} \left(\begin{matrix} r_1\sigma_1 & r_3\sigma_3 \\ r_2\sigma_2 & r_4\sigma_4 \end{matrix} \middle| u_{21}; u_{43}; i\omega_p \right)_{\chi \rightarrow 0} &= K_{e-\omega}^{-1} \left(\begin{matrix} r_1\sigma_1 & r_3\sigma_3 \\ r_2\sigma_2 & r_4\sigma_4 \end{matrix} \middle| u_{21}; u_{43}; i\omega_p \right) \\ &= K_M^{E-1} \left(\begin{matrix} r_1\sigma_1 & r_3\sigma_3 \\ r_2\sigma_2 & r_4\sigma_4 \end{matrix} \middle| u_{21}; u_{43}; i\omega_p \right) - \frac{\delta(u_{21})\delta(u_{43})}{\hbar^2 c^2 V} \\ &\times \sum_{Q \in \text{BZ}} \langle r_1, \sigma_1 | \hat{j}_\alpha(Q) | r_2, \sigma_2 \rangle D_{\alpha\beta}^{(0)}(Q; i\omega_p) \langle r_4, \sigma_4 | \hat{j}_\beta(-Q) | r_3, \sigma_3 \rangle \end{aligned} \quad (1b)$$

$$S_{\alpha\beta}^{-1} \left(\begin{matrix} l & l' \\ \mathbf{K} & \mathbf{K}' \end{matrix} \middle| i\omega_p \right)_{\chi \rightarrow 0} = S_{\alpha\beta}^{(0)^{-1}} \left(\begin{matrix} l & l' \\ \mathbf{K} & \mathbf{K}' \end{matrix} \middle| i\omega_p \right). \quad (1c)$$

In the above equation and throughout this paper we use the same notation as in paper 1. K_M^E is a two-particle electron-hole Green function for 'mechanical' excitons, which takes into account the Elliott exchange interaction (see definition (42b) of paper 1). The photon self-energy $\Pi_{\alpha\beta}^{(e-\omega)}$ is obtained from equation (43c) of paper 1 by taking the limit $\chi_{\alpha\beta}(z|\xi) \rightarrow 0$:

$$\begin{aligned} \Pi_{\alpha\beta}^{(2)}(Q; i\omega_p)_{\chi \rightarrow 0} &= \Pi_{\alpha\beta}^{(e-\omega)}(Q; i\omega_p) = \frac{1}{\hbar^2 c^2 V} \langle r_2, \sigma_2 | \hat{j}_\alpha(Q) | r_1, \sigma_1 \rangle \\ &\times K_M^E \left(\begin{matrix} r_1\sigma_1 & r_3\sigma_3 \\ r_2\sigma_2 & r_4\sigma_4 \end{matrix} \middle| u_{21} = 0; u_{43} = 0; i\omega_p \right) \langle r_3, \sigma_3 | \hat{j}_\beta(-Q) | r_4, \sigma_4 \rangle. \end{aligned} \quad (1d)$$

In the zero-order approximation the spectrum of the excitonic polaritons $\omega_\nu^{(0)}(Q)$ are solution of the following equation:

$$\det \left\| \left(\frac{\omega_\nu^{(0)}}{c} \right)^2 \varepsilon_{\alpha\beta}^{(e-\omega)}(Q; \omega_\nu^{(0)}) - \delta_{\alpha\beta} Q^2 + Q_\alpha Q_\beta \right\| = 0 \quad (2a)$$

where the tensor $\varepsilon_{\alpha\beta}^{(e-\omega)}$ follows from equation (43a) of paper 1 in the limit $\chi_{\alpha\beta}(z|\xi) \rightarrow 0$:

$$\varepsilon_{\alpha\beta}(Q; i\omega_p)_{\chi \rightarrow 0} = \varepsilon_{\alpha\beta}^{(e-\omega)}(Q; i\omega_p) \delta_{\alpha\beta} - \frac{4\pi\hbar c^2}{(i\omega_p)^2} \Pi_{\alpha\beta}^{(e-\omega)}(Q; i\omega_p). \quad (2b)$$

Let us define the photon polarisation vectors $e_\alpha(i; Q)$ ($i = 1, 2, 3$) in a manner that $e_\alpha(3; Q) = Q_\alpha/|Q|$ and the following normalisation conditions must be fulfilled:

$$\sum_{\alpha=x,y,z} e_\alpha(i; Q) e_\alpha^*(j; Q) = \delta_{ij} \quad \sum_{i=1,2,3} e_\alpha(i; Q) e_\beta^*(i; Q) = \delta_{\alpha\beta}.$$

In the new basis the photon Green function (1a) and the dielectric tensor (2a) assume the form

$$D_{ij}^{(e-\omega)}(Q; i\omega_p) = \sum_{\alpha,\beta} e_\alpha(i; Q) D_{\alpha\beta}^{(e-\omega)}(Q; i\omega_p) e_\beta^*(j; Q) \quad (3a)$$

$$\varepsilon_{ij}^{(e-\omega)}(Q; i\omega_p) = \sum_{\alpha,\beta} e_\alpha(i; Q) \varepsilon_{\alpha\beta}^{(e-\omega)}(Q; i\omega_p) e_\beta^*(j; Q). \quad (3b)$$

For simplicity we will consider only crystals with a cubic point group. In this case, the symmetry considerations imply $D_{ij}^{(e-\omega)}(Q; i\omega_p) = \delta_{ij} D_i(Q; i\omega_p)$ and $\varepsilon_{ij}^{(e-\omega)}(Q; i\omega_p) = \delta_{ij} \varepsilon(Q; i\omega_p)$ where, for ($i = 1, 2$)

$$D_i(Q; i\omega_p) = 4\pi\hbar c^2 / [(i\omega_p)^2 \varepsilon(Q; i\omega_p) - c^2 Q^2] \quad (4a)$$

$$D_3(Q; i\omega_p) = 4\pi\hbar c^2 / (i\omega_p)^2 \varepsilon(Q; i\omega_p). \quad (4b)$$

In the case of cubic crystals there are two types of excitonic polaritons: transverse normal modes $\omega_\nu^{(0)}(Q)$ and longitudinal ones $\Omega_\nu^{(0)}(Q)$. They satisfy the following equations:

$$[\omega_\nu^{(0)}(Q)]^2 \varepsilon(Q; \omega_\nu^{(0)}) = c^2 Q^2 \quad (5a)$$

$$\varepsilon(Q; \Omega_\nu^{(0)}(Q)) = 0. \quad (5b)$$

The contributions of the two types of polaritons to the analytical continuations of the functions (4a) and (4b) are

$$D_i(Q; \omega) \rightarrow \frac{4\pi\hbar c^2}{\omega \rightarrow \omega_\nu^{(0)} \{(\partial/\partial\omega)[\omega^2 \varepsilon(Q; \omega)]\}_{\omega=\omega_\nu^{(0)}}} \left(\frac{1}{\omega - \omega_\nu^{(0)} + i0^+} \right) \quad (5c)$$

$$D_3(Q; \omega) \rightarrow \frac{4\pi\hbar c^2}{\omega \rightarrow \Omega_\nu^{(0)} (\Omega_\nu^{(0)})^2 \{(\partial/\partial\omega)\varepsilon(Q; \omega)\}_{\omega=\Omega_\nu^{(0)}}} \left(\frac{1}{\omega - \Omega_\nu^{(0)} + i0^+} \right). \quad (5d)$$

2.2. Excitation spectra in the case of an instantaneous electron-phonon interaction

In this section our attention will be focused on step (ii) of the first approach, that is the scattering from one polariton state to another due to the emission (Stokes scattering) or absorption (anti-Stokes scattering) of a phonon.

A more realistic approximation than the zero-order one is to assume that the excitonic polaritons and the phonons interact via an instantaneous electron–phonon interaction. In this case the two-particle electron–hole Green function and the photon propagator have common poles, while the poles of the phonon Green function are located at the energies $\hbar\tilde{\Omega}_\mu(Q)$ and describe the renormalised phonon energies due to the interaction with the photons (see §4 of paper 1). The new phonon frequencies $\tilde{\Omega}_\mu(Q)$ and the corresponding eigenvectors $\varepsilon_\alpha^k(\mu; Q)$ can be obtained from a standard eigenvector–eigenvalue problem.

Let us now turn to a detailed discussion of the Bethe–Salpeter equation for the two-particle electron–hole Green function. The Green function \tilde{K} ‘dressed’ by the instantaneous electron–phonon interaction satisfies the Bethe–Salpeter equation

$$\tilde{K}^{-1} \begin{pmatrix} y & x'' \\ x & y'' \end{pmatrix} \tilde{K} \begin{pmatrix} x'' & y' \\ y'' & x' \end{pmatrix} = \delta(x - x') \delta(y - y') \quad (6a)$$

where the inverse propagator \tilde{K}^{-1} can be written in the form

$$\tilde{K}^{-1} \begin{pmatrix} y & x' \\ x & y' \end{pmatrix} = K_{e-\omega}^{-1} \begin{pmatrix} y & x' \\ x & y' \end{pmatrix} - \Delta I \begin{pmatrix} y & x' \\ x & y' \end{pmatrix}. \quad (6b)$$

Here $K_{e-\omega}^{-1}$ is defined by equation (1b) and ΔI is an additional term to the kernel of the Bethe–Salpeter equation due to the scattering by phonons.

Since in this paper we confine our attention to the first-order Raman effect, we consider only the contributions to the kernel ΔI that involve a single-phonon line. Those contributions can be obtained from the kernel $\delta\Sigma/\delta G$ of the Bethe–Salpeter equation (equation (44) of paper 1) in the limit of an instantaneous electron–phonon interaction. To do that, we must first obtain an equation (Edward’s equation) for the electron–phonon vertex. This equation can be easily obtained by means of the Schwinger equation (37c) of paper 1 after differentiation over the phonon source, by using the method of Legendre transforms. The corresponding Edward’s equation has the form

$$\begin{aligned} \theta_\alpha(y, x|\xi) &= \theta_\alpha^{(e)}(y, x|\xi) + \left[K_M^{E-1} \begin{pmatrix} y & x' \\ x & y' \end{pmatrix} - \Gamma_\beta^{(0)}(y, x|z) \right. \\ &\quad \left. \times D_{\beta\gamma}^{(0)}(z, z') \Gamma_\gamma^{(0)}(y', x'|z') \right] G(x', y'') G(x'', y') \theta_\alpha(y'', x''|\xi) \end{aligned} \quad (7a)$$

where we have introduced the effective electron–phonon vertex

$$\begin{aligned} \theta_\alpha^{(e)}(y, x|\xi) &= i \frac{\delta(u - u')}{\hbar^2 c^2 V} \sum_Q \sum_{G_n, G_m} \sum_{\omega_p} \exp\{-i[\mathbf{Q} \cdot \mathbf{R}_l - \omega_p(u - w)]\} \\ &\quad \times (i\omega_p) \langle r, \sigma | \hat{j}_\beta(-\mathbf{Q} - G_n) | r', \sigma' \rangle \\ &\quad \times D_{\beta\gamma}^{(e)}(\mathbf{Q} + G_n, \mathbf{Q} + G_m; i\omega_p) P_{j\alpha}^k(\mathbf{Q} + G_m). \end{aligned} \quad (7b)$$

Here the functions $D_{\alpha\beta}^{(e)}$ and $P_{\alpha\beta}^k$ have been defined by equations (21a) and (16c) of paper 1 and R_l is the unit cell vector. In the limit of an instantaneous electron–phonon interaction we have to take into account only the instantaneous part of $\theta_\alpha^{(e)}$. Proceeding

as in paper 1 we obtained the instantaneous part of the electron–phonon vertex (7b) in the form

$$\begin{aligned} \theta_{\alpha}^{(e)INST}(y, x | \xi) &= \frac{4\pi e}{\hbar V} \delta(u - u') \delta(u - w) \sum_Q \sum_{G_n, G_m} \exp[-iQ \cdot R_I] \\ &\times \langle r, \sigma | \exp[i(Q + G_n) \cdot \hat{x}] | r', \sigma' \rangle \\ &\times \frac{1}{|Q + G_n|} \varepsilon_{\parallel}^{(e)-1}(Q + G_n; Q + G_m; 0) \frac{(Q + G_m)_{\beta}}{|Q + G_m|} P_{\beta\alpha}^{\kappa}(Q + G_m) \end{aligned} \tag{8}$$

where \hat{x} denotes a coordinate operator, and $\varepsilon_{\parallel}^{(e)-1}$ is obtained from the longitudinal part of the dielectric matrix $\varepsilon_{\alpha\beta}^{(e)}(Q + G_n; Q + G_m; 0)$ by a matrix inversion procedure (equation (31e) of paper 1).

Furthermore, it is convenient to write the kernel ΔI in (k, ω) representation, taking into account the one-particle band structure of semiconductors. By using kp perturbation theory, we have introduced (Glinskii and Koinov 1987) the basis $|S, n, k^i\rangle$, where k^i is a vector of the Brillouin zone near to the point k_0^i ; k_0^i is the vector of the conduction band minimum or the valence band maximum; i is the number of the equivalent extremum; S stands for the type of the irreducible representation of the point-group symmetry at the point k_0^i ; n is the index of the degenerate states at the point k_0^i that have the same transformation properties as the basis functions obeying the irreducible representation S . It should be noted that the basis $|S, n, k^i\rangle$ is chosen in a manner that

$$\langle S_1, n_1, k_1^{i_1} | \hat{H} | S_2, n_2, k_2^{i_2} \rangle = \delta_{i_1 i_2} \delta_{k_1^{i_1} k_2^{i_2}} H_{n_1 n_2}^{S_1 S_2}(k_1^{i_1})$$

where $H_{n_1 n_2}^{S_1 S_2}(k_1^{i_1})$ are the matrices for the kp Hamiltonian, determining the one-particle band structure.

In the limit of an instantaneous electron–phonon interaction, the Bethe–Salpeter equation (6a), written in the new basis, assumes the form

$$\bar{K}^{-1} \begin{pmatrix} 1 & 3 \\ 2 & 4 \end{pmatrix} \Big| i\omega_p \Big) \bar{K} \begin{pmatrix} 3 & 5 \\ 4 & 6 \end{pmatrix} \Big| i\omega_p \Big) = \delta_{13} \delta_{26} \tag{9a}$$

where the following abbreviations have been used $|S_1, n_1, k_1^{i_1}\rangle = |1\rangle$:

$$\bar{K} \begin{pmatrix} 1 & 3 \\ 2 & 4 \end{pmatrix} \Big| u_{21} = 0; u_{43} = 0; i\omega_p \Big) = \bar{K} \begin{pmatrix} 1 & 3 \\ 2 & 4 \end{pmatrix} \Big| i\omega_p \Big).$$

The renormalised polariton energies are just the poles of the function \bar{K} . Thus the contribution from the polariton state $\omega_{\nu}(Q)$ to the above Green function is

$$\bar{K} \begin{pmatrix} 1 & 3 \\ 2 & 4 \end{pmatrix} \Big| z \Big) = \frac{\bar{\varphi}_{21}^{\nu Q} \bar{\varphi}_{43}^{\nu Q*}}{z - \omega_{\nu}(Q) + i0^+} + \Delta \bar{K} \begin{pmatrix} 1 & 3 \\ 2 & 4 \end{pmatrix} \Big| z \Big) \tag{9b}$$

where $\Delta \bar{K}$ is a term regular at $z = \omega_{\nu}(Q)$. For a given wavevector Q , the poles $\omega_{\nu}(Q)$ and the corresponding wave functions $\bar{\varphi}_{21}^{\nu Q}$ are the solutions of the Bethe–Salpeter equation

$$\bar{K}^{-1} \begin{pmatrix} 1 & 3 \\ 2 & 4 \end{pmatrix} \Big| \omega_{\nu}(Q) \Big) \bar{\varphi}_{43}^{\nu Q} = 0. \tag{9c}$$

Taking into account only the single-phonon-line contributions to the kernel ΔI , and

using equations (38b), (44a)–(44g) of paper 1, the inverse two-particle electron-hole propagator

$$\tilde{K}^{-1} \left(\begin{array}{cc} 1 & 3 \\ 2 & 4 \end{array} \middle| \omega_\nu \right)$$

is obtained in the form

$$\begin{aligned} \tilde{K}^{-1} \left(\begin{array}{cc} 1 & 3 \\ 2 & 4 \end{array} \middle| \omega_\nu(Q) \right) &= K_M^{E-1} \left(\begin{array}{cc} 1 & 3 \\ 2 & 4 \end{array} \middle| \omega_\nu(Q) \right) \\ &\quad - \frac{1}{\hbar^2 c^2 V} \langle 1 | \hat{j}_\alpha(Q) | 2 \rangle D_{\alpha\beta}^{(0)}(Q; \omega_\nu) \langle 4 | \hat{j}_\beta(-Q) | 3 \rangle \\ &\quad - \Delta I \left(\begin{array}{cc} 1 & 3 \\ 2 & 4 \end{array} \middle| \omega_\nu(Q) \right) \end{aligned} \tag{10a}$$

where the kernel ΔI is given by the following equation:

$$\begin{aligned} \Delta I \left(\begin{array}{cc} 1 & 3 \\ 2 & 4 \end{array} \middle| \omega_\nu(Q) \right) &= \sum_\mu \sum_k [N_+(\tilde{\Omega}_\mu(\mathbf{k})) W_{\mu,k} \left(\begin{array}{cc} 1 & 5 \\ 2 & 6 \end{array} \right) \\ &\quad \times K_{e-\omega} \left(\begin{array}{cc} 5 & 7 \\ 6 & 8 \end{array} \middle| \omega_\nu(Q) - \tilde{\Omega}_\mu(\mathbf{k}) \right) W_{\mu,-k} \left(\begin{array}{cc} 7 & 3 \\ 8 & 4 \end{array} \right)] \\ &\quad + \sum_\mu \sum_k [N_-(\tilde{\Omega}_\mu(\mathbf{k})) W_{\mu,k} \left(\begin{array}{cc} 1 & 5 \\ 2 & 6 \end{array} \right) \\ &\quad \times K_{e-\omega} \left(\begin{array}{cc} 5 & 7 \\ 6 & 8 \end{array} \middle| \omega_\nu(Q) + \tilde{\Omega}_\mu(\mathbf{k}) \right) W_{\mu,-k} \left(\begin{array}{cc} 7 & 3 \\ 8 & 4 \end{array} \right)]. \end{aligned} \tag{10b}$$

Here

$$N_+(\omega) = n(\omega) + 1 \quad N_-(\omega) = n(\omega) \quad n(\omega) = [\exp(\hbar\omega/k_B T) - 1]^{-1}$$

is the Bose–Einstein distribution function of phonons. The electron–phonon effective vertex $W_{\mu,k}$ in the instantaneous limit has the form

$$\begin{aligned} W_{\mu,Q} \left(\begin{array}{cc} 1 & 3 \\ 2 & 4 \end{array} \right) &= - \frac{4\pi e}{\hbar \sqrt{V}} \left(\frac{\hbar}{2M_0 V_0} \right)^{1/2} \sum_{G_n, G_m} \frac{1}{|Q + G_n|} \varepsilon_{\parallel}^{(e)-1}(Q + G_n; Q + G_m; 0) \\ &\quad \times \frac{(Q + G_m)_\alpha}{|Q + G_m|} P_{\alpha\beta}^\kappa(Q + G_m) \varepsilon_\beta^\kappa(\mu, Q) \times [\delta_{24} \langle 1 | \exp[i(Q + G_n) \cdot \mathbf{x}] | 3 \rangle \\ &\quad - \delta_{13} \langle 4 | \exp[i(Q + G_n) \cdot \mathbf{x}] | 2 \rangle]. \end{aligned} \tag{11}$$

Here M_0 is the mass of the atoms in the unit cell, and V_0 is the volume of the cell. The symbol δ_{12} denotes $\delta_{s_1 s_2} \delta_{n_1 n_2} \delta_{i_1 i_2} \delta_{k_1 k_2}$. The terms in parentheses correspond to the interactions of the electron in the conduction band and the hole of the valence band with the phonon.

In order to obtain the solutions of equation (9c) we introduce the Green function for 'mechanical' excitons which is 'dressed' by electron-phonon interaction, and which takes into account the Elliott exchange interaction

$$\tilde{K}_M^E^{-1} \begin{pmatrix} 1 & 3 \\ 2 & 4 \end{pmatrix} \omega_\nu = K_M^E^{-1} \begin{pmatrix} 1 & 3 \\ 2 & 4 \end{pmatrix} \omega_\nu - \Delta I \begin{pmatrix} 1 & 3 \\ 2 & 4 \end{pmatrix} \omega_\nu. \quad (12)$$

Proceeding as in paper 1, it is easy to show that for a given wavevector Q the solutions $\omega_\nu(Q)$ of equation 9(c) are solutions of the following equation:

$$\det \left\| \left(\frac{\omega_\nu}{c} \right)^2 \tilde{\epsilon}_{\alpha\beta}(Q, \omega_\nu) - \delta_{\alpha\beta} Q^2 + Q_\alpha Q_\beta \right\| = 0 \quad (13)$$

where

$$\tilde{\epsilon}_{\alpha\beta}(Q, \omega_\nu) = \delta_{\alpha\beta} - \frac{4\pi\hbar c^2}{\omega_\nu^2} \tilde{\Pi}_{\alpha\beta}(Q, \omega_\nu). \quad (14a)$$

Here the proper self-energy part $\tilde{\Pi}_{\alpha\beta}$ has the form

$$\tilde{\Pi}_{\alpha\beta}(Q, \omega_\nu) = \frac{1}{\hbar^2 c^2 V} \langle 2 | \hat{j}_\alpha(Q) | 1 \rangle \tilde{K}_M^E \begin{pmatrix} 1 & 3 \\ 2 & 4 \end{pmatrix} \omega_\nu \langle 3 | \hat{j}_\beta(-Q) | 4 \rangle. \quad (14b)$$

This proper self-energy part contains both a real and an imaginary part. The real part contributes to a shift in the energy $\hbar\omega_\nu^{(0)}(Q)$ of the excitonic polaritons, while their damping rate is proportional to the imaginary part of $\tilde{\Pi}_{\alpha\beta}(Q, \omega_\nu)$.

2.3. Theory of first-order Raman effect

Since the damping rate of the polariton mode is given by the imaginary part of the corresponding pole $\omega_\nu(Q)$ of the two-particle electron-hole Green function, the decay rate of the number of quanta in the incident radiation is equal to $2 \text{Im } \omega_\nu(Q)$ (Mills and Burstein 1969).

In the cubic crystals there are two types of excitonic polaritons. For a given wavevector Q , the transverse polariton modes $\omega_\nu(Q)$ are solutions of the following equation:

$$\omega_\nu^2 \tilde{\epsilon}(Q, \omega_\nu) = c^2 Q^2 \quad (15a)$$

where

$$\tilde{\epsilon}(Q, \omega_\nu) = 1 - (4\pi\hbar c^2 / \omega_\nu^2) \tilde{\Pi}(Q, \omega_\nu) \quad (15b)$$

$$\tilde{\Pi}(Q, \omega_\nu) = \sum_{\alpha, \beta} e_\alpha(i; Q) \tilde{\Pi}_{\alpha\beta}(Q, \omega_\nu) e_\beta^*(i; Q). \quad (15c)$$

Here $i = 1$ or 2 and the phonon-polarisation vectors $e_\alpha(i; Q)$ have been introduced in § 2.1.

It is worth noting that the kernel (10b) involves only a single-phonon-line contribution, while the two-particle Green function K_M^E , and therefore the proper self-energy $\tilde{\Pi}(Q, \omega_\nu)$ obtained by the ladder graph approximations, contain higher-order phonon contributions.

In the specific case, where the electron–phonon interaction is small, one can solve equation (15a) by expanding $\omega_\nu(Q)$ in a power series with respect to the small parameter of the electron–phonon interaction

$$\omega_\nu(Q) = \omega_\nu^{(0)} + \Delta\omega_\nu^{(1)}(Q) + i\delta\omega_\nu^{(1)}(Q) + \dots \quad (16)$$

where $\Delta\omega_\nu^{(1)}$ and $\delta\omega_\nu^{(1)}$ are the real and imaginary parts of the lowest-order contribution to the excitonic polariton dispersion relation $\omega_\nu^{(0)}(Q)$ due to the electron–phonon interaction. Since we treat this interaction as a small perturbation, the frequency shift $\Delta\omega_\nu^{(1)}$ can be neglected.

In order to obtain $\delta\omega_\nu^{(1)}$ we separate the lowest-order contribution to $\tilde{\epsilon}(Q, \omega_\nu^{(0)})$ as follows:

$$\tilde{\epsilon}(Q, \omega_\nu^{(0)}) = \epsilon(Q, \omega_\nu^{(0)}) - \frac{4\pi\hbar c^2}{(\omega_\nu^{(0)})^2} \delta\tilde{\Pi}^{(1)}(Q, \omega_\nu^{(0)}) \quad (17a)$$

where $\epsilon(Q, \omega_\nu^{(0)})$ can be obtained by means of equations (2)–(3) of the present paper and equations (35) from our previous work (Glinskii and Koinov 1987):

$$\epsilon(Q, \omega_\nu^{(0)}) = 1 - \frac{4\pi}{(\omega_\nu^{(0)})^2 V} \sum_l \left(\frac{|j_x^l(Q)|^2}{(\omega_\nu^{(0)}(Q) - \omega_l(Q)} - \frac{|j_x^l(-Q)|^2}{\omega_\nu^{(0)}(Q) + \omega_l(Q)} \right) \quad (17b)$$

where $\omega_l(Q)$ is the dispersion relation of the ‘mechanical’ excitons, when the Elliott exchange interaction was taken into account, and $j_\alpha^l(Q)$ is the exciton current.

Since the energy of the incident photon has been assumed to be in the region where the exciton $\omega_l(Q)$ and photon $\Omega_{pt}(Q) = cQ$, dispersion relations are intersected, the condition $\omega_l(Q) \neq \omega_\nu^{(0)}(Q)$ must be fulfilled. In this energy region the dielectric function (21b) is real since the imaginary part of K_M^E is equal to zero.

The lowest-order contribution $\delta\tilde{\Pi}^{(1)}$ in equation (17a) can be obtained from equations (14b) and (15c):

$$\begin{aligned} \delta\tilde{\Pi}^{(1)}(Q, \omega_\nu^{(0)}) &= \frac{1}{\hbar^2 c^2 V} \langle 2 | \hat{j}_\alpha(Q) e_\alpha(i; Q) | 1 \rangle K_M^E \begin{pmatrix} 1 & 3 \\ 2 & 4 \end{pmatrix} \left| \omega_\nu^{(0)}(Q) \right\rangle \\ &\times \Delta I \begin{pmatrix} 3 & 5 \\ 4 & 6 \end{pmatrix} \left| \omega_\nu^{(0)}(Q) \right\rangle K_M^E \begin{pmatrix} 5 & 7 \\ 6 & 8 \end{pmatrix} \left| \omega_\nu^{(0)}(Q) \right\rangle \langle 7 | \hat{j}_\beta(-Q) e_\beta(i; -Q) | 8 \rangle. \end{aligned} \quad (17c)$$

Since $\epsilon(Q; \omega_\nu^{(0)})$ is real, from equation (15a) follows

$$\delta\omega_\nu^{(1)}(Q) = (2\pi\hbar/Q) V_E^{\nu Q} \text{Im} \delta\tilde{\Pi}^{(1)}(Q, \omega_\nu^{(0)}) \quad (18a)$$

where the ‘energy transport’ velocity $V_E^{\nu Q}$ is defined by

$$V_E^{\nu Q} = 2c^2 Q \{ (\partial/\partial\omega) [\omega^2 \epsilon(Q, \omega)]_{\omega=\omega_\nu^{(0)}(Q)} \}^{-1}. \quad (18b)$$

In order to obtain an exact expression for $\text{Im} \delta\tilde{\Pi}^{(1)}$ one has to know the exact form of the two-particle propagator

$$K_{e-\omega} \begin{pmatrix} 1 & 3 \\ 2 & 4 \end{pmatrix} \left| \omega \right\rangle.$$

For ω near to $\omega_\nu^{(0)}(Q)$ we have

$$K_{e-\omega} \begin{pmatrix} 1 & 3 \\ 2 & 4 \end{pmatrix} \left| \omega \right\rangle = \varphi_{21}^{\nu Q} \varphi_{43}^{\nu Q*} / (\omega - \omega_\nu^{(0)}(Q) + i0^+) + \Delta K_{e-\omega} \begin{pmatrix} 1 & 3 \\ 2 & 4 \end{pmatrix} \left| \omega \right\rangle \quad (19a)$$

where $\Delta K_{e-\omega}$ is a term regular at $\omega = \omega_\nu^{(0)}(Q)$.

In this paper we are primarily interested in the RRS processes: that means the polariton mode $\omega_\nu^{(0)}(Q)$ emits (Stokes scattering) or absorbs (anti-Stokes) a phonon of a wavevector k and energy $\hbar\tilde{\Omega}_\mu(k)$, thus making a transmission to the final polariton state $\omega_\nu^{(0)}(Q') = \omega_\nu^{(0)}(Q) \pm \tilde{\Omega}_\mu(k)$. For those processes we can neglect the regular term in equation (19a). Since the resonant terms of $K_{e-\omega}$ and $D_i(Q, \omega)$ are connected by equations (45) of paper 1, the following equation has to take place:

$$\begin{aligned} \varphi_{2\Gamma}^{\nu Q} \varphi_{4\Gamma}^{\nu Q*} &= \frac{1}{\hbar^2 c^2 V} \frac{2\pi\hbar}{Q} V_{\text{E}}^{\nu Q} \sum_{i=1,2} K_{\text{M}}^{\text{E}} \begin{pmatrix} 1 & 5 \\ 2 & 6 \end{pmatrix} \omega_\nu^{(0)}(Q) \\ &\times \langle 5 | \hat{j}_\alpha(-Q) e_\alpha(i; -Q) | 6 \rangle \langle 8 | \hat{j}_\beta(Q) e_\beta(i; Q) | z \rangle \\ &\times K_{\text{M}}^{\text{E}} \begin{pmatrix} 7 & 3 \\ 8 & 4 \end{pmatrix} \omega_\nu^{(0)}(Q). \end{aligned} \quad (19b)$$

If one takes into account only the resonant term in equation (19b), then the decay rate of the number of quanta in the incident radiation $\Gamma(\nu \rightarrow \nu'; Q)$ that comes from the processes in which an excitonic polariton mode (ν, Q) emits or absorbs a phonon of a wavevector k , thus making a transition to the final polariton state $(\nu', Q + k)$, is obtained in the following form:

$$\begin{aligned} \Gamma_\pm(\nu \rightarrow \nu'; Q) &= (2\pi/\hbar V) \sum_\mu \sum_k |M_{\mu,k}(Q, \omega_\nu^{(0)}(Q); Q \\ &+ k, \omega_{\nu'}^{(0)}(Q + k)|^2 N_\pm(\tilde{\Omega}_\mu(k)) \delta[\hbar\omega_{\nu'}^{(0)}(Q + k) \\ &\pm \hbar\tilde{\Omega}_\mu(k) - \hbar\omega_\nu^{(0)}(Q)] \end{aligned} \quad (20a)$$

where

$$\begin{aligned} M_{\mu,k}(Q, \omega_\nu^{(0)}(Q); Q + k, \omega_{\nu'}^{(0)}(Q + k)) &= \sum_l \sum_{l'} \sum_{j=1,2} \{e_\alpha(i; Q) \\ &\times S^l(Q; \omega_\nu^{(0)}(Q)) W_{\mu,k}(l, Q; l', Q + k) S_{\beta}^{l'*} \\ &\times (Q + k; \omega_{\nu'}^{(0)}(Q + k)) e_\beta^*(j, Q + k) \\ &+ e_\alpha^*(i; -Q) S_\alpha^{l*}(-Q; -\omega_\nu^{(0)}(Q)) W_{\mu,k}^*(l, -Q; l', -Q - k) \\ &\times S_{\beta}^{l'}(-Q - k; -\omega_{\nu'}^{(0)}(Q + k)) e_\beta(j; -Q - k)\} \end{aligned} \quad (20b)$$

and

$$\begin{aligned} S_\alpha^l(Q; \omega_\nu^{(0)}(Q)) &= \left(\frac{2\pi}{V}\right)^{1/2} \frac{1}{(\omega_\nu^{(0)}(Q))^{1/2}} \frac{j_\alpha^l(Q)}{[\omega_\nu^{(0)}(Q) - \omega_l(Q)]} \\ &\times \left\{ 1 + \frac{2\pi}{\omega_\nu^{(0)}(Q)V} \sum_{l'} \left[\frac{|j_x^{l'}(Q)|^2}{[\omega_\nu^{(0)}(Q) - \omega_{l'}(Q)]^2} \right. \right. \\ &\left. \left. - \frac{|j_x^{l'}(-Q)|^2}{[\omega_\nu^{(0)}(Q) + \omega_{l'}(Q)]^2} \right] \right\}^{-1/2}. \end{aligned} \quad (20c)$$

Here $W_{\mu,k}$ is the electron-phonon matrix element

$$W_{\mu,k}(l, Q; l', Q + k) = \Psi_{43W_{\mu,k}}^{lQ*} \begin{pmatrix} 3 & 1 \\ 4 & 2 \end{pmatrix} \Psi_{21}^{l'Q+k} \quad (20d)$$

where Ψ_{21}^{lQ} are the wavefunctions of the ‘mechanical’ excitons, when the Elliott exchange interaction was taken into account, and $|\bar{2}\rangle = \hat{T}|s_2, n_2, k_2^i\rangle - \hat{T}$ is the time inversion operator. In equation (20d) the even and odd indices indicate the states from the valence and conduction band respectively.

Let us now consider the specific case when the incident radiation is in the frequency region below $\omega_l(Q)$ associated with the smallest band gap. In this case, however, the normal mode $\omega_v^{(0)}(Q)$, as well as the final polariton state $\omega_v^{(0)}(Q + k)$, are on the lowest-frequency branch of the polariton dispersion relation. It is easily seen that if (i) we do not consider the local-field effects, and (ii) we put the renormalised phonon frequencies $\bar{\Omega}_\mu$ equal to the ‘bare’ ones, equations (24) give the same results as those obtained by Mills and Burstein (1969).

Furthermore, we will consider the RRS by optical phonons in polar semiconductors. A characteristic feature of the optical processes in polar semiconductors is the predominance of the interaction of the photons with LO phonons as compared with phonons of the other types. Thus in cubic polar semiconductors with two atoms per cell, $\bar{\Omega}_{LO}(k) \approx \bar{\Omega}_{LO}(0) = \omega_0$, and by neglecting the short-range part of the effective electron-phonon vertex (11) in comparison to its long-range part, we have

$$W_{LOk} \begin{pmatrix} 1 & 3 \\ 2 & 4 \end{pmatrix} = - \left\{ \frac{2\pi\omega_0 e^2}{\hbar V} \left(\frac{1}{\kappa_\infty} - \frac{1}{\kappa_0} \right) \right\}^{1/2} [\delta_{24} \delta_{s_1 s_3} \delta_{n_1 n_3} \delta_{i_1 i_3} \delta_{k_1^i, k_3^i+k} - \delta_{13} \delta_{s_2 s_4} \delta_{n_2 n_4} \delta_{i_2 i_4} \delta_{k_2^i, k_4^i+k}] \quad (21)$$

where κ_∞ and κ_0 are the high- and low-frequency limit of the dielectric matrix (see § 4 of paper 1).

Finally, we can obtain an expression for the Raman efficiency per unit length per unit solid angle $(1/L)dS/d\Omega_s$. Proceeding in a similar way as in the paper by Mills and Burstein (1969), one sees that the Raman efficiency can be written in the form

$$\frac{1}{L} \frac{dS}{d\Omega_s} = \frac{N_\pm(\omega_0)(Q+k)^2 |M_{LO,k}(Q, \omega_v^{(0)}(Q); Q+k, \omega_v^{(0)}(Q+k))|^2}{4\pi^2 \hbar^2 V_E^v \cdot V_g^{v,Q+k}} \quad (22)$$

where $V_g^{v,Q} = d\omega_v^{(0)}(Q)/dQ$ is the group velocity.

3. Results and discussions

In order to obtain a simple understanding of the corrections to the Raman efficiency due to local-field effects, we consider direct-gap semiconductors with T_D point-group symmetry. We assume that the lowest conduction band has a Γ_6 symmetry and the uppermost valence band is the one with Γ_8 symmetry.

For a given wavevector the transverse polariton modes $\omega_v^{(0)}(Q)$ satisfy equation (15a), where in the case of a simple one-oscillator model $\varepsilon(Q, \omega)$, defined by equation (17b), assumes the form

$$\varepsilon(Q, \omega) = \varepsilon_b \left(1 + \frac{E_L^2 - E_T^2}{E_T^2(Q) - \hbar^2 \omega^2} \right). \quad (23)$$

Here ε_b is the ‘background’ dielectric constant, $E_T(Q) = E_T + \hbar^2 Q^2/2M_{exc}$ is the

dispersion of the ‘mechanical’ excitons when the Elliott exchange interaction was taken into account, and M_{exc} is the effective exciton mass. E_T and $E_L = E_T + \Delta_{LT}$ denote the energies of the transverse and longitudinal excitons at $Q = 0$; Δ_{LT} is the longitudinal–transverse splitting.

If the local-field effects are neglected, the dielectric function $\epsilon(Q, \omega)$ is equal to the $G_n = G_m = 0$ component of the dielectric matrix $\epsilon(Q + G_n, Q + G_m; \omega)$. The last matrix is defined by means of the two-particle Green function of ‘mechanical’ excitons (see equation (18) of paper 1). The ground state of the ‘mechanical’ excitons in our case is transforming as $\Gamma_1 \otimes \Gamma_6 \otimes \Gamma_8 = \Gamma_3 \oplus \Gamma_4 \oplus \Gamma_5$. Thus, if local-field effects are neglected, we have the dielectric function (23), where $E_T = E_{\Gamma_3 \oplus \Gamma_4 \oplus \Gamma_5}$.

If local-field effects are taken into account the dielectric function $\epsilon(Q, \omega)$ can be obtained by means of the Green function of ‘mechanical’ excitons when the Elliott exchange interaction is taken into account. The Elliott exchange splits the Γ_5 excitons from the $\Gamma_3 \oplus \Gamma_4$ excitons. Therefore, E_T in equation (23) is given by $E_T = E_{\Gamma_3 \oplus \Gamma_4} + \Delta_T$, where the splitting Δ_T is proportional to the following expression (Rossler and Trebin 1981):

$$\Delta_T \sim \sum_{G_n} \frac{4\pi e^2}{|G_n|} \left| \langle R | \exp(-iG_n \cdot \hat{x}) | z \rangle \right|^2.$$

Here $|z\rangle$ and $|R\rangle$ denote the orbital parts of the zone-centre Bloch functions for the valence band and for the conduction band respectively. The longitudinal part of the exchange interaction splits the longitudinal exciton Γ_{5L} from the transverse excitons Γ_{5T} . Thus one can write $E_T = E_{\Gamma_{5T}} = E_{\Gamma_3 \oplus \Gamma_4} + \Delta_T$ and $E_L = E_{\Gamma_{5T}} + \Delta_{LT}$.

Let us turn to a detailed discussion of the contributions of the local-field effects to the Raman efficiency. Those corrections are determined by the frequency dependence of the matrix element $M_{LO,K}$ and by the changes in the group and ‘energy transport’ velocities.

Using equations (20) it is not difficult to see that the corrections to the matrix element $M_{LO,K}$ due to the local-field effects are determined by the frequency dependence of $S_\alpha^i(Q; \omega_\nu^{(0)}(Q))$. Taking into account equation (23), we find that $S_\alpha^{\Gamma_5} \sim (E_L^2 - E_T^2)^{1/2}$, so the local-field corrections to the matrix element $M_{LO,K}$ may become more important from one material to another with increasing ratio $\Delta_T/E_{\Gamma_3 \oplus \Gamma_4}$.

We have calculated the dispersion of the excitonic polaritons, the group and ‘energy transport’ velocities for copper halides. The excitonic polariton parameters used in the numerical calculations are as follows (Levy *et al* 1985):

CuCl	CuBr
$\Delta_{LT} = 0.0212 \text{ eV}$	$\Delta_{LT} = 0.0122 \text{ eV}$
$\Delta_T = 0.0690 \text{ eV}$	$\Delta_T = 0.0170 \text{ eV}$
$E_{\Gamma_3 \oplus \Gamma_4} = 3.200 \text{ eV}$	$E_{\Gamma_3 \oplus \Gamma_4} = 2.9627 \text{ eV}$
$M_{\text{exc}} = 2.5 m_0$	$M_{\text{exc}} = 0.93 m_0$
$\epsilon_b = 5.0$	$\epsilon_b = 5.4$

Since the ratio $\Delta_T/E_{\Gamma_3 \oplus \Gamma_4}$ in CuBr is a little less than in CuCl, the corrections due to the local-field effects to the group and ‘energy-transport’ velocities in CuCl should be

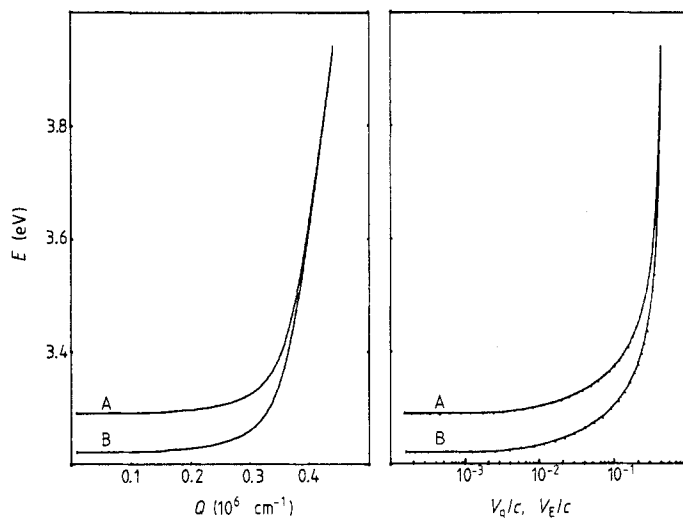


Figure 1. Dispersion of the upper excitonic polariton branch, group velocity and 'energy-transport' velocity (dotted line) in CuCl, calculated from a one-oscillator model: A, with; B, without local-field effects. The coincidence between the calculated values of V_g and V_E is in agreement with the experimental results (Levy *et al* 1985).

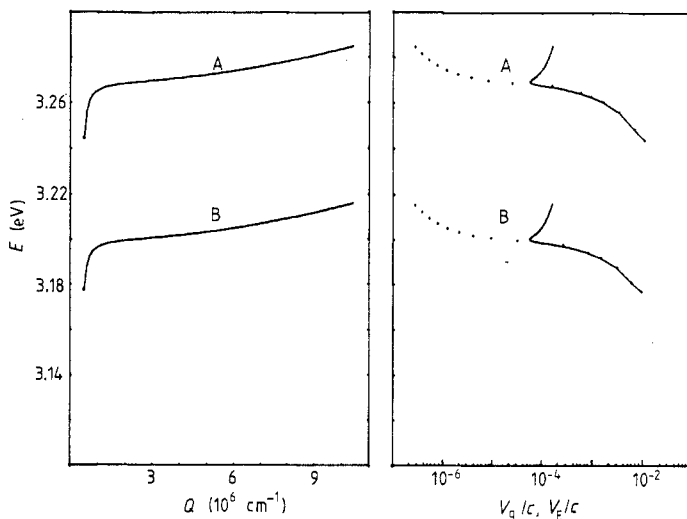


Figure 2. Dispersion of the lower excitonic polariton branch, group velocity and 'energy-transport' velocity (dotted line) in CuCl, calculated from a one-oscillator model: A, with; B, without local-field effects.

higher. The results for CuCl are presented in figures 1 and 2. We find: (i) the local-field effects modify significantly both the upper and the lower polariton dispersions: (ii) in the region $0.25 < Q < 0.35 \times 10^6 \text{ cm}^{-1}$ the changes in the group and 'energy transport' velocities due to the local-field effects are of the order of 20–30%.

In conclusion, some comments should be made concerning the problem of an electron and hole interacting with each other via a Coulomb potential, while each of them is interacting via the Frohlich term with the longitudinal optical phonons. From equation (12) one sees that the Coulomb potential is screened by the high-frequency dielectric constant, while the kernel (10b) is just the lowest-order phonon exchange contribution to the electron-hole interaction. Taking the approximation

$$K_{e-\omega} \left(\begin{array}{c} 1 \\ 2 \end{array} \begin{array}{c} 3 \\ 4 \end{array} \middle| \omega_{\nu}^{(0)}(Q) \pm \tilde{\Omega}_{\mu}(k) \right) \approx K^{(0)} \left(\begin{array}{c} 1 \\ 2 \end{array} \begin{array}{c} 3 \\ 4 \end{array} \middle| \omega_{\nu}^{(0)}(Q) \pm \tilde{\Omega}_{\mu}(k) \right)$$

and using equation (10b), we can rederive the results by Oswald and Egri (1983) for the effective electron-phonon interaction in polar semiconductors.

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