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Self-consistent approach to the theory of Wannier excitons in polar semiconductors

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Abstract. A theory of Wannier excitons in polar semiconductors that provides a simultaneous treatment of electron–phonon and exchange interactions, local-field effects and band degeneracy is formulated. The Bethe–Salpeter equation for the exciton Green function is reduced to an effective eigenvalue equation, taking into account the frequency dependence of the dielectric function that screens the electron–hole interactions. In comparison with the Coulomb attraction between the electrons and holes in a rigid lattice, screened by the optical constant, our effective potential contains an additional term due to the exchange of longitudinal polaritons. It is shown that in the case of $\Gamma_6 \times \Gamma_8$ excitons our treatment leads to a decrease of the electron–phonon vertex in comparison to the Frohlich one $\Gamma_q = i(2\pi e^2\omega_0/q^2\varepsilon^*)^{1/2}$ by the factor $[\![1 - \{[3 - (\varepsilon_\infty/\varepsilon_0)]/[1 - (\varepsilon_\infty/\varepsilon_0)]\}\Delta_{LT}/E_T]\!]^{1/2}$, where E_T and Δ_{LT} are the energy of the transverse exciton and its longitudinal–transverse splitting at the $Q = 0$ point, and $\omega_0 = \Omega_{TO}(\varepsilon_0/\varepsilon_\infty)^{1/2}$ according to the Lyddane–Sachs–Teller relation.

1. Introduction

The concept of Wannier excitons interacting with phonons is needed for interpretation of optical spectra of semiconductors. In polar semiconductors the interaction with optical phonons dominates as compared with the interaction with phonons of other types. In fact, most of present work on the exciton–LO phonon system in polar crystals is an extension of the Frohlich Hamiltonian (Frohlich *et al* 1950), derived for describing the interaction between electrons and phonons. According to the Frohlich model, excitons in polar crystals are considered as two excitons, e.g. an electron and a hole in polar semiconductors interact to a good approximation via a Coulomb interaction screened by the optical dielectric constant. The Frohlich model was studied by different mathematical techniques: perturbation theory (Wang and Matsuura 1974), variational calculations (Hattori 1976, Pollmann and Buttner 1977, Rossler and Trebin 1981), path-integral methods (Adamowski *et al* 1981), Green function methods (Shindo 1970, Mahanti and Varma 1972, Sac 1972, Klochihin 1980) and the equation-of-motion method (Oswald and Egri 1983). The Frohlich theory is certainly the correct picture as long as the electron and hole that constitute the exciton are well separated, and therefore interact individually with phonons. In real crystals, however, there exist well defined composite excitations (polaritons), formed by the coupling of excitons, phonons and photons (Koinov and Glinskii 1988, 1989, Glinskii and Koinov 1989). Unfortunately, these exact states are so complicated that they are almost of no use in calculating observable quantities. For this reason we decompose our electron–phonon–photon system into

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two subsystems, excitonic polaritons and phonons, with the following properties: the eigenstates of the first one are simple enough to be treated exactly by suitable mathematical techniques, while the interaction between two subsystems is small enough to be treated by perturbation theory.

In view of the fact that most papers are based on the Frohlich Hamiltonian, there may be a need to clarify the motivations for our approach. First, this approach is free from the assumption that electrons and holes couple separately to LO-phonon branch. Secondly, the many-body aspect of the bound state (electron–hole–LO phonon) can be dealt with more systematically, so all possible correlation and screening effects are included in principle. Thirdly, this approach provides a simultaneous treatment of electron–phonon and exchange interactions, local-field corrections and band degeneracy.

2. Model

In this paper we consider the non-relativistic bound-state problem in the system of interacting electrons, phonons and photons. In terms of the field theory we deal with a boson (photon) field $A_\alpha(z)$ interacting with a fermion (electron) field $\bar{\Psi}(y)$ (or $\Psi(x)$) and with a boson (phonon) field $u_\lambda(\zeta)$ at zero temperature. Here $y = \{\boldsymbol{r}, \sigma, t\}$, $x = \{\boldsymbol{r}', \sigma', t'\}$, $z = \{\boldsymbol{\rho}, t\}$ and $\zeta = \{\boldsymbol{R}_l, \kappa, t\}$ are composite variables: $\boldsymbol{r}, \boldsymbol{r}', \boldsymbol{\rho}$ are radius vectors; $\alpha, \beta, \lambda, \mu$ label the Cartesian coordinates; $\boldsymbol{R}_l$ is a radius vector of the lth unit cell, $l = 1, 2, \ldots, N$; $\kappa = 1, 2, \ldots, s$ characterises the atoms in the unit cell. There exist s atoms in a primitive cell, and the crystal consists of N cells.

The system under consideration has the following action

$$S = S_0^{(e)} + S_0^{(\omega)} + S_0^{(\Omega)} + S^{(e-\omega)} + S^{(\omega-\Omega)}. \tag{1}$$

Here the action $S_0^{(\omega)}$ describes the photon field, while the material system (semiconductor) is described by the action $S_0^{(e)}$ of non-interacting electrons in a periodic lattice potential and by the action $S_0^{(\Omega)}$ for bare phonons. The radiation and the matter interact via an electron–radiation interaction and phonon–radiation interaction, described by the actions $S^{(e-\omega)}$ and $S^{(\omega-\Omega)}$, respectively (Koinov and Glinskii 1988).

As was mentioned above, the 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 other types. For this reason, we can neglect the short-range part of the photon–phonon interaction in comparison to its long-range part. In this approximation one has for the action

$$S^{(\omega-\Omega)} = A_\alpha(z)\chi^{(0)}_{\alpha\lambda}(z|\zeta)u_\lambda(\zeta) \tag{2a}$$

where the long-range part of the photon–phonon vertex has the form

$$\chi^{(0)}_{\alpha\lambda}(z|\zeta) = \frac{1}{V_Q}\sum_{BZ}\int_{-\infty}^{+\infty}\frac{d\omega}{2\pi}\exp\{i[\boldsymbol{Q}\cdot(\boldsymbol{\rho}-\boldsymbol{R}_l) - \omega(t-t')]\}\left(\frac{-i\omega}{hc}\right)P^{\kappa}_{\alpha\lambda}(\boldsymbol{Q}). \tag{2b}$$

In the above equations the summation–integration convention, that the repeated variables are summed up or integrated over, is assumed. $P^{\kappa}_{\alpha\lambda}(\boldsymbol{Q})$ are phenomenological parameters, determining the polarisation of the crystal at the point ρ due to the atomic displacements $u^{l\kappa}_\lambda$.

The fundamental point in our approach is that all quantities of interest can be expressed in terms of appropriate Green functions. If we investigate the optical properties of semiconductors in the range of the fundamental absorption spectra, the discussions naturally centre on the behaviour of the excitons, since the absorption spectra are intimately related to the imaginary part of the dielectric function. The latter is primarily connected with the exciton Green function. In fact, the dielectric function can be written in the following form (Koinov and Glinskii 1988)

$$\varepsilon_{\alpha\beta}(Q,\omega)=\delta_{\alpha\beta}-\frac{4\pi\hbar c^2}{\omega^2}[\Pi^{(1)}_{\alpha\beta}(\boldsymbol{Q},\omega)+\Pi^{(2)}_{\alpha\beta}(\boldsymbol{Q},\omega)] \tag{3}$$

where the proper self-energies of the photon $\Pi^{(1)}_{\alpha\beta}$ and $\Pi^{(2)}_{\alpha\beta}$ can be obtained from equations (43) of the above-mentioned paper by neglecting the short-range part of the photon–phonon vertex and by making the analytic continuations off the set points $i\omega_p$ along the imaginary axis into the appropriate half of the ω plane. The first term $\Pi^{(1)}_{\alpha\beta}$ describes the scattering of photons from phonons due to the long-range part of the photon–phonon interaction

$$\Pi^{(1)}_{\alpha\beta}(\boldsymbol{Q},\omega)=\frac{\omega^2}{M_0V_0\hbar c^2}\sum_{\lambda}\frac{Z_\alpha(\lambda,\boldsymbol{Q})Z^*_\beta(\lambda,\boldsymbol{Q})}{\omega^2-\Omega^2_\lambda(\boldsymbol{Q})} \tag{4a}$$

where $Z_\alpha(\lambda,\boldsymbol{Q})=P^\kappa_{\alpha\mu}(\boldsymbol{Q})e^\kappa_\mu(\lambda,\boldsymbol{Q})$. Here $\hbar\Omega_\lambda(\boldsymbol{Q})$ are the energies of bare phonons with wavevectors Q in the Brillouin zone and λ branch index, $e^\kappa_\mu(\lambda,\boldsymbol{Q})$ denotes the phonon eigenvectors, and M_0 and V_0 are the mass of the atoms in the unit cell and its volume.

The second term $\Pi^{(2)}_{\alpha\beta}(\boldsymbol{Q},\omega)$ represents the combined effect of electron–photon and photon–phonon interactions on the photon scattering processes

$$\Pi^{(2)}_{\alpha\beta}(\boldsymbol{Q},\omega)=\frac{1}{\hbar^2c^2V}\langle 2|\hat{\jmath}_\alpha(\boldsymbol{Q})|1\rangle K_\Omega\left(\begin{matrix}1 & 3\\ 2 & 4\end{matrix}\middle|\omega\right)\langle 3|\hat{\jmath}_\beta(-\boldsymbol{Q})|4\rangle \tag{4b}$$

where $\hat{\jmath}_\alpha(\boldsymbol{Q})$ is a single-particle current operator, and K_Ω is the exciton Green function. In the above equation we have used the basis $|1\rangle=|s_1,n_1,k^i_1\rangle$ by $\boldsymbol{k}\cdot\boldsymbol{p}$ perturbation theory (see Glinskii and Koinov 1987). $\boldsymbol{k}^i$ is a vector of the Brillouin zone, near to $\boldsymbol{k}^i_0$; $\boldsymbol{k}^i_0$ 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 $\boldsymbol{k}^i_0$; n is the index of the degenerate states at the point $\boldsymbol{k}^i_0$ that have the same transformation properties as the basis functions obeying the irreducible representation s.

3. Bethe–Salpeter equation for the exciton Green function

The exciton four-time-variables Green function

$$K_\Omega\left(\begin{matrix}1 & 3\\ 2 & 4\end{matrix}\middle|t_2-t_1;t_4-t_3;t_1-t_3\right)$$

has the following Fourier transform

$$K_\Omega\begin{pmatrix}1 & 3\\ 2 & 4\end{pmatrix}\Big| t_2-t_1; t_4-t_3; t_1-t_3\Big) = \int_{-\infty}^{+\infty}\frac{d\Omega}{2\pi}\frac{d\Omega'}{2\pi}\frac{d\omega}{2\pi}\exp\{i[\Omega(t_2-t_1)-\Omega'(t_4-t_3) - \omega(t_1-t_3)]\}K_\Omega\begin{pmatrix}1 & 3\\ 2 & 4\end{pmatrix}\Big|\Omega;\Omega';\omega\Big). \tag{5a}$$

The exciton Green function

$$K_\Omega\begin{pmatrix}1 & 3\\ 2 & 4\end{pmatrix}\Big|\omega\Big)$$

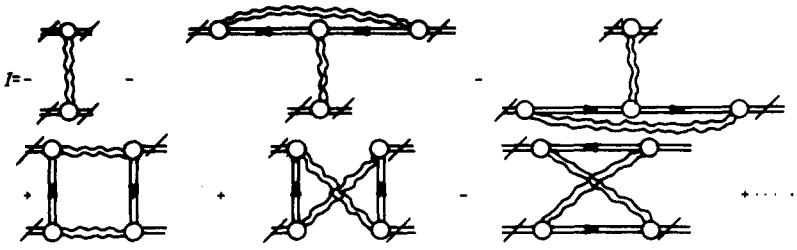
which is needed for determining the self-energy part $\Pi^{(2)}_{\alpha\beta}$, is defined as

$$K_\Omega\begin{pmatrix}1 & 3\\ 2 & 4\end{pmatrix}\Big|\omega\Big) = \int_{-\infty}^{+\infty}\frac{d\Omega}{2\pi}\frac{d\Omega'}{2\pi}K_\Omega\begin{pmatrix}1 & 3\\ 2 & 4\end{pmatrix}\Big|\Omega;\Omega';\omega\Big). \tag{5b}$$

From 12(a) and (42) of Koinov and Glinskii (1988) one can write the following Bethe–Salpeter equation for the exciton Green function

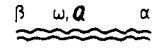
$$\int_{-\infty}^{+\infty}\frac{d\Omega'}{2\pi}\left[K^{(0)-1}\begin{pmatrix}1 & 3\\ 2 & 4\end{pmatrix}\Big|\Omega;\Omega';\omega\Big) - I\begin{pmatrix}1 & 3\\ 2 & 4\end{pmatrix}\Big|\Omega,\Omega';\omega\Big) - I_E\begin{pmatrix}1 & 3\\ 2 & 4\end{pmatrix}\Big|\omega\Big)\right] \times K_\Omega\begin{pmatrix}3 & 5\\ 4 & 6\end{pmatrix}\Big|\Omega';\Omega'';\omega\Big) = \delta(\Omega-\Omega'')\delta_{15}\delta_{26} \tag{6}$$

where $K^{(0)-1}$ is the inverse free two-particle propagator and I_E is the Elliott exchange interaction. In (6) the following abbreviation has been introduced: $\delta_{12} = \delta_{s_1s_2}\delta_{n_1n_2}\delta_{i_1i_2}\delta_{k_1^{i_1}k_2^{i_2}}$. The kernel I represents the sum of all connected diagrams with two lines entering and two leaving such that no part can be disconnected by cutting a pair of electron–hole lines:

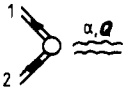


(7a)

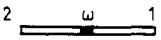
The rules making up the contribution of the diagrams to the algebraic form are



$D_{\alpha\beta}(\boldsymbol{Q},\omega)$ is the photon Green function



$(1/\hbar c V^{1/2})\langle 1|\hat{j}_\alpha(\boldsymbol{Q})|2\rangle$ is the electron–photon vertex



$G(2,1;\omega)$ is the one-particle electron (or hole) Green function.

(7b)

From (6) one can conclude that the Bethe–Salpeter equation is not closed for

$$K_\Omega\left(\begin{matrix}1 & 3\\ 2 & 4\end{matrix}\middle|\,\omega\right),$$

because the integration over Ω' on the left-hand side cannot be carried out immediately on account of the dynamic character of the kernel I.

Quite generally, if we restrict the range of frequencies ω in the Fourier transform (5*a*) to the neighbourhood of position $\pm\omega_n(\boldsymbol{Q})$, which are just the exciton eigenstates with a band index n and a wavevector $\boldsymbol{Q}$ within the Brillouin zone, we may write for the four-time-variables Green function

$$K_\Omega\left(\begin{matrix}1 & 3\\ 2 & 4\end{matrix}\middle|\,\Omega;\Omega';\omega\right)=\frac{F_{\bar{2}1}^{n\boldsymbol{Q}}(\Omega)F_{\bar{4}3}^{n\boldsymbol{Q}}(\Omega')^*}{\omega-\omega_n(\boldsymbol{Q})+\mathrm{i}0^+}-\frac{F_{\bar{1}2}^{n\boldsymbol{Q}}(\Omega)^*F_{\bar{3}4}^{n\boldsymbol{Q}}(\Omega')}{\omega+\omega_n(\boldsymbol{Q})+\mathrm{i}0^+}$$
$$+R\left(\begin{matrix}1 & 3\\ 2 & 4\end{matrix}\middle|\,\Omega;\Omega';\omega\right) \tag{8}$$

where $R(\omega)$ is a term regular at $\omega=\pm\omega_n(\boldsymbol{Q})$. In the above equation $|\bar{2}\rangle=\hat{T}|s_2,n_2,\boldsymbol{k}_2^{i_2}\rangle=|s_2^*,n_2,-\boldsymbol{k}_2^{i_2}\rangle$, where $\hat{T}$ is the time inversion operator. The functions $F_{\bar{2}1}^{n\boldsymbol{Q}}(\Omega)$ are assumed to be non-zero only in the case when the states $|\bar{2}\rangle$ and $|1\rangle$ stand for the hole and electron states, respectively.

Let us define the exciton wavefunction $F_{\bar{2}1}^{n\boldsymbol{Q}}$, which is the probability amplitude at one time to find an electron at state $|1\rangle$ and a hole at state $|\bar{2}\rangle$

$$F_{\bar{2}1}^{n\boldsymbol{Q}}=\delta_{\boldsymbol{Q},\boldsymbol{k}_1^{i_1}+\boldsymbol{k}_2^{i_2}}\int_{-\infty}^{+\infty}\frac{\mathrm{d}\Omega}{2\pi}F_{\bar{2}1}^{n\boldsymbol{Q}}(\Omega). \tag{9a}$$

From equations (4*b*), (5*b*) and (8) it follows that in the region of frequencies ω such that $\omega\simeq\omega_n(\boldsymbol{Q})$ the self-energy part $\Pi_{\alpha\beta}^{(2)}$ assumes the form

$$\Pi_{\alpha\beta}^{(2)}(\boldsymbol{Q},\omega)=\frac{1}{\hbar^2c^2V}\sum_n\left(\frac{j_\alpha^n(\boldsymbol{Q})j_\beta^n(\boldsymbol{Q})^*}{\omega-\omega_n(\boldsymbol{Q})+\mathrm{i}0}-\frac{j_\alpha^n(-\boldsymbol{Q})^*j_\beta^n(-\boldsymbol{Q})}{\omega+\omega_n(\boldsymbol{Q})+\mathrm{i}0}\right) \tag{9b}$$

where $j_\alpha^n(\boldsymbol{Q})$ is the exciton current

$$j_\alpha^n(\boldsymbol{Q})=\sum_{\bar{2},1}\langle\bar{2}|\hat{j}_\alpha(\boldsymbol{Q})|1\rangle F_{\bar{2}1}^{n\boldsymbol{Q}}. \tag{9c}$$

An exact equation for $F_{\bar{2}1}^{n\boldsymbol{Q}}(\Omega)$ can be derived from the Bethe–Salpeter equation (6) making the integration along the contour enclosing the point $\omega_n(\boldsymbol{Q})$ in the complex plane ω. The result is

$$F_{\bar{2}1}^{n\boldsymbol{Q}}(\Omega)=-\mathrm{i}G(1,3;\Omega+\omega_n)G(\bar{4},\bar{2};\Omega)\int_{-\infty}^{+\infty}\frac{\mathrm{d}\Omega'}{2\pi}\left[I\left(\begin{matrix}3 & 5\\ \bar{4} & \bar{6}\end{matrix}\middle|\,\Omega;\Omega';\omega_n\right)\right.$$
$$\left.+I_{\mathrm{E}}\left(\begin{matrix}3 & 5\\ \bar{4} & \bar{6}\end{matrix}\middle|\,\omega\right)\right]F_{\bar{6}5}^{n\boldsymbol{Q}}(\Omega'). \tag{10}$$

Our further aim, however, is to extract the dominant contributions to the kernel I of the exact equation (10), in order to reduce the Bethe–Salpeter equation to an effective eigenvalue equation to determine the exciton wavefunctions $F_{\bar{2}1}^{n\boldsymbol{Q}}$ and corresponding energies $\hbar\omega_n(\boldsymbol{Q})$.

4. Relevant approximations

In order to derive an effective eigenvalue equation, which determines the exciton energies $\hbar\omega_n(\boldsymbol{Q})$ and corresponding wavefunctions, some approximations have to be made. They are as follows.

(i) In the gauge we have used (the scalar potential is set equal to zero) one can separate the direct electron–hole interaction (the first diagram in equation (7*a*)) into an instantaneous (Coulomb) part and a retardation part. This can be done by writing the photon propagator as the sum of longitudinal and transverse parts. Since the retardation part is proportional to $(e^2/\hbar c)^2$, it can be neglected in comparison to the Coulomb part.

In crystals with a cubic point group the dielectric function $\varepsilon_{\alpha\beta}(\boldsymbol{Q}, \omega)$, defined by equations (3), (4*a*) and (9*b*), and the longitudinal part of the photon Green function have the forms

$$\varepsilon_{\alpha\beta}(\boldsymbol{Q}, \omega) = \delta_{\alpha\beta}\varepsilon(\boldsymbol{Q}, \omega) \tag{11a}$$

$$D_{\parallel}(\boldsymbol{Q}, \omega) = D_{\alpha\beta}(\boldsymbol{Q}, \omega)\boldsymbol{Q}_\alpha\boldsymbol{Q}_\beta/\boldsymbol{Q}^2 = \frac{4\pi\hbar c^2}{\omega^2}\varepsilon^{-1}(\boldsymbol{Q}, \omega). \tag{11b}$$

Let us denote by $\Omega_\nu(\boldsymbol{Q})$ the longitudinal polaritons (or longitudinal normal modes) in the crystal, which can be determined by the equation $\varepsilon(\boldsymbol{Q}, \Omega_\nu) = 0$. Then, from (11*b*) it follows that

$$D_{\parallel}(\boldsymbol{Q}, \omega) \underset{\omega\to\Omega_\nu}{\to} \frac{4\pi\hbar c^2}{\Omega_\nu^2\{[\partial\varepsilon(\boldsymbol{Q}, \omega)/\partial\omega]|_{\omega=\Omega_\nu}\}} \frac{1}{\omega_\nu - \Omega(\boldsymbol{Q}) + \mathrm{i}0^+}. \tag{11c}$$

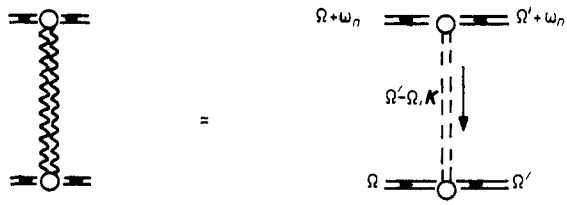
Thus, by making use of the Kramers–Kronig relation and from equation (11) we can write for the time-order inverse dielectric function

$$\varepsilon^{-1}(\boldsymbol{Q}, \omega) = \varepsilon_\infty^{-1}(\boldsymbol{Q}) - \sum_\nu \left(\left.\frac{\partial\varepsilon(\boldsymbol{Q}, \omega)}{\partial\omega}\right|_{\omega=\Omega_\nu}\right)^{-1} \times \left(\frac{1}{\Omega_\nu(\boldsymbol{Q}) - \omega - \mathrm{i}0^+} + \frac{1}{\Omega_\nu(\boldsymbol{Q}) + \omega - \mathrm{i}0^+}\right) \tag{12}$$

where we have introduced the high-frequency dielectric function

$$\varepsilon_\infty^{-1}(\boldsymbol{Q}) = \lim_{\omega\to+\infty} \varepsilon^{-1}(\boldsymbol{Q}, \omega).$$

After the above approximations, the first term in equation (7*a*) assumes the form



(13*a*)

and the rules making up the contribution of the above diagram are

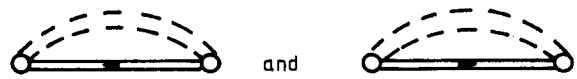
$$\langle 1|\exp(-i\boldsymbol{K}\cdot\hat{\boldsymbol{x}})|3\rangle$$

$$\langle\bar{4}|\exp(i\boldsymbol{K}\cdot\hat{\boldsymbol{x}})|\bar{2}\rangle \tag{13b}$$

$$\frac{4\pi e^2}{VK^2}\left[\varepsilon_\infty^{-1}(\boldsymbol{K})-\sum_\nu\left(\frac{\partial\varepsilon(\boldsymbol{K},\omega)}{\partial\omega}\bigg|_{\omega=\Omega_\nu}\right)^{-1}\right.$$

$$\left.\times\left(\frac{1}{\Omega_\nu(\boldsymbol{K})-\omega-i0^+}+\frac{1}{\Omega_\nu(\boldsymbol{K})+\omega-i0^+}\right)\right].$$

(ii) The single-particle Green functions entering (10) satisfy Dyson's equations with the mass operators written as the sum of a Hartree part and a screened Fock part (Koinov and Glinskii 1988, 1989, Glinskii and Koinov 1989). By extracting from the mass operators only the screened Fock terms



one can write the following Dyson's equations

$$G^{-1}(3,1;\Omega)=G^{(0)-1}(3,1;\Omega)-\sum_{\mathrm{e}}(3,1;\Omega) \tag{14a}$$

$$G^{-1}(\bar{2},\bar{4};\Omega)=G^{(0)-1}(\bar{2},\bar{4};\Omega)-\sum_{\mathrm{h}}(\bar{2},\bar{4};\Omega). \tag{14b}$$

Let us assume that the single-particle electron $\phi^{s_1}_{nl_1}(\boldsymbol{k}^{i_1}_1)$ and hole $\phi^{s_2}_{n_2l_2}(\boldsymbol{k}^{i_2}_2)$ wavefunctions are known by a previous solution of a band eigenvalue problem (in a Hartree approximation) (Glinskii and Koinov 1987)

$$H^{s_1}_{n_1n_1'}(\boldsymbol{k}^{i_1}_1)\phi^{s_1}_{n_1'l_1'}(\boldsymbol{k}^{i_1}_1)=E_{l_1}(\boldsymbol{k}^{i_1}_1)\phi^{s_1}_{n_1l_1}(\boldsymbol{k}^{i_1}_1) \tag{15a}$$

$$H^{s_2}_{n_2n_2}(\boldsymbol{k}^{i_2}_2)\phi^{s_2}_{n_2'l_2}(\boldsymbol{k}^{i_2}_2)=E_{l_2}(\boldsymbol{k}^{i_2}_2)\phi^{s_2}_{n_2l_2}(\boldsymbol{k}^{i_2}_2). \tag{15b}$$

Here $H^{s_1}_{n_1n_1'}(\boldsymbol{k}^{i_1}_1)$ and $H^{s_2}_{n_2n_2}(\boldsymbol{k}^{i_2}_2)$ are the corresponding Luttinger matrices. If we can solve equations (15), then the single-particle Green functions in the quasi-particle approximation are

$$G^{(0)}(1,3;\Omega)=\Delta_{13}\sum_{l_1}\frac{\phi^{s_1}_{n_1l_1}(\boldsymbol{k}^{i_1}_1)\phi^{s_1}_{l_1n_3}(\boldsymbol{k}^{i_1}_1)^*}{\Omega-(1/\hbar)E_{l_1}(\boldsymbol{k}^{i_1}_1)+i0^+} \tag{16a}$$

$$G^{(0)}(\bar{4},\bar{2};\Omega)=\Delta_{\bar{4}\bar{2}}\sum_{l_2}\frac{\phi^{s_2}_{n_4l_2}(\boldsymbol{k}^{i_2}_2)\phi^{s_2}_{l_2n_2}(\boldsymbol{k}^{i_2}_2)^*}{\Omega-(1/\hbar)E_{l_2}(\boldsymbol{k}^{i_2}_2)-i0^+} \tag{16b}$$

where the following abbreviation has been introduced: $\Delta_{13}=\delta_{s_1s_3}\delta_{i_1i_3}\delta_{k^{i_1}_1k^{i_3}_3}$.

The corresponding electron and hole mass operators, according to the above-mentioned approximations, have the forms

$$\sum_{\mathrm{e}}(3,1;\Omega)=\Delta_{13}\frac{4\pi e^2}{V}\sum_{\boldsymbol{K}}\frac{1}{\boldsymbol{K}^2}\sum_\nu\left(\frac{\partial\varepsilon(\boldsymbol{K},\omega)}{\partial\omega}\bigg|_{\omega=\Omega_\nu}\right)^{-1}$$

$$\times\sum_{l_1}\frac{\phi^{s_1}_{n_3l_1}(\boldsymbol{k}^{i_1}_1+\boldsymbol{K})\phi^{s_1}_{l_1n_1}(\boldsymbol{k}^{i_1}_1+\boldsymbol{K})^*}{\Omega-\Omega_\nu(\boldsymbol{K})-(1/\hbar)E_{l_1}(\boldsymbol{k}^{i_1}_1+\boldsymbol{K})+i0^+} \tag{17a}$$

$$\sum_{\mathrm{h}}(\bar{2},\bar{4};\Omega)=\Delta_{\bar{2}\bar{4}}\frac{4\pi e^2}{V}\sum_{\boldsymbol{K}}\frac{1}{\boldsymbol{K}^2}\sum_{\nu}\left(\frac{\partial\varepsilon(\boldsymbol{K},\omega)}{\partial\omega}\bigg|_{\omega=\Omega_\nu}\right)^{-1}$$
$$\times\sum_{l_2}\frac{\phi^{s_2}_{n_2^{\prime}l_2}(\boldsymbol{k}_2^{i_2}-\boldsymbol{K})\phi^{s_2}_{l_2^{\prime}n_4}(\boldsymbol{k}_2^{i_2}-\boldsymbol{K})^*}{\Omega+\Omega_\nu(\boldsymbol{K})-(1/\hbar)E_{l_2}(\boldsymbol{k}_2^{i_2}-\boldsymbol{K})-\mathrm{i}0^+}. \tag{17b}$$

In the conclusion of this section some comments should be made concerning the differences between our approach and others, which are based on the Frohlich theory. The main difference is that in our approach the effective interaction between electrons and holes is not only the interaction screened by the optical constant Coulomb attraction but the interaction due to the exchange of the longitudinal polaritons as well. In the Frohlich model the last interaction is due to the exchange of the phonon polaritons, which are formed by the coupling of photons with optical phonons. Thus, due to the retardation effects in the Frohlich theory the effective interaction between electrons and holes depends on the exciton binding energy. In our method this effective interaction depends not only on the binding energy but on the exciton wavefunctions as well, so one can say we have a self-consistent approach.

5. Effective eigenvalue equation

In the case when the retardation effects (or the effects due to the exchange of the longitudinal polaritons) are neglected, the solution of the Bethe–Salpeter equation (10) has the following form

$$F^{n\boldsymbol{Q}}_{\bar{2}\bar{1}}(\Omega)=\sum_{l_1l_2}\{\phi^{s_1}_{n_1l_1}(\boldsymbol{k}_1^{i_1})\phi^{s_2}_{l_2n_2}(\boldsymbol{k}_2^{i_2})^*R^{\{1,\bar{2}\},n\boldsymbol{Q}}_{l_1l_2}(\Omega)$$
$$\times[\Omega-\omega_n(\boldsymbol{Q})-(1/\hbar)E_{l_1}(\boldsymbol{k}_1^{i_1})+i0^+]^{-1}[\Omega-(1/\hbar)E_{l_2}(\boldsymbol{k}_2^{i_2})-\mathrm{i}0^+]^{-1}\}. \tag{18a}$$

Here we have used the abbreviation $\{1,\bar{2}\}=\{s_1,i_1,\boldsymbol{k}_1^{i_1};s_2,i_2,\boldsymbol{k}_2^{i_2}\}$ and the function $R(\Omega)$ is a regular function in the frequency plane with the following property

$$R^{\{1,\bar{2}\},n\boldsymbol{Q}}_{l_1l_2}[(1/\hbar)E_{l_1}(\boldsymbol{k}_1^{i_1})-\omega_n(\boldsymbol{Q})]=R^{\{1,\bar{2}\},n\boldsymbol{Q}}_{l_1l_2}[(1/\hbar)E_{l_2}(\boldsymbol{k}_2^{i_2})]. \tag{18b}$$

Further, our aim is to extract by using perturbation theory the contributions (due to the exchange of longitudinal polaritons) to the screened Coulomb attraction, in a manner such that equations (18) hold. If one takes into account the operators Σ_{e} and Σ_{h} and the first diagram in (7*a*), the following eigenvalue equation for the exciton wavefunction $F^{n\boldsymbol{Q}}_{\bar{2}\bar{1}}$ can be obtained

$$\sum_{3,\bar{4}}\Big\{\delta_{\bar{2}\bar{4}}\Delta_{13}H^{s_1}_{n_1n_3}(\boldsymbol{k}_1^{i_1})-\delta_{13}\Delta_{\bar{2}\bar{4}}H^{s_2}_{n_4n_2}(\boldsymbol{k}_2^{i_2})$$
$$+I_{\mathrm{C}}\begin{pmatrix}1&3\\\bar{2}&\bar{4}\end{pmatrix}+I_{\mathrm{E}}\begin{pmatrix}1&3\\\bar{2}&\bar{4}\end{pmatrix}+\sum_{\boldsymbol{K}}\sum_{\nu}$$
$$\times\Big[U_{\nu,\boldsymbol{K}}\begin{pmatrix}1&5\\\bar{2}&\bar{6}\end{pmatrix}K^{(0)}\left(\begin{matrix}5&7\\\bar{6}&\bar{8}\end{matrix}\Big|\omega_n(\boldsymbol{Q})-\Omega_\nu(\boldsymbol{K})\right)$$
$$\times U_{\nu,-\boldsymbol{K}}\begin{pmatrix}7&3\\\bar{8}&\bar{4}\end{pmatrix}\Big]\Big\}F^{n\boldsymbol{Q}}_{\bar{4}\bar{3}}=\hbar\omega_n(\boldsymbol{Q})F^{n\boldsymbol{Q}}_{\bar{2}\bar{1}}. \tag{19}$$

Here I_C and I_E are the screened Coulomb attraction and the Elliott exchange interaction. The leading term of I_C is diagonal in the band indices

$$I_C\begin{pmatrix}1 & 3\\ \bar{2} & \bar{4}\end{pmatrix} = -\frac{4\pi e^2}{V\varepsilon_\infty}\delta_{s_1s_3}\delta_{i_1i_3}\delta_{n_1n_3}\delta_{s_2s_4}\delta_{i_2i_4}\delta_{n_2n_4}$$
$$\times \delta_{k_1^{i_1}+k_2^{i_2},k_3^{i_3}+k_4^{i_4}}\frac{1}{|k_1^{i_1}-k_3^{i_3}|^2} + \Delta I_C + \Delta I_C^{k\cdot p}. \tag{20a}$$

The corrections ΔI_C and $\Delta I_C^{k\cdot p}$ are due to the short-range part of the Coulomb potential and to the $k\cdot p$ interaction with other bands (Bir and Pikus 1974, Glinskii and Koinov 1987). The Elliott exchange interaction has the form

$$I_E\begin{pmatrix}1 & 3\\ \bar{2} & \bar{4}\end{pmatrix} = \delta_{k_1^{i_1}+k_2^{i_2},k_3^{i_3}+k_4^{i_4}}\frac{4\pi e^2}{V}\sum_{G_n\neq 0}\frac{1}{|G_n|^2}$$
$$\times \langle 1|\exp(\mathrm{i}G_n\cdot\hat{x})|\bar{2}\rangle\langle\bar{4}|\exp(-\mathrm{i}G_n\cdot\hat{x})|3\rangle. \tag{20b}$$

In (19) the effective electron–longitudinal polariton vertex $U_{\nu,K}$ is defined as

$$U_{\nu,K}\begin{pmatrix}1 & 3\\ \bar{2} & \bar{4}\end{pmatrix} = \mathrm{i}\left(\frac{4\pi e^2}{VK^2}\right)^{1/2}\left(\left|\frac{\partial\varepsilon(K,\omega)}{\partial\omega}\right|_{\omega=\Omega_\nu}\right)^{-1/2}$$
$$\times[\delta_{\bar{2}\bar{4}}\langle 1|\exp(-\mathrm{i}K\cdot\hat{x})|3\rangle - \delta_{13}\langle\bar{4}|\exp(-\mathrm{i}K\cdot\hat{x})|\bar{2}\rangle]. \tag{21a}$$

The free two-particle propagator has the form (Glinskii and Koinov 1987)

$$K^{(0)}\left(\begin{matrix}1 & 3\\ \bar{2} & \bar{4}\end{matrix}\middle|\,\omega\right) = \Delta_{13}\Delta_{\bar{2}\bar{4}}\sum_{l_1l_2}[\![\phi_{n_1l_1}^{s_1}(k^{i_1})\phi_{l_1n_3}^{s_1}(k_1^{i_1})^*\,\phi_{n_4l_2}^{s_2}(k_2^{i_2})\phi_{l_2n_2}^{s_2}(k_2^{i_2})^*$$
$$\times\{\omega-(1/\hbar)[E_{l_1}(k_1^{i_1})-E_{l_2}(k_2^{i_2})]+\mathrm{i}0^+\}^{-1}]\!]. \tag{21b}$$

It is easy to see that the other diagrams in (7*a*) modify (19), and the new form can be obtained by replacing the last term in the left-hand side of (19) by the following term

$$\sum_K\sum_\nu\left[U_{\nu,K}\begin{pmatrix}1 & 5\\ \bar{2} & \bar{6}\end{pmatrix}K^{(0)}\left(\begin{matrix}5 & 7\\ \bar{6} & \bar{8}\end{matrix}\middle|\,\omega_n(Q)-\Omega_\nu(K)\right)U_{\nu,-K}\begin{pmatrix}7 & 3\\ \bar{8} & \bar{4}\end{pmatrix}\right]F_{\bar{4}3}^{nQ}$$
$$\to\sum_K\sum_\nu\left[U_{\nu,K}\begin{pmatrix}1 & 5\\ \bar{2} & \bar{6}\end{pmatrix}K_{e\text{-}\omega}\left(\begin{matrix}5 & 7\\ \bar{6} & \bar{8}\end{matrix}\middle|\,\omega_n(\Omega)-\Omega_\nu(K)\right)\right.$$
$$\left.\times U_{\nu,-K}\begin{pmatrix}7 & 3\\ \bar{8} & \bar{4}\end{pmatrix}\right]F_{\bar{4}3}^{nQ}. \tag{22a}$$

Where the two-particle propagator $K_{e\text{-}\omega}$ satisfies the Bethe–Salpeter equation

$$\left[(K^{(0)})^{-1}\left(\begin{matrix}1 & 3\\ 2 & 4\end{matrix}\middle|\,\omega\right) - I_C\begin{pmatrix}1 & 3\\ 2 & 4\end{pmatrix} - I_E\begin{pmatrix}1 & 3\\ 2 & 4\end{pmatrix}\right.$$
$$\left.\times\,\mathrm{I}_{\mathrm{exc}}\left(\begin{matrix}1 & 3\\ 2 & 4\end{matrix}\middle|\,\omega\right)\right]K_{e\text{-}\omega}\left(\begin{matrix}3 & 5\\ 4 & 6\end{matrix}\middle|\,\omega\right) = \delta_{15}\delta_{26} \tag{22b}$$

where I_{exc} is the long-range exchange interaction

$$I_{exc}\begin{pmatrix}1 & 3\\ 2 & 4\end{pmatrix}\omega\Big) = \frac{1}{\hbar^2 c^2 V}\sum_{Q}\langle 1|\hat{j}_\alpha(-\boldsymbol{Q})|2\rangle D^{(e)}_{\alpha\beta}(\boldsymbol{Q},\omega)\langle 4|\hat{j}_\beta(\boldsymbol{Q})|3\rangle \tag{22c}$$

and $D^{(e)}_{\alpha\beta}$ is the photon Green function, screened by the dielectric constant, which accounts for all other bands, not explicitly considered in the above equation

$$D^{(e)}_{\alpha\beta}(\boldsymbol{Q},\omega) = \frac{4\pi\hbar c^2}{\varepsilon_\infty\omega^2 - c^2\boldsymbol{Q}^2}\left(\delta_{\alpha\beta} - \frac{c^2\boldsymbol{Q}_\alpha\boldsymbol{Q}_\beta}{\varepsilon_\infty\omega^2}\right). \tag{22d}$$

The term given by (22*a*) plays an important role in the theory of the first-order resonant Raman scattering of light due to LO phonons (Koinov 1989).

6. Γ_6–Γ_8 excitons

Let us restrict ourselves to the case of T_d direct-band-gap polar semiconductors with two atoms per cell. We assume that the exciton state is formed by the Γ_6 conduction and Γ_8 valence band, neglecting the influence of the split-off band. In this case we have for the conduction band

$$E_{l_1}(\boldsymbol{k}) = E_c(\boldsymbol{K}) = E_G + (\hbar^2\boldsymbol{k}^2/2m_c)$$

and

$$E_{l_2}(\boldsymbol{k}) = E_l(\boldsymbol{k}) = -\{A\boldsymbol{k}^2 \pm [B\boldsymbol{k}^4 + (D^2 - 3B^2)(\boldsymbol{k}_x^2\boldsymbol{k}_y^2 + \boldsymbol{k}_x^2\boldsymbol{k}_z^2 + \boldsymbol{k}_y^2\boldsymbol{k}_z^2)]^{1/2}\}$$

for the light ($l = 1, 2$) and heavy ($l = 3, 4$) hole bands. The Luttinger matrices $H_{cc'}(\boldsymbol{k})$ and $H_{vv'}(\boldsymbol{k})$ ($c = \pm 1/2$, $v = \pm 3/2, \pm 1/2$), as well as the corresponding wavefunctions $\phi_{vl}(\boldsymbol{k})$, are given in Bir and Pikus (1974).

Let us now turn to a detailed discussion of equation (19). In the zero-order approximation the electrons and holes interact via an instantaneous Coulomb interaction screened by ε_∞. In this approximation the eight-fold degenerate ground state Γ_6–Γ_8 is decomposed by Elliott exchange into exciton states with Γ_3, Γ_4 and Γ_5 symmetry. If the Schrödinger equation (19) ($U_{v,\boldsymbol{K}} = 0$) can be solved analytically, one may obtain the dielectric function $\varepsilon(\boldsymbol{K},\omega)$ by using equations (3), (4*a*) and (9*b*).

In the case of a simple one-oscillator model, when the $\boldsymbol{K}$ dependence of the dielectric function is not taken into account, one may write for $\varepsilon(\omega)$

$$\varepsilon(\omega) = \varepsilon_\infty\left(\frac{\Omega_{LO}^2 - \omega^2}{\Omega_{TO}^2 - \omega^2} + \frac{E_L^2 - E_T^2}{E_T^2 - \hbar^2\omega^2}\right) \tag{23a}$$

where ε_∞ is the background dielectric constant, which accounts for all oscillators not explicity considered in the above model. Ω_{TO} is the transverse optical frequency, associated with the eigenvectors of the dynamic matrix for the bare phonon problem. In (23*a*) we have introduced the frequency

$$\Omega_{LO}^2 = \Omega_{TO}^2 + \frac{4\pi Z^2}{M_0 V_0 \varepsilon_\infty} \tag{23b}$$

where $Z = Z_\alpha(\text{TO}, \boldsymbol{Q})\boldsymbol{Q}_\alpha/|\boldsymbol{Q}|$ is the effective charge. E_T is the energy of the Γ_5 exciton at the point Γ of the Brillouin zone, and $E_L = E_T + \Delta_{LT}$. where Δ_{LT} is the longitudinal–transverse splitting.

For semiconductors $\Delta_{LT} \ll E_T$ (Levy *et al* 1985). Thus, the solutions of the equation $\varepsilon(\Omega_\nu) = 0$ are $\hbar\Omega_1 \simeq E_l$ and $\Omega_2 = \omega_0[1 - (\Delta_{LT}/E_T)]$, where we have introduced the LO-phonon frequency $\omega_0 = \Omega_{TO}(\varepsilon_0/\varepsilon_\infty)^{1/2}$ according to the Lyddane–Sachs–Teller relation ($\varepsilon_0 = \varepsilon(\omega = 0)$).

Further, in order to solve (19) self-consistently, we must calculate $U_{\nu,K}$ for $\nu = 1, 2$, according to (21*a*). It is easy to see that the exchange of longitudinal polaritons of type Ω_1 leads to an additional term to the screened Coulomb interaction that is of order (Δ_{LT}/E_G), and therefore we may conclude that this exchange gives a very small contribution to the exciton binding energy. In real crystals that exchange can be neglected.

From equations (21*a*) and (23*a*) the effective vertex $U_{\nu,K}$ is obtained in the following form

$$U_{2,K}\begin{pmatrix} 1 & 3 \\ \frac{}{2} & \frac{}{4} \end{pmatrix} = i\left(\frac{2\pi e^2 \tilde{\omega}_0}{V K^2 \varepsilon^*}\right)^{1/2} [\delta_{\bar{2}\bar{4}}\langle 1|\exp(-i\boldsymbol{K}\cdot\hat{\boldsymbol{x}})|3\rangle - \delta_{13}\langle\bar{4}|\exp(-i\boldsymbol{K}\cdot\hat{\boldsymbol{x}})|\bar{2}\rangle] \tag{24a}$$

where $(\varepsilon^*)^{-1} = \varepsilon_\infty^{-1} - \varepsilon_0^{-1}$ and

$$\tilde{\omega}_0 = \omega_0\left(1 - \frac{3 - (\varepsilon_\infty/\varepsilon_0)}{1 - (\varepsilon_\infty/\varepsilon_0)}\Delta_{LT}/E_T\right)^{1/2}. \tag{24b}$$

The effective vertex (24*a*) is identical to Frohlich's vertex, but with the renormalised frequency $\tilde{\omega}_0$ instead of ω_0, defined by the Lyddane–Sachs–Teller relation. For CuBr one has $\hbar\omega_0 = 20$ meV, $\varepsilon_0 = 6$, $\varepsilon_\infty = 5.4$, $\Delta_{LT} = 12.2$ meV, $E_T = 2.9644$ eV (Levy *et al* 1985). In this case we have $\Omega_2 \simeq \omega_0$, but

$$\left(1 - \frac{3 - (\varepsilon_\infty/\varepsilon_0)}{1 - (\varepsilon_\infty/\varepsilon_0)}\Delta_{LT}/E_T\right) = 0.91$$

and the Frohlich interaction is reduced by about 9%.

Finally, we end this paper by writing the explicit form of equation (19) for the $\Gamma_6 \times \Gamma_8$ exciton with wavevector $\boldsymbol{Q} = 0$:

$$\sum_{c',v'}\sum_{\boldsymbol{p}}\Bigg(E_c(\boldsymbol{k})\delta_{vv'}\delta_{\boldsymbol{kp}} + H_{v'v}(\boldsymbol{k})\delta_{cc'}\delta_{\boldsymbol{kp}} - \frac{4\pi e^2}{V\varepsilon_\infty}\frac{1}{|\boldsymbol{k}-\boldsymbol{p}|^2}\delta_{cc'}\delta_{vv'} + \frac{4\pi e^2}{V}\sum_{\boldsymbol{G}_n\neq 0}\frac{1}{|\boldsymbol{G}_n|^2}\langle c|\exp(-i\boldsymbol{G}_n\cdot\hat{\boldsymbol{x}})|\bar{v}\rangle\langle\bar{v}'|\exp(i\boldsymbol{G}_n\cdot\hat{\boldsymbol{x}})|c'\rangle\delta_{\boldsymbol{kp}} - I_{vv'}^{(\Sigma)}(\boldsymbol{k};E_B)\delta_{cc'}\delta_{\boldsymbol{kp}} + I_{vv'}^{(\Omega)}(\boldsymbol{k},\boldsymbol{p};E_B)\delta_{cc'}\Bigg)F_{c'v'}(\boldsymbol{p}) = E_B F_{cv}(\boldsymbol{k}) \tag{25a}$$

where the exciton binding energy $E_B = E_G - \hbar\omega_{1s}(\boldsymbol{Q} = 0)$.

The first four terms, which are the kinetic energies of the electron–hole pair, the screened Coulomb interaction between them and Elliott exchange, form the Schrödinger equation for the excitons in a rigid lattice (Rossler and Trebin 1981). In comparison to an exciton in a rigid lattice, equation (25*a*) contains additionally a self-energy correction $I^{(\Sigma)}$ to the kinetic energy as well as an effective electron–hole interaction $I^{(\Omega)}$ due to the exchange of longitudinal polaritons

$$I_{vv'}^{(\Sigma)}(\boldsymbol{k};E_B) = \frac{2\pi e^2\hbar\tilde{\omega}_0}{V\varepsilon^*}\sum_{\boldsymbol{q}}\frac{1}{\boldsymbol{q}^2}\sum_{l=1}^{4} \times\left(\frac{\phi_{v'l}(\boldsymbol{k})\phi_{lv}^*(\boldsymbol{k})}{\hbar\omega_0 + E_B + E_c(\boldsymbol{k}+\boldsymbol{q}) + E_l(\boldsymbol{k})} + \frac{\phi_{v'l}(\boldsymbol{q}-\boldsymbol{k})\phi_{lv}^*(\boldsymbol{q}-\boldsymbol{k})}{\hbar\omega_0 + E_B + E_c(\boldsymbol{k}) + E_l(\boldsymbol{q}-\boldsymbol{k})}\right) \tag{25b}$$

$$I_{vv'}^{(\Omega)}(\boldsymbol{k},\boldsymbol{p};E_B)=\frac{2\pi e^2\hbar\tilde{\omega}_0}{V\varepsilon^*}\frac{1}{|\boldsymbol{k}-\boldsymbol{p}|^2}\sum_{l=1}^{4}$$
$$\times\left(\frac{\phi_{v'l}(\boldsymbol{k})\phi_{lv}^*(\boldsymbol{k})}{\hbar\omega_0+E_B+E_c(\boldsymbol{p})+E_l(\boldsymbol{k})}+\frac{\phi_{v'l}(\boldsymbol{p})\phi_{lv}^*(\boldsymbol{p})}{\hbar\omega_0+E_B+E_c(\boldsymbol{k})+E_l(\boldsymbol{p})}\right). \quad (25c)$$

It is easy to prove that $I^{(\Sigma)}$ is definitely different from the sum of the corresponding free-polaron self-energy corrections, defined by equations (17).

A critical remark should be made concerning the accuracy of the numerical results, obtained eventually by solving equations (25). In order to solve (25) one needs the input bare-mass parameters. But, the parameter sets obtained, for example from magneto-optical data or from cyclotron resonance, must be interpreted as polaron-mass parameters. Thus, we need some additional procedure of converting the polaron-mass parameters into the bare-mass parameters. One may well ask whether this conversion does not affect the accuracy of the numerically calculated exciton binding energy. Despite the above remark, equations (25) were evaluated analytically in the simple case of parabolic bands neglecting the Elliott exchange (see Oswald and Egri 1983). As can be seen, the binding energy agrees well with other theories and experimental values for a number of direct-band-gap semiconductors.

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