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Non-equilibrium screening and plasmons in a coherently pumped semiconductor

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Abstract. The dielectric function of a coherently pumped semiconductor is calculated in the RPA to arbitrary order in the pump field. The theory starts with the longitudinal intraband polarization function. The diagonal and off-diagonal retarded one-particle Green's functions as well as the distribution functions are derived by means of the non-equilibrium Keldysh formalism. Exact analytical expressions follow within the collisionless regime and the rotating wave approximation. In this regime of the optical Stark effect the wavevector and frequency dependences of the resulting dielectric function are discussed for several values of the Rabi frequency and detuning between the pump frequency and energy gap. In the high-frequency limit the electron-hole pairs excited virtually exhibit the same Drude behaviour as real free carriers. For small frequencies and wavevectors there are remarkable discrepancies. The metallic properties of the virtual two-component plasma are depressed due to a finite transition energy. Moreover, contrary to what is found in the equilibrium case plasmons are strongly Landau damped already, even for vanishing wavevectors. Nevertheless, a screening function within the plasmon pole approximation is prepared.

Recent advances in ultrashort pulse spectroscopy allow the observation of electronic renormalization induced by intense coherent laser beams in semiconductors. Experiments under non-resonant excitation have revealed light-induced changes of excitonic energies and corresponding oscillator strengths [1–3]. In a time-resolved pump and probe measurement this optical Stark effect of the excitons is characterized by a saturating blue shift sometimes accompanied by a bleaching of the 1s light- and heavy-hole resonances with increasing pump intensity. The main effect is due to the direct dressing of electrons (e) and holes (h) excited virtually by pump photons. However, it is well known [4] that the properties of semiconductors optically excited above the gap are strongly influenced by the interactions with the real electrons and holes. This is also expected in the case of coherently driven virtual populations of e-h pairs present in the regime of the optical Stark effect [5–7]. Moreover, it is shown theoretically that such carriers screen applied static electric fields [8].

A first microscopic description of intense-laser-field effects has been formulated by Schmitt-Rink and Chemla [9] within the unscreened Hartree-Fock approximation. On the basis of the non-equilibrium Green's function theory and including many-body effects Schmitt-Rink *et al* [10] developed a detailed theory of the optical Stark effect. In this work we want to complete the description of the virtually excited electron-hole pairs by calculating explicitly the corresponding screening function as well as discussing the plasmons accompanying the vibrations of the two-component carrier gas.

For this purpose we start from the common expression for the longitudinal dielectric function in the random-phase approximation (RPA). Neglecting local-field effects one has in the non-equilibrium case

$$\epsilon(\mathbf{q}, \omega, T) = \epsilon_b [1 - v(\mathbf{q}) P_{\text{intra}}(\mathbf{q}, \omega, T)] \quad (1)$$

with the background dielectric constant ϵ_b and the spatially Fourier transformed Coulomb potential $v(\mathbf{q}) = 4\pi e^2 / \epsilon_b q^2$. In the low-frequency limit which is relevant for screening properties the wavevector- and frequency-dependent retarded polarization function $P(\mathbf{q}, \omega, T)$ can be replaced by the intraband one. It is Fourier transformed with respect to the relative time $t = t_1 - t_2$ and depends explicitly on the centre time $T = \frac{1}{2}(t_1 + t_2)$ due to its non-equilibrium character. In the screening limit effects of the e-h interaction can be neglected. Hence, the polarization function can be represented within the RPA as

$$P_{\text{intra}}(\mathbf{q}, t, T) = -2i\hbar \int \frac{d^3k}{(2\pi)^3} \sum_{ij} \left[G_{ij}^R(\mathbf{k} + \mathbf{q}, t_1, t_2) G_{ji}^<(\mathbf{k}, t_2, t_1) - \text{cc} \right] \quad (2)$$

by one-particle retarded Green's functions and distribution functions

$$\begin{aligned} G_{ij}^R(\mathbf{k}, t_1, t_2) &= \frac{1}{i\hbar} \theta(t_1 - t_2) \left\langle \left[a_{i\mathbf{k}}(t_1), a_{j\mathbf{k}}^\dagger(t_2) \right]_+ \right\rangle \\ G_{ij}^<(\mathbf{k}, t_1, t_2) &= -\frac{1}{i\hbar} \theta(t_1 - t_2) \left\langle a_{i\mathbf{k}}^\dagger(t_2) a_{j\mathbf{k}}(t_1) \right\rangle. \end{aligned} \quad (3)$$

Here, $a_{j\mathbf{k}}^\dagger(t)$ ($a_{j\mathbf{k}}(t)$) represents a creation (annihilation) operator of an electron in the Bloch state $|j\mathbf{k}\rangle$ with energy $E_j(\mathbf{k})$. In all practical cases we restrict ourselves to a two-band model with an empty conduction band ($j = 1$) and a filled valence band ($j = 2$) characterized by an energy gap E_g and positive band masses m_j , i.e. $E_j(\mathbf{k}) = (-1)^{j+1} (E_g/2 + \hbar^2 k^2/2m_j)$. The optical transition between the two bands near $\mathbf{k} = 0$ is assumed to be dipole-allowed and described by a constant dipole matrix element μ .

The one-particle functions (3) can be exactly calculated under some simplifying assumptions.

(i) The interaction with the coherent (quasi-) monochromatic pump field, $E - p \exp(-i\omega - pt) + \text{cc}$, is treated in the rotating wave approximation (RWA), keeping resonant terms only. The slow time variation of the field amplitude during the pulse action is omitted, applying the approximation of slowly varying non-equilibrium.

(ii) Only non-interacting electrons and holes are considered. The only self-energy in the problem is the radiative one. This restriction is not necessary. The electron-electron interaction can be simply taken into account within the (screened) Hartree-Fock approximation [5-7,10,11]. However, for pump frequencies in the energy gap far from the 1s exciton resonance these renormalization effects are small [11].

(iii) Relaxation processes are neglected assuming very short pulses. More strictly we consider a time regime with $1/\omega_p \approx 10^{-15}\text{s} \ll T \ll \tau_{\text{relaxation}} \approx 10^{-12}\text{s} \ll \tau_{\text{recombination}} \approx 10^{-9}\text{s}$.

Within the slowly varying non-equilibrium, the collisionless regime and the rotating frame the one-particle Green's functions can be obtained by applying the Keldysh

technique. One finds [4,7,10-12] for the retarded Green's functions

$$G_{ij}^R(\mathbf{k}, T + t/2, T - t/2) = (1/i\hbar)\Theta(t) \times \begin{pmatrix} e^{-\frac{1}{2}\omega_p t}(u_{\mathbf{k}}^2 e_{\omega_1} + v_{\mathbf{k}}^2 e_{\omega_2}) & e^{-i\omega_p T} u_{\mathbf{k}} v_{\mathbf{k}} (e_{\omega_1} - e_{\omega_2}) \\ e^{+\frac{1}{2}\omega_p t} u_{\mathbf{k}} v_{\mathbf{k}} (e_{\omega_1} - e_{\omega_2}) & e^{\frac{1}{2}\omega_p t} (v_{\mathbf{k}}^2 e_{\omega_1} + u_{\mathbf{k}}^2 e_{\omega_2}) \end{pmatrix} \quad (4)$$

(where $e_{\omega_1} \equiv e^{-i\omega_1(\mathbf{k})t}$ and $e_{\omega_2} \equiv e^{-i\omega_2(\mathbf{k})t}$) with the definitions

$$\hbar\omega_{1/2}(\mathbf{k}) = \frac{1}{2} \left[E_1(\mathbf{k}) + E_2(\mathbf{k}) \pm \hbar\sqrt{\Delta^2(\mathbf{k}) + \Omega_R^2} \right] \quad (5)$$

$$u_{\mathbf{k}}^2 = \frac{1}{2} \left(1 + \frac{\Delta(\mathbf{k})}{\omega_1(\mathbf{k}) - \omega_2(\mathbf{k})} \right) \quad v_{\mathbf{k}}^2 = \frac{1}{2} \left(1 - \frac{\Delta(\mathbf{k})}{\omega_1(\mathbf{k}) - \omega_2(\mathbf{k})} \right)$$

the resonance detuning

$$\hbar\Delta(\mathbf{k}) = E_1(\mathbf{k}) - E_2(\mathbf{k}) - \hbar\omega_p \quad (6)$$

and the Rabi frequency

$$\hbar\Omega_R = 2\mu E_p \quad (7)$$

which, in general, can depend on the centre time T via the slow variation of the pump pulse amplitude.

Taking into account the renormalization effects, the generalization is straightforward. The one-particle energies $E_j(\mathbf{k})$ ($j = 1, 2$) and the Rabi frequency Ω_R have to be replaced by the renormalized ones [5-7,10,11]. Results for the distribution functions $G_{ij}^<(\mathbf{k}, T + \frac{1}{2}t, T - \frac{1}{2}t)$ follow in the same manner. In expression (4) $\Theta(t)$ has to be replaced by -1 and $f_j(\mathbf{k})e^{-i\omega_j(\mathbf{k})t}$ ($j = 1, 2$) is used instead of $e^{-i\omega_j(\mathbf{k})t}$, where the electron distribution functions $f_j(\mathbf{k})$ in the conduction ($j = 1$) or valence ($j = 2$) band are the diagonal elements of the reduced density matrix before or after the short pump pulse excitation (i.e. in the equilibrium or quasiequilibrium) and if $\Delta(\mathbf{k}) > 0$. Otherwise, i.e. if $\Delta(\mathbf{k}) < 0$ the band characters of $f_1(\mathbf{k})$ and $f_2(\mathbf{k})$ have to be interchanged. We mention that the resulting reduced density matrix elements are $n_{ij}(\mathbf{k}, T) = -i\hbar G_{ij}^<(\mathbf{k}, 0, T)$ or, more strictly,

$$n_{ij}(\mathbf{k}) = f_j(\mathbf{k})\delta_{ij} + v_{\mathbf{k}} [f_1(\mathbf{k}) - f_2(\mathbf{k})] \begin{pmatrix} -v_{\mathbf{k}} & u_{\mathbf{k}} e^{-i\omega_p T} \\ u_{\mathbf{k}} e^{i\omega_p T} & v_{\mathbf{k}} \end{pmatrix} \quad (8)$$

fulfils the rules of invariance of trace and determinant.

Applying the one-particle functions G_{ij}^R from (4) and the corresponding ones for $G_{ij}^<$ the Fourier transform of the retarded intraband polarization function (2) takes the form ($\epsilon \rightarrow +0$)

$$P_{\text{intra}}(q, \omega, T) = \frac{4}{\hbar} \int \frac{d^3\mathbf{k}}{(2\pi)^3} \sum_{i,j=1}^2 \frac{f_j(\mathbf{k}) [\omega_i(\mathbf{k} + \mathbf{q}) - \omega_j(\mathbf{k})]}{(\omega + i\epsilon)^2 - (\omega_i(\mathbf{k} + \mathbf{q}) - \omega_j(\mathbf{k}))^2} \times [(u_{\mathbf{k}} u_{\mathbf{k}+\mathbf{q}} + v_{\mathbf{k}} v_{\mathbf{k}+\mathbf{q}})^2 \delta_{ij} + (u_{\mathbf{k}+\mathbf{q}} v_{\mathbf{k}} - u_{\mathbf{k}} v_{\mathbf{k}+\mathbf{q}})^2 (1 - \delta_{ij})]. \quad (9)$$

In the limit considered the only dependence on T is due to the Rabi frequency, i.e. the slow variation of the pump field amplitude E_p . For vanishing pump field, i.e. $u_k = \Theta(\Delta(k))$ and $v_k = \Theta(-\Delta(k))$, expression (9) changes into the intraband part of the well-known Ehrenreich-Cohen formula. In the following we focus our attention on the screening by the virtually excited electrons and holes. Therefore we assume $f_1(k) = 0$ and $f_2(k) = 1$ for pump frequencies below the gap. Using the symmetry properties one finds for the relative change of the dielectric function (1) due to the virtual carriers $\Delta\epsilon(q, \omega) = \epsilon(q, \omega, T)/\epsilon_b - 1$ the explicit representation

$$\Delta\epsilon(q, \omega) = -v(q) \frac{4}{\hbar} \int \frac{d^3k}{(2\pi)^3} (u_{k+q}v_k - u_kv_{k+q})^2 \times \frac{\omega_1(k+q) - \omega_2(k)}{(\omega + i\epsilon)^2 - (\omega_1(k+q) - \omega_2(k))^2}. \quad (10)$$

The resulting dielectric function obeys a generalized f -sum rule

$$\int_{-\infty}^{\infty} \frac{d\omega}{\pi} \omega \operatorname{Im} \Delta\epsilon(q, \omega) = \omega_{\text{pl}}^2 \quad (11)$$

$$\omega_{\text{pl}} = -v(q) \frac{2}{\hbar} \int \frac{d^3k}{(2\pi)^3} \left(1 - \frac{\Delta(k)}{\sqrt{\Delta^2(k) + \Omega_R^2}} \right) \sum_{j=1}^2 (-1)^j [E_j(k) - E_j(k+q)]$$

with a corresponding screened plasma frequency ω_{pl} determined by the number of virtual electron-hole pairs N ; more explicitly

$$\omega_{\text{pl}} = \frac{4\pi e^2}{\epsilon_b m^*} N \quad \frac{1}{m^*} = \frac{1}{m_1} + \frac{1}{m_2} \quad (12)$$

$$N = 2 \int \frac{d^3k}{(2\pi)^3} \frac{1}{2} \left(1 - \frac{\Delta(k)}{\sqrt{\Delta^2(k) + \Omega_R^2}} \right).$$

The squares of ω_{pl} and N are plotted versus the Rabi frequency, i.e. the square root of the pump intensity, in figure 1 for fixed detuning $\Delta(0)$. Excitonic units $R_{\text{exc}} = m^* e^4 / 2\epsilon_b^2 \hbar^2$ and $a_{\text{exc}} = \epsilon_b \hbar^2 / m^* e^2$ and GaAs parameters are used. Both quantities show a nearly quadratic dependence indicating that the low-intensity limit $\omega_{\text{pl}} = \Omega_R (R_{\text{exc}} / \hbar \Delta(0))^{1/4}$ is widely valid. As a consequence of the sum rule (11) the coherently driven two-component e-h gas shows a classical Drude behaviour, $\Delta\epsilon(q, \omega) = -\omega_{\text{pl}}^2 / (\omega + i\epsilon)^2$, in the high-frequency limit. In this limit virtually excited electrons and holes act in the same manner as real carriers.

On the other hand the low-frequency limit which governs the screening properties is much more complicated. Only for vanishing wavevectors is an analytical expression for the dielectric function $\Delta\epsilon(0, \omega)$ possible. From formula (10) it follows, in the non-resonant case, $\Delta(0) > 0$, that for the imaginary part

$$\operatorname{Im} \Delta\epsilon(0, \omega) = \Theta \left(|\omega| - [\Delta^2(0) + \Omega_R^2]^{1/2} \right) \times \frac{4}{3} \left(\frac{R_{\text{exc}}}{\hbar} \right)^{1/2} \frac{\Omega_R^2}{\omega^3} \left[\sqrt{\omega^2 - \Omega_R^2} - \Delta(0) \right]^{3/2} \frac{1}{\sqrt{\omega^2 - \Omega_R^2}}. \quad (13)$$

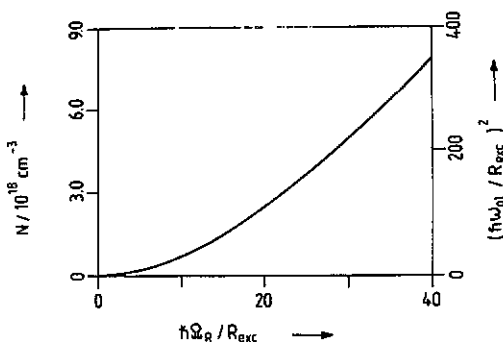


Figure 1. Density of virtual electron-hole pairs N and corresponding square of plasma frequency ω_{pl}^2 versus the Rabi frequency for fixed detuning $\hbar\Delta(0)/R_{exc} = 10$.

The step function in $\text{Im } \Delta\epsilon$ indicates the appearance of an energy gap in the spectrum of the virtual electron gas determined by detuning and pump intensity. As a consequence the virtual e-h gas exhibits insulating properties in the low-frequency limit in contrast to a real electron gas. The real part is given by the corresponding Cauchy relation. If $\omega = 0$ this integral can also be solved analytically. One finds for the relative change of the dielectric constant of the system due to the virtual e-h pairs

$$\Delta\epsilon(0,0) = \frac{8}{9} \left(\frac{R_{exc}}{\hbar\Delta(0)} \right)^{1/2} \left(\frac{\Omega_R}{\Delta(0)} \right)^2 s^{5/2} \frac{d^2}{ds^2} P_{-\frac{1}{2}}(s) \Big|_{s=1/\sqrt{1+(\Omega_R/\Delta(0))^2}} \quad (14)$$

with $P_{-\frac{1}{2}}(s)$ as a Legendre function. For large detuning or/and small pump intensities expression (14) can be simplified to

$$\Delta\epsilon(0,0) = \frac{1}{16} \left(\frac{R_{exc}}{\hbar\Delta(0)} \right)^{5/2} \left(\frac{\hbar\Omega_R}{R_{exc}} \right)^2$$

. This means that for realistic Rabi frequencies and for detunings that are not too small one expects relative changes of the material dielectric constant due to virtual carriers of the order of one per cent.

The wavevector dependence of the change in relative screening $\Delta\epsilon(q,0) = \epsilon(q,0,T)/\epsilon_b - 1$ is shown in figure 2 for a fixed detuning $\hbar\Delta(0) = 10 R_{exc}$. To make evident the effect of virtual carriers, large pump intensities $\hbar\Omega_R > R_{exc}$ are considered. The strongest effects are found for vanishing wavevectors. There is a strong increase with rising pump intensity. However, the screening effect of the coherently pumped electron-hole pairs is weak. Only if the detuning is drastically reduced is the screening increase remarkable. A reduction by a factor of two produces a gain of approximately one order of magnitude.

Our results for the relative change of the static screening are the same as obtained by Côté and Haug [7] using the generalized random phase approximation if the renormalization of the Rabi frequency is taken into account. The question is whether such small changes of the screening can be observed in experiment. In principle this should be possible studying the optical Stark effect [1-3]. The observed blue shift of the excitons is influenced by the actual screening. However, in the limit of large

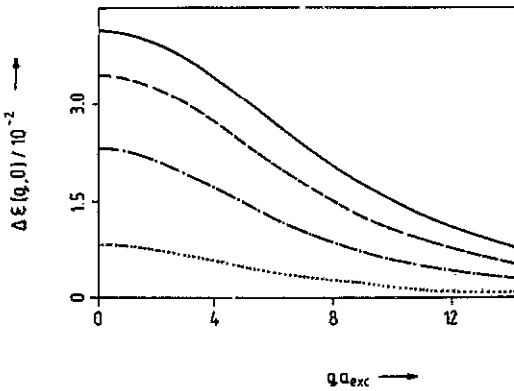


Figure 2. Relative change of the static dielectric function versus wavevector for different pump intensities and fixed detuning $\hbar\Delta(0) = 10 R_{exc}$; $\hbar\Omega_R/R_{exc} = 20$ (solid line), $= 15$ (dashed line), $= 10$ (dot-dashed line), and $= 5$ (dotted line).

detuning the Stark shift is essentially determined by changes of the kinetic energy of the excitons, i.e. the main contribution results from the renormalization of the single-particle energies, as can be shown by numerical calculations [11]. In the near-resonance case if the pump frequency approaches the nominal exciton position a stronger change in the screening properties due to virtual carriers is expected. However, in this limit more detailed studies of all contributions are necessary [13].

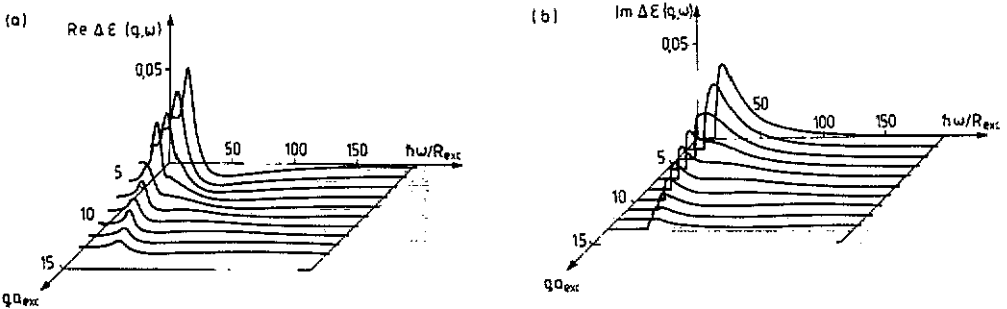


Figure 3. Real (a) and imaginary (b) part of the relative change of the dielectric function for $\hbar\Omega_R = \hbar\Delta(0) = 10 R_{exc}$.

For this time regime characterized above, the complete wavevector and frequency dependences of the relative change of the dielectric function (10) are plotted in figure 3 for fixed Rabi frequency $\hbar\Omega_R = 10 R_{exc}$ and detuning $\hbar\Delta(0) = 10 R_{exc}$. Figure 3 clearly indicates the damped oscillator behaviour of the screening system with oscillator frequency and oscillator damping which increase with rising wavevector. For frequencies that are not too small, the dielectric function in figure 3 can be fairly well represented by

$$\Delta\epsilon(q, \omega) = \frac{\omega_{pl}^2}{\Omega^2(q) - \omega^2 - i\omega\Gamma(q)} \tag{15}$$

with a relatively large oscillator frequency

$$\Omega(q) = \frac{\omega_{pl}}{\sqrt{\Delta\epsilon(q, 0)}} \tag{16}$$

in accordance with the small relative change of the dielectric constant. In the low-intensity limit and for small wavevectors this oscillator frequency is simply given by the detuning—more strictly by $\Omega(0) \approx 4\Delta(0)$. For arbitrary wavevectors $\Omega(q)$ is given fairly well by the zero of $\text{Re } \Delta\epsilon(q, \omega) = 0$. The damping of the oscillator $\Gamma(q)$ can be extracted from the peak position

$$\omega = \left\{ [\Omega^4(q) + \Gamma^4(q)/4]^{1/2} - \Gamma^2(q)/2 \right\}^{1/2}$$

in the imaginary part. One estimates $\Gamma(q) \approx (5/6)^{1/2} \Omega(q)$.

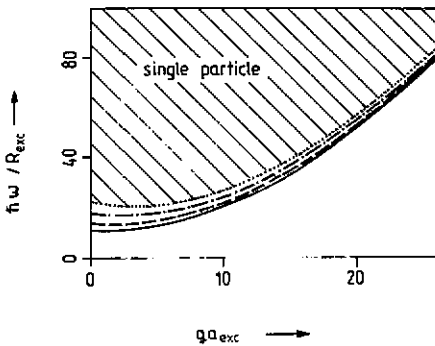


Figure 4. Pair excitation spectrum for the virtual e-h gas for a fixed detuning $\hbar\Delta(0) = 10 R_{exc}$; $\hbar\Omega_R / R_{exc} = 5$ (solid line), $= 10$ (dashed line), $= 15$ (dot-dashed line), and $= 20$ (dotted line).

The approximate representation (15) of the dielectric function, which includes the plasmon-pole approximation well known from the classical electron gas [14], makes one question whether plasma vibrations are possible in the coherently driven virtual e-h pair gas. The answer to this is given by the pair excitation spectrum as shown in figure 4. The single-particle spectrum defined by $\text{Im } \epsilon(q, \omega, T) \neq 0$ ($\text{Im } \Delta\epsilon(q, \omega) \neq 0$) is represented by the hatched region. In comparison to a real two-component neutral e-h gas one observes two remarkable changes. First, in the excitation spectrum of the virtual e-h gas there is a gap. The continuum of single-particle excitations starts for $\omega > [\Delta^2(0) + \Omega_R^2]^{(1/2)}$. Second, for high frequencies and small wavevectors one finds no single-particle excitations in the real e-h gas in contrast to what is found in the case considered. The last point is important for the observation of plasmons. The plasma oscillates spontaneously when $\text{Re } \epsilon(q, \omega, T) = 0$ ($\text{Re } \Delta\epsilon(q, \omega) = -1$) and $\text{Im } \epsilon(q, \omega, T) = 0$ ($\text{Im } \Delta\epsilon(q, \omega) = 0$) are simultaneously fulfilled. This can only happen outside the area of single-particle excitations. However, for the realistic parameters under consideration, i.e. $4\Delta(0) \approx \Omega_R$, the condition $\text{Re } \Delta\epsilon(q, \omega) = -1$ is always accompanied by $\text{Im } \Delta\epsilon(q, \omega) \neq 0$, i.e. by single-particle excitations. Hence, if plasmons appear they are strongly Landau damped in the sense of formula (15).

Real plasmons with frequencies $\Omega(q)$ ($\text{Re } \Delta\epsilon(q, \Omega(q)) = -1$) can only occur if these frequencies are smaller than the edge of the continuum of single-particle excitations, i.e. for $\Omega(0) < [\Delta^2(0) + \Omega_R^2]^{(1/2)}$. However, this relation can only be fulfilled in the limit of vanishing detuning. In this limit the strong renormalization of the band edges neglected here has to be taken into account.

In summary, we find that in a coherently pumped semiconductor far from equilibrium there are virtually excited electron-hole pairs which give rise to an additional screening. In the high-frequency limit the screening carrier gas exhibits a Drude behaviour as a real electron gas. However, in the low-frequency limit the properties of the virtual electron-hole gas are drastically changed. There is a gap for single-particle excitations and the collective excitations are strongly damped even for vanishing wavevectors.

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