Theory of the band-tail absorption saturation in polar semiconductors

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A theory is presented which allows one to calculate the absorption and its saturation in the band-tail region of polar semiconductors for elevated temperatures. The influence of the LO phonons and of the electron-hole plasma on the dynamically broadened band tail is considered. The problem is decomposed into the calculation of the broadening and the shift of the electron-hole pair states. Both the broadening and the shift are temperature and density dependent, although the shift of the 1*s*-exciton line depends mainly on the temperature alone. The calculation of the absorption saturation includes a self-consistent determination of the plasma density in terms of the intensity of the exciting laser light. The saturation of the absorption sets in after the light intensity exceeds a critical value which increases with decreasing frequency of the exciting laser light.

I. INTRODUCTION AND MODEL

The absorption saturation plays an important role in the nonlinear optical switching devices, which operate in the band-tail region.[1,2] In pure semiconductors the tail of the linear absorption below the band gap is known to vary exponentially with the frequency due to the influence of longitudinal optical (LO) phonons on the optically excited electron-hole pair. The exponential decrease of the linear absorption is called Urbach's rule[3] and is found in a wide class of semiconductors. The exponential frequency variation at high temperatures can arise from the influence of the LO phonon on the relative motion and on the center-of-mass motion of an electron-hole pair. In the first case the ionization of an exciton in the fluctuating electric field of the phonons is the dominant process.[4] In the second case the localization or self-trapping of a tightly bound exciton in the deformable lattice[5] is causing the absorption below the band gap.

In a previous investigation,[6] it has been shown how the application of a cluster expansion allows us to treat, at least in principle, the intraband and interband transitions of the exciton caused by the LO phonons on equal footing.

Here we will extend this treatment and calculate the nonlinear absorption for a laser beam with arbitrary intensity. It is known experimentally[2] that the band-tail absorption is difficult to saturate, but no detailed theoretical model existed so far of this saturation process, which is often of crucial importance for optical semiconductor switching devices. Because these switching devices are mainly used without cooling at room temperature, we will make high-temperature approximations wherever necessary to simplify the treatment.

We start from a two-band model with one electron band and one hole band both with parabolic dispersion. We include in our treatment the Coulomb interaction, the interaction with LO phonons, and the coherent pump field $E(R,t)$ $\hbar=1$ is used throughout this text.

The Hamiltonian H is given by

$$\begin{aligned} H=&\sum_k (E_g+\varepsilon_k^e)c_k^\dagger c_k+\sum_k \varepsilon_k^h d_k^\dagger d_k+\sum_q \omega_L b_q^\dagger b_q \\ &+\sum_{k,k_1,q} V_q(\tfrac{1}{2}c_{k+q}^\dagger c_{k_1-q}^\dagger c_{k_1}c_k+\tfrac{1}{2}d_{k+q}^\dagger d_{k_1-q}^\dagger d_{k_1}d_k-c_{k+q}^\dagger d_{k_1-q}^\dagger d_{k_1}c_k) \\ &+\sum_{k,q} M_q(b_q+b_{-q}^\dagger)(c_{k+q}^\dagger c_k-d_{k+q}^\dagger d_k)-\sum_{k,p} g_k[E_p(t)c_k^\dagger d_{-k+p}^\dagger+E_p^*(t)d_{-k+p}c_k]\ . \end{aligned} \tag{1.1}$$

c_k and d_k are the annihilation operators of an electron and a hole with momentum k, respectively, b_k is the annihilation operator of a LO phonon. The kinetic energies of the electrons (e) and the holes (h) are

$$\varepsilon_k^e=\frac{k^2}{2m_e} \quad \text{and} \quad \varepsilon_k^h=\frac{k^2}{2m_h}\ , \tag{1.2a}$$

m_e and m_h are their effective masses. E_g is the direct energy gap. The bare Coulomb potential is

$$V_q=\frac{4\pi e^2}{\epsilon_\infty q^2}\ . \tag{1.2b}$$

The matrix element for the Fröhlich coupling is

$$M_q^2=\frac{2\pi e^2\omega_L}{\epsilon^* q^2}\ , \tag{1.2c}$$

with $1/\epsilon^* = 1/\epsilon_\infty - 1/\epsilon_0$ and ω_L is the LO-phonon energy. ϵ_0 and ϵ_∞ are the low- and high-frequency dielectric constants. The electric field $E(R,t)$ Fourier transformed into momentum space is denoted by $E_p(t)$. The dipole matrix element between the valence and the conduction band is obtained by the $\mathbf{k}\cdot\mathbf{p}$ perturbation theory as

$$g_k \simeq \frac{g}{1+\frac{k^2}{2m_e}E_g} , \tag{1.2d}$$

where $g^2 \simeq Hn_2\Delta_{LT}/4\pi$ and Δ_{LT} is the longitudinal transversal splitting of the lowest exciton. The field $E(R,t)$ is a solution of the wave equation

$$\left[\Delta_R - \frac{n^2}{c^2}\frac{\partial^2}{\partial t^2}\right] E(R,t) = -\frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}\sum_p e^{-ip\cdot R}\sum_k g_k(\bar{S}^\dagger_{k,-k+p} + \text{H.c.}) . \tag{1.3}$$

Here, n is the background refractive index and c is the velocity of light. The macroscopic polarization function is determined by the macroscopic electron-hole pair amplitude $\bar{S}^\dagger_{k,-k+p}$, which is an appropriate average of the electron-hole pair operator

$$S^\dagger_{k,-k+p} = c^\dagger_k d^\dagger_{-k+p} . \tag{1.4}$$

The macroscopic electron-hole pair amplitude will be called pair amplitude. If spatial dispersion has to be included, the finite size of the bulk can be taken into account by using Pekar's boundary condition for the polarization.

The excited semiconductor is assumed to be in a quasiequilibrium state in which a thermal electron-hole plasma exists whose density is given by the external field $E(R,t)$. Therefore the pair amplitude is the thermal average of the electron-hole pair operator.

Our first aim is to calculate the broadening of the pair amplitude. It is assumed that the broadening is mainly due to the coupling to LO phonons and plasmons. For this purpose we use the cluster expansion method,[7] which reproduces in first order the exact result for a structureless, localized particle interacting with phonons.[8] Applying this method to the polaron problem, i.e., one electron interacting with the LO phonons, gives reliable results for the retarded electron Green's function $G^r(k,t) = -i\Theta(t)\langle c_k(t)c^\dagger_k(0) + c^\dagger_k(0)c_k(t)\rangle_T$.[7,9] $\langle\cdots\rangle_T$ denotes the thermal average. Due to the conservation of the total momentum of the system, the calculation is simplified considerably.

Generalizing this approach to our many-particle system, some simplifications have to be made in order to get a problem, which can be solved. First, some approximations are made in our equation for the pair amplitude which can be tackled with the cluster expansion method. The resulting equation for the pair amplitude $\bar{S}_{k,-k+p}(t)$ contains as an inhomogeneity the driving field $E(R,t)$. The homogeneous part of this equation is an effective Schrödinger equation for one electron-hole pair interacting with longitudinal excitations. These excitations are the diagonalized LO-phonon–plasmon modes. The approximations are valid if the temperature is sufficiently high.

The solution of this equation is complicated by the fact that the relative electron-hole momentum is not conserved. The disturbance of the center-of-mass motion of the electron-hole pair can be treated in the same way as for the polaron problem, but for the relative motion the straightforward cluster expansion is not tractable. Therefore, an effective coupling for each internal state of the electron-hole pair is introduced. These internal states are eigenstates of an effective Hamiltonian for the relative motion which includes the static influence of the interaction with the longitudinal excitations.

We illustrate the simplifications made in deriving the equation for the pair amplitude for the simple model of an electron gas interacting with phonons. The calculation of the retarded self-energy $\Sigma^r(k,\omega)$ gives within the Bohm-Pines random-phase approximation (RPA) and the screened potential approximation[10]

$$\Sigma^r(k,\omega) = -\frac{1}{\beta}\sum_{q,z_B} \mathcal{V}_s(q,z_B)\mathcal{G}^0(k-q,\omega+i\delta-z_B) . \tag{1.5}$$

$\mathcal{V}_s(k,z_B)$ is the screened potential and $\mathcal{G}^0(k,z_F)$ is the bare electron Green's function. Both are Matsubara functions. Here $z_B = i2n\pi/\beta$ is a complex boson frequency, and $z_F = i(2n+1)\pi/\beta$ is a complex fermion frequency. The screened potential is given within the plasmon-pole approximation[11] by

$$\mathcal{V}_s(q,\omega) \simeq V_q + \sum_j (M'_{jq})^2 \frac{2\omega'_{jq}}{\omega^2 - (\omega'_{jq})^2} . \tag{1.6}$$

ω'_{jq} are the diagonalized phonon-plasmon modes and M'_{jq} are the corresponding matrix elements. Their explicit values are of no relevance at this point. The Matsubara sum in the above equation can then be evaluated explicitly and yields

$$\Sigma^r(k,\omega) = -\sum_q V_q n(\varepsilon^e_{k+q}) + \sum_{j,q}(M'_{jq})^2\left[\frac{N(\omega'_{jq}) + n(\varepsilon^e_{k+q})}{\omega+i\delta-\varepsilon^e_{k+q}+\omega'_{jq}} + \frac{1+N(\omega'_{jq})-n(\varepsilon^e_{k+q})}{\omega+i\delta-\varepsilon^e_{k+q}-\omega'_{jq}}\right] . \tag{1.7}$$

Here, $N(\omega)$ is the Bose distribution and $n(\varepsilon)$ is the Fermi function.

One possible generalization of the cluster expansion approach would result in the following approximation for the retarded Green's function:

$$G^r(k,t) = -i\Theta(t)\exp\left[-i\left[\varepsilon^e_k - \sum_q V_q n(\varepsilon^e_{k+q})\right]t + F_k(t)\right] , \tag{1.8a}$$

The cumulant F_k is given by

$$F_k(t)=\sum_{j,q}[M'_{jq}(q)]^2\left[[N(\omega'_{jq})+n(\varepsilon^e_{k+q})]\left[\frac{it}{\varepsilon^e_{k+q}-\varepsilon^e_k-\omega'_{jq}}-\frac{1-e^{-i(\varepsilon^e_{k+q}-\varepsilon^e_k-\omega'_{jq})t}}{(\varepsilon^e_{k+q}-\varepsilon^e_k-\omega'_{jq})^2}\right]\right.$$
$$\left.+[1+N(\omega'_{jq})-n(\varepsilon^e_{k+q})]\left[\frac{it}{\varepsilon^e_{k+q}-\varepsilon^e_k+\omega'_{jq}}-\frac{1-e^{-i(\varepsilon^e_{k+q}-\varepsilon^e_k+\omega'_{jq})t}}{(\varepsilon^e_{k+q}-\varepsilon^e_k+\omega'_{jq})^2}\right]\right]. \tag{1.8b}$$

In Eq. (1.8b) the terms proportional to t give the correlation energy shift comparable to the second term in Eq. (1.7). The broadening is mainly due to the exponential terms in the function $F_k(t)$. Its order of magnitude is determined by the longitudinal excitation energies ω'_{jq}, the matrix elements M'_{jq}, and the temperature-dependent factors $[N(\omega'_{jq})+n(\varepsilon^e_{k+q})]$ and $[1+N(\omega'_{jq})-n(\varepsilon^e_{k+q})]$. If the temperature is sufficiently high, the relation $N(\omega'_{jq})>n(\varepsilon^e_{k+q})$ holds sufficiently well. In this case the contribution of the Fermi functions in the cumulant $F_k(t)$ to the broadening is negligible as can be checked numerically. Thus we use an approximation for the Green's function

$$G^r(k,t)=-i\Theta(t)\exp\left[-i\left[\varepsilon^e_k+\tilde{\Sigma}_k\right]t+\tilde{F}_k(t)\right], \tag{1.9a}$$

which contains two distinct parts. The self-energy contribution

$$\tilde{\Sigma}_k=-\sum_q V_q n(\varepsilon^e_{k+q})+\sum_{j,q}(M'_{jq})^2\left[\frac{1}{\varepsilon^e_k-\varepsilon^e_{k+q}+\omega'_{jq}}-\frac{1}{\varepsilon^e_k-\varepsilon^e_{k+q}-\omega'_{jq}}\right]n(\varepsilon^e_{k+q}) \tag{1.9b}$$

is the screened exchange energy. It is proportional to the plasma density. The second contribution is given by the cumulant

$$\tilde{F}_k(t)=\sum_{j,q}[M'_{jq}(q)]^2\left[N(\omega'_{jq})\left[\frac{it}{\varepsilon^e_{k+q}-\varepsilon^e_k-\omega'_{jq}}-\frac{1-e^{-i(\varepsilon^e_{k+q}-\varepsilon^e_k-\omega'_{jq})t}}{(\varepsilon^e_{k+q}-\varepsilon^e_k-\omega'_{jq})^2}\right]\right.$$
$$\left.+[1+N(\omega'_{jq})]\left[\frac{it}{\varepsilon^e_{k+q}-\varepsilon^e_k+\omega'_{jq}}-\frac{1-e^{-i(\varepsilon^e_{k+q}-\varepsilon^e_k+\omega'_{jq})t}}{(\varepsilon^e_{k+q}-\varepsilon^e_k+\omega'_{jq})^2}\right]\right], \tag{1.9c}$$

which depends on the density through the screened potential only. In $\tilde{F}_k$ the Fermi functions of F_k are omitted. The physical origin of the cumulant $\tilde{F}_k$ is derived from the interaction of one electron with the longitudinal excitations.

For this simple example this splitting is not necessary, because one can use Eq. (1.8) directly. But for the calculation of the pair amplitude this procedure allows the treatment of the plasmon-phonon induced sidebands. However, Eq. (1.8) is less rigorous than in the polaron problem.

II. DERIVATION OF THE EQUATION OF MOTION FOR THE PAIR AMPLITUDE

From the Heisenberg equation for the electron-hole pair operator $S^\dagger$ one obtains the equation of motion for the pair amplitude

$$\bar{S}^*_{k,-k+p}=\langle S^\dagger_{k,-k+p}\rangle_T, \tag{2.1}$$

where $\langle\cdots\rangle_T$ denotes the thermal average appropriate for a semiconductor which is in a state of quasiequilibrium. The resulting equation is

$$i\frac{\partial}{\partial t}\bar{S}^*_{k,-k+p}=-(E_g+\varepsilon^e_k+\varepsilon^h_{-k+p})\bar{S}^*_{k,-k+p}+\sum_q V_q\bar{S}^*_{k+q,-k-q+p}-\sum_q\langle(S^\dagger_{k-q,-k+p}-S^\dagger_{k,-k-q+p})\Phi_q\rangle_T$$
$$+\sum_{p_1}g_k E^*_{p_1}(t)(\delta_{p,p_1}-\langle d^\dagger_{-k+p}d_{-k+p_1}\rangle_T-\langle c^\dagger_k c_{k+p-p_1}\rangle_T), \tag{2.2}$$

where $g_{k+p-p_1}\simeq g_k$ has been taken. The potential operator Φ_q is given by

$$\Phi_q=V_q\rho_q-M_q A_q, \tag{2.3a}$$

with the abbreviations

$$A_q=b_q-b^\dagger_{-q}\quad\text{and}\quad\rho_q=\sum_k(c^\dagger_{k-q}c_k-d^\dagger_{k-q}d_k). \tag{2.3b}$$

An external charge ρ^x is introduced whose potential is added to Φ:

$$\Phi_q^x=\Phi_q+V_q\rho_q^x \ . \tag{2.4}$$

The external charge ρ^x is needed to derive self-consistent equations of motion with the functional derivative technique. Switching to a special interaction picture defined by

$$\hat{S}^+(t)=U(t,-\infty)S^+(t)U(-\infty,t) \ , \tag{2.5}$$

where the S-matrix $U(t,t_0)$ is given by

$$U(t,t_0)=T\exp\left[-i\int_{t_0}^{t}dt_1\sum_q\rho_q^x(t_1)[\hat{\Phi}_q^x(t_1)-\hat{\Xi}_q^x(t_1)]\right] , \tag{2.6}$$

where T is the time-ordering operator, will allow us to derive an effective equation of motion for the pair amplitude, which contains the coupling of the pair amplitude to the diagonalized LO-phonon–plasmon modes as has been described in the first chapter. They are used instead of the superposition Φ_q of charge-density excitations and LO phonons. The potential generated from the diagonalized LO-phonon–plasmon modes is denoted by Ξ_q. Furthermore, the sum of the potential Ξ_q and the potential of the external charge ρ^x is called $\Xi_q^x=\Xi_q+V_q\rho_q^x$. Ξ_q is given in terms of the phonon-plasmon mixed-mode operators:[12,13]

$$\begin{aligned}\Xi_q(t)&\simeq-\sum_{j=1,2}^{2}M_{jq}A_{jq}(t)\\&=-\sum_{j=1,2}^{2}M_{jq}(e^{-i\omega_{j,q}t}b_{j,q}+e^{i\omega_{j,-q}t}b_{j,-q}^{\dagger}) \ .\end{aligned} \tag{2.7}$$

The Hamiltonian H_M for these longitudinal eigenmodes is

$$H_M=\sum_{j,q}\omega_{j,q}b_{j,q}^{\dagger}b_{j,q} \ . \tag{2.8}$$

The matrix elements M_{jq} and the energies ω_{jq} of the eigenmodes are obtained from the requirement that the responses of Φ and Ξ to the external charge ρ^x are approximately the same. Thus we impose on Ξ the following condition:

$$\left\langle\frac{\delta\hat{\Xi}_q^x(t)}{\delta\rho_{q_1}^x(t_1)}\right\rangle_T\Bigg|_{\rho^x=0}-\left\langle\frac{\delta\hat{\Phi}_q^x(t)}{\delta\rho_{q_1}^x(t_1)}\right\rangle_T\Bigg|_{\rho^x=0}\simeq 0 \ . \tag{2.9}$$

The derivative $\delta\hat{\Phi}/\delta\rho^x$ at $\rho^x=0$ yields

$$\begin{aligned}&\frac{\delta}{\delta\rho_{q_1}^x(t_1)}U(-\infty,t)\Phi_q(t)U(t,-\infty)\Big|_{\rho^x=0}\\&\qquad=-i\Theta(t-t_1)[\Phi_q(t),\Phi_{q_1}(t_1)] \ ,\end{aligned} \tag{2.10}$$

which is a retarded response function. In the derivation of Eq. (2.10) we have used

$$[\Phi_{q_1}(t_1),\Xi_{q_2}(t_2)]=0 \ , \tag{2.11}$$

which holds because Ξ and Φ operate on different spaces and because the time evolution of Ξ is determined by H_M only. The derivative $\delta\langle\hat{\Phi}^x\rangle_T/\delta\rho^x$ gives the first term of the expansion of the self-consistent RPA result for the screened interaction $V_s^r(q,t)$ (Ref. 10) in terms of the polarization. This derivative will be replaced always by $V_s^r(q,t)$, i.e.,

$$\frac{\delta\langle\hat{\Phi}_q^x(t)\rangle_T}{\delta\rho_{q_1}^x(t_1)}\Bigg|_{\rho^x=0}\sim\delta(q+q_1)V_s^r(q,t-t_1) \ , \tag{2.12}$$

because the full sum of the RPA should be contained in the final results. Within the plasmon-phonon-pole approximation the dynamically screened Coulomb potential $V_s^r(q,\omega)$ can be decomposed into the sum of the bare Coulomb interaction and the contribution due to the interaction with longitudinal excitations. It is given by

$$\begin{aligned}V_s^r(q,\omega)\simeq&\frac{4\pi e^2}{\varepsilon_\infty q^2}-\frac{2\pi e^2\omega_{\rm L}}{\varepsilon^* q^2}\frac{2\omega_{\rm L}}{\omega_{\rm L}^2-(\omega+i\delta)^2}\\&-\frac{2\pi e^2\omega_p^2}{\varepsilon_0 q^2\omega_q}\frac{2\omega_q}{\omega_q^2-(\omega+i\delta)^2} \ .\end{aligned} \tag{2.13a}$$

In Eq. (2.13a) we neglect the coupling between plasmons and phonons to simplify the following numerical evaluation. The electron-phonon interaction is therefore unscreened. The effective dispersion of the plasma oscillation is

$$\omega_q^2=\omega_p^2\left[1+\frac{q^2}{\kappa^2}\right]+\left[\frac{\varepsilon_q^e+\varepsilon_q^h}{2}\right]^2 \tag{2.13b}$$

as suggested by Zimmerman[14] and Rice.[15] Here

$$\omega_p=\left[\frac{4\pi e^2N}{m\varepsilon_\infty}\right]^{1/2} \tag{2.13c}$$

is the plasma frequency, and

$$\kappa=\left[\frac{4\pi e^2}{\varepsilon_\infty}\left[\frac{\partial N^e}{\partial\mu^e}+\frac{\partial N^h}{\partial\mu^h}\right]\right]^{1/2} \tag{2.13d}$$

is the inverse screening length. Therefore, the dispersion relations ω_{jq} and the matrix elements M_{jq} are given for the LO phonons by

$$M_{1q}^2=\frac{2\pi e^2\omega_{\rm L}}{\varepsilon^* q^2}, \quad \omega_{1q}=\omega_{\rm L} \ , \tag{2.14a}$$

and for the plasmons by

$$M_{2q}^2=\frac{2\pi e^2\omega_p^2}{\varepsilon_0 q^2\omega_q}, \quad \omega_{2q}=\omega_q \ . \tag{2.14b}$$

The influence of the phonon-plasmon coupling on the first phonon-plasmon sidebands is discussed in more detail in Ref. 16.

From Eqs. (2.2) and (2.6) we get the equation of motion for $\langle\hat{S}^+\rangle_T$:

$$i\frac{\partial}{\partial t}\langle \hat{S}^{\dagger}_{k,-k+p}\rangle_T = -(E_g+\varepsilon^e_k+\varepsilon^h_{-k+p})\langle \hat{S}^{\dagger}_{k,-k+p}\rangle_T + \sum_q V_q \langle \hat{S}^{\dagger}_{k+q,-k-q+p}\rangle_T$$
$$- \sum_q \langle (\hat{S}^{\dagger}_{k-q,-k+p} - \hat{S}^{\dagger}_{k,-k-q+p})\hat{\Phi}^x_q\rangle_T$$
$$+ \sum_{p_1} g_k E^*_{p_1}(t)(\delta_{p,p_1} - \langle \hat{d}^{\dagger}_{-k+p}\hat{d}_{-k+p_1}\rangle_T - \langle \hat{c}^{\dagger}_k \hat{c}_{k+p-p_1}\rangle_T) . \tag{2.15}$$

$\hat{S}^{\dagger}$ can be cast into the form

$$\hat{S}^{+}(t) = [U^{\dagger}(\infty,-\infty)][TS^{+}(t)U(\infty,-\infty)] , \tag{2.16}$$

T denotes the time-ordering operator. $U(\infty,-\infty)$ is abbreviated by U from now on. Our aim is to calculate the thermal average $\langle \hat{S}^{\dagger}(t)\rangle_T$, where $\langle \hat{S}^{\dagger}(t)\rangle_T$ is the solution of Eq. (2.15). For the trace $\langle \cdots \rangle_T$ one needs to calculate the diagonal elements $\langle i|\hat{S}^{\dagger}(t)|i\rangle$. If $\rho^x(t)$ contains only frequencies which cannot induce resonant transitions, if it vanishes as $|t|$ approaches infinity, and if the complete set of states $|i\rangle$ is nondegenerate, it can be assumed that

$$\langle i|U^{\dagger}|j\rangle \propto \delta_{ij} , \tag{2.17}$$

which gives

$$\langle i|\hat{S}^{+}(t)|i\rangle = \langle i|U^{\dagger}|i\rangle\langle i|TS^{+}(t)U|i\rangle . \tag{2.18}$$

The functional derivative technique is used to handle the dependence on $\Phi-\Xi$ in the equation for $\hat{S}^{\dagger}$. Let $|i\rangle$ be an arbitrary state from the set which is needed to calculate the trace $\langle \cdots \rangle_T$. The following identity holds:

$$\langle i|[\hat{S}^{\dagger}_{k-q,-k+p}(t)-\hat{S}^{\dagger}_{k,-k-q+p}(t)][\hat{\Phi}^x_q(t)-\hat{\Xi}^x_q(t)]|i\rangle = i\frac{\delta\langle i|[\hat{S}^{\dagger}_{k-q,-k+p}(t)-\hat{S}^{\dagger}_{k,-k-q+p}(t)]|i\rangle}{\delta\rho^x_q(t^-)}$$
$$+\langle i|\hat{\Phi}^x_q(t)-\hat{\Xi}^x_q(t)|i\rangle\langle i|\hat{S}^{\dagger}_{k-q,-k+p}(t)-\hat{S}^{\dagger}_{k,-k-q+p}(t)|i\rangle . \tag{2.19}$$

t^- denotes a time which is infinitesimally earlier than t. The last term on the right-hand side of Eq. (2.19) vanishes at $\rho^x=0$ because

$$\langle i|\Xi_q(t)|i\rangle = 0 \quad \text{and} \quad \langle i|\Phi_q(t)|i\rangle \propto \delta_{q,0} \quad \text{for} \quad \rho^x \to 0 .$$

Equation (2.19) is introduced into Eq. (2.15). Since we do not use the resulting equation for calculating any self-consistent relations, we can take its limit $\rho^x=0$ which is

$$i\frac{\partial}{\partial t}\langle S^{\dagger}_{k,-k+p}(t)\rangle_T = -(E_g+\varepsilon^e_k+\varepsilon^h_{-k+p})\langle S^{\dagger}_{k,-k+p}(t)\rangle_T + \sum_q V_q\langle S^{\dagger}_{k+q,-k-q+p}(t)\rangle_T$$
$$- \sum_q \langle \Xi^x_q(t)[S^{\dagger}_{k-q,-k+p}(t)-S^{\dagger}_{k,-k-q+p}(t)]\rangle_T$$
$$- \frac{1}{-i}\sum_q \frac{\delta\langle[\hat{S}^{\dagger}_{k-q,-k+p}(t)-\hat{S}^{\dagger}_{k,-k-q+p}(t)]\rangle_T}{\delta\rho^x_q(t^-)}\bigg|_{\rho^x=0}$$
$$+ g_k E^*_p(t) - \sum_{p_1} g_k E^*_{p_1}(t)\langle d^{\dagger}_{-k+p}(t)d_{-k+p_1}(t)\rangle_T$$
$$- \sum_{p_1} g_{k+p-p_1}E^*_{p_1}(t)\langle c^{\dagger}_k(t)c_{k+p-p_1}(t)\rangle_T . \tag{2.20}$$

The functional derivative $\delta\langle \hat{S}^{\dagger}\rangle_T/\delta\rho^x$ in Eq. (2.20) can be decomposed as

$$\frac{\delta\langle \hat{S}^{\dagger}_{k-q,-k+p}(t)\rangle_T}{\delta\rho^x_q(t^-)}\bigg|_{\rho^x=0} = \left\langle \frac{\delta\hat{c}^{\dagger}_{k-q}(t)}{\delta\rho^x_q(t^-)}\bigg|_{\rho^x=0} d^{\dagger}_{-k+p}(t)\right\rangle_T + \left\langle c^{\dagger}_{k-q}(t)\frac{\delta\hat{d}^{\dagger}_{-k+p}(t)}{\delta\rho^x_q(t^-)}\bigg|_{\rho^x=0}\right\rangle_T . \tag{2.21}$$

The next step is to calculate the factors $\langle(\delta\hat{c}^{\dagger}/\delta\rho^x)d^{\dagger}\rangle_T$ and $\langle c^{\dagger}(\delta\hat{d}^{\dagger}/\delta\rho^x)\rangle_T$. The equation of motion for $c^{\dagger}$ is

$$-i\frac{\partial}{\partial t}c^{\dagger}_k(t) = (E_g+\varepsilon^e_k)c^{\dagger}_k(t) + \sum_q c^{\dagger}_{k-q}(t)\Phi_q(t) - \sum_p g_k E^*_p(t)d_{-p+k}(t) . \tag{2.22}$$

Using again the interaction representation (2.5) and the successive procedure, we find by multiplying the resulting equation with $d^{\dagger}_{k'}(t')$ (where $t'<t$) from the right

$$\left[-i\frac{\partial}{\partial t}-E_g-\varepsilon_k^e\right]\langle\hat{c}_k^\dagger(t)d_{k'}^\dagger(t')\rangle_T-\sum_q\langle\hat{\Xi}_q^x(t)\hat{c}_{k-q}^\dagger(t)d_{k'}^\dagger(t')\rangle_T$$
$$=\frac{1}{-i}\sum_q\left\langle\frac{\delta\hat{c}_{k-q}^\dagger(t)}{\delta\rho_q^x(t^-)}d_{k'}^\dagger(t')\right\rangle_T-\sum_p g_kE_p^*(t)\langle\hat{d}_{-p+k}(t)d_{k'}^\dagger(t')\rangle_T\,, \tag{2.23a}$$

where we have omitted the contributions which correspond to the last term of Eq. (2.19). In the limit $\rho^x\to 0$ its contribution will be proportional to terms of the form

$$\left\langle i\left|\frac{\delta\hat{\Xi}_q^x(t)}{\delta\rho_{q_1}^x(t_1)}\right|i\right\rangle-\left\langle i\left|\frac{\delta\hat{\Phi}_q^x(t)}{\delta\rho_{q_1}^x(t_1)}\right|i\right\rangle\Bigg|_{\rho^x=0}\,. \tag{2.23b}$$

Such terms are omitted because they are of the order of the difference $\langle\delta\hat{\Xi}^x/\delta\rho^x\rangle_T-\langle\delta\hat{\Phi}^x/\delta\rho^x\rangle_T$.

We are searching for an effective one-particle equation for $\langle\hat{c}^\dagger(t)d^\dagger(t')\rangle_T$ in Eq. (2.23a). For this purpose we use a screened Hartree-Fock approximation. Introducing the operator

$$G_\sigma^{-1}(k,t;k_1,t_1)=\delta(t-t_1)\left[\left[-i\frac{\partial}{\partial t}-E_g-\varepsilon_k^e\right]\delta(k-k_1)-\langle\hat{\Xi}_{k-k_1}^x(t)\rangle_T\right]+\Sigma(k,t;k_1,t_1)\,, \tag{2.24}$$

where a yet unknown self-energy Σ is introduced which has to describe the screened exchange with the plasma. G_σ^{-1} is the inverse of the Green's function

$$G_\sigma(k,t|k_1,t_1)=-i\langle T\bar{c}_k^\dagger(t)\bar{c}_{k_1}(t_1)\rangle_T\,, \tag{2.25a}$$

where $\bar{c}_k^\dagger(t)$ contains the time development due to Eq. (2.24). Furthermore,

$$\int dt_1\sum_{k_1}G_\sigma^{-1}(k,t;k_1,t_1)G_\sigma(k_1,t_1;k_2,t_2)=\delta(t-t_2)\delta(k-k_2)\,, \tag{2.25b}$$

$$\int dt_1\sum_{k_1}G_\sigma^{-1}(k,t;k_1,t_1)\langle\hat{c}_{k_1}^\dagger(t_1)d_{k'}^\dagger(t')\rangle_T=0\,. \tag{2.25c}$$

Using G_σ^{-1}, Eq. (2.23a) can be put into the form

$$\int dt_1\sum_{k_1}G_\sigma^{-1}(k,t;k_1,t_1)\langle\hat{c}_{k_1}^\dagger(t_1)d_{k'}^\dagger(t')\rangle_T=\int dt_1\sum_{k_1}\Sigma(k,t;k_1,t_1)\langle\hat{c}_{k_1}^\dagger(t_1)d_{k'}^\dagger(t')\rangle_T$$
$$+i\sum_{k_1}\left\langle\frac{\delta\hat{c}_{k_1}^\dagger(t)}{\delta\rho_{k-k_1}^x(t^-)}d_{k'}^\dagger(t')\right\rangle_T+\sum_q\langle\hat{\Xi}_q^x(t)\hat{c}_{k-q}^\dagger(t)d_{k'}^\dagger(t')\rangle_T$$
$$-\sum_q\langle\hat{\Xi}_q^x(t)\rangle_T\langle\hat{c}_{k-q}^\dagger(t)d_{k'}^\dagger(t')\rangle_T-\sum_p g_kE_p^*(t)\langle\hat{d}_{-p+k}(t)d_{k'}^\dagger(t')\rangle_T\,. \tag{2.26}$$

The self-energy is defined through the requirement

$$\int dt_1\sum_{k_1}\Sigma(k,t|k_1,t_1)\langle\hat{c}_{k_1}^\dagger(t_1)d_{k'}^\dagger(t')\rangle_T+i\sum_{k_1}\left\langle\frac{\delta\hat{c}_{k_1}^\dagger(t)}{\delta\rho_{k-k_1}^x(t^-)}d_{k'}^\dagger(t')\right\rangle_T=0\,. \tag{2.27}$$

The derivative of G_σ^{-1} with respect to ρ^x is

$$\frac{\delta G_\sigma^{-1}(k,t;k_1,t_1)}{\delta\rho_{p_2}^x(t_2)}=-\delta(t-t_1)\frac{\delta\langle\hat{\Xi}_{k-k_1}^x(t)\rangle_T}{\delta\rho_{p_2}^x(t_2)}+\frac{\delta\Sigma(k,t;k_1,t_1)}{\delta\rho_{p_2}^x(t_2)}\,. \tag{2.28}$$

The derivative of Eq. (2.26) with respect to ρ^x yields

$$\frac{\delta\langle\hat{c}_k^\dagger(t)d_{k'}^\dagger(t')\rangle_T}{\delta\rho_{k_2}^x(t_2)}=\int dt_1\sum_{k_1}G_\sigma(k,t|k_1,t_1)\left[\sum_{k_3}\frac{\delta\langle\hat{\Xi}_{k_1-k_3}^x(t_1)\rangle_T}{\delta\rho_{k_2}^x(t_2)}\langle\hat{c}_{k_3}^\dagger(t_1)d_{k'}^\dagger(t')\rangle_T\right.$$
$$\left.-\int dt_3\sum_{k_3}\frac{\delta\Sigma(k_1,t_1;k_3,t_3)}{\delta\rho_{k_2}^x(t_2)}\langle\hat{c}_{k_3}^\dagger(t_3)d_{k'}^\dagger(t')\rangle_T\right]$$
$$+G_\sigma\frac{\delta}{\delta\rho^x}(\langle\hat{\Xi}^x\hat{c}^\dagger d^\dagger\rangle_T-\langle\hat{\Xi}^x\rangle_T\langle\hat{c}^\dagger d^\dagger\rangle_T)-G_\sigma\frac{\delta}{\delta\rho^x}gE^*\langle\hat{d}d^\dagger\rangle_T\,. \tag{2.29}$$

The term containing the field E^* is neglected because we are interested in the linear response of the pair amplitude only. Correlations induced by the derivatives $\delta\Sigma/\delta\rho^x$ and $\delta(\langle\hat{\Xi}^x\hat{c}^\dagger d^\dagger\rangle_T-\langle\hat{\Xi}^x\rangle_T\langle\hat{c}^\dagger d^\dagger\rangle_T)/\delta\rho^x$ are neglected as well; they describe vertex corrections. With these approximations we find

$$\int dt_3\sum_{k_3}\Sigma(k_1,t_1;k_3,t_3)\langle\hat{c}^\dagger_{k_3}(t_3)d^\dagger_{k'}(t')\rangle_T\simeq -i\int dt_4\sum_{k_3,k_4,k_5}G_\sigma(k_3,t_1;k_4,t_4)\frac{\delta\langle\hat{\Xi}^x_{k_4-k_5}(t_4)\rangle_T}{\delta\rho^x_{k_1-k_3}(t_1^-)}\langle\hat{c}^\dagger_{k_5}(t_4)d^\dagger_{k'}(t')\rangle_T\ . \tag{2.30}$$

The correlation function $\langle\delta\hat{\Xi}/\delta\rho^x\rangle_T$ is the retarded dynamically screened Coulomb potential V_s^r. We replace the retarded function V_s^r by its static limit V_s which is consistent with Eq. (2.17). Taking the thermal average of the Green's function yields finally the following approximation for $\langle[\delta\hat{c}^\dagger(t)/\delta\rho^x(t^-)]d^\dagger(t')\rangle_T$:

$$\begin{aligned}\left\langle\frac{\delta\hat{c}^\dagger_k(t)}{\delta\rho^x_q(t^-)}d^\dagger_{k'}(t')\right\rangle_T&\simeq\int dt_1\sum_{k_1}V_s^r(q,t^--t_1)G_\sigma(k,t;k_1,t_1)\langle\hat{c}^\dagger_{k_1+q}(t_1)d^\dagger_{k'}(t')\rangle_T\\&\simeq\sum_{k_1}V_s(q)G_\sigma(k,t;k_1,t^-)\langle\hat{c}^\dagger_{k_1+q}(t)d^\dagger_{k'}(t')\rangle_T\\&\simeq iV_s(q)n^e_k(t)\langle\hat{c}^\dagger_{k+q}(t)d^\dagger_{k'}(t')\rangle_T\ .\end{aligned} \tag{2.31}$$

Now we are able to calculate the functional derivatives

$$\begin{aligned}&\frac{1}{-i}\sum_q\frac{\delta\langle[\hat{c}^\dagger_{k-q}(t)\hat{d}^\dagger_{-k+p}(t)-\hat{c}^\dagger_k(t)\hat{d}^\dagger_{-k+p-q}(t)]\rangle_T}{\delta\rho^x_q(t^-)}\bigg|_{\rho^x=0}\\&\simeq\frac{1}{-i}\sum_q V_s(q)[in^e_{k-q}(t)\langle\hat{c}^\dagger_k(t)d^\dagger_{-k+p}(t)\rangle_T+(-i)n^h_{-k+p}(t)\langle c^\dagger_{k-q}(t)\hat{d}^\dagger_{-k+p+q}(t)\rangle_T\\&\qquad-in^e_k\langle\hat{c}^\dagger_{k+q}(t)d^\dagger_{-k+p-q}(t)\rangle_T-(-i)n^h_{-k+p-q}(t)\langle c^\dagger_k(t)\hat{d}^\dagger_{-k+p}(t)\rangle_T]\Big|_{\rho^x=0}\\&=-\sum_q V_s(q)(n^e_{k-q}+n^h_{-k+p-q}\bar{S}^*_{k,-k+p}+V_s(q)(n^e_k+n^h_{-k+p})\bar{S}^*_{k+q,-k+p-q}\ ,\end{aligned} \tag{2.32}$$

and insert the result into Eq. (2.20). The relation $\langle\Xi S\rangle_T=\langle\Xi\bar{S}\rangle_T$ holds, where the thermal average reduces there to

$$\langle\Xi\bar{S}\rangle_T=\frac{\mathrm{Tr}(e^{-\beta H_M}\Xi\bar{S})}{\mathrm{Tr}(e^{-\beta H_M})}\ . \tag{2.33}$$

With this procedure we obtain the following effective equation of motion for the pair amplitude $\bar{S}$:

$$\begin{aligned}i\frac{\partial}{\partial t}\bar{S}_{k,-k+p}=&\left[E_g+\left[\varepsilon^e_k-\sum_q V_{s,q}n^e_{k+q}\right]+\left[\varepsilon^h_{-k+p}-\sum_q V_{s,q}n^h_{-k+p-q}\right]\right]\hat{S}_{k,-k+p}\\&-\sum_q(V_q-V_{s,q}n^e_k-V_{s,q}n^h_{-k+p})\bar{S}_{k+q,-k+p-q}\\&-\sum_q\langle\Xi_q(t)(\bar{S}_{k-q,-k+p}-\bar{S}_{k,-k+p-q})\rangle_T-g_k(1-n^e_k-n^h_{-k+p})E_p(t)\ .\end{aligned} \tag{2.34}$$

The screened exchange energies[10] are the terms proportional to the Fermi functions n^i_k $(i=e,h)$, whose quasichemical potentials μ^i are given in terms of the plasma density $N=N^i=\sum_k n^i_k$. The potential $\Xi_q(t)$ contains the contributions of the fluctuating field A_q of the LO phonons and the fluctuating plasma density ρ_q, therefore it contains among other things the missing part of the correlation energy. If the whole correlation energy is calculated statically but on equal footing, by choosing $\Xi=0$ in the above derivation, the resulting equation for the pair amplitude is

$$\begin{aligned}i\frac{\partial}{\partial t}\bar{S}_{k,-k+p}=&\left[E_g+\varepsilon^e_k+\varepsilon^h_{-k+p}-\sum_q V_{s,q}(n^e_k+n^h_k)-\sum_q(V_q-V_{s,q})\right]\bar{S}_{k,-k+p}\\&-\sum_q V_{s,q}(1-n^e_k-n^h_{-k+p})\bar{S}_{k+q,-k+p-q}-g_k(1-n^e_k-n^h_{-k+p})E_p(t)\ ,\end{aligned} \tag{2.35}$$

which is the inhomogeneous integral equation for the pair amplitude driven by a coherent field, in the form which has been derived and investigated by Haug *et al.*[1,10,17] and Zimmermann *et al.*[18] In Eq. (2.34) the normal static RPA self-energy shifts in form of the screened exchange term and the Coulomb-hole term are obtained. Naturally the procedure which gives the Coulomb-hole term can be used only if the static limit

$$\sum_q (V_q - V_{s,q}) = V(r=0) - V_s(r=0)$$

is finite, which does not hold if the LO phonons are included in the correlation energy. In this case the Rayleigh-Schrödinger perturbation theory can be used to obtain the energy shifts, as in Eq. (1.9b). Equation (2.35) would not allow the calculation of the phonon-plasmon sidebands, but Eq. (2.34) still contains the dynamics due to the phonon-plasmon excitations and thus it allows the calculation of the sidebands. The thermal average of Ξ_q cannot be evaluated directly, because its solution involves correlations between $\Xi_q(t)$ and $\Xi_{q'}(t')$, whose expectation value is nonzero, while the expectation value of Ξ_q itself vanishes.

If Eq. (2.34) if Fourier transformed to real space, it gives in principle the band-edge equation of Stahl and Balslev.[19]

III. APPROXIMATE CALCULATION OF THE PAIR AMPLITUDE

As mentioned in the first chapter, Eq. (2.34) is now solved by diagonalizing the interaction of the internal degrees of freedom of the pair amplitude with the longitudinal excitations Ξ_q approximately. To facilitate the notation, we describe only the exciton–LO-phonon problem for polar semiconductors[6] for demonstrating the derivation of the general result. The equation to be solved is

$$i\frac{\partial}{\partial t}\bar{S}_{k,-k+p_0} = (E_g + \varepsilon_k^e + \varepsilon_{p_0-k}^h)\bar{S}_{k,-k+p_0} - \sum_q V_q \bar{S}_{k+q,-k+p_0-q} - \sum_q M_q \langle A_q(t)(\bar{S}_{k-q,-k+p_0} - \bar{S}_{k,-k+p_0-q})\rangle_T - g_k \Theta(t)\mathcal{E}_{p_0} e^{i\omega_0 t} , \tag{3.1}$$

where the explicit time dependence of the coherent field

$$E(R,t) = \mathcal{E}_{p_0}\Theta(t) e^{-i(\omega_0 t - p_0 \cdot R)}, \quad \Theta(t) = \begin{cases} 1 \text{ for } t>0 \\ 0 \text{ for } t<0 \end{cases}, \tag{3.2}$$

with frequency ω_0 and wave vector p_0 has been inserted into Eq. (3.1). The field is switched on at time $t=0$. Let the set $\{\psi_n, \varepsilon_n\}$ be the exact eigenfunctions and eigenvalues of the homogeneous part of Eq. (3.1). Then the absorption spectra are given by

$$\alpha(\omega_0) = \zeta \,\mathrm{Im}\left\langle \sum_n \frac{|\psi_n(r=0)|^2}{\varepsilon_n - \omega_0 - i\delta} \right\rangle_T, \quad \delta > 0 , \tag{3.3}$$

where only the resonant terms have been considered. The explicit dependence on the phonon coordinates has to be eliminated by a thermal average. The prefactor ζ is approximately given by $\zeta \simeq E_g 4\pi^2 g^2 a_0^3/cn$, where a_0 is the bare exciton radius.

Since the exact wave functions ψ_n are unknown, one has to express α in terms of a perturbation series. The set of eigenfunction and eigenvalues of the Schrödinger equation for the exciton without coupling to the phonons is denoted by $\{\psi_n^0, \varepsilon_n^0\}$. The absorption constant α is now expressed by

$$\alpha(\omega_0) = \zeta' \,\mathrm{Im} \sum_{n,m} \psi_n^0(r=0)\psi_m^{0*}(r=0) G_{nm}(\omega_0 + i\delta) . \tag{3.4}$$

$G_{nm}(\omega + i\delta)$ is the Fourier-transformed retarded one-particle exciton Green's function, which describes the evolution of the state ψ_n^0 at time zero to the state ψ_m^0 at time t, under the interaction with the phonons. So the band tail which consists of the phonon sidebands is contained in $G_{nm}(t)$. A straightforward application of the cluster expansion to the calculation of $G_{nm}(t)$ leads to the exponential of a time-dependent infinite dimensional matrix. This matrix mixes the internal states of the exciton. However, such a complicated form cannot be handled. In order to overcome this problem, a statically screened electron-hole interaction potential is introduced, generating a set of eigenfunctions and eigenvalues $\{\chi_n, \nu_n\}$. This set diagonalizes Eq. (3.1) with respect to the internal motion approximately, and results in an effective coupling between each of those states and the LO phonons. With this approximation Eq. (3.4) reduces to

$$\alpha(\omega_0) \simeq \zeta' \,\mathrm{Im} \sum_n |\chi_n(r=0)|^2 G_{nn}^{\chi}(\omega_0 + i\delta)\Big|_{p=0} . \tag{3.5}$$

G_{nn}^{χ} is a similar Green's function as G_{nn}, but evaluated for the set $\{\chi_n, \nu_n\}$. χ_n and ν_n are determined by a pair equation with the pair interaction $V_{s,q} = \varepsilon_\infty/\varepsilon_0 V_q$, which is statically screened by phonons:

$$\nu_n \chi_n(k) = \left[E_g + \frac{k^2}{2\mu} + \frac{p^2}{2M}\right]\chi_n(k) - \sum_{k_1} V_s(k-k_1)\chi_n(k_1) . \tag{3.6}$$

Here, $\mu = (1/m_e + 1/m_h)^{-1}$ is the relative mass and $M = m_e + m_h$ is the total exciton mass. The quantum number $n = p, m$ contains the total momentum p and the quantum number for the internal states of the electron-hole pair which is denoted by m. Equation (3.1) for the electron-hole amplitude can be represented by a Hamiltonian $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_I$. In real space this Hamiltonian is given by

$$\mathcal{H}_0 = E_g - \frac{\Delta_{r_e}}{2m_e} - \frac{\Delta_{r_h}}{2m_h} - V_s(r_e - r_h) + \omega_L \sum_q b_q^\dagger b_q \tag{3.7a}$$

and

$$\mathcal{H}_I = V_s(r_e - r_h) - V(r_e - r_h) + \sum_q M_q (b_q + b_{-q}^\dagger)(e^{iqr_e} - e^{iqr_h}) . \tag{3.7b}$$

H_0 is diagonalized by $\{\chi_{mp},\nu_{mp}\}=\{e^{ip\cdot R}\phi_m(r),\nu'_m+\nu''_p\}$. $r=r_e-r_h$ is the relative coordinate, and ν'_m is the associated eigenenergy. R is the center-of-mass coordinate and $\nu''_p=p^2/2M$ is the corresponding eigenenergy. Time-dependent first-order perturbation theory for the evolution of the amplitude

$$\bar{S}_{mp}(t)=e^{-i\nu_{mp}t}\hat{S}_{mp}(t) \tag{3.8}$$

gives

$$\begin{aligned}\hat{S}_{mp}(t)=\hat{S}_{mp}(0)&-i\int_0^t dt_1\sum_{m_1}(V_s-V)_{mm_1}e^{-i(\nu_{m_1,p}-\nu_{m,p})t_1}\hat{S}_{m_1p}(0)\\&+(-i)^2\int_0^t dt_1\int_0^{t_1}dt_2\sum_{q,m_1,m_2}Y_{mm_1}(q)Y_{m_1m_2}(-q)\langle A_q(t_1)A_{-q}(t_2)\rangle_T\\&\times e^{-i(\nu_{m_1,p+q}-\nu_{m,p})t_1}e^{-i(\nu_{m_2,p}-\nu_{m_1,p+q})t_2}\hat{S}_{m_2p}(0)\ ,\end{aligned} \tag{3.9}$$

with

$$Y_{mm_1}(q)=M_q\langle\phi_m|\{\exp[(i\mu/m_e)\mathbf{q}\cdot\mathbf{r}]-\exp[(-i\mu/m_h)\mathbf{q}\cdot\mathbf{r}]\}|\phi_{m_1}\rangle\ , \tag{3.10}$$

$$(V_s-V)_{mm_1}=\langle\phi_m|(\tilde{V}_s-V)|\phi_{m_1}\rangle\ , \tag{3.11}$$

$$\langle A_q(t_1)A_{-q}(t_2)\rangle_T=(N_L+1)e^{-i\omega_L(t_1-t_2)}+N_Le^{i\omega_L(t_1-t_2)}\ , \tag{3.12}$$

and

$$N_L=\frac{1}{e^{\beta\omega_L}-1}\ . \tag{3.13}$$

$\langle\phi_m|\cdots|\phi_{m_1}\rangle\equiv\int\phi^*_m(r)\cdots\phi_{m_1}(r)d^3r$ is the ordinary scalar product. The integration over t_1 and t_2 yields

$$\begin{aligned}\hat{S}_{mp}(t)=\hat{S}_{mp}(0)&+\sum_{m_1}(V_s-V)_{mm_1}\frac{e^{-i(\nu_{m_1,p}-\nu_{m,p})t}-1}{\nu_{m_1,p}-\nu_{m,p}}\hat{S}_{m_1p}(0)\\&-\sum_{q,m_1,m_2,\delta}Y_{mm_1}(q)Y_{m_1m_2}(-q)N_\delta\left[-\frac{(e^{-i(\nu_{m_2,p}-\nu_{m,p})t}-1)}{(\nu_{m_2,p}-\nu_{m_1,p+q}-\delta\omega_L)(\nu_{m_2,p}-\nu_{m,p})}\right.\\&\left.+\frac{e^{-i(\nu_{m_1,p+q}-\nu_{m,p}+\delta\omega_L)t}-1}{(\nu_{m_2,p}-\nu_{m_1,p+q}-\delta\omega_L)(\nu_{m_1,p+q}-\nu_{m,p}+\delta\omega_L)}\right]\hat{S}_{m_2p}(0)\ .\end{aligned} \tag{3.14}$$

Here the parameter δ takes the values ±1, with $N_-=N_L$ and $N_+=N_L+1$. It is not possible to take only the diagonal part of Eq. (3.14) as an approximation, because the continuum contribution would diverge. With respect to the relative electron-hole motion the static limit is taken

$$\omega_L\gg\nu'_m-\nu'_j\ , \tag{3.15}$$

which holds if $E_{1s}\leq\omega_L$. With this approximation (3.14) yields

$$\begin{aligned}\hat{S}_{mp}(t)-\hat{S}_{mp}(0)\simeq&\sum_{q,m_1,m_2,\delta}Y_{mm_1}(q)Y_{m_1m_2}(-q)N_\delta\left[\frac{1-e^{-i(\nu''_{p+q}-\nu''_p+\delta\omega_L)t}}{(\nu''_{p+q}-\nu''_p+\delta\omega_L)^2}\right]\hat{S}_{m_2p}(0)\\&+\sum_{q,m_2,\delta}\left[\sum_{m_1}Y_{mm_1}(q)Y_{m_1m_2}(-q)\frac{N_\delta}{\nu''_p-\nu''_{p+q}-\delta\omega_L}+(\tilde{V}_s-V)_{mm_2}\right]\left[\frac{e^{-i(\nu_{m_2,p}-\nu_{m,p})t}-1}{\nu_{m_2,p}-\nu_{m,p}}\right]\hat{S}_{m_2p}(0)\ .\end{aligned} \tag{3.16}$$

Now the sum over m_1 can be carried out:

$$\sum_{m_1}Y_{mm_1}(q)Y_{m_1m_2}(-q)=M_q^2[2\delta_{m,m_2}-2\langle\phi_m|\cos(q\cdot r)|\phi_{m_2}\rangle]\ .$$

The contribution of $(V_s - V)_{mm_2}$ cancels a part within the large parentheses of the second term on the right-hand side of Eq. (3.16), if the limit $\omega_L \to \infty$ is used. Explicitly they are

$$\lim_{\omega_L \to \infty} \left[\sum_{\delta,q} 2M_q^2 [\phi_m | \cos(\mathbf{q}\cdot\mathbf{r}) | \phi_{m_2}] \frac{N_\delta}{\nu_p'' - \nu_{p+q}'' - \delta\omega_L} + (\phi_m | V_s - V | \phi_{m_2}) \right] = 0 . \tag{3.17}$$

Therefore this contribution is omitted. The remaining part in the second term is a self-energy

$$-it \left[\sum_{\delta,q} 2M_q^2 \frac{N_\delta}{\nu_p'' - \nu_{p+q}'' - \delta\omega_L} \right] \hat{S}_{mp}(0) . \tag{3.18}$$

Here however, the limit $\omega_L \to \infty$ is not taken, because $\sum_q M_q^2/\omega_L$ would diverge. By this procedure one avoids the introduction of a cutoff vector in the summation over q, and the localization aspect of the center of mass remains included. The first term on the right-hand side of Eq. (3.16) is responsible for the phonon sidebands. We use the following approximation for the matrix elements

$$\langle \phi_m | [2 - 2\cos(\mathbf{q}\cdot\mathbf{r})] | \phi_{m_2} \rangle \simeq 2\delta_{m,m_2}(1 - e^{-a_m^2 q^2}) = f_m(q)\delta_{m,m_2} . \tag{3.19}$$

The cancelations which are derived from $\exp\{-a_m^2 q^2\}$ in the effective matrix element $f_m(q)$ are necessary only if excitonic bound states with a small Bohr radius are treated. a_m is taken as the Bohr radius, thus $f_m(q) \equiv 2$ suffices for our calculations. Therefore, Eq. (3.16) reduces to

$$\hat{S}_{mp}(t) = \hat{S}_{mp}(0) + \left[\sum_{q,\delta} 2M_q^2 N_\delta \left[\frac{-it}{\nu_p'' - \nu_{p+q}'' - \delta\omega_L} - \frac{1 - e^{-i(\nu_{p+q}'' - \nu_p'' + \delta\omega_L)t}}{(\nu_{p+q}'' - \nu_p'' + \delta\omega_L)^2} \right] \right] \hat{S}_{mp}(0) = [1 + F_p(t)] \hat{S}_{mp}(0) , \tag{3.20}$$

where F_p is the cumulant which is independent from m in this case. The cluster expansion is the exponential resummation of the perturbation theoretic series; this gives

$$\hat{S}_{mp}(t) = e^{F_p(t)} \hat{S}_{mp}(0) , \tag{3.21}$$

and

$$G^{\chi}_{mp,mp}(t) = \exp[-i\nu_{mp}t + F_p(t)] \tag{3.22}$$

for the Green's function G^{χ}. The sum in Eq. (3.5) can thus be written as

$$\alpha(\omega_0) = \zeta \, \mathrm{Im} \left[i \int_0^\infty dt \, e^{i\omega_0 t} \left[\sum_m |\chi_{m,p=0}(r=0)|^2 e^{-i\nu_m' t} \right] e^{F_{p=0}(t)} \right] . \tag{3.23}$$

The time-dependent integrand in the above formula factorizes into two parts. The factor in the parentheses is calculated from the set $\{\chi_{m0}, \nu_{m0}\}$, which depends on the statically screened Coulomb potential. This accounts for the screening due to the phonons. The second factor describes an effective scattering of the center of mass, which accounts for the localization of the center of mass. This factor is responsible for the phonon sidebands of the related spectrum.

The additional scattering due to the plasmons is easily included into the cumulant F_p in Eq. (3.20); it is in principle the same calculational procedure. If the plasma density is low, the condition for the application of the quasistatic limit is not fulfilled for the plasmon energy, which is the correspondant condition to inequality (3.15). However, for increasing plasma density the situation becomes better, because the exciton energy decreases and the plasmon energy increases. The last task is to take the screened exchange terms of Eq. (2.34) into account, i.e., one has to calculate the set of eigenfunctions and eigenvalues $\{\bar{\chi}_{m,p=o}, \bar{\nu}_m' + \bar{\nu}_{p=0}''\}$ of the following equation:

$$\bar{\nu}_m' \bar{\chi}_{m0}(k) = \left[E_g + \frac{k^2}{2\mu} - \sum_q V_{s,q}(n^e_{k+q} + n^h_{-k-q}) \right] \bar{\chi}_{m0}(k) - \sum_q V_{s,q}(1 - n^e_k - n^h_{-k}) \bar{\chi}_{m0}(k+q) . \tag{3.24}$$

The functions n_k^i and $\sum_q V_{s,q} n^i_{k+q}$ depend only on k^2. For these functions the dependence on k^2 will be replaced by a mean value k_m^2 for all occurrences in Eq. (3.24). If $\bar{\chi}_{m0}$ is a bound state, $k_m^2 = -\langle \bar{\chi}_{m0} | \Delta | \bar{\chi}_{m0} \rangle$ is used, and if $\bar{\chi}_{m0}$ is a continuum solution, $k_m^2 = p_m^2$ is taken, where p_m is determined from the asymptotics and $\bar{\chi}_{m0}(r)$, i.e., $\bar{\chi}_{m0}(r) \sim \sin[p_m r + O(\ln r)]$. This approximation is denoted by $\langle n_k^i \rangle_m$ and $\langle \sum_q V_{s,q} n^i_{k+q} \rangle_m$. It is exact for zero plasma density and for a sufficiently high plasma density in which case the Coulomb potential is completely screened. Thus Eq. (3.24) can be transformed into real space, which yields

$$\bar{\nu}_m' \bar{\chi}_{m0}(r) = \left[E_g - \frac{\Delta_r}{2\mu} - \left\langle \sum_q V_{s,q} n^e_{k+q} \right\rangle_m - \left\langle \sum_q V_{s,q} n^h_{-k-q} \right\rangle_m \right] \bar{\chi}_{m0}(r) - V_s(r)(1 - \langle n^e_k \rangle_m - \langle n^h_{-k} \rangle_m) \bar{\chi}_{m0}(r) . \tag{3.25}$$

The statically screened Coulomb potential $V_{s,q}$ is obtained from Eq. (2.13a):

$$V_{s,q}=\frac{4\pi e^2}{\epsilon_0 q^2}\frac{\omega_q^2-\omega_p^2}{\omega_q^2} , \tag{3.26}$$

which can be transformed to r space directly. In order to have analytical solutions the resulting potential $V_s(r)$ is fitted to the Hulthén potential[20]

$$V_s(r)\simeq\frac{e^2\kappa'}{\epsilon_0}\frac{1}{e^{\kappa' r}-1} , \tag{3.27}$$

where $\kappa'=\kappa$ can be chosen for the most purposes. Let the bound states be labeled by m; then their contributions to the probability amplitudes are

$$|\bar{\chi}_m(r=0)|^2 =(\kappa')^2 N_m^2\,|{}_2F_1(2\alpha_m+1+m,\,1-m;2\alpha_m+1;1)|^2 , \tag{3.28}$$

${}_2F_1(a,b;c;z)$ is the hypergeometric function[21] and

$$N_m^{-2}=\frac{1}{4\pi\kappa'}\sum_{j,n=0}^{m} b_j^{(m)}b_n^{(m)}\frac{1}{2\alpha_m+j+n} , \tag{3.29a}$$

where the normalization constants N_m are obtained by normalizing the wave functions given in Ref. 22 to unity. The coefficients $b_j^{(m)}$ are

$$b_0^{(m)}=1 , \tag{3.29b}$$

$$b_m^{(m)}=(-1)^m\frac{(2\alpha_m+1+m)_{m-1}}{(2\alpha_m+1)_{m-1}} , \tag{3.29c}$$

$$b_j^{(m)}=-m(2\alpha_m+m)\frac{(2\alpha_m+1+m)_{j-1}(1-m)_{j-1}}{(2\alpha_m+1)_m m!} \quad \text{if } 0<j<m . \tag{3.29d}$$

Pochhammer's symbol $(z)_n$ which occurs in (3.29c) and (3.29d) is defined by $(z)_n=\Gamma(z+n)/\Gamma(z)$, where $\Gamma(z)$ is the Γ function.[21] The parameter α_m, the eigenenergy $\bar{\nu}'_m$ and the mean momenta k_m are given by

$$\alpha_m=\frac{\beta_m^2-m^2}{2m} \quad \text{with} \quad \beta_m^2=\frac{2\mu}{(\kappa')^2}\frac{e^2\kappa'}{\epsilon_0}(1-n^e_{k_m}-n^h_{-k_m}) . \tag{3.30}$$

$$\bar{\nu}'_m=-\alpha_m^2\frac{(\kappa')^2}{2\mu} , \tag{3.31}$$

and

$$-\langle\bar{\chi}_m|\Delta|\bar{\chi}_m\rangle\simeq\alpha_m^2(\kappa')^2=k_m^2 . \tag{3.32}$$

The maximum number of bound states is determined through the condition $\alpha_m>0$. The system of Eqs. (3.30) and (3.31) has to be solved numerically, because they are interdependent.

Let the states of the continuum be labeled by ξ. The wave functions given in Ref. 22 are superimposed such that they describe standing waves which are normalized to Dirac's δ function $\delta_{\xi,\xi'}$ by using the procedure which is described in a book of Sommerfeld.[23] Therefore the corresponding probability amplitudes are

$$|\bar{\chi}_\xi(r=0)|^2=(\kappa')^3\frac{|\xi-\beta_\xi^2\mathrm{Im}[\Psi(1+\gamma_\xi-i\xi)+\Psi(1-\gamma_\xi-i\xi)]|^2\,|\Gamma(1-2i\xi)|^2}{2\pi^2|\Gamma(1+\gamma_\xi-i\xi)|^2\,|\Gamma(1-\gamma_\xi-i\xi)|^2} , \tag{3.33}$$

where the Ψ function[21] is defined by $\Psi(z)=(d/dz)\ln\Gamma(z)$, and the sum over ξ means

$$\sum_\xi=\int_0^\infty d\xi . \tag{3.34}$$

The parameter γ_ξ, the eigenenergy $\bar{\nu}'_\xi$, and the mean momenta k_γ are given by

$$\gamma_\xi=(\beta_\xi^2-\xi^2)^{1/2} ,$$

with

$$\beta_\xi^2=\frac{2\mu}{(\kappa')^2}\frac{e^2\kappa'}{\epsilon_0}(1-n^e_{k_\xi}-n^h_{-k_\xi}) , \tag{3.35}$$

$$\bar{\nu}'_\xi=\frac{\xi^2(\kappa')^2}{2\mu} , \tag{3.36}$$

and

$$p_\xi^2=\xi^2(\kappa')^2=k_\xi^2 . \tag{3.37}$$

IV. RESULTS AND DISCUSSION

The resulting formula for the absorption coefficient is

$$\alpha(\omega_0)=\zeta\,\mathrm{Im}\left[i\int_0^\infty e^{i\omega_0 t}dt\,e^{F_0(t)}\left(\sum_m|\bar{\chi}_{m,0}(r=0)|^2(1-\langle n_k^e\rangle_m-\langle n^h_{-k}\rangle_m)e^{-i\bar{\nu}'_m t}\right)\right] , \tag{4.1}$$

with

$$F_0(t)=\sum_{j,q}\sum_{\delta=\pm1}2M_{jq}^2(q)N_\delta(\omega_{jq})\left[\frac{it}{q^2/2M+\delta\omega_{jq}}-\frac{1-e^{-i(q^2/2M+\delta\omega_{jq})t}}{(q^2/2M+\delta\omega_{jq})^2}\right]-\gamma_0 t . \tag{4.2}$$

γ_0 accounts for the zero-temperature linewidth of the absorption lines and is added phenomenologically.

In Eq. (4.2) the slow time dependence of the electron-hole density has been eliminated adiabatically. On the slow time scale the evolution of the density is described through a rate equation[10]

$$\frac{\partial N(t)}{\partial t} = \frac{\alpha(\omega_0, N(t))I(t)}{\omega_0} - \frac{N(t)}{\tau} . \tag{4.3}$$

For the range of excitation intensities and for the elevated temperatures which we consider, the linear absorption is always larger than the two-photon absorption.[24] The last term is the rate of the electron-hole recombination, which is assumed to be linear in N. τ is the corresponding decay time. For the wide-gap semiconductors under consideration the linear decay dominates over the nonlinear Auger recombination rate at all plasma densities which can be excited by band-tail absorption. The assumed spatial homogeneity is present if the diffusion length $\sqrt{D\tau}$ of the electron-hole plasma is longer than the crystal length.

For the numerical evaluations we use parameters appropriate for wurtzite CdS. We use masses and dielectric constants which are averaged over the crystal anisotropy. The used values are[25] $m_e = 0.2m_0$, $m_h = 1.0m_0$, $\varepsilon_\infty = 5.9$, $\varepsilon_0 = 8.6$, $\omega_L = 37$ meV, and $\Delta_{LT} = 2$ meV, where m_0 is the free electron mass. The exciton binding energy obtained from these values is 31 meV, the $T = 0$ exciton polaron shift $2\alpha\omega_L$ is 80 meV, where α is the exciton-Fröhlich coupling constant, which has the value of 1.1. Figures 1 and 2 show the absorption spectra of a weak probe beam testing a sample with various given plasma densities N for two different equilibrium temperatures. E_x is the exciton rydberg, and E_g is the bare band gap. The densities are

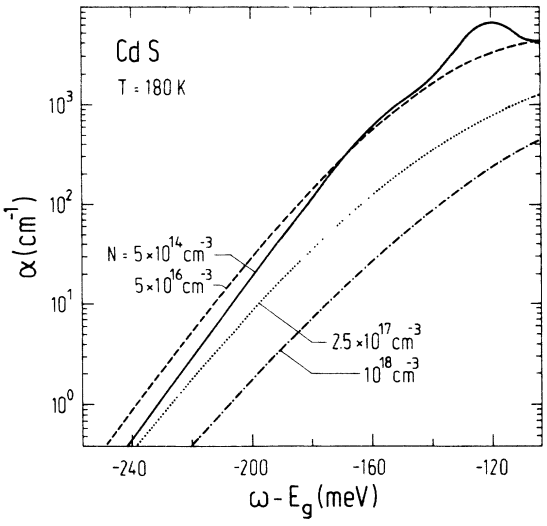

FIG. 1. Calculated absorption spectra at the absorption edge for various plasma densities N with an equilibrium temperature of 180 K. The absorption α is induced by the interaction with LO phonons and plasmons. It is plotted vs the light energy ω measured from the bare band gap E_g.

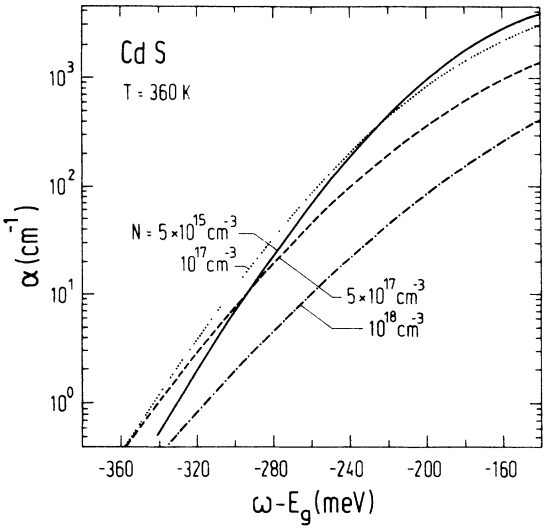

FIG. 2. The same as in Fig. 1 but at an equilibrium temperature of 360 K.

varied over a range which does not result in optical gain. The 1s-exciton line is smeared out. The influence of the exchange terms on the position of the 1s-exciton line cancels each other to a large extent. Two other sources of influence on its position remain, namely a red shift caused by the interaction with the longitudinal excitations and a blue shift which is derived from the decreasing binding energy of the exciton.[10,26] When the density increases, phase-space filling sets in, causing a net blue shift of the absorption edge. The band tail is dominated by the energy of the LO phonons ω_L, unless it is higher than the plasmon energy ω_p. Nevertheless, as the density increases, the tail broadens.

In the connection with Urbach's rule[3] one frequently discusses the quantity $\sigma = k_B T \partial \ln(\alpha(\omega_0))/\partial\omega_0$, which is almost temperature independent in the high-temperature regime. k_B is Boltzmann's constant. In Fig. 3 the calculated plasma density dependence of σ is shown. In the low-density region σ is practically constant. After some critical density has been exceeded σ decreases with increasing density. From Fig. 3 one obtains the heuristic relation

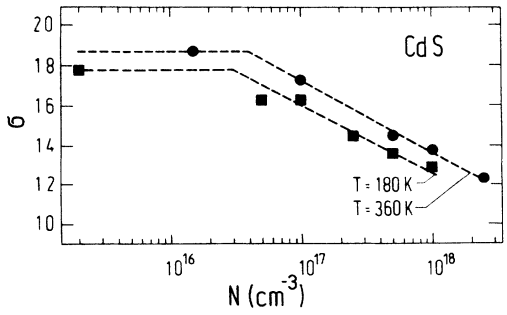

FIG. 3. Steepness parameter σ as a function of the plasma density N for the equilibrium temperatures of 180 and 360 K.

$$\sigma(T,N)\simeq\sigma(T,0)-0.17\ln\left[1+\frac{N}{N_0}\right],$$

$$N_0\simeq 5\times 10^{16}\ \mathrm{cm}^{-3}\ . \quad (4.4)$$

For zero plasma density and $T=360$ K the value of σ is 1.9, which has to be compared with the experimental value of 2.2 for CdS.[27]

Next we calculate for a single stationary beam the excited plasma density self-consistently. Figures 4 and 5 show the resulting absorption spectra (straight lines) for various intensities. The dotted curves are the low-density curves from the Figs. 1 and 2, which we show in order to facilitate the comparison. The spectra exhibit in the tail region two distinctly different regions. The low-energy region of the absorption tail changes very little with intensity and shows no saturation. In the successive high-energy region of the absorption tail a pronounced saturation occurs with increasing light intensity. The absorption saturates in this frequency range, when it exceeds a critical value. The parameter σ is independent of the temperature and of the density in the saturation region. Its numerical value is 0.37 ± 0.03.

Figures 6 and 7 show the pump-intensity dependence of the absorption α for various pump frequencies ω_0. The experimental situation equals the previous one. The slightly increasing absorption in the low-intensity regime is caused by the energy renormalization due to the optically excited *e-h* plasma. In the saturation region the absorption vanishes with increasing density as $\sqrt{I}$. In this region the saturation is described by the heuristic formula

$$\alpha(\omega_0,N(I))\propto\frac{\exp\left[\frac{0.37}{k_BT}(\omega_0-E_g+\tilde{E})\right]}{\sqrt{I}}$$

$$\text{with } \tilde{E}\simeq 30\ \text{meV}\ . \quad (4.5)$$

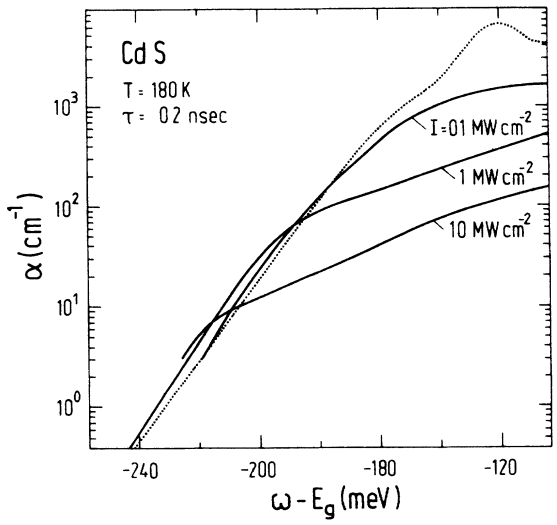

FIG. 4. Self-consistently calculated absorption spectra for various light intensities with an equilibrium temperature of 180 K.

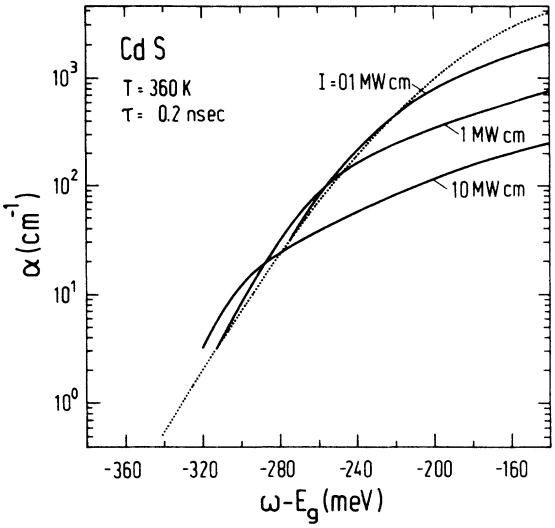

FIG. 5. The same as in Fig. 4 but at an equilibrium temperature of 360 K.

The curves in Figs. 4–7 are calculated with $\tau=0.2$ ns. Equation (4.4) shows how the curves will scale for different recombination times τ. Our theoretical treatment explains indeed the experimentally known fact that it becomes increasingly difficult to saturate the absorption if the pump beam is tuned far below the band gap. However, at the present time no systematical measurements of the band-tail absorption saturation are available, which could directly be compared to our results. The saturation of the resonant exciton absorption has been measured for CdS (Refs. 28–30) and for GaAs.[31] The ob-

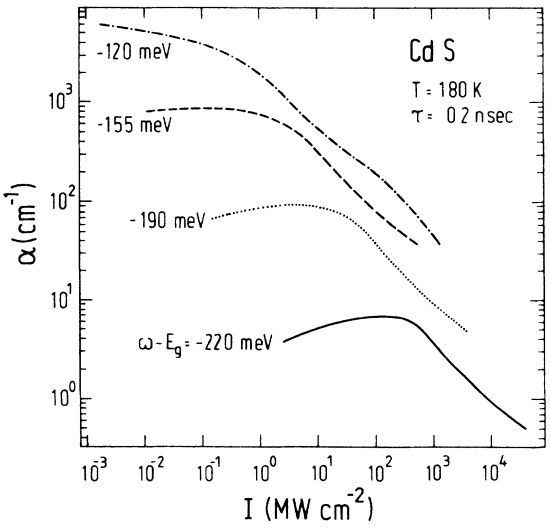

FIG. 6. Self-consistently calculated absorption depicted vs the light intensity for various frequencies of the light. The equilibrium temperature is 180 K.

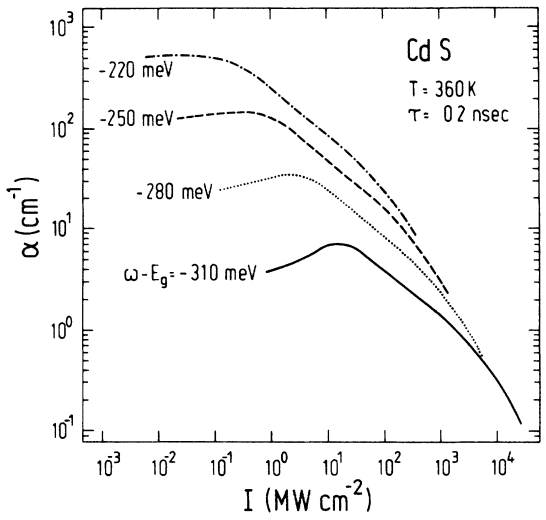

FIG. 7. The same as in Fig. 6 but at an equilibrium temperature of 360 K.

served absorption decrease determined in Ref. 29 is in agreement with our calculations. The transmission experiments on thin CdS platelets[29,26] cannot be compared directly with our results, because they are determined by the intensity-dependent changes of the absorption and refraction.

In summary, we have calculated the band-tail absorption of a polar semiconductor for arbitrary plasma densities in the high-temperature regime, where the plasma is generated by a stationary pump beam and relaxes fast into a thermal distribution. For zero plasma density the band tail is described by Urbach's rule. Our theory reproduces the measured slope parameter σ quite well. We have shown that under high-excitation conditions this rule still holds, but the slope parameter σ becomes plasma density dependent. We are not aware of systematic measurements in order to check these results in detail. Our theory yields the saturation of the absorption in the band-tail region. It explains, in accordance with the experiments, that it becomes increasingly difficult to saturate the absorption if the pump beam is tuned far below the band gap.

ACKNOWLEDGMENTS

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