Quantum kinetic effects in the optical absorption of semiconductor quantum-dot systems

M. Lorke, T. R. Nielsen, J. Seebeck, P. Gartner and F. Jahnke
Institute for Theoretical Physics, University of Bremen, 28334 Bremen, Germany
E-mail: mlorke@itp.uni-bremen.de

Abstract. A microscopic theory is used to study the optical properties of semiconductor quantum dots. The dephasing of the coherent polarization due to carrier-carrier Coulomb interaction and carrier-phonon interaction is determined from quantum kinetic equations. We investigate the density dependence of the dephasing mechanisms, and compare the relevance of various interaction processes. The failure of frequently used approximations based on the GKBA with free single-particle energies is demonstrated for pure dephasing processes involving only the localized quantum-dot states.

1. Introduction
Optical properties of semiconductors critically depend on dephasing processes, which are dominated by the electron-phonon interaction and, at elevated carrier densities, by the Coulomb interaction of carriers. We study the influence of both mechanisms on the optical absorption and gain spectra for a system of self-assembled InGaAs quantum dots (QDs) which are of intense current interest due to various technological applications in novel light emitting devices. Self-assembled QDs are characterized by localized QD states with discrete energies below a continuum of delocalized two-dimensional wetting layer (WL) states.

The theoretical description of interaction processes involving the localized QD states and the interplay between localized and delocalized states poses new challenges. Due to pure dephasing processes involving only discrete states, which turn out to be of critical importance, some frequently used approximations such as the free generalized Kadanoff-Baym ansatz (free GKBA) break down.

We present a quantum kinetic theory of the dephasing due to many-body effects of carrier-carrier Coulomb interaction and electron-LO-phonon interaction for an InGaAs QD-WL system. Previous studies [1, 2] for quantum well (QW) or QD-WL systems consider only the Coulomb interaction of carriers In Ref. [2] only a sub-Class of possible interaction processes in the QD-WL system is considered. We analyze the dephasing from all possible scattering processes due to carrier-carrier and carrier-phonon interaction. Pure dephasing processes (which do not change population) require a treatment beyond the free GKBA. We calculate the scattering processes for renormalized quasi-particles where the renormalization is self-consistently determined from the scattering self-energies. This approach includes the description of the carrier-phonon interaction in the polaron picture. The corresponding contributions for the carrier-Kinetics in the QD system have been studied in Ref. [3] for the carrier-carrier Coulomb interaction and in Ref. [4] for the interaction with LO phonons.
In general we find strong density dependent broadening and line-shift effects in the optical spectra of QDs due to Coulomb interaction which results in a non-Lorentzian lineshape for the QD resonances. The electron-phonon interaction gives rise to more complicated optical spectra stemming from phonon replicas and hybridizations in the polaronic spectral functions for the discrete states.

2. Theory of optical absorption

We consider the Coulomb interaction of carriers, the Fröhlich coupling to LO-phonons and the interaction with a classical optical light field via the dipole coupling. Our model system consists of two confined QD shells, which we label as s-shell for the ground state and p-shell for the double degenerate excited state, energetically below a quasi-continuum of delocalized WL states [3]. We assume a density of QDs on the WL of

$$n_{\text{QD}} = 1 \cdot 10^{10} \text{cm}^{-2}.$$  

For optical interband processes, we investigate the dynamics of the transition amplitude

$$\psi_\alpha(t) = -i\hbar G_{\alpha,eh}(t, t).$$

Here and in the following, $\alpha$ combines the quantum numbers for either WL or QD states. For linear optical spectra, determined in response to a weak optical field, changes of the electron and hole population, given by

$$f_{e\alpha}(t) = -i\hbar G_{\alpha,ee}(t, t)$$

and

$$f_{h\alpha}(t) = -i\hbar G_{\alpha,hh}(t, t),$$

respectively, can be neglected. Throughout this paper, the calculations are performed for quasi-equilibrium distributions for electrons and holes. To determine the dynamics of the transition amplitude $\psi_\alpha$, we solve quantum kinetic equations in Fourier space [1, 2]

$$\left(\hbar \omega - e_{\alpha}^{c,\text{HF}} - e_{\alpha}^{h,\text{HF}}\right)\psi_\alpha(\hbar \omega) + \left[1 - f_{e\alpha}^c - f_{e\alpha}^h\right] \Omega_{\alpha}^{\text{HF}}(\hbar \omega) = S^{\text{Coul}}_{\alpha}(\hbar \omega) + S^{\text{Phon}}_{\alpha}(\hbar \omega).$$  \hspace{1cm} (1)

The left hand side of Eq. (1) contains the contribution due to Hartree-Fock (HF) Coulomb interaction, which give rise to single-particle renormalizations $e_{\alpha}^{c,\text{HF}} = e_{\alpha}^c + \Sigma_{\alpha}^{c,\text{HF}}$ with

$$\Sigma_{\alpha}^{c,\text{HF}} = \sum_\beta \left( V_{\alpha \beta \alpha - V_{\alpha \beta \beta}} \right) f_{\beta}^a,$$

and to the excitonic resonance of the WL and QD states via

$$\Omega_{\alpha}^{\text{HF}}(\hbar \omega) = d \cdot E(\hbar \omega) + \sum_\beta V_{\alpha \beta \beta} \psi_{\beta}(\hbar \omega).$$

Note that in a spatially inhomogeneous QD-WL system the QD Hartree terms which are due to interaction with carriers in the same dot remain present, while all other Hartree contributions vanish because of global charge neutrality. This is discussed in detail in Ref. [5]. The correlation contributions due to Coulomb interaction and electron-LO-phonon interaction, which cause the dephasing and additional energy renormalizations, will be outlined in the following sections. For the present paper we assume in a first approximation that the dephasing contributions due to both mechanisms can be evaluated separately.

The scattering integrals on the R.H.S. of Eq. (1) for both Coulomb and carrier-phonon interaction can be separated into a part diagonal in the state index $\alpha$ for the transition amplitude (diagonal dephasing $\Gamma^{\text{DD}}$) and a off-diagonal part ($\Gamma^{\text{OD}}$) which mixes the coherent interband transition amplitudes for all states, i.e.

$$S_{\alpha}(\hbar \omega) = -\Gamma^{\text{DD}}_{\alpha}(\hbar \omega)\psi_{\alpha}(\hbar \omega) + \sum_{\alpha_1} \Gamma^{\text{OD}}_{\alpha \alpha_1}(\hbar \omega)\psi_{\alpha_1}(\hbar \omega).$$  \hspace{1cm} (2)

3. Coulomb correlations

3.1. Theory

The correlation contributions due to Coulomb interaction, $S^{\text{Coul}}_{\alpha}(\hbar \omega)$, are evaluated in the second order Born approximation. To close the set of equations for the interband transition amplitude $\psi$ we additionally apply the generalized Kadanoff-Baym ansatz (GKBA) [6, 7]. Within this
approach the correlation contributions due to Coulomb interaction read

\[ \Gamma^\text{DD}_\alpha(h\omega) = i \sum_{a,b=e,h} \sum_{a_1a_2a_3} W_{a_2a_3a_1} [2W_{a_2a_3a_1}^* - W_{a_2a_1a_3}^*] \]

\[ \times g \left( \frac{h\omega - \varepsilon^b_{a} - \varepsilon^a_{a_1} + (\varepsilon^a_{a_2})^* - \varepsilon^a_{a_3}}{1 - f_{a_2}^a f_{a_3}^a f_{a_1}^a + (f - 1 - f) f_{a_2}^b f_{a_3}^b f_{a_1}^b} \right) \]

\[ + 2W_{a_2a_3a_1}^* W_{a_2a_3a_1} \]

\[ \times g \left( \frac{h\omega - \varepsilon^b_{a} - \varepsilon^a_{a_1} + (\varepsilon^a_{a_2})^*}{1 - f_{a_2}^a f_{a_3}^a f_{a_1}^a + (f - 1 - f) f_{a_2}^b f_{a_3}^b f_{a_1}^b} \right), \]

and

\[ \Gamma^\text{OD}_{\alpha_1}(h\omega) = i \sum_{a,b=e,h} \sum_{a_2a_3} W_{a_2a_3a_1} [2W_{a_2a_3a_1}^* - W_{a_2a_1a_3}^*] \]

\[ \times g \left( \frac{h\omega - \varepsilon^b_{a} - \varepsilon^a_{a_1} + (\varepsilon^a_{a_2})^*}{1 - f_{a_2}^a f_{a_3}^a f_{a_1}^a + (f - 1 - f) f_{a_2}^b f_{a_3}^b f_{a_1}^b} \right) \]

\[ + 2W_{a_2a_3a_1}^* W_{a_2a_3a_1} \]

\[ \times g \left( \frac{h\omega - \varepsilon^b_{a} - \varepsilon^a_{a_1} + (\varepsilon^a_{a_2})^*}{1 - f_{a_2}^a f_{a_3}^a f_{a_1}^a + (f - 1 - f) f_{a_2}^b f_{a_3}^b f_{a_1}^b} \right). \]

The effective single-particle energies \( \varepsilon = \varepsilon^a - i\gamma^a \) appearing in the function \( g(\Delta) = \frac{1}{\Delta} \) include renormalized energies as well as the corresponding quasi-particle damping, as elaborated below. In the dephasing terms we find direct and exchange contributions, containing the screened interaction matrix elements \( W_{a_2a_3a_1} \), which are evaluated according to Ref. [3]. The availability of initial and final states for the dephasing processes is described by the population factors.

In a Markovian calculation, the dephasing rates would be frequency independent, whereas in our calculation the frequency dependence of the non-Markovian dephasing rates reflects the memory effects in the time domain. It was shown in Ref. [2] that for a QD-WL system with contributions of discrete states to the single-particle spectrum, the frequency dependence of the dephasing rates is important. Then the damping of a QD resonance should be determined by the dephasing at the corresponding interband transition energy. This important effect would not be included in the Markov approximation.

A frequently used approximation for QW or bulk systems is the free GKBA where unrenormalized energies are used in the scattering integrals. This leads to the limit \( g(\Delta) = \pi \delta(\Delta) + iP_{\Delta}^{1/2} \) where \( P \) denotes the principal value integral. The approximation fails completely when processes involving only the discrete QD states are taken into account, because the \( \delta \)-functions are not integrated out and the result is not well defined. Also a finite broadening of the \( \delta \)-function, which would be justified e.g. by the interaction of carriers with acoustic phonons, would lead to unphysical results. Then the resonance denominator of \( g(\Delta) \) can give rise to additional resonances at the wrong energies, as we will show in Section 3.2.

As the relative strength of the different processes is temperature and density dependent, it is important to include all possible dephasing channels in the calculation (for a discussion of the scattering channels in the framework of carrier kinetics see [3]).

**Self-consistent single-particle energy renormalizations** The renormalizations of the single-particle energies should be treated on the same footing as the dephasing processes itself, since they originate from the same self-energy.

At elevated carrier densities and temperatures, the Coulomb interaction is expected to cause only an energy shift accompanied by a broadening. Then we can apply a pole-approximation,
The retarded GF of the interacting system can be written as

\[
G_{\alpha,\alpha}^{\text{ret}}(\hbar \omega) = \frac{1}{\hbar \omega - \tilde{\varepsilon}_\alpha} = \frac{1}{\hbar \omega - \varepsilon^a_\alpha + i\gamma^a_\alpha} .
\]

(5)

With this ansatz, we define a complex effective single-particle energy, which consists of the renormalized energy \(\varepsilon^a_\alpha\) and the corresponding quasi-particle damping \(\gamma^a_\alpha\).

Within the pole approximation, effective single-particle energies are calculated self-consistently according to

\[
\tilde{\varepsilon}^a_\alpha = \varepsilon^a_\alpha + \Sigma^a_\alpha + \Sigma^a_{\text{ret}}(\tilde{\varepsilon}^a_\alpha) ,
\]

(6)

with the retarded self energy

\[
\Sigma^a_{\text{ret}}(\hbar \omega) = \sum_{b=e,h} \sum_{\alpha_1 \alpha_2 \alpha_3} W_{\alpha_2 \alpha_3 \alpha_1} \left[ 2W_{\alpha_2 \alpha_3 \alpha_1} - W_{\alpha_1 \alpha_2 \alpha_3} \right] 
\]

\[
\times g \left( \hbar \omega - \tilde{\varepsilon}_\alpha^a + \left( \tilde{\varepsilon}_\alpha^b \right)^* - \tilde{\varepsilon}_\alpha^c \right) \left[ (1 - f^a_{\alpha_2}) f^a_{\alpha_3} f^a_{\alpha_1} + (f \rightarrow 1 - f) \right] 
\]

\[
+ 2W_{\alpha_2 \alpha_3 \alpha_1} W_{\alpha_1 \alpha_2 \alpha_3} 
\]

\[
\times g \left( \hbar \omega - \tilde{\varepsilon}_\alpha^a - \tilde{\varepsilon}_\beta^b + \left( \tilde{\varepsilon}_\gamma^c \right)^* \right) \left[ f^b_{\alpha_2} \left( 1 - f^b_{\alpha_3} \right) f^a_{\alpha_1} + (f \rightarrow 1 - f) \right] .
\]

The imaginary part of the energies ensures that the denominator of the g-functions do not vanish. In other words, the self-consistent scheme avoids the energy-conserving \(\delta\)-functions, which would be meaningless over the discrete spectrum.

### 3.2. Results

In Fig. 1 optical absorption spectra for the combined QD-WL system for various carrier densities are shown. The excitonic resonance of the WL, p-shell, and s-shell can be identified at around -15meV, -90meV, and -150meV, respectively.

**Figure 1.** Imaginary part of the optical susceptibility for the combined QD-WL system and various carrier densities. For better visibility the inset shows a magnification of the QD absorption (T=300K).

For increasing carrier density, we observe a transition from absorption to gain, accompanied by a strong damping and a pronounced red-shift of the resonances, which has also been observed...
in experiments [8]. The s-shell resonance shows a clear saturation effect due to Pauli blocking. In contrast the gain at the p-shell resonance increases further and shows no saturation for the densities investigated here. Note that it is important to include all possible dephasing channels in the calculation, since their relative strength is density and temperature dependent. Equally important is the inclusion of the exchange scattering term, since it causes changes in the dephasing of about 30%.

The lineshape of the p-shell resonance is shown in Fig. 2. For low carrier densities, the dephasing due to Coulomb interaction leads to an approximately Lorentzian lineshape of the resonance. In the density regime close to transparency, the dephasing is increased and a clear non-Lorentzian character of the lineshape emerges.

As already mentioned, it is important to use renormalized energies in the Coulomb dephasing rates. In Fig. 3 the artifacts of a calculation with free single-particle energies in the scattering integrals are shown (dashed line) in comparison to the result with self-consistently renormalized energies (solid line). We observe a multitude of peaks, stemming from the resonance denominator of $g(\Delta)$ in Eqs. (3) and (4). The mixing of renormalized and free energies leads to a splitting of the interband transition lines. Furthermore, lines corresponding to the free s-shell and p-shell transition appear, when the free GKBA is used.

![Figure 3. Imaginary part of the optical susceptibility for the QD resonances and a carrier density of $1 \times 10^{10}$ cm$^{-2}$ with (dashed line) and without (solid line) self-consistently renormalized single-particle energies in the scattering integrals (T=77K). For illustrative purposes only the processes causing the artifacts are included.](image)

**Figure 3.** Imaginary part of the optical susceptibility for the QD resonances and a carrier density of $1 \times 10^{10}$ cm$^{-2}$ with (dashed line) and without (solid line) self-consistently renormalized single-particle energies in the scattering integrals (T=77K). For illustrative purposes only the processes causing the artifacts are included.

4. Carrier-phonon interaction

4.1. Theory

As discussed in Ref. [4] the carrier-phonon interaction in QDs requires a non-pertubative treatment within the polaron picture. To compute the LO-phonon contributions to the dephasing and energy renormalizations to Eq. (1) we use the random-phase approximation (RPA) and the GKBA with the retarded carrier Green’s function (GF) calculated in the polaron picture [4].
Then the correlation contributions due to LO-phonons are
\[
\Gamma^{\text{DD}}_{\alpha}(\hbar \omega) = i \sum_{a,b=e,h} \sum_{b \neq a} M^2_{\text{LO}} V_{\alpha \beta \alpha \beta} \left\{ (1 - f^a_\beta) \left[ (1 + N_{\text{LO}}) G^{a,b}_{\beta,\alpha} (\hbar \omega - \hbar \omega_{\text{LO}}) + N_{\text{LO}} G^{a,b}_{\beta,\alpha} (\hbar \omega + \hbar \omega_{\text{LO}}) \right] + f^a_\beta \left[ (1 + N_{\text{LO}}) G^{a,b}_{\beta,\alpha} (\hbar \omega + \hbar \omega_{\text{LO}}) + N_{\text{LO}} G^{a,b}_{\beta,\alpha} (\hbar \omega - \hbar \omega_{\text{LO}}) \right] \right\},
\]
and
\[
\Gamma^{\text{OD}}_{\alpha \beta}(\hbar \omega) = i \sum_{a,b=e,h} \sum_{b \neq a} M^2_{\text{LO}} V_{\alpha \beta \alpha \beta} \left\{ (1 - f^a_\beta) \left[ (1 + N_{\text{LO}}) G^{b,\alpha}_{\beta,\alpha} (\hbar \omega - \hbar \omega_{\text{LO}}) + N_{\text{LO}} G^{b,\alpha}_{\beta,\alpha} (\hbar \omega + \hbar \omega_{\text{LO}}) \right] + f^a_\alpha \left[ (1 + N_{\text{LO}}) G^{b,\alpha}_{\beta,\alpha} (\hbar \omega + \hbar \omega_{\text{LO}}) + N_{\text{LO}} G^{b,\alpha}_{\beta,\alpha} (\hbar \omega - \hbar \omega_{\text{LO}}) \right] \right\},
\]
where \( M^2_{\text{LO}} = 4 \pi \alpha \frac{\hbar}{\sqrt{2m}} (\hbar \omega_{\text{LO}})^{3/2} \). We use for \( G^{a,b}_{\beta,\alpha}(\hbar \omega) \) a convolution of two retarded polaronic GFs, so that the full polaronic effects are included in Eqs. (8) and (9). A more detailed discussion of the polaronic effects in a QD-WL system, such as phonon replicas and hybridizations in the spectral function, is given in Ref. [4].

4.2. Results
In contrast to our results for Coulomb interaction in Fig. 1, the optical spectra including dephasing due to carrier-phonon interaction displays a more complicated structure, since various kinds of polaronic features are revealed (Fig. 4).

**Figure 4.** Imaginary part of the optical susceptibility for the combined QD-WL system and various carrier densities with dephasing due to carrier-phonon interaction. The inset is a logarithmic representation that reveals the polaronic features of the spectra (T=300K).

**Figure 5.** Imaginary part of the optical susceptibility for the combined QD-WL system and various carrier densities with dephasing due to Coulomb and carrier-phonon interaction. The inset shows a magnification of the QD resonances (T=300K).

One can identify phonon replicas of the fundamental interband transitions which are modified by the hybridization in the polaronic states. This leads to a strong non-Lorentzian lineshape of
the QD resonances. The damping of the resonances at higher carrier densities is rather weak, and a saturation of the s-shell gain is not found. This shows that at elevated carrier densities, the Coulomb scattering of carriers needs to be included.

5. Dephasing due to carrier-phonon and Coulomb interaction
In this section we combine the dephasing contributions due electron-phonon and Coulomb interaction. For simplicity and illustrative purposes we evaluate the total dephasing by adding the dephasing contributions due to both mechanisms. In Fig. 5 the resulting absorption spectra are shown.

As in the case of dephasing only due to Coulomb interaction (Fig. 1) we find the bleaching and red-shift of the resonances and also the saturation of the s-shell gain. We observe that the Coulomb interaction is clearly the dominant dephasing mechanism for high carrier densities, although even in the gain regime some of the polaronic features remain present. For example, the high energetic side of the p-shell resonance shows a second peak at low densities (arrow 1), which becomes completely damped out with increasing density. On the other hand, the peak on the low energetic side of the p-shell resonance (arrow 2) is almost not visible at low densities but becomes more pronounced in the gain regime.

It is expected that these polaronic features vanish at high densities as the polaronic features in the spectral function should be damped out by Coulomb effects. This requires a self-consistent treatment of the spectral properties with inclusion of both Coulomb and polaronic effects.

6. Conclusion
A quantum kinetic theory is used to study the influence of dephasing processes on optical absorption and gain spectra in semiconductor quantum dot systems. It is shown, that the use of free energies in the dephasing rates for the carrier-carrier Coulomb interaction can lead to unphysical results. A non-Markovian treatment with self-consistently renormalized energies is necessary to treat pure dephasing processes which play a dominant role in QD systems. We observe a density dependent red-shift and a pronounced density dependent damping due to Coulomb correlations, in agreement with experiments. The electron-phonon interaction leads to polaronic features in the optical spectra via the hybridization and phonon-replica which contribute to a non-Lorentzian lineshape of the QD resonances.

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