Influence of carrier-carrier and carrier-phonon correlations on optical absorption and gain in quantum-dot systems

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A microscopic theory is used to study the optical properties of semiconductor quantum dots. The dephasing of a coherent excitation and line-shifts of the interband transitions due to carrier-carrier Coulomb interaction and carrier-phonon interaction are determined from a quantum kinetic treatment of correlation processes. We investigate the density dependence of both mechanisms and clarify the importance of various dephasing channels involving the localized and delocalized states of the system.

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I. INTRODUCTION

In recent years, semiconductor quantum dots (QDs) have been studied extensively due to possible applications in optoelectronic devices like LEDs, lasers, or amplifiers 1,2. In the rapidly emerging field of quantum information technology, QDs have been successfully used to demonstrate the generation of single photons or correlated photon pairs 3,4,5. Furthermore, the strong coupling regime for QD emitters in optical microcavities has been demonstrated 6,7. A common aspect in fundamental studies and practical applications of QDs is the critical role of dephasing processes. They determine the homogeneous linewidth of the QD resonances, limit the coherence properties of QD lasers and their ultrafast emission dynamics, and have a strong influence on coherent optical nonlinearities. Moreover, dephasing processes are intimately linked to lineshifts of the QD resonances.

Optical studies of QDs have been recently focused on self-assembled systems which are typically grown in the Stranski-Krastanoff mode. The resulting QDs are randomly distributed on a two-dimensional wetting layer (WL). The interplay of carriers in the localized QD and delocalized WL states is mediated by the Coulomb interaction of carriers in addition to carrier-phonon interaction.

Theoretical studies of absorption and gain in QDs have been accomplished on the basis of multilevel optical Bloch equations 8, or semiconductor Bloch equations (SBE) with screened exchange and Coulomb hole contributions 9. However, in these approaches the dephasing is merely a parameter entering the calculation via a $T_2$-time. In Ref. 10 the dephasing due to Coulomb interaction has been calculated using SBE with correlation contributions due to Auger-like WL-assisted capture processes (see below) where scattering integrals have been evaluated in terms of free-carrier energies. A more elaborate analysis of dephasing due to Coulomb interaction in quantum wells (QWs), which included non-Markovian scattering integrals based on renormalized energies, revealed quantitative modifications of the results 11. We show that for QD systems the situation is different due to the appearance of localized states with a discrete spectrum. The calculation of scattering integrals in terms of free-carrier energies breaks down for processes due to Coulomb interaction which involve only localized states. It turns out that these are among the dominant processes for QDs containing more than one confined shell. For other processes involving localized states, their energy renormalization is also of enhanced importance. In the first part of this paper we present a non-Markovian treatment of dephasing due to Coulomb interaction where energy renormalizations under high-density conditions typical for QD lasers are self-consistently included and the importance of various scattering channels is analyzed.

For the interaction of QD carriers with LA-phonons, extensive work has been devoted to the low temperature regime, 12,13,14. It is generally acknowledged that acoustic phonons dominate the dephasing at low temperatures. Nevertheless, several low temperature PL measurements 16,17 revealed LO-phonon replicas. In Ref. 15 it was suggested that for temperatures above 100K the dephasing due to carrier-LO-phonon interaction is of growing importance. The interaction of QD carriers with LO phonons is strongly influenced by hybridization effects 18, which require the application of the polaron picture. A quantum-kinetic description of carrier-phonon scattering based on polarons has recently been used to explain ultrafast carrier capture and relaxation processes in QDs 19. In the second part of this paper, we present the corresponding treatment of dephasing processes. Then the combined influence of Coulomb interaction and carrier-phonon interaction on optical absorption and gain spectra is analyzed for quasi-equilibrium excitation conditions.

In Refs. 20 and 21 PL spectroscopy measurements revealed homogeneous linewidths of several meV at room temperature. Comparable results have also been found...
by four-wave mixing experiments. Our microscopic calculation can reproduce these experimental findings even though a direct quantitative comparison, which is not the purpose of this paper, would require better knowledge of the specific QD parameters as well as possible improvements of our QD model.

The primary focus of this work is on QD optical spectra in the room-temperature high-density regime relevant for laser applications. For this situation, a quantum kinetic theory for carrier-carrier interaction and carrier scattering with LO-phonons is developed. It is shown that non-Markovian effects and energy renormalizations play an essential role in QD systems. The theory is evaluated to compare the contributions of various scattering channels to dephasing and line-shifts as well as their excitation density dependence. Studies of the temperature dependence are restricted to $T \geq 150\text{K}$ where the inclusion of LA-phonons is not necessary.

Our investigations show that the efficiency of dephasing processes depends strongly on the carrier density in the system. For low carrier densities and room temperature, the electron-LO-phonon interaction is the dominant mechanism, leading to the appearance of additional side-peaks in the optical spectra due to polaronic hybridization effects. Even though for high carrier densities the Coulomb interaction becomes the dominant mechanism, the electron-LO-phonon interaction remains important as it continues to contribute to the linewidth and line-shape of the QD resonances.

For typical InGaAs QD parameters, a QD density of $10^{10}\text{cm}^{-2}$ on the WL and carrier densities around $10^{10}\text{cm}^{-2}$ at room temperature, we obtain a homogeneous linewidth of the ground state resonance with a full width at half maximum (FWHM) of 3.2meV in agreement with the experimental findings of Ref. [20]. For lower carrier densities and decreasing temperature, we can reproduce the observed dephasing reduction while for elevated carrier densities and decreasing temperature, the reduction of the dephasing due to LO-phonons is compensated by increasing dephasing due to Coulomb scattering.

II. THEORY OF OPTICAL ABSORPTION & GAIN CALCULATIONS

The coherent optical excitation of semiconductors can be described in terms of interband transition amplitudes $\psi_\alpha$ and population functions $f_\alpha^e = \langle e_\alpha | e_\alpha \rangle$ and $f_\alpha^h = \langle h_\alpha | h_\alpha \rangle$ for electrons and holes, respectively. For the annihilation and creation operators of electrons and holes, $\alpha$ combines the quantum numbers for either WL or QD states. The dynamics of $\psi_\alpha(t)$ and $f_\alpha^e, h(t)$ in response to the optical field $E(t)$ is determined by the SBE [22] which can be used to incorporate many-body interaction effects to describe dephasing, energy renormalization and screening as demonstrated for QWs in Ref. [23]. The optical absorption or gain coefficient can be defined via the response of the system to a weak optical field $E(\omega)$ in terms of the linear optical susceptibility $\chi(\omega) = P(\omega)/E(\omega)$. Here $P(\omega)$ is the Fourier transform of the macroscopic polarization $P(t) = \sum_\alpha d_\alpha^\dagger \psi_\alpha$ with the interband dipole matrix element $d_\alpha$.

A solution linear in the optical probe field $E(t)$ implies that changes of the electron and hole population $f_\alpha^e, h$ due to $E(t)$ can be neglected. In the following we consider quasi-equilibrium distributions for electron and holes which are approximately realized in injection-current driven devices. Alternatively the carriers can be generated with an optical pre-pulse where a sufficient delay of the probe pulse ensures the the excited carriers are thermalized and that coherent effects of the pump pulse can be neglected.

In this regime, the Fourier transform of the coherent interband transition amplitude $\psi_\alpha$ obeys the equation

$$\left(\hbar \omega - e_\alpha^e \frac{\varepsilon}{\varepsilon} - e_\alpha^h \frac{\varepsilon}{\varepsilon} \right) \psi_\alpha(\hbar \omega) + \left[1 - f_\alpha^e - f_\alpha^h \right] \Omega_\alpha(\hbar \omega) = S_\alpha^\text{Coul}(\hbar \omega) + S_\alpha^\text{Phon}(\hbar \omega).$$  \(1\)

The single particle energies entering Eq. [1] include already renormalizations due to Hartree-Fock (HF) Coulomb effects according to $\varepsilon_\alpha^e = e_\alpha^e + \Sigma_\alpha^{\varepsilon\text{HF}}$ and $\varepsilon_\alpha^h = e_\alpha^h + \Sigma_\alpha^{\varepsilon\text{HF}}$ with $\alpha = e, h$. Here $e_\alpha$ are the free-carrier energies and $\Sigma_\alpha^{\varepsilon\text{HF}} = \sum_\beta (V_{\alpha\beta}\psi_\beta - V_{\alpha\beta}\psi_\beta^\dagger) f_\beta^\dagger$ combines the Hartree and exchange Coulomb self-energy. In a spatially homogeneous system like bulk semiconductors or for the in-plane motion for QWs, the Hartree terms cancel due to local charge neutrality. In the QD-WL system, the absence of local charge neutrality leads to Hartree contributions for the QD states. The influence of the WL states can be incorporated approximately within screened QD Hartree contributions as discussed in detail in Ref. [21]. The Coulomb exchange contributions to the interband Rabi energy $\Omega_\alpha(\hbar \omega) = d \cdot E(\hbar \omega) + \sum_\beta V_{\alpha\beta}\psi_\beta(\hbar \omega)$ give rise to the excitonic resonance of the WL and QD states.

Without the terms $S_\alpha^\text{Coul}(\hbar \omega)$ and $S_\alpha^\text{Phon}(\hbar \omega)$ we would recover the well-known semiconductor Bloch equations formulated in an arbitrary eigenfunction basis. $S_\alpha^\text{Coul}(\hbar \omega)$ and $S_\alpha^\text{Phon}(\hbar \omega)$ are used to describe correlation contributions which lead to dephasing of the coherent polarization and to the corresponding additional energy renormalizations due to Coulomb interaction and carrier-phonon interaction, respectively. These terms will be discussed in the following sections. A general feature of the following description is that the dephasing contributions for both the Coulomb interaction and the carrier-phonon interaction can be separated into a part which is diagonal in the state index $\alpha$ for the transition amplitude (diagonal dephasing $\Gamma^{DD}$) and a corresponding off-diagonal part ($\Gamma^{DD}$) which mixes the coherent interband transi-
The imaginary part of $\Gamma^{DD}$ describes a dephasing (damping) of the coherent polarization which can be expressed as a state and frequency dependent $T_2$ time. The contribution of $\Gamma^{DD}$ is however partly compensated by the off-diagonal contribution $\Gamma^{OD}$. The real parts of $\Gamma^{DD}$ and $\Gamma^{OD}$ give rise to additional renormalizations of the energies on the L.H.S. of Eq. (1).

with the screened Coulomb interaction matrix elements $W_{a_\alpha a_\alpha}$ discussed in Appendix A. Note that $\tilde{\epsilon}_a = \frac{1}{a} \alpha - i\gamma_\alpha$ appearing in the function $g(\Delta) = \frac{1}{3}$ are effective complex single-particle energies which combine renormalized energies $\epsilon_\alpha$ as well as the corresponding quasi-particle damping $\gamma_\alpha$. This is elaborated in more detail below.

As for the particle scattering [27] we have direct and exchange contributions which are proportional to $2W_{a_\alpha a_\alpha}$ and $W_{a_\alpha a_\beta} W_{a_\alpha a_\beta}$, respectively. The population factors describe the availability of initial and final states. For the above discussed excitation conditions, the population factors are time independent and we have restricted ourselves to the contributions linear in the transition amplitude $\psi_\alpha$.

The frequency dependence of $\Gamma^{DD}$ and $\Gamma^{OD}$ in Eqs. (3) and (4) reflect the non-Markovian treatment of the dephasing processes. Indeed, in this case, the product of the type $\Gamma_\psi$ of Eq. (4) amounts, in the time domain, to a convolution integral describing memory effects. The Markovian limit is obtained by pulling out from this integral the slowly varying component of $\psi_\alpha(t)$. Then the fast component $e^{i(\epsilon_\alpha + \tilde{\epsilon}_a) t}$ $(a \neq b)$ fixes the frequency value at $\omega = \omega_a = \frac{1}{2}(\tilde{\epsilon}_a + \tilde{\epsilon}_b)$ for $\Gamma^{DD}$ and $\omega = \omega_a$ for $\Gamma^{OD}$, so that in this limit both dephasing contributions become frequency independent (see Appendix C for details). If one combines the Markov approximation with the use of free-carrier energies in the scattering contributions, the diagonal dephasing is given by the sum of in- and out-scattering rates, $\text{Im}(\Gamma^{DD}) = S^{\text{in}}_\alpha + S^{\text{out}}_\alpha$. Here $S^{\text{in, out}}_\alpha$ are the rates in the Markovian kinetic equation for the carrier population, as defined e.g. in Ref. [27]. Results of these equations for optical spectra have been studied in detail for QW systems. If one restricts the analysis to diagonal dephasing contributions $\Gamma^{DD}$ and neglects off-diagonal dephasing terms $\Gamma^{OD}$, damping of the excitonic resonances is grossly overestimated [28]. Non-Markovian calculations further reduce the interaction-induced broadening and line-shift of the excitonic resonances in QW systems [11].

For the following discussion we would like to point out that one has to compare on one side the Markov approximation with a non-Markovian treatment and on the other side the use of free-carrier energies versus renormalized

$$S_\alpha(h\omega) = - \Gamma^{DD}_\alpha(h\omega) \psi_\alpha(h\omega) + \sum_{\alpha_i} \Gamma^{OD}_{\alpha\alpha_i}(h\omega) \psi_{\alpha_i}(h\omega).$$

(2)
energies in the scattering integrals. The non-Markovian treatment is more crucial in QD systems compared to QWs, since the discrete part of the spectrum with large energy separation emphasizes the frequency-dependence of the dephasing rates.10

For the use of free-carrier energies in the scattering contributions, the limit $\gamma_a^a \to 0, \gamma_a^b > 0$ leads to $\gamma(\Delta) = \pi \delta(\Delta) + iP \frac{1}{\Delta}$ where $P$ denotes the principal value integral. This approximation implies serious difficulties in a QD system when processes are taken into account which involve only discrete states. In these cases the $\delta$-functions are not integrated out and thus the results are not well defined. Note that this is not a artifact of the Markov approximation but also applies to the non-Markovian calculation. Even if one would introduce a finite broadening of the $\delta$-function, e.g., due to interaction of carriers with acoustic phonons, the non-Markovian calculation still yields unphysical results. This is because the spectrum predicted by Eq. 10 has peaks at the energies of the L.H.S. renormalized by the correlation contributions on the R.H.S. and hence, $\hbar \omega$ samples renormalized interband transitions. If they are mixed with free-carrier energies in the correlation contributions Eqs. 13 and 14, via $g(\hbar \omega - \Delta)$, then artificially also free carrier transitions and hybridization effects between all these energies can appear in the optical spectra, which are absent when self-consistently renormalized energies are used.

Another important point, which complicates the treatment of the coupled QD-WL system, is the existence of many scattering channels which are often partly neglected (for a discussion of the scattering channels in the framework of carrier kinetics see Ref. 27). This can strongly influence the results as we will show in Section 11.

**Self-consistent single-particle energy renormalizations**

The renormalization of the single-particle energies, which enter in the dephasing rates, originates from the same interaction processes that determine the dephasing itself. Technically, they can be traced back to the same many-body self-energy. The renormalized energies can be obtained from the poles of the retarded Green’s function (GF) in Fourier space. In the absence of interaction, the retarded GF is given by

$$G_{a,a,\text{ret}}^0(\hbar \omega) = \frac{1}{\hbar \omega - e_a^a + i\delta}$$

with the single-particle energy $e_a^a$ and $\delta \to 0, \delta > 0$. At elevated carriers densities, it is a reasonable approximation, that the main effect of the Coulomb interaction is a shift of the single-particle energy and the addition of a quasi-particle broadening such that the single-pole structure of the retarded GF remains valid. This picture corresponds to the Landau theory of a Fermi liquid and leads to the retarded GF of the interacting system

$$G_{a,a}(\hbar \omega) = \frac{1}{\hbar \omega - \bar{e}_a^a} = \frac{1}{\hbar \omega - e_a^a + i\gamma_a^a}.$$  

With this ansatz, we define a complex effective single-particle energy $\bar{e}_a^a = e_a^a - i\gamma_a^a$, which consists of a renormalized energy $e_a^a$ and the corresponding quasi-particle damping $\gamma_a^a$. In the pole approximation, the self-consistency requirement leads to

$$\bar{e}_a^a = e_a^a + \Sigma_{\alpha,\text{HF}} + \Sigma_{\alpha,\text{ret}}^a(\bar{e}_a^a)$$

with the retarded self-energy

$$\Sigma_{\alpha,\text{ret}}^a(\hbar \omega) = -i \sum_{b=e,h} \sum_{a_1a_2a_3} \times \left\{ W_{a_1a_2a_3}^a a_{a_2a_3} + 2W_{a_1a_2a_3}^a - W_{a_1a_2a_3}^a \right\} g \left( \hbar \omega - \bar{e}_a^a + (\bar{e}_a^a)^* - \bar{e}_{a_2}^a \right) \left[ (1 - f_{a_2}) f_{a_1} f_{a_1} + (f \to 1 - f) \right]$$

$$\times \left[ f_{a_2} (1 - f_{a_1}) f_{a_1} + (f \to 1 - f) \right].$$

The close connection between the diagonal dephasing and the retarded self-energy $\Sigma_{\alpha,\text{ret}}^a$ can be expressed as

$$\Gamma_{\alpha,\text{DD}}^a(\hbar \omega) = \sum_{a,b=e,h} \sum_{b \neq a} \Sigma_{\alpha,\text{ret}}^a(\hbar \omega - \bar{e}_b^a).$$

**B. Results**

Throughout this paper, results are presented for a carrier and lattice temperature of 300K and a density of
QDs on the WL of \(n_{\text{QD}} = 10^{10} \text{cm}^{-2}\). Further details of the QD model are given in Appendix A.

In Fig. 1 we show optical absorption spectra for the combined QD-WL system and various carrier densities. Excitation-induced dephasing and energy renormalizations due to Coulomb interaction have been included according to Eqs. (3),(4) and (7),(8). Identifiable are the excitonic resonance of the WL at around -15meV as well as the p-shell and s-shell resonances at about -90meV and -150meV, respectively. We observe a strong damping of these resonances with increasing carrier density which is accompanied by a pronounced red shift of the QD lines. The transition from absorption to gain takes place at a density \(\sim 1 \times 10^{11} \text{cm}^{-2} (\sim 5 \times 10^{11} \text{cm}^{-2})\) for the s-shell (p-shell). An important point for practical applications is the saturation of the optical gain for the s-shell accompanied by an increasing red-shift. This is due to a combination of state filling and saturation of dephasing at the s-shell resonance. In contrast the gain at the p-shell resonance increases further and shows no saturation for the densities investigated here.

**Importance of different dephasing processes and frequently used approximations**

The Coulomb matrix elements allow to distinguish between different dephasing processes in complete analogy to the carrier scattering [27]. If all four indices of \(W_{\alpha_2\alpha_3\alpha_1}\) are WL states, we refer to the corresponding processes as WL relaxation. They describe dephasing and energy shifts of the WL states in the optical spectra. Apart from the fact that we have to include OPW corrections to the interaction matrix elements for a proper description of the coupled QD-WL system [27], this resembles the case of a QW system. Since the processes involving QD states (except intra-dot processes, see below) are discussed in great detail in Ref. [27] we will only give a brief survey here.

If one of the four indices of \(W_{\alpha_2\alpha_3\alpha_1}\) corresponds to a QD state, we consider the dephasing mechanism as WL assisted carrier-capture, because in the corresponding scattering process an electron or a hole is captured from the WL to the QD [27]. Likewise we refer to processes with two QD state indices as WL assisted relaxation and to processes with three QD state indices as dot-assisted processes. Intra-dot processes are described when all four indices of \(W_{\alpha_2\alpha_3\alpha_1}\) correspond to QD states. These scattering events are not important in the carrier dynamics, because we only consider two confined shells for electrons and holes and therefore such scattering processes cannot redistribute carriers. However, they clearly provide additional dephasing of the coherent polarization. Note that the intra-dot processes are not the only events which produce dephasing without redistribution of carriers. Such processes, in which two carriers switch their states, also appear in the WL assisted relaxation and in the WL relaxation.

**FIG. 1:** Imaginary part of the optical susceptibility for the combined QD-WL system including interaction-induced dephasing and line shifts due to Coulomb interaction for various total carrier densities. The inset shows a scale up of the QD resonances.

**FIG. 2:** Imaginary part of the optical susceptibility for the s-shell resonances at a carrier density of \(2 \times 10^{11} \text{cm}^{-2}\). The result including all considered Coulomb scattering processes (thick solid line) is compared to calculations where only certain classes of processes are evaluated.

Figure 2 shows that WL assisted relaxation (dash-dotted line) and intra-dot scattering (thin solid line),
which include the processes leading to dephasing without redistribution of carriers, are the most important contributions, while WL assisted capture (dotted line) and dot-assisted (dashed line) processes cause a rather small dephasing. However, since this picture can change with carrier density, it is important to test the influence of all dephasing channels.

In Fig. 3 we compare the optical susceptibility from the full calculation (solid line) with a result where only the diagonal dephasing contributions were taken into account (dotted lines). For the excitonic resonance of the

\[
\chi_\alpha(\omega) = \frac{1}{\pi} \int dE \frac{\text{Im} \chi(\omega - i\Gamma_{\text{DD}}(\omega) + i\Gamma_{\text{OD}}(\omega))}{(E - E_g)^2 + \Gamma_{\text{DD}}(\omega) + \Gamma_{\text{OD}}(\omega)},
\]

with exchange scattering

\[
\Gamma_{\text{DD}}(\omega) = \frac{1}{\pi} \int dE \frac{\text{Im} \chi(\omega - i\Gamma_{\text{DD}}(\omega) + i\Gamma_{\text{OD}}(\omega))}{(E - E_g)^2 + \Gamma_{\text{DD}}(\omega) + \Gamma_{\text{OD}}(\omega)},
\]

without exchange scattering

WL, the absence of the off-diagonal dephasing leads to an overestimation of the linewidth by roughly a factor of two under the considered high-density conditions. Using only the diagonal dephasing contributions turns out to be a reasonable approximation for the lowest QD resonances while for the excited QD transition the lineshape is not fully reproduced. Off-diagonal dephasing contributions are less important for the QD resonances due to the rather large spectral separation between the QD states and the WL states and for the same reason off-diagonal contributions are stronger for the p-shell than for the s-shell.

Note that the foregoing discussion applies to the non-Markovian treatment. If one neglects the off-diagonal dephasing and uses the Markov approximation, again the dephasing is grossly overestimated when all relevant scattering processes are included. For the discussed QD model, transitions due to localized states are completely damped out in this approximation.

The influence of the exchange term in the dephasing rates, which is often disregarded, is also investigated in Fig. 3. Neglecting the exchange scattering in the dephasing contributions clearly overestimates the homogeneous linewidth by about 30%, thus pointing out that the exchange terms are almost as important as the direct term and should be included in the calculation.

**Renormalization of single-particle states**

As discussed above, the single particle renormalizations are of critical importance for the proper determination of the dephasing rates. In Fig. 4, the single particle renormalizations due to Coulomb interaction are shown. The Fock and correlation contributions lead to a decrease of the single-particle energies with increasing carrier density.

![FIG. 4: Renormalized single-particle energies of the QD states for electrons and holes as a function of the total carrier density in the system (a) and corresponding single-particle broadening(b).](image-url)
efficiency. Additionally at very high carrier densities the intra-dot processes are suppressed due to Pauli-blocking. These effects result in a strong decrease of the single-particle broadening.

IV. CARRIER-PHONON INTERACTION

A. Theory

In this section, we compute correlation contributions in the optical spectra due to interaction of carriers with LO-phonons. In contrast to the frequently applied time-dependent perturbation theory, it has been pointed out in Ref. [10] that the interaction involving localized QD carriers requires a description in the polaron picture. In Ref. [19], quasi-particle renormalizations described by a polaronic retarded GF have been used with in a quantum-kinetic theory to evaluate scattering processes and populations changes. We extent this treatment to the polarization dynamics to analyze the corresponding dephasing and interband-energy renormalizations.

Within the random-phase approximation (RPA) and the GKBA, the correlation contributions due to LO-phonons are

\[
\Gamma_{\alpha}^{DD}(\hbar \omega) = i \sum_{a,b,e,h}^{b \neq a} \frac{M_{LO}^2}{\varepsilon^2/\varepsilon_0} V_{\alpha \beta \alpha \beta} \left\{ (1 - f^e_{\beta}) \left[ (1 + N_{LO}) G_{\beta,\alpha}^{a,b}(\hbar \omega - \hbar \omega_{LO}) + N_{LO} G_{\beta,\alpha}^{a,b}(\hbar \omega + \hbar \omega_{LO}) \right] \right. \\
+ f^e_{\beta} \left[ (1 + N_{LO}) G_{\beta,\alpha}^{a,b}(\hbar \omega + \hbar \omega_{LO}) + N_{LO} G_{\beta,\alpha}^{a,b}(\hbar \omega - \hbar \omega_{LO}) \right] \right\}
\]

and

\[
\Gamma_{\alpha}^{OD}(\hbar \omega) = i \sum_{a,b,e,h}^{b \neq a} \frac{M_{LO}^2}{\varepsilon^2/\varepsilon_0} V_{\alpha \beta \alpha \beta} \left\{ (1 - f^e_{\beta}) \left[ (1 + N_{LO}) G_{\beta,\alpha}^{b,a}(\hbar \omega - \hbar \omega_{LO}) + N_{LO} G_{\beta,\alpha}^{b,a}(\hbar \omega + \hbar \omega_{LO}) \right] \right. \\
+ f^e_{\beta} \left[ (1 + N_{LO}) G_{\beta,\alpha}^{b,a}(\hbar \omega + \hbar \omega_{LO}) + N_{LO} G_{\beta,\alpha}^{b,a}(\hbar \omega - \hbar \omega_{LO}) \right] \right\}
\]

where the prefactor \( M_{LO}^2 = 4\pi\alpha \frac{\hbar}{\sqrt{2m}} (\hbar \omega_{LO})^{3/2} \) includes the polar coupling strength \( \alpha \), the reduced mass \( m \) and the phonon energy \( \hbar \omega_{LO} \). In this equations we have introduced

\[
G_{\beta,\alpha}^{a,b}(\omega) = \int d\tau e^{i\omega \tau} G_{\beta}^{a,ret}(\tau) G_{\alpha}^{b,ret}(\tau)
\]

which represents the Fourier transform of a product of two retarded polaronic GFs. Please note that the structure of this quantity can be traced back to the change from the \( e, v \) picture to the \( e, h \) picture. Through these functions polaronic renormalization effects such as phonon replicas and hybridization between the localized states [14] are included in Eqs. (10) and (11). Details for the calculation of the retarded polaronic GF are given in Appendix B.

B. Results

The high-density spectra with dephasing due to Coulomb interaction exhibit only three resonances, namely s-shell, p-shell and the excitonic resonance of the WL. Absorption spectra with correlation contributions due to interaction of carriers with LO phonons are shown in Fig. 5. Polaronic renormalizations of the single-particle states lead to a more complicated resonance structure for the interband transitions displayed in the inset of Fig. 5. One can identify phonon replicas and results of the hybridization of the single-particle states. For example, the s-shell resonances has a shoulder on the lower energetic side due to hybridization of the corresponding electron state. Energetically above the resonances of the s-shell and the p-shell several peaks due to phonon replicas and their hybridizations can be observed. The non-Lorentzian character of the lineshapes is even more pronounced as for the spectra with Coulomb interaction. On the other hand, the damping of the resonances at higher densities remains weak.

V. DEPHASING DUE TO CARRIER-PHONON AND COULOMB INTERACTION

In this section, we investigate the combined influence of the carrier-carrier Coulomb scattering and the interaction of carrier with LO phonons. It turns out that polaronic resonances in the single-particle spectral function are strongly damped out due to Coulomb scattering of carriers even at low carrier densities, so that a pole
approximation for the single-particle properties of the interacting system is reasonable. On the other hand, interband energy renormalizations and dephasing contributions due to the interaction of carriers with LO phonons continue to be important. Since our goal is a calculation of optical spectra for the QD-WL system under the influence of correlation effects at elevated carrier densities, we use for the description of single-particle properties a retarded GF obeying Eq. (B1) with the free carrier energy in the L.H.S. replaced by $\tilde{\epsilon}_\alpha = \epsilon_\alpha + \Delta_\alpha - i\gamma_\alpha$, which is determined by Eqs. (7) and (8). In other words the polaron is obtained by dressing with the phonon interaction not the free particles but the quasi-particles obtained by Coulomb renormalization. Rewriting the retarded GF as

$$G_{\alpha}^{\text{ret}}(\tau) = G_{\alpha}^{\text{a}}(\tau) e^{-\frac{i}{\hbar}(\Delta_\alpha - i\gamma_\alpha)\tau}. \tag{13}$$

we separate in $G_{\alpha}^{\text{a}}(\tau)$ the phonon renormalization effects which are, of course, influenced by the presence of the Coulomb interaction. (The equation obeyed by $G$ still contains $\Delta$ and $\gamma$.) The finite life-time of these quasi-particles produces in general sufficient damping to reduce the polaronic GF $G_{\alpha}^{\text{a}}$ to a single-pole structure. This pole is used instead of $\epsilon_\alpha$ in Eq. (6). In this way the Coulombian and the polaronic problems become coupled and have to be solved self-consistently. The iterative solution to this problem converges rapidly. Results of the spectral function of QD and WL states are shown in Fig. 6. Even for low carrier densities, polaronic structures are strongly broadened as a result of the dominant role of damping due to Coulomb scattering. (For low carrier densities the exponential decay $e^{-\frac{2\pi}{\hbar}\tau}$ due to Coulomb interaction, which is superimposed to the polaronic function $G_{\alpha}^{\text{a}}(\tau)$ in Eq. (13), might somewhat overestimate the damping of the polaronic resonances. Nevertheless at higher carrier densities we expect a strong broadening of the polaron satellites.)

In Fig. 7 calculated absorption spectra are shown, which include correlations due to carrier-carrier scattering Eqs. (6)-(11), and interaction with LO-phonons, Eqs. (13)-(14), both evaluated with self-consistently renormalized single-particle energies. As in the result for dephasing due to Coulomb interaction, Fig. 11 we see the bleaching and red-shift of the resonances due to many-body interactions and also the saturation of the s-shell gain. Although we observe that the Coulomb interaction is the clearly dominant dephasing mechanism for high carrier densities, we also infer from a comparison of Fig. 8 with Fig. 11 that even in the gain regime the electron-phonon interaction gives rise to a clear increase in the dephasing. Nevertheless polaronic features are absent in the spectra, since the complicated multi-peak structure of the spectral function is completely damped out due to Coulomb effects.

For intermediate carrier densities around $5 \times 10^{10}$ cm$^{-2}$ both types of interaction processes are equally important.
Comparing the results in Fig. 8, one can conclude for our situation that taking only the Coulomb dephasing mechanism into account underestimates the dephasing of the ground state transition by roughly a factor of two, while the damping of the WL is even dominated by carrier-phonon interaction. For higher carrier densities, however, this picture changes as can be seen in Fig. 9. Using a carrier density of $2 \times 10^{11}$ cm$^{-2}$, the Coulomb interaction is clearly the dominant mechanism for the QD resonances. For the excitonic resonance of the WL, the two mechanisms are equally important even at this rather high carrier density, where we are already in the gain regime for the s-shell transition.

**Comparison with Experiments**

In most experiments, the emission from an ensemble of QDs is studied such that the additional inhomogeneous broadening of the QD resonances due to size and composition fluctuations contributes. However, recent experiments [20, 21] have been performed at room temperature on single QDs. In Fig. 1(a) of Ref. [21] optical spectra are displayed for different excitation intensities. Although a direct comparison of the excitation dependence is not possible, due to uncertainties in the carrier densities generated in those experiments, the general features of the spectra are identical. A clear distinction between the QD transitions and the excitonic resonance of the WL is seen. The QD resonances show a density dependent bleaching and a pronounced red shift due to many-body correlations while the spectral position of the WL is almost unchanged and the WL resonance is only bleached out. This means that the red-shift of the QD resonances cannot be attributed to band-gap shrinkage effects of the WL.
The observed homogeneous linewidth in this experiment is 8-13meV depending on the excitation intensity. This is larger than our findings, but one has to take into account that the QD, investigated in Ref. [21], seems to have three confined electronic shells which results in more dephasing channels. A better comparison is possible with the results of Ref. [20] because two types of QDs are investigated, where one type resembles our model in the respect that it is supposed to have two confined electronic shells. Regarding the s-shell, we can infer from Fig. 3 of Ref. [20] that the observed homogeneous linewidth is slightly above 3meV at room temperature which is similar to our result of 3.2meV. The calculated value is practically unchanged for a carrier density range from $5 \times 10^8$ to $10^{10}$ cm$^{-2}$.

For a better comparison with experiments, we have also calculated the temperature dependence of the homogeneous linewidth in a temperature range in which the influence of LA-phonons remains small. As shown in Fig. 10 we obtain for low carrier densities (where carrier-carrier scattering is sufficiently weak) the expected reduction of dephasing with decreasing temperature as seen in Ref. [20]. (A quantitative comparison would require a better knowledge of QD parameters and phonon modes in the QD-WL system.) For the case of a higher excitation density of $10^{10}$ cm$^{-2}$, the line-broadening is almost temperature independent due to a balancing of different scattering channels. The dephasing due to LO-phonons decreases with temperature as in the low-density case. However, the Coulomb interaction is no longer negligible for this carrier density. With decreasing temperature, the WL states are less populated while the occupation of QD states increases. This enhances intra-dot relaxation processes and provides stronger dephasing due to Coulomb interaction.

![FIG. 10: Temperature dependence of the linewidth (full halfwidth) for the QD ground state resonance and different carrier densities.](image)

VI. CONCLUSION

We have calculated the excitation induced dephasing and lineshifts for a QD-WL system on a microscopic basis. Both Coulomb and LO-phonon contributions to the homogeneous linewidths are found to be equally important for elevated carrier densities. The role of self-consistent single-particle energy renormalizations in the scattering integrals is emphasized. For the Coulomb interaction the relative importance of various scattering channels has been analyzed.

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APPENDIX A: QUANTUM DOT MODEL SYSTEM AND COULOMB MATRIX ELEMENTS

We focus on lens-shaped InGaAs QDs which are located on top of a two dimensional WL. Such systems typically consist of localized QD states energetically below a quasi-continuum of delocalized WL states for electrons and holes. In the present paper we assume the single particle spectrum as depicted in Fig. 1 of Ref. [27]. For the in plane motion we assume a parabolic confinement potential, leading to harmonic oscillator like single particle wavefunctions. To account for the finite depth of the confinement potential, we take into account two confined shells for electrons and holes, which we refer to as s-shell for the ground state and p-shell for the double degenerated excited state, due to their angular momentum properties. We choose a level spacing of 40meV for electrons and 15meV for holes so that the free carrier transitions appear at -55meV relative to the WL bandedge for the p-shell and at -110meV for the s-shell. The WL states are modeled by orthogonalized plane waves (OPW) [27]. For the motion in growth direction (z-direction) we assume an infinite height potential, leading to harmonic oscillator like single particle spectrum as depicted in Fig. 1 of Ref. [27].

Under this assumptions we can construct the Coulomb matrix elements

$$V_{\alpha\beta\gamma\delta} = \frac{1}{A} \sum_q V_q$$

$$\times \int d^2 \varphi \varphi^* \varphi^* \varphi^* e^{-iq\cdot\varphi}$$

$$\times \int d^2 \varphi' \varphi^* \varphi^* \varphi^* e^{iq\cdot\varphi'} \; , \quad (A1)$$

consisting of overlap integrals between single-particle wavefunctions and the Coulomb potential $V_q$ as given in Ref. [27]. For the screening we use a generalization of the static Lindhard formula which is also explained in detail in Ref. [27]. This procedure leads to the replacement
$V_q \to W_q$ in Eq. A1 for the matrix elements $W_{\alpha \beta \gamma \delta}$. Because we are working in the envelope approximation with equal envelope wavefunctions, we do not need to consider band indices for the Coulomb matrix elements. All other parameters are chosen as in Ref. 27.

**APPENDIX B: RETARDED POLARONIC GREEN’S FUNCTION**

The retarded polaronic GFs obey the equation of motion

$$
\left[ i\hbar \frac{\partial}{\partial \tau} - e_{\alpha}^a \right] G_{\alpha}^{\text{ret}}(\tau) = \delta(\tau)
+ \int d\tau' \Sigma_{\alpha}^{\text{ret}}(\tau - \tau') G_{\alpha}^{\text{ret}}(\tau') .
$$

(B1)

The corresponding retarded self-energy is given in RPA by

$$
\Sigma_{\alpha}^{\text{ret}}(\tau) = i\hbar \sum_{\beta} G_{\beta \alpha}^{\text{ret}}(\tau) D_{\beta \alpha}^{\text{ret}}(-\tau) .
$$

(B2)

We assume for the calculations restricted to electron-phonon interaction that the polaronic retarded GF is not influenced by population effects. This has been verified for a bulk system in Ref. 28.

Assuming that the phonon system is in thermal equilibrium, the phonon propagator (combined with the interaction matrix elements) is given by

$$
i\hbar D_{\beta \alpha}^{\text{LO}}(\tau) = M^2 F_{\beta \alpha \gamma \delta} V_{\beta \alpha \gamma \delta}
\times \left[ N_{\text{LO}} e^{-i\omega_{\text{LO}}\tau} + (1 + N_{\text{LO}}) e^{i\omega_{\text{LO}}\tau} \right] .
$$

(B3)

In the following we explicitly consider the direct e-e and h-h interaction contributions of the diagonal dephasing, which corresponds to Eq. (4), we obtain

$$
S_{\alpha}(t) = \frac{1}{\hbar^2} \sum_{a,b=\epsilon,h} \sum_{\gamma \delta \alpha \beta} 2W_{\alpha \beta \gamma \delta} W_{\alpha \beta \gamma \delta}^* F_{\alpha \beta \gamma \delta} (t) .
$$

(C2)

**APPENDIX C: CONNECTION BETWEEN MEMORY EFFECTS AND FREQUENCY DEPENDENCE IN THE SCATTERING INTEGRALS**

In this section we is show how the non-Markovian (Markovian) scattering integrals lead to frequency dependent (independent) dephasing terms. The time domain formulation of the SBE with correlation contributions due to Coulomb interaction is given by 22

$$
(i\hbar \frac{\partial}{\partial t} - e_{\alpha}^{\text{HF}} - e_{\alpha}^{\text{H}} \psi_{\alpha}(t) + [1 - f_{\alpha}^e - f_{\alpha}^h] \Omega_{\alpha}^{\text{HF}}(t) = -i\hbar S_{\alpha}^{\text{Con}}(t) .
$$

(C1)

We assume static screening where the Coulomb matrix elements depend only parametrically on time via the population functions. For the discussed excitation conditions both $f$ and $W$ are time independent. With the ansatz

$$
G_{\alpha}^{\text{ret}}(t,t') = \frac{i}{\hbar} \Theta(t - t')e^{-\frac{\tau}{\hbar}} e_{\alpha}^{\text{H}}(t-t') ,
$$

(C4)
With the Fourier transform \( \psi_\alpha(t) = \int \frac{d\omega}{2\pi} e^{-i\omega t} \psi_\alpha(\omega) \) and using the integral relation

\[
\int_{-\infty}^{t} dt' e^{i\Delta(t-t')} = \frac{i\hbar}{\Delta}, \quad \text{Im}\Delta > 0 \quad (C5)
\]

we arrive at Eq. 1 and the terms of Eq. 3 which are discussed here.

For the Markov approximation one separates from the transition amplitude a phase factor, which rapidly oscillates with the (renormalized) interband transition energy, via the ansatz

\[
\psi_\alpha(t) = e^{\frac{i}{\hbar} \left( \varepsilon_\alpha^a + \varepsilon_\alpha^b \right) t} \tilde{\psi}_\alpha(t). \quad (C6)
\]

Now one assumes that a weakly time dependent \( \tilde{\psi}_\alpha(t') \) can be replaced by the transition amplitude at the actual time \( t \), \( \tilde{\psi}_\alpha(t) \), thus neglecting the memory effects of \( \tilde{\psi}_\alpha \). Using the integral relation (C5) again, one arrives at an expression similar to Eq. 3 but with

\[
g \left( \hbar \omega - \tilde{\varepsilon}_\alpha^a - \tilde{\varepsilon}_\alpha^b - \tilde{\varepsilon}_\alpha^c + \left( \tilde{\varepsilon}_\alpha^a \right)^* \tilde{\varepsilon}_\alpha^d - \tilde{\varepsilon}_\alpha^e \right)
\]

replaced by

\[
g \left( \tilde{\varepsilon}_\alpha^a - \tilde{\varepsilon}_\alpha^b + \left( \tilde{\varepsilon}_\alpha^a \right)^* \tilde{\varepsilon}_\alpha^d - \tilde{\varepsilon}_\alpha^e \right).
\]

[1] Y. Masumoto and T. Takagahara, eds., *Semiconductor Quantum Dots* (Springer-Verlag, Berlin, 2002), 1st ed.
[2] P. Michler, ed., *Single Quantum Dots* (Springer-Verlag, Berlin, 2003), 1st ed.
[3] P. Michler, A. Imamoglu, M. D. Mason, P. J. Carson, G. F. Strouse, and S. K. Buratto, Nature 406, 968 (2000).
[4] E. Moreau, I. Robert, L. Manin, V. Thierry-Mieg, J. M. Gerard, and I. Abram, Phys. Rev. Lett. 87, 183601 (2001).
[5] M. Pelton, C. Santori, J. Vuckovic, B. Zhang, G. S. Solomon, J. Plant, and Y. Yamamoto, Phys. Rev. Lett. 89, 236302 (2002).
[6] J. P. Reithmaier, G. Sek, A. Löffler, C. Hofmann, S. Kuhn, S. Reitzenstein, L. V. Keldysh, V. D. Kulakovskii, T. L. Reinecke, and A. Forchel, Nature 432, 197 (2004).
[7] T. Yoshie, A. Scherer, J. Hendrickson, G. Khitrova, H. M. Gibbs, G. Rupper, C. Ell, O. B. Shchekin, and D. G. Deppe, Nature 432, 200 (2004).
[8] Y. Z. Hu, H. Gießen, N. Peyghambarian, and S. W. Koch, Phys. Rev. B 53, 4814 (1996).
[9] H. C. Schneider, W. W. Chow, and S. W. Koch, Phys. Rev. B 64, 115315 (2001).
[10] H. C. Schneider, W. W. Chow, and S. W. Koch, Phys. Rev. B 70, 235308 (2004).
[11] G. Manzke and K. Henneberger, phys. stat. sol. (b) 234, 233 (2002).
[12] A. V. Uskov, A.-P. Jauho, B. Tromborg, J. Mørk, and R. Lang, Phys. Rev. Lett. 85, 1516 (2000).
[13] B. Krummheuer, V. M. Axt, and T. Kuhn, Phys. Rev. B 65, 195313 (2002).
[14] E. A. Muljarov and R. Zimmermann, Phys. Rev. Lett. 85, 1516 (2000).
[15] P. Borri, W. Langbein, S. Schneider, U. Woggon, R. L. Sellin, D. Ouyang, and D. Bimberg, Phys. Rev. Lett. 87, 157401 (2001).
[16] H. Htoon, D. Kulik, O. Baklenov, A. L. H. Jr., T. Takagahara, and C. K. Shih, Phys. Rev. B 63, 241303(R) (2001).
[17] R. Oulton, J. J. Finley, A. I. Tartakovskii, D. J. Mowbray, M. S. Skolnick, M. Hopkinson, A. Vasanelli, R. Ferreira, and G. Bastard, Phys. Rev. B 68, 235301 (2003).
[18] T. Inoshita and H. Sakaki, Phys. Rev. B 56, 4355 (1997).
[19] J. Seebeck, T. R. Nielsen, P. Gartner, and F. Jahnke, Phys. Rev. B 71, 125327 (2005).
[20] M. Bayer and A. Forchel, Phys. Rev. B 65, 041308(R) (2002).
[21] K. Matsuda, K. Ikeda, T. Saiki, H. Saito, and K. Nishi, Appl. Phys. Lett. 83, 2250 (2003).
[22] H. Haug and S. W. Koch, *Quantum Theory of the Optical and Electronic Properties of Semiconductors* (World Scientific Publ., Singapore, 1994), 3rd ed.
[23] F. Jahnke, M. Kira, and S. W. Koch, Z. Physik B 104, 559 (1997).
[24] T. R. Nielsen, P. Gartner, M. Lorke, J. Seebeck, and F. Jahnke, Phys. Rev. B 72, 235311 (2005).
[25] P. Lipavský, V. Špička, and B. Velický, Phys. Rev. B 34, 6933 (1986).
[26] H. C. Tso and N. J. Morgenstern Horing, Phys. Rev. B 69, 235314 (2004).
[27] P. Gartner, L. Bányai, and H. Haug, Phys. Rev. B 60, 14234 (1999).