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Luminescence of a semiconductor quantum dot system

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Abstract. A microscopic theory is used to study photoluminescence of semiconductor quantum dots under the influence of Coulomb and carrier-photon correlation effects beyond the Hartree-Fock level. We investigate the emission spectrum and the decay properties of the time-resolved luminescence from initially excited quantum dots. The influence of the correlations is included within a cluster expansion scheme up to the singlet-doublet level.

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1 Introduction

Semiconductor quantum dots (QDs) attract considerable attention due to their potential for fundamental studies as well as device applications, such as cavity-quantum electrodynamics, lasers and non-classical light sources [1,2]. QDs allow for a carrier confinement in all three dimensions with a discrete atomic-like density of states corresponding to the localized states. Carriers in such "artificial atoms" are often described in terms of excitons, i. e., fully correlated electron-hole pairs. However, this simplified picture should be treated with care since the elementary quasi-particles in semiconductors are electrons and holes, and the degree of correlations depends on the many-body Coulomb interaction and on the carrier-photon interaction.

The influence of Coulomb-correlated multi-exciton states on optical spectra of QDs has been investigated by several groups [3–9]. Much less is known about the influence of correlations on the spontaneous recombination dynamics. Time-resolved photoluminescence (PL) measurements provide direct access to the efficiency of carrier scattering processes after optical excitation with short pulses [10] and to the modification of the spontaneous emission lifetime for QDs in optical cavities due to the Purcell effect [2,11,12]. The theoretical description is rather challenging, because it requires not only a computation of carrier scattering and correlations, but also a full quantum mechanical treatment of the light field. One promising approach for the description of PL is the equation of motion technique [13]. The resulting semiconductor luminescence equations (SLE) have previously been used to study PL spectra [14] and exciton formation [15] in a quantum well system, and the PL decay dynamics of an ensemble of QDs embedded into a microcavity [16]. It is well-known that the equation of motion technique leads to a hierarchy of correlation functions due to the Coulomb interaction between carriers and due to the carrier-photon interaction. A systematic way to truncate this hierarchy is the cluster expansion method [17].

In this paper, we apply the equation of motion technique to QDs coupled to a quantized light field in free space where correlations are included up to the so-called singlet-doublet level. Our numerical results show that it is necessary to go beyond the singlet level in order to obtain well-defined positive luminescence spectra. Moreover, we demonstrate that the higher order correlations can have a strong impact on the luminescence decay dynamics.

The paper is organized as follows: In Section 2 our model system and the Hamiltonian are introduced. In Section 3 the equations of motion for the carriers in the QDs and the quantized light field are derived. The PL spectrum is studied in Section 4 and the PL decay is investigated in Section 5.

2 System and Hamiltonian

We consider self-assembled QDs, where the discrete states, corresponding to three-dimensional carrier confinement, are located energetically below a quasi-continuum of delocalized states, which corresponds to the two-dimensional motion in a wetting layer (WL). To study PL, the system may be off-resonantly excited by an optical pulse which creates carriers in the WL, from where the carriers relax

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quickly into the localized QD states [18,19]. At low temperatures, the carriers populate solely the QD states. Then the WL states are mainly important for carrier-scattering processes if the excitation involves the quasi-continuum. For the recombination dynamics due to carrier-photon interaction, the unpopulated WL states are of negligible importance and are therefore not included in our calculation. Coulomb correlations between the discrete QD states and the energetically displaced quasi-continuum of the WL are much weaker than those between QD states.

In order to study PL, we treat both the carrier dynamics and the electromagnetic field quantum-mechanically. The total Hamiltonian for the system has the following contributions:

$$H = H^0_{\text{carr}} + H_{\text{Coul}} + H_{\text{ph}} + H_{\text{D}}. \tag{1}$$

The Hamiltonian which describes the system of valence- and conduction-band electrons interacting via Coulomb interaction has the two parts

$$H^0_{\text{carr}} = \sum_\nu \varepsilon^c_\nu \, c^\dagger_\nu c_\nu + \sum_\nu \varepsilon^v_\nu \, v^\dagger_\nu v_\nu, \tag{2}$$

$$H_{\text{Coul}} = \frac{1}{2} \sum_{\alpha'\nu\nu'\alpha} \left[V^{cc}_{\alpha'\nu,\nu'\alpha} \, c^\dagger_{\alpha'} c^\dagger_\nu c_{\nu'} c_\alpha + V^{vv}_{\alpha'\nu,\nu'\alpha} \, v^\dagger_{\alpha'} v^\dagger_\nu v_{\nu'} v_\alpha \right] + \sum_{\alpha'\nu\nu'\alpha} V^{cv}_{\alpha'\nu,\nu'\alpha} \, c^\dagger_{\alpha'} v^\dagger_\nu v_{\nu'} c_\alpha. \tag{3}$$

The free Hamiltonian H^0_{carr} contains information about the single-particle spectrum $\varepsilon^{c,v}_\nu$ and describes a system of non-interacting charge carriers. The Coulomb interaction between the carriers is accounted for in H_{Coul}. The operators c_ν ($c^\dagger_\nu$) annihilate (create) electrons in the one-particle states $|\nu\rangle$ of energy ε^c_ν. The corresponding operators and single-particle energies for valence-band electrons are v_ν ($v^\dagger_\nu$) and ε^v_ν, respectively. The explicit form of the single-particle wave function $\langle \mathbf{r}|\nu,\lambda\rangle = \psi_{\nu,\lambda}(\mathbf{r})$ enters the description via the Coulomb matrix elements

$$V^{\lambda\lambda'}_{\alpha'\nu,\nu'\alpha} = \int \mathrm{d}^3r \int \mathrm{d}^3r' \; \psi^{\lambda *}_{\alpha'}(\mathbf{r}) \psi^{\lambda' *}_{\nu}(\mathbf{r}') V(\mathbf{r}-\mathbf{r}') \psi^{\lambda'}_{\nu'}(\mathbf{r}') \psi^{\lambda}_{\alpha}(\mathbf{r}) \tag{4}$$

with the band index $\lambda = c, v$ and the Coulomb potential $V(\mathbf{r}) = e^2/4\pi\epsilon_0\epsilon r$. The dielectric constants of the vacuum and the background material are given by ϵ_0 and ϵ, respectively.

Since the main physical effects discussed below arise from the many-body interaction, we consider a simple model for the single-particle states. It has been shown that in the case of flat, cylindrically symmetric QDs the single-particle bound-state wave functions in the plane of larger extension (perpendicular to the growth direction) are well approximated by those of a two-dimensional harmonic oscillator [20]. Due to the rotational symmetry around the QD axis, the corresponding angular momentum is a good quantum number. We consider the first two confined shells of such a system, which are denoted by s and p according to their in-plane symmetry. The s-shell is only spin-degenerate, while the p-shell has an additional angular-momentum two-fold degeneracy. To account for the strong confinement in growth direction, we use an infinite potential well to model the corresponding finite extension of the wave-function. Only the energetically lowest state due to the confinement in growth direction will be considered. Further details of the QD model are discussed in [18], where Figure 1 displays the considered energy levels.

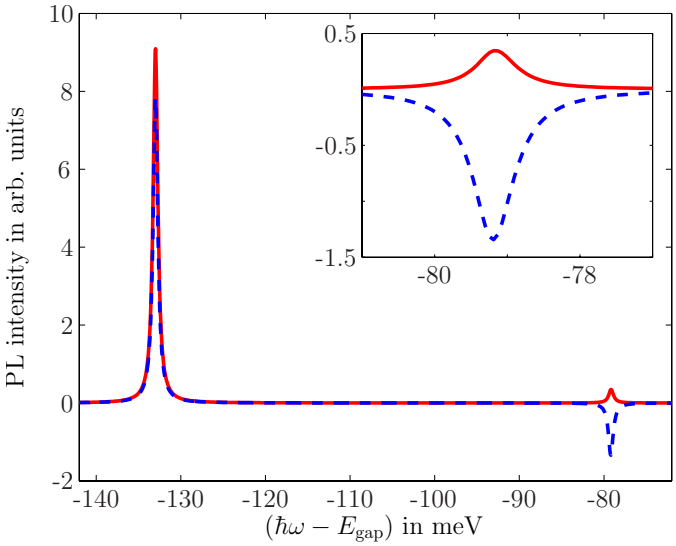

Fig. 1. PL spectrum after 50 ps of time evolution. The solid line corresponds to the full calculation, the dashed line to the singlet (Hartree-Fock) factorisation. The inset shows a magnification of the intensity at the p-shell.

We use a quantum mechanical description of the electromagnetic field to formulate the theory of QD luminescence. The free electromagnetic field is given by the Hamiltonian [21]

$$H_{\text{ph}} = \sum_\xi \hbar\omega_\xi \left(b^\dagger_\xi b_\xi + \frac{1}{2} \right), \tag{5}$$

where ξ labels the mode, $\hbar\omega_\xi$ is the photon energy, and the operators $b^\dagger_\xi$ (b_ξ) create (destroy) a photon in the mode ξ. In free space, the label ξ contains the wave vector $\mathbf{q}$ and the polarization vector of the electromagnetic field $\mathbf{e}_{\text{p}}(\mathbf{q})$, with the index $\text{p} = \pm$. The mode frequencies are then given by $\omega_\xi = c|\mathbf{q}|$, with c being the speed of light, and the explicit form of the modes is $\mathbf{U}_\xi(\mathbf{r}) = \mathbf{e}_{\text{p}}(\mathbf{q}) e^{i\sqrt{\epsilon}\,\mathbf{qr}}$.

The light-matter interaction Hamiltonian in dipole approximation reads

$$H_{\text{D}} = -i \sum_{\xi,\,\alpha\nu} \left(g_{\xi\alpha\nu} \, c^\dagger_\alpha v_\nu b_\xi + g_{\xi\alpha\nu} \, v^\dagger_\alpha c_\nu b_\xi \right) - \text{h.c.} \tag{6}$$

The resonant elementary process associated with this Hamiltonian is the transition of an electron from the valence into the conduction band (or vice versa) by absorption (emission) of a photon. The matrix elements $g_{\xi\alpha\nu}$, which describe the coupling between the mode ξ of the electromagnetic field and the carrier transition between

states $|\alpha\rangle$ and $|\nu\rangle$, are given by

$$g_{\xi\alpha\nu} = E_\xi \int d^3r\ \psi_\alpha^{c*}(\mathbf{r}) e\mathbf{r}\mathbf{U}_\xi(\mathbf{r})\psi_\nu^v(\mathbf{r})\ , \tag{7}$$

where $E_\xi = \sqrt{\hbar\omega_\xi/2\epsilon\epsilon_0 V}$ and V is the normalization volume. In envelope-function approximation [22] the wavefunction $\psi_\alpha^c(\mathbf{r})$ and $\psi_\nu^v(\mathbf{r})$ can now be decomposed into an envelope part, which varies only slightly over a unit cell, and the rapidly oscillating Bloch-factor $u_{\mathbf{k}\approx 0}(\mathbf{r})$. Taking into account that the electromagnetic field is approximately constant over the extent of a QD and considering equal envelopes for the conduction- and valence-band electrons, one finds

$$g_{\xi\alpha\nu} = E_\xi \mathbf{d}_{cv}\mathbf{U}_\xi(\mathbf{r}_0)\delta_{\alpha\nu} \equiv g_{\xi\nu}\delta_{\alpha\nu}. \tag{8}$$

Here, $\mathbf{d}_{cv}$ are the interband matrix elements and $\mathbf{r}_0$ is the position of the QD. From equation (8) it follows that, within the envelope-function approximation, optical transitions occur only between the s-shells or the $p_\pm$-shells of electrons in the valence and conduction band.

3 QD semiconductor luminescence equations

The aim of our description is the calculation of the time-resolved PL and the corresponding emission spectrum. We assume that PL takes place in the incoherent regime where the influence of a coherent polarization can be neglected. Examples are incoherent carrier excitations or coherent excitation of higher states with rapid dephasing and carrier relaxation. We determine operator averages within the rotating wave approximation using the equation of motion technique [13]. It is a well-known difficulty that the equation of motion of an average of N operators couples to $N+2$ operator averages due to the two-particle interaction Hamiltonian (3). In order to numerically calculate operator averages, this hierarchy of equations must be truncated in an unambiguous way. We use the cluster expansion method [17], which has previously been applied in the microscopic description of quantum wells [15], where one directly formulates equations of motion for correlations. The correlation of an operator average with N operators is obtained by subtracting all possible factorisations into correlations of maximal $N-2$ operators. This way one ensures that correlations of a certain order contain no lower order correlation effects. The truncation is then applied to all correlations containing more than $N_{\max}$ operators [17]. In this work, we use the so-called singlet-doublet truncation scheme, i. e., we include correlations with up to four operators. Schematically, denoting the fermionic operators c, v by a, we obtain equations of the structure

$$\frac{d}{dt}\langle a^\dagger a\rangle\Big|_{\mathrm{H_{Coul}}} \propto \langle a^\dagger a^\dagger aa\rangle\ , \tag{9}$$

where we factorize into singlets and introduce the correlation, denoted by δ, of four operators according to

$$\begin{aligned}\langle a^\dagger a^\dagger aa\rangle &= \langle a^\dagger a^\dagger aa\rangle_{\mathrm{HF}} + \delta\langle a^\dagger a^\dagger aa\rangle \\ &= \langle a^\dagger a\rangle\langle a^\dagger a\rangle + \delta\langle a^\dagger a^\dagger aa\rangle.\end{aligned} \tag{10}$$

For a four-operator average, the singlet factorization corresponds to the Hartree-Fock approximation. The equation of motion for the four-operator correlation leads to averages of six operators, which we factorize according to

$$\begin{aligned}\langle a^\dagger a^\dagger a^\dagger aaa\rangle = \langle a^\dagger a\rangle\langle a^\dagger a\rangle\langle a^\dagger a\rangle + \langle a^\dagger a\rangle\,\delta\langle a^\dagger a^\dagger aa\rangle \\ + \delta\langle a^\dagger a^\dagger a^\dagger aaa\rangle\end{aligned} \tag{11}$$

into singlet, singlet-doublet and triplet contributions. Note that all possible combinations of averages and correlations must be taken. Truncating at singlet-doublet level corresponds to neglecting the last term, and consequently higher-order correlations. We also encounter averages of fermionic and bosonic operators. Realizing that a photon operator b ($b^\dagger$) corresponds to *two*-carrier operators [23], as can be seen from formally solving the equation of motion for a single photon operator, we can apply the truncation scheme to mixed operator averages in a straightforward way.

Writing down Heisenberg's equation of motion for the photon number, we find

$$i\hbar\frac{d}{dt}\langle b_\xi^\dagger b_\xi\rangle = 2i\,\mathrm{Re}\sum_\nu g_{\xi\nu}^*\langle b_\xi^\dagger v_\nu^\dagger c_\nu\rangle\ , \tag{12}$$

which couples to the photon-assisted polarization amplitude $\langle b_\xi^\dagger v_\nu^\dagger c_\nu\rangle$. The corresponding equation of motion is given by

$$\begin{aligned}i\hbar\frac{d}{dt}\langle b_\xi^\dagger v_\nu^\dagger c_\nu\rangle = (\tilde\varepsilon_\nu^c - \tilde\varepsilon_\nu^v - \hbar\omega_\xi - i\Gamma)\langle b_\xi^\dagger v_\nu^\dagger c_\nu\rangle \\ + (f_\nu^c - f_\nu^v)\sum_\alpha V_{\nu\alpha\nu\alpha}\langle b_\xi^\dagger v_\alpha^\dagger c_\alpha\rangle \\ + i\,g_{\xi\nu}f_\nu^c(1-f_\nu^v) + i\sum_\alpha g_{\xi\alpha}C^{\mathrm{x}}_{\alpha\nu\nu\alpha}.\end{aligned} \tag{13}$$

The free evolution is determined by the renormalized energies $\tilde\varepsilon_\nu^c = \varepsilon_\nu^c - \sum_\alpha V_{\nu\alpha\nu\alpha}f_\alpha^c$ and $\tilde\varepsilon_\nu^v = \varepsilon_\nu^v - \sum_\alpha V_{\nu\alpha\nu\alpha}f_\alpha^v$, the resonance frequency ω_ξ of the optical mode ξ, and a phenomenological dephasing Γ which broadens the spectral lines. The term in the second line is analogous to the quantum well case, where it gives rise to the excitonic PL below the band gap [13]. Here it introduces the corresponding excitonic resonances for the QD states due to the interband Coulomb exchange interaction. The source term of spontaneous emission in the last line enters the theory naturally due to the quantization of the light field. We have omitted the term representing stimulated emission/absorption, which contributes for example if an external field is resonant with the considered transitions or if a resonator provides feedback for the emitted photons [16]. Before evaluating the correlation term in the last line of equation (13), we give the time evolution of the

carrier population

$$i\hbar\frac{d}{dt}f_\nu^c = -2i\ \mathrm{Re}\sum_\xi g_{\xi\nu}^*\langle b_\xi^\dagger v_\nu^\dagger c_\nu\rangle + 2i\ \mathrm{Im}\sum_{\alpha\alpha'\nu'} V_{\nu\alpha'\alpha\nu'}\left(C^c_{\nu\alpha'\alpha\nu'} - C^{\mathrm{x}}_{\alpha'\nu\alpha\nu'}\right) \tag{14}$$

$$i\hbar\frac{d}{dt}f_\nu^v = 2i\ \mathrm{Re}\sum_\xi g_{\xi\nu}^*\langle b_\xi^\dagger v_\nu^\dagger c_\nu\rangle - 2i\ \mathrm{Im}\sum_{\alpha\alpha'\nu'} V_{\nu\alpha'\alpha\nu'}\left(C^v_{\nu\alpha'\alpha\nu'} - C^{\mathrm{x}}_{\alpha'\nu\alpha\nu'}\right). \tag{15}$$

In the following, we consider s-states with zero angular momentum and p-states with angular momentum of ±1, which, as we now explain, allows us to take $\langle a_\nu^\dagger a_{\nu'}\rangle = f_\nu^a\,\delta_{\nu\nu'}$. Initially all expectation values but the population in the s- and p-shells are set to zero. The rotational symmetry of the system and the resulting conservation of angular momentum ensures that all off-diagonal terms $\langle a_\nu^\dagger a_{\nu'}\rangle$ with $\nu \neq \nu'$ describe forbidden transitions and remain zero during the time evolution. Therefore, in all equations expectation values of two carrier operators are restricted to populations. An inclusion of higher angular momentum states is straightforward, but unnecessary at low temperatures and left out for transparency. Furthermore, polarisation-like averages of the form $\langle v_\nu^\dagger c_\nu\rangle$ vanish in the incoherent regime [13].

The interband correlations are defined according to equation (10) as

$$\begin{aligned} C^{\mathrm{x}}_{\alpha'\nu\nu'\alpha} &= \delta\langle c_{\alpha'}^\dagger v_\nu^\dagger c_{\nu'} v_\alpha\rangle \\ &= \langle c_{\alpha'}^\dagger v_\nu^\dagger c_{\nu'} v_\alpha\rangle - \langle c_{\alpha'}^\dagger v_\nu^\dagger c_{\nu'} v_\alpha\rangle_{\mathrm{HF}} \\ &= \langle c_{\alpha'}^\dagger v_\nu^\dagger c_{\nu'} v_\alpha\rangle + f_{\nu'}^c f_\nu^v\,\delta_{\nu\alpha}\delta_{\nu'\alpha'}\,. \end{aligned} \tag{16}$$

The time evolution of the interband correlations are given by the equation of motion

$$\begin{aligned} i\hbar\frac{d}{dt}C^{\mathrm{x}}_{\alpha'\nu\nu'\alpha} &= -\left(\varepsilon^c_{\alpha'} + \varepsilon^v_\nu - \varepsilon^c_{\nu'} - \varepsilon^v_\alpha\right)\langle c_{\alpha'}^\dagger v_\nu^\dagger c_{\nu'} v_\alpha\rangle \\ &- \sum_{\nu_2\nu_3\nu_4}\Big[V_{\nu_1\nu_2\nu_3\alpha'}\langle c_{\nu_4}^\dagger(c_{\nu_2}^\dagger c_{\nu_3} + v_{\nu_2}^\dagger v_{\nu_3})v_\nu^\dagger c_{\nu'} v_\alpha\rangle \\ &\quad + V_{\nu_4\nu_2\nu_3\nu}\langle c_{\alpha'}^\dagger v_{\nu_4}^\dagger(c_{\nu_2}^\dagger c_{\nu_3} + v_{\nu_2}^\dagger v_{\nu_3})c_{\nu'} v_\alpha\rangle \\ &\quad - V_{\nu'\nu_2\nu_3\nu_4}\langle c_{\alpha'}^\dagger v_\nu^\dagger(c_{\nu_2}^\dagger c_{\nu_3} + v_{\nu_2}^\dagger v_{\nu_3})c_{\nu_4} v_\alpha\rangle \\ &\quad - V_{\alpha\nu_2\nu_3\nu_4}\langle c_{\alpha'}^\dagger v_\nu^\dagger c_{\nu'}(c_{\nu_2}^\dagger c_{\nu_3} + v_{\nu_2}^\dagger v_{\nu_3})v_{\nu_4}\rangle\Big] \\ &- i\sum_\xi\Big[g_{\xi\alpha'}^*\langle b_\xi^\dagger v_{\alpha'}^\dagger v_\nu^\dagger c_{\nu'} v_\alpha\rangle - g_{\xi\alpha}^*\langle b_\xi^\dagger c_{\alpha'}^\dagger v_\nu^\dagger c_{\nu'} c_\alpha\rangle \\ &\quad + g_{\xi\nu'}\langle b_\xi c_{\alpha'}^\dagger v_\nu^\dagger v_{\nu'} v_\alpha\rangle - g_{\xi\nu}\langle b_\xi c_{\alpha'}^\dagger c_\nu^\dagger c_{\nu'} v_\alpha\rangle\Big] \\ &- i\hbar\frac{d}{dt}\langle c_{\alpha'}^\dagger v_\nu^\dagger c_{\nu'} v_\alpha\rangle_{\mathrm{HF}}\,. \end{aligned} \tag{17}$$

Here, the first line is due to the free carrier Hamiltonian (2), the first sum due to the carrier Coulomb interaction (3), and the second sum due to the light-matter interaction (6). Note that the time derivative of the singlet factorization must be subtracted in order to obtain the pure four-operator correlation.

Evaluating equation (17) in singlet-doublet factorization leads to

$$\begin{aligned} i\hbar\frac{d}{dt}C^{\mathrm{x}}_{\alpha'\nu\nu'\alpha} &= \left(\varepsilon^c_{\nu'} - \varepsilon^v_\nu - \varepsilon^c_{\alpha'} + \varepsilon^v_\alpha\right)C^{\mathrm{x}}_{\alpha'\nu\nu'\alpha} \\ &+ V_{\nu'\alpha\nu\alpha'}\left[(1-f^c_{\alpha'})(1-f^v_\nu)\,f^c_{\nu'}f^v_\alpha\right. \\ &\left. - f^c_{\alpha'}f^v_\nu\,(1-f^c_{\nu'})(1-f^v_\alpha)\right] \\ &+ \sum_{\beta\beta'}\Big\{(f^v_\nu - f^v_\alpha)\,V_{\beta\alpha\nu\beta'}\left(C^{\mathrm{x}}_{\alpha'\beta\nu'\beta'} + C^c_{\alpha'\beta\nu'\beta'}\right) \\ &+ (f^c_{\alpha'} - f^c_{\nu'})\,V_{\nu'\beta\beta'\alpha'}\left(C^{\mathrm{x}}_{\beta\nu\beta'\alpha} + C^v_{\beta\nu\beta'\alpha}\right)\Big\} \\ &+ \sum_{\beta\beta'}\Big\{f^c_{\beta'}V_{\beta'\beta\beta'\alpha'}C^{\mathrm{x}}_{\beta\nu\nu'\alpha} + f^v_{\beta'}V_{\beta'\beta\beta\nu'}C^{\mathrm{x}}_{\alpha'\beta\nu'\alpha} \\ &- f^c_\beta\,V_{\nu'\beta\beta'\beta}\,C^{\mathrm{x}}_{\alpha'\nu\beta'\alpha} - f^v_{\beta'}V_{\alpha\beta\beta'\beta}\,C^{\mathrm{x}}_{\alpha'\nu\nu'\beta'}\Big\} \\ &+ \sum_{\beta\beta'}(f^c_\beta + f^v_\beta)\big[V_{\beta'\beta\beta\alpha'}C^{\mathrm{x}}_{\beta'\nu\nu'\alpha} + V_{\beta'\beta\beta\nu}\,C^{\mathrm{x}}_{\alpha'\beta'\nu'\alpha} \\ &- V_{\nu'\beta\beta\beta'}C^{\mathrm{x}}_{\alpha'\nu\beta'\alpha} - V_{\alpha\beta\beta\beta'}C^{\mathrm{x}}_{\alpha'\nu\nu'\beta'}\big] \\ &+ \sum_{\beta\beta'}\Big\{(f^v_\alpha - f^c_{\alpha'})\,V_{\beta\alpha\beta'\alpha'}\,C^{\mathrm{x}}_{\beta\nu\nu'\beta'} \\ &- (f^v_\nu - f^c_{\nu'})\,V_{\nu'\beta\nu\beta'}\,C^{\mathrm{x}}_{\alpha'\beta\beta'\alpha} \\ &+ (1 - f^v_\alpha - f^c_{\nu'})\,V_{\nu'\alpha\beta\beta'}\,C^{\mathrm{x}}_{\alpha'\nu\beta'\beta} \\ &+ (f^c_{\alpha'} - 1 + f^v_\nu)\,V_{\beta\beta'\nu\alpha'}\,C^{\mathrm{x}}_{\beta\beta'\nu'\alpha} \\ &+ (f^c_{\nu'} - f^c_{\alpha'})\,V_{\nu'\beta\alpha'\beta'}\,C^{\mathrm{x}}_{\beta\nu\beta'\alpha} \\ &+ (f^v_\alpha - f^v_\nu)\,V_{\beta\alpha\beta'\nu}\,C^{\mathrm{x}}_{\alpha'\beta\nu'\beta'}\Big\} \\ &- i\sum_\xi\delta_{\alpha\alpha'}\delta_{\nu\nu'}\left[g^*_{\xi\alpha}(f^v_\alpha - f^c_\alpha)\langle b_\xi^\dagger v_\nu^\dagger c_\nu\rangle\right. \\ &\left. + g_{\xi\nu}(f^v_\nu - f^c_\nu)\langle b_\xi c_\alpha^\dagger v_\alpha\rangle\right]. \end{aligned} \tag{18}$$

The first three lines of equation (18) correspond to a calculation of C^{x} in singlet approximation. A theory restricted to this level was used in [24,14] for quantum wells and in [16] for QDs. Note that the singlet factorization of six-operator averages already goes beyond the Hartree-Fock approximation. The terms in lines four and five can be shown to provide screening according to the Lindhard theory for the Coulomb potential in equation (13). The next two lines contain exchange-like scattering terms, followed by two lines of Hartree-like scattering terms, identified by the indices on the Coulomb matrix elements. The terms in the fourth sum are Coulomb renormalisations due to scattering between all possible states. The last two lines finally contain the contribution due to the light-matter interaction (6).

Similar equations for the correlations $C^c_{\alpha'\nu\nu'\alpha} = \delta\langle c_{\alpha'}^\dagger c_\nu^\dagger c_{\nu'} c_\alpha\rangle$ and $C^v_{\alpha'\nu\nu'\alpha} = \delta\langle v_{\alpha'}^\dagger v_\nu^\dagger v_{\nu'} v_\alpha\rangle$, obtained along the same lines, are provided in the appendix. Note that all Coulomb correlations can only redistribute carriers without changing the total population $\sum_\nu f_\nu^{c,v}$. Therefore, the changes of the carrier densities in each band are determined only by the photon-assisted polarisation. From combination of equations (12) and (14), one readily

obtains

$$\frac{d}{dt}\left(\sum_{\nu} f_{\nu}^{c} + \sum_{\xi}\langle b_{\xi}^{\dagger} b_{\xi}\rangle\right) = 0, \tag{19}$$

which reveals that a decrease (increase) of population in the conduction band is balanced by the increase (decrease) of the total photon number.

4 Photoluminescence spectrum

We now present results for the numerical solution of the equations derived in the previous section. The equations are solved in the time domain using a fourth order Runge-Kutta method. The material parameters are those of [8] for an InGaAs QD system and we consider a density of QDs on the WL of $3 \times 10^{10}\,\mathrm{cm}^{-2}$ and a gap energy of 1.52 eV. We assume that the excitation involves only carriers with one spin polarization, e. g., due to excitation with circular polarized light. It is not the purpose of this paper to study the dynamics of the carrier generation and relaxation. Since these processes are much faster than the recombination, we assume a quasi-equilibrium distribution of carriers with given carrier density and temperature as an initial state for our calculation. Within such an assumption, the initial values for the correlation functions C^{x}, C^{e}, and C^{v} remain open. For an unexcited system, all correlation functions are zero and build up during the carrier generation process according to their equations of motion. Starting a calculation with quasi-equilibrium population and vanishing correlation functions can lead to an abrupt build-up of correlations, resulting in unphysical oscillations of the correlation matrix elements which carry over to the population dynamics. To avoid these unphysical results, we numerically determine the initial conditions for the correlation functions from their equations of motion. For this purpose, we perform a separate calculation, where the carrier populations are adiabatically ramped up to their equilibrium values. The resulting steady-state solutions of the equations of motion for the correlation functions provide the starting point of the time evolution discussed in the following. We define the time-dependent luminescence spectrum according to [25, 26] and consider the limit of high frequency resolution of a detector to obtain

$$I(\omega) = \frac{d}{dt}\sum_{\xi}\langle b_{\xi}^{\dagger} b_{\xi}\rangle\Big|_{|\mathbf{q}|=\frac{\omega}{c}} . \tag{20}$$

Here the mode label contains $\xi = \{\mathbf{q}, \mathbf{e}_{\mathrm{p}}\}$. Using equation (12) leads to

$$I(\omega) = \frac{2}{\hbar}\sum_{\nu} |g_{q\nu}|^{2}\,\mathrm{Re}\,\widetilde{\langle b_{q}^{\dagger} v_{\nu}^{\dagger} c_{\nu}\rangle} , \tag{21}$$

where $\widetilde{\langle b_{\xi}^{\dagger} v_{\nu}^{\dagger} c_{\nu}\rangle} = \langle b_{\xi}^{\dagger} v_{\nu}^{\dagger} c_{\nu}\rangle / g_{\xi\nu}$ has been introduced. By means of this redefinition, the resulting quantity can be shown to depend only on the photon energy $\hbar\omega = \hbar c q$ and neither on the direction nor on the polarization of the mode ξ. Then the angular part and the polarization can be integrated out in equation (20), yielding the quantity $|g_{q\nu}|^{2}$ in equation (21) according to the Wigner-Weißkopf theory [21]. The total photon number is obtained from

$$I_{\mathrm{tot}} = \int d\omega\; I(\omega) . \tag{22}$$

The PL spectrum after a time evolution of 50 ps is plotted in Figure 1 for a temperature of 30 K and an initial carrier density of $1.5 \times 10^{10}\,\mathrm{cm}^{-2}$. Scaled relative to the bandgap, the two peaks correspond to the s-shell resonance at about -133 meV and the p-shell transition at about -79 meV. Both peaks are red shifted due to the Coulomb interaction from the non-interacting energies $\varepsilon_{p}^{c}+\varepsilon_{p}^{v} = -111$ meV and $\varepsilon_{s}^{c} + \varepsilon_{s}^{v} = -55.5$ meV, respectively. Furthermore, the peak height is increased due to the Coulomb interaction. The results of the calculation on the singlet-doublet level are compared to those obtained in Hartree-Fock approximation in Figure 1. The PL at the s-shell is slightly enhanced, while the p-shell PL becomes negative. Contrary to an absorption spectrum, where a negative peak corresponds to gain, this result is unphysical and an artefact of the Hartree-Fock approximation at low temperatures. If the correlations (16) are factorized on singlet-doublet level, the spectrum is positive for all temperatures.

5 Time-resolved photoluminescence

In this section we study the influence of the correlations on the time-resolved PL and on the corresponding population dynamics. Before we discuss the numerical results obtained by the solution of the full set of equations derived in Section 3, it is instructive to analyze the source term in equation (13) in more detail to gain a deeper understanding of the decay behavior of PL in a semiconductor. In the derivation of equation (13) the source term of spontaneous emission $g_{\xi\nu} f_{\nu}^{c}(1-f_{\nu}^{v}) + \sum_{\alpha} g_{\xi\alpha} C^{\mathrm{x}}_{\alpha\nu\nu\alpha}$ originates from the operator average $\sum_{\alpha} g_{\xi\alpha}\langle c_{\alpha}^{\dagger} v_{\alpha} v_{\nu}^{\dagger} c_{\nu}\rangle$. A simplified discussion is possible if one completely neglects the Coulomb interaction of carriers. Then the source term of spontaneous emission with the four-operator expectation value (resulting from the carrier-photon interaction) can be evaluated in two limiting cases. The first is the Hartree-Fock approximation, where correlations among the carriers are neglected. The second corresponds to the two-level approximation, where one assumes that the relevant physics is determined by one confined shell for electrons and for holes. The latter is of particular interest, since two-level models are frequently used in the discussion of QDs.

Within the Hartree-Fock approximation, the source term of spontaneous emission is solely determined by the product $f_{\nu}^{c}(1-f_{\nu}^{v})$. In the absence of Coulomb interaction, one can formulate the stationary solution of equation (13) for the case of slowly varying populations to obtain

$$\begin{aligned}\frac{d}{dt} f_{\nu}^{c} &= -\frac{2}{\hbar}\,\mathrm{Re}\sum_{\xi}\frac{i\,|g_{\xi\nu}|^{2} f_{\nu}^{c}(1-f_{\nu}^{v})}{\varepsilon_{\nu}^{c} - \varepsilon_{\nu}^{v} - \hbar\omega_{\xi} - i\Gamma} \\ &= -\frac{f_{\nu}^{c}(1-f_{\nu}^{v})}{\tau_{\nu}} ,\end{aligned} \tag{23}$$

where $1/\tau_\nu$ is the Wigner-Weißkopf rate of spontaneous emission for $\Gamma \to 0$. From this equation it is obvious that the decay of the population f_ν^c is non-exponential, unless f_ν^v is held constant by some mechanism, like background doping. Furthermore, the rate of decay depends on the carrier density and is higher for larger population. This behavior carries over to the PL according to equation (19). Due to the non-exponential PL decay a simple decay rate cannot be used to characterize the PL dynamics. This might appear surprising, since from a two-level atom an exponential decay behavior is known. Let us consider this two-level approximation as the second limiting case. Carrier indices are dropped as they are not needed if one restricts the population dynamics to one confined electron and hole level. In the de-excited (excited) state for this two-level system the electron is in the valence (conduction) band state. For the two spin polarizations, which are decoupled in the absence of Coulomb interaction, the independent processes then involve only the excitation of a single electron. This has a significant consequence for the evaluation of the source term of spontaneous emission. Within the two-level approximation, the successive application of more than one annihilation operator always yields zero, so that the source term $\langle c^\dagger v v^\dagger c \rangle$ reduces to f^c. Along the same lines discussed above, instead of equation (23) one obtains

$$\frac{d}{dt} f^c = -\frac{f^c}{\tau}, \tag{24}$$

which corresponds to the exponential decay behavior known from the two-level system. In this case, $1/\tau$ is the Wigner-Weißkopf rate for the considered two-level transition. From this analysis we deduce that the exponential decay within the two-level approximation stems from the fact, that the excitation of a QD conduction-band carrier is rigidly linked to the absence of a QD valence-band carrier. In the electron-hole picture, this corresponds to fully correlated electron-hole pair. On the operator level this is expressed as $c^\dagger v v^\dagger c = c^\dagger c$. In a semiconductor, however, many carriers can be present and the correlations are subject to scattering and dephasing processes and must therefore be explicitly calculated.

Now we turn to the numerical results of the SLE including Coulomb interaction. In Figure 2 we show the evolution of the time-resolved PL on a nanosecond timescale. Again, the initial carrier density is taken to be $1.5 \times 10^{10}\,\mathrm{cm}^{-2}$. For the solid line all correlations up to the singlet-doublet level have been included. The result of a calculation in the Hartree-Fock approximation, which corresponds to uncorrelated carriers, is shown as dashed line. In this case, the decay is clearly non-exponential, which is in accordance with equation (23). Comparing these two results, it is obvious that carriers are strongly correlated on the singlet-doublet level. However, our approach so far does not account for any kind of dephasing of the correlations. A physical mechanism for such dephasing is phonon scattering. Hoyer et al. have studied phonon scattering on a microscopic level for a quantum-well system. It is shown that dephasing of correlations is indeed

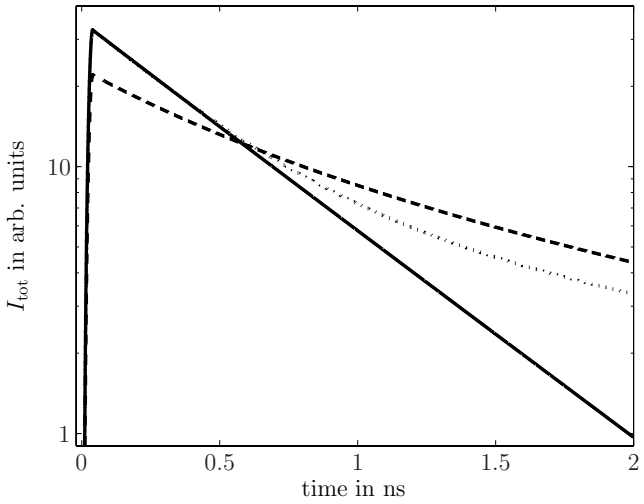

Fig. 2. Logarithmic plot of the time evolution of the quantum dot photoluminescence. The dashed line corresponds to the calculation in singlet factorization and the solid line to the singlet-doublet level. For the dotted line the described phonon contributions were added to the singlet-doublet calculation.

provided, although this enters only via higher-order triplet terms [15,27]. Extensions beyond the singlet-doublet factorization are not within the scope of this work. Nevertheless, we can account for the main features of phonon scattering, which is dephasing of correlations and relaxation of the carrier population towards thermal equilibrium at the lattice temperature, on a phenomenological level. A constant dephasing term $-i\gamma C^{\mathrm{x}}_{\alpha'\nu\nu'\alpha}$ is added on the right hand side of equation (18) and the scattering is treated within relaxation-time approximation by introducing

$$\frac{d}{dt} f_\nu^{c,v}\Big|_{\mathrm{relax}} = -\frac{f_\nu^{c,v} - F_\nu^{c,v}(T)}{\tau_{\mathrm{relax}}^{c,v}}\,, \tag{25}$$

in equations (14) and (15), where $F_\nu^{c,v}(T)$ is a Fermi-Dirac distribution at temperature T, evaluated for every timestep at the present carrier density.

Several recent experimental and theoretical investigations address the efficiency of carrier-phonon interaction in QDs. In the low-temperature regime, interaction of carriers with LA-phonons [28–30] provide the dominant dephasing mechanism while at elevated temperatures the interaction of carriers with LO-phonons leads to very efficient dephasing [19]. The temperature dependence of the dephasing has been studied, e.g., in four-wave experiments [31]. Even for the weakest dephasing values obtained there, we find that the influence of carrier correlations on the PL decay is strongly reduced.

To demonstrate the effect of a very weak constant dephasing rate $\gamma = 0.001\,\mathrm{meV}$ has been used for the dotted line in Figure 2. The correlations are drastically reduced so that a non-exponential signature of the decay is regained on longer timescales.

Our numerical analysis shows that the results presented here are rather insensitive to the exact value of the relaxation time $\tau_{\mathrm{relax}}^{c,v}$. For the discussed example, typical

values of $\tau_{\mathrm{relax}} = 1\,\mathrm{ps}$ for electrons and holes are taken [19]. Hoyer et al. have shown that a constant dephasing γ causes unphysical heating of the system. However, this effect is only weak for a small value for the dephasing and, additionally, the scattering term (25) counteracts the heating.

Clearly such a phenomenological treatment of the dephasing via phonons cannot cover the spectral side-bands known from an interaction of the QD carriers with the continuum of acoustic phonons [29,32]. For this an inclusion of a microscopic description of the electron-phonon interaction would be necessary and is left for future work. Here the important point is that *any* kind of dephasing of the correlations will lead to deviations from an exponential decay.

6 Conclusion

A theory for semiconductor quantum dots interacting with the quantized light field has been used to describe the spontaneous emission properties. The influence of carrier-carrier correlations due to Coulomb as well as carrier-photon interaction has been studied using the cluster-expansion technique on the singlet-doublet level. For the time-resolved PL, we have discussed two limiting cases of i) uncorrelated carriers (Hartree-Fock approximation), leading to a nonexponential decay; and ii) fully-correlated carriers (two-level approximation), resulting in an exponential decay. Our numerical results reveal that correlations of the excited electrons and holes, treated on the singlet-doublet level, lead to an exponential PL decay, provided that dephasing of the correlations can be neglected. This indicates that with a theory formulated in the single-particle electron-hole basis, the presented inclusion of correlations due to Coulomb and carrier-photon interaction allows to recover the limit of strongly correlated carriers, which are typically viewed as excitonic excitations. In turn, it is also shown that weak dephasing of the correlations leads to a clear departure from the exponential decay of the PL signal. This result provides a possible intrinsic explanation for experimental observations of non-exponential PL decay, which is in contrast to extrinsic effects like coupling to dark excitons or inhomogeneous broadening effects.

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Appendix A: Equation of motion for intraband correlations

The intraband correlations can be obtained in analogy to the derivation of equation (18). We restrict ourselves here to the equation of motion for the correlations of the conduction-band electrons. A similar equation can be given for the valence-band carriers by exploiting the symmetry properties of the Hamiltonian (1).

$$
\begin{aligned}
i\hbar\frac{d}{dt}C^{c}_{\alpha'\nu\nu'\alpha} = &-\left(\varepsilon^{c}_{\alpha'}+\varepsilon^{c}_{\nu}-\varepsilon^{c}_{\nu'}-\varepsilon^{c}_{\alpha}\right)C^{c}_{\alpha'\nu\nu'\alpha}\\
&-\left(V^{*}_{\alpha'\nu\nu'\alpha}-V^{*}_{\alpha'\nu\alpha\nu'}\right)\left[(1-f^{c}_{\nu})(1-f^{c}_{\alpha'})\,f^{c}_{\nu'}f^{c}_{\alpha}\right.\\
&\left.-f^{c}_{\nu}f^{c}_{\alpha'}\,(1-f^{c}_{\nu'})(1-f^{c}_{\alpha})\right]\\
&+\sum_{\beta\beta'}\Big\{(f^{c}_{\nu'}-f^{c}_{\nu})\,V_{\nu'\beta\beta'\nu}\,C^{c+\mathrm{x}}_{\alpha'\beta\alpha\beta'}\\
&-(f^{c}_{\alpha'}-f^{c}_{\alpha})\,V_{\alpha\beta\beta'\alpha'}\,C^{c+\mathrm{x}}_{\nu\beta\nu'\beta'}\\
&-(f^{c}_{\nu'}-f^{c}_{\alpha'})\,V_{\nu'\beta\beta'\alpha'}\,C^{c+\mathrm{x}}_{\nu\beta\alpha\beta'}\\
&+(f^{c}_{\nu}-f^{c}_{\alpha})\,V_{\alpha\beta\beta'\nu}\,C^{c+\mathrm{x}}_{\alpha'\beta\nu'\beta'}\Big\}\\
&+\sum_{\beta\beta'}\Big\{f^{c}_{\beta'}V_{\beta'\beta\beta'\alpha'}C^{c}_{\beta\nu\nu'\alpha}+f^{c}_{\beta'}V_{\beta'\beta\beta'\nu}\,C^{c}_{\alpha'\beta\nu'\alpha}\\
&-f^{c}_{\beta}\,V_{\nu'\beta\beta'\beta}\,C^{c}_{\alpha'\nu\beta'\alpha}-f^{c}_{\beta}\,V_{\alpha\beta\beta'\beta}\,C^{c}_{\alpha'\nu\nu'\beta'}\Big\}\\
&+\sum_{\beta\beta'}\Big\{(f^{c}_{\beta}+f^{v}_{\beta})\,V_{\beta'\beta\beta\alpha'}C^{c}_{\beta'\nu\nu'\alpha}\\
&+(f^{c}_{\beta}+f^{v}_{\beta})\,V_{\beta'\beta\beta\nu}\,C^{c}_{\alpha'\beta'\nu'\alpha}\\
&-(f^{c}_{\beta}+f^{v}_{\beta})\,V_{\nu'\beta\beta\beta'}C^{c}_{\alpha'\nu\beta'\alpha}\\
&-(f^{c}_{\beta}+f^{v}_{\beta})\,V_{\alpha\beta\beta\beta'}C^{c}_{\alpha'\nu\nu'\beta'}\Big\}\\
&+\sum_{\beta\beta'}\Big\{(1-f^{c}_{\alpha}-f^{c}_{\nu'})\,V_{\nu'\alpha\beta'\beta}\,C^{c}_{\alpha'\nu\beta\beta'}\\
&-(1-f^{c}_{\alpha'}-f^{c}_{\nu})\,V_{\beta\beta'\nu\alpha'}C^{c}_{\beta\beta'\nu'\alpha}\\
&-(f^{c}_{\alpha'}-f^{c}_{\alpha})\,V_{\alpha\beta\alpha'\beta'}C^{c}_{\beta\nu\nu'\beta'}\\
&-(f^{c}_{\nu'}-f^{c}_{\alpha'})\,V_{\beta\nu'\beta'\alpha'}C^{c}_{\beta\nu\beta'\alpha}\\
&-(f^{c}_{\nu'}-f^{c}_{\nu})\,V_{\beta\nu'\beta'\nu}\,C^{c}_{\alpha'\beta\beta'\alpha}\\
&-(f^{c}_{\nu}-f^{c}_{\alpha})\,V_{\alpha\beta\nu\beta'}C^{c}_{\alpha'\beta\nu'\beta'}\Big\}
\end{aligned}
\tag{A.1}
$$

Here we have used the abbreviation $C^{c+\mathrm{x}}_{\alpha'\nu\nu'\alpha} = C^{c}_{\alpha'\nu\nu'\alpha} + C^{\mathrm{x}}_{\alpha'\nu\nu'\alpha}$. The terms can be interpreted in analogy to equation (18). However, the contribution due to the light-matter interaction vanishes for C^{c} and C^{v}. We found the influence of the correlations C^{c} and C^{v} to be negligible in comparison to the electron-hole correlations C^{x} for the studied parameters.

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