A Theoretical Analysis of Instantaneous Coulomb Renormalizations in a Single Quantum Dot Pump-Probe Experiment

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Abstract. We present a theoretical analysis of Coulomb-correlation effects on the optical spectra of a single negatively charged quantum dot. The considered initial and final many particle states are adapted and motivated by a recent two-color pump-probe experiment. We demonstrate that the lowest energy transition is noticeably affected by exchange interactions if the initial state consists of a hot trion. It turns out that for initial states as obtained after spin-conserving carrier relaxation new absorption and emission lines emerge.

1. Introduction
In a recent publication femtosecond two-color pump-probe experiments on a strongly confined and singly charged quantum dot (QD) have been reported that allowed for an analysis of the optically induced dynamics on a picosecond timescale [1]. In these experiments, a pump pulse initialized the QD by a resonant excitation of a higher shell while the probe pulse was in resonance with the lowest energy transition. It was found that for short positive delay times between pump and probe pulse the fundamental trion line (FTL) vanished. To explain this result instantaneous Coulomb renormalizations of the respective transition energies have been suggested. For larger positive delay times the experiment reported optical gain on the FTL, which was traced back to spin-conserving intraband relaxations.

We study, motivated by the above mentioned experiment, the influence of Coulomb-correlation effects on the FTL. In particular we will show that this absorption line is noticeably affected when a pump pulse creates an exciton in a higher shell. Furthermore, we show that after spin-conserving intraband relaxations of the excited carriers new optical transition lines around the FTL emerge which can be assigned to transitions from the trion state either into a single electron state or to a charged biexciton state.

2. Theoretical Model
In order to study the influence of Coulomb interactions on the optical transitions in a strongly confined QD we model the QD in the envelope-function approximation by confinement potentials for electrons and holes given by an anisotropic harmonic oscillator potential. We assume that
the confinement in growth direction is much stronger than in the lateral direction. Therefore, only the lowest energy state in the growth (z-)direction is considered which is of heavy-hole type. The confinement lengths in the in-plane directions x and y are taken to be similar, such that the single particle states $|i\rangle$ are organized in shells referred to as $s, p, d, \ldots$. Furthermore, we assume identical envelopes for conduction-band electrons and valence-band holes, such that the Coulomb matrix elements for electron-electron-, hole-hole-, and electron-hole-interaction are the same for a given combination of oscillator quantum numbers $i, j, k, l$, i.e., $V_{ijkl}^{ee} = V_{ijkl}^{eh} = V_{ijkl}^{hh}$ [2, 3]. The Hamiltonian conserves the total number of electrons $N_e$ and holes $N_h$ as well as the spin of the electrons ($S^e, S^e_z$) and holes ($S^h, S^h_z$). As a consequence, the Hamiltonian in a basis of uncorrelated many particle states consists of subblocks for fixed numbers of electrons and holes and constant spin configurations. We have calculated the Coulomb-correlated multi-exciton states by numerically diagonalizing these subblocks taking into account the twelve lowest single particle states. Finally, the coupling to an external light field is described within the usual dipole and rotating wave approximation giving rise to the standard selection rules for excitonic transitions. The optical absorption or stimulated emission spectra resulting from this coupling are then evaluated using Fermi’s golden rule. In order to account for various line broadening mechanisms we have replaced the $\delta$-function in Fermi’s golden rule by a Lorentzian. We consider for the following results a single CdSe QD of ellipsoidal shape with the lengths $l_x = 2.6$ nm, $l_y = 2.8$ nm and $l_z = 1.0$ nm. The conduction band mass $m_e = 0.13 m_0$, the valence band mass $m_h = 0.45 m_0$, as well the dielectric constant $\epsilon_r = 9.57$ are taken from Ref. [4].

3. Results

Let us first discuss optical transitions of a negatively charged QD from the ground state to a charged exciton (trion) state with two electrons and one hole. In Fig. 1 the calculated absorption spectrum including the 20 lowest trion states is plotted. The energies are given relative to the FTL. The absorption spectrum shows three main absorption lines. Our numerical results indicate that these states are dominated by a single uncorrelated electron-hole state with weak admixture of other states. The first line corresponds to the optically allowed FTL where all particles in the final state are in the $s$-shell, the two electrons having opposite spin projections forming a singlet state, i.e., $S^e = 0$. The second and third line are triplet states displaying excited trion transitions to the $p$-shell which are split due to the anisotropic in-plane confinement potential. Around these main excited transition lines a bunch of weaker absorption lines appears which correspond to strongly mixed singlet states. The comparison of our calculated spectra with the photoluminescence (PL) and the photoluminescence excitation spectra (PLE) measured in Ref. [1] reveals a good quantitative and qualitative agreement. This allows us to identify the electronic states which were resonantly excited in the pump-probe experiment. For this purpose, we have also plotted the spectral envelopes of the pump (red) and probe (thin dashed blue line) spectra.

![Figure 1](image_url)  

**Figure 1.** (color online) Absorption spectrum of a singly charged quantum dot. Energies are given with respect to the lowest energy transition. The spectral envelopes of the pump pulse (thin red line) and probe pulse (thin dashed blue line) used by the two color pump-probe experiment in Ref. [1] are also depicted.
Figure 2. (color online) Schematic illustration of the analyzed pump-probe experiment. The pump pulse excites a negatively charged QD and creates a hot trion. The following probe pulse is in the spectral range of the FTL and measures optical transitions to a hot charged biexciton (states (1) and (2)). After spin-conserving carrier relaxation new optical transitions emerge resulting in stimulated emission to a single electron state (states (3) and (4)) and absorption to a charged biexciton in the ground state (state (5)).

When applying a $\pi$-pulse in resonance to a higher shell a hot electron-hole pair is created transferring the initial single electron state to a hot trion state. The electrons in this optically initialized QD may have parallel or antiparallel spins (see Fig. 2). The probe-pulse in the spectral region of the FTL now gives rise to transitions to a charged biexciton state with three electrons and two holes, where the holes may have spins with parallel or antiparallel orientation (states labeled (1) and (2) in Fig. 2).

Figure 3 shows in part (a) again the absorption spectrum of the unexcited QD, i.e., the FTL. In part (b) the spectrum measured by a probe pulse in the presence of the hot trion is plotted. We clearly see that the FTL is now split into two lines with approximately half of the original intensity. The lines correspond to the transitions to the charged biexciton states labeled (1) and (2) in Fig. 2. The energetically lower line results from the transition to the (hole-)triplet states, $S_h = 1$, whereas the higher line is related to the transition to the (hole-)singlet state, $S_h = 0$. Indeed, it turns out that the energy separation ($\sim 14$ meV) is close to the hole exchange matrix element $V_{hhsp}$ between holes in $s$- and $p$-states. Hence, the reported vanishing FTL may be traced back to strong exchange interactions between holes in different shells.

The exchange interaction is effective only for carriers in different orbital shells and, therefore, vanishes when the hot trion relaxes to its lowest energy level. The times for spin-conserving energy relaxation processes are typically of the order of several picoseconds [5, 6]. As is shown in Fig. 2, we now have to distinguish two cases. If the electrons in $s$- and $p$-shell have opposite spin orientations, both electron and hole will relax to their respective ground states. If they have the same spin orientation, electron relaxation is suppressed due to Pauli blocking and only the hole will relax. This has important consequences for the absorption spectrum of a subsequent probe pulse in the spectral range of the FTL. In the former case the electronic $s$-shell is completely filled and the pump pulse will give rise to a stimulated emission leading to the final single electron state (3). This is exactly the inverse process of the fundamental trion absorption line. Thus the corresponding spectrum shown in Fig. 3(c) shows a gain line at zero energy (red solid line). In the latter case both an emission and an absorption process are possible. The final state in the emission process is an electron in the $p$-shell (4) while the absorption process leads to a
Figure 3. (Color online) Optical spectra in the region of the FTL resulting from a QD initially (a) in the negatively charged ground state, (b) in a hot trion state, and (c) after spin-conserving relaxation (see also Fig. 2). The red solid curve in (c) refers to the initial state with both electrons in the s-shell, the green dashed line to the state with one electron in the s- and one in the p-shell.

charged biexciton in the ground state (5). As can be seen in Fig. 3(c) (green dashed line) the emission gives rise to a gain line (4) which is strongly blue-shifted with respect to the FTL. Here the electron-electron exchange interaction considerably lowers the energy. The absorption line corresponding to the transition to the charged biexciton is ∼1 meV red-shifted with respect to the FTL showing that here exchange interactions do not strongly affect the electronic states and Coulomb-correlations are rather small. It should be mentioned that this transition has not been observed in the discussed pump-probe experiment Ref. [1], although it is very close to the FTL. However, it turns out that the exact spectral position of this line strongly depends on geometry parameters, in particular the relative confinements length of electron and hole and it may easily be shifted outside of the spectral window detected in the experiment.

4. Conclusions

We have analyzed the influence of Coulomb-renormalizations on the optical spectra of a single negatively charged QD. The considered initial and final Coulomb-correlated multi-exciton states have been chosen according to states excited in a two-color pump-probe experiment. We have shown that the FTL is noticeably affected by exchange interactions as long as carrier relaxation has not started. This result may explain a reported vanishing of this lowest energy transition in a recent experiment. For initial states resulting from a spin-conserving energy relaxation new optical transition lines emerge exhibiting both absorption and emission.

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