INTRABAND RELAXATION OF p-SHELL EXCITONS IN DISK-SHAPED QUANTUM DOTS*

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We study the generation of p-shell excitons in optically active disk-shaped quantum dots subjected to ultrafast optical pulses. The single-particle spectral properties are obtained from the four-band kp theory, whereas the Coulomb interaction is taken into account within the configuration-interaction approach, particular attention being payed to configuration mixing due to electron-hole correlations. The effect of intraband relaxation processes is included in the non-unitary dynamics derived from the von-Neumann Lindblad equation. We find that the fast hole relaxation processes drive the p-shell excitons to intermediate states which eventually evolve to s-shell excitons *via* slower electron relaxation.

Key words: Excitons in quantum dots, Intraband relaxation effects.

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1. INTRODUCTION

One of the most promising approaches towards implementing quantum hardware [1] is based on exciton dynamics in optically active quantum dots (QD). There are a lot of extensive studies in which self-assembled quantum dots were investigated both experimentally [2, 3] and theoretically [4, 5]. The latter studies mostly concentrate on systems with two levels, while we model a more reallistic QD. Therefore we take into account both s and p shells, which requires considering configurations mixing effects and intraband relaxation processes (IBR). The aim of this work is to reveal the effects of the IBR on the evolution of p-shell excitons for a disk-shaped QD subjected to ultrafast laser pulses. The evolution of more complex states, such as the sp biexcitons was investigated in our recent work [6], the two-color protocol described there being similar to the conditional C-NOT gate proposed in Ref.[5]. Our simulations include the Coulomb interaction within the configuration interaction model. We stress the fact that when the p-shell states are taken into account one cannot use a 4×4 evolution operator as in the case of the two level system [2] because IBR

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activates states which are otherwise forbidden. The paper is organized as follows: Section 2 presents the formalism, Section 3 contains the numerical results and their discussion. We conclude in Section 4.

2. FORMALISM

The valence band wave functions were obtained by diagonalizing the 4×4 Kohn-Luttinger H_{KL} Hamiltonian [7], within the anvelope function approximation [8], w.r.t to the single-particle basis $|J_z, m_z, n, l\rangle := |\phi_{m_z nl}\rangle|J_z\rangle$, $|J_z\rangle$ being the band-edge Bloch states and $\phi_{m_z nl}$ the appropriate envelope functions for cylindrical confinement with vanishing Dirichlet boundary conditions (see Ref. [6] for further details). Here m_z is the quantum azimuthal number, n is the other quantum number associated with the transverse confinement and l is the quantum number associated with the confinement along the vertical z axis. The total orbital quantum number $F_z = J_z + m_z$ is a good quantum number for a given spinor. The conduction band wave functions ψ_j^c were found by diagonalizing a 2×2 single-band effective mass Hamiltonian H_C in the basis $\{|S_z, m_z, n, l\rangle\}$. The set of non-interacting many-body states (MBS) of $H_{KL} + H_C$ are denoted by $\{\nu\}_{\nu=1,N_{MB}}$. We proceed further by writing the fully interacting Hamiltonian $H_0 = H_{KL} + H_C + W$ in terms of creation and annihilation operators for electrons and holes in the non-interacting basis described above. W represents the Coulomb interaction Hamiltonian. The interacting states (denoted here by bold letters) can be written as linear combinations of non-interacting MBS: $|\boldsymbol{\nu}\rangle = \sum_{\nu'} C_\nu^{\boldsymbol{\nu}} |\nu'\rangle$.

The von Neumann-Lindblad equation for the density operator ρ (see *e.g.* Ref.[10]) reads as follows :

$$i\hbar\dot{\rho}(t) = [H_0 + V_R(t), \rho(t)] + i \sum_{\lambda=C,V} \mathcal{L}_\lambda[\rho(t)], \tag{1}$$

where $V_R(t)$ is the semi-classical interband Hamiltonian describing light-matter interactions. $\mathcal{L}_\lambda$ is the Lindblad "collapse" operator which describes the IBR between single particle states from the band λ.

The relaxation rates are defined as the inverses of the relaxation times which were taken from literature. The spin-conserving hole relaxation time τ_h is about few picoseconds or even less than 1 ps, spin-conserving relaxation time for electrons τ_e exceeds 10ps [6], the spin-flip processes in the conduction band $\tau_s = 10\tau_e$ and, finally, the hole spin relaxation is neglected as the measured values are of order of nanoseconds.

We numerically solved the Lindblad equation on the relevant Fock subspace of the interacting MBS which contain the relevant states: the ground state (*i.e.* filled valence band, empty conduction band) and all exciton and biexciton states.

The diagonal element of the density operator represents the population of a particular many-body configuration whose notation is $P(\boldsymbol{\nu}) = \langle \boldsymbol{\nu} | \rho(t) | \boldsymbol{\nu} \rangle$.

3. NUMERICAL RESULTS

Let H be the height of the cylinder and R its radius. Then for small aspect ratios $\frac{H}{2R}$ the LH-HH mixing is small [9]. In this case a Luttinger spinor has only one component and it can be denoted by $|J_z, m_z\rangle$.
Let us consider the dynamics of p-shell exciton for $R = 15$nm and $H = 5$nm CdTe QD. The lowest energy fully interacting p exciton is denoted by $\mathbf{X}_{p\downarrow}$ and is a mixture of non-interacting electron-hole states:

$$|\mathbf{X}_{p\downarrow}\rangle = C_{\downarrow_{p_-}} |\downarrow_{p_-} \Uparrow_{p_+}\rangle + C_{\downarrow_{p_+}} |\downarrow_{p_+} \Uparrow_{p_-}\rangle + C_{s\downarrow} |\downarrow_s \Uparrow_s\rangle, \tag{2}$$

where $p_\pm$ are the p sub-shells corresponding to the values of $m_z = +1$ and $m_z = -1$ for the quantum azimuthal number m_z. The dominant character of the state $\mathbf{X}_{p\downarrow}$ in (2) is p-like because the weight $|C_{\downarrow_{p_-}}|^2 \approx |C_{\downarrow_{p_+}}|^2 \gg |C_{s\downarrow}|^2$. We observe that both p-shell states $|\downarrow_{p_-} \Uparrow_{p_+}\rangle$ and $|\downarrow_{p_+} \Uparrow_{p_-}\rangle$ contribute substantially to the exciton state while the weight of the noninteracting s-shell state is very small. This behavior is explained by looking at the non-conserving matrix element of the interband interaction $V_{pp} = \langle \downarrow_{p_+} \Uparrow_{p_-} |W^{eh}| \downarrow_{p_-} \Uparrow_{p_+}\rangle$ which leads to the mixing of the two p-shell states. The matrix element V_{pp} exceeds the gap within the p shell and therefore a strong mixing is expected [6].
The lowest energy exciton $\mathbf{X}_{p\downarrow}$ is generated by a resonant σ_+ pulse. At $t = 0$ the system is in the state $|0\rangle$ for which there are no holes in the valence band and no electrons in the conduction band. Due to the small splitting within the p shell the two Kramers doublets are simultaneously activated by the optical pulse. We have checked that the contribution from the biexciton $|\mathbf{B}_{pp}\rangle \sim |\downarrow_{p_-} \downarrow_{p_+} \Uparrow_{p_+} \Uparrow_{p_-}\rangle$ is very small due to the rather large (~ 8 meV) binding energy. In the absence of the IBR the occupations of the exciton state and of the initial state display Rabi oscillations (not shown). The two spin-conserving hole relaxation processes within the p shell are $\Uparrow_{p_-} \to \Uparrow_{p_+}$ and $\Downarrow_{p_+} \to \Downarrow_{p_-}$. On the other hand the s shell receives holes from both $p_\pm$ subshells. The effect of the relaxation processes on the p-shell exciton is shown in Fig. 1 (a). The simulations were done by setting the ground state as the initial state. The σ_+ pulse is applied at $t = 0$ and then switched off at $t = 9.6$ps. One notices the damped Rabi oscillations of the optically generated p-shell exciton and the exponential decay once the pulse is stopped. The population of the ground state becomes vanishingly small once the pulse is switched off. The s-shell exciton state $\mathbf{X}_{s\downarrow}$ is filled uniformly due to IBR processes which we now discuss in more detail. Already by looking at Fig. 1(a) we can guess that the dissipative dynamics does not involve only the states $\mathbf{X}_{p\downarrow}$, $\mathbf{X}_{s\downarrow}$ and the ground state; if this was the case one would

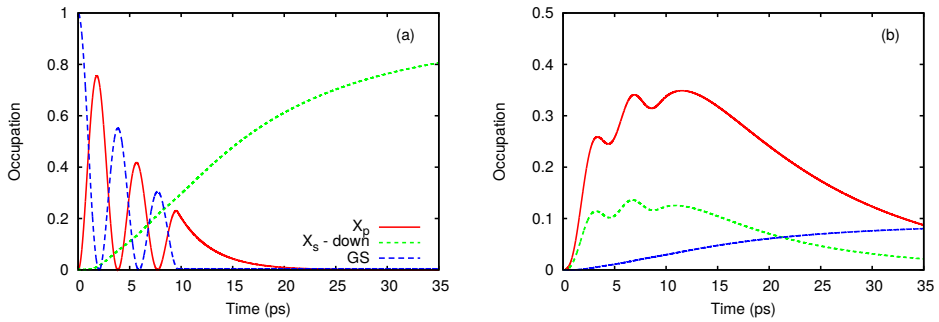

Fig. 1 – (Color on-line) a) IBR effects on the dynamics of the p-shell exciton. The populations $P(0)$ and $P(\mathbf{X}_{p\downarrow})$ exhibit damped Rabi oscillations while occupation of the s-shell exciton $\mathbf{X}_{s_\downarrow}$ increases uniformly. b) The occupation of the intermediate states: solid (red) line - $P(\mathbf{X}^{\downarrow_{p_+}\Uparrow_s}_{\downarrow_s\Uparrow_{p_+}}) + P(\mathbf{X}^{\downarrow_{p_-}\Uparrow_s}_{\downarrow_s\Uparrow_{p_-}})$, dashed (green line) - $P(\mathbf{X}^{\downarrow_s\Uparrow_{p_+}}_{\downarrow_{p_+}\Uparrow_s}) + P(\mathbf{X}^{\downarrow_s\Uparrow_{p_-}}_{\downarrow_{p_-}\Uparrow_s})$, dotted (blue) line - $P(\mathbf{X}_{s\uparrow})$. The relaxation times for electrons and holes are $\tau_h = 4$ps, $\tau_e = 30$ps. The electric field is $E = 30$kV/cm.

check that the occupations of these states sum to 1.

Thus it turns out that the relaxation of $\mathbf{X}_{p\downarrow}$ to s-shell excitons is mediated by intermediate states. This can be understood by discussing first the relaxation processes in the non-interacting case. One can easily see that the exciton $|\downarrow_{p_+}\Uparrow_{p_-}\rangle$ decays to s-shell excitons through multiple 'paths' (we list for simplicity only states accessible *via* spin-conserving relaxation processes):

$$|\downarrow_{p_+}\Uparrow_{p_-}\rangle \Longrightarrow |\downarrow_{p_+}\Uparrow_s\rangle \longrightarrow |\downarrow_s\Uparrow_s\rangle, \tag{3}$$

$$|\downarrow_{p_+}\Uparrow_{p_-}\rangle \longrightarrow |\downarrow_s\Uparrow_{p_-}\rangle \Longrightarrow |\downarrow_s\Uparrow_s\rangle. \tag{4}$$

Here the double (simple) arrows mark that the relaxation process implies holes (electrons). Since the hole relaxation is the fastest process the path given in Eq.(3) will be more favorable and the intermediate state $|\downarrow_s\Uparrow_{p_-}\rangle$ on the 'slow' path (see Eq.(4)) will be less populated. The states $|\downarrow_{p_+}\Uparrow_s\rangle$ and $|\downarrow_s\Uparrow_{p_-}\rangle$ are optically dark.

In the interacting case the configuration mixing comes into play and we find that the p-shell exciton (see Eq.(2)) relaxes *via* four intermediate states which are mostly made of two non-interacting configurations:

$$|\mathbf{X}^{\downarrow_{p_\pm}\Uparrow_s}_{\downarrow_s\Uparrow_{p_\pm}}\rangle = C_{p_\pm,s}|\downarrow_{p_\pm}\Uparrow_s\rangle + c_{s,p_\pm}|\downarrow_s\Uparrow_{p_\pm}\rangle, \tag{5}$$

$$|\mathbf{X}^{\downarrow_s\Uparrow_{p_\pm}}_{\downarrow_{p_\pm}\Uparrow_s}\rangle = C_{s,p_\pm}|\downarrow_s\Uparrow_{p_\pm}\rangle + c_{p_\pm,s}|\downarrow_{p_\pm}\Uparrow_s\rangle. \tag{6}$$

In the above definitions the upper configuration is the dominant one, that is its weight exceeds the weight of the lower configuration, *i.e.* $|C|^2 \gg |c|^2$.

The dynamics of the intermediate states is given in Fig. 1(b) (see the caption for more details). It turns out that the two states given in Eq.(5) behave in a very similar way

so for simplicity we display only the total occupation of these states and the total occupation of the two states defined in Eq.(6). As expected the intermediate states are populated much faster than the s-shell excitons but they deplete considerably at $t = 35$ps. There is also a clear connection between the filling of the p-shell exciton and the increased population of the intermediate states. In particular, the minima of $P(\mathbf{X}_{p_\downarrow})$ correspond to local minima of the intermediate states population. The most important observation from Fig. 1(b) is that the states $\mathbf{X}^{\downarrow_{p_+}\Uparrow_s}_{\downarrow_s\Uparrow_{p_+}}$ and $\mathbf{X}^{\downarrow_{p_-}\Uparrow_s}_{\downarrow_s\Uparrow_{p_-}}$ are being populated much faster and also that their occupation exceeds the population of the other intermediate states. This happens because the dominant many-body configurations in these states (namely $|\downarrow_{p_+}\Uparrow_s\rangle$ and $|\downarrow_{p_-}\Uparrow_s\rangle$) are filled up *via* fast hole relaxation process. On the other hand, the states $\mathbf{X}^{\downarrow_s\Uparrow_{p_+}}_{\downarrow_{p_+}\Uparrow_s}$ and $\mathbf{X}^{\downarrow_s\Uparrow_{p_-}}_{\downarrow_{p_-}\Uparrow_s}$ are configurations which mostly experience electron relaxation, so their charging will be slower. The slow spin-flip processes are less effective so the steady-state population of the spin-up exciton $\mathbf{X}_{s\uparrow}$ does not exceed 8%.

4. CONCLUSIONS

We studied the dynamics of p-shell excitons in a CdTe QD subjected to electromagnetic pulses. The geometry of the sample and the Coulomb interaction effects were taken into account. We introduced IBR processes within the von Neumann-Lindblad equation. We find that the Rabi oscillations of the p-shell exciton are strongly damped in the presence of IBR. The latter leads to the filling of intermediate states which are optically inactive.

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