

Coulomb effects in semiconductor quantum dots

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Abstract. Coulomb correlations in the optical spectra of semiconductor quantum dots are investigated using a full-diagonalization approach. The resulting multi-exciton spectra are discussed in terms of the symmetry of the involved states. Characteristic features of the spectra like the nearly equidistantly spaced *s*-shell emission lines and the approximately constant *p*-shell transition energies are explained using simplified Hamiltonians that are derived taking into account the relative importance of various interaction contributions. Comparisons with previous results in the literature and their interpretation are made.

PACS. 78.67.Hc Quantum dots – 71.35.-y Excitons and related phenomena

1 Introduction

Recent experimental investigations of optical spectra from individual self-assembled semiconductor quantum dots (QDs) [1] renewed the interest in fundamental quantum-mechanical effects of confined interacting few-carrier systems and their theoretical description [2–6]. The few-carrier systems in individual QDs are interesting candidates for potential applications in quantum optics like sources of single photons or entangled photon pairs [7].

Apart from the experimental relevance, the problem of few charge carriers in the discrete states of a given confinement potential is also a paradigm for an interacting few-carrier system, that can be solved without further approximations. A discussion of the exact (but complicated) results in terms of simplified pictures is therefore desirable.

In the past, electronic states and the resulting dipole transition have been calculated in box-like confinement potentials using exact diagonalization [2] as well as in a two-dimensional system with harmonic confinement potential [3] with a limited number of configurations considered in the diagonalization. Furthermore, configuration interaction calculations for a numerically determined strain-induced confinement potential [6] and density functional calculations of the energy level structure and luminescence spectra [8] have been performed.

In this paper we study the influence of Coulomb correlations on the optical spectra of QDs. Starting from the localized single-particle states in flat, cylindrically symmetric QDs, described in the envelope function picture, the interacting states are obtained from the full diagonalization of the Hamiltonian including Coulomb interaction. From these states, optical emission spectra are computed

for an increasing number of excitons assumed to be in their (interacting) ground states. Even though the Hamiltonian considered here is the same as in reference [3], the full diagonalization results differ in several respects from the approximate picture obtained there.

Despite the complexity of the spectra, characteristic features appear that can be explained in terms of simplified Hamiltonians. These are obtained from the original Hamiltonian by retaining dominant terms and neglecting less important ones, such that, on the one hand, the essential spectral features are preserved and, on the other hand, analytic results can be deduced. The aim is to get a more intuitive picture of the influence of Coulomb effects and to provide an alternative to a numerically demanding full diagonalization approach. The trade-off between accuracy and simplicity can be reached in several ways and we give here two examples whose merits and shortcomings are assessed against the full calculation result.

A first case is represented by a diagonal Hamiltonian, which is obtained by retaining only the direct and exchange terms, and which describes fairly well the main spectral features. The role of the Coulomb exchange interaction in the ladder-like structure of the lower part (*s*-lines) of the emission spectrum, recently observed in references [4,9] can be easily accounted for in this description.

For explaining the absence of a similar ladder structure in the upper part (*p*-lines) of the spectrum a more careful handling of the Coulomb interaction is required. This is done in the second simplified approach described by an adiabatic Hamiltonian. One encounters here the so-called ‘hidden symmetry’. This property was discussed in a series of papers [10–13]. Nevertheless, the proof of the argument makes use of the assumption that one deals with

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a single degenerate shell, while in fact several, not weakly interacting shells are always present. Therefore it is by no means clear if the ‘hidden symmetry’ result holds, and if it does why and in what form. We show that in the adiabatic approach, and due to certain peculiarities of the problem, the ‘hidden symmetry’ can be recovered, albeit with the parameters renormalized by the interaction between the shells. The discussion of the conditions in which this happens sheds light on the limits of validity of the ‘hidden symmetry’ argument.

2 Hamiltonian

To describe the system of QD electrons and holes under the influence of Coulomb interaction we use the Hamiltonian $H = H_0 + H_{Coul}$ in the envelope-function approximation,

$$\begin{aligned}
H_0 &= \sum_{i\sigma} \varepsilon_i^e e_{i\sigma}^\dagger e_{i\sigma} + \sum_{i\sigma} \varepsilon_i^h h_{i\sigma}^\dagger h_{i\sigma}, \\
H_{Coul} &= \frac{1}{2} \sum_{\substack{ijkl \\ \sigma\sigma'}} V_{ij,kl}^{ee} e_{i\sigma}^\dagger e_{j\sigma'}^\dagger e_{k\sigma} e_{l\sigma} \\
&\quad + \frac{1}{2} \sum_{\substack{ijkl \\ \sigma\sigma'}} V_{ij,kl}^{hh} h_{i\sigma}^\dagger h_{j\sigma'}^\dagger h_{k\sigma} h_{l\sigma} \\
&\quad - \sum_{\substack{ijkl \\ \sigma\sigma'}} V_{ij,kl}^{he} h_{i\sigma}^\dagger e_{j\sigma'}^\dagger e_{k\sigma} h_{l\sigma}. \quad (1)
\end{aligned}$$

Here $e_{i\sigma}$ ($e_{i\sigma}^\dagger$) are annihilation (creation) operators of electrons with spin σ in the one-particle orbital states $|i\rangle$ of energy ε_i^e . The corresponding operators and single-particle energies for holes are $h_{i\sigma}$ ($h_{i\sigma}^\dagger$) and ε_i^h , respectively. The Coulomb interaction matrix elements are given by

$$\begin{aligned}
V_{ij,kl}^{\lambda\lambda'} &= \int d^3r \int d^3r' \xi_{i,\lambda}^*(\mathbf{r}) \xi_{j,\lambda'}^*(\mathbf{r}') \\
&\quad \times V(\mathbf{r} - \mathbf{r}') \xi_{k,\lambda'}(\mathbf{r}') \xi_{l,\lambda}(\mathbf{r}) \quad (2)
\end{aligned}$$

with the band index $\lambda = e, h$, the single-particle wave function $\langle \mathbf{r} | i, \lambda \rangle = \xi_{i,\lambda}(\mathbf{r})$, and the Coulomb potential $V(\mathbf{r}) = e^2/4\pi\epsilon_0\epsilon_r r$, where ϵ_r is the background dielectric constant.

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 [14]. Due to the rotational symmetry around the dot axis the corresponding angular momentum is a good quantum number. For the strong confinement in the 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 confinement in the growth direction will be considered.

A particular situation which is often employed in the literature and will be adopted in the following is the so-called symmetric case, in which one assumes identical envelopes for the valence- and conduction-band electrons,

Table 1. Non-zero Coulomb matrix elements in units of E_c . The indices refer only to the z -component of the angular momentum $m = 0$ for the s -shell and $m = \pm 1$ for the p -shell. The horizontal lines divide the matrix elements into three groups: direct – (top), exchange – (bottom) and other terms.

(i, j, k, l)	$V_{ij,kl}/E_c$
(0, 0, 0, 0)	1.1197
(0, 1, 1, 0), (1, 0, 0, 1)	0.8690
(0, -1, -1, 0), (-1, 0, 0, -1)	0.8690
(1, 1, 1, 1)	0.7935
(1, -1, -1, 1), (-1, 1, 1, -1)	0.7935
(-1, -1, -1, -1)	0.7935
(0, 0, 1, -1), (0, 0, -1, 1), (1, -1, 0, 0), (-1, 1, 0, 0)	0.2507
(0, 1, 0, 1), (1, 0, 1, 0)	0.2507
(0, -1, 0, -1), (-1, 0, -1, 0)	0.2507
(1, -1, 1, -1), (-1, 1, -1, 1)	0.1753

i.e. $\xi_{i,e} = \xi_{i,h}^* = \xi_i$. This assumption holds exactly for QDs modelled by a box-like potential and is found to be a good approximation for oscillator potentials. Note that for the same state index i the electrons and holes have opposite angular momenta. With the assumption of identical envelopes all Coulomb matrix elements are related to the electron-electron ones

$$V_{ij,kl}^{hh} = V_{kl,ij}^{ee} \quad \text{and} \quad V_{ij,kl}^{he} = V_{lj,ki}^{ee}, \quad (3)$$

and the superscript ee of the latter will be skipped in what follows.

The Hamiltonian is fully determined by the effective masses $m_e = 0.065 m_0$ and $m_h = 0.17 m_0$ for the electrons and holes, respectively, the dielectric constant $\epsilon_r = 13.69$ as well as the oscillator length $l_{osc} = 5.4$ nm (describing the in-plane confinement) and the QD height $L = 4$ nm. From these parameters we obtain the single-particle energies with constant spacing $\hbar\omega_e = \hbar^2/m_e l_{osc}^2 = 40.20$ meV and $\hbar\omega_h = 15.37$ meV as well as the energy scale of the Coulomb matrix elements $E_c = e^2/4\pi\epsilon_0\epsilon_r l_{osc} = 19.48$ meV. In the following we will restrict ourselves to a QD containing only s - and p -shells for both electrons and holes. In this case the orbital of the single-particle state $|i\rangle$ can be uniquely identified by its angular-momentum, $m = 0$ for the s -shell and $m = \pm 1$ for the two p -states.

The six-fold integral in equation (2) can be analytically reduced to a one-dimensional integral, which is determined numerically. Due to the cylindrical symmetry of the problem we have angular momentum conservation: $V_{ij,kl} \propto \delta_{m_i+m_j, m_k+m_l}$. All non-zero Coulomb matrix elements are listed in Table 1.

3 Exact diagonalization

In a semiconductor QD the finite height confinement potential leads to a finite number of localized states as well as to energetically higher delocalized states. When the influence of the delocalized states on the discrete QD spectrum is neglected, the eigenvalue problem has a finite (albeit large) dimension and can be solved without further

approximations. In this way the Coulomb interaction between the different possible configurations of carriers in the available bound states is fully taken into account.

As the Hamiltonian conserves the total number of electrons N_e and holes N_h , the Hamiltonian matrix falls into subblocks with basis states corresponding to uncorrelated many-particle states of the form

$$|\phi\rangle = \prod_{\sum_i n_i^e = N_e} (e_i^\dagger)^{n_i^e} \prod_{\sum_j n_j^h = N_h} (h_j^\dagger)^{n_j^h} |0\rangle. \quad (4)$$

Moreover, the Hamiltonian (1) commutes with the total (electron plus hole) angular momentum $l_z = l_z^e + l_z^h$, with the total spin of electrons, \mathbf{S}_e^2, S_e^z , as well as with the total spin of holes, \mathbf{S}_h^2, S_h^z . This rather rich symmetry can be used for separating even smaller Hamiltonian subblocks and for predicting degeneracies. A list of good quantum numbers includes therefore $N_e, N_h, l_z, \mathbf{S}_e^2, \mathbf{S}_h^2, S_e^z, S_h^z$ and the eigenstates are degenerate with respect to S_e^z, S_h^z . By numerical diagonalization of these subblocks one gets the Coulomb-correlated states and the corresponding eigenvalues, classified according to the above-mentioned symmetries.

The coupling of the QD with the optical field is described by the interband dipole operator

$$P = \sum_{\sigma} P_{\sigma} = d \sum_{i,\sigma} h_{i,\sigma} e_{i,-\sigma} \quad (5)$$

and its hermitian conjugate, where d is the interband dipole matrix element. The operator P describes the recombination of “mirror-symmetric” pairs, i.e., having opposite z -components of the angular momentum and spin. The diagonality in the orbital index i is a consequence of the identical envelopes of the electron and hole states. The matrix elements of P define the QD emission as given by Fermi’s golden rule,

$$I(\omega) = \frac{2\pi}{\hbar} \sum_f |\langle \phi_f | P | \phi_i \rangle|^2 \delta(E_i - E_f - \hbar\omega), \quad (6)$$

where the correlated initial (i) and final (f) states and their energies are calculated as described above. The final state has, of course, one electron-hole pair less than the initial one. The hermitian conjugate operator P^\dagger creates mirror-symmetric pairs and appears in a similar formula for the light absorption.

Next, one has to specify the initial states for which the optical spectra will be calculated. In the following we restrict ourselves to situations where optical excitation leads to the same number of electrons and holes within the QD, $N_e = N_h = N^X$ where N^X stands for the number of electron-hole pairs (in the following loosely called excitons). It is further assumed that energy relaxation of carriers within the QD is considerably faster than carrier recombination such that the initial state for the recombination process with given N^X is the corresponding multi-exciton state with the lowest energy (N^X exciton

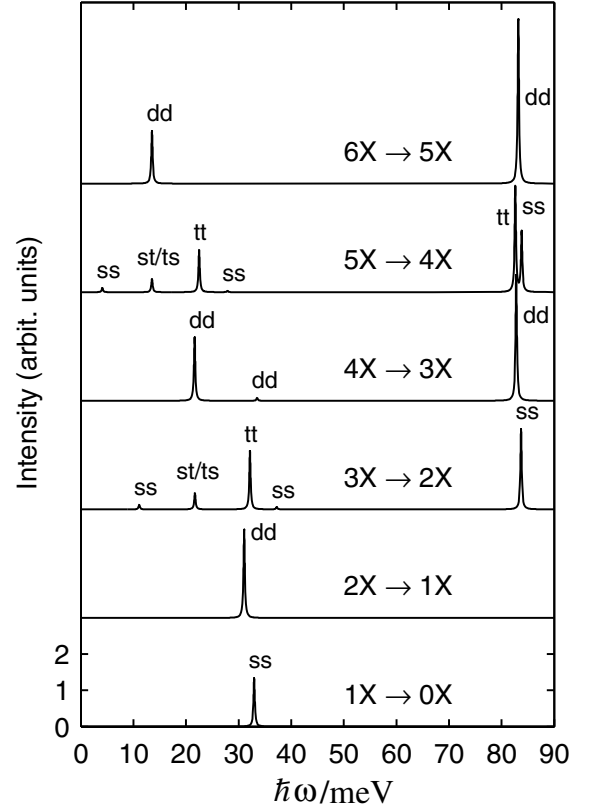


Fig. 1. Ground state emission spectra for a quantum dot with different number of excitons. The labels indicate the total spin of the final state for electrons and holes, respectively: s-singlet, d-doublet, t-triplet. The total angular momentum z -component is always zero. All energies are measured relative to the band-gap.

ground state). Moreover, changes of the carrier spin during relaxation are neglected [15]. Since optical excitation only creates multi-exciton states where electrons and holes have opposite S_z values, only states with vanishing total z -component of the spin are considered as initial states in equation (6).

The analysis of Coulomb-correlated multi-exciton states shows that for even N^X , the symmetry of the ground state is singlet-singlet (ss), i.e., $S_e = S_h = 0$. These states are nondegenerate. On the other hand, for odd N^X one has doublet-doublet (dd) ground states with $S_e = S_h = 1/2$ which are four times degenerate. For the above discussed choice of the initial states for the recombination process only two states contribute to the emission formula with a weighting factor of 1/2 each. The other two states are also dipole-allowed, but are eliminated from the emission formula because their total S_z is nonzero. (By the same arguments one may be concerned about eliminating the states not having total angular momentum zero but, as expected, no such ground states occur.) The final state can be any (ground or excited) state of the system with one exciton removed.

The results of an evaluation of equation (6) based on Coulomb-correlated multi-exciton states are shown in Figure 1 for an initial filling of one up to six electron-hole

pairs. For better visibility the δ -functions are represented by narrow Lorentzians centered at the energy $\hbar\omega = E_i - E_f$ (measured relative to the band gap) and having the area equal to the oscillator strength $|\langle\phi_f|P|\phi_i\rangle|^2$.

Several features are immediately obvious. First, there is a clear spectral separation between the higher energy p -lines for $\hbar\omega > 80$ meV (obtained by removing one exciton from the p -shell) and the lower energy s -lines (for $\hbar\omega < 35$ meV). Second, the position of the s -lines are arranged approximately in a descending ladder as the number of excitons increases, while the energy of the p -lines show a remarkable stability. The last fact was attributed to a ‘hidden symmetry’ property, which will be discussed below. The dipole operator P has no simple commutation relation with the spin operators $\mathbf{S}_e^2, \mathbf{S}_h^2$ and therefore the spin symmetry of the final state is not determined by that of the initial state [16]. This is why one encounters as final states all possible spin symmetries (ss, dd, tt, st, ts), as indicated in the figure.

By restricting the present discussion to s - and p -shells only, the Hamiltonian of equation (1) is identical (up to nonessential differences in the parameters) to the case analyzed in [3]. Nevertheless, the full diagonalization procedure used here leads to different relative line intensities and, more importantly, to the appearance of new emission lines (the st- and ts-lines are missing in [3]).

4 Approximate Hamiltonians

The interaction between different state configurations, as given by the exact diagonalization procedure, shows a quite high degree of complexity and therefore the results are not immediately intuitive. Even though we have considered electron-hole pairs that are optically created in mirror-symmetric states, the Coulomb interaction mixes them strongly with configurations in which the electrons and holes are *not* arranged symmetrically. For instance, promoting two holes from the s -shell to the p -shell is energetically less costly than promoting one electron and one hole. The second case is more symmetric, but the first produces a state which is energetically closer and therefore participating stronger in the exact interacting eigenstate. This may explain the disagreement with the line intensities found in [3], where only symmetric states are considered.

On the other hand, the relatively regular structure of the emission lines seems to indicate that an intuitive picture should be possible. This is achieved by turning to approximate, simpler Hamiltonians which allow analytic solutions and at the same time retain the essential features of the full problem.

One such simplified Hamiltonian can be obtained as follows. By examining Table 1 one sees that the largest Coulomb matrix elements are the direct ones, $V_{ij,ji} = D_{ij} = D_{ji}$. Their contribution to H_{Coul} can be expressed solely in terms of the occupation number operators, $\hat{n}_{i\sigma}^e = e_{i\sigma}^\dagger e_{i\sigma}$ and $\hat{n}_{i\sigma}^h = h_{i\sigma}^\dagger h_{i\sigma}$, and therefore is diagonal in the noninteracting basis, equation (4). The same is

true for the exchange matrix elements $V_{ij,ij} = X_{ij} = X_{ji}$ with $i \neq j$ provided one includes their contribution only in the e-e and h-h interaction terms involving the same spin ($\sigma = \sigma'$). In this way one obtains the diagonal Hamiltonian

$$H_d = \sum_i \left(\varepsilon_i^e - \frac{1}{2} D_{ii} \right) \hat{n}_i^e + \sum_i \left(\varepsilon_i^h - \frac{1}{2} D_{ii} \right) \hat{n}_i^h + \frac{1}{2} \sum_{i,j} D_{ij} (\hat{n}_i^e - \hat{n}_i^h) (\hat{n}_j^e - \hat{n}_j^h) - \frac{1}{2} \sum'_{i,j,\sigma} X_{ij} (\hat{n}_{i,\sigma}^e \hat{n}_{j,\sigma}^e + \hat{n}_{i,\sigma}^h \hat{n}_{j,\sigma}^h). \quad (7)$$

The prime in the last summation indicates that $i = j$ terms have to be omitted and $\hat{n}_i^{e,h} = \hat{n}_{i\uparrow}^{e,h} + \hat{n}_{i\downarrow}^{e,h}$. Of course, for this Hamiltonian there is no configuration interaction. The non-correlated states are eigenstates and the eigenvalues are derived by the above formula by inserting the corresponding occupation numbers.

If the states are symmetrically populated, $n_{i\sigma}^e = n_{i,-\sigma}^e = n_{i\sigma}^X$, their energy is further simplified:

$$E = \sum_i (\varepsilon_i^e + \varepsilon_i^h - D_{ii}) n_i^X - 2 \sum_{i < j} X_{ij} n_{i,\sigma}^X n_{j,\sigma}^X. \quad (8)$$

In this model, the exciton energy is $E_i^X = \varepsilon_i^e + \varepsilon_i^h - D_{ii}$ where the binding energy results from the direct electron-hole Coulomb attraction. When the electron and hole envelope functions are identical, the excitons are not only globally but also locally neutral and the direct electrostatic interaction between different excitons vanishes. In the approximate Hamiltonian (7) the only interaction between excitons comes from the exchange terms and takes place between excitons with the same spin structure [17]. For instance, the biexcitonic binding energy is zero in this approximation (the full model gives a small binding energy of about 2 meV). A comparison of the diagonal model with the full result is given in Figure 2. The approximate spectrum indeed shows a ladder-like structure of the s -lines, with the corresponding spacing in good agreement with the full calculation and given by the exchange interaction between carriers in the s - and p -shell, $2X_{sp}$. According to equation (8) the energy change for the removal of one s -exciton with a given spin structure is proportional to the number of p -excitons having the same spin structure. Adding equal contributions from electrons and holes the coefficient is $2X_{sp}$. Also, in the case of an odd number of excitons one has two s -lines, depending on whether the spins in the removed s -exciton agree or not with the spin orientation in the majority of the p -excitons. For an even number of excitons this splitting does not occur. These main spectral features have been described previously [4,9], here we show which terms of the full Hamiltonian are responsible for them and that these terms can be included in an exactly solvable approximate Hamiltonian H_d .

Even though the diagonal Hamiltonian gives an intuitive picture of the main features in terms of uncorrelated

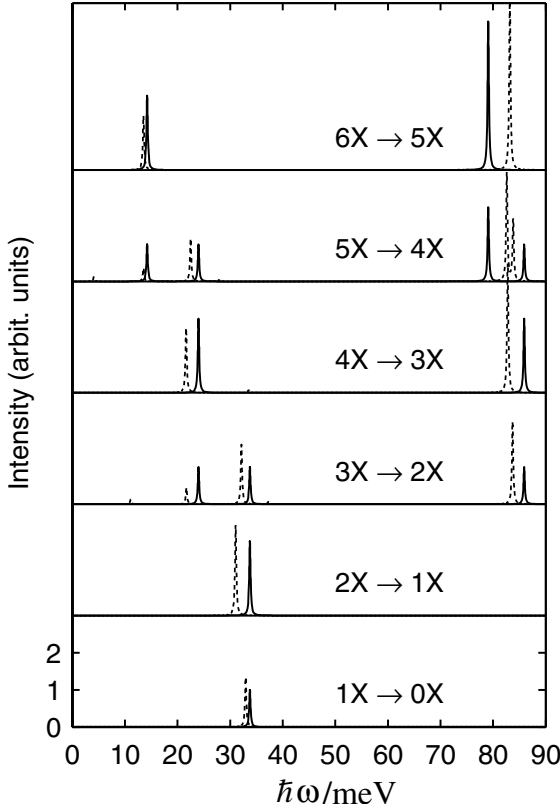


Fig. 2. Comparison of results from the diagonal Hamiltonian, equation (8), (solid line) and the full diagonalization (dashed line) for quantum dot emission spectra with increasing number of excitons.

states, the following shortcomings of this model need to be discussed. The ground state of the 4-exciton problem is inaccurately given by the diagonal model as a triplet-triplet state, as predicted by Hund's rule. It is known that Hund's rule does not always apply in QDs and this is such a case. The true ground state with an energy slightly below this triplet-triplet state has a singlet-singlet symmetry, as mentioned in the previous section. It was this singlet-singlet state which was used as initial state in the emission spectrum obtained from the diagonal Hamiltonian, solid line in Figure 2, in order to make the comparison meaningful. Also the diagonal Hamiltonian entails a ladder-like structure for the p -lines too, with a spacing of $2X_{pp}$, which is not confirmed by the full calculation. As discussed below, this stems from neglecting important pieces from the interaction.

Regarding the p -shell emission, one can refine the approximate Hamiltonian in the following way. An examination of the ground states for a situation with three or more excitons, calculated with the full diagonalization procedure of Section 3, shows that it is safe to assume that in such states the s -shell is completely filled. The states with full s -shell configurations appear with a weighting factor of at least 0.95. Therefore, as far as the ground states and the lower excited states are concerned, it is possible to construct an approximate Hamiltonian describing the

fully interacting p -states following 'adiabatically' an external field provided by the 'frozen' s -carriers. Practically this is obtained from equation (1) along the same lines as before, but this time one enforces diagonality *only with respect to the s -occupation numbers*. In other words, one discards only those terms which contain s -state creation or annihilation operators and cannot be expressed in terms of s -state occupation numbers. In this way, one obtains fully correlated p -states at given s -orbital fillings.

In the resulting 'adiabatic' Hamiltonian $H_{ad} = H_{ad}^{(s)} + H_{ad}^{(p)} + H_{ad}^{(sp)}$ we have separated the terms describing the s - and p -shell as well as the s - p -interaction. The p -shell part retains the form of equation (1) with the summation restricted to the p -orbitals. Therefore, in the following equations, the indices i, j, k, l label only p -states while for the s -states the explicit subscript s is taken. Using the symmetry relations of the Coulomb matrix elements, equation (3), and bringing close the operators with the same spin, $H_{ad}^{(p)}$ can be rewritten as

$$\begin{aligned}
 H_{ad}^{(p)} = & \sum_{i\sigma} \varepsilon_i^e \hat{n}_{i\sigma}^e - \frac{1}{2} \sum_{ijk\sigma} V_{ij,kj} e_{i\sigma}^\dagger e_{k\sigma} \\
 & + \sum_{i\sigma} \varepsilon_i^h \hat{n}_{i\sigma}^h - \frac{1}{2} \sum_{ijk\sigma} V_{ij,kj} h_{i\sigma}^\dagger h_{k\sigma} \\
 & + \frac{1}{2} \sum_{\substack{ijkl \\ \sigma\sigma'}} V_{ij,kl} \left(e_{i,\sigma}^\dagger e_{l,\sigma} - h_{l,\sigma}^\dagger h_{i,\sigma} \right) \\
 & \times \left(e_{j,\sigma'}^\dagger e_{k,\sigma'} - h_{k,\sigma'}^\dagger h_{j,\sigma'} \right) \quad (9)
 \end{aligned}$$

where the new one-particle Coulomb terms result from the reordering of operators. The s -shell part and the s - p -interaction are similar to the diagonal Hamiltonian, equation (7),

$$\begin{aligned}
 H_{ad}^{(s)} = & \left(\varepsilon_s^e - \frac{1}{2} D_{ss} \right) \hat{n}_s^e + \left(\varepsilon_s^h - \frac{1}{2} D_{ss} \right) \hat{n}_s^h \\
 & + \frac{1}{2} D_{ss} (\hat{n}_s^e - \hat{n}_s^h)^2, \quad (10)
 \end{aligned}$$

$$\begin{aligned}
 H_{ad}^{(sp)} = & D_{sp} (\hat{n}_s^e - \hat{n}_s^h) (\hat{n}_p^e - \hat{n}_p^h) \\
 & - X_{sp} \sum_{\sigma} (\hat{n}_{s,\sigma}^e \hat{n}_{p,\sigma}^e + \hat{n}_{s,\sigma}^h \hat{n}_{p,\sigma}^h), \quad (11)
 \end{aligned}$$

where $\hat{n}_{s,p}^{e,h}$ stands for the total population of electrons or holes on the s - or p -shell, respectively. It is important to note that, with the Coulomb matrix elements listed in Table 1, in the one-particle Coulomb terms of equation (9) only the contributions with $i = k$ appear. They are responsible for a renormalization of the one-particle energies $\varepsilon_i^{e,h}$. Moreover, these renormalized energies do not depend on the index i , so that $H_{ad}^{(p)}$ describes a 'single degenerate

shell' [3,10,13], and can be rewritten as

$$\begin{aligned}
H_{ad}^{(p)} = & \left(\varepsilon_p^e - \frac{1}{2}D_{pp} - \frac{1}{2}X_{pp} \right) \hat{n}_p^e \\
& + \left(\varepsilon_p^h - \frac{1}{2}D_{pp} - \frac{1}{2}X_{pp} \right) \hat{n}_p^h \\
& + \frac{1}{2} \sum_{\substack{ijkl \\ \sigma\sigma'}} V_{ij,kl} \left(e_{i,\sigma}^\dagger e_{l,\sigma} - h_{i,\sigma}^\dagger h_{i,\sigma} \right) \\
& \times \left(e_{j,\sigma'}^\dagger e_{k,\sigma'} - h_{k,\sigma'}^\dagger h_{j,\sigma'} \right). \quad (12)
\end{aligned}$$

With the help of the adiabatic Hamiltonian H_{ad} the full emission spectrum can be explained as follows. For the s -lines the arguments showing the formation of a ladder with the spacing of $2X_{sp}$ are as in the case of the diagonal Hamiltonian H_d . The energetic position of the p -shell emission can be deduced from the commutation relation of H_{ad} with the p -shell dipole-transition operator P_p , defined as in equation (5) but with the sum restricted only to p -states. It is readily verified, that

$$\left[e_{i,\sigma}^\dagger e_{j,\sigma} - h_{j,\sigma}^\dagger h_{i,\sigma}, P_p \right] = 0 \quad (13)$$

and therefore P_p commutes with the last two lines of equation (12). This is the core of the 'hidden symmetry' property [10,13] showing that the interaction part plays no role in this argument. The commutation of P_p with the occupation number operators of the shells is rather obvious, because the application of P_p corresponds to a population reduction by one electron-hole pair in the p -states and no change in the s -states. Correspondingly, one can verify $[\hat{n}_p^{e,h}, P_p] = -P_p$ and $[\hat{n}_s^{e,h}, P_p] = 0$. Such simple relations arise only when commuting P_p with the total number operator of the p -shell, not with individual number operators. This is why it is important to have degenerate shells.

Assuming, that the s -states are fully occupied ($n_{s,\sigma}^{e,h} = 1$) one obtains from these results

$$[H_{ad}, P_p] = -(\varepsilon_p^e + \varepsilon_p^h - D_{pp} - X_{pp} - 2X_{sp}) P_p. \quad (14)$$

This shows that the removal of one p -exciton is accompanied by an energy decrease which does not depend on the number of excitons. The value of this energy,

$$\begin{aligned}
\mathcal{E}_p^X &= \varepsilon_p^e + \varepsilon_p^h - D_{pp} - X_{pp} - 2X_{sp} \\
&= E_p^X - X_{pp} - 2X_{sp}, \quad (15)
\end{aligned}$$

is in excellent agreement with the exact diagonalization result. Indeed, using the values in Table 1 one obtains $\mathcal{E}_p^X = 82.5$ meV. The emission spectrum associated with the adiabatic Hamiltonian consists therefore of p -lines having all this common value, while the s -line ladder remains the same as given by the diagonal Hamiltonian.

The 'hidden symmetry' argument, as discussed by Wojs and Hawrylak, [3,10,13] is proven on the assumptions that (i) one has identical envelopes in the two bands and (ii) the one-particle levels form 'a single degenerate

shell'. In these hypotheses one gets a constant energy value at the removal of each exciton, and this value depends only on the parameters of this shell.

Nevertheless, the Hamiltonian under discussion here, as generally used in the literature, is rather describing several interacting shells, so that is not obvious why (if at all) the argument holds. In the present case the answer is contained in the adiabatic Hamiltonian. In it the p -shell is the 'single degenerate shell' because (i) the s -shell is 'frozen' and higher shell are absent in the considered situation and (ii) the field created by the s -shell carriers does not remove the degeneracy of the p -shell. In such conditions the adiabatic Hamiltonian obeys exactly the 'hidden symmetry' commutation relations. In this picture the energy for the removal of a p -exciton, equation (15), contains also terms coming from the s - p -interaction, and it is this value that is in agreement with the full diagonalization.

This discussion also shows the validity range of the 'hidden symmetry'. For instance, the presence of higher shells (but also depending on the actual model parameters) may spoil the argument. This seems to be the case described in [4], where the p -lines are not independent on the exciton number, but are arranged in a descending ladder too. Also, the field of the 'frozen' states may remove some of the degeneracy of the shell in question. For example if the outer shell is a d -shell, the states with zero angular momentum and those with angular momentum ± 2 will experience the field created by the s -shell carriers differently.

One may argue that by approximating the full Hamiltonian, equation (1), with the adiabatic one, equations (9–11), the interaction inside the p -shell is still too complicated to allow analytic diagonalization, i.e. the 'hidden symmetry' property is a simple relation between otherwise complex, strongly correlated states. Nevertheless, the Fock subspace generated by the p -orbitals is significantly smaller and this is in itself a numerical simplification. Symmetry arguments also can be used to reduce the blocks to be diagonalized and at least the ground states can be obtained analytically. A procedure for obtaining analytical eigenstates is the repeated applications of the raising operator P_p^\dagger on the 'vacuum' (full s -shell, empty p -shell) state [3,10,13].

In summary, for semiconductor QDs with finite height two-dimensional harmonic confinement potential the multi-exciton emission spectra are discussed on the basis of a full diagonalization of the Hamiltonian including Coulomb interaction for the localized states. The characteristic features are s -shell lines arranged approximately in a descending ladder with increasing exciton number as well as nearly constant energetic position of the p -shell lines, provided that these are the only confined shells. Based on the relative importance of the various Coulomb matrix elements, a simplified Hamiltonian has been constructed which is diagonal in the single-particle states. It explains the s -shell emission properties, that appear as soon as the p -shell population starts to contribute, in terms of the s - p -exchange interaction X_{sp} . The results are two main s -shell lines for odd number of excitons

separated by $2X_{sp}$, while the spectrum is dominated by a single line for an even exciton number. The almost constant energetic position of the p -shell emission ('hidden symmetry') is discussed in terms of Coulomb correlated p -shell carriers in the presence of a completely filled s -shell. It is also shown that the arguments for the 'hidden symmetry' break down as soon as higher confined shells contribute. Depending on the coupling strength of higher shells to the p -shell the energetic stability of the p -lines is expected to disappear gradually.

The proposed simplified Hamiltonians give a more intuitive picture of the rich properties of the emission spectra from the Coulomb-correlated QD carriers. They also might be an alternative to a demanding full diagonalization scheme when Coulomb interaction in the presence of three and more electron-hole pairs is important. Finally, only a diagonal Hamiltonian allows to discuss physical processes in terms of single-particle states. Our comparison of results from full and diagonal Hamiltonian shows to what extent this is justified.

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16. The operator P commutes with the total spin and therefore some authors, e.g. [2,4], prefer the quantum numbers \mathbf{S}^2, S_z instead of $\mathbf{S}_e^2, \mathbf{S}_h^2$, as being conserved during emission. Nevertheless, P_\uparrow and P_\downarrow separately do not commute with \mathbf{S} so that if one is interested in the polarization of the emitted light there is no advantage in classifying the states by the total spin
17. Since we only consider electron-hole pairs with opposite z -components of the spin, the exciton spin is always zero. Exchange interaction between two excitons is possible when the z -components of the spin for the two electrons agree, which is then also true for the holes