The electronic structure and optical properties of type-II InP/InGaP self-assembled quantum dots and quantum-dot molecules

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Abstract. The electron, hole, and exciton spectra in single quantum dot and quantum-dot molecules consisting of three vertically arranged type-II InP/InGaP self-assembled quantum dots are modelled by the \( \mathbf{k} \cdot \mathbf{p} \) theory. Strain and band mixing are explicitly taken into account in the single particle models of the electronic structure, while an exact diagonalization approach is adopted to compute the exciton states. Reasonably good agreement with magneto-photoluminescence measurements on single InP self-assembled quantum dots is found. As a result of the selection rules, the exciton ground state in the single quantum dot is dark. For the holes, the interplay of strain and mixing enables binding at large distances between the dots in quantum-dot molecules. The exciton states are strongly influenced by the holes, which are able to turn bonding behavior of exciton levels into antibonding for thick spacers.

INTRODUCTION

Recently, there has been an increasing interest in self-assembled quantum dots (SAQD’s) in which carriers can be strongly confined [1]. Various material combinations and dot shapes [1] have been realized by the Stranski-Krastanow mode of epitaxial growth. The density, size, composition, and shape of the quantum dots are determined by the growth conditions (e.g. growth temperature, deposition time, flux etc.). From a theoretical point of view, the calculation of the strain distribution precedes the quantum mechanical modelling of the SAQD’s electronic structure. A handful of approaches to compute the strain are at one’s disposal [2]. One of them being the continuum mechanical (CM) model [2], which perfectly fits a continuum model of electronic structure like the \( \mathbf{k} \cdot \mathbf{p} \) theory, and also may take account of the crystal anisotropy. In contrast to SQD structures, applications of the \( \mathbf{k} \cdot \mathbf{p} \) theory to quantum-dot molecules (QDM’s) are scarce [3].

In this paper, we aim to provide a consistent explanation of the spatial localization of electrons, holes, and excitons in single disk-shaped InP/InGaP quantum dot and vertical stacks composed of these dots. Without strain, the InP/InGaP quantum dot is a type-II structure, but the strain reverts the band alignment to type I. We study the magnetoexciton in a single cylindrical InP/InGaP SAQD [4], and compare the diamagnetic shift with the experimental results [5]. Furthermore, the electron, hole, and exciton states in symmetric stacks of three strained dots are followed as they evolve from the respective SQD states.
THE MODEL

In our approach the strain distribution is extracted from the CM model, which was solved numerically using the finite-element method [6]. The wavefunctions of electrons, holes, and excitons are expanded on the cylinder. Referring to the large band gaps of the constituent materials, the electrons are considered uncoupled from the valence bands, while the mixing between the heavy-hole \((hh)\), light-hole \((lh)\), and the split-off \((so)\) band is explicitly taken into account through the \(6 \times 6\) Pikus-Bir \(\mathbf{k} \cdot \mathbf{p}\) Hamiltonian [6]. For the hole states we use the symbol \(nXf_h\sigma\), where \(X\) denotes the lowest absolute value of the envelope angular momentum \(l\) for the six hole basis states, \(f_h\) is the total angular momentum of the hole in units of \(\hbar\) \((Fzh = f_h\hbar)\), \(n\) denotes the principal quantum number, and \(\sigma\) is the parity of the state.

The exciton parity \(\sigma_{exc}\) is a good quantum number for the axially symmetric exciton, while the total angular momentum composed of the angular momentum of the hole \(Fzh\) and the angular momentum of the electron,

\[ F_{exc} = f_{exc}\hbar = (s + l_e)\hbar - F_{zh}, \]

is also conserved. Here, \(s\hbar\) denotes the electron spin, which is included in the symbol of the exciton state, \(nXf_{exc}\sigma\), where the meaning of the other symbols is similar to the one used for the holes. As a figure of merit for the recombination of an exciton, we use the oscillator strength. Overall the exciton states are fourfold degenerate, which arises from the double degeneracy of both the electron and the hole, and thus exciton states are arranged in quartets.

RESULTS AND DISCUSSION

The material and band structure parameters are taken as in Ref. [6]. For the single quantum dot radius is taken as \(R = 8\) nm, while the dot height is varied in the range \(2.40-2.70\) nm to explore the influence of the dot height on the exciton energies and the diamagnetic shift. For the height of the dots in the quantum dot molecules, we choose three different values, \(h = 2, 3, \) and \(4\) nm, while the thickness of the InGaP spacer varied from \(d = 0\) to \(20\) nm.

The energy levels in 2.5 nm high InP/InGaP SAQD are shown in Figs. 1(a-c). Because the strain confines heavy holes inside the quantum dot and their envelope angular momentum equals zero in the \(1S^{\pm}_{\pm3/2}\) states, these states are the ground hole states in the quantum dot for the whole explored range of \(B\). The magnetic field removes Kramers degeneracy between even and odd parity hole states, and the ground state in each hole shell becomes even, with no noticeable anticrossings in the explored range of \(B\).

We show in Fig. 2(a) the energy of the \(S_{11}^+\) exciton state as it varies with the magnetic field. This exciton state is found to be optically active for in-plane polarized light and to have the lowest energy among all \(bright\) excitons. The best agreement between theory and experiment is found for \(h = 2.55\) nm, which equals approximately 9 monolayer high InP layer. The localization of the exciton states affects the oscillator strength as depicted in Fig. 2(b) for the optically active states in the \(1Q_1^+\) quartet [3].

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The four highest-energy valence-band states in InP/InGaP SQD of: (a) $S^{±}_{3/2}$, (b) $S^{±}_{1/2}$, and (c) $P^{±}_{5/2}$ symmetries. The dot radius amounts to $R = 8$ nm, and the dot height is $h = 2.5$ nm.

The $1S^{±}_{3/2}$ and $1S^{±}_{1/2}$ hole levels in the QDM composed of three quantum dots are shown in Figs. 3(a) and (b), respectively. Decreasing the thickness of the spacer in the regime of hole localization labelled by I in Figs. 3(a) and (b), a maximum in the energy of the $1S^{±}_{3/2}$ and $1S^{±}_{1/2}$ states is reached, after which the bonding behavior follows. Then the hole localization enters the II regime, where the confinement of the light holes in the spacer suppresses the confinement of the heavy holes inside the dots. In the III regime of hole localization, i.e. for $d < 3$ nm, there exists a near degeneracy of states in doublets, which is mainly a result of the existence of large effective quantum dots for the light holes in the matrix [3].

These peculiarities are reflected in the exciton energy levels as function of $d$, as shown in Fig. 3(c) for the lowest energy $Q^{±}_{1}$ and $Q^{±}_{2}$ quartets in the 2, 3, and 4 nm high quantum dots, respectively. All exciton energies in the QDM exhibit triplet ordering and overshoots on the SQD exciton energy levels.
FIGURE 3. The valence band levels in QDM as a function of the spacer thickness for: (a) $S_{3/2}$ states and (b) $S_{1/2}$ states. The dot height is 3 nm. (c) Variations of the energies of the even exciton states in QDM with the spacer thickness. The lowest energy $Q_{1}^{+}$ and $Q_{2}^{+}$ exciton quartets are shown.

CONCLUSION

The hole and exciton states in flat cylindrically shaped InP/InGaP self-assembled quantum dots was modelled using the multiband $k \cdot p$ theory and exact diagonalization approach, respectively. The light holes are mainly responsible for the coupling of the hole states in the quantum-dot molecule. Moreover, the exciton energies in the quantum-dot molecule exhibit blue shifts with respect to the energies in the single quantum dot.

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REFERENCES