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Vladan Mlinar
Antwerp
June, 2007
Chapter 1

Introduction

The question is not what you look at, but what you see.

Henry David Thoreau

1.1 Quantum dots - interesting physics and various applications

Semiconductor quantum dots (QDs) are three-dimensional (3D) artificial semiconductor based structures with size of a few nanometers in all three directions (Shchukin et al. 2003, Bimberg et al. 1998). Their confinement, smaller than de Broglie wavelength in semiconductors, leads to a discrete energy spectrum and a delta-function atomic-like density of states, which enables the analogy with real atoms. Therefore, QDs are often referred to as artificial atoms although containing from $10^5$ to $10^6$ atoms. Furthermore, the coupling between QDs to obtain new functional units, leads to a formation of quantum dot molecules (QDM) (Schedelbeck et al. 1997), and extends the analogy between real and artificial atoms to the molecular realm (Kouwenhoven et al. 2001).

The growth conditions mainly determine the properties of QDs and QDMs. Various techniques have been used in fabrication of QDs (Alivisatos 1996, Leonard et al. 1993), but small size of dots ($\sim 10$nm), self-assembling and possibility to produce billions of highly uniform dots per cm$^2$ of semiconductor wafer in a single technological step make Stranski-Krastanov (SK) growth mode among all other methods widely used in the QD formation (Shchukin et al. 2003, Bimberg et al. 1998, Shchukin and Bimberg 1999). Such a formation of QDs, driven by local strain fields, not only that enables formation of defect-free (coherent) islands, but also enables the formation of vertically stacked QDs, i.e. QDMs. Namely, the creation of vertically stacked QDs is impelled by the strain that surrounds a quantum dot in the lower lying layer which enforces the location of the next QD on top of the first one (Xie et al. 1995, Heitz et al. 1998).

The occurrence of strain fields in and around the dots and significant intermixing, tightly connected with the SK growth mode, as well as 3D confinement present in the QD systems that leads to multi-band and inter-valley mixing, bring a head new features of QDs and QDMs as compared to real atoms and molecules.
For example, He et al. (2005a) have shown that while the electron loading to the QD follows the Aufbau principle and Hund’s rule, as known from atomic physics, the hole loading violates both, the Aufbau principle and Hund’s rule. Furthermore, placing a QD in an external magnetic field opens up a new playground for an investigation of spin-related properties making the magnetic field an interesting tuning parameter. Alternatively, presence of a magnetic field enables a study of a competition between quantum confinement, Coulomb interaction and magnetic field confinement, experimentally observable through magneto-photoluminescence measurements. In principle, manipulation of the QDs’ properties is driven by current and potential applications of QD systems, ranging from QD lasers (Kirstaedter et al. 1994), a possible physical representation of a quantum bit (Troiani et al. 2003), to nanosensors used to detect DNA molecules (Zhang et al. 2005).

Tasks put in front of theoreticians are far from being trivial. The electronic and optical properties of QD system for both, fundamental study and potential QD application, have to be extracted from the detail theoretical models that include a realistic geometry and composition profile of the QDs, as obtained from experiment, to take into account the strain distribution in and around the dots, effects of piezoelectricity, multi-band and inter-valley mixing, originating from the 3D confinement, and also to describe effects of external electric and/or magnetic fields. Qualitative agreement between theoretical findings and experimental data, as well as the predictive power of the theory depend not only on the complexity of the employed model, but also on the accuracy of the experimental input on the structural properties of QDs. The latter may alter the calculated electronic structure of the QD system so strongly, that agreement with the experiment of even thorough models becomes problematic, and theoretical predictions questionable.

In this thesis we consider SK fabricated QDs made of III-V zinc blende semiconductors (see Figs. 1.1 and 1.2) with a goal to provide a consistent theoretical description of the electronic and optical properties of such QDs grown on arbitrary substrate orientation and/or placed in an external magnetic field.

In what follows, we further elaborate the above mentioned statements one by one. First, an overview of the existing semiconductor nanostructures is made in Sec. 1.1.1 Sec. 1.1.2 is devoted to the novel QD based optoelectronics devices. The objectives of this thesis are put forward in Sec. 1.1.3 Prior to answering these objectives, we recapitulate techniques
1.1. Quantum dots - interesting physics and various applications

Figure 1.2: (a) First Brillouin zone of a zinc-blende semiconductor. (b) A typical band structure: GaAs.

used for the QDs’ growth in Sec. 1.2 and Sec. 1.3. Structural characterization of QD system is explained in Sec. 1.4. Also, we address the optical measurements performed on such systems in Sec. 1.5 and finally, theoretical approaches employed for the modelling of the electronic and optical properties of the QD systems are discussed in Sec. 1.6. At the end of the Introduction, in Sec. 1.7 we give the details (derivation, advantages, and disadvantages) of our theoretical approach.

1.1.1 From bulk to quantum nanostructures: wells, wires, dots

In semiconductors the electron energy is a multivalued function of momentum resulting in energy bands, continuous density of states, and gaps whereas the primarily interest is in the valence band and conduction band. Within this thesis we focus only on zinc-blende based direct gap materials, meaning that the conduction band minimum and valence band maximum are at the $\Gamma$ point.\footnote{Strictly speaking, the valence band maximum is slightly shifted from $\Gamma$ point for zinc-blende semiconductors.} Crystal structure of zinc-blende lattice is shown in Fig. 1.1 and the first Brillouin zone with points of high symmetry is shown in Fig. 1.2(a). For most applications we are interested in what happens near the top of the valence band and the bottom of the conduction band (around $\Gamma$ point in our case). These states originate from the atomic levels of the valence shell in the elements making up the semiconductor. As an illustration, a typical band structure of a direct gap zinc-blende semiconductor, GaAs, is plotted in Fig. 1.2(b).

A semiconductor heterostructure is called to be of reduced dimensionality, when the motion of at least one type of charge carriers is confined in at least one direction within a spatial extent comparable to the de-Broglie wavelength $\lambda_B = 1.22 nm / \sqrt{E_{kin}}$ (eV) of the carriers. The carriers momentum in that direction is quantized and its energy spectrum is given by the discrete solutions of the Schrödinger equation, the eigenenergies. As a consequence the carrier has a non-vanishing minimum kinetic energy, the quantum confinement energy. For
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![Density of states of the bulk, quantum well, quantum wire, and quantum dot.](image)

**Figure 1.3:** Density of states of the bulk, quantum well, quantum wire, and quantum dot.

Localization of carriers in semiconductor nanostructures: Depending on the band gap of the constituent semiconductors forming nanostructure, three cases can be distinguished. The structures are called type I if both, electrons and holes, are confined in the same structure (well, wire or dot), i.e. when the electron and hole wavefunctions have a significant spatial overlap. When only one carrier type is spatially confined, the structure is called type II. For this case the unbound carrier type normally gets localized in the vicinity of the low-dimensional structure due to Coulomb interaction and sometimes also by strain. A structure is said to be of type III, if not only that both, electrons and holes, are confined in the same structure, as in the case of type I structures, but also the conduction and the valence bands are overlapping. Fig. 1.4 illustrates our discussion. Namely, direct $\Gamma$ valley energy gap for zinc-blende semiconductors as it varies with the lattice constant is shown in Fig. 1.4(a), and the examples of type I, type II, and type III structures are plotted in Fig. 1.4(b).

Density of states of nanostructures: A reduction in the dimensionality (from bulk material towards wells, wires, or dots) leads to changes in the density of states (D(E)) of the quantum confined carrier. Corresponding density of states for the case of bulk material, QW, QWR, and QD are also shown in Fig. 1.3 As an important consequence, the dependence of D(E) on the dimensionality impacts the thermally induced broadening of the carrier distribution n(E), which is given by the product $n(E) = D(E) \times f(E)$, where f(E) is the Fermi function. The large confinement energies decrease thermal broadening, i.e. for large confinement energies the sublevel splitting $E_{i+1} - E_i \gg k_B T$, and only one sublevel gets thermally occupied reducing the thermal broadening. It is, therefore, expected that the carrier distribution function for QDs to be independent of temperature.
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The peculiar features of quantum dots:
Although the QD-real atom analogy can give some information about properties of QD system, the interesting features actually arise due to specific growth conditions (existence of strain field), 3D confinement, Coulomb interactions present in multi-charged QDs. QDs’ electronic structure is strongly influenced by the strain field and this dependence is reflected through the modification of energy band gaps, lifting up the heavy-hole light-hole degeneracy at zone center (Chao and Chuang 1992, Bimberg et al. 1998), and piezoelectric field (Grundmann, Stier and Bimberg 1995, Stier et al. 1999, Bester and Zunger 2005, Bester, Zunger, Wu and Vanderbilt 2006, Miglioratio et al. 2006) arising from the non negligible shear components of the strain tensor. In case of 3D confinement, the confinement energies of QDs are much larger than those of quantum wells because of the strong confinement of the carriers not only in one direction (e.g. along growth axis as in the case of QWs) but also in the layer plane. The confinement and hence the localization of electron and hole states comes into play due to their resulting spread in \( k \)-space, and the accompanying contributions far from the \( \Gamma \) point. As a consequence, the effect of band non-parabolicity becomes very important, and is reflected through strong band mixing and even inter-valley mixing. Depending on the QDs’ geometry and composition profile, numerous levels can be confined in the conduction band and even more in the valence band (Stier et al. 1999, Pryor 1998). In the case of more than one charge carrier (electron, hole, or both) localized in a QD, many-body states are formed automatically, with different energies as compared to simple conduction and valence band states (see Sec. 1.6.4). Rich emission

Figure 1.4: (a) Direct \( \Gamma \)-valley energy gap as a function of lattice constant for the zinc blende form of 12 III-V binary compound semiconductors (points) and some of their random ternary alloys (curves) at zero temperature (taken from the publication of Vurgaftman et al. (2001)). (b) Illustration of type I, type II, and type III types of confinement in nanostructures. Red (blue) lines refer to valence (conduction) band.
spectra depending on selection rules, dipole lengths, or polarizations, of such a system open playground for both, fundamental studies and potential device applications of QDs.

**Quantum dots for device application:** If one wants to use QD systems for device application, three conditions have to be fulfilled: (i) Large sublevel splitting, implying an upper boundary for the structure size and a lower one for the depth of the attractive potential. (ii) The localization energy of the carriers should be large as compared to the thermal energy. Otherwise, the confined carriers can be thermally activated to leave the low-dimensional structure at elevated temperatures. This condition includes the need for at least one confined carrier state, which is only fulfilled for QDs exceeding a certain critical size. In general, the requirement for a large localization energy is equivalent to a lower boundary for the spatial extent and the depth of the attractive potential. (iii) Size and composition uniformity of QDs in an ensemble are required for e.g. QD laser or QD infrared photodetectors, but not for mode-locking or semiconductor optical amplifiers that need such non-uniformity in order to obtain picosecond pulse width or wavelength multiplexing.

1.1.2 Towards novel applications

The persistent interest in QDs is driven by the multitude of their actual and potential applications ranging from novel lasers (Kirstaedter et al. 1994, Bimberg et al. 1997, Tatebayashi et al. 2005, Heinrichsdorff et al. 2000), and optical amplifiers (Uskov et al. 2004) to physical representations of a quantum bit (Troiani et al. 2003, DiVincenzo 1995, Imamoglu et al. 1999), or single polarized photon sources emitting "quantum bits" (Benson et al. 2000). For the illustrative purposes, we address here applications of QDs for lasers, quantum dot infrared photodetectors, optical amplifiers, and single photon sources.

**Quantum dot laser:** Arakawa and Sakaki (1982) predicted a temperature independent threshold current for a laser based on QDs with infinitely high barriers. From the experimental point of view, (In,Ga)As/GaAs QDs are usually used as an active medium. In order to provide a confinement energy comparable to the thermal energy at room temperature, an In(Ga)As QD in a GaAs matrix should have a size in the range 5-20 nm. The first MBE-grown InGaAs/GaAs QD laser was demonstrated by Kirstaedter et al. (1994). The optimization of the growth parameters and the laser design since this first report has led to QD lasers with low threshold current densities and high characteristic temperatures, thus confirming the early predictions. As with the demonstration of the advantages of the quantum well laser that preceded it, the full promise of the quantum dot laser must await advances in the understanding of the materials growth and optimization of the laser structure. Although the self-assembled dots have provided an enormous stimulus to work in this field, there remain a number of critical issues involving their growth and formation: greater uniformity of size,
1.1. Quantum dots - interesting physics and various applications

Figure 1.5: (a) Schematic of a QD laser structure. (b) P-I characteristics and emission spectrum of a quantum dot laser diode (http://cqd.eecs.northwestern.edu/research/qdots.php).

Controllable achievement of higher quantum dot density, and closer dot-to-dot interaction range will further improve laser performance. Better understanding of carrier confinement dynamics and capture times, and better evaluation of loss mechanisms, will further improve device characteristics. As an illustration, we show a QD laser diode in Fig. 1.5(a), and P-I characteristic and emission spectrum in Fig. 1.5(b). Stimulated emission was observed at 995 nm with an injection current of 400 mA (These results were reported at the Center for Quantum Devices at the Northwestern University).

It should be noted that the spatial localization of carriers brought about by the quantum dot confinement may play a role in the "anomalous" optical efficiency of GaN-based materials, which is exceptional in light of the high concentration of threading dislocations (∼108 - 1010 cm⁻²) that currently plague this material system. The localization imposed by the perhaps natural nanostructure of the GaN materials may make the dislocation largely irrelevant to the purely optical (but not to the electrical) behavior of the material.

Quantum dot infrared photodetectors: Multi-stack (In,Ga)As quantum dot growth on both, InP and GaAs, have been used for infrared photodetector (QDIP). Such QDIPs have many advantages compared to the conventional quantum well infrared photodetector (QWIP), including: higher responsivity, higher temperature operation, higher light coupling to normal incidence light, and capability of narrow band nability. Illustration of the cross section of a QDIP is shown in Fig. 1.6(a). A GaAs-based QDIP operating at 9 µm at 77K, utilizes a InGaAs/GaAs Dot-in-a-Well structure (DWELL) with In-GaP barriers.

http://cqd.eecs.northwestern.edu/research/qdots.php
Figure 1.6: (a) Illustration of the cross section of a QDIP. (b) Thermal imaging of the human body taken by the 256×256 InGaAs/GaAs/InGaP DWELL FPA (http://cqd.eecs.northwestern.edu/research/qdots.php).

In addition, the QDIP focal plane array (FPA) was also demonstrated, based on the InGaAs/InGaP/GaAs QDIP with a 256×256 format. The image could be seen at temperatures as high as 120 K. Thermal image of human hand as imaged by QDIP is shown in Fig. 1.6(b) (These results were reported at the Center for Quantum Devices at the Northwestern University).

Optical amplifiers: *(from the PhD thesis of Lämmlin (2007))*. An optical amplifier is conceptually a gain medium through which light is transmitted and amplified by stimulated emission. The carrier population in the active region of the semiconductor optical amplifier (SOA) waveguide is inverted by electrical pumping. An input signal coupled to this waveguide travels along the device and experiences amplification by stimulated emission. At the opposite end the amplified light exits the semiconductor chip. A semiconductor optical amplifier is very similar to a semiconductor laser with the main difference that the resonator of the laser is removed. Ideally, an optical mode is travelling only once through the SOA which is also called travelling wave amplifier. Light can be generated and amplified by stimulated emission from electron-hole recombination in an active semiconductor region. The high carrier densities needed for this condition are created in a semiconductor by optical excitation or preferably current injection. Therefore, a structure is needed that is able to confine and guide light in a dielectric waveguide and that provides a carrier injection and confinement as well.

A pn-heterojunction is used for carrier injection and confinement. It is realized by a narrow-gap semiconductor, which can be p-, n-type or undoped, that is sandwiched between two higher-gap p- and n-type semiconductor layers. The carriers that are injected into the

3http://cqd.eecs.northwestern.edu/research/qdots.php
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Figure 1.7: (a) Schematic, in vertical growth direction (z) of a GaAs/AlGaAs pn-heterojunction with two QD layers in the active region. (b) Energy band edges of a forward-biased double heterostructure. Electrons and holes are trapped in the potential well formed by the conduction/valence band (CB/VB) edge discontinuity and are then captured in the wetting layer and QD states. (c) Spatial profile of the index of refraction which creates a dielectric waveguide in the active region layer. (d) Intensity profile of the fundamental optical mode in a three-layer waveguide (taken from the PhD thesis of Lämmlin (2007)).

active region are prevented from diffusing out by means of the potential barrier due to the difference between the energy gaps of the active layer and the doped claddings. This structure does not only inject and confine electrons and holes at the heterojunction, also the index of refraction of the lower-gap region is larger than for the higher-gap material. The energy band diagram, the refractive index distribution and the fundamental mode of a double heterostructure, including two QD layers in the active region, at forward bias are depicted in Fig. 1.7.

Using QDs in the active region brings a head several improvements as compared to standard SOA. For example a wide spectral bandwidth is expected, the unique dependence on
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Figure 1.8: (a) DBR micro-cavity post structure and the map of the energy density. (b) Photon correlation histogram for a QD on resonance with the cavity under pulsed, resonant excitation (taken from publication of Vuckovic et al. (2003)).

the linewidth enhancement factor and consequently regions of decoupled gain and refractive index propose interesting cross gain and phase characteristics, the temperature performance of the devices is improved, ultrafast gain relaxation can help to build SOAs working at high speed, and their saturation output power could be larger than for conventional SOAs as the dominant saturation mechanism is spectral hole burning at large pump current.

Single photon sources: An ideal triggered source of single photons emits one and only one photon in each pulse that is different from a “classical” source such as an attenuated laser, where the number of photons in each pulse typically follows a Poisson distribution. Complete characterization of a single-photon source should address the following issues:

(i) Reduction of the two-photon probability relative to a Poisson distribution. (ii) The collection efficiency is enough to allow high repetition frequency. (iii) The photons can be extracted with high efficiency into free space or a fiber. (iv) The radiative lifetime of the emission is short to have good coherence and indistinguishability.

Usually self-assembled InAs quantum dots in GaAs were investigated for usage as single photon sources. These structures have advantages as single-photon emitters, including larger quantum confinement energies and high radiative efficiency. The operation temperature can be increased to ~10-20K with little degradation of performance, and up to ~50K with line broadening.

Single-photon emission can be enhanced with distributed Bragg reflector (DBR) cavity (Pelton et al. 2002, Vuckovic et al. 2003), or photonic-crystal cavity. In Fig. 1.8(a) we show DBR micro-cavity post structure and the map of the energy density. Results obtained
from photon correlation measurements are shown in Fig. 1.8(b). The distance between the peaks corresponds to the repetition period of pulses from the Ti:sapphire laser. The vanishing central peak indicates a strong antibunching and a large suppression of multiphoton sources (Vuckovic et al. 2003).

One problem with quantum dots as single-photon sources is that each one has a unique emission wavelength and radiative lifetime. Therefore, two single-photon sources built with quantum dots would not likely be compatible with each other for information processing operations. Another solid-state system that could potentially work much better in this respect is impurity-bound excitons. Furthermore, if one locks out a single QD, the main requirement to create an on-demand entangled photon source, is that photons are distinguishable by polarization only i.e. linear polarization must be less than the homogeneous linewidth of the emission. However, the existence of the spin splitting of the intermediate exciton level results in polarization-correlated photon pairs. Such a splitting is attributed to the breaking of the in-plane symmetry of the electron-hole exchange interaction originating from e.g. QDs’ shape or strain anisotropies.

Recently, Seguin et al. (2005) reported a variation by one order of magnitude as well as a sign inversion of the exciton fine structure splitting with the QD size. Furthermore, it was very recently reported by Seguin et al. (2006) and Ellis et al. (2007) that the exciton splitting of an individual QD can be tuned through zero by thermal annealing (see Sec. 1.3.3 for details about thermal annealing). In Fig. 1.9 we show a polarization dependent PL from a QD for successive anneals. The excitonic fine-structure splitting decreases from 170±20µeV to less than 20µeV after the second annealing step (taken from the publication of Seguin et al. (2006)).

1.1.3 The scope of the thesis

There has been a great deal of experimental work devoted to the fabrication of dots with desired properties e.g. good size homogeneity, known composition profile, emission at the

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4The localized impurity-bound exciton recombination line is narrow, determined entirely by the basic properties of its host semiconductor, and when embedded in a cavity identical photon emitters should be possible.
desired wavelengths etc. Once the dots are fabricated their structural and optical properties are being investigated experimentally. First, information about QDs’ geometrical properties and composition profile are extracted from the structural characterization, also information about strain components (see Sec. 1.4.1), and even about confined electron wavefunctions (see Sec. 1.4.1). Finally, optical measurements are performed on such dots. In addition, dots can be placed in an external magnetic or an electric field.

This thesis has two goals:

I. To provide an elegant theoretical model for studying the electronic and optical properties of self-assembled QDs taking into account all the relevant effects, where dots can be grown on high index planes or placed in an external magnetic or electric field (Mlinar and Peeters 2007b). In that respect, three of my main contributions are:

(i) Reporting the existence of nonphysical solutions in multi-band effective mass model applied to nanostructures caused by the ill-defined boundary conditions at the nanostructure-barrier interface.

(ii) Proposal of an improved theoretical model for the electronic structure calculations of nanostructures placed in an external magnetic field (Mlinar et al. 2005).

(iii) Numerical implementation of 3D model for the electronic structure calculation of an arbitrary 3D nanostructure grown on high index planes and/or in presence of an external magnetic field (Mlinar and Peeters 2007b).

II. To explain the underlying physics of the less investigated QD systems. In that respect, we study:

(i) The influence of the substrate orientation on the electronic and optical properties of InAs QDs and InAs QDM. The later contain up to eight lateral and vertically coupled QDs. Our theory provide a guideline for the variation of the electronic and optical properties of QDs going from the well investigated [001] grown QDs to [11k] grown QDs, where $k = 1, 2, 3$ (Mlinar and Peeters 2006a, Mlinar and Peeters 2006b, Mlinar and Peeters 2007a, Mlinar and Peeters 2007c).

(ii) Two cases of type II QDs have been discussed: InP/InGaP QDs and molecules in an external magnetic field (Mlinar et al. 2006, Tadić et al. 2005), and InAs QDs capped with Ga(As,Sb). In both considered cases holes are placed outside the dot and in the former case are localized due to strain, and in the latter due to Coulomb interaction with electrons in the dot. Novel features of such systems are expected.

(iii) The electronic and optical properties of unstrained self-assembled QDs with precisely known sizes and shape representing an ideal test case for a theoretical model. Our findings are compared with the experimental results (Mlinar et al. 2007).
1.2 Fabrication of quantum dots

The reliable production of QDs offers outstanding opportunities for optical and electronic technologies as well as the development of new technologies. First attempts to produce QD systems with sufficient optical quality were based on conventional post-growth lithography and etching methods or epitaxy on patterned substrates. The main problem with these techniques is introduction of interface damages and impurities, and relatively poor resolution of post-growth lithography methods. The more successful techniques are based on in-situ growth of self-assembled QDs, where nucleation at desired sites is promoted introducing nonplanar features or strained patterns. Furthermore, nano-capillarity effects on nonplanar surfaces are utilized to generate QD systems that form inherently at prescribed position on a substrate. In general, demands in the fabrication of QD systems are ranging from precise position control i.e. to achieve ordered QD systems, or tailored optical emission and absorption, to effective integration with photonic devices such as optical cavities, waveguides, and photonic crystals.

In what follows, we briefly introduce various techniques used in QD fabrication. It is our intention with this Section to draw the attention of the reader on the most important techniques employed to QD manufacturing without going into the details. For a detailed review on growth techniques see Bimberg et al. (1998) or the more recent work by Shchukin et al. (2003).

1.2.1 Lithographic techniques

Firstly, lithographic patterning and etching of QW structures, techniques that include selective intermixing of QW, or use of stressors have been employed for QD fabrication. QDs with various interesting features have been produced, but are not the subject of the present thesis. The widely used lithographic techniques are: optical lithography and holography, X-ray lithography, electron and focused ion beam lithography, and scanning tunnelling microscopy.

1.2.2 Self-organization concept

Spontaneous formation of periodically ordered domain structures in solids with periodicity much larger than the lattice parameters can nucleate if the homogeneous state of the system is thermodynamically unstable and the system undergoes a phase transition into an inhomogeneous state. Then, a coordinate-dependent order parameter becomes a source of a long range field (in our case strain). A multidomain state of the solid is energetically more favorable than the single-domain state because the multidomain state provides compensation of the long-range field at large distances outside the domain structure (Shchukin et al. 2003, Bimberg et al. 1998).
When the lattice constants of the substrate and the crystallized material differ considerably, only the first deposited monolayers crystallize in the form of epitaxial, strain layers with the lattice constant equal to that of the substrate. When the crystal thickness exceeds certain value, a significant strain is accumulated in the layer which leads to the break-down of such an ordered structure and to the spontaneous creation of randomly distributed islands of regular shape and similar sizes. The shape and average size of these islands depend mainly on factors such as the strain intensity in the layer as related to the misfit of the lattice constants, the temperature at which the growth occurs, and the growth rate. The phase transition from the epitaxial structure to the random arrangements of islands is called Stranski-Krastanov (SK) transition. When the process of crystallization is terminated shortly after reaching the phase transition, the islands evolve to the state of quasi-equilibrium in which they assume the shape of pyramids or lenses formed on a thin wetting layer. The QDs formed in the SK growth are called self-assembled QDs and are the subject of this thesis. Therefore, whenever we refer to QDs, we are talking about self-assembled QDs.

1.3 Stranski-Krastanow growth mode

As discussed above, SK growth mode for a fabrication of QDs uses the natural lattice mismatch between the substrate and the deposited material. The main advantages of the SK growth mode are fabrication of defect-free QDs of small sizes, homogeneity in sizes and shapes of QDs, formation of ordered arrays of 3D coherent islands, and fairly convenient growth processes. The main problems, however, are non-uniformity of QDs on a sample due to the QDs geometry and composition variation during the growth process. These drawbacks appear to be the most dominant limiting factors in the application of these systems to optoelectronic devices.

Using suitable growth conditions by Molecular Beam Epitaxy (MBE) and Metal-Organic Chemical-Vapour Deposition (MOCVD) it is possible to fabricate arrays and stacks of small QDs. QDs’ density is around $4 \times 10^{10}$ cm$^{-2}$, and various geometries for coherent islands have been reported: lenses, square-based or elongated pyramids and truncated pyramids with different facet orientations. In what follows we briefly introduce those two growth techniques.

1.3.1 Molecular Beam Epitaxy

MBE takes place in high vacuum or ultra high vacuum ($10^{-8}$ Pa) and is used for very clean growth when a limited amount of deposited material is necessary. The most important aspect of MBE is the slow deposition rate (0.001 to 0.3 $\mu$m/minute), which allows the films to grow epitaxially, but on the other hand, it requires proportionally better vacuum in order to achieve the same impurity levels as other deposition techniques. MBE growth technique
allows studies on epitaxy, because through the reflecting high-energy electron diffraction (RHEED) technique it is possible to follow the growth layer after layer and detect the formation of 3D structures. A computer controls shutters in front of each furnace, allowing precise control of the thickness of each layer, down to a single layer of atoms. An MBE apparatus consists of a UHV chamber, with several solid-state sources (Knudsen cells) directed to the substrate, which usually can rotate to make the epitaxy more homogeneous. In solid-source MBE, ultra-pure elements such as gallium and arsenic are heated in separate quasi-Knudsen effusion cells until they begin to slowly evaporate. The evaporated elements then condense on the wafer, where they may react with each other. In the example of gallium and arsenic, single-crystal gallium arsenide is formed. The term “beam” simply means that evaporated atoms do not interact with each other or any other vacuum chamber gases until they reach the wafer, due to the long mean free paths of the beams.

1.3.2 Metal-Organic Chemical-Vapour Deposition

MOCVD is a chemical vapour deposition method of epitaxial growth of materials, especially compound semiconductors from the surface reaction of metalorganics compounds or metal hydrides containing the required chemical elements. In contrast to MBE, the growth of crystals is by chemical reaction and not physical deposition, where formation of the epitaxial layer occurs by final pyrolysis of the constituent chemicals at the substrate surface. As compared to MBE that takes place in high vacuum or ultra high vacuum, MOCVD takes place at moderate pressures (2 to 100 kPa). As such this technique is preferred for the formation of devices incorporating thermodynamically metastable alloys.

1.3.3 Annealing of quantum dots

Annealing is a post-growth procedure to tune the properties of nanostructures by subjecting it to heat treatment. It enables the manipulation with the energy levels of self-assembled QD ensembles by tuning the intersublevel energy-spacing and the number of confined states. Namely, the interdiffusion of self-assembled QDs can be treated by annealing (e.g. thermal or by laser), and three main effects of alloy intermixing were demonstrated in QDs: (i) The emission has been strongly blue-shifted. (ii) The intersublevel spacing can be tuned. (iii) The inhomogeneous broadening can be linearly decreased, for example ranking from a FWHM of 46 meV down to smaller than 15 meV for rapid thermal annealing and 8 meV in the most extreme case of laser annealing as reported by Allen et al. (2000). Furthermore, Seguin et al. (2006) and Ellis et al. (2007) showed that thermal annealing may be used for substantial reduction of the excitonic fine-structure splitting.
1.4 Structural characterization

Information about size, shape, and composition profile of fabricated QDs are of fundamental importance for understanding their electronic and optical properties. The primary methods for structural characterization can be divided into direct imaging methods and diffraction methods (Shchukin and Bimberg 1999). In what follows, we highlight widely used techniques and address their advantages and disadvantages.

1.4.1 Direct imaging methods

Three experimental techniques will be discussed here: Atomic force microscopy (AFM), scanning tunnelling microscopy (STM), and transmission electron microscopy (TEM).

Actually, STM, AFM, as well as the near-field scanning optical microscopy (NSOM), which will not be discussed here, belong to the scanning probe microscopy (SPM). SPM is currently a well established technique in surface science to diagnose deposited and etched surfaces and to elucidate the associated surface chemistry on the atomic scale. Another use of SPM is to modify surfaces, producing nanostructures whose properties display many quantum characteristics usually associated with individual molecules.

Atomic Force Microscopy: AFM plan-view measurements are performed on uncovered islands. The main advantage of this method is the possibility to reveal the morphology of a surface on a atomic level since AFM has atomic resolution of typically a few nanometers in lateral direction, and of 0.1nm resolution in the growth direction. Of course, the actual resolution depends on the specific size and shape of the tip whereas tip effects can modify the measured height of the dot, and to a large extent the lateral size of the dot.

Actually, the AFM as well as the STM, as will be shown later, deliver a convolution of the island geometry with tip-surface interaction function. Also, the AFM measurements are usually not performed at the growth temperature but a sample is first cooled down. It results in severe modifications of the dot size and density due to changes in growth conditions. Tracing the
1.4. Structural characterization

changes of the QD morphology with decreasing temperature as well as in the dependence of the AFM measurements on the rate of cooling is a tedious task.

Furthermore, the AFM can be used for the fabrication of high-quality InAs QD arrays (Kim et al. 2006). The highly aligned defect-free nanoholes as nucleation sites were fabricated by using AFM tip-induced nano-oxidation and subsequent atomic-hydrogen etching/cleaning. Next, using the droplet epitaxy on artificially patterned nanoholes, Kim et al. (2006) fabricated laterally arrayed InAs QDs with an interdot distance of 100 nm. We show in Fig. [L10] the AFM image with a cross-sectional profile for the nano-oxide dots grown by AFM tip-induced oxidation.

**Scanning Tunneling Microscopy:** As in the case of AFM, STM is normally used to image surface without causing damage or introducing modifications (plane view measurements). This plan-view provide us with information about the surface, but details of the material that are not surface specific remain hidden for this method. In contrast, cross sectional STM denoted as xSTM, XSTM, or X-STM, provides the properties of the material such as concentration and types of defects in the semiconductor. For the case of heterostructures, cross sectional imaging is useful for probing the structure of the interfaces between neighboring layers, as well as for studying the detailed properties of the individual layers themselves. X-STM is performed on surfaces prepared by in situ cleavage and which are not reconstructed. Fig. [L11] illustrates the difference between STM and X-STM imaging. Note also that problems connected with the cooling of a sample, present in the case of AFM and STM measurements are not encountered if X-STM experiments are performed on covered samples.

X-STM has been widely used for the structural characterization of QDs (see e.g. work of Eisele et al. (1999) or Çelebi et al. (2006)). For illustrative purposes, we show a detailed X-STM image of a selected threefold stack of QDs in Fig. [L12] as taken from Eisele et al. (1999). It is desirable to obtain a quantitative evaluation of chemical composition as a function of position in the heterostructure bringing a head a question of what is imaged by STM.

There are two main mechanisms for the observed contrast (Eisele et al. 1999): (i) The dependence of the tunnelling probability on the local electronic structure. It means that the material of the nanostructure with its lower band gap locally provides more electronic states for tunnelling. In the case of InAs/GaAs system it means that this effect will lead to protrusions at InAs as compared to GaAs. (ii) The displacement of the cleavage face (in the case shown in Fig. [L11] - (110) cleavage face) due to strain in the underlying material. In the case of InAs/GaAs system, the compressive strain in the InAs regions is partly relaxed upon cleavage leading to a smooth elevation of the surface. Therefore, in order to image the chemical contrast, the long-range height variation due to strain relaxation has to be separated (This is shown in Fig. [L12(b)]). X-STM has been proven to be an useful tool for the estima-
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Figure 1.11: Illustration of the STM and X-STM imaging.

Figure 1.12: (a) Detailed X-STM image of a selected threefold stack of QDs. (b) The same image after background subtraction, with a schematic outline of the dot boundary at the cleavage surface. (taken from publication of Eisele et al. (1999)).

...tion of the compositional inhomogeneity (Liu et al. 2000). For example, Liu et al. (2005) studied the shape and compositional inhomogeneity of In$_{0.5}$Ga$_{0.5}$As QDs by X-STM finding...
that while the dots are of truncated pyramidal shape, an In-rich core has an inverted-triangle shape with its apex at the center of the island base. Their findings are illustrated in Fig. 1.13. Important consequence of such a composition profile is that the effective size of the dot is completely different than expected, having smaller size than would be supposed.

For completeness, let us add that, although STM does not damage or modify a surface, as mentioned above, its tip can also be used to control the nanostructures’ nucleation site (Kohmoto et al. 1999, Nishikawa et al. 2001). Namely, some material may be deposited through voltage pulses from the STM tip. Note that AFM is used for similar purposes too, as discussed in previous paragraph.

**Wave function mapping:** X-STM enables imaging of wave function amplitudes in QDs. Namely, Grandier et al. (2000) investigated the conduction band (CB) states of InAs quantum boxes embedded in GaAs with the spectroscopic ability of the STM, observing at room temperature, for the first time, standing wave patterns in the InAs boxes associated with the lowest CB states. Furthermore, low-temperature scanning tunnelling spectroscopy (STS) was used to perform a systematic study of the shape of the squared wave function $|\psi_i(x,y)|^2$ (Maltezopoulos et al. 2000). Maltezopoulos et al. (2000) investigated freestanding QDs deposited on GaAs(001) and got full access to the geometrical shape of the QDs, their single-particle peaks in the density of states, and the spatial distribution of the corresponding $|\psi_i(x,y)|^2$. Fig. 1.14 (a) shows I(V) curve recorded above the QD (black line), and I(V) curve recorded on the wetting layer (grey line). Two current steps are observed on the QD. $\frac{dI}{dV}$
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Figure 1.14: Single QD, W tip: (a) I(V) curves recorded on the QD (black line) and on the wetting layer (grey line), $V_{stab}=1.6$ V, $I_{stab}=50$ pA; (b) dI/dV curves recorded simultaneously with (a), $V_{mod}=28$ mV; (c),(d) spatially resolved dI/dV data at $V_{sample}=0.89$ V and $V_{sample}=1.14$ V, respectively. (taken from the publication of Maltezopoulos et al. (2000)).

The curve is shown in Fig. 1.14(b), where the two steps appear as two peaks and are caused by the two quantized states of the QD. The dI/dV image recorded at the first and second peak of Fig. 1.14(b) are shown in Fig. 1.14(c) and 1.14(d), respectively. The first peak has a circular symmetric intensity distribution, as expected for (000) state, while the second one has a pronounced node in the center as expected for a (100) state, i.e. a p-state.

Transmission Electron Microscopy: TEM and high resolution TEM (HRTEM) provide information about covered islands which is the main strength of these methods. Namely, most applications of QDs require covered dots with optimized sizes and shapes. Indeed, the shape and size of the QDs as well as information about the coherent or incoherent nature of islands can be obtained with high accuracy using TEM. For example, top-view TEM may indicate the base of the dot (square, circle, elliptic, elongate etc.), but has problems with the estimation of the dots' height due to the strain fields. HRTEM however provides full information about QD size and shape. Cross-sectional TEM provides details about the island diameter and shape, but it samples fewer islands (Schukin and Bimberg 1999). As an
1.4. Structural characterization

**Figure 1.15**: A [110] cross-sectional TEM micrograph of a single InP QD overgrown with nominally 20 nm of Ga\textsubscript{x}In\textsubscript{1-x}P. White arrows indicate domains with higher degree of ordering in the Ga\textsubscript{x}In\textsubscript{1-x}P islands. The inset shows the diffraction pattern for the sample. (taken from publication of Hakanson, Sass, Johansson, Pistol and Samuelson (2002)).

example, we show in Fig. 1.15 a [110] cross-sectional TEM micrograph of a single InP QD overgrown with nominally 20 nm of Ga\textsubscript{x}In\textsubscript{1-x}P as reported by Hakanson, Sass, Johansson, Pistol and Samuelson (2002).

Actually, by combining the AFM measurements on islands before overgrowth with the TEM measurements after overgrowth, a "more complete" picture of the effects present in the QD morphology can be extracted.

**Strain mapping from HRTEM images**: Improved point to point resolution of HRTEM images allows their usage for the characterization of the structure parameters and processes in semiconductor nanostructures at atomic scale. In particular, the elastic stress present during the heteroepitaxy induces various phenomena and consequently requires to be evaluated (Tillmann et al. 1999). Recent advances in digital imaging and image-processing techniques offer the possibility of locally determining the elastic strain of materials at atomic scale using HRTEM images. On the other hand, the reliability of strain profiles relies on the assumption of a constant spatial relationship between the intensity maxima in the image and the relative positions of the atomic columns in the specimen. This is not true in all cases, due to some known effects, such as thin foil relaxation, local crystal tilts, surface relaxation and the appearance of shifts in lattice fringes due to thickness and/or composition variation across the material (Galindo et al. 2005).

Peak finding and geometric phase are the two main approaches for strain mapping whereas
both of them are based on a comparison of the image features with reference lattice, in order to determine the distortions of the unit cells in the image. The local lattice distortions are then used to obtain the local strain distributions of the crystal. Peak finding methods (Bierwolf et al. 1993, Seitz et al. 1998, Kret et al. 2001) work in real space, building a two-dimensional reference lattice associated to a non-distorted region of the material, and identifying the local displacements of a grid that is built up from the set of intensity maxima in the HRTEM image. The strain field is obtained as a derivative of the local discrete displacement field that was calculated at each node of the lattice. In particular, the selection of pairs of peaks along two non-collinear directions, gives us the possibility to determine precisely the strains around dislocations (Galindo et al. 2005).

Geometric phase methodology, developed by Hýtch et al. (1998), works in Fourier space and consists of filtering the image with an asymmetric filter at a Bragg spot of the Fourier transform of a HRTEM lattice image and performing an inverse Fourier transformation. The phase component of the resulting complex image gives information about local displacement in a given direction. Local strain is derived by analyzing the derivative of the displacement obtained from two non-collinear components. For completeness, we note that peak finding approaches have some advantages when the image has different materials across the image,
1.4. Structural characterization

Figure 1.17: (Ga(Sb,As)/GaAs QDs on (In,Ga)As/GaAs seed QDs: Determination of the local lattice parameter $a$ in the [001] direction by means of peak finding method (a), and geometric phase method (b). (taken from the publication of Neumann et al. (2006)).

because of the global character of the Fourier transform, whereas the geometric phase exhibits superior performances in the case of strain factor determination around dislocations (Galindo et al. 2005, Guerrero et al. 2007). As an example, we show a simulated HRTEM image of the InAs$_{0.5}$P$_{0.5}$/InP heterostructure as well as the strain profile obtained after applying the geometric phase method in Fig. 1.16. These results are reported by Guerrero et al. (2007).

As an example of an application of the strain mapping to QDs, we show the results of Neumann et al. (2006) on double layer of (In,Ga)As/GaAs and (Ga(Sb,As)/GaAs QDs grown on GaAs (001) substrates. The positions of the (Ga(Sb,As)/GaAs QDs is directly correlated with the (In,Ga)As/GaAs in the lower lying layer by the strain field. Determination of the local lattice parameter $a$ (that further leads to strain distribution) in the [001] direction of such a double layer system by means of peak finding method and geometric phase method is shown in Fig. 1.17(a) and (b), respectively.

1.4.2 Diffraction methods

Three diffraction methods (Shchukin and Bimberg 1999), reflection high-energy electron diffraction (RHEED), reflectance anisotropy spectroscopy (RAS), and X-ray diffraction, will be discussed in this subsection.

Reflection high-energy electron diffraction: RHEED is a highly surface-sensitive ultra-high vacuum technique used for the monitoring of the MBE growth and its value lies in the fact that it enables in situ monitoring of the formation of the 3D islands.
Reflectance anisotropy spectroscopy: RAS enables the monitoring of the asymmetry of the dielectric properties of the surface. Identification of surface reconstruction and oscillations due to monolayer growth.

X-ray diffraction: It is useful for structural investigation after growth. The problem in this approach is the week diffraction signal of the dots which is a consequence of the much larger dots’ size (∼ 10nm than the probing wavelength (∼0.1nm). However, X-ray diffraction has been shown to be a powerful tool for measuring the alloy composition of InGaAs in the wetting layer.

1.5 Optical measurements

Optical spectroscopy is a very powerful and important tool for the investigations of the electronic structure of semiconductor nanostructures. The well known steady-state optical measurements are photoluminescence (PL), photoluminescence excitation (PLE), and single dot spectroscopy techniques, such as cathodoluminescence (CL) or STM induced luminescence (STML). However, due to the overlapping of the spectral repartitions, steady-state measurements have not allowed to discriminate between the different contributions of each luminescent center. On the other hand, time-resolved spectroscopy is well known to be powerful to supply for such problem, notably when the involved time constants are very different.

Through this thesis we compare our theoretical findings to steady-state optical measurements, therefore in what follows we address PL, PLE in Sec. 1.5.1 and single dot spectroscopy techniques in Sec. 1.5.2.

1.5.1 Photoluminescence

The emission of light from a semiconductor is the result of the recombination of an electron and a hole. This is what is called luminescence. The electron and the hole can be created in different ways. The usual method is to optically create the carriers. We call this photoluminescence (PL). The wavelength of the emitted light is determined by the energy difference between the involved electron and hole states. By analyzing the emitted photons, information about the states can be obtained. Some processes involved in the experiments can be seen in Fig. 1.18. As an example, the PL spectra of InAs/GaAs and InAs/GaSb QDs is shown in Fig. 1.19a. The excitation power is varied and it was found that the peak position of PL spectra exhibits a significant blueshift under a moderate excitation level. The observed blueshift was explained by the band bending effect due to the spatially separated photoexcited carriers in a type-II band alignment in InAs/GaSb system. Type II systems are discussed in Chapter 5 of this thesis. Furthermore, PL measurements are often carried out in the presence of an external magnetic field (Schildermans et al. 2005, Hayne
1.5. Optical measurements

Figure 1.18: Processes involved in a photoluminescence experiment. The non-resonant photons are absorbed, creating hot electrons and holes. The carriers thermalize and can be captured in a dot. At low excitation power density, the carriers relax to the bottom of the potential well since this process is much faster than the recombination process.

et al. 2000, Hayne et al. 2001, Godefroo et al. 2004). These magneto-photoluminescence (magneto-PL) measurements enable a study of competition between quantum confinement, Coulomb interaction and magnetic field confinement. In Fig. 1.19(b) an example of magneto-PL of InAs/GaAs QDs grown on [113] substrate orientation is shown for magnetic field applied in the growth direction.

The photoluminescence excitation (PLE) spectrum is obtained by detecting the intensity variation of an emission line while changing the excitation energy. The detected photoluminescence can be used to probe the absorption in the empty states of the dot.

1.5.2 Single-dot spectroscopy

Optical study of an ensemble of quantum dots yields a blurred picture due to the size dispersion of QDs related to the growth method.

As we have already shown in Sec. 1.2 the nucleation is a random process and no matter how good control we have of the epitaxy, the dots will not be identical in terms of size and strain and they will not live in physically identical neighborhoods. Therefore, in order to avoid statistical broadening of the spectra and to obtain maximum information, individual objects must be studied.
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Figure 1.19: (a) Excitation power dependence of the PL spectra of InAs/GaAs QDs (dotted lines) and of InAs/GaSb QDs (solid lines) (taken from the publication of Chen et al. (2007)). (b) The center of mass of the PL peak ($E_{\text{CM}}$) as a function of the magnetic field applied parallel to the growth direction. The inset shows the normalized luminescence spectra of the ensemble without field (full line) and for a magnetic field of 50.8 T parallel to the growth direction (dashed line) (taken from the publication of Godefroo et al. (2004)).

The selection of the photon emission from a single quantum dot can be done in a variety of ways. Using optical microscopy, studies of a single dot in lithographically defined etched mesa structures (Marzin et al. 1994, Kuther et al. 1998), and nano-apertures in a metal (Hessman et al. 1996) have been performed. In order to increase the spatial resolution of the experiment it is also possible to combine optical microscopy with excitation methods that only address a small enough volume for a single dot to be excited. This has been done using cathodoluminescence (CL) spectroscopy (Grundmann, Christen, Ledentsov, Böhmer, Bimberg, Ruvimov, Werner, Richter, Gösele, Heydenreich, Ustinov, Egorov, Zhukov, Kopev and Alferov 1995), near-field scanning optical microscopy (Saiki et al. 1998), and STM induced luminescence (STML) (Hakanson, Johansson, Persson, Johansson, Pistol, Montelius and Samuelson 2002). STML allows investigation of single QDs correlating the spectrally resolved luminescence and the topography of the investigated nanostructures. Fig. 1.20 shows STML spectra from two different InP/(In,Ga)P dots having the same size (marked by crosses in the inset). The luminescence spectra have approximately the same shape and are shifted less than 10 meV with respect to each other. Note that the spectra from different dots are generally similar with respect to energy range and peak spacing, but the intensity ratio of the peaks, as well as the overall intensity may vary.

Ota et al. (2004) reported the single-dot spectroscopy of a InAs QD. The electronic structure of a single self-assembled InAs quantum dot was studied by probing elastic single-
Figure 1.20: STM induced luminescence spectra acquired with the tip positioned on top of two dots, denoted A and B in the inset. (taken from publication of Hakanson, Johansson, Persson, Johansson, Pistol, Montelius and Samuelson (2002)).

electron tunnelling through a single pair of weakly coupled dots where the s and the p shell were identified.

1.6 Modelling of self-assembled quantum dots

In previous sections we explained growth procedures, structural characterization of fabricated QDs, and the optical measurements that are usually performed on such QD systems. A thorough theoretical analysis of the electronic and optical properties of QDs should include a realistic geometry and composition profile of the QD, take into account the strain distribution in and around the dots, effects of piezoelectricity, and band nonparabolicity. The electronic structure calculations of QDs and QDMs are usually performed using $k \cdot p$ (Stier et al. 1999, Pryor 1998, Mlinar et al. 2006, Mlinar and Peeters 2006a, Tomić et al. 2006, Sheng and Leburton 2001b, Kurdi et al. 2006) and atomistic methods (He et al. 2005b, Bester and Zunger 2005, Jaskólski et al. 2006).

The steps performed in the theoretical modelling and link to the experimental data are explained in the following subsection. Overview of the widely used approaches for the calculation of the strain distribution, single-particle states, and many-particle states is presented in Sec. 1.6.2, Sec. 1.6.3, and Sec. 1.6.4 respectively. Finally, our theoretical approach is described in Sec. 1.6.5.
1.6.1 Theory versus experiment

We define the steps in the modelling of QDs electronic and optical properties and specify at which point experimental data are linked to a theoretical model. A theoretical approach for modelling optical properties of self-assembled QDs is illustrated in Fig. 1.21.

Size, shape, and composition profile of a model QD and/or QDM as they enter our calculations are extracted from structural characterization (Sec. 1.4). For the embodiment of a 3D model QD, we distinguish two cases. First, if the available PL measurements are performed on an ensemble of QDs, the information about the individual characteristics of the QDs are lost, and therefore we consider our model QD as a representative of the ensemble of QDs. Second, if the experimentalists provide us with single dot spectroscopy data (see Sec. 1.5.2), our model QD has to have the exact geometry and composition profile as the dot which the experiments were performed on. Only in that case one can expect that the measured optical spectra is correctly interpreted.

Subsequent to the QD model creation, strain distribution in and around such a model dot is calculated. For that purpose, several approaches were put forward: isotropic model, continuum elasticity model, valence force field model etc. The basic ideas behind those
approaches as well as main differences are discussed in Sec. 1.6.2. What is important to clear up at this point is the fact that strain distribution is calculated under the assumption that dot is already formed. Namely, the dot made of one material is embedded in a surrounding material with different lattice constant and the elastic energy of such a system is minimized giving us the components of the strain tensor. It is clear that size, shape, composition profile as well as the growth on high index planes influence significantly the strain distribution. Elastic constants that characterize the constituent materials forming the dot, have values extracted from experiment, and enter the calculations regardless of the employed model. In Fig. 1.21 this is illustrated with gray boxes. Also, as we have shown under paragraph "Strain mapping from HRTEM images" in Sec. 1.4.1 it is possible to extract strain components from HRTEM images of the sample, enabling a comparison between calculated components of the strain tensor and strain components extracted from these HRTEM images. Let us also add that neither one of above mentioned approaches models the variation of the strain distribution during the QDs’ growth and such a modelling is beyond the scope of this thesis.

As a next step in the theoretical modelling, single particle states are calculated. Since the QD system contains from $10^5$ to $10^6$ atoms, the usage of semi-empirical theories becomes necessary. In Sec. 1.6.3 we briefly discuss main features of commonly used models: single-band model, $k \cdot p$ models, tight binding, and empirical pseudopotential models. Regardless of the employed approach, effective-mass or atomistic, the models are parameterized by a set of experimentally obtained band structure parameters from the bulk material (gray box in Fig. 1.21). From the calculated electron and hole energy levels of the QD, the dependence to QDs’ size, shape, or interfacial strain can be established. A comparison to the experimental data at this stage can be retrieve from PL experiment, used to probe the absorption in the empty states of the dot (see Sec. 1.5.1), or wave function mapping (see paragraph "Wave function mapping" in Sec. 1.4.1) as illustrated in Fig. 1.21. If measurements on QDs are performed in external electric or magnetic field, these external field effects enter the calculations at this step, as indicated with blue arrow pointing from "external electric or magnetic fields” box to "single particle states” box in Fig. 1.21.

The nature of many-particle interaction, such as electron-hole Coulomb effects, electron-hole exchange interaction, or correlations can be extracted from configuration-interaction calculations as explained in Sec. 1.6.4. Absorption spectra and transition energies are further calculated enabling a direct comparison with the available experimental data. This comparison is especially interesting if the single dot spectroscopy of such a dot had been done.

### 1.6.2 Strain calculations

**Isotropic model:** Isotropy-elasticity (IE) model was proposed by Davies (1998) and is based on the Eshelby’s theory of inclusions (Eshelby 1957). It was shown that the elastic field may be derived from a scalar potential that obeys a Poisson equation with the lattice
mismatch as charge density. The displacement was made analogous to the electric field providing a simple way to visualize the distortion around a dot.

**Continuum elasticity:** In the continuum mechanical (CM) model (Pryor et al. 1998, Jogai 2000), the functional form of the elastic energy is minimized in order to obtain the distribution of the displacement in the structure, wherefrom the strain is extracted. The crystal structure anisotropy is included in this model. Structures based not only on the zinc-blende but also on the wurtzite semiconductors can be considered by this method.

**Valence force field:** The valence force field (VFF) model of Keating (1966) and Martin (1970) is an atomistic approach, where the elastic energy is expressed as a sum of the potential energies located in atomic bonds. VFF contains only two elastic parameters which are easily derived from the experimental elastic constants. More complex approaches rely on Stillinger-Weber (SW) or Tersoff potentials which have seven elastic parameters.

**Comparison of the models:** IE is a continuum approach that assumes homogeneous elastic properties in contrast to CM where the anisotropy of the elastic constants is taken into account. Tadić, Peeters and Janssens (2002) compared IE and CM models when applied to InP/(In,Ga)P QDs, and shown that both the electron and hole energies are underestimated if the strain distribution is calculated by the IE, but in principle, qualitative differences are not expected to appear between IE and CM models for the case of disk-like QDs (Tadić, Peeters, Janssens, Korkusinski and Hawrylak 2002). Since IE and CM models belong to continuum approaches, they are not able to describe the strain at the level of the crystal unit cell where atomistic approaches are needed. Also, the large strains that exist between lattice-mismatched III-V semiconductors bring the CM outside the linearity regime, and the continuum approaches at the limit of the validity whereas the VFF approach would give a satisfactory results (Pryor et al. 1998). It is important to mention that continuum approaches "see" geometrical symmetry rather than the real atomistic symmetry of a system. For example, for InAs/GaAs pyramidal QDs, CM sees $C_{4v}$ symmetry, whereas VFF sees correct atomistic $C_{2v}$ symmetry (Pryor et al. 1998).

### 1.6.3 Single particle states

**Single band model:** The single-band models is the simplest and first developed approach based on the effective-mass theory. It actually assumes bands to be completely decoupled (e.g. see band structure of GaAs shown in Fig. 1.2). The electron and heavy or light hole are treated with two independent Schrödinger equations whereas the effective-mass parameters from some special point in the Brillouin zone are adapted.
**k·p model:** The more advanced approach based on the effective mass approximation that includes band mixing, is a multi-band k·p method (Luttinger and Kohn 1955, Luttinger 1956, Kane 1957, Pidgeon and Brown 1966, Weiler et al. 1978). It uses a minimal set of parameters that are determined empirically from experiments. By means of a perturbative approach, it provides a continuum in the wave vector $k$ of the energy bands in the vicinity of some special point in the Brillouin zone. More details about multi-band approach are given in Sec. [1.7].

**Empirical tight binding model:** In the empirical tight-binding model (ETBM) introduced by Slater and Koster (1954), the electronic states are considered to be linear combinations of atomic (s,p,d,...) orbitals. The Hamiltonian matrix elements between the atomic orbital states are introduced as free parameters to be determined by fitting the band gaps and band curvatures, i.e. effective masses at critical points in the BZ (Chadi and Cohen 1974). Depending on the number of orbitals and nearest neighbors used to represent the states, the ETBM requires that the overlap integrals be determined in terms of the measured direct and indirect band gaps and/or effective masses in the bulk material.

**Empirical pseudopotentials:** In the empirical pseudopotential model, the crystal potential is represented by a linear superposition of atomic potentials, which are modified to obtain good fits to the experimental direct and indirect band gaps and effective masses. It is the most complex approach and includes "all" the effects present in the system under study. For an application to nanostructures see work of Wang and Zunger (1996), Zunger (1998), Shumway et al. (2001), Bester and Zunger (2005), or Bester, Zunger, Wu and Vanderbilt (2006).

**Comparison of the different models:** In the case of strong confinement, as e.g. in QDs, the carriers are confined in all three directions making the non-parabolicity of the bands of the underlying semiconductors (used to construct the heterostructure) very important. Therefore, decoupling of bands, as is done in the single-band model, introduces significant errors in energy prediction. However, a 3D single band model that includes realistic QDs’ geometry is often accepted as a good compromise especially by the experimental part of the QD community. Implementation of a multi-band model appears as a reasonable compromise between the simplicity and inclusion of most relevant effects present in here studied III-V semiconductor QDs. Advantages of this approach are easy treatment of strain effects, external electric and magnetic field, growth on high index surfaces. Main drawbacks of the multi-band model are limited number of bands used in expansion (not all relevant band mixing may not be included), omission of atomistic symmetry (only geometric one is present). However, as to come to comparison with experiment, calculated optical properties (optical absorption spectra, transition energies etc.) extracted from multi-band theory agree
remarkable well with the available experimental data. Although microscopic semiempirical pseudopotential methods are more accurate, they are still limited in the same way as the \( \mathbf{k} \cdot \mathbf{p} \) technique because of the present lack of accuracy of the input parameters. The limitations of the multi-band calculations are given in Sec. [1.7.4](#).

### 1.6.4 Exciton complexes

The elementary optical excitation of a quantum dot is the exciton (X). In the dots, the exciton is split into four different states arising from spin. Two of these exciton ground states have anti-parallel spin and have optically allowed transitions to the quantum dot ground state. The two other dark states have parallel spin and are optically forbidden. Furthermore, a quantum dot charged with a single electron has a spin-1/2 ground state that has two substates, spin up and spin down. An exciton together with this additional electron creates a negatively charged exciton (X\(^-\)), also known as a negative trion. Alternatively, an exciton together with additional hole creates a positively charged exciton (X\(^+\)), also called a positive trion. A second exciton puts the quantum dot into a biexciton state (XX). Also the bi-exciton has four emission lines, given by the four different final states of the exciton. Furthermore, dots can be filled by more than one biexciton making the emission spectra much reacher and indeed more interesting for investigation.

Presence of more than one charge carrier in a QD leads to the appearance of direct Coulomb interaction, exchange effects, and correlation between the carriers. Prior to detailed analysis of many-body problem in QDs, let us first define the quantum confinement/Coulomb effects ratio that actually "selects" the theoretical approach (Stier 2001, Bimberg et al. 1998):

(i) The strong confinement regime—the electron and hole wave functions are largely uncorrelated. Actually, the Coulomb effects are only a small correction to the dominating quantization of the kinetic energy, meaning that electron and hole sublevel separations are much larger than the Coulomb interaction energy. In our case of QDs, strong confinement regime means that the dot radius is smaller than the bulk exciton radius. (ii) The intermediate confinement regime—while electron sublevel separations are larger than the Coulomb interaction energy, the hole sublevel separations are smaller. As a consequence, the hole energy is quantized by the electrostatic potential of the electron orbital. (iii) The weak confinement regime—the electron and hole sublevel spacings are comparable to or larger than the Coulomb interaction energy. The electron and hole form pairs whose center of mass motion is quantized by the confinement potential. In our case of QDs, the weak confinement means that the exciton radius is smaller than the dot radius.

**Direct Coulomb interaction:** As being in the regime of strong confinement, the many-body effects act as a perturbation to the confinement. Actually, the direct Coulomb interaction...
1.6. Modelling of self-assembled quantum dots

1.6.5 How do we approach modelling?

Following the steps given in a flow diagram in Fig. 1.21 our goal is to develop a model for the electronic structure calculations of QDs that takes into account the influence of strain, piezoelectricity, growth on high index planes, and effects of external fields, in particular a magnetic field. As one could see from the previous discussion in Secs. 1.6.2, 1.6.3 and 1.6.4 the main differences between models appear in strain and single-particle state calculation.

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6Important for single photon sources as explained in Sec. 1.1.2
In our approach, we create the QD structure on a 3D rectangular grid which enables us to define the parameters of our model at each node of the grid. The strain distribution is calculated in the framework of continuum elasticity theory, and the single particle states are extracted from the nonsymmetrized eight-band Hamiltonian. For the electronic structure calculations of QDs placed in an external magnetic field, an implementation of an eight band $k \cdot p$ theory appears to be the most elegant. Our model properly takes into account the effect of the substrate orientation by rotation of the coordinate system in the way that one coordinate coincides with the growth direction (see Sec. 3.2 for details). The effects of a tilted external magnetic field are taken into account through the Zeeman effect and employing a gauge invariant scheme based on Wilson’s formulation of the lattice gauge theory (see Sec. 3.3 for details). In next section we give a short overview of the multi-band effective mass theory, and its application to nanostructures. Details of our numerical approach are given in Chapter 3.

1.7 Multi-band effective mass theory

The effective-mass theory is a very well established method to obtain the band structure in the case of weak perturbing inhomogeneous semiconductor potentials (Luttinger and Kohn 1955, Luttinger 1956, Kane 1957, Weiler et al. 1978, Weiler 1981). Electron and hole energies near the band extrema in the presence of external magnetic and electric fields and in a crystal containing a shallow impurity were successfully described by this theory (Luttinger 1956, Kane 1957, Pidgeon and Brown 1966).

1.7.1 Multi-band effective mass model: Basic ideas

The basic idea behind the effective mass theory is actually rather simple: Near the band edges the electrons can be described to behave as if they are in free space except their masses taking some effective value $m^*$. Therefore, the electron states are described by a single Schrödinger equation around the given point of the Brillouin zone, assuming the (conduction) band parabolic.\(^7\) In what follows we shall focus on the band structure description around the $\Gamma$ point discussing only the case of direct gap semiconductors (see Fig. 1.22). The physical accuracy of this simple effective mass approach decreases as one wanders away from the $\Gamma$ point since the band non-parabolicity originating from band mixing, or inter-valley mixing, starts to be important. In the framework of the effective-mass approach, this limitation was overcome by employing the model referred to as $k \cdot p$ theory (Luttinger 1956, Kane 1957, Pidgeon and Brown 1966). It is based on selecting a few 3D-periodic Bloch orbitals taken from the Brillouin zone center which single particle wavefunctions are expanded in. Namely,\(^8\)

\(^7\)Limitations of the eight band $k \cdot p$ model are given in Sec. 1.7.4

\(^8\)Strictly speaking such a picture is valid only at the high symmetry points of the Brillouin zone.
1.7. Multi-band effective mass theory

In \textbf{k} \cdot \textbf{p} theory, the coupling between the bands of interest\(^9\) (set A) and all other bands (set B) is reduced to zero whereas the coupling between the bands of interest was not affected by this transformation. At the same time the effective masses in the bands of interest are renormalized to take into account the influence of the not included bands in set A (Löwdin renormalization, see the work of Löwdin (1951)). As a next step, a fitting procedure is used to obtain the effective-mass parameters (Pidgeon and Brown 1966)\(^{10}\). Depending on the problem one is dealing with, the number of bands in the set A can vary from four up to thirty. Namely, \(4 \times 4\) \textbf{k} \cdot \textbf{p} model includes only heavy holes and light hole bands (see Fig. 1.22(b))\(^{11}\) and is used when the energy gap and split-off energy are large enough so band mixing between heavy or light hole and the conduction band and split-off band can be neglected. For example, such a model has been employed by Pedersen and Chang (1996) to describe confined electron and hole energy levels in GaAs/(Al,Ga)As QDs with parabolic in-plane potential and step like potential in the growth direction. Furthermore, to take into account the split-off band in materials with small split-off energy, basis set A was extended to include the split-off band, resulting in the \(6 \times 6\) \textbf{k} \cdot \textbf{p} model. In this case, it is assumed that the

\(^9\)In our case of eight band \textbf{k} \cdot \textbf{p} theory - the lowest lying conduction band and three top most valence bands, see Fig. 1.22(b) and Chapter 4 for details

\(^{10}\)Parameters are determined empirically and the choice of the parameter set is discussed in detail in Sec. 5.5

\(^{11}\)Due to the spin, one ends up with four bands.
energy gap is large enough so the band mixing between valence band states and lowest laying conduction band can be neglected. For example, this model was used to investigate the electronic structure of InP/(In,Ga)P QDs (Tadić, Peeters and Janssens 2002). In the case of materials with narrow band gap, e.g. InAs, the lowest lying conduction band has to be added into the set A, resulting in the $8 \times 8 \mathbf{k} \cdot \mathbf{p}$ model, used in this thesis. Further extension of the basis set are possible, resulting in the $14 \times 14$ (Pfeffer and Zawadzki 1990, Kurdi et al. 2003), or even $30 \times 30 \mathbf{k} \cdot \mathbf{p}$ (Richard et al. 2005, Kurdi et al. 2006) models. It is intuitively clear that with the inclusion of more bands, the validity of the model is extended further from the zone center. For example, employing $30 \times 30 \mathbf{k} \cdot \mathbf{p}$ gives us the description of the whole Brillouin zone (Richard et al. 2005). For completeness, we add that in the case of indirect gap materials, such as AlAs, or Si, eight-band $\mathbf{k} \cdot \mathbf{p}$ theory is not sufficient, and usage of more bands in recommended, e.g. Si/Ge QW and QDs have been successfully investigated in the framework of 30 band $\mathbf{k} \cdot \mathbf{p}$ theory (Kurdi et al. 2006).

1.7.2 Derivation of the model for bulk

As we have already mentioned, a loosely bound outer shell electron in a semiconductor crystal can be best described by a particle travelling in a slowly varying periodic potential. The one electron Schrödinger equation reads:

$$
\left(\frac{\mathbf{p}^2}{2m_0} + V(\mathbf{r}) + \hat{H}_{so}\right)\Psi_{\mathbf{k}}(\mathbf{r}) = E_{\mathbf{k}}\Psi_{\mathbf{k}}(\mathbf{r}) ,
$$

where the first, second, and third term on the right side correspond to the kinetic energy of the electron, the periodic crystal potential, and spin-orbit interaction respectively. The spin-orbit interaction, $\hat{H}_{so}$, is given by:

$$
\hat{H}_{so} = \frac{\hbar}{4m_0^2c^2}(\nabla V \times \mathbf{p}) \cdot \mathbf{\sigma} + \frac{\hbar}{4m_0^2c^2}(\nabla V \times \mathbf{k}) \cdot \mathbf{\sigma} ,
$$

where $\sigma$ is the Pauli operator. Note that the main contribution to the spin orbit interaction comes from the core region of the atom where $\nabla V$ and $\mathbf{p}$ are very large (the first term on right side of Eq. 1.2), whereas the second term is usually neglected since $\mathbf{k}$ is at most 1/2 a reciprocal lattice vector and hence small compared to $\mathbf{p}$.

The Bloch function is given by:

$$
\Psi_{\mathbf{k}}(\mathbf{r}) = \exp(i\mathbf{kr})u_{\mathbf{n}}(\mathbf{r}) .
$$

where $u_{\mathbf{n}}(\mathbf{r})$ has the periodicity of $V(\mathbf{r})$, $\mathbf{k}$ lies in the first Brillouin zone, and $n$ is the band index.

Substituting Eq. (1.3) into Eq. (1.1) gives (see e.g. Kane (1957)): 
\[ \left[ \frac{p^2}{2m_0} + V(r) + \frac{\hbar^2 k^2}{2m_0} + \frac{\hbar}{m_0} \mathbf{k} \cdot \mathbf{p} + \frac{\hbar}{4m_0^2 c^2} (\nabla V \times \mathbf{p}) \cdot \sigma \right] \psi_{nk}(r) = E_n(k) \psi_{nk}(r) , \tag{1.4} \]

This approach leads to the \( \mathbf{k} \cdot \mathbf{p} \) approximation named after the 4th term appearing in the Hamiltonian of Eq. (1.4). For fixed value \( k = k_0 \), Eq. (1.4) has a complete set of eigenfunctions \( \psi_{nk}(r) \) which completely span the space of functions periodic in the real space Bravais lattice. This set of states is called \( \mathbf{k} \cdot \mathbf{p} \) representation (Kane 1957).

We use \( \Gamma_0 \) point \( (k_0 = 0) \) as the basis for a \( \mathbf{k} \cdot \mathbf{p} \) representation\(^{12}\) and theoretically the entire zone can be covered. For example, Richard et al. (2005) showed that 15 bands (30 including spin-orbit interaction) can give the description of the whole Brillouin zone.

One can write \( \psi_{nk}(r) \) in terms of \( \psi_{n'k_0}(r) \):

\[
\psi_{nk}(r) = \sum_{n'} N c_{n'n} \psi_{n'k_0}(r) , \tag{1.5}
\]

where \( N \) is the number of bands used in the expansion.

Substituting Eq. (1.5) into Eq. (1.4), then multiplying Eq. (1.4) on the left by \( \psi_{n'k_0} \), and finally performing the integration over the unit cell gives the \( k_0 \cdot p \) representation:

\[
\sum_{n'} \left\{ [E_n(k_0) + \frac{\hbar^2 (k^2 - k_0^2)}{2m_0}] \delta_{nn'} + \frac{\hbar}{m_0} (k - k_0) \cdot \langle \psi_{nk} | \mathbf{p} | \psi_{n'k_0} \rangle \right\} c_{n'n} = E_n(k) c_{nn'} . \tag{1.6}
\]

In this thesis we are concentrated on the region of the conduction and valence band extrema where most carriers sit (around the \( \Gamma \) point). The topmost valence states and the lowest-lying conduction band are close together and well separated from all other band edges (e.g. see Fig. 1.22(a)). This fact enables division of all states into two sets: set A containing the bands of interest (in our case, the lowest lying conduction band and three topmost valence states) and set B containing remaining bands that have non-negligible effect on the states in A (see also discussion in Sec. 1.7.1). The interactions between A and B are removed, whereas interactions in A are perturbatively renormalized to account for the influence of the bands in set B (Löwdin 1951).

Let us first consider the case without the spin-orbit interaction. The valence band maximum is derived from atomic p functions, X, Y, Z, which remain degenerate under the tetrahedral point group of the zinc-blende lattice to give the representation \( \Gamma_1^5 \). The conduction band is derived from S functions which give the tetrahedral representation \( \Gamma_1 \). Therefore,

\(^{12}\)\( \Gamma_0 \) point is the most suitable to use as a basis, since it is also the point of highest symmetry in the zone.
Eq. (1.5) written in the matrix form for N=4, where the influence of all bands outside s,p subspace is included perturbatively, has the following form:

\[
H_4 = \begin{pmatrix}
D_e & B_{k_x}k_z + iP_{k_x} & B_{k_x}k_z + iP_{k_y} & B_{k_x}k_y + iP_{k_z} \\
B_{k_x}k_z - iP_{k_x} & D_{h_1} & -N'k_xk_y & -N'k_xk_z \\
B_{k_x}k_z - iP_{k_y} & -N'k_xk_y & D_{h_2} & -N'k_yk_z \\
B_{k_x}k_z - iP_{k_z} & -N'k_xk_z & -N'k_yk_z & D_{h_3}
\end{pmatrix},
\]

(1.7)

Here, \(D_e = E_c + A_\gamma k^2\), \(D_{h_1} = E_v - L'k_x^2 - M(k_y^2 + k_z^2)\), \(D_{h_2} = E_v - L'k_y^2 - M(k_x^2 + k_z^2)\), \(D_{h_3} = E_v - L'k_z^2 - M(k_x^2 + k_y^2)\). \(E_v\) denotes the energy of the conduction band minimum, and \(E_c\) is the energy of the valence band maximum. \(P\) is the Kane matrix element defined as \(P = -i(h/m)\langle S|p_x|X\rangle\) and \(B\) arises because of the inversion asymmetry of zinc-blende semiconductors. The number of independent parameters in Eq. (1.7) comes from the symmetry arguments (Luttinger 1956, Weiler et al. 1978, Weiler 1981). The parameters \(P, E_v, E_c\) come from the direct interaction between s and p wavefunctions, whereas \(A_\gamma, B, L', M, N'\) result from the second order interactions due to the L"owdin (1951) perturbation involving the states outside s-p subspace. \(A_\gamma\) is the electron effective mass renormalized to include influence of remote bands (see also Sec. 2.2). \(B\) has a finite value in zinc-blende semiconductors, but parameter \(B\) is usually neglected (Stier 2001). In what follows we shall assume the inversion symmetry of the system (B=0). \(L', M, N'\) can be expressed in the terms of scaled Luttinger (1956) parameters \(\gamma_i\), where \(i=1,2,3\):

\[
L' = (\hbar^2/2m)(\gamma_1 + 4\gamma_2), \quad M = (\hbar^2/2m)(\gamma_1 - 2\gamma_2), \quad N' = (\hbar^2/2m)(6\gamma_5),
\]

where \(m\) is the electron mass. Parameters \(\gamma_i\) were first introduced by Luttinger (1956) to represent the second order interactions involving the states outside p subspace. In our case, in which the conduction band is treated together with the valence bands i.e. s-p subspace, the Luttinger parameters have to be scaled to remove perturbative interaction with the lowest lying conduction band. In this thesis original or valence-band Luttinger parameters are denoted as \(\gamma_i\), and scaled ones as \(\gamma_i\), where \(i=1,2,3\). Scaled Luttinger parameters are connected to the valence-band Luttinger parameters by the relations (Pidgeon and Brown 1966):

\[
\gamma_1 = \gamma_1' - E_P/3E_g, \quad \gamma_2 = \gamma_2' - E_P/6E_g, \quad \gamma_3 = \gamma_3' - E_P/6E_g,
\]

where \(E_g\) is the fundamental band gap, and \(E_P\) is related to the Kane matrix element by \(E_P = 2mP^2/\hbar^2\).

Luttinger parameters can be written in a manner which explicitly reveals the contribution of all other states of symmetry \(\Gamma_1, \Gamma_{15}, \Gamma_{12}, \) or \(\Gamma_{25}\) (Foreman 1993). This states are compatible with the s, p, d, and f orbitals respectively. The contribution of \(\Gamma_{25}\) symmetry is neglected here.\(^{13}\) The Luttinger parameters can be written as (Foreman 1993):

\(^{13}\)Contribution of f orbitals to the valence electronic structure of semiconductors is insignificant, and therefore usually neglected (Foreman 1993).
\[ \gamma_1 = -1 + 2\sigma + 4\pi + 4\delta, \]
\[ \gamma_2 = \sigma - \pi + 2\delta, \]
\[ \gamma_3 = \sigma + \pi - \delta, \]
(1.9)

where \(\sigma, \pi,\) and \(\delta\) are defined by:
\[ \sigma = \frac{1}{3m} \sum_{i} (|X| p_x |u_i|^2 / (E_j - E_v) - E_p / 6E_g), \]
\[ \pi = \frac{1}{3m} \sum_{i} (|X| p_y |u_i|^2 / (E_j - E_v)), \]
\[ \delta = \frac{1}{6m} \sum_{i} (|X| p_x |u_i|^2 / (E_j - E_v)). \]

\(m\) is the free electron mass, \(p\) is the momentum operator, and \(E_v\) is the valence band energy in the absence of spin orbit interaction. The sum is over the basis states \(u_i\) of all remote bands of a given symmetry. Note that from the sum that goes over basis states of the \(\Gamma_1\) symmetry (\(\sigma\) term in Eq. (1.10)), the lowest-lying conduction band is subtracted (second term of right-hand side of expression for \(\sigma\)) since the interaction between the lowest-lying conduction band and three topmost valence bands is already explicitly treated in our model.\(^{14}\)

When the spin-orbit interaction is included, the same s-p subspace is used but also the spin degree of freedom is taken into account. As a consequence we have eight states instead of four. The spin-orbit interaction splits the \(\Gamma_{15}\) states forming irreducible representations \(\Gamma_8\) and \(\Gamma_7\). Namely, coupling of angular momentum with the spin gives two possible values of total angular momentum \((j=3/2\) and \(j=1/2)\). The four states \(|3/2, j_z\rangle\) form the irreducible representation \(\Gamma_8\), whereas two \(|1/2, j_z\rangle\) form the irreducible representation \(\Gamma_7\).

Spin-orbit interaction may be represented by the single parameter, \(\Delta\), which is the spin-orbit splitting of the p states:
\[ \Delta = -3i \left( \frac{\hbar}{4m_e c^2} \right) \langle X | \nabla V \times p | Z \rangle. \]
(1.11)

It is most convenient to work with eigenfunctions of the total angular momentum \(|j, j_z\rangle\) which diagonalize spin-orbit interaction.\(^{15}\) The explicit form of the eight-band model used in this thesis is given in Chapter 2 as well.

For completeness, let us add that if the crystal structure has inversion symmetry the bands are double degenerate with respect to spin (Kramers theorem). In the theoretical description of zinc-blende semiconductors, such as GaAs, InAs, AlAs etc. it is assumed that inversion symmetry is approximately satisfied due to the small difference in the constituent atoms.

\(^{14}\)The lowest-lying conduction band belongs to the set A.

\(^{15}\)The proper linear combinations of p functions and spin functions are taken to diagonalize the spin orbit interaction (Luttinger and Kohn 1955, Luttinger 1956).
Presence of weak perturbing inhomogeneous semiconductor potentials leads to the introduction of the envelope function (Luttinger and Kohn 1955, Luttinger 1956). It means that detailed wavefunction is split into slow varying (envelope part) $F_n(r)$ and the cell periodic and more oscillatory Bloch functions $u_{nk}(r)$ satisfying the Schrödinger’s equation with band-edge energies:

$$\Psi(r) = F_n(r)u_{nk}(r).$$ \hspace{1cm} (1.12)

Implementation of strain effects and external magnetic field effect within this theory, is given in Chapter 2.

1.7.3 Application to nanostructures

Due to its simplicity, easy implementation of the strain effects, effects of an applied magnetic field, or growth on high index surfaces, the multi-band effective theory has been widely accepted as a reasonably good theoretical tool for the electronic structure description of semiconductor nanostructures. It has been proven to give a remarkable good agreement with the experimental data when employed to nanostructures, ranging from QW to QDs, or recently to quantum rings. Here, we shall briefly introduce the main stumble points in the application of the multi-band effective-mass theory to nanostructures.

When applied to nanostructure, the effective-mass parameters present in the multi-band model become spatially dependent, commonly varying abruptly through the interface between the nanostructure and the surrounding barrier material. Application of the multi-band Hamiltonian to a nanostructure can be understood as an application of the multi-band Hamiltonian, as in the bulk case, for the nanostructure (one bulk material), and the second application for the barrier material (second bulk material). Boundary conditions are then applied to connect solutions from both sides of the interface. Also, it is assumed for all constituent materials to have the identical $\Gamma$ point. In general, such a Hamiltonian does not have to be Hermitian, and its Hermiticity actually depends on the choice of the boundary conditions at the heterointerface of a nanostructure. Although clear from previous Section, we stress here that the validity of the effective mass approximation is broken when applied to nanostructures, since the condition of the weak perturbing potential is violated at the heterointerface. Indeed, the arbitrariness in the choice of the boundary conditions, as well as an unjustified application of the effective mass theory to abruptly varying potentials (beyond the valid range of the model) are the main weak points of this approach. Depending on the treatment of the boundary, several approaches were put forward, that can be divided into three groups:

(i) First one is based on the matching of bulk solutions across an interface, as we mentioned above, and the generalization of the bulk equation to the case of spatially varying composition (Eppenga et al. 1987). This approach has been widely accepted in the literature and no unexpected or nonphysical behavior of the calculated electronic spectra of any of the modelled nanostructures has been reported. The main weakness of
this approach is the absence of the possibility to trace and estimate errors coming from the employed boundary conditions, since they were ad hoc introduced only taking care about the Hermiticity of the Hamiltonian. Validity of this approach we shall discuss in the next Chapter.

(ii) Second approach is based on the development of the envelope function approach for nanostructures, with an intention to resolve both, the boundary condition problem as well as the validity of the effective-mass approach when applied to nanostructures (Burt 1992, Burt 1999). Namely, the envelope function expansion with zone-center eigenfunctions was applied to an entire nanostructure\textsuperscript{16} making the envelope functions unique and circumventing the boundary condition problem. The later is simply resolved by the fact that envelope functions are smooth and continuous everywhere for well behaved wavefunctions, even at the abrupt interface\textsuperscript{17}. From the practical point of view, the multiband Hamiltonian developed in the framework of Burts’ approach differs from the one discussed under (i) in the additional terms arising because of the abrupt variation of the effective-mass parameters at the interface of the heterostructure. Such a Hamiltonian was first developed by Foreman (1993), and this approach is usually referred to as Burt-Foreman approach. Development of the Hamiltonian in the framework of this approach is given in next Chapter. It is worth mentioning, that all simplicity of the conventional approach with respect to the modelling of strain in nanostructures, effects of an external fields, growth on high index surfaces is preserved in this approach as well. Strain effects are incorporated in an adiabatic manner, as discussed in previous section, where the part of the inhomogeneity effects on the band-structure is accounted for through the terms coupling the conduction and valence bands (Zhang 1994).

(iii) The third approach was proposed by Foreman (2005) and is based on a new first-principle envelope-function theory that claims to include all non-zero, first-principle derived terms (Foreman 2006b, Foreman 2007). The envelope functions are formulated in terms of atoms rather than bulk compounds, whereas a traditional bulk-crystal description can be obtained from the atomistic formulation via linear transformation of variables.

The price one has to pay by losing all the elegance of the standard multi-band approach (straight-forward inclusion of the strain effects, external magnetic field, etc.), on behalf of inclusion of the all the non-zero terms (some of them not even evaluated) in the "correct" Hamiltonian is simply too high. Namely, the main difficulty in modelling the electronic

\textsuperscript{16}The same $U_{nc}(R)$ are used throughout even though there are regions where they are not eigenstates of the local Hamiltonian.

\textsuperscript{17}The limited range of wavevectors is used in their plane expansion
properties of QDs comes from a relative uncertainty of the input parameters (e.g. exact QD geometry, the composition, or the effect of the In segregation). These parameters will depend on the growth conditions and even a slight variation on their values (e.g. composition) can lead to strong deviations on the calculated electronic structures. In this respect, it is not to be expected that the additional interface parameters that complicate the model significantly, will introduce improvement in the modelling. Therefore, the analysis of the two approaches given under (i) and (ii) will be discussed in next Chapter in more details.

1.7.4 Limitations of the model

The main limitations of \( k \cdot p \), as can be find in the literature (Zunger 1998, Bester and Zunger 2005, Wang et al. 2000, Wood and Zunger 1996), are: Insufficient number of Bloch functions used in the expansion of the wavefunctions (see Sec. 1.7.1), assumption of identical Bloch functions through whole nanostructure, boundary conditions at the nanostructure-barrier interface, geometric versus real atomistic symmetry, and heavy dependence of the electronic states on the even small variation of the band structure parameters.

The \( k \cdot p \) theory is valid only in the vicinity of the zone center. Reducing the dimensionality from the bulk case to the quantum dot, leads to a significant increase of the confinement. This as a consequence leads to a spread in \( k \)-space, and the accompanying contributions far from the \( \Gamma \) point of electron and hole states. Realistic model has to be able to include this non-parabolicity, and as concerned the multi-band effective mass model, increasing the number of Bloch functions in the expansion leads to the extension of the validity further from the \( \Gamma \) point. So in the case of 30 bands, the whole Brillouin zone is described and in contrast to the expected dramatically increase of the number of parameters (Zunger 1998), it turned out that only 10 and 11 input parameters for \( O_h \) and \( T_d \) group, respectively are required. Eight bands, as used in this thesis are sufficient to describe the electronic and optical properties of QDs made of direct gap III-V zinc-blende semiconductors, whereas the dots are limited in size to the effective radius larger than 2nm. Proper choice of the empirically obtained parameter set may improve the validity of the theory away from the zone center, as discussed in Sec. 3.5. Boundary condition problem as well as the assumption of identical Bloch functions through the whole nanostructure were indeed serious drawbacks in the implementation of the multi-band effective mass, but they were overcome by the nonsymmetrized approach (Burt 1992, Foreman 1997, Mlinar et al. 2005, Foreman 2006b, Nilsson et al. 2006). However, the boundary condition problem in the 30 band model, where the closed basis set is used (no influence of the set B - see Sec. 1.7 on the effective mass parameters is taken into account) is still not resolved. The main unresolved problem of the continuum model such as here implemented multi-band effective mass theory is the absence of the possibility to feel the atomistic symmetry of the real system (Bester and Zunger 2005). There were several attempts to resolve this issue by adding \( ad \) \( hoc \) introduced terms at the interface, but without
great success. It is honest to say that this model simply does not see atomistic symmetry and should not be used for such a purposes. The introduction of the additional parameters to reflect the underlying symmetry, were fit to describe the experiment, but the predictive power of such an approach is questionable. In summary, the model is rather successfully used for the description of PL and PLE measurements on QD systems, even for the description of single dot spectroscopy (Rastelli et al. 2004), but subtle details, originating from the atomistic symmetry such as degree of the optical anisotropy (since electron and hole wavefunctions are influenced by this symmetry) could not be estimated from this approach. Question of the parameters dependence as well as the choice of an appropriate parameter set is discussed in Sec. 3.5.

1.8 Outline of the thesis

The thesis is organized as follows.

Chapter 2 is devoted to the intrinsic problems of multiband envelope function theory at the transition from a bulk description to a heterostructure description. Previously unknown influence of different boundary conditions at the hetero-interface of a nanostructure on the calculated confined electron and hole states is demonstrated, and we propose “more correct” boundary conditions that should be used if the nanostructure is placed in an external magnetic field. Details of the development of a nonsymmetrized Hamiltonian for nanostructures in a magnetic field are given as well.

Employing the nonsymmetrized Hamiltonian as developed in Chapter 2, we present a full three-dimensional model for the electronic structure calculation of semiconductor QDs and QDMs grown on high index surfaces and/or in the presence of an external tilted magnetic field in Chapter 3. Also, we give details of the numerical implementation of our 3D model. Special attention is payed on preserving the gauge invariance by employing a gauge invariant scheme based on Wilson’s formulation of lattice gauge theory.

The model consistently developed in Chapters 2 and 3 enable us to model electronic and optical properties of various QD systems making comparison with experiment or providing theoretical predictions. In this thesis we have chosen to investigate electronic and optical properties of QDs and QDMs grown on [11k] substrates, where k = 1, 2, 3 (Chapter 4), type II InP/InGaP QDs and InAs QDs capped with GaAsSb (Chapter 5), and recently fabricated unstrained GaAs QDs in an external magnetic field (Chapter 6).

In Chapter 4 we discuss the influence of the substrate orientation on the electronic and optical properties of QDs and QDMs, and point out the role of piezoelectricity for InAs/GaAs QDs grown on [11k], where k = 1, 2, 3, 4, 5, 7, 9 and for QDMs containing eight InAs/GaAs QDs grown on [11l], where l = 1, 2, 3. Type II InP/InGaP QDs and InAs QDs capped with GaAsSb in the presence of an external magnetic field are investigated in Chapter 5, where we compare our findings with the available experimental data. Finally, Chapter 6 is devoted to
the investigation of the influence of interface smoothing and effects of an external magnetic field on the electronic and optical properties of recently fabricated unstrained GaAs/AlGaAs QDs. Due to the nonexistence of strain and reduced uncertainties in the size and shape, such QDs represent an ideal test case for electronic structure models.
Chapter 2

An eight-band $k\cdot p$ model for nanostructures

This may be viewed as a modern confirmation of Luttinger’s original work demonstrating the importance of including antisymmetric terms in the Hamiltonian.

Bradley A Foreman (2006b)

Abstract

In the framework of the Burt-Foreman theory a nonsymmetrized eight-band effective-mass Hamiltonian is derived for nanostructures in the presence of a magnetic field. The Hamiltonian is tested for the case of a cylindrical quantum dot with parabolic in-plane confinement potential in a perpendicular magnetic field. We compare the results of our nonsymmetrized model with the single-band and conventional multiband calculations, where ad-hoc symmetrization is used. The model is tested on GaAs/Al$_0.3$Ga$_0.7$As, GaAs/AlAs and InAs/GaAs quantum dots, where strain is not included in the model in order to resolve the influence of the boundary on the electronic structure. In structures with a large difference of Luttinger parameters between the constituent materials, like InAs/GaAs quantum dots, the conventional multiband models lead to non-physical high magnetic field solutions which are substantially different from those obtained from the nonsymmetrized Hamiltonian and single-band model for the ground state. A similar behavior is observed for the case of InAs/GaAs quantum wells, where energy levels as a function of $k_z$ are analyzed. This discrepancy is attributed to an overestimation of band mixing in conventional models because of the inappropriate treatment of the boundary.

2.1 Validity of the envelope function theory for nanostructures

The effective-mass theory is a very well established method to obtain the band structure in the case of weak perturbing inhomogeneous semiconductor potentials (Luttinger and Kohn 1955, Luttinger 1956, Kane 1957, Weiler et al. 1978, Weiler 1981). Electron and hole energies near the band extrema in the presence of external magnetic and electric fields and in a crystal containing a shallow impurity, were successfully described by
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This theory (Luttinger 1956, Kane 1957, Pidgeon and Brown 1966). It is also well suited for valence bands, but because of the degeneracy between the heavy-hole and light-hole bands, the multiband approach should be applied. The majority of current theoretical work on the calculations of the electronic structure is based on the Luttinger-Kohn Hamiltonian (Luttinger and Kohn 1955, Luttinger 1956) for valence bands and Pidgeon-Brown Hamiltonian (Kane 1957, Pidgeon and Brown 1966) to consider mixing between valence and conduction bands. Although the validity of the effective mass approximation is broken (the weak perturbing potential condition is violated), it has nevertheless been proven to be a useful method to compute the electronic structure of semiconductor nanostructures, owing to its simplicity and reasonable agreement with experiment. For this purpose, the appropriate bulk multiband Hamiltonian is used in each constituent material separately, and the envelope functions on either side of the interface are connected by applying ad-hoc symmetrization rules as proposed by Eppenga et al. (1987). We refer to this approach as the conventional multiband model.

An envelope function theory for nanostructures was developed by Burt (1992). The model is valid for the case of abrupt interfaces and it relies only on the slow variation of the envelope functions (Burt 1992, Burt 1999, Foreman 1998b). In the honor of the authors who developed it, this theory is sometimes referred to as Burt-Foreman (BF) theory. The fact that the Hamiltonian as a whole is Hermitian, but the matrix elements themselves are not necessarily so, was disregarded in the conventional multiband approach. The operator ordering with respect to the band parameters reflects the boundary conditions at the abrupt interface. This ordering for the nonsymmetrized multiband Hamiltonian was systematically derived by introducing asymmetry parameters, first for quantum wells and superlattices (Foreman 1993, Stavrinou and van Dalen 1997), and subsequently for arbitrary three-dimensional confinement in quantum dots (Pokatilov et al. 2001). However, the influence of an external magnetic field was not addressed up to now.

Furthermore, it has been argued very recently that most derivations of effective-mass equations for heterostructures, including Burt’s derivation, do not take into account all perturbative corrections up to the same order and that the additional approximation introduced by Foreman (1993), resulting in the asymmetry parameters, was incorrect (Foreman 2005). Several terms with different operator ordering were omitted, and the asymmetric term in the nonsymmetrized Hamiltonian was found to be incorrect which lead to the proposal (Foreman 2005) of a new first-principle envelope-function theory (FPEFT). A numerical implementation of the FPEFT (Foreman 2005) was very recently reported (Foreman 2007). The valence subband structure of GaAs/AlAs, GaAs/Al\(_x\)Ga\(_{1-x}\)As, and In\(_{0.53}\)Ga\(_{0.47}\)As/InP (001) superlattices was calculated using FPEFT and the obtained results were compared to those extracted from plane-wave pseudopotential total energy calculations. The valence subband structure was reproduced accurately over a wide energy range using FPEFT.

However, performing the electronic structure calculations of an arbitrary 3D nanostruc-
ture, that takes into account strain effects, growth on high index planes, and/or effects of an external magnetic field is a rather tedious task in the framework of the proposed FPEFT. All "elegancy" in the treatment of e.g. strain effects, effects of an applied magnetic field present in conventional as well as in the BF multi-band theory gets lost on behalf of the inclusion of the all non-zero, first-principle derived terms (Foreman 2006b, Foreman 2007) and the description of the whole Brillouin zone in the FPEFT. Through this work we will focus on the investigation of III-V nanostructures, in particular QDs, with conduction band minima and valence band maximum at the $\Gamma$ point. We calculate electronic and optical properties of such nanostructures comparing our findings with the available experimental data. The most elegant way to do so is by employing the multiband effective mass theory that captures most of the relevant effects present in the above mentioned system (band mixing, strain effects, piezoelectricity, magnetic field or electric field effects). The application of the conventional symmetrized Hamiltonian is still wide-spread and numerous experimental data were described in the framework of this theory, although the treatment of the interface in this model is problematic. The consequences of the ill-defined boundary conditions in the conventional model used to calculated the electronic structure of the nanostructure are still unknown. Therefore, we modify here the BF theory to include effects of an external magnetic field and implement it as a first correction to the usual approach. As argued above many subtle effects may not be included, but no non-physical solutions are found within this BF theory, in contrast to the results obtained with the conventional approach (see Sec. 2.3.2 for more details).

The aim of this chapter is to answer the following questions: (i) What is the form of the nonsymmetrized Hamiltonian in the case of a nanostructure in an external magnetic field? (ii) What is the correct interpretation of the interface asymmetric term present in the BF theory? (iii) What are the consequences of the ill-defined boundary conditions in the conventional approach on the calculated electronic structure of nanostructures?

### 2.2 Nonsymmetrized Hamiltonian for nanostructures in a magnetic field

In the BF theory, the multiband Hamiltonian for bulk crystals is modified so that (Burt 1992, Burt 1999, Foreman 1993): (i) the operators are ordered with respect to the effective mass parameters, which leads to specific boundary conditions at the atomically abrupt interface; (ii) the light-hole $|3/2, \pm 1/2\rangle$ and the split-off $|1/2, \pm 1/2\rangle$ states are coupled due to the local inversion asymmetry caused by the variation of the material composition.

The Hamiltonian for bulk crystals without spin-orbit coupling written in the basis $|S\rangle$, $|X\rangle$, $|Y\rangle$, $|Z\rangle$ of the periodic parts of the zone-center Bloch functions, is given by (Kane

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1It is assumed that inversion asymmetry of the zinc-blende lattice can be neglected
2. An eight-band k-p model for nanostructures

1957, Foreman 1997):

\[ H_4 = \begin{pmatrix} D_e & iPK_x & iPK_y & iPK_z \\ -iPK_x & D_{h1} & -N'k_xk_y & -N'k_xk_z \\ -iPK_y & -N'k_xk_y & D_{h2} & -N'k_yk_z \\ -iPK_z & -N'k_xk_z & -N'k_yk_z & D_{h3} \end{pmatrix}, \quad (2.1) \]

Here, \( D_e = E_v + \Delta k^2 \), \( D_{h1} = E_v' - L'k_x^2 - M(k_y^2 + k_z^2) \), \( D_{h2} = E_v' - L'k_y^2 - M(k_x^2 + k_z^2) \), \( D_{h3} = E_v' - L'k_z^2 - M(k_x^2 + k_y^2) \), \( E_v \) denotes the energy of the conduction band minimum, \( E_v' = E_v + \Delta / 3 \), where \( E_v \) is the energy of the valence band maximum, and \( \Delta \) is the spin-orbit split-off energy. \( P \) is the Kane matrix element defined as \( P = -i(h/m)\langle S|P_i|X \rangle \), \( L' = (\hbar^2/2m)(\gamma_1 + 4\gamma_2), M = (\hbar^2/2m)(\gamma_1 - 2\gamma_2), N'' = (\hbar^2/2m)(6\gamma_1) \), where \( m \) is the electron mass. Parameters \( \gamma_1 \) are related to the valence-band Luttinger parameters \( \gamma_\ell \) by the relations \( \gamma_1 = \gamma_1' = E_P/3E_g \), \( \gamma_2 = \gamma_2' - E_P/6E_g \), \( \gamma_3 = \gamma_3' - E_P/6E_g \), where \( E_g \) is the fundamental band gap, and \( E_P \) is related to the Kane matrix element by \( E_P = 2mP^2/\hbar^2 \). \( A_c \) is calculated from the expression \( A_c = \hbar^2/2m_e - 2P^2/3E_g - P^2/3(E_g + \Delta) \).

The operator ordering for the zero magnetic field case, proposed by Foreman (1997), is given by:

\[ \begin{align*}
    PK_{x_i} &= \frac{1}{2} \{ P_1 \hat{k}_{x_i} + \hat{k}_{x_i}P_2 \}, \\
    MK_{x_i}^2 &= \hat{k}_{x_i}M\hat{k}_{x_i}, \\
    NK_{x_i}N'_{x_j} &= \hat{k}_{x_i}N'_{x_j} + \hat{k}_{x_j}N'_{x_i}.
\end{align*} \quad (2.2) \]

where \( x_i, x_j = x, y, z; x_j \not= x_i \). \( P_1, P_2 \) are asymmetry parameters defined by Pokatilov et al. (2001). \( N'_{x} \) in Eq. (2.4) is the contribution to \( N' \) from the \( \Gamma_1 \) and \( \Gamma_{12} \) bands, while \( N' \) represents the influence of the \( \Gamma_{15} \) and \( \Gamma_{25} \) bands. If one neglects the \( \Gamma_{25} \) bands Eq. (2.4) becomes (Foreman 1997):

\[ \hat{k}_{x_i}N'_{x_j} + \hat{k}_{x_j}N'_{x_i} = 3(\hbar^2/2m)(\hat{k}_{x_i}(\gamma_3 + \chi)\hat{k}_{x_j} + \hat{k}_{x_j}(\gamma_3 - \chi)\hat{k}_{x_i}), \quad (2.5) \]

where \( \chi \) is called the asymmetry parameter in the notation of Pokatilov et al. (2001), and is given by \( \chi(r) = (2\gamma_2(r) + 3\gamma_3(r) - \gamma_1(r) - 1)/3 \).

2.2.1 Influence of an external magnetic field

If the operator ordering, given by Eqs. (2.2), (2.3), and (2.4) is implemented in the Hamiltonian for bulk material in the presence of a finite magnetic field (Luttinger and Kohn 1955) \( (k \rightarrow -i \nabla - eA/\hbar) \), it may be shown that the asymmetry parameter, which should only give a contribution at the interface, also contributes away from the interface. As a matter of fact, Eq. (2.5) can be written as:

\[ \frac{3}{2m} \hbar^2 \left( -i \frac{\partial(\gamma_3 + \chi)}{\partial x_i} \right) \hat{k}_{x_i} - i \frac{\partial(\gamma_3 - \chi)}{\partial x_j} \hat{k}_{x_j} + \gamma_3 \{ \hat{k}_{x_i}, \hat{k}_{x_j} \} + \chi \{ \hat{k}_{x_i}, \hat{k}_{x_j} \}, \quad (2.6) \]
The first two terms represent the correct boundary conditions. They arise from the different contributions of the remote bands of symmetry $\Gamma_1$, $\Gamma_{12}$ (the first term), and $\Gamma_{15}$ (the second term). In the case of the ad-hoc symmetrization these two terms also exist but in a different form $3(-i\partial \chi J_3/\partial x \hat{k}_{xj} - i\partial \chi J_3/\partial x \hat{k}_{xj})$, obtained by taking $\chi = 0$ in Eq. (2.5) and they follow from the fact that each matrix element of the Hamiltonian in the ad-hoc symmetrization has to be Hermitian, which is shown to be incorrect (Burt 1992, Burt 1999, Foreman 1997). The third term represents the anticommutator of the operators $\hat{k}_{xv}, \hat{k}_{xj}$. Note that both in the ad-hoc symmetrization and the bulk Hamiltonian, the third term appears. It arises from the symmetry of the crystal (Luttinger and Kohn 1955, Luttinger 1956). The fourth term is the commutator of the operators $\hat{k}_{vx}, \hat{k}_{xj}$ and it is proportional to the asymmetry parameter $\chi$. It plays a role not only at the boundary but through the whole nanostructure. The correct interpretation of this term is very important. We demonstrate that this term is essentially the same as the one introduced by Luttinger (1956). For the case of bulk semiconductors in a magnetic field, the commutator of different components of $k$, multiplied by the antisymmetric constant $K/2$ represents the antisymmetric term (Eq. (12) of Luttinger (1956)):

$$D_{XY} = C\{k_x, k_y\} + \frac{1}{2}K[k_x, k_y] .$$  \hspace{1cm} (2.7)

As we showed in Eq. (2.6), in the Burt-Foreman approach this antisymmetric term appears naturally in the derivation and it exists in bulk materials as well as in nanostructures. If one compares Eq. (2.7) and Eq. (2.6) in the case of bulk material (the first two terms in Eq. (2.6) are then equal to zero), one can obtain a direct relation between the antisymmetric constant $K$ and the asymmetry parameter $\chi$ from the BF theory. Note that we treat the conduction band together with the valence bands, so the hole effective masses are written in terms of the scaled Luttinger parameters $\gamma_i$ (with $i = 1, 2, 3$), as it was shown at the beginning of this section. If one excludes the conduction band from the model, $\gamma_i$ reduces to $\gamma^L_i$, and, as a consequence, $\chi$ reduces to $\chi^L = (2\gamma^L_2(r) + 3\gamma^L_3(r) - \gamma^L_1(r) - 1)/3$. Due to the fact that only the valence bands are treated explicitly in Luttinger’s Hamiltonian, $K$ is related to $\chi^L$ by $K = -(\hbar^2/m)3\chi^L$. Note that this term vanishes for zero magnetic field when $\hat{k}_{xv}, \hat{k}_{xj}$ commute. Further, we can relate the asymmetry parameter $\chi$ to the dimensionless constant $\kappa$, which was introduced by Luttinger (1956): $(\hbar^2/2m)(3\kappa + 1) = -K$. To be consistent, we denote Luttinger’s $\kappa$ as $\kappa^L$. In our case, in which the conduction band is treated together with the valence bands (see Sec. 1.7.2), the scaled $\kappa$ is related to the Luttinger’s $\kappa^L$ by $\kappa = \kappa^L - E_p/(6E_d)$. The direct relation between $\kappa^L$ and the asymmetry parameter $\chi^L$ from the BF theory is then given by $\kappa^L = \chi^L - 1/3$. Note that it is equivalent to Eq. (23) of Pidgeon and Brown (1966) where $\kappa^L = \gamma^L_1 + (2/3)\gamma^L_2 - (1/3)\gamma^L_3 - 2/3$. Inclusion of the conduction band into the model leads to the replacement $\kappa^L \rightarrow \kappa$ and $\chi^L \rightarrow \chi$, resulting in the relation

$$\kappa = \chi - \frac{1}{3} ,$$  \hspace{1cm} (2.8)
2.2.2 Final form of the Hamiltonian

When the spin-orbit interaction is included the basis of the periodic parts of the Bloch functions is given by:

\[
\begin{align*}
|1/2, 1/2\rangle &= |S \uparrow\rangle, \\
|1/2, -1/2\rangle &= |S \downarrow\rangle, \\
|3/2, 3/2\rangle &= \frac{1}{\sqrt{2}} (|X \uparrow\rangle + i|Y \uparrow\rangle), \\
|3/2, 1/2\rangle &= \frac{1}{\sqrt{6}} (|X \uparrow\rangle + i|Y \downarrow\rangle - 2|Z \uparrow\rangle), \\
|3/2, -1/2\rangle &= \frac{1}{\sqrt{6}} (|X \uparrow\rangle - i|Y \uparrow\rangle + 2|Z \downarrow\rangle), \\
|3/2, -3/2\rangle &= \frac{1}{\sqrt{3}} (|X \downarrow\rangle - i|Y \downarrow\rangle), \\
|1/2, 1/2\rangle &= \frac{1}{\sqrt{3}} (|X \uparrow\rangle + i|Y \downarrow\rangle + |Z \uparrow\rangle), \\
|1/2, -1/2\rangle &= \frac{1}{\sqrt{3}} (|X \uparrow\rangle - i|Y \downarrow\rangle - |Z \downarrow\rangle).
\end{align*}
\]

In the single-group representation the \( \Gamma_8 \times \Gamma_8 \) parameters \( \gamma_1, \gamma_2, \gamma_3, \kappa \) also involve the split-off band \( \Gamma_7 \), and this representation is the one which we will use. However, in the full double-group picture the \( \Gamma_7 \times \Gamma_7 \) and \( \Gamma_7 \times \Gamma_8 \) parameters are independent of the \( \Gamma_8 \times \Gamma_8 \) ones (Weiler et al. 1978, Weiler 1981). Following the procedure by Weiler et al. (1978) the complete \( 8 \times 8 \) nonsymmetrized Hamiltonian is given by

\[
\hat{H} = \begin{pmatrix} H_{11}(k) & H_{12}(k) \\ H_{21}(k) & H_{22}(k) \end{pmatrix},
\]

where \( H_{ij}(k) \) are \( 4 \times 4 \) matrices given by:

\[
H_{11}(k) = \begin{pmatrix} D_{\uparrow\downarrow} & C_{\uparrow\downarrow} + 2(N_1 + \frac{1}{2}) & iV_+ & \sqrt{\frac{3}{2}} V_0 \\ C_{\uparrow\downarrow} + 2(N_1 + \frac{1}{2}) & D_{\uparrow\downarrow} & 0 & -\frac{1}{\sqrt{3}} V_+ \\ -iV_+ & 0 & D_{\uparrow\uparrow} & -S_- - i\sqrt{3} \sqrt{\gamma} C_{\uparrow\downarrow} - i\sqrt{3} (\kappa + \frac{3}{2} q) H_- \\ \sqrt{\frac{3}{2}} V_0 & -\frac{1}{\sqrt{3}} V_1 & -S_- + i\sqrt{3} \sqrt{\gamma} C_{\uparrow\downarrow} + i\sqrt{3} (\kappa + \frac{3}{2} q) H_+ & D_{\uparrow\uparrow} \end{pmatrix},
\]

\[
H_{12}(k) = \begin{pmatrix} i\frac{1}{2} V_- & 0 & \frac{1}{2} V_0 & \sqrt{\frac{3}{2}} V_- \\ i\frac{1}{2} V_0 & -V_- & i\frac{1}{2} V_+ & -\frac{1}{\sqrt{3}} V_0 \\ -R & \frac{1}{2} (C_{\uparrow\downarrow} + q H_+) & \sqrt{\frac{3}{2}} (1 + \kappa) H_- & i\sqrt{2} R \\ -C - \frac{1}{2} i(\gamma_2 + q) H_- & \sqrt{\frac{3}{2}} (1 + \kappa) H_- & -i\sqrt{2} \Sigma_2 - i\sqrt{3} (\kappa + \frac{3}{2} q) H_+ & \sqrt{\frac{3}{2}} (1 + \kappa) H_- \end{pmatrix},
\]

\[2\text{Choice of a parameter set used in } k \cdot p \text{ model and consequences of single-group approximation for } \Gamma_7 \times \Gamma_7, \Gamma_7 \times \Gamma_8, \text{ and } \Gamma_8 \times \Gamma_8 \text{ are discussed in Sec. 3.5.}\]
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\[ H_{21}(k) = \begin{pmatrix}
-\frac{i}{\sqrt{3}} V^t_0 - i\sqrt{\frac{3}{2}} V^t_0 & -R^t & -C^t + 2i\frac{1}{2} C_{pq} + i\frac{1}{2}(\kappa + \frac{3}{4} q) H_+ - R^t \\
0 & -V^t_0 - \frac{i}{2}(C_{pq} + q H_-) & -R^t \\
-\frac{i}{\sqrt{3}} V^t_0 - i\sqrt{\frac{3}{2}} V^t_0 & \frac{i}{\sqrt{3}} S^t_+ + \frac{1}{2}(1 + \kappa) H_+ & -i\sqrt{2}(Q_+ + H_+)^t \\
\sqrt{\frac{3}{2}} V^t_0 & -i\sqrt{\frac{3}{2}} V^t_0 & -i\sqrt{2} R^t \\
\end{pmatrix}, \quad (2.13) \]

\[ H_{22}(k) = \begin{pmatrix}
D_{hh-} & S^t_+ - i\sqrt{\frac{3}{2}} C_{pq} - i\sqrt{2} \Sigma_- \sqrt{\frac{1}{2} (1 + \kappa) H_+} & i\sqrt{2}(Q_+ - H_+) \\
S_+ + i\sqrt{\frac{3}{2}} C_{pq} & D_{hh-} & i\sqrt{2} R^t \\
-\frac{i}{\sqrt{2}} \Sigma^t_- + \frac{1}{2}(1 + \kappa) H_- & -i\sqrt{2} R & D_{oo+} \\
-i\sqrt{2}(Q_+ - H_+) & \sqrt{\frac{1}{2} (1 + \kappa) H_-} & C^t - i\sqrt{2} \Sigma^t_- \sqrt{\frac{1}{2} (1 + \kappa) H_-} \\
\end{pmatrix}, \quad (2.14) \]

where

\[ D_{ch\pm} = E_v + T \pm \frac{\hbar^2}{2m} (2N_1 + 1) i[\hat{k}_x, \hat{k}_y] \pm 2i C_{N_1}, \quad (2.15) \]

\[ D_{hh\pm} = E_v - (P_\pm + Q_\pm) \mp \frac{\hbar^2}{2m} 3(\kappa + (9/4) q) i[\hat{k}_x, \hat{k}_y] \mp (27/4) i C_{q_1}, \]

\[ D_{hh\pm} = E_v - (P_\pm - Q_\pm) \mp \frac{\hbar^2}{2m} (\kappa + (1/4) q) i[\hat{k}_x, \hat{k}_y] \pm (1/4) i C_{q_1}, \]

\[ D_{oo\pm} = E_v - (P_\pm \mp Q_\pm) \mp \frac{\hbar^2}{2m} (2\kappa + 1) i[\hat{k}_x, \hat{k}_y], \]

\[ \hat{k}_+ = \frac{k_x + ik_y}{\sqrt{2}}, \quad \hat{k}_- = \frac{k_x - ik_y}{\sqrt{2}}, \quad (2.16a) \]

\[ H_c = \frac{\hbar^2}{2m} (1 + \kappa)[\hat{k}_x, \hat{k}_y], \quad (2.16b) \]

\[ H_\pm = \pm \frac{\hbar^2}{2m} \sqrt{2}[k_\pm, k_c], \quad (2.16c) \]
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\[ V_\pm = \frac{1}{2}(P_1 \hat{k}_\pm + \hat{k}_\pm P_2) , \]
\[ V_0 = \frac{1}{2}(P_1 \hat{k}_z + \hat{k}_z P_2) , \]
\[ T = \hat{k}_+ A_\pm \hat{k}_- + \hat{k}_- A_\pm \hat{k}_+ + \hat{k}_z A_\pm \hat{k}_z , \]
\[ P_\pm = \frac{\hbar^2}{2m}(\gamma_1 \{\hat{k}_\pm, \hat{k}_\mp\} + (\hat{k}_\pm(\gamma_1 - 2\chi))\hat{k}_\mp + (\hat{k}_\mp(\gamma_1 + 2\chi))\hat{k}_\pm + \hat{k}_z \gamma_1 \hat{k}_z) , \]
\[ Q_\pm = \frac{\hbar^2}{2m}(\gamma_2 \{\hat{k}_\pm, \hat{k}_\mp\} + (\hat{k}_\pm(\gamma_2 - \chi))\hat{k}_\mp + (\hat{k}_\mp(\gamma_2 + \chi))\hat{k}_\pm + 2\hat{k}_z \gamma_2 \hat{k}_z) , \]
\[ R = \sqrt{3} \frac{\hbar^2}{2m} (\hat{k}_+(\gamma_2 - \gamma_1)\hat{k}_+ + \hat{k}_-(\gamma_2 + \gamma_1)\hat{k}_-) , \]
\[ S_\pm = \pm i\sqrt{6} \frac{\hbar^2}{2m}(\gamma_3 \{\hat{k}_\pm, \hat{k}_\mp\} + (\hat{k}_\pm(\gamma_3 + \chi))\hat{k}_\mp + (\hat{k}_\mp(\gamma_3 - \chi))\hat{k}_\pm) , \]
\[ \Sigma_\pm = \pm i\sqrt{6} \frac{\hbar^2}{2m}(\gamma_3 \{\hat{k}_\pm, \hat{k}_\mp\} + (\hat{k}_\pm(\gamma_3 - \chi))\hat{k}_\mp + (\hat{k}_\mp(\gamma_3 + \chi))\hat{k}_\pm) , \]
\[ C = -i2\sqrt{2} \frac{\hbar^2}{2m}(\hat{k}_-\chi)\hat{k}_z - (\hat{k}_z\chi)\hat{k}_-). \]
\[ C_{q\pm} = \pm \sqrt{2} \frac{\hbar^2}{2m}(\hat{k}_\pm q)\hat{k}_z - (\hat{k}_z q)\hat{k}_\pm) . \]
\[ C_{q_z} = \frac{\hbar^2}{2m}(\hat{k}_z q)\hat{k}_y - (\hat{k}_y q)\hat{k}_z) . \]
\[ C_{N_1\pm} = \pm 2\sqrt{2} \frac{\hbar^2}{2m}(\hat{k}_\pm N_1)\hat{k}_z - (\hat{k}_z N_1)\hat{k}_\pm) . \]
\[ C_{N_1_z} = \frac{\hbar^2}{2m}((\hat{k}_z N_1)\hat{k}_y - (\hat{k}_y N_1)\hat{k}_z) . \]

Parameter $N_1$ was introduced by Weiler et al. (1978) and contributes to the electron $g$ factor, whereas $q$ is the parameter introduced by Luttinger (1956) and to leading order is proportional to the spin-orbit splitting of the remote B states (see Secs. 1.7.1 and 1.7.2 for discussion about division of all states into two sets: set A containing the bands of interest and set B containing remaining bands). Because of a reduced symmetry at the interface, also an additional coupling is caused by these parameters $q$ and $N_1$ at the interface. It is included in the Hamiltonian Eq. (2.10) through $C_{q\pm}$, $C_{q_z}$, $C_{N_1\pm}$, and $C_{N_1_z}$ given by Eqs. (2.17), but in our numerical calculations in Sec. 2.3 we assume that these two parameters are zero. Values of parameter $N_1$ for various III-V semiconductors are not known, whereas the values for $q$ as
given by Lawaetz (1971), are 0.04, 0.04, 0.03, 0.13, 0.01 and 0.01 for GaAs, InAs, AlAs, GaSb, InP, and GaP respectively.

In Eq. (2.10) \( \dagger \) denotes Hermitian conjugation. We avoided to use complex conjugation of operators and transpose of operators as proposed by Pokatilov et al. (2001). Without magnetic field \( \hat{k}_i^0 = -\hat{k}_x \), and \( \hat{k}_i^+ = -\hat{k}_z \), but in the case of a magnetic field \( \hat{k}_i^0 \neq -\hat{k}_x \), and \( \hat{k}_i^+ \neq -\hat{k}_z \). We adopt the convention introduced by Foreman (1993).

In Eqs. (2.16g), (2.16h) \{\( \hat{k}_\pm, \hat{k}_z \)\} = \( \hat{k}_z \hat{k}_\pm + \hat{k}_\pm \hat{k}_z \), and in Eqs. (2.16j), (2.16k) \{\( \hat{k}_\pm, \hat{k}_z \)\} = \( \hat{k}_\pm \hat{k}_z + \hat{k}_z \hat{k}_\pm \).

In the case of a nanostructure in a perpendicular magnetic field, the Zeeman terms on the diagonal and those which couple light-hole and split-off bands remain, and all others become zero. This case will be considered in the next section where a cylindrical quantum dot in a perpendicular magnetic field is analyzed.

If the asymmetry parameter \( \chi \) is taken equal to zero, this Hamiltonian has the same form as the conventional 8 \( \times \) 8 multiband Hamiltonian for the magnetic field case.

**Strain Hamiltonian:** In the case of strained semiconducting heterostructures in a magnetic field an additional strain dependent part should be added to the heterostructure Hamiltonian:

\[
H = H_k + H_s
\]

where \( H_k \) denotes the kinetic part given by Eq. (2.10) and \( H_s \) is the strain dependent part.

In what follows we give the explicit terms of the strain dependent part of the Hamiltonian. As adapted from Pryor (1998), the strain Hamiltonian in the basis given in Eq. (2.9) becomes

\[
\hat{H}_s = \begin{pmatrix}
a_e e & 0 & -i\sqrt{3}v & -\sqrt{2}u & -iv & 0 & -iu & -\sqrt{2}v \\
0 & a_e e & 0 & i\sqrt{3}v & -i\sqrt{2}u & \sqrt{3}v & -i\sqrt{2}v & u \\
-i\sqrt{2}u & 0 & -p - q & is & r & 0 & -\frac{1}{\sqrt{3}}s & -i\sqrt{2}r \\
i\sqrt{2}u & 0 & -is & -p - q & 0 & r & i\sqrt{2}q & -\frac{1}{\sqrt{3}}s \\
i\sqrt{3}v & 0 & r & 0 & -p + q & -is & -\frac{3}{2}s^3 & i\sqrt{2}q \\
0 & i\sqrt{3}v & r & 0 & -is & -p + q & -\frac{3}{2}s^3 & -i\sqrt{2}r \\
iu & i\sqrt{2}u & -\frac{1}{\sqrt{2}}s^3 & -i\sqrt{2}q & -\sqrt{3}v & i\sqrt{2}r & -a_e e & 0 \\
-\sqrt{2}v & u & i\sqrt{2}r & -\sqrt{3}v & -i\sqrt{2}q & -\frac{1}{\sqrt{2}}s^3 & 0 & -a_e e \\
\end{pmatrix}
\]

(2.19)

where

\[
p = a_e (e_{xx} + e_{yy} + e_{zz}),
\]

(2.20a)

\[
q = b [e_{zz} - \frac{1}{2}(e_{xx} + e_{yy})],
\]

(2.20b)

\[
r = \frac{\sqrt{3}}{2} b (e_{xx} + e_{yy}) - ide_{yy},
\]

(2.20c)
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\[ s = -d(e_{xz} - ie_{yz}), \quad (2.20d) \]

\[ u = \frac{1}{\sqrt{3}}P_0 \sum_j e_{z j} k_j, \quad (2.20e) \]

\[ v = \frac{1}{\sqrt{6}}P_0 \sum_j (e_{x j} - ie_{y j}) k_j, \quad (2.20f) \]

In the previous equations $a_v$ is the hydrostatic valence-band deformation potential, $a_c$ the conduction-band deformation potential, $e_{ij}$ is the strain tensor, where $i, j$ runs over $x, y, z$, and $b$ and $d$ are the shear deformation potentials.

Numerical implementation of the presented Hamiltonian for modelling arbitrary three-dimensional nanostructures in an external magnetic field and grown on high-index planes is presented in Chapter 5.

2.3 Comparison of different models

2.3.1 Model quantum dot

In order to test the nonsymmetrized Hamiltonian, we calculate the electronic structure of a cylindrical quantum dot with parabolic in-plane confinement potential and compare the results with those of the conventional multiband models. A hard-wall potential of rectangular shape is assumed along the $z$-direction, and the case of a perpendicular magnetic field is considered. The axial approximation is adopted. It corresponds to ignoring the square terms that are not axially symmetric about the $z$ axis (Eq. (2.16i)). Since the confinement potential has cylindrical symmetry, the $z$ component of the total angular momentum $F$ can be introduced as a good quantum number, $F_z = f_z \hbar$ (Pedersen and Chang 1996, Pedersen and Chang 1997). The $z$ projection of the total angular momentum $F$ can be written as $F_z = J_z + L_z$, where $J_z$ is the $z$ component of the angular momentum of the band-edge Bloch function and $L_z$ is the $z$ component of the envelope angular momentum. In the presence of a magnetic field, the operators $k_\pm$ take the form:

\[ k_\pm = -ie^{\pm i\phi} \left( \frac{\partial}{\partial \rho} \mp \frac{i}{\rho} \frac{\partial}{\partial \phi} \mp \frac{\rho}{2l_c^2} \right), \quad (2.21) \]

in cylindrical coordinates. Here $l_c$ is the magnetic length given by $l_c = (\hbar/eB)^{1/2}$.

The states are described by the Hamiltonian:

\[ H = H_{kin} + V(\rho, z), \quad (2.22) \]

where $H_{kin}$ presents the kinetic part given in Eq. (2.10) and $V(\rho, z)$ is a diagonal matrix containing potentials for the valence and conduction band. For GaAs/Al$_{0.3}$Ga$_{0.7}$As the
8-band effective mass Hamiltonian is employed, while for GaAs/AlAs, InAs/GaAs we use the 6-band effective-mass Hamiltonian. AlAs is indirect-gap semiconductor, so coupling between the valence and conduction band cannot be modelled by the $8 \times 8$ Hamiltonian. In the case of an InAs/GaAs quantum dot the full picture can be obtained by employing the $8 \times 8$ Hamiltonian (Pryor 1998). But here, we are interested in the influence of the boundary conditions on the electronic structure calculations of the valence band. In order to analyze the influence of the boundary conditions we have already excluded the strain from our calculations. For the valence band the boundary conditions extracted from the conventional approach are significantly different than those extracted from the nonsymmetrized one. By resolving the problem one gets a more precise treatment of the abrupt interface in the framework of the BF multiband effective-mass theory for the valence band and one could further incorporate these boundary conditions in the full $8 \times 8$ nonsymmetrized Hamiltonian. It should be pointed out that generalized envelope function theory should include additional material parameters, whose values can be obtained from the first-principles calculations or experiments on heterostructures.

For the parabolic quantum dot, $V(\rho, z) = V_\parallel(\rho) + V_\perp(z)$, where $V_\parallel(\rho)$ and $V_\perp(z)$ are given by:

$$V_\parallel(\rho) = \frac{1}{2} \omega_0^2 m_i \rho^2,$$

$$V_\perp(z) = \begin{cases} \Delta E_i & \text{for } |z| > h/2, \\ 0 & \text{for } |z| < h/2. \end{cases}$$

where $i = c, h, m_e$ is the electron mass, $m_h = m_0/(\gamma_1 + \gamma_2)$ is the heavy-hole mass, and $\omega_0$ is the characteristic frequency of the lateral confinement potential.

If the quantum dot is symmetric in the $z$-direction, the parity of the wave function is a good quantum number. The spinor of the even valence-band state has the form (Tadić, Peeters and Janssens 2002):

$$F_+ = [F_{cb}^+, F_{ch}^+, F_{lh}^+, F_{hh}^+, F_{so}^+, F_{so}^-]_s.$$

The states, for a given quantum number $f_z$, are denoted by $nX_{f_z}^{par}$ (Pedersen and Chang 1997), where $n$ is the label of the state for given $f_z$, $X$ denotes the states with lowest $|l|$ among bands in the basis, and $par$ represents the total parity of the state (+ for even parity, − for odd parity). It is obvious that crossings between states of the same total parity and the same $f_z$ are forbidden, due to the fact that states are classified with respect to $f_z$, parity, and $n$. The ordering of the even and odd states depends on the dimensions of the quantum dot.

The envelope functions in both the valence and conduction bands are expanded into

$$\chi_{n,l,s}^\pm = C(n,l,s) \Phi_{nl}(\rho, \phi) f_s^\pm(z).$$
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The in-plane part is given by

\[ \Phi_{nl}(\rho, \phi) = \frac{1}{\sqrt{2\pi}} \exp(i\phi) \exp(-\rho^2/2a^2)(\rho/a)L_n^{||}(\rho^2/a^2), \]  

(2.27)

while the z-dependent part is given by

\[ f^+_s(z) = \frac{1}{\sqrt{L_z}} \cos \frac{m\pi z}{2L_z}, \quad m = 1, 3, 5, \ldots, \]  

(2.28)

\[ f^-_s(z) = \frac{1}{\sqrt{L_z}} \sin \frac{m\pi z}{2L_z}, \quad m = 2, 4, 6, \ldots. \]  

(2.29)

In Eqs. (2.26, 2.27, 2.28, 2.29), \( C(n, l, s) \) is a normalization constant, \( L_n^{||}(x) \) is the generalized Laguerre polynomial, \( L_z \) denotes the half-height of the expansion cylinder, and \( l \) is computed for the given \( f \) and \( j \). The length \( a \) is related to the magnetic length \( l_c \) and the harmonic length \( l_0 \) through

\[ a = \left( \frac{2l_0^2l_c^2}{l_0^4 + 4l_c^4} \right)^{1/2}. \]

We assumed a step variation of the material parameters, for example for \( \gamma_i = \gamma_{im} + (\gamma_{id} - \gamma_{im}) (\theta(z+w) - \theta(z-w)) \), where index \( m \) denotes the value of the Luttinger parameter in the matrix and index \( d \) the value of this parameter in the dot, \( w \) is the half-height of the dot and \( \theta \) is the Heaviside step function. The parameters used for our model quantum dot are based on \( \Gamma \)-point band structure parameters and are given in Table 2.1.

**Table 2.1: Material parameters used for the electronic structure calculations (Vurgaftman et al. 2001)**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Units</th>
<th>GaAs</th>
<th>AlAs</th>
<th>InAs</th>
</tr>
</thead>
<tbody>
<tr>
<td>( m^*_c )</td>
<td>( m_0 )</td>
<td>0.067</td>
<td>0.15</td>
<td>0.026</td>
</tr>
<tr>
<td>( \gamma_1^L )</td>
<td></td>
<td>6.98</td>
<td>3.76</td>
<td>20.00</td>
</tr>
<tr>
<td>( \gamma_2^L )</td>
<td></td>
<td>2.06</td>
<td>0.82</td>
<td>8.5</td>
</tr>
<tr>
<td>( \gamma_3^L )</td>
<td></td>
<td>2.93</td>
<td>1.42</td>
<td>9.2</td>
</tr>
<tr>
<td>( \kappa^L )</td>
<td></td>
<td>1.28</td>
<td>0.12</td>
<td>7.68</td>
</tr>
<tr>
<td>( \Delta_0 )</td>
<td>eV</td>
<td>0.341</td>
<td>0.28</td>
<td>0.39</td>
</tr>
<tr>
<td>( E_g )</td>
<td>eV</td>
<td>1.519</td>
<td>–</td>
<td>0.417</td>
</tr>
<tr>
<td>( E_P )</td>
<td>eV</td>
<td>28.8</td>
<td>21.1</td>
<td>21.5</td>
</tr>
</tbody>
</table>

In our numerical implementation, the half-height of the expansion cylinder is assumed to be approximately four times larger than the half-height of the dot. Eight in-plane and forty \( z \)-dependent basis functions were employed in the calculation. If the basis size is increased, the energy levels shift by less than 0.8 meV.

Hole energy levels are given with respect to the top of the valence band of the barrier material.
2.3. Comparison of different models

2.3.2 Nonsymmetrized versus conventional model

The energy levels of the holes are computed as they vary with the magnetic field for a range of confinement strengths and thicknesses of our model quantum dot. We tested the nonsymmetrized \(8 \times 8\) multiband Hamiltonian by comparing the energy levels with those extracted from the respective conventional one. Also, \(6 \times 6\) multiband nonsymmetrized and conventional models are compared. The difference between the nonsymmetrized and the conventional Hamiltonian arises from the different treatment of the interface in these approaches. As previously discussed, the asymmetry parameter arises at the interface and contributes to the effective mass parameters in the nonsymmetrized Hamiltonian, while in the conventional Hamiltonian this parameter equals zero. Consequently, we expect that agreement between these models strongly depends on the structural parameters, and therefore we compare these two models as a function of the magnetic field for different materials in the dot and the barrier.

**GaAs/Al\(_{0.3}\)Ga\(_{0.7}\)As system:** The magnetic field dependence of the three lowest hole energy levels of \(S_{-3/2}^+\) symmetry in a GaAs/Al\(_{0.3}\)Ga\(_{0.7}\)As quantum dot is shown in Fig. 2.1. The \(6 \times 6\) and \(8 \times 8\) nonsymmetrized and conventional Hamiltonians are compared. With varying in-plane confinement potential as well as height of the dot, the two models give similar results, but notice that the discrepancy between the two models is larger for the more strongly confined quantum dot (Fig. 2.1(b)). By explicitly including the lowest conduction band (\(8 \times 8\) model), results do not change qualitatively with respect to those from the \(6 \times 6\) model (Fig. 2.1). For the analyzed quantum dot the lowest hole energy levels for sixteen different symmetries are shown in Fig. 2.2 obtained by the \(6 \times 6\) nonsymmetrized multiband Hamiltonian. The expected behavior of energy levels as a function of magnetic field is observed, and it was already analyzed by Pedersen and Chang (1997).

**GaAs/AlAs system:** For GaAs/AlAs we used only the \(6 \times 6\) nonsymmetrized and conventional Hamiltonians. Energy levels of holes as a function of magnetic field are shown in Fig. 2.3. In this case the difference in Luttinger parameters in the dot and barrier is larger than in GaAs/Al\(_{0.3}\)Ga\(_{0.7}\)As (see Table I), and we notice that the difference between the two approaches becomes more pronounced. However, the energy level behavior as a function of the magnetic field, is qualitatively the same, only an energy shift is observed.

There is a good qualitative agreement between these two models, and the single band model (dotted lines in Figs. 2.1, 2.3), for the ground state, in both GaAs/Al\(_{0.3}\)Ga\(_{0.7}\)As and GaAs/AlAs as well. However, in the case of a large in-plane confinement potential, when band mixing becomes more important, the discrepancy in the results obtained by the single-band and the multiband models is more pronounced (see Figs. 2.1(b), 2.3(b)). Another discrepancy is also observed for low values of the magnetic field. The Zeeman energy term
in the single-band model tends to decrease the energy, but with increasing the value of the magnetic field the kinetic energy part becomes dominant which further leads to the expected behavior of the energy level as a function of the magnetic field. In the multiband model, however, band-mixing plays an important role, and it partially suppresses the effect of the Zeeman energy term. For high values of the magnetic field, a fully agreement between the results of the single-band model and the multiband model for the ground state is obtained.

**InAs/GaAs system:** Next we consider the case of an InAs/GaAs quantum dot in a magnetic field. The results are shown in Figs. 2.4(a) and 2.4(b), which exhibit the ground and first excited hole energy level in a quantum disk with $\hbar \omega_0 = 6$ meV and $\hbar \omega_0 = 10$ meV, respectively. In order to fully resolve the influence of the boundary on the electronic structure, strain was not included in the model although the lattice mismatch is 7.2%. The effect of
2.3. Comparison of different models

Figure 2.2: The lowest hole energy levels for sixteen different symmetries as a function of the magnetic field in GaAs/Al$_0.3$Ga$_{0.7}$As quantum dot: $\hbar \omega_0 = 10$ meV, and $h = 10$nm. The results are given for the $6 \times 6$ nonsymmetrized multiband Hamiltonian.

strain will be addressed at the end of this section. Results obtained by the nonsymmetrized Hamiltonian and the single band model show a good agreement, but those obtained by the conventional models differ appreciably. For high values of the magnetic field, the values of the energy levels obtained in the framework of conventional $\mathbf{k} \cdot \mathbf{p}$ theory, become non-physical. Indeed, note that for the $h = 10$ nm high dot (Fig. 2.4), and for values of the magnetic field $B \geq 27$ T, the energy levels lie above the top of the valence band (the top of the valence band of InAs is at 0.18734eV.) With increasing height of the dot, the effect of the interface boundary becomes less important, and consequently the difference between the energies computed by the different models becomes smaller. Only when the quantum dot thickness is larger than $\sim 60$ nm, the results obtained by the two models become identical in the considered magnetic field range. We found that such a behavior is due to the $S$ element in the Hamiltonian.

\begin{equation}
S_\pm = \pm i \sqrt{6} \frac{\hbar^2}{2m} (2 \gamma_3 \hat{k}_z - i \frac{d(\gamma_3 - \chi)}{dz}) \hat{k}_\pm. \tag{2.30}
\end{equation}

The influence of the boundary on the “conventional” $S$ element is determined by the factor $d(\gamma_3 - \chi)/dz$ in Eq. (2.30) and is $(\gamma_3 - \gamma_m)/(\gamma_3 - \chi_d) \approx 135$ times larger than in the nonsymmetrized $S$ element for the InAs/GaAs dot under study. As a consequence, band mixing is overestimated in the conventional Hamiltonian, which is large for materials with a large difference in Luttinger parameters. This overestimation leads to nonphysical solutions for high magnetic field values, as $d(\gamma_3 - \chi)/dz$ is multiplied by $k_\pm$ in Eq. (2.30), which depends on the magnetic field. Note that any other dependence of $\hat{k}_\pm$ (e.g. on the parabolic confinement strength) will cause such an amplification as well. The same behavior can be expected in the more simple case of a quantum well in the absence of a magnetic
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Figure 2.3: The three lowest hole energy levels of $S^{\pm 1/2}$ symmetry as a function of the magnetic field in a GaAs/AlAs quantum dot: (a) $\hbar \omega_0 = 6$ meV, $h = 6$ nm, (b) $\hbar \omega_0 = 30$ meV, $h = 4$ nm.

field, where $\hat{k}_\pm \to k_\pm$ is now a number. In this case we calculate the hole energy levels as a function of $k_t$ ($k_t = \sqrt{k_x + k_y}$). This is shown in Fig. 2.5 for quantum well growth in the [001] direction. In this case, $k_t$ artificially amplifies the effect of the boundary and it, as well, leads to nonphysical solutions, i.e. the energy levels, for high values of $k_t$, lie above the top of the valence band. It should be pointed out that this comparison was made just to illustrate the influence of the boundary on the electronic structure calculations for nanostructures and to stress that the observed nonphysical solutions as a function of a magnetic field for a quantum dot in a magnetic field are a consequence of the ill-defined boundary conditions at the abrupt interface.

In order to illustrate the difference between the conventional and nonsymmetrized approaches, the probability densities in the ground state at $B = 40$ T for the InAs/GaAs quantum dot and at $k_t = 0.5$ nm$^{-1}$ for the InAs/GaAs quantum well determined by the nonsymmetrized
and conventional Hamiltonians are shown in Figs. 2.6 and 2.7 respectively. Notice that the state shown in Fig. 2.6(a) is heavy hole like, which explains the good agreement between the singe-band and the nonsymmetrized model displayed in Fig. 2.4. In the conventional approach, however, there is a strong mixing between the heavy and light hole, and the holes...
An eight-band k·p model for nanostructures

Figure 2.5: The three lowest hole energy levels of InAs/GaAs quantum well as a function of $k_t$. Results are given for the nonsymmetrized Hamiltonian (solid line), and the conventional $6 \times 6$ Hamiltonian (dashed line). Width of the well varies from 10 nm to 60 nm.

are localized near the boundaries in the $z$ direction. One can notice a similar behavior in the quantum well case, where holes, according to the conventional approach, are also localized near the boundary (Fig. 2.7).

Let us also shortly discuss the results for the single-band model for the InAs/GaAs quantum dot. These conclusions obtained previously from the comparison of the single-band and the multiband model for GaAs/Al$_{0.3}$Ga$_{0.7}$As and GaAs/AlAs are valid for the InAs/GaAs
2.4 Conclusions and discussion

In the framework of the Burt-Foreman theory a nonsymmetrized eight-band Hamiltonian for nanostructures was extended to include a magnetic field. For zero-magnetic field case,

quantum dot as well. However, in the latter case the discrepancy between the single-band model and the nonsymmetrized multiband model, as found with increasing height of the dot, appears at higher magnetic field. This is a consequence of the lower spatial confinement in the z-direction, which leads to a relatively stronger influence of the in-plane part and band mixing caused by the magnetic field. Notice that with increasing the value of the confinement potential from $\hbar \omega_0 = 6$ meV (Fig. 2.4(a)) to $\hbar \omega_0 = 10$ meV (Fig. 2.4(b)), the influence of the magnetic field is reduced, and therefore the discrepancy between single-band and multiband models appears at higher values of the magnetic field.

For completeness, the lowest hole energy levels for sixteen different symmetries are shown in Fig. 2.8 for an InAs/GaAs quantum dot in the magnetic field, obtained by the $6 \times 6$ nonsymmetrized multiband Hamiltonian.

Inclusion of strain in the analysis leads to a splitting of the heavy hole and light hole bands (Chao and Chuang 1992, Foreman 1994) which consequently decreases band-mixing and partially suppresses the discussed effect due to the incorrect boundary conditions used in conventional $\mathbf{k} \cdot \mathbf{p}$ theory.

**Figure 2.6:** The probability density of the $1S_{-3/2}$ state, $\hbar \omega_0 = 10$ meV, calculated for InAs/GaAs using the (a) nonsymmetrized Hamiltonian and the (b) conventional Hamiltonian, for a magnetic field of $B = 40$ T. Darker region denote higher probability density.
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Figure 2.7: The probability density for the InAs/GaAs quantum well case at $k_t = 0.5$ nm$^{-1}$ using the (a) nonsymmetrized Hamiltonian, and (b) conventional Hamiltonian. Contributions of the different heavy and light hole components are also shown.

Figure 2.8: The lowest hole energy levels for sixteen different symmetries as a function of the magnetic field in an InAs/GaAs quantum dot: $\hbar\omega_0 = 10$ meV, and $h = 10$nm. The results are given for the $6 \times 6$ nonsymmetrized multiband Hamiltonian.

our Hamiltonian reduces to the nonsymmetrized one of Pokatilov et al. (2001), and if the asymmetry parameter $\chi$ is taken equal to zero, our Hamiltonian has the same form as the conventional $8 \times 8$ multiband Hamiltonian in non-zero magnetic field. We showed that the asymmetry parameter $\chi$ introduced in the BF theory plays a role not only at the boundary but through the whole nanostructure in the case of an applied magnetic field. We demonstrate that this term is essentially the same as the fourth Luttinger parameter $\kappa$ (see Eq. 2.8). We note that the antisymmetric part of the "valence-band" part of nonsymmetrized Hamiltonian is actually the valence-band Rashba coupling (Foreman 2006b). Recently, Foreman
(2006b) reported the existence of additional interface terms claiming to be of the same order of magnitude as the valence-band Rashba term incorporated in our Hamiltonian although the numerical values of these additional terms were unknown. However, Foreman (2006b) concludes that the dominant-magnetic field dependent interface term is indeed the one included in our Hamiltonian and that the additional terms would just introduce quantitative changes in our numerical results. Our numerical results presented in Sec. 2.3 indeed support this fact since the results extracted from our nonsymmetrized Hamiltonian did not show any non-physical behavior in all of the considered cases in contrast to the results obtained with the conventional approach. In the later case the non-physical results were caused by the choice of material parameters for the nanostructure (see Sec. 2.3.2).

We tested our newly developed Hamiltonian by applying it to the case of a parabolic confined cylindrical quantum dot in a perpendicular magnetic field and compared our findings to those obtained from the conventional Hamiltonian. In general, we found that the agreement between our nonsymmetrized model and the conventional models strongly depends on the difference in the structural parameters between the quantum dot and the barrier material. Comparing the energy levels as a function of magnetic field in the case of InAs/GaAs quantum dot and energy levels as a function of $k_t$ for InAs/GaAs quantum well, a similar non-physical behavior was found, and it is a direct consequence of the unappropriate treatment of the boundary. As argued in Sec. 2.1 many subtle effects may not be included, we stress here once more that no non-physical solutions are found within this BF theory in contrast to the results obtained with the conventional approach.

For completeness, let us add that treatment of the interface in the framework of the envelope function approximation, has attracted a lot of attention and that some of the drawbacks of the envelope function approach were tried to be resolved by adding additional interface terms. We give here several examples. Rodina et al. (2002) proposed general boundary conditions for the multiband effective-mass theory that required the conservation of the probability flux density normal to the interface and the self-adjointness of the multiband Hamiltonian. Ivchenko et al. (1996) modified standard multiband Hamiltonian by introducing an additional term that couples heavy-hole - light-hole even at the center of the Brillouin zone. This heavy-hole - light-hole coupling, as interpreted by Ivchenko et al. (1996), is caused by reduced translational and point symmetries of the system arising from the interface in the nanostructure. It was shown that this anisotropic effect can also be modelled within the BF theory (Foreman 1998a). Szmulowicz et al. (2004) developed a modified eight band envelope function approximation formalism incorporating this anisotropic effect of Ivchenko et al. (1996). The latter effect is not considered in conventional multiband nor in the BF envelope-function approximation. At this point we stress that non of the above mentioned approaches discussed either the validity of the envelope function approach for nanostructures or consequences of incorrect treatment of the interface on the electronic structure of a nanostructure.
Chapter 3

Numerical implementation of the three-dimensional k·p model

I tell you: One must still have chaos in one, to give birth to a dancing star.

Friedrich Nietzsche

Abstract

A full three-dimensional model for the calculation of the electronic structure of semiconductor quantum dots (QD) and molecules (QDM) grown on high index surfaces and/or in the presence of an external magnetic field is presented. The strain distribution of the dots is calculated using continuum elasticity and single-particle states are extracted from the nonsymmetrized eight-band \( k \cdot p \) theory. The model properly takes into account the effects of different substrate orientation by rotation of the coordinate system in the way that one coordinate coincides with the growth direction, whereas the effects of a tilted external magnetic field are taken into account through the Zeeman effect and employing a gauge invariant scheme based on Wilson’s formulation of lattice gauge theory. We also discuss the influence of the parameter set on the calculated electronic structure.

3.1 Choice of the model

The 3D model of the self-assembled QDs has been already implemented by several groups for quantum dots grown in [001] direction (Stier et al. 1999, Pryor 1998, Sheng and Leburton 2001b, Tomić et al. 2006). On the other hand, the 3D modeling of the electronic and optical properties of QDs grown on high index planes and/or placed in an external tilted magnetic field in the framework of eight-band \( k \cdot p \) theory have not attracted a lot of attention. Therefore, we present here our theoretical approach for modelling arbitrary 3D nanostructures (dots, molecules, or rings) grown on high index planes and/or in the presence of an external magnetic field. We address the main points in our numerical implementation.

Steps in the modelling of QDs’ electronic and optical properties and points at which experimental data are lined to our model are as follows:
(i) Size, shape, and composition profile of our model QD and/or QDM as they enter our calculations are extracted from structural characterization, e.g. by cross section scanning tunnelling microscopy or HRTEM (Bimberg et al. 1998, Shchukin et al. 2003, Bimberg and Ledentsov 2003). At this point it is important to note that strain and piezoelectric fields in and around the dots and compositional intermixing during the QD growth lead to uncertainties in the determination of their geometry and composition and therefore limits the predictive power of the theoretical models.

(ii) In the second step we calculate the strain distribution in and around the dots using continuum elasticity theory (Grundmann, Stier and Bimberg 1995, Pryor et al. 1998, Jogai 2000). The character of the strain is determined by the hydrostatic $Tr(e)$ part of the strain tensor influencing the conduction and valence band profiles and the biaxial part $B(e)$ of the strain tensor influencing the valence band mixing. The off-diagonal components of the strain tensor lead to a piezoelectric potential which as a consequence influences the distribution of the electron and hole wave function inside the dot.

(iii) Using diagonalization of the eight-band $k\cdot p$ Hamiltonian, including strain and piezoelectric potential, the confined electron and hole energy levels are obtained as a next step in our modelling. Note that $k\cdot p$ theory is a semi-empirical theory, and is parameterized by a set of experimentally obtained twelve parameters at the $\Gamma$ point. For the present case of nanostructures, for each of the materials forming the nanostructure, the values of the parameters are taken from the corresponding bulk material. At this point it is important to stress that no adjustable parameters are present in this model (for more details see Sec. 3.5).

(iv) As a final step in our model, we calculate the energy of the exciton complexes and the absorption spectra. It is at this point where we compare our findings with the experimental data and two cases can be distinguished. First, if the available photoluminescence measurements are performed on an ensemble of QDs, the information about the individual characteristics of the QDs are lost, and therefore we consider our model QD as a representative of the ensemble of QDs. Second, if the experimentalists provide us with single dot spectroscopy data, our model QD has to include the exact geometry and composition profile as the ones of the dot which the experiments were performed on. Only in that case one can expect that the measured optical spectra is correctly interpreted.

In what follows, we explain our model in detail.
3.2 Modelling of quantum dots grown on high index surfaces

Growth of QDs on high index (hkl) surfaces can be included in our model in two ways. First, one can keep the employed Hamiltonian related to the [001] coordinate system and define the structure in the [hkl] direction. Although the optical properties can be extracted from such a model as reported by Cornet et al. (2006), the underlying physical features of the QD system cannot be traced. The second approach is based on the rotation of the coordinate system and the employed Hamiltonian, in the way that the Cartesian coordinate z' coincides with the growth direction as shown in Fig. 3.1.

The general [hkl] coordinate system \((x', y', z')\) is then related to the conventional [001] coordinate system \((x, y, z)\) through the transformation matrix \(U=U(\phi, \theta)\) (Henderson and Towe 1995, Henderson and Towe 1996):

\[
U = \begin{pmatrix}
cos \phi \cos \theta & sin \phi \cos \theta & -sin \theta \\
-sin \phi & cos \phi & 0 \\
cos \phi \sin \theta & sin \phi \sin \theta & cos \theta
\end{pmatrix}, \quad (3.1)
\]

The angles \(\phi\) and \(\theta\) represent the azimuthal and polar angles, respectively, of the [hkl] direction relative to the [001] coordinate system. The azimuthal angle and the polar angle are specified in terms of indices \(h, k, \) and \(l\) and are given by:

\[
tan \phi = \frac{k}{h}, \quad tan \theta = \frac{\sqrt{h^2 + k^2}}{l}.
\quad (3.2)
\]

For the case of [11k] grown QDs and QDM, addressed in the Chapter [4] the Eq. (3.2) reduces to \(\phi = \pi/4, \theta = arctan(\sqrt{2}/k)\).

3.2.1 Strain distribution

The strain components related to the rotated coordinate system, are denoted by \(\varepsilon'_{ij}\), where \(i, j = x, y, z\), and are derived by applying the unitary transformation to the stiffness coefficient \(C_{ij}\) tensors according to the relationship:

\[
C'_{ijkl} = \sum_{mnop} U_{im}U_{jn}U_{ko}U_{lp}C_{mnop}.
\quad (3.3)
\]

Furthermore, the relation between \(\varepsilon'_{ij}\) and \(\varepsilon_{ij}\), where \(\varepsilon_{ij}\) are the strain components related to the [001] coordinate system, is given by:

\[
\varepsilon_{ij} = \sum_{kl} U_{ki}U_{lj}\varepsilon'_{kl}.
\quad (3.4)
\]
3. Numerical implementation of the three-dimensional $k \cdot p$ model

The shear strains induce a piezoelectric polarization which creates fixed charges and piezoelectric potentials. In the case of QDs and QDMs grown on high index planes the magnitude of the polarization field is proportional to the off-diagonal components of the strain tensor related to the [001] coordinate system. In order to illustrate the implementation of the model for the study of the influence of substrate orientation on the strain distribution in and around a QD, a lens shaped InAs/GaAs QD with radius $R = 7.91$nm and height $h = 4.52$nm has been chosen. Since most of the experimental work has been done on the QDs grown on [11k] substrates, where $k=1,2,3,...$, we show the calculated strain distribution in QDs grown on these high index planes. The variation of the hydrostatic component of the strain tensor with the substrate orientation along growth direction is shown in Fig. 3.2.

Thorough analysis of the influence of the substrate orientation on strain distribution in and around QDs is given in Chapter 4, Sec. 4.3.
3.2. Modelling of quantum dots grown on high index surfaces

Figure 3.2: Hydrostatic part of the strain tensor for lens shaped QD with radius $R = 7.91$ nm and height $h = 4.52$ nm along growth direction, as it varies with substrate orientation.

3.2.2 An eight-band Hamiltonian

The appropriate Hamiltonian for the [hkl] crystallographic orientation is derived by rotating the angular and crystallographic orientation in accordance to the relationship (Henderson and Towe 1995, Henderson and Towe 1996, Baraff and Gershoni 1991):

$$k_i' = \sum_j U_{ij} k_j,$$  

where $i, j = x, y, z$.

We applied here the Hamiltonian, as derived by Mlinar et al. (2005) (see previous Chapter for the details), to our QD structures. We stress that, by nature of its derivation, the nonsymmetrized Hamiltonian is valid for any growth direction, and one can simply replace the old $k$ operators by their projections in the new frame, as given above (Stavrinou and van Dalen 1997).

In order to illustrate the implementation of the model for the electronic structure calculation of QDs grown on high index planes, a lens shaped InAs/GaAs QD with radius $R = 7.91$ nm and height $h = 4.52$ nm has been chosen. As in the example given in Sec. 3.2.1 for influence of the substrate orientation on the strain distribution in and around a QD, we study QDs grown on [11k] substrates, where $k=1,2,3,...$. The variation of the electron and hole energy levels with the substrate orientation is shown in Fig. 3.3. Thorough analysis of the influence of the substrate orientation on single particle states in QDs is given in Chapter 4, Sec. 4.4.1.
3. Numerical implementation of the three-dimensional \( k \cdot p \) model

**Figure 3.3:** Electron (upper panel) and hole (lower panel) energy levels as they vary with the substrate orientation for lens shaped QD with radius \( R = 7.91\text{nm} \) and height \( h = 4.52\text{nm} \). The energy levels are given with respect to the top of the valence band of GaAs.

### 3.3 Gauge invariant inclusion of an external magnetic field

In the framework of the multi-band effective-mass theory, a magnetic field is included through Peierls substitution in the wave-vector and by adding the Zeeman energy term (Luttinger 1956, Pidgeon and Brown 1966, Weiler et al. 1978). However, it has been proven that Peierls substitution in the Hamiltonian, which subsequently is discretized on a grid, may lead to breaking of gauge invariance (Governale and Ungavelli 1998, Pryor and Flatté 2006). As a result of such a straightforward implementation, large errors in the estimates of the eigenvalues and corresponding eigenvectors may occur, producing solutions that do not reflect the actual physics, but rather ill-employed numerics. Therefore, a discretization scheme which preserves gauge invariance is required. Here, we implement a gauge invariant grid discretization based on Wilson’s formulation of lattice gauge theory (Wilson 1974).

#### 3.3.1 Gauge invariant discretization scheme

For the discretized Hamiltonian we have to impose the following condition (Wilson 1974): \( \text{The discretized Hamiltonian has to maintain all its physical properties if we perform a gauge transformation of the vector potential.} \) Or more formally: If the vector potential transforms as \( \vec{A} \rightarrow \vec{A} + \nabla \chi \), then a wave function representing a physical state of the system transforms as \( \psi(\vec{r}) \rightarrow G(\vec{r})\psi(\vec{r}) \), where \( G(\vec{r}) = \exp(-i(e/h)\chi(\vec{r})) \) (\( e \) - electron charge, \( h \) Planck constant). As a consequence, the Hamiltonian transforms as \( H \rightarrow G(\vec{r})HG^\dagger(\vec{r}) \).

Furthermore, considering a uniform discretization grid, one can define the lattice operator
3.3. Gauge invariant inclusion of an external magnetic field

\[ U_j(\xi) : \]

\[ U_j(\xi) = \exp(-i((e/\hbar)a_{latt}A_j(\xi))) , \] (3.6)

where \( j = x, y, z \), and \( a_{latt} \) is the grid spacing, and \( \xi = (l a_{latt}, m a_{latt}, n a_{latt}) \) defines the position on the grid. The lattice operator transforms as:

\[ U_j(\xi) \rightarrow G^\dagger(\xi)U_j(\xi)G(\xi) , \] (3.7)

Note that the \( U_j(\xi) \) can also be understood as the link variable between two points on the grid. Previous relations lead to the correct discretization scheme, and as an example, the discretization of the second derivative is given by:

\[ \frac{\partial^2 \psi}{\partial x^2} \rightarrow \frac{U_j^\dagger(i, j, k)\psi_{i+1,j,k} - 2\psi_{i,j,k} + U_j(i + 1, j, k)\psi_{i-1,j,k}}{a_{latt}^2} . \] (3.8)

We stress that the correct choice of the boundary conditions at the edges of the calculation box is very important. While the phase of the wave function is preserved on the inner points of the grid, as shown above, the phase should also be preserved at the boundary. Imposing Dirichlet boundary conditions ensures phase preserving at the edges of the computational box.

For completeness, let us add that Wilson’s formulation, as presented here, is oriented more to the numerical implementation, and does not explicitly exploit the periodicity of the crystal potential. In contrast, a general theory for the nonperturbative Bloch solution of Schrödinger’s equations in the presence of a constant magnetic field was recently proposed (Trelakis 2003) where an equivalent quantum system with a periodic vector potential was obtained using a single gauge transformation based on a lattice of magnetic flux lines.

3.3.2 A numerical example

From the practical point of view, for symmetric structures in an external magnetic field, where both, electrons and holes are localized in the dot, implementation of a simple discretization scheme, i.e. by simply implementing the Peierls substitution in the wave vector and setting the origin of the coordinate system at the center of symmetry of the structure, would not lead to a qualitatively different (incorrect) behavior of the eigenenergies and eigenvectors with variation of the magnetic field. Although the results so obtained are based on an inconsistent formalism, the results turn out to agree rather well with those obtained through a correct interpretation. As an example, we show the electron and hole states of the single [001] grown lens shaped QD of radius \( R = 13.56 \text{nm} \), and height \( h = 5.65 \text{nm} \) placed in an external magnetic field, where the magnetic field is varied in the range \( 0 \text{T} - 40 \text{T} \), and is directional dependent (see Fig. 3.4).
3. Numerical implementation of the three-dimensional k·p model

Figure 3.4: (a) Electron (upper panel) and hole (lower panel) energy levels as they vary with magnetic field for [001] grown lens shaped QD. Three different directions of a magnetic field are shown: Magnetic field parallel to the growth direction (z-axis)-black solid curve, magnetic field in lateral direction (x-axis)-red dashed curve, and magnetic field applied in the direction $\pi/4$ between the x and z axis-blue dotted lines. (b) The square modulus of the electron and hole ground state wave functions for $B=0\,\text{T}$, and $B_{xz}=40\,\text{T}$.

If the origin of the coordinate system is placed in center of the dot, the difference in electron and hole energies and wave functions between two implementations is negligible. The competition between the quantum confinement and effects of an external magnetic field on the electron and hole energy levels of the lens shaped QD are shown in Fig. 3.4(a). The largest influence on the electron and hole energy levels is found for a magnetic field applied in the growth directions (the weakest lateral confinement). In Fig. 3.4(b) we show the electron and hole square modulus of the wave function at $B = 0\,\text{T}$ and $B = 40\,\text{T}$, where the role of the magnetic field is reflected through the additional confinement and Zeeman splitting. Variation of the origin of the coordinate system, however, changes the eigenenergies and leads to numerically caused asymmetric wave functions. The problem also arises in the treatment of magnetic field effects of asymmetric structures, or e.g. type II QDs (where hole or electron are localized outside the dot) or QDM (see e.g. work of Mlinar et al. (2006)).
3.4 Numerical implementation

The experimental uncertainties in the geometry and composition profile of a QD or QDM enforce us the usage of numerical procedures capable for easy handling of variations of 3D input structures. Therefore, we will create our model structure on a rectangular grid which enables us to define parameters of our model at each node of the grid. In what follows we discuss the numerical implementation of the 3D model for calculation of the strain distribution, single-particle states, and Coulomb interaction. Numerical procedures employed in our model are illustrated in Fig. 3.5.

3.4.1 Continuum elasticity theory for the strain distribution

Strain is determined by global minimization of the action integral for elastic strain (Grundmann, Stier and Bimberg 1995, Pryor et al. 1998, Jogai 2000). The model takes into account the finite size of the dots and includes the anisotropy of the elastic modulus. The components of the strain tensor are calculated directly from the displacement $u$ that minimizes the action integral for the strain tensor (Pryor et al. 1998, Jogai 2000). The displacements are discretized at the nodes of the grid representing their first derivatives by finite differences.
The first derivative is averaged over the eight permutations of forward and backward differences (Pryor et al. 1998, Jogai 2000). Typically, the grid consists of \( N = 189 \times 189 \times 159 \) nodes. We choose a grid spacing equal to the lattice constant of the material that surrounds the dot. For example, for InAs/GaAs QDs, the grid spacing equals \( a_{lat}(GaAs) \), which gives a typical computational box size of \( \approx 107\text{nm} \times 107\text{nm} \times 90\text{nm} \). Furthermore, we assume that strain is completely relaxed at the edges of the computational box. Numerically, a matrix equation \( Au = b \) is solved by the conjugate gradient method (CGM), where \( A \) is a \( 3N \times 3N \) matrix, and \( b \) is a known vector of size \( 3N \).

### 3.4.2 Piezoelectricity

Effect of piezoelectricity represents the formation of a polarization field due to the presence of off-diagonal components of the strain tensor. Relation between the polarization \( P \) and strain tensor \( \varepsilon_{ij} \), in the linear regime, is given by (Grundmann, Stier and Bimberg 1995, Stier et al. 1999, Bester and Zunger 2005):

\[
P_i = \sum_{ijk} e_{ijk} \varepsilon_{jk},
\]

where \( e_{ijk} \) is tensor of third rank.

For crystals with zinc-blende structure, all components of \( e_{ij} \) are zero except \( e_{14} = e_{25} = e_{36} \).

The piezoelectric charge \( \rho_{\text{piezo}} \) is obtained from the divergence of the polarization:

\[
\rho_{\text{piezo}} = -\nabla P,
\]

The piezoelectric potential \( V_{\text{piezo}} \) is extracted from the Poisson equation, taking into account the image charge effects due to the discontinuous dielectric constants \( \varepsilon_r \) at the heterointerfaces:

\[
\rho_{\text{piezo}} = \varepsilon_0 \nabla [\varepsilon_r(\mathbf{r}) \nabla V_{\text{piezo}}(\mathbf{r})].
\]

Although these image charge effects are not significant for our embedded QDs, they become very important in the case of uncapped QDs. The Poisson equation is discretized on a larger grid than the one used for the strain calculations, we take typically \( N = 199 \times 199 \times 199 \), and is solved by CGM.

### 3.4.3 Single particle states

For the calculation of confined electron and hole states, we significantly reduce the size of our 3D grid in all three directions. The reason lies in the fact that while strain decays slowly, with a power law, the electron and hole wave functions have an exponential decay. Therefore, our grid for the electronic structure calculation is typically \( N = 69 \times 69 \times 59 \). In the case of
the QDMs containing eight dots, as will be shown later, a larger grid of $N = 79 \times 79 \times 79$ was used. Eight coupled Schrödinger equations including the strain Hamiltonian and the piezoelectric potential are discretized by the finite difference method, whereas special care is taken for the boundary nodes between the dot and the surrounding material (Hwang et al. 2004). The appropriate effective-mass parameters are taken from (Lawaetz 1971), and grid spacing are chosen such that no numerically caused spurious solutions appear (Cartoixá et al. 2003). In our calculation we use a grid spacing equal to the lattice constant of the material that surrounds the dots. Therefore, in the numerical experiment presented in next Section, the grid spacing equals $a_{\text{latt}}(GaAs)$. The finite-difference scheme is tested by checking the consistency and stability (Bilbao 2001). In order to fulfill the condition of stability, the problematic operators for discretization are: $\partial / \partial i, \partial^2 / \partial i \partial j$ where $i, j = x, y, z$, and $i \neq j$. The discrete approximation to the first derivatives, from the physical point of view, should exhibit the same spectral characteristic, i.e. it should be non-dissipative and non-dispersive (Mahesh 1998, Grogger 2006). Non-dissipative approximation is a necessary condition for numerical stability to be independent of the propagation direction.

Confined electron and hole states are found by diagonalization of the discretized Hamiltonian, with dimensions of several million by several million. The Hamiltonian is diagonalized twice, once to obtain the confined electron states and a second time for the confined hole states. In this work the numerical implementation of Geus (1999) of the Jacobi-Davidson (JD) algorithm (Sleijpen and der Vorst 1996) is used for extracting eigenvalues and eigenvectors of the matrix.

### 3.4.4 Large-scale eigenvalue problem

From the eigensolver we expect to give us a few (up to 50) inner eigenvalues and eigenvectors around the given searching energy. The JD algorithm is actually a combination of the Jacobi’s orthogonal component correction (JOCC) approach and the Davidson method. Beside the actual eigensolver loop, JD involves the solution of the linear system, i.e. the inner loop of the solver where the correction equation is solved. In the present implementation the Quasi Minimal Residual Simplified with preconditioning is used as the linear system solver. An overall acceleration of the nested loop (linear system / eigenvector) is achieved if, and only if, the JOCC preconditioner succeeds to increase the spectral gap of the desired eigenvalue so much that the entire additional effort spent for calculating the sophisticated correction vector is more than compensated. Finding the highly efficient, easy to calculate, preconditioner is a very tedious task and it is determined by the structure of the matrix.

### 3.4.5 Few particle states

As we have already discussed in Sec. 1.6.4 the presence of more than one electron or hole confined in the QD lead to the formation of few-particle states with different energies and
optical properties. Effect of the direct Coulomb interaction, exchange effects, and correlation are modelled by CI (e.g. see work by Stier et al. (1999), or Stier (2001)).

In this thesis only excitons are considered. Direct Coulomb interaction between electron and hole is calculated for QDs and QDMs grown on high index surfaces in Chapter 4 for InP/(In,Ga)P QDs and QDMs in an external magnetic field in Chapter 5, and full CI was employed for calculation of excition states of unstrained GaAs/(Al,Ga)As QDs in a magnetic field in Chapter 6.

In principle, exciton Hamiltonian \( H_X \) has following form:

\[
H_X = H_c - H_v - \frac{e^2}{4\pi\varepsilon_0\varepsilon_r(r_c,r_v)} \frac{1}{|r_c - r_v|},
\]

where \( H_c \) (\( H_v \)) is single particle Hamiltonian\(^1\) and \( \varepsilon_r \) is spatial dependent dielectric constant. If the direct Coulomb interaction is included only, energy of the exciton is given by:

\[
E_X = E_0^{(c)} - E_0^{(v)} - \frac{e^2}{4\pi\varepsilon_0} \left\langle \frac{|\psi_0^{(c)}(r)|^2|\psi_0^{(v)}(r)|^2}{\varepsilon_r(r_c,r_v)|r_c - r_v|} \right\rangle,
\]

where \( E_0^{(c)} \) (\( E_0^{(v)} \)) is electron (hole) ground state energy, and second term on right-hand side of the Eq. (3.13) is Coulomb energy \( (E_X) \) given in terms of the single particle wave functions \( \psi_0^{(c)}(r) \) and \( \psi_0^{(v)}(r) \) (electron and hole ground state wave functions, respectively). This is the Hartree approximation, neglecting the exchange and correlation effects. Numerically, Coulomb energy should be extracted from the sixfold integral, but using Green’s formula, this integral can be decomposed into a solution of the Poisson equation and 3D integration (Stier 2001).

\[
\nabla \left[ \varepsilon_r(r_c) \nabla U^{(c)}(r_c) \right] = \frac{e^2}{4\pi\varepsilon_0} \psi_0^{(c)}(r)^* \psi_0^{(c)}(r),
\]

\[
E_X = \langle \psi_0^{(v)} | U^{(c)} | \psi_0^{(v)} \rangle
\]

3.5 Choice of the parameter set

As we have already stressed the \( \mathbf{k} \cdot \mathbf{p} \) theory is a semiempirical theory, parameterized by a set of twelve parameters\(^2\) obtained from the experiment. For the case of QDs, for each of the materials forming the QD, the values of the parameters are taken from the corresponding bulk material. However, values of each of these parameters, as extracted from different

\(^1\)\( c \) and \( v \) denote that operators act only on the CB or VB part of the Hamiltonian, respectively.

\(^2\)Note that our model neglects terms arising from the inversion asymmetry of zinc-blende semiconductors, since they are assumed to be small.
3.5. Choice of the parameter set

experiments, vary in the wide range (for an overview see work of Vurgaftman et al. (2001)). For example, for GaAs first Luttinger parameter $\gamma_1$ has values in the range 6.79-7.2, or the optical matrix parameter $E_P$ in the range 25.5-29 eV. In the case of InAs bulk material, values of $E_P$ can be find in the range of 21.5-22.2 eV. Situation is even worse for parameters that enter strain part of the Hamiltonian, e.g. hydrostatic CB deformation potential $a_c$ varies from -6.3eV and -5.08eV to -18.3eV and -11.7eV for GaAs and InAs, respectively. As a consequence, Wang et al. (2000), Zunger (1998), and Fu et al. (1997) concluded that rather good agreement between the results of the $k \cdot p$ model and experiments was just a consequence of adjusting (or fitting) of the $k \cdot p$ parameter set to describe particular experiment. Actually, it was suggested that the choice of the parameter sets for semiconductors forming a nanostructure was arbitrary in $k \cdot p$ model. Under this assumption, Wang et al. (2000) extracted $k \cdot p$ parameters from the band structure of the given semiconductor calculated using empirical pseudopotential method (Wood and Zunger 1996, Wang et al. 2000) and compared different models. However, it turned out that such values of $k \cdot p$ parameters as extracted from calculated band structure, differ significantly from all previously reported values. Efficacious example is the value of Luttinger parameters for InAs as proposed by Wang et al. (2000): Luttinger parameters $\gamma_1$, $\gamma_2$, and $\gamma_3$ for InAs have values 10.6, 4.37, and 4.9, respectively which is approximately 2 times smaller than usually reported values (band parameters for III-V semiconductors can be found in publication of Vurgaftman et al. (2001)). Similar situation is with GaAs, where three Luttinger parameters, as calculated by Wang et al. (2000), have approximately 1.5 times smaller values than previously reported ones. The question that arises is how the effective mass parameters are defined within the effective-mass approach. Furthermore, it has been shown that depending on the employed parameter set $k \cdot p$ model may produce nonphysical solutions in the band gap (Foreman 2006a, Cartoixá et al. 2003). What is the origin of these solutions, and how can they be eliminated from the model? In order to clear up those issues, in what follows we explain how the parameters that enter $k \cdot p$ model are obtained, what causes the existence of nonphysical solution in the model, and give a guideline how to choose parameter sets in the case of a nanostructure.

Parameter fitting: In eight band $k \cdot p$ theory, the coupling between the lowest lying conduction band and three top most valence bands (set A) is explicitly included in the model, whereas the coupling to all other bands (set B) is reduced to zero. At the same time the effective masses in the bands of interest are renormalised to take into account the influence of the bands that are not included in the set A. Effective-mass parameters (i.e. $k \cdot p$ parameter set) are obtained experimentally by following a fitting procedure of Pidgeon and Brown (1966). Therefore, $k \cdot p$ parameter set is fitted to the experimental data, the same data (in addition to direct and indirect band gaps) the atomic potentials in the empirical pseudopotentials model are fitted to. Hence, when comparing $k \cdot p$ model with more complex models such as empirical pseudopotential model, the same experimental data have to be used for fitting procedure
3. Numerical implementation of the three-dimensional $k \cdot p$ model

and than comparison should be made. The fact that the effective-mass parameters extracted from the band structure calculated using empirical pseudopotential model differ substantially from experimental values, does not imply that such modified values should be used as the input for $k \cdot p$ model. Let us also add that improper fitting procedure in the $k \cdot p$ model may result in an unstable parameter set leading to the nonphysical behavior of the theoretical model (for more details see work of Foreman (2006b)).

Furthermore, as already mentioned in Sec. 2.2.2 in the full double-group picture the $\Gamma_7 \times \Gamma_7$ and $\Gamma_7 \times \Gamma_8$ parameters are independent of the $\Gamma_8 \times \Gamma_8$ ones (Weiler et al. 1978, Weiler 1981, Mlinar et al. 2005, Foreman 2006b) although the single-group approximation is widely accepted. The single-group approximation actually means that for $\Gamma_7 \times \Gamma_7$, $\Gamma_7 \times \Gamma_8$, and $\Gamma_8 \times \Gamma_8$ effective-mass parameters (Luttinger parameters) have same values, those of $\Gamma_8 \times \Gamma_8$ parameters. Introduced error affects primarily split-off band, and therefore not relevant for our theoretical model since split-off band is usually not of direct experimental interest.

**Spurious solutions:** Spurious solutions are defined as eigenstates of $k \cdot p$ Hamiltonian with large wave vectors which arise from small Hamiltonian matrix elements of order $k^2$ (van Alfthan et al. 2005, Yang and Chang 2005), but are also often understood as a consequence of instability with respect to small changes of the input parameters (Bastard 1988). They appear in eight band $k \cdot p$ model, but not in four or six band models. In principle, spurious solutions exist in the band gap, and actually lead to the change of the physical character of the model system from semiconducting to metallic. They depend on the experimental data fit procedures, and this uncertainty enters theoretical model (Foreman 2006a) depending on the employed (unknown in advance) parameter set. If spurious solutions appear for the given parameters set, they can be eliminated either by changing the parameters set, or by slight modification of conduction-valence band interaction i.e. by modification of the optical matrix element $E_P$.

There is also a class of spurious solutions that are consequence of the ill-employed numerics (Cartoixà et al. 2003). Namely, Cartoixà et al. (2003) demonstrated existence of spurious solution depending on the grid spacing when the finite-difference method was applied. They proposed a way to eliminate those spurious solutions through relation between the grid spacing and Luttinger parameters. However, consistent numerical approach will not lead to such artificial solutions.

**Parameter set for nanostructure:** The only difference that arises in the case of a nanostructure, as compared to bulk case is presence of nanostructure-barrier interface. and arbitrariness in the treatment of the variation of parameters through the interface (see Sec. 1.7.3)

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3It is assumed that the Kane interband parameter B is equal zero. Note that such an approximation does not influence existence of spurious solutions (Foreman 2006a).
and Chapter [2]. It is clear that each of materials forming the nanostructure have to fulfil criteria described in previous two paragraphs. The spurious solutions can appear as a consequence of the variation of the parameter $E_P$ through the interface. The easiest way to eliminate these solutions, if they appear, is to set value of $E_P$ to be constant through all nanostructure. Introduced error in that case is of order of several meV i.e. comparable to error introduced by the numerical approach.

3.6 Summary

We presented a full 3D model for electronic structure calculations of a 3D nanostructure of an arbitrary size, shape, and composition profile. Within this thesis developed model is employed for the investigation of the electronic and optical properties of QD and QDM grown on high index planes, or placed in an external magnetic field. However, this model is also applicable for studying the electronic structure of quantum ring or nanowires of the finite length. The strain distribution is calculated in the framework of continuum elasticity theory, and the single particle states are extracted form the nonsymmetrized eight-band Hamiltonian. Our model properly takes into account the effect of the substrate orientation by rotation of the coordinate system in the way that one coordinate coincides with the growth direction. The effects of a tilted external magnetic field are taken into account through the Zeeman effect and employing a gauge invariant scheme based on Wilson’s formulation of the lattice gauge theory. Calculation of few particle states in QDs is performed in the way widely accepted in QD community.

Further development: In its present form, the program does not include periodic boundary conditions relevant for the study of arrays of QDs. The modification should include the strain part of the code (for details see work of Jogai (2000)), as well as the boundary conditions for the electronic structure calculations especially important for preserving the gauge invariance (The phase of the wave function should be preserved at the edges of the computational box!). Furthermore, the present implementation allows easy variation of the used model Hamiltonian, enabling the extension to fourteen-band model (Pfeffer and Zawadzki 1990, Kurdi et al. 2003), or thirty band $k \cdot p$ model (Richard et al. 2005, Kurdi et al. 2006).
Chapter 4

Quantum dots grown on high-index surfaces

Abstract

We predict variation of electronic and optical properties of InAs/GaAs quantum dots (QDs) and quantum dot molecules (QDM) with the substrate orientation. The QDs’ and QDMs’ transition energies are obtained for high index substrates [11k], where k = 1, 2, 3 and are compared with [001]. We find that: (i) The QD size in the growth direction determines the degree of influence of the substrate orientation: the flatter the dots, the larger the difference from the reference [001] case. (ii) We predicted the variation of the transition energies of QDM as a function of substrate orientation and inter-dot distances in the eight QD molecule showing that the variation of the inter-dot distance qualitative changes the transition energy dependence on the substrate orientation. For example, for [111] grown molecules, changing the inter-dot distance varies the transition energies up to 50 meV. (iii) We showed that the size of the QD in the growth direction determines the influence of the (In,Ga)As capping layer on the optical properties of [11k] grown InAs QDs, where k = 1, 2, 3. For flat dots, increase of In concentration in the capping layer leads to a decrease of the transition energy, as is the case of [001] grown QDs, whereas for large dots an increase of the In concentration in the capping layer is followed by an increase of the transition energy up to a critical concentration of In, after which the optical transition energy starts to decrease. Furthermore, we point out the role of piezoelectricity for InAs/GaAs QDs grown on [11k], where k = 1, 2, 3, 4, 5, 7, 9 and for QDMs containing eight InAs/GaAs QDs grown on [11l], where l = 1, 2, 3.

4.1 Why are quantum dots grown on high-index surfaces important?

The strong dependence of the morphological properties of self-assembled QDs on growth conditions has been already discussed in Chapter I. The strained layer epitaxy causes the occurrence of varying strain and piezoelectric fields in and around the dots and significant compositional intermixing. As a consequence, uncertainties in the QDs’ geometry and composition appear. This turns out to be the main drawback for the various applications of QDs ranging from novel lasers (Bimberg and Ledentsov 2003, ?), Heinrichsdorff
et al. 2000), and optical amplifiers (Uskov et al. 2004) to physical representations of a quantum bit (Troiani et al. 2003, DiVincenzo 1995, Imamoglu et al. 1999) or single polarized photon sources emitting “quantum bits” (Benson et al. 2000). Furthermore, it also limits the predictive power even of detailed theoretical models of electronic and optical properties (He et al. 2005b, Stier et al. 1999, Sheng and Leburton 2001a, Mlinar et al. 2006). We note here that most experimental (Fry et al. 2000) and theoretical (He et al. 2005b, Stier et al. 1999, Sheng and Leburton 2001a, Mlinar et al. 2006) studies were performed on QDs grown on [001] substrates. The question is if the QDs’ growth on high index planes is going to bring improvements in e.g. size dispersion of QDs, or optical properties (emission at telecom wavelengths of 1.3\,\mu m or 1.55\,\mu m).

Indeed, interest has moved towards QDs grown on high index surfaces where a substantial amount of experimental work has been done (Gong et al. 2006, Lytvyn et al. 2005, Godefroo et al. 2004, Temko et al. 2003a, Temko et al. 2003b, Xu et al. 2005). It was shown that growth of QDs on high index planes has several practical advantages. For example, growth on a [113]B substrate leads to good quality QD structures with high densities and low size dispersion which are useful for QDs based lasers (Caroff et al. 2005). Namely, Caroff et al. (2005) recently reported very promising optimized growth procedure of highly dense uniformly sized InAs QDs on [113]B InP substrate with laser emission on the ground state transition at 1.59\,\mu m at room temperature. Their findings are shown in Fig. 4.1. Very recently it was found that planar and vertical ordering in QD lattices can be controlled by substrate orientation enabling 3D growth ranging from a chainlike pattern to a square-like lattice of QDs (Schmidbauer et al. 2006). From a physics point of view, different substrate orientations result in different planar projections of conduction and valence bands of the constituent crystals forming QD. As a consequence, the photoluminescence energy is expected to change with the substrate orientation. It is, therefore, of fundamental importance to understand the underlying physical features of such systems.

We are interested in finding the main differences in electronic and optical properties of high index grown QDs as compared to well investigated [001] grown QDs. For that purpose we consider the well investigated [001] grown InAs/GaAs QDs. Since experimentalists were mainly interested in [11k] grown QDs, where k = 1,2,...,9, in our theoretical investigation we...
4.2 Model quantum dot and molecule

The influence of the substrate orientation is more pronounced if the degree of lattice mismatch between the dot and the barrier is larger as it is the case for InAs/GaAs QDs, where the lattice mismatch is ~ 7.2%. We create such a model InAs/GaAs QD grown on [11k] substrates, where k=1,2,3.

The parameters used for our structure are based on Γ-point band structure parameters and are given in Table 4.1.

We tested various dot shapes and sizes. Here we present the results for two different dot shapes: lens and truncated pyramid (see Fig. 4.2), and three different sizes as given in Table 4.2. Only in Sec. 4.3.2 where we discuss the effect of piezoelectricity, we consider a lens shaped QD with radius R = 12.5nm and height h = 4.52nm, and wetting layer of thickness 2.28nm and In concentration of 20%.

Our model QDM consists of eight identical lens shaped QDs, as shown in Fig. 4.1(c) (Sec. 3.2), with radius R = 7.91nm, and height h = 4.52nm. We consider four different molecules, classified by the distance between the dots in the lateral direction $d_{\text{lat}}$ and the distance between the dots in the vertical direction $d_{\text{ver}}$ as given in Table 4.3. In the model of the QDM the wetting layer was not included.
Table 4.1: Material parameters used for the electronic structure calculations.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>[Ref.]</th>
<th>Unit</th>
<th>GaAs</th>
<th>InAs</th>
<th>In$<em>x$Ga$</em>{1-x}$As</th>
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<td>[a]</td>
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<td>$C_{11}$</td>
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<td>[e]</td>
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<td>$1/[(1 - x)/3.28 + x/9.29]$</td>
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<td>$\kappa$</td>
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<td>1.72</td>
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Table 4.2: Dimensions of lens shaped and truncated pyramidal model QDs used in our calculations

<table>
<thead>
<tr>
<th>lens</th>
<th>$R$ (nm)</th>
<th>$h$ (nm)</th>
<th>pyramid</th>
<th>$b$ (nm)</th>
<th>$h$ (nm)</th>
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</thead>
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<td>$L_1$</td>
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<td>$P_1$</td>
<td>14.7</td>
<td>3.4</td>
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<td>$L_2$</td>
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<td>3.84</td>
<td>$P_2$</td>
<td>18</td>
<td>3.6</td>
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<td>$L_3$</td>
<td>10.17</td>
<td>10.17</td>
<td>$P_3$</td>
<td>22</td>
<td>4.5</td>
</tr>
</tbody>
</table>
4.3 Strain distribution

4.3.1 Hydrostatic and biaxial components of strain tensor

In order to determine the character of the strain for dots grown on [11k] substrates, we decompose the calculated strain tensor into an isotropic part $Tr(e) = \varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz}$ and a biaxial part $B = \sqrt{(\varepsilon_{xx} - \varepsilon_{yy})^2 + (\varepsilon_{yy} - \varepsilon_{zz})^2 + (\varepsilon_{zz} - \varepsilon_{xx})^2}$, where $\varepsilon_{\alpha\beta}$ denotes the $\alpha\beta$ component of the strain tensor.

**Single quantum dot:** The strain profiles along the growth direction across the lens shaped L3 and truncated pyramidal P1 QDs are shown in Figs. 4.3(a) and (b) respectively. First, for [001] grown QDs, the isotropic strain is negative (compressive) inside the dot and tends to zero rapidly in the barrier. This isotropic strain is increased in [11k] grown flat QDs regardless of the dot shape, and the largest increase was found for [111] grown dots. However, this is no longer the case for larger dots (pyramidal, half-spherical, or conoidal QDs), where variation of the substrate orientation keeps the isotropic strain almost constant in the growth direction of the dot. Second, for [001] grown QDs the biaxial strain is non zero inside the dot and reaches a significant amount into the barrier decaying very slowly to zero. For flat dots the biaxial strain is almost constant inside the dot (Fig. 4.3(b)), while for the larger dots it has a distinct minimum in the QD (Fig. 4.3(a)). QD growth on [11k] surfaces does not modify the qualitative behavior of biaxial strain but just decreases the biaxial component regardless of the dot size and shape. Third, for all the considered dot sizes and shapes shear strains are increased for [11k] growth. As an example, we show in Fig. 4.3(c), for the L3 QD, the $e_{xz}$ strain component as it varies with substrate orientation. These shear components leads to a strongly asymmetric piezoelectric potential for [11k] grown QDs influencing the distribution of electron and hole wave function inside the dot.

**Quantum dot molecule:** Hydrostatic strain profiles of the M1 QDM are shown in Fig. 4.4. Fig. 4.4(a) shows a cross section of the hydrostatic strain profile along the growth direction.
Quantum dots grown on high-index surfaces

Figure 4.3: Isotropic $Tr$ (solid lines) and biaxial part $B$ (dashed lines) of the strain tensor for lens-shaped (a) and truncated pyramidal (b) QD. The gray surfaces in (a) and (b) represent the dots in the growth direction. (c) $\varepsilon_{xz}$ strain component of L3 QD in the plane demonstrating an increase of the shear strain with changing substrate orientation.

and along the diagonal through the lateral dots in the molecule. The hydrostatic strain profile in the plane perpendicular to the growth direction, at the bottom of the second layer of QDs is shown in Fig. 4.4b. The largest strain coupling between the dots is observed for [113] grown QDM, whereas the strain coupling between the dots for [111] grown QDM is significantly reduced. This can be understood by looking at the hydrostatic strain for a single QD (see Figs. 4.3a and b). For [111] the hydrostatic component is almost completely localized inside the dot. Only the case of the M1 molecule is shown since with increasing distance between the dots the coupling reduces.

Increasing the inter-dot distance does not change the above conclusions on the strain coupling and we stress that in all considered cases [111] grown molecules have almost negligible coupling whereas the [113] grown molecules have the largest strain coupling.
4.3. Strain distribution

Figure 4.4: Contour plot of the hydrostatic strain for M1 QDM (a) in a plane along the growth direction and the diagonal of the lateral dots in the molecule, and (b) in the plane perpendicular to the growth direction, at the bottom of the second layer of the QDs.

4.3.2 Role of Piezoelectricity

Single quantum dot: Fig. 4.5 shows the piezoelectric potential of [11k] grown QDs, where \( k = 1, 2, 3, 4, 5, 7, 9 \), whereas the minimal and maximal values of the piezoelectric potential are given in Table 4.4. The piezoelectric potential of [001] grown QDs is also shown in Fig. 4.5.
Figure 4.5: Piezoelectric potential for a QD with isosurfaces at ±31mV for different substrate orientation (blue -31mV, red +31mV).
for comparative reasons. The largest piezoelectric potential is obtained for [111] grown QDs. The question is if all the relevant components of the strain tensor are included in calculation of the piezoelectric potential. Omitting some of the components of the strain tensor may lead to the loosing the symmetry of certain crystallographic direction (e.g. three fold symmetry of the [111] direction). Going from the [111] grown QDs to [119] the value of the piezoelectric potential is lowered, and for [119] grown QDs, it approaches the case of our reference case of [001] grown QD (although asymmetric as compared to the reference case). Note that it was recently argued by Bester, Wu, Vanderbilt and Zunger (2006) that the second order piezoelectric term, usually not included in the model, is of the same order of magnitude as the linear term, and is responsible for the lowering of the piezoelectric potential. Moreover, the significantly reduced influence of the effect of piezoelectricity on the electron and hole wave functions in InAs/GaAs QD has been reported by the same authors (Bester, Zunger, Wu and Vanderbilt 2006). However, a different derivation of the second order piezoelectric term was also reported very recently (Miglioratio et al. 2006), showing a smaller contribution of the second order piezoelectricity to the total piezoelectricity effect as compared to the one proposed by Bester, Wu, Vanderbilt and Zunger (2006). Therefore, in this work, we included only the linear term as is widely accepted in the literature (Grundmann, Stier and Bimberg 1995, Stier et al. 1999, Bester and Zunger 2005).

**Quantum dot molecule:** For QDMs, the role of piezoelectricity is increased as compared to the single dot case. Fig. 4.6 shows the piezoelectric profile of the [11k] grown QDM, where k=1,2,3. The reference case of the [001] grown molecule is shown as well. The very complex piezoelectric potential is a consequence of the fact that the dots forming the molecule are very close to each other (~2nm). Increasing the distance between the dots leads
4. Quantum dots grown on high-index surfaces

4.4 Electronic and optical properties of [11k] grown nanos-structures

4.4.1 Single quantum dot

Electronic structure: Using a diagonalization of the eight-band Hamiltonian, including the strain and the piezoelectric potential, confined electron and hole energy levels are obtained numerically, which are shown in Fig. 4.7. In the upper panels of Figs. 4.7(a) and (b), the lowest lying electron energy levels of lens shaped and truncated-pyramidal QDs are shown. Variation of the electron energy levels with the substrate orientation depends on the dot size in the growth direction and is mainly influenced by the hydrostatic strain: For smaller dots the influence of the substrate orientation on the electron energy levels is larger.
The situation with the hole states is more complex. In the lower panels of Figs. 4.7(a) and (b) the hole energy levels are shown. First, one should know that, for [11k] substrate orientation, different valence bands interact even at the zone center, preventing us from classifying the hole states as a heavy-hole or light-hole. Such a simplified, but useful picture was widely used in studies of [001] grown QDs, although, strictly speaking, this heavy-hole, light-hole classification even for [001] grown QDs, is valid only at the zone center. Next, origin of the variation of hole energy levels with the substrate orientation can be traced back to the
4. Quantum dots grown on high-index surfaces

Figure 4.8: Transition energies as they vary with the substrate orientation for lens shaped (upper panel) and truncated pyramidal (lower panel) QDs (Classification of QDs by size and shape is given in Table 4.2).

The competition of several effects: (i) hydrostatic component of the strain tensor responsible for the shift of the valence bands downwards, (ii) biaxial component of the strain tensor influencing the degree of the valence bands mixing, (iii) variation of the hole effective mass with the substrate orientation, since it can significantly alter the effects of the size quantization in QD. The hole effective mass along given momentum direction can be obtained by determining the energy dispersion relation from the kinetic part of the Hamiltonian, and for the bulk case, it is largest for the [111] surface. As a result of the competition of the three above effects, we can distinguish two opposite cases as a function of QD size in the growth direction: (i) flat dots, where the variation of hole energy levels with the substrate orientation is mainly influenced by the hydrostatic strain, and (ii) large dots (see Fig. 4.7 L3 QD), where the band mixing resulting from the kinetic part of the Hamiltonian, and reduced biaxial part of the strain tensor, has a dominant influence on the variation of the hole levels with substrate orientation. We stress that the shape of the dot does not influence the above conclusions.

Transition energies: Including direct Coulomb interaction in our calculations, the variation of the transition energy with the substrate orientation and QD size and shape is shown in Fig. 4.8. For flat dots, the transition energies of [11k] grown QDs are increased with respect to those of the reference [001] grown QDs. The largest difference from the reference case exhibits [111] grown QD, whereas [113] grown QDs have transition energies that are closest to the ones of [001]. Situation with the larger dots is opposite, where the transition energies...
of [11k] grown large QDs are smaller as compared to [001] grown QDs. These findings are a direct consequence of previous discussion for single particle states. An intermediate case is illustrated by the example of P3 QD, where the transition energies of [112] and [113] grown QDs are lower than the one of the [001] grown QD, but the transition energy of the [111] grown QD is higher.

4.4.2 Quantum dot molecules

**Electronic structure:** Electron and hole confined states are extracted from the eight-band Hamiltonian including strain and piezoelectricity, and are shown in Fig. 4.9 for our model QDM. As we have already shown for the single QD case in Sec. 4.4.1, electron states are mainly influenced by hydrostatic strain, while for hole states, there is a competition between strain effects and band mixing (increased for growth on high index surfaces). For the QDM the situation is much more complex. The electron ground state is mainly determined by the hydrostatic component of the strain tensor, whereas the electron ground state of the [111] grown QDM is always the highest in energy, and the one of the [113] grown QDM is the closest energetically to the reference case of [001] grown QDM. The qualitative behavior of the electron ground state in QDM is not influenced neither by the distance between the dots in the vertical direction, $d_{ver}$, nor by the inter-dot distances in the lateral direction, $d_{lat}$. The excited electron states, however, strongly depend on the variation of the inter-dot distances in the vertical direction. In contrast, the main influence on the hole states is coming from the kinetic part of the Hamiltonian and the biaxial component of the strain tensor suppressing the influence of the hydrostatic component of the strain tensor. Increasing the inter-dot distances in the lateral direction, from M1 to M2 QDM, the lowest hole ground state energy changes from the one of [112] grown M1 QDM to [111] grown M2 QDM. The increase of the inter-dot distances in vertical direction, from M1 QDM to M3 QDM, has the same impact on the variation of the hole ground state energy with substrate orientation. However, the increase of the inter-dot distances in lateral and vertical direction, from M1 QDM to M4 QDM, the lowest energy has the hole ground state for [112] growth and is not altered by the change of inter-dot distances.

**Transition energies:** Transition energies of QDM, including direct Coulomb interaction in our calculations, as they vary with the substrate orientation are shown in Fig. 4.10. Depending on the distances between the dots forming the molecule, variation of the transition energies with the substrate orientation can be rather small, as is the case of the M3 molecule, or can qualitatively change their behavior with the variation of M1 and M2 against M4. For example, for [111] grown molecules changing the inter-dot distance varies the transition energies up to 50meV. Therefore, our predictions reported here, can be used as a guideline for potential application of such a system, in e.g. optoelectronics. Note that a recent experi-
4. Quantum dots grown on high-index surfaces

Figure 4.9: Electron (upper panels) and hole (lower panels) energy levels as they vary with the substrate orientation for QDM. The electron and hole energy levels are given with respect to the top of the valence band of GaAs. The distance between the dots in lateral and growth direction is varied as well.

4.5 Optical properties of (In,Ga)As capped InAs quantum dots

To achieve long wavelength emission, e.g. larger than 1.3µm in a QD laser diode, or alternatively user-defined detection wavelength, e.g. for quantum dot infrared photodetectors, DWELL structures were introduced (Lin et al. 2007, Gong et al. 2006). Namely, optical properties of a QD are tuned by size and chemical composition of QW layer, where the QD is embedded in.

In a widely investigated DWELL system, [001] grown InAs QD embedded in In_xGa_{1-x}As QW, variation of the In concentration in the capping layer as well as the thickness of the layer influence the hydrostatic component of the strain tensor and consequently the transition energies: Increase of the In concentration in the QW leads to a decrease of the transition energy.
4.5. Optical properties of \((\text{In},\text{Ga})\text{As capped InAs quantum dots}\)

What is happening in the case of QDs grown on \([11k]\) substrates, where \(k=1,2,3\)? How does the \((\text{In},\text{Ga})\text{As capping layer influence the optical properties of the InAs QDs grown on [11k] substrates? In this Section we answer these questions and provide a guideline for the variation of the transition energy of [11k] grown QDs, as function of the capping layers thickness and chemical profile, and for different dot composition.

4.5.1 \textbf{Transition energy vs In concentration in the capping layer}

Prior to understanding how the capping layer influences the transition energies of \([11k]\) grown InAs QDs, one has to know the effect on the transition energies of QD growth on \([11k]\) substrates \((k=1,2,3)\). As shown in previous Section, the origin of the variation of the transition energy with the substrate orientation can be traced back to the competition of several effects: (i) hydrostatic component of the strain tensor is responsible for a shift of the conduction band upwards and the valence bands downwards, (ii) biaxial component of the strain tensor influences the degree of the valence band mixing, and (iii) variation of the hole effective mass with the substrate orientation, which can significantly alter the effects of the size quantization in the QD. Actually, we have shown that the QD size in the growth direction determine which of the three above mentioned effects will be the dominant one, regardless on the dot shape. Therefore, we consider here two model lens-shaped QDs with different height: \(L_1\) QD with radius \(R=9.04\text{nm}\) and height \(h=4.52\text{nm}\), and \(L_2\) QD with radius
4. Quantum dots grown on high-index surfaces

R=9.04nm and height h=9.04nm. The thickness of the capping layer is assumed to be the same as the height of the dots, whereas the In concentration in the capping layer is varied from 0 to 30%. Our theoretical approach was described in details in Sec. 3.2, and our model QD is constructed as described in Sec. 4.2.

What will happen when both effects, QD growth on high index surfaces and capping, are present? Transition energies of L1 and L2 QD, extracted from our numerical calculations, as they vary with substrate orientation and In concentration in the capping layer, are shown in Figs. 4.11(a) and (b), respectively. One can see that for both, [001] grown L1 and [001] grown L2 QD, an increase of the In concentration in the capping layer leads to a decrease of the transition energy. On the other hand, our findings on the transition energies versus In concentration for [11k] grown QD, depend heavily on the dot size in the growth direction. Let us first discuss the case of the L1 QD.

L1 QD: The variation of the transition energy with the In concentration does not depend qualitatively on the substrate orientation, i.e. with increase of In concentration the transition energy decreases, as was the case for [001] grown QDs [black line in Fig. 4.11(a)]. This is a simple consequence of the variation of the hydrostatic component of the strain tensor with the substrate orientation and In concentration in the capping layer, as shown in Fig. 4.12(a). The hydrostatic component of the strain tensor of [11k] grown QDs reduces with the increase of the In concentration, as in the case of [001] grown QDs. The substrate orientation only determines the degree of the influence of the capping layer on the hydrostatic strain, and consequently on the transition energy. Namely, for [111] grown QDs, transition energies decrease slower with the increase of the In concentration, whereas same dependence for
4.5. Optical properties of (In,Ga)As capped InAs quantum dots

Figure 4.12: Hydrostatic component of the strain tensor as it varies with the substrate orientation and In concentration in the capping layer of L1 QD (a) and L2 QD (b).

Figure 4.13: Variation of the electron and hole energy levels of [001] (short dash) and [111] (dash-dot-dot) L2 QD with In concentration in the capping layer. Insets show the variation of the biaxial component of the strain tensor with In concentration of [001] and [111] grown L2 QD.

[113] grown QDs is similar to the reference case of [001] grown QDs.

L2 QD: For the L2 QD surprising results are obtained: with the increase of the In concentration in the capping layer the transition energies of [11k] grown L2 QD increase, exactly the opposite to the dependence for [001] grown L2 QD. After the concentration of In in the capping layer reaches some critical value, ~35%, ~20%, and ~10%, for [111], [112], and [113] grown L2 QDs, respectively, the transition energy starts to decrease with consecutive increase of In concentration in the capping layer [it starts to follow the pattern of [001]
4. Quantum dots grown on high-index surfaces

Figure 4.14: $\Delta E_{\text{trans}} = E_{\text{trans}}(x) - E_{\text{trans}}(0)$, where $x$ is In concentration in the capping layer, of L2 QDs, versus In concentration in the capping layer when (a) the dot composition is varied from pure InAs to $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}$ and $\text{In}_{0.75}\text{Ga}_{0.25}\text{As}$; (b) the thickness of the capping layer, $d$, is varied from $d=h$, where $h$ is the dot height, to $d=\frac{h}{3}$, $d=\frac{2h}{3}$, and $d=\frac{4h}{3}$.

grown L2 QD. What is the origin of such a behavior? Dependence of the hydrostatic strain on the substrate orientation and In concentration in the capping layer is shown in Fig. 4.12(b) demonstrating that the increase of the In concentration leads to a decrease of the hydrostatic strain, as in the case of [001] grown QDs. We single out the most pronounced case, [111] grown L2 QD, and show in Fig. 4.13 calculated electron and hole ground state for $x=0\%$ and $x=30\%$ In concentration in capping layer. As a comparison, we show the results for [001] grown L2 QD as well. Clearly, the variation of the hole ground state, which is most strongly influenced by the strain, with the capping leads to an increase of the transition energy. It is caused by the increase of the biaxial component of the strain, as shown in the inset of Fig. 4.13. Actually, the competition between the increased biaxial component of the strain tensor, responsible for the decrease of the valence band mixing, and the decrease of the hydrostatic strain with increase of the In concentration determine the transition energy. Note also that even for [001] grown L1 QD, biaxial strain is increased, but the hydrostatic strain has a dominant influence on the transition energy.

4.5.2 Dot composition and the thickness of the capping layer

At the end we address how the above conclusions are affected by the variation in the dot composition and the thickness of the capping layer, since those are the uncertainties to be expected in the experiment. For that purpose we modified the L2 QD composition from pure InAs to $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}$ and $\text{In}_{0.75}\text{Ga}_{0.25}\text{As}$, and thickness of the capping layer, $d$, from $d=h$, where $h$ is the L2 dot height, to $d=\frac{h}{3}$, $d=\frac{2h}{3}$, and $d=\frac{4h}{3}$. Our results are shown in Figs. 4.14(a) and (b).

An increase of the transition energy with increase of the In concentration in the cap-
4.6 Comparison with experiment

In this section we compare calculated exciton diamagnetic shift to the position of the PL peak of an ensemble of InAs/GaAs QDs grown on [001] and [113]B substrates and placed in an external magnetic field applied parallel to the growth direction (Godefroo et al. 2004).

Structural characterization (Henini et al. 1997) of these InAs QDs grown both, on [001] and [113] substrates show significant influence of the substrate orientation on the final QD size and shape. Guided by the experimental results of Henini et al. (1997) we assume our model QD to be a flat truncated pyramid ($b=25\text{nm}$, $h=2.5\text{nm}$, see Fig. 4.2(b)). Since the Zeeman splitting of the single particle orbitals is smaller than the experimental PL-broadening, the transition energy of exciton is averaged and then compared to the experimental data.

Let us first discuss the PL spectra of these InAs QDs for different growth interruptions in the absence of a magnetic field. Results of Godefroo et al. (2004) are shown in Fig. 4.15. One can see that growth interruption leads to a variation of the transition energies with substrate orientation. Therefore, based on our previous theoretical analysis (see Sec. 4.4.1) where we...
showed that QD size in the growth direction determines the degree of the influence of the substrate orientation, we vary the height of our model QD. Our results for both, QDs grown on [001] and [113] substrates, are shown in Fig. 4.16. Our calculations reproduce the experimental results of Godefroo et al. (2004) qualitatively and to a large degree also quantitatively. In general, PL-peak as a function of the magnetic field shows a parabolic behavior for low fields and linear for high fields. Typical example is shown in Fig. 4.16(a). However, for [113] grown QDs, deviation from the standard low magnetic field behavior just for certain growth conditions is reported in the experiment of Godefroo et al. (2004). Results of our calculations do not reproduce such a particular behavior. Existence of the negative diamagnetic shift, as detected in the experiment, is however less than 1meV.

### 4.7 Conclusions and discussion

In conclusion, our 3D $k\cdot p$ calculation predict the dependence of the transition energies of InAs/GaAs QDs on substrate orientation. We show that the QD size in the growth direction determines the degree of the influence of the substrate orientation on the electronic and optical properties of [11k] grown QDs, whereas the influence of the shape is of secondary importance. The flatter the dots are, the larger the difference from the reference [001] case. We predicted the variation of the transition energies of QDM as a function of substrate orientation and inter-dot distances in the 8 QD molecule showing that the variation of the inter-dot distance qualitative changes the transition energy dependence on the substrate orientation. For example, for [111] grown molecules, changing the inter-dot distance varies
4.7. Conclusions and discussion

the transition energies up to 50meV. Although composition intermixing and shape variation related to the growth conditions can quantitatively influence our results, the presented work should be understood as a guideline for the variation of the electronic and optical properties of QDs going from the well investigated [001] grown QDs to [11k] grown QDs, where k = 1,2,3. We showed the variation of the piezoelectric potential with the substrate orientation for QDs grown on [11k], where k = 1,2,3,4,5,7,9 and QDMs containing eight QDs grown on [11l], where l = 1,2,3. The [111] grown QDs and QDMs exhibit the largest piezoelectric potential. At this stage of the investigation of the electronic and optical properties of QDs and QDMs grown on [11k] substrates, we did not investigate separately contribution from the piezoelectric effect on the energy levels and consequently on the transition energies of a QD or QDM.

Furthermore, we showed that the size of the QD in the growth direction determines the influence of the (In,Ga)As capping layer on the optical properties of [11k] grown InAs QDs, where k=1,2,3. For flat dots, increase of In concentration in the capping layer leads to a decrease of the transition energy, as is the case of [001] grown QDs, whereas for large dots an increase of the In concentration in the capping layer is followed by an increase of the transition energy up to a critical concentration of In, after which the optical transition energy starts to decrease.
Chapter 5

Type II Quantum Dots

Abstract

Hole and exciton energy levels in single and vertically coupled double and triple InP/In$_x$Ga$_{1-x}$P quantum dot molecules placed in an external magnetic field are analyzed. The size of the dots and the inter-dot distance in the quantum dot molecule (QDM) determine which one of the exciton quartets will have the lowest energy and determine also which one of the individual states in the quartet will be the ground state in the presence of an external magnetic field. Competition between confinement, quantum mechanical coupling, and strain influence the exciton diamagnetic shift in double and triple QDM. We found that the available experimental data [M. Hayne et al., Phys. Rev. B 62, 10324 (2000); Appl. Phys. Lett. 79, 45 (2001)] are successfully described by one of the optically active exciton states of the lowest lying exciton quartet.

5.1 Importance of type II quantum dots

Most of the previous theoretical as well as experimental studies have been performed on type-I QDs. However, interesting features of type-II QDs started to attract more attention (Tadić, Peeters and Janssens 2002, Janssens et al. 2003). Type II InP/InGaP QDs and InAs QDs capped with GaAsSb in presence of an external magnetic field are investigated in this Chapter. In both considered cases holes are placed outside the dot and in former case are localized due to strain, and in the latter case due to Coulomb interaction with electrons in the dot. Novel features of such systems are expected. In addition, we compare our findings with the available experimental data.

This chapter is organized as follows. In Sec. 5.2 we discuss electronic and optical properties of InP/InGaP QDs and vertically coupled QDMs containing up to three QDs. Also, the influence of an external magnetic field is investigated, and our results on the diamagnetic shift are compared with recent experimental data (Hayne et al. 2000, Hayne et al. 2001). In Sec. 5.3 we discuss the influence of Ga(As,Sb) capping layer on the electronic and optical properties of such a QD system since the Sb-based capping makes these dots type-II, placing the hole in the capping layer. Influence of an external magnetic field is addressed as well.
5. Type II Quantum Dots

5.2 InP/In$_x$Ga$_{1-x}$P system in an external magnetic field

Although attracted less attention, InP/InGaP QDs turned out to exhibit a lot of interesting fundamental physics and may also be important for potential applications, e.g., for red lasers (Zundel et al. 1998). From the fundamental point of view, it is interesting that with changing geometrical properties (size and shape) of the dot, one can control the position of the hole. For example, with increasing thickness of the dot it is possible to move the heavy hole out of the disk, and to create a type II system for the heavy holes (Janssens et al. 2003). In the case of a QDM the modified strain distribution in and around the dots directly affects the effective confinement potentials for the holes. Thus the interdot distance appears now as a new controllable parameter and a fundamental question about the spatial localization of holes and excitons in such a QDM arises. Furthermore, if an external magnetic field is introduced in such a system, additional confinement due to the magnetic field is present. Since we are dealing with dots having sizes of a few tens of nanometers this additional magnetic confinement significantly influences the electronic structure of the molecule in the magnetic field range of interest ($B \leq 50T$). Recently, photoluminescence measurements on double and triple InP/InGaP QDMs in a magnetic field up to 50T, have been carried out (Hayne et al. 2000, Hayne et al. 2001). In order to qualitatively explain those magneto-photoluminescence measurements, one has to employ a model that takes into account the strain field in and around the dots as well as the multi-band mixing.

The InP/InGaP dot systems were previously studied theoretically: Single InP/InGaP QD was studied in the absence of any external fields by Pryor et al. (1997) and Tadić, Peeters and Janssens (2002), then in the presence of an external magnetic field by Janssens et al. (2003) and Tadić et al. (2005). Furthermore, Janssens et al. (2004) and Tadić and Peeters (2005) extended their approach to model InP/InGaP QDMs. Janssens et al. (2004) discussed the influence of strain on the exciton properties in triple QDM in the case a magnetic field was present. They assumed the dots to be disk-like, strain was included through an isotropic model, and excitonic properties were calculated within the single-band effective mass approximation. Nonetheless, the multiband model offers a more comprehensive description of the exciton states, with additional details not accessible by the single-band model. As a matter of fact, the heavy-hole and light-hole exciton states are not separate entities in the multiband \( k \cdot p \) model. Furthermore, the polarization sensitive selection rules are appropriately taken into account only within the multiband model. Therefore, the approach of Janssens et al. (2004) was extended by Tadić and Peeters (2004) where the continuum mechanical model was employed to model strain and the multi-band effective mass approximation was used to calculate the electronic structure.

The aim of the present Section is to provide a consistent explanation of the spatial localization of holes and excitons in InP/InGaP QDs and QDMs, to investigate the behavior of the hole and exciton energy levels in the presence of an external magnetic field and to compare...
5.2. InP/In$_{x}$Ga$_{1-x}$P system in an external magnetic field

Figure 5.1: The double (a) and triple InP/InGaP QDM (b) placed inside our simulation area (dashed lines), with radius $R$, and height $2H_z$. Dots are separated by an InGaP layer of thickness $d$. The system (a) and (b) are symmetric with respect to the $z=0$ plane. Dots are identical with radius $R$ and thickness $h$ where the Cartesian coordinates $x, y, z$ coincide with the [100], [010] and [001] crystallographic directions (c).

the calculated exciton diamagnetic shift to the position of the photoluminescence peak from Hayne et al. (2000) and Hayne et al. (2001).

5.2.1 Model quantum dot and molecule

Our models for the double- and triple-QDM consisting of self-assembled InP QD grown in the [001] direction are shown in Fig. 5.1. The dots are assumed to be flat disks, which is a good approximation for the large radius dots (Hayne et al. 2000, Hayne et al. 2001) and we take them identical.

The Cartesian coordinates $x, y, z$ are taken along the [100], [010] and [001] crystallographic directions, respectively. In our theoretical approach, the shape of the dots is assumed to be cylindrical. $R$ and $h$ are radius and height of the dots respectively (Fig. 5.1(c)), and $d$ is the distance between the dots. The stack of two and three quantum dots is symmetric with respect to the $z = 0$ plane where we assume equal distance between the dots and furthermore...
we neglect the wetting layer in our calculations. The molecules are placed inside a large cylinder of radius $R_t$ and height $2H_z$. The strain calculations as well as the electronic structure calculations are performed within this cylinder: (i) For the strain calculations we use the continuum mechanical model, where the displacements of the first-order finite elements are discretized onto a nonuniform grid. (ii) The electron, hole and exciton envelope functions are expanded within the cylinder. The parameters used for our structure are based on $\Gamma$-point band structure parameters and are given in Table 5.1.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Unit</th>
<th>InP</th>
<th>In$<em>{0.29}$Ga$</em>{0.71}$P</th>
</tr>
</thead>
<tbody>
<tr>
<td>$a$</td>
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<td>5.6532</td>
</tr>
<tr>
<td>$a_c$</td>
<td>eV</td>
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<td>$-7.5$</td>
</tr>
<tr>
<td>$a_v$</td>
<td>eV</td>
<td>0.4</td>
<td>0.40</td>
</tr>
<tr>
<td>$b$</td>
<td>eV</td>
<td>$-2.0$</td>
<td>$-1.9$</td>
</tr>
<tr>
<td>$C_{11}$</td>
<td>GPa</td>
<td>102.2</td>
<td>121.7</td>
</tr>
<tr>
<td>$C_{12}$</td>
<td>GPa</td>
<td>57.6</td>
<td>60.1</td>
</tr>
<tr>
<td>$C_{44}$</td>
<td>GPa</td>
<td>46.0</td>
<td>58.2</td>
</tr>
<tr>
<td>$\varepsilon_r$</td>
<td></td>
<td>12.61</td>
<td>12.61</td>
</tr>
<tr>
<td>$E_0$</td>
<td>eV</td>
<td>1.424</td>
<td>1.97</td>
</tr>
<tr>
<td>$\Delta_0$</td>
<td>eV</td>
<td>0.11</td>
<td>0.095</td>
</tr>
<tr>
<td>$m_e$</td>
<td>m$_0$</td>
<td>0.0795</td>
<td>0.092</td>
</tr>
<tr>
<td>$\gamma_1$</td>
<td></td>
<td>4.95</td>
<td>5.24</td>
</tr>
<tr>
<td>$\gamma_2$</td>
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<td>1.53</td>
</tr>
<tr>
<td>$\gamma_3$</td>
<td></td>
<td>2.35</td>
<td>2.21</td>
</tr>
</tbody>
</table>

Taking into account the symmetry of the molecule, the strain calculations can be restricted to the first octant. Details of the strain calculations can be found in work of Tadić, Peeters and Janssens (2002), Tadić and Peeters (2004) and Pryor et al. (1998). Piezoelectric effect is not considered in this Chapter, since it has been shown that piezoelectric effect has negligible influence (comparable to the numerical error) on the electron and hole states for disk-shaped QDs (Bester and Zunger 2005). In the present InP/InGaP quantum dot molecules only the valence sub-band mixing was explicitly included through the axial symmetric $6 \times 6$ multi-band Hamiltonian, while the mixing between conduction and valence band could be omitted because of the large band gaps of the considered constituent materials. In the axial approximation the Hamiltonian is rotationally invariant around the $z$ axis, which in our case leads to neglecting both the bulk in-plane warping of all valence bands and the

$^1$The low-lying states in the quantum dots are not significantly affected by the wetting layer
shear strains and averaging the diagonal components of the strain tensor over the azimuthal angle. As a consequence of the axial symmetry, the $z$ component of the total angular momentum can be introduced as a good quantum number (Pedersen and Chang 1996, Pedersen and Chang 1997), $F_z = f_z \hbar$. The $z$ projection of the total angular momentum can be written as $F_z = J_z + L_z$, where $J_z$ is the $z$ component of the angular momentum of the band-edge Bloch function and $L_z$ is the $z$ component of the envelope angular momentum. Furthermore, if the quantum dots are symmetric with respect to the $z = 0$ plane, which is fulfilled in our case, the parity of the wave function is a good quantum number. For example, the spinor of the even valence-band state has the form (Tadić, Peeters and Janssens 2002):

$$F_z^+ = \left[ F_{zh}^+ , F_{zh}^- , F_{so}^- , F_{so}^+ , F_{hh}^+ , F_{hh}^- \right] ,$$

(5.1)

Here $hh$ denotes the heavy hole, $lh$ the light hole, and $so$ the spin-orbit split-off band. The ± sign in the superscript of the envelope functions denotes the parity of the envelope function (+ (−) for even (odd) envelope function). The hole states for a given quantum number $f_z$, are denoted by $nX_{f_z}^{par}$ (Pedersen and Chang 1997), where $n$ is the label of the state for given $f_z$, $X$ denotes the minimum value of $|l|$ in the chosen basis set for the valence band states, and $par$ represents the parity of the state (+ (−) for even (odd) parity). The exciton energy states are computed using the exact-diagonalization approach, where the electron-hole exchange interaction is neglected. The latter interaction is not important in type II systems. The exciton wave function is expanded into pairs formed by the zone-center single-particle electron and hole wave functions,

$$\Psi_{exc}(r_e, r_h) = \sum_{s,jh} F_{s,jh}(r_e, r_h) | s \rangle | jh \rangle ,$$

(5.2)

where $s$ denotes the electron spin, $j_h$ is the magnetic quantum number of the holes and $F_{s,jh}$ is the envelope function of the exciton. The $z$ component of the total angular momentum ($F_{exc}^z = F^z_e - F^z_h$) is a good quantum number for the exciton. The parity of the exciton can be introduced as a good quantum number (Tadić and Peeters 2004). As a consequence, the exciton states can be classified by electron spin, $z$ projection of angular momentum of the exciton and the parity. The exciton states are denoted as $nX_{f_{exc}}^{par}$. Since the electron-hole exchange interaction is neglected in our calculations, each exciton level is four-fold degenerate in the absence of a magnetic field (Tadić et al. 2005), because of spin degeneracy and Kramers degeneracy of the holes. Examples of these quartets are

$$Q_1^+ = \begin{bmatrix} S_{s^1}^{+1} \mid (x,y) & S_{s^2}^{+2} \mid (x,y) & S_{s^3}^{+2} \mid (x,y) & S_{s^4}^{+2} \mid (x,y) \\ S_{s^1}^{+1} \mid (x,y) & S_{s^2}^{+2} \mid (x,y) & S_{s^3}^{+2} \mid (x,y) & S_{s^4}^{+2} \mid (x,y) \end{bmatrix} ,$$

(5.3a)

$$Q_2^- = \begin{bmatrix} S_{s^1}^{-1} \mid (x,y) & S_{s^2}^{+2} \mid (x,y) & S_{s^3}^{-1} \mid (x,y) & S_{s^4}^{+1} \mid (x,y) \\ S_{s^1}^{-1} \mid (x,y) & S_{s^2}^{+2} \mid (x,y) & S_{s^3}^{-1} \mid (x,y) & S_{s^4}^{+1} \mid (x,y) \end{bmatrix} ,$$

(5.3b)

where the optical active states in the quartet are indicated by a box and the subscripts $(x,y)$ and $z$ for in-plane and $z$ polarized light, respectively. Recombination of an exciton depends
5. Type II Quantum Dots

on the polarization sensitive selection rules for the zone center wave functions and the selection rules resulting from the conservation of the envelope angular momentum. The oscillator strength is used as a figure of merit for the recombination of an exciton (Eq. (5.4))

\[ f = \frac{2}{m_0 E_{\text{exc}}} |M_{\text{exc}}|^2, \]  

(5.4)

where \( E_{\text{exc}} \) denotes the recombination energy, \( m_0 \) is the free electron mass, and \( M_{\text{exc}} \) is the transition matrix element given by:

\[ M_{\text{exc}} = \int_{r_e, r_h} \varepsilon p_h \Psi_{\text{exc}} \, d r_e d r_h, \]  

(5.5)

where \( \varepsilon \) denotes the polarization vector, \( \Psi_{\text{exc}} \) is the wave function composed of valence band electron envelope functions and the conjugate complex of the conduction band electron envelope functions, \( p_h \) acts only on the valence band zone center periodic parts of the Bloch functions, and the integration is performed over electron and hole coordinates. Details of the calculations of the electron, hole and exciton energy spectra were already given elsewhere (Tadić, Peeters and Janssens 2002, Tadić and Peeters 2004). In the case of a magnetic field, the vector potential should be incorporated into the \( k \) operator of the Hamiltonian (Luttinger 1956, Mlinar et al. 2005). The magnetic field also gives rise to a Zeeman energy term which removes Kramers degeneracy which further leads to lifting of the degeneracy of the exciton quartets.

To show more clearly the exciton magnetic field dependence, we analyze the exciton diamagnetic shift, defined as

\[ \Delta E_{\text{exc}} = E_{\text{exc}}(B) - E_{\text{exc}}(B = 0 T). \]  

(5.6)

5.2.2 Single quantum dot

We present here the results of our calculation on the exciton states in self-assembled InP QDs embedded in In\(_{0.49}\)Ga\(_{0.51}\)P matrix, subject to magnetic fields up to 50 T. The 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.

**Hole energies:** The ground state has an angular momentum \( F_{\text{zh}} \) for which the magnitude of the envelope angular momentum of the heavy or light hole equals zero, as found in \( S_{\pm \frac{1}{2}} \) and \( S_{\pm \frac{3}{2}} \) states, respectively. For larger total angular momentum, like \( (5/2)\hbar \), the higher kinetic energy reduces the energy (the energy axis pointing upwards from the valence to the conduction band). The energy levels in 2.5 nm high InP/InGaP QD are shown in Figs. 5.2(a-c). Four highest-energy valence band states of the symmetry \( S_{\pm \frac{1}{2}}, S_{\pm \frac{3}{2}}, \) and \( P_{\pm \frac{5}{2}} \) are
5.2 \textit{InP/In}_{1-x}\text{Ga}_x\text{P system in an external magnetic field}

Figure 5.2: The four highest-energy valence-band states in InP/InGaP QD of: (a) $S_{\pm 3/2}$, (b) $S_{\pm 1/2}$, and $P_{\pm 5/2}$ symmetries. The dot radius amounts to $R = 8$ nm, and the dot height is $h = 2.5$ nm.

Figure 5.3: (a) The energy of the $1S_{\pm 1}^{-}$ exciton state as it varies with the magnetic field. The exciton energy is computed for five values of the dot height $h$, and for a dot radius $R = 8$ nm. The experimental results of Hayne et al. (2000) are also included. (b) Splitting of the $1Q_1$ and $2Q_1$ exciton quartets in the magnetic field.

displayed in the figure. Because the strain confines heavy holes inside the quantum dot and their envelope angular momentum equals zero in the $1S_{\pm 3/2}$ 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 $B$. The anticrossings are found between the lower energy valence-band states of all three symmetries.
**Comparison with experiment:** The shape and the dimensions of the fabricated quantum dots are not known accurately. But, in flat-shaped quantum dots as analyzed here, the exciton energy is most strongly affected by the change of the dot height. Hence, for a few values of dot height, we show in Fig. 5.3(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. Due to Zeeman splitting at low magnetic field ($B < 10$ T), the exciton energy decreases when the dot height increases, but at larger magnetic fields, the exciton energy increases, and the dependence might be modelled by $\sim B^2$ dependence, which resembles the result from a simple single-band effective mass approach. The best agreement between theory and experiment is found for $h = 2.55$ nm, which equals approximately 9 monolayer high InP layer. When the dot height varies slightly, the exciton energy shifts by a few meV, but the diamagnetic shift is almost not affected by this variation.

The $1S_{11}^+$ state belongs to the $Q_1$ exciton quartet (see Eq. 5.3a). This quartet is also composed of the $S_{11}^+$ bright exciton for the in-plane polarization, while the other two states appear dark. The magnetic field splits each $Q_1$ quartet into individual states, as shown in Fig. 2(b). The schemes of splitting are similar for $1Q_1$ and $2Q_1$ quartets, with the bright excitons having energies located between the dark ones.

**Oscillator strength:** The portion of the exciton wavefunction relevant to calculate the oscillator strength is determined by the condition $r_e = r_h$. In order to have a large value of the oscillator strength, the exciton should be appropriately localized. The $1S_{11}^+$ exciton state is $s$-like, localized inside the quantum dot, as shown in Fig. 3(a), where the probability density at $r_e = r_h$ is shown. On the contrary, the probability density of the $S_{01}^+$, which is optically active for $z$ polarized light, consists of both $s$-like and $p$-like clouds, as shown in Fig. 5.4(b). The localization of the exciton states affects the oscillator strength as depicted in Figs. 5.4(c) and (d) for the optically active states in the $1Q_1$ and $1Q_2$ quartets. The increase of the oscillator strength is small for the bright $Q_1$ states, while the enlargement of the $s$-like cloud in the $S_{01}^+$ state causes a sharp increase of the oscillator strength when the magnetic field exceeds 30 T. In the $S_{01}^+$ exciton state, a decrease of the $s$-like cloud is found, and therefore the oscillator strength decays towards zero as the magnetic field increases.

### 5.2.3 Double quantum dot molecules: Hole states

Here, we discuss the influence of the size (height and radius) of the dots forming the molecule, the inter-dot distance and the magnetic field on the hole energy levels of a double QDM. To simplify our model we consider two vertically aligned identical quantum dots forming a QDM. The electron wave function is symmetrically distributed over the dots, and its qualitative behavior is not significantly influenced by interdot distance and the dots size (Janssens
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Figure 5.4: The probability density of the bright exciton states at $r_e = r_h$: (a) $1S_{1/2}$ and $1S_{3/2}$ and (b) $1S_{0/2}$ and $1S_{0/2}$. The oscillator strength of exciton recombination for: (c) $1S_{1/2}$ and $1S_{3/2}$ states and (d) for $1S_{0/2}$ and $1S_{0/2}$ states. The dot radius is $R = 8$ nm, the dot height is $h = 2.55$ nm, and the magnetic field equals zero.

et al. 2004, Tadić and Peeters 2004). The hole energy levels as a function of the magnetic field for two interdot distances $d=1.5$nm and $d=5$nm are shown in Figs. 5.5 and 5.6 respectively.

Inter-dot distance of 1.5nm: Two values of the dot radius, 8nm and 10nm, and dots height, 2nm and 4nm, are considered in Fig. 5.5. Notice that the size of the dots in the molecule influences the ordering of the hole energy levels. For the dots radius $R=8$nm and height $h=2$nm the hole ground state is $1S_{1/2}^+$, but when we increase the radius to 10nm the hole ground state becomes $1S_{3/2}^+$. If the dots height is increased as well (to 4nm), the hole ground state becomes $1S_{1/2}^-$. Decreasing the dot radius to 8nm, but keeping the height unchanged ($h=4$nm) does not change the character of the hole ground
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Figure 5.5: Hole energy levels as function of the magnetic field for dot radii $R=8\text{nm}$ and heights $h=2\text{nm}$ (a), dot radii $R=10\text{nm}$ and heights $h=2\text{nm}$ (b), dot radii $R=8\text{nm}$ and heights $h=4\text{nm}$ (c), and dot radii $R=10\text{nm}$ and heights $h=4\text{nm}$ (d). Interdot distance is 1.5nm. Black arrows indicate the transition fields of the hole ground state.

state. Because the hole energy levels $1S_{\pm\frac{1}{2}}$ and $1S_{\pm\frac{3}{2}}$ and $1S_{\pm\frac{1}{2}}$ and $1S_{\pm\frac{3}{2}}$ for dot height $h=4\text{nm}$ and for both dot radius $R=8\text{nm}$ and $R=10\text{nm}$, are nearly degenerate we show in the insets of Figs. 5.5(c) and 5.5(d) the lowest hole energy levels as they vary with magnetic field. Small interdot distance prevents the hole to sit between the dots which leaves us three possibilities. The hole can sit above and below the stack, inside the dots or outside the dots in the radial direction. The location of the holes is determined by the strain field as well as by the Coulomb interaction with the electron whose wave function is symmetrically distributed over the dots. Location of the hole ground state is shown in Fig. 5.7.

Only for a dot radius $R=10\text{nm}$ and height $h=2\text{nm}$ we find that the hole ground state is localized in the dots and it is heavy-hole like, in all other considered cases the hole sits above and below the stack and it is light-hole like. Terms heavy-hole like and light hole like are
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Figure 5.6: The same as Fig. 5.5 but now for an interdot distance of 5nm.

used just to stress the dominant contribution of one of the three bands (heavy hole, light hole, spin-orbit band) to the hole ground state.

Presence of a magnetic field removes Kramers degeneracy between even and odd parity hole states (Fig. 5.5). For example, hole states $1S_{1/2}^+$ and $1S_{-1/2}^-$ are degenerate in the absence of a magnetic field, but lifting the degeneracy and splitting of those two energy levels come forward as a consequence of an applied magnetic field. Increasing the dot radius leads to a stronger influence of the magnetic field through magnetic field induced band-mixing and Zeeman splitting. Furthermore, if one follows the behavior of the hole ground state energy as a function of the magnetic field (see Fig. 5.5), one can see that the character of the hole ground state changes in the case of a dot radius $R=8$nm and height $h=2$nm. The first hole ground state transition occurs for $B=10$T where the hole ground state transits from $1S_{1/2}^+$ to $1S_{-1/2}^-$, and then at $B=45$T, the hole ground state changes to $1S_{3/2}^-$. Using the simplified but useful heavy- and light-hole like states picture, one can see that the transition at $10$T
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keeps the light-hole like ground state, but changes only the z-projection of the total angular momentum (from \(1S_{1/2}^+\) to \(1S_{1/2}^-\)), and that the hole ground state transition at \(B \approx 45T\) is a transition from the light-hole like ground state to the heavy-hole like ground state (from \(1S_{3/2}^+\) to \(1S_{3/2}^-\)). Reason for the second transition can be found in the stronger effect of the magnetic field on the heavy-hole like states. Note that the in-plane heavy-hole mass is lighter than the in-plane light-hole mass, and the lighter the in-plane mass is, the larger is the magnetic field energy. Increasing the dot radius to \(R=10\,\text{nm}\), a transition from the state \(1S_{3/2}^+\) to \(1S_{3/2}^-\) will occur for a magnetic field larger than 50T (see Fig. 5.5(b)). In contrast, when increasing the dot height no hole ground state transition is found as a function of the magnetic field.

**Inter-dot distance of 5nm:** For interdot distance \(d=5\,\text{nm}\), the two dots in the molecule are mainly coupled by the strain field since strain decays by a power law, and electron and hole wave functions decay exponentially. Hole energy levels as a function of the magnetic field are shown in Fig. 5.6. As in the case of interdot distance \(d=1.5\,\text{nm}\), quantum dots geometrical properties influence the reordering of the hole states. In the absence of a magnetic field, for a dot height of 2nm, the hole ground state is \(1S_{3/2}^-\) for both dots radius \(R=8\,\text{nm}\) and \(R=10\,\text{nm}\). But for a larger dots height of \(h=4\,\text{nm}\) the hole ground state is \(1S_{1/2}^-\) when the dots radius is \(R=8\,\text{nm}\) and \(1S_{5/2}^-\) when the dots radius is increased to \(R=10\,\text{nm}\). Compared to the case of interdot distance \(d=1.5\,\text{nm}\) where the hole could not sit between the dots, for this interdot distance \(d=5\,\text{nm}\) there is also a possibility for the hole to be localized between the dots. The corresponding hole ground state probability densities are shown in Fig. 5.8.

For the dot height 2nm, the hole ground state is heavy hole like and it is mainly located inside the dots, where for the dot radius \(R=8\,\text{nm}\), there is a contribution of the light-hole bands in between the two dots. However, for a dot height of \(h=4\,\text{nm}\), the hole sits in between the dots (see Fig. 5.8) for both dots radius \(R=8\,\text{nm}\) and \(R=10\,\text{nm}\) and it is light-hole like.

Compared to the case of interdot distance \(d=1.5\,\text{nm}\), where hole ground state transitions were observed as a function of the magnetic field for dot heights of \(h=2\,\text{nm}\), magnetic field induced hole ground state transitions for the case \(d=5\,\text{nm}\) are found for a dot height of \(h=4\,\text{nm}\). For both values of the dot radius \(R=8\,\text{nm}\) and \(R=10\,\text{nm}\), the transition occur at \(B \approx 5T\). For dot radius \(R=8\,\text{nm}\), a transition from the state \(1S_{1/2}^-\) to \(1S_{3/2}^-\) is observed, while for dot radius \(R=10\,\text{nm}\), a transition from the state \(1S_{5/2}^-\) to \(1S_{3/2}^-\) is observed - in both cases from the light-hole like ground state to the heavy-hole like ground state. One can see that whenever the hole ground state for \(B=0\,\text{T}\) is heavy hole like a magnetic field will not induce any transition of the hole ground state because of the larger shift of the heavy-hole like states in a magnetic field.

Let us also briefly discuss the kinks in the hole energy levels as a function of a magnetic field (for example, see the hole state \(1S_{3/2}^-\) in Fig. 5.5(b) or the hole state \(1S_{3/2}^-\) in Fig. 5.6(b)). We have already discussed crossings between the lowest lying hole states of either
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Figure 5.7: Double QDM: the contour plot of the hole ground state probability density for $B=0T$ and $B=40T$, for dot radii $R=8\text{nm}$ and heights $h=2\text{nm}$ (a), dot radii $R=10\text{nm}$ and heights $h=2\text{nm}$ (b), dot radii $R=8\text{nm}$ and heights $h=4\text{nm}$ (c), and dot radii $R=10\text{nm}$ and heights $h=4\text{nm}$ (d). Interdot distance is $1.5\text{nm}$. The dotted lines outline the position of the dots.

Figure 5.8: The same as Fig. 5.7 but now for interdot distance of $5\text{nm}$.

different parity or $z$-projection of total angular momentum (Figs. 5.5 and 5.6). Since the hole states are classified with respect to their $z$ component of total angular momentum, their parity
and their principal quantum number (see discussion in Sec. 5.2.1), crossings between the states of the same z component of total angular momentum and the same parity are forbidden. Thus, kinks in the hole energy level as a function of the magnetic field are a consequence of the anticrossing of that hole state with the hole state of the same parity and the same z component of total angular momentum but different principal quantum number (Pedersen and Chang 1996, Pedersen and Chang 1997, Tadić, Peeters and Janssens 2002).

5.2.4 Double quantum dot molecules: Exciton states

We investigate now the lowest exciton levels for a double QDM as a function of the magnetic field. We vary dots radius, height and inter-dot distances in the molecule.

Fixed dots’ height to 2 nm: For interdot distance d=3 nm and d=8 nm (see Fig. 5.9) we take the dot height 2 nm and vary the dot radii in the range 6-10 nm. For the case with interdot distance 3 nm (see Fig. 5.9(a)) and dot radii 6 and 8 nm the lowest exciton levels belong to the \( Q_1^- \) exciton quartet, and all individual states in that quartet are optically inactive (inset of Fig. 5.9(a) for R = 6 nm, and R = 8 nm) and the first optically active quartet is \( Q_2^- \) (Fig. 5.9(a) for R = 6 nm, and R = 8 nm).

For dot radius R = 10 nm the lowest exciton levels belong to the \( Q_1^- \) exciton quartet as well, but the lowest optically active exciton levels belong to the \( Q_1^+ \) exciton quartet (Fig. 5.9(a) and inset, for R=10nm). The exciton ground state is \( S_{1\downarrow} \). Thus, for a fixed interdot distance and dot height, the size of the dots in the lateral direction determines which exciton quartet will be the ground state. Presence of a magnetic field lifts the degeneracy and the four-fold degenerate exciton energy splits into four levels. Some of those individual states are optically active (bright exciton) and others not (dark exciton).

Exciton probability densities for interdot distance d=3 nm and for three different values of the dot radius R=6 nm, R=8 nm, R=10 nm are shown in Figs. 5.10(a), 5.10(b) and 5.10(c), respectively. Only the probability density of the lowest lying optical active exciton state belonging to the first optical active exciton quartet for B=0T, B=20T, and B=40T are plotted. For example, for the case of the dot radius R=8 nm, the exciton wave function of the state \( S_{-1\downarrow} \) of the \( Q_2^- \) quartet is shown in Fig. 5.10(b). One can see that increasing the dot radius from R=6 nm to R=8 nm tends to place the exciton more inside the dots, where for R=10 nm, the exciton is completely localized inside the dots. Presence of the magnetic field does not lead to a redistribution of the exciton wave function, but just enhances the electron-hole overlap. With increasing interdot distance to d=8 nm, changing the dot radius does not influence the exciton ground state quartet which is always \( Q_1^- \) (insets of Fig. 5.9(b)) and the first optically active quartet is \( Q_2^+ \) (Fig. 5.9(b)).

Let us also briefly discuss the optical activity of the individual states belonging to the lowest exciton quartets. For example, for the dot radius R=8 nm, the states of the \( Q_2^- \) exciton
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Figure 5.9: Splitting of the lowest lying optically active exciton quartet into four branches as a function of magnetic field for a double InP/InGaP QDM. Dot radius is varied from 6 nm to 10 nm, with fixed thickness of the dot $h=2$ nm and fixed interdot distance $d=3$ nm (a) and interdot distance $d=8$ nm (b). The insets depict the ground state quartet, but where all the individual states are optically inactive.

The exciton quartet is split in a pair of two branches: lower lying $S_{−1\downarrow}^{−}$ and $S_{0\downarrow}^{−}$ and upper lying $S_{+1\uparrow}^{+}$ and $S_{0\uparrow}^{−}$. In Fig. 5.11(a), oscillator strength for exciton recombination as a function of the magnetic field is shown. All states are optically active, $S_{+1\uparrow}^{+}$, $S_{−1\downarrow}^{−}$ for $x,y$ polarization of the light, and $S_{0\uparrow}^{−}$ and $S_{0\downarrow}^{−}$ for $z$ polarization.

However, optical activity for $x$ and $y$ polarization is an order of magnitude smaller than...
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Figure 5.10: The probability density of the lowest lying optical active exciton state belonging to the first optical active exciton quartet for \( B=0 \)T, \( B=20 \)T, and \( B=40 \)T in a double InP/InGaP QDM. The exciton wave function for dot radius \( R=6 \)nm (a), \( R=8 \)nm (b), and \( R=10 \)nm (c) are shown. Dot thickness is fixed to \( h=2 \) nm and the interdot distance to \( d=3 \) nm.

for \( z \) polarization (see Fig. 5.11(a)). Next, as an example of the optical activity for the \( Q_{1}^{+} \) quartet, oscillator strength for exciton recombination as a function of the magnetic field for the exciton \( Q_{1}^{+} \) quartet for \( d=3 \) nm and dot radius \( R=10 \) nm is calculated. Only two individual states, \( S_{11}^{\uparrow}\tilde{1} \) and \( S_{11}^{\downarrow}\tilde{1} \), of the exciton \( Q_{1}^{+} \) quartet are optically active, and only for \( x,y \) polarization. The oscillator strength as a function of the magnetic field is shown in Fig. 5.11(b) which is two orders of magnitude larger than for the \( 1Q_{1}^{+} \) state of Fig. 5.11(a). There is no optically activity for \( z \) polarization.

Influence of the variation of the dots' heights: For completeness, influence of the variation of interdot distance, dots height and radius on the double QDM magneto-exciton states
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5.2.5 Comparison with experiment

We compare our theoretical results with experimental data from Hayne et al. (2000) and Hayne et al. (2001) for triple and double quantum dot molecules. It should be pointed out that the shape and size of the dots are not known very accurately. Strain and alloy fluctuations will affect, for example, the localization of the charged particles and may lead to an asymmetric distribution of the particles along the z-direction. Transmission electron microscopy showed that dots in the triple QDM analyzed by Hayne et al. (2000) were most likely lens or disc
Figure 5.12: Splitting of the lowest lying optically active exciton quartet as a function of magnetic field in a double InP/InGaP QDM for interdot distance d=1.5nm (a) and d=5nm (b). Exciton levels for two values of the dot radius R=8nm and R=10nm, and two values of the dot height h=2nm and h=4nm are plotted.

Our theoretical findings for the exciton diamagnetic shift for triple QDM are compared with the experimental results of the position of the photoluminescence peak (squares). The results are shown in Figs. 5.13 [5.14] and 5.15 and are in good agreement with the experiment. Experimental data are always well described by the lowest lying exciton quartet, but not always with the lowest optically active individual exciton state belonging to that quartet.
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Note that the main goal of this comparison is not to fit our results to the experimental curves, but to explain the experimental results in the framework of the applied theory. Thus, we did not fit any effective mass parameter. Zundel et al. (1997) reported the growth details of these stacked QDs and it is well known that lateral dot size fluctuation and fluctuation in the total height of the dot stack (height of the dots + interdot distances) determine the PL linewidth and that the influence of fluctuations in the height of the dots reduces once they become coupled in the stack. Furthermore, the smaller the interdot distance the smaller the influence of height fluctuations (Hayne et al. 2000, Zundel et al. 1997). How do we approach the problem? We choose a QDM which we consider to be representative for the dots in the ensemble. Since the experiments on the ensemble of QDs average the finer details present in single QDs, we assume disk-shaped QDs. Next, we take into account the known fact of how the fluctuation of the dots height and lateral size depend on the interdot distances, so that for small interdot distances lateral dot size fluctuation are more important (samples D and C from (Hayne et al. 2000), and sample A from (Hayne et al. 2001)), and for larger interdot distances, when dots behave "more independent" (sample B from (Hayne et al. 2000)) fluctuation of the height of the dots is more pronounced. Thus, in order to compare our theoretical findings for the exciton diamagnetic shift for double and triple QDM to the experimental results for small interdot distances \(d=2\) nm, and \(d=4\) nm (samples D and C from (Hayne et al. 2000) for triple QDM and sample A from (Hayne et al. 2001) for double QDM) our QDM representative of the ensemble consists of three or two identical QDs of radius \(R=10\) nm, height \(h = 2\) nm, and in the case of larger interdot distance \(d = 8\) nm (sample B from (Hayne et al. 2000)) our QDM consists of three identical QDs of radius \(R=8\) nm, and height \(h = 3\) nm. The most pronounced discrepancy between our results and the experimental data is found for interdot distance \(d=4\) nm. We attribute this to a dominant influence of the strain between the dots (see discussion in previous section). For the interdot distance \(d=2\) nm as well as for interdot distance \(d=8\) nm, a good agreement is achieved.

**Triple QDM with inter-dot distance of 2 nm:** Fig. 5.13(b) shows the result for the exciton diamagnetic shift of the lowest exciton levels for a triple QDM with interdot distance \(d=2\) nm.

The behavior of the individual exciton states of the lowest quartet, in this case the \(Q_2^+\) quartet, as a function of the magnetic field is depicted in Fig. 5.13(a) where in the inset of the figure we show the oscillator strength for the states \(S_{01}^+\) and \(S_{01}^-\) of the \(Q_2^-\) quartet. All exciton states of the \(Q_2^-\) quartet are optical active. However, for x and y light polarization (\(S_{1-1}^+\) and \(S_{-1-1}^-\) of the \(Q_2^-\) quartet) the oscillator strength (not shown) is approximately 10\% of the oscillator strength for z light polarization (\(S_{01}^+\) and \(S_{01}^-\) of the \(Q_2^-\) quartet). A good agreement between the experimental data and the optical active \(S_{01}^+\) and \(S_{01}^-\) states of the \(Q_2^-\) exciton quartet are found. As diamagnetic shift carries information about the lateral confinement and the Coulomb interaction, increasing the dot radius in our model decreases the lateral confinement in our dots and increases the influence of the magnetic field. One can see from
the inset of Fig. 5.13 (a) that the oscillator strength of the $S_{0\uparrow}^-$ state tends to zero for magnetic fields above 40T, while the oscillator strength of $S_{0\downarrow}^+$ just slightly decreases with increasing magnetic field. It is easy to see from Fig. 5.13 (b) that the experimental data fits best the $S_{0\uparrow}^-$ state, but the difference between the $S_{0\uparrow}^-$ and $S_{0\downarrow}^+$ energies are small and are within the experimental accuracy of a photoluminescence experiment on an ensemble of QDM. For the completeness, the diamagnetic shift of the optically active exciton low-lying quartet were the nominal experimental parameters are used ($R=8$ nm, $h=2$ nm, $d=2$ nm) is compared to the experimental data in the inset of the Fig. 5.13 (b). Even in that case, experimental data are described by the lowest lying exciton quartet.

**Triple QDM with inter-dot distance of 4 nm:** With increasing distance between the dots we actually decrease the quantum mechanical coupling, as well as the influence of strain on the confinement in the dot. It is however known that the strain decays more slowly (with power law) than the electron and hole wavefunction (exponential decay), and thus strain will dominantly influence the carrier localization and the exciton diamagnetic shift for this interdot distance. Fig. 5.14 (a) shows the individual exciton states of the lowest quartet, in this case the $Q_{11}^+$ quartet, as a function of the magnetic field. As in the case of interdot distance $d=2$ nm, for $d=4$ nm, the radius of the dots is taken to be 10 nm and height of the dots is 2 nm. In the inset of the figure we show the oscillator strength for the states $S_{2\downarrow}^+$ and $S_{1\downarrow}^+$ of the $Q_{11}^+$ quartet. The other two exciton states $S_{2\uparrow}^-\downarrow$ and $S_{2\downarrow}^+$ of the $Q_{11}^+$ quartet are dark. The exciton diamagnetic shift in the case of the triple QDM, for interdot distances of 4 nm
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Figure 5.14: The same as Fig. 5.13 but now for interdot distance $d=4$ nm. Inset of (a) is now for the $x,y$ light polarization, and in inset of (b) calculated exciton diamagnetic shift for experimentally given size of the dots ($R=8$ nm, $h=2$ nm, $d=4$ nm) is compared with experimental data.

As in the case of interdot distance 2 nm, the experimental data are described by the optically active states belonging to the lowest lying exciton quartet. However, the experimental curve fits the exciton state $S_{1\uparrow}^{-}$ which lies above the optical active state $S_{1\downarrow}^{+}$. If one looks at the oscillator strengths in the inset of Fig. 5.14 (a), the oscillator strength of $S_{1\uparrow}^{-}$ tends to zero for values of magnetic field larger than 40 T, while the oscillator strength of $S_{1\downarrow}^{+}$ increases with magnetic field. Thus it is questionable that the experimental data are described by the state $S_{1\uparrow}^{-}$ and the agreement is probably coincidental. As was already pointed out, for the considered inter-dot distance 4 nm, the dots are mainly coupled by strain. On the other hand, the strain field is very sensitive to the dot size and shape and we assumed in our model three identical vertically aligned disk-like quantum dots. Note that the same model has been employed to study the exciton diamagnetic shift in a single InP/InGaP QD (see Sec. 5.2.2 and (Tadić et al. 2005)), and when comparing with experiment a good agreement was found for a dot height of 2.55 nm, and radius 8 nm. Thus, our assumption of disk-like quantum dots is justified. However, following the mechanism of growth of a quantum dot molecule, where a pronounced minimum in the elastic strain energy at the surface directly above the dot leads to the preferential growth of a second layer of dots vertically aligned with the first and further to the vertically aligned third layer, modifies the strain distribution in and around dots and directly affects the effective confinement potentials for carriers. Because of the difference in strain fields in the top and bottom dots, it is not likely that the quantum dots in the QDM are identical. It is therefore possible that the exciton is mainly localized in the largest quantum dot. A more precise study should include the yet unknown dot size variation in the stack of dots. As in the case of interdot distance $d=2$ nm, in the inset of Fig. 5.14 (b) we show the diamagnetic shift of the optically active exciton low-lying quartet were the nominal experi-
mental parameters are used (R=8nm, h=2nm, and d=4nm) as compared to the experimental data.

**Triple QDM with inter-dot distance of 8 nm:** With a further increase of the inter-dot distance from d=4nm to d=8nm, coupling due to strain decreases as well. The ground state exciton quartet for the triple QDM in this case is $Q^{-2}$, and it is shown in Fig. 5.15(a). In the inset of Fig. 5.15(a) the oscillator strength for the states $S_{01}^+$ and $S_{01}^-$ of the $Q_2^-$ quartet are shown. As in the case of interdot distance d=2nm, for x, y light polarization ($S_{+1}^+$ and $S_{-1}^-$ of $Q_2^-$ quartet) the oscillator strength (not shown) is approximately 10 % of the oscillator strength for z light polarization ($S_{01}^+$ and $S_{01}^-$ of $Q_2^-$ quartet). In Fig. 5.15(b), a comparison between our findings and the experimental ones (Fig. 5.15(b)) is shown for dot radius R=8nm, height h=3nm, and interdot distance d=8nm. One can see that the experimental data fit the exciton state $S_{01}^+$ as well as $S_{01}^-$. The difference between the two curves is too small to be discriminated experimentally and we may conclude that there is good agreement between our theoretical results and the experimental data. For the completeness, the diamagnetic shift of the optically active exciton low-lying quartet were the nominal experimental parameters are used (R=8nm, h=2nm, and d=8nm) is compared to the experimental data in the inset of the Fig. 5.15(b).

**Double QDM with inter-dot distance of 4 nm:** Let us also briefly discuss the results for a double QDM. The ground state exciton quartet is $Q_2^-$ and it is shown in Fig. 5.16(a) for dot radius R=10 nm, thickness h=2 nm, and interdot distance d=4 nm. Oscillator strength for the exciton recombination for z light polarization is shown in the inset of Fig. 5.16(a). The exciton diamagnetic shift calculated from our model is compared with the experimental data in Fig. 5.16(b). In the inset of the Fig. 5.16(b) we show the diamagnetic shift of the
optically active exciton low-lying quartet where nominal experimental parameters are used (R=8nm, h=2nm, and d=4nm).

From Fig. 5.16 (b) one can see that the experimental data fit the upper lying exciton S_{0}^{+}, for which the oscillator strength tends to zero for a magnetic field higher that 40T (see inset of Fig. 5.16 (a)). This is the same discrepancy as was found for the triple QDM when the interdot distance is d=4nm (see Fig. 5.14). We have no clear explanation for this interchange of exciton levels.

**5.3 GaAs_{1−x}Sb_{x} capped InAs quantum dots**

Increased interest in GaAs based optoelectronics devices emitting at wavelengths of 1.5μm and higher, enforced the search for growth protocols that enables to produce QDs that meet these requirements. There has been several reports on the achieved PL in the range 1.5-1.7μm (Maximov et al. 1999, Mowbray et al. 2006, Xin et al. 2003, Ru et al. 2003, Ripalda et al. 2005). For example, Maximov et al. (1999) grown InAs/GaAs QDs at unusually low temperatures of 325°C and reported PL emission at 1.7μm.

Also, it has been shown by Xin et al. (2003) that QDs’ growth on metamorphic buffer layers may lead to emission at 1.63μm. Furthermore, Ru et al. (2003) obtained PL at 1.48μm by growing double layers of InAs QDs capped with InGaAs. Capping of (In,Ga)As QDs with a GaAsSb layer has been proposed as an alternative way to obtain emission around 1.5μm by Liu et al. (2005), Ripalda et al. (2005), and Mowbray et al. (2006). Liu et al. (2005) observed PL up to 1.4μm using GaAsSb capping layers on InAs QDs. Ripalda et al. (2005) measured room temperature PL at 1.62μm from InAs QDs capped with 8nm thick GaAsSb capping layer with 17% Sb content is shown in Fig. 5.17. As compared to capping with GaAs, the presence of Sb during capping leads to a blue shift of the emission and a broadening of the
main PL peak. It is important to stress that a for concentration of 17% Sb in the capping layer, a type II band alignment is present placing the holes in the capping layer.

Very recently, Mowbray et al. (2006) fabricated InAs QDs capped with GaAsSb where the concentration of Sb in the 7nm thick capping layer was varied in the range 14% to 26%, and also reported that increasing Sb concentration in the capping layer not only resulted in a blue shift of the emission but also in a decrease of the QD size. The latter is a consequence of the fact that Sb acts as a surfactant leading to a decrease of the QDs’ size. PL at 10K and at room temperature (RT) of these dots is shown in Fig. 5.18 (a) and (b), respectively. As compared to dots of Ripalda et al. (2005), these dots emit at ≈1.6µm for 26% Sb at RT. We have shown in the previous section, for the case of InP/InGaP QDs, that holes are localized around the dot due to the strain fields, and that variation of geometrical properties may lead to a type II → type I transition. A similar case has been recently observed for SiGe/Si QDs, where the electron is localized in a ring around the dot that was created by strain effects (Liu et al. 2002). What is happening in the case of InAs QDs capped with GaAsSb? Does the strain force the hole to sit close (around) to the dot or it is just Coulomb interaction that attracts the hole to the dot? Consequently, how is this charge localization being reflected through experimentally
5.3. GaAs$_{1-x}$Sb$_x$ capped InAs quantum dots

Figure 5.19: (a) X-STM image of InAs QD capped with GaAs$_{1-x}$Sb$_x$, where x \approx 25\%. The capping layer is 5-6nm thick, whereas the wetting layer is 2.5nm thick with the \sim 10\% In concentration, and free of Sb. The dot is of 9-10 nm height, and of square base with base length of \sim 23nm. (b) Model of an InAs QD capped with GaAs$_{1-x}$Sb$_x$.

observed variation of the PL spectrum.

Additionally, Hayne et al. (2007) have used the same samples to perform low and high excitation PL-measurements using high magnetic fields parallel and perpendicular to the growth direction. They reported unusual behavior in the position of the PL peak with magnetic field for both considered directions of the magnetic field. For all investigated samples with concentration of Sb capping layer in the range 14-26\%, the exciton energy shift is often found to deviate from the expected parabolic and linear behavior (e.g. see Sec. 5.2.3 or Sec. 6.4.2), showing a decrease of the PL-energy with increasing magnetic field. Note that a similar behavior was observed in the case of [113] grown InAs QDs in an external magnetic field (Godefroo et al. 2004). The origin of such a behavior is unknown.

5.3.1 Theoretical model

Size, shape, and composition profile of the model QD as they enter our calculations are extracted from structural characterization by X-STM (Drouzas et al. 2006). X-STM pictures of one InAs QD capped with GaAs$_{1-x}$Sb$_x$ are shown in Fig. 5.19(a) for group III sensitive elements (In,Ga). Drouzas et al. (2006) suggested that the dots have approximately pyramidal shape, with height of 9-10nm and basis length of \approx 24nm and capping layer’ thickness of 5-6nm and a 2.5nm thick wetting layer with 10\% In concentration that is free of Sb.

Our model QD is shown in Fig. 5.19(b). We construct pyramidal QD of 9nm height and
basis length of 24nm. We introduce slight deviation from pyramidal shape, but this will not influence our conclusions given below. Namely, we change the shape of the QD from pyramidal to truncated pyramidal for fixed value of the dot height. In the next section, we show the results for truncated pyramidal InAs QD capped with Ga(As,Sb). The parameters used for our structure are based on Γ-point band structure parameters and are given in Table 4.1, whereas the parameter set used for the GaAs\(_{1-x}\)Sb\(_x\) capping layer is given in Table 5.2.

### Table 5.2: Material parameters used for the electronic structure calculations.

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<th>Parameter</th>
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<th>GaSb</th>
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[a] taken from Stier et al. (1999).  
[c] taken from Wang et al. (2004).  
[e] taken from Levinshtein et al. (1996).  

### 5.3.2 Electronic and optical properties

Introduction of the capping layer in InAs/GaAs system leads to a reduction of the hydrostatic strain, and consequently to the blue shift in the emission. What is happening when InAs dot is capped with Ga(As,Sb)? It is important to note that we adapt values of Sb concentration as reported from the experiment (Mowbray et al. 2006). In order to understand underlying...
5.3. GaAs$_{1-x}$Sb$_x$ capped InAs quantum dots

Figure 5.20: (a) Hydrostatic component of the strain tensor, in the growth direction as it varies with the Sb concentration in the capping layer. (b) Isosurface of piezoelectric charge at $\rho = 0.08e/\text{nm}^3$ (left), and isosurface of piezoelectric potential at $V_{\text{piezo}}=43\text{meV}$ (magenta $-V_{\text{piezo}}$, cyan $+V_{\text{piezo}}$).

physics of such a system, we investigate modification of the band profiles caused by the capping layer, electron and hole energy states, as well as the transition energies. We are currently performing a thorough theoretical study of this system with the intention to investigate effect of an external magnetic field and compare our findings with the magneto-PL data. Note that negative diamagnetic shift is reported in the experiment (Hayne et al. 2007).

In Fig. 5.20 we show the variation of the hydrostatic component of the strain tensor (in the growth direction) with Sb concentration in the capping layer. As compared to the case of (In,Ga)As capping layer, the reduction of the strain is significantly smaller. For completeness, we show piezoelectric charge density and piezoelectric potential in Fig. 5.20(b).

Strain modified confining potentials (see e.g. work by Pryor (1999) for definition) in QD growth direction as they vary with Sb concentration in the capping layer are shown in Fig. 5.21. One can easily see that variation of the Sb concentration in the given range (from 14% to 26%) does not influence confining potential of electrons, demonstrating that the electron will be localized in the dot. On the other hand, for holes (both light and heavy holes) is preferable to sit outside the dot. In contrast to the case of InP/InGaP QD and QDM where hole was localized near the dot due to strain, in this case hole sits near the dot only because the dot is charged with electrons. Note that peaks in the confining potential as can be seen in Fig. 5.21 and are just numerical artifacts.

Transition energy of Ga(as,Sb) capped InAs QD as it varies with the Sb concentration is shown in Fig. 5.22. Variation of the Sb concentration in the capping layer mainly influences hole states, as already discussed for the previous figure.
5. Type II Quantum Dots

Figure 5.21: (a) X-STM image of InAs QD capped with GaAs$_{1-x}$Sb$_x$, where $x \approx 25\%$. The capping layer is 5-6nm thick, whereas the wetting layer is 2.5nm thick with the $\sim 10\%$ In concentration, and free of Sb. The dot is of 9-10 nm height, and of square base with base length of $\sim 23$nm. (b) Model of an InAs QD capped with GaAs$_{1-x}$Sb$_x$.

5.4 Conclusions

Sizes of the dots in the molecule and inter-dot distance determine whether the hole in the InP/InGaP QDMs sits in the dots, in between the dots or in the matrix near the bases of the stack. We found that for small inter-dot distance ($< 3$nm) the hole settles in the matrix near the bases of the stack or in the dots, while for larger inter-dot distances the hole can be localized in between the dots or in the dots depending on the dot size. Whenever for $B=0T$ the hole ground state is heavy hole like the magnetic field is unable to induce a transition of the hole ground state.

Our theoretical findings for the exciton diamagnetic shift for single, triple, and double InP QDM are compared with the experimental results on the position of the photoluminescence peak.

For single InP/(In,Ga)P QDs we found a reasonably good agreement between theory and experiment for the magnetic field dependence of the PL energy. The magnetic field splits the exciton quartets, and the bright excitons having lower angular momenta have energies located between the dark ones. The oscillator strength for in-plane polarized light is found to be an order of magnitude larger than for $z$ polarized light, which is ascribed to the difference in the probability density of the optically active states for two the polarization directions.

For the case of QDMs, the inter-dot distance de-
5.4. Conclusions

termines whether quantum mechanical coupling (electron-hole overlap) or strain dominantly affect the exciton diamagnetic shift. For small inter-dot distance (< 3nm) quantum mechanical coupling is the dominant effect and the calculated diamagnetic shift shows good agreement with the experiment. With increasing inter-dot distance (~4nm) strain instead of quantum mechanical coupling affects dominantly the exciton diamagnetic shift, and experimental data does not fit the lowest lying optically active individual exciton state, but the higher one, which also belongs to the lowest exciton quartet. The strain field is very sensitive to the dot size and shape and we assumed in our model three identical vertically aligned disk-like quantum dots. However, following the mechanism of growth of a quantum dot molecule, where a pronounced minimum in the elastic strain energy at the surface directly above the dot leads to the preferential growth of a second layer of dots vertically aligned with the first and further to the vertically aligned third layer, modifies the strain distribution in and around dots and directly affects the effective confinement potentials for carriers. Because of the difference in strain fields in the top and bottom dots, it is not likely that the quantum dots in the QDM are identical. It is therefore possible that the exciton is mainly localized in the largest quantum dot. A more precise study should include the yet unknown dot size variation in the stack of dots. A similar discrepancy was found for the double QDM with inter-dot distance d=4nm. Furthermore, for triple QDM with inter-dot distance d=8nm the dots act "more independently" and the calculated exciton diamagnetic shift shows good agreement with the experiment data.

In the case of Ga(As,Sb) capped InAs QDs, electron is localized inside the dot, whereas hole is siting in the capping layer. Electrons in the dots attract hole to sit near the dot, but not strain, as was the case of InP/InGaP QDs. Variation of the transition energy with the Sb concentration in the capping layer is determined by the variation of the hole energy levels.
Chapter 6

Unstrained GaAs/Al\textsubscript{x}Ga\textsubscript{1−x}As Quantum Dots

Abstract

Magneto-optics of unstrained GaAs/Al\textsubscript{x}Ga\textsubscript{1−x}As quantum dots are investigated theoretically in the presence of an external magnetic field. Single particle states, exciton binding energies and the exciton diamagnetic shift are calculated with a confinement potential based on atomically resolved STM pictures. The degree of interface intermixing is treated as a variable. The electronic structure of the dot in the presence of a magnetic field is calculated using eight-band \textbf{k·p} theory including a magnetic field. We find that varying interface roughness sensitively affects the interband but hardly the intraband energies. For magnetic fields applied both in growth direction and perpendicular to it (for \textit{B} ≤ 50T), we find good agreement between our predicted exciton diamagnetic shift and recent experimental magneto-photoluminescence data (N. Schildermans et al. Phys. Rev. B 72, 115312 (2005)). The inherent coupling of valence- and conduction band taken into account in the eight-band \textbf{k·p} model explains well the observed experimental results.

6.1 Advantages of GaAs/Al\textsubscript{x}Ga\textsubscript{1−x}As system

The morphological properties of QDs grown by strained layer epitaxy strongly depend on the details of the growth conditions (Stoddart et al. 1998, Shchukin et al. 2003, Shchukin and Bimberg 1999, Maes et al. 2002). The occurrence of varying strain and piezoelectric fields in and around the dots and significant compositional intermixing lead to uncertainties in the determination of their geometry and composition e.g. by HRTEM (Bimberg et al. 1998, Shchukin et al. 2003, Bimberg and Ledentsov 2003). Thus the predictive power even of detailed theoretical models of electronic and optical properties (Grundmann, Stier and Bimberg 1995, Stier et al. 1999, He et al. 2005b, Narvaez et al. 2005, Mlinar et al. 2006, Mlinar and Peeters 2006a) is limited by this uncertainty.

Recently, unstrained GaAs/Al\textsubscript{x}Ga\textsubscript{1−x}As QDs were fabricated through an ingenious multistep approach based on a combination of hierarchical self-assembly and \textit{in situ} etching (Rastelli et al. 2004). Due to the nonexistence of strain and reduced uncertainties in the size and shape, such QDs represent an ideal test case for electronic structure models.

Let us shortly discuss the growth procedure of such unstrained GaAs QDs as introduced by Rastelli et al. (2004). First, a template of SK-grown InAs/GaAs (001) islands was created.
Then, the islands were "converted" into nanoholes on a GaAs surface by GaAs overgrowth followed by in situ etching. Self-assembled nanoholes are then transferred to an AlGaAs surface, filled with GaAs, and finally overgrown with AlGaAs. The AFM images of GaAs nanoholes prior and after overgrowth, as reported by Rastelli et al. (2004), are shown in Fig. 6.1 (a) and (b) respectively. It is expected that the QDs’ morphology is given by the shape of AlGaAs nanoholes whereas the question of the Al-Ga intermixing at the interface was not addressed in the work of Rastelli et al. (2004).

It was also shown that the average hole depth decreases as the AlGaAs overgrowth proceeds and a statistical analysis on the hole morphology as a function of thickness D is given.
6.1 Advantages of GaAs/Al$_x$Ga$_{1-x}$As system

Figure 6.2: (a) Filtered XSTM image of a GaAs/AlGaAs QD at negative sample bias of 2.0 V. Filtering was applied in order to pronounce the differences between GaAs and AlGaAs. Arrows mark the GaAs/AlGaAs/GaAs structure. The AlGaAs/GaAs interfaces are marked with white lines, and the reversed truncated pyramidal shape of the GaAs/AlGaAs QD is indicated by a black dashed line. (b) Filtered XSTM image of a GaAs/AlGaAs QD at positive sample bias of +2.3 V. The AlGaAs/GaAs interfaces are marked with white lines, and the QD shape is indicated by the dashed black line. The XSTM image is filtered in order to emphasize the AlGaAs layer (taken from the publication of Lenz et al. (2006)).

in Fig. 6.1 (c). Furthermore, Rastelli et al. (2004) reported the shrinking of the nanoholes in the [1 10] direction while the size of nanoholes in the [110] direction remained approximately unchanged, as shown in Fig. 6.1 (d). This effect was attributed to the anisotropy in surface diffusivity of Al and Ga, since the diffusivity along the [1 10] direction is higher than along the [110], and nanoholes tend to fill first in the former direction.

Also, PL of an ensemble of such QDs revealed an appreciably smaller inhomogeneous broadening, in the range between 8 to 15 meV depending on the growth conditions, indicating a good size homogeneity. The QDs have a typical lateral size of 35-65nm and a thickness of $\sim$6nm. The question is if one can assume that the shape of the dot remains unchanged after the nanoholes were filled with GaAs as suggested by Rastelli et al. (2004). Recent cross section scanning tunnelling microscopy (X-STM) experiments demonstrate that the interfaces between GaAs and AlGaAs layers are not abrupt (Lenz et al. 2006) as previously observed for GaAs quantum wells (Hermann et al. 1991). Namely, Lenz et al. (2006) showed that Al-Ga intermixing at the interface occurs during the growth leading to changes of the aluminium/gallium content at the heterojunctions, as shown in Fig. 6.2. In Fig. 6.2 (a), a X-STM image of AlGaAs/GaAs/ALGaAs structure taken for negative bias are shown, whereas
taking the negative bias implies that the group V atoms are predominantly imaged (see Chapter 1 for more details). The XSTM image of another GaAs QD is shown in Fig. 6.2(b) for positive bias imaging the group III atoms. In Figs. 6.2(a) and (b) the interfaces of the AlGaAs/GaAs nanostructure (white lines) and the GaAs/AlGaAs QD (black dashed lines) are marked. Influence of such an intermixing on the electronic and optical properties was not addressed.

Furthermore, PL measurements as well as single dot spectra were performed on these GaAs/AlGaAs QDs and are compared to the theoretical findings extracted from an eight-band $k \cdot p$ model showing good agreement (Rastelli et al. 2004). The zero-field PL measurements were subsequently extended to magnetic field up to 50T by Schildermans et al. (2005) and a single-band model modified to include conduction band non-parabolicity (Ekenberg 1989) was implemented to describe the magnetic field dependence (Sidor et al. 2006). However, a consistent theoretical study for the interpretation of the non-zero magnetic field experimental data has not been reported.

The aim of this chapter is to answer the following questions: (i) How does the interface smoothing influence the optical properties of GaAs/Al$_x$Ga$_{1-x}$As QDs? (ii) How do the electronic properties of GaAs QD depend on the direction of an applied magnetic field? (iii) Can magneto-photoluminescence data of Schildermans et al. (2005) be described in the framework of an eight-band $k \cdot p$ model and consequently is it possible to estimate conduction band non-parabolicity of these dots?

### 6.2 Model quantum dot

The unique fabrication process used by Rastelli et al. (2004) made it possible to extract the QD structure with great detail: Prior to the final step of QD deposition the surface profile (see Fig. 6.3(a), red dotted line) has been scanned using STM while preserving the ultra high vacuum conditions during epitaxy. The shape of the resulting QD is determined by this pattern and taken as the origin of our primary model QD. Next, a series of model QDs is derived from the primary one based on a systematic variation of Fickian diffusion induced interface roughness (see Fig. 6.4). The parameter N describes the degree of intermixing (N=0: no intermixing; N=9: strong intermixing). The parameter N is actually the number of smoothing steps at the interface. One smoothing step at one point of the grid describing the structure is taken as average value of the material parameters of the structure on the neighboring points of the grid.

**Electronic structure:** The single particle energies of our model QDs are extracted from an eight-band $k \cdot p$ Hamiltonian (Stier et al. 1999), where the magnetic field is incorporated through standard Peierls substitution (Luttinger 1956) in the wave-vector and Zeeman energy term (Mlinar et al. 2005). The parameters used for our structure are based on $\Gamma$-point band
Figure 6.3: (a) Schematic sample structure of unstrained GaAs/AlGaAs quantum dots. The structure of the implemented model QD (c) is derived from STM measurements (b) taken from the publication of Rastelli et al. (2004). The red dotted line in (a) indicates the position where the STM picture is taken from.

structure parameters and are given in Table 6.1. Note that parameters used for GaAs in this Chapter differ from those used in Chapter 4 also for GaAs (see Table 4.1). As we have already discussed in Sec. 3.5 choice of the parameters set may lead to the existence of spurious solutions as well as can cause numerical difficulties (e.g. it can influence the convergency of an eigenvalue solver). Because of the relatively large size of the structure
unstrained GaAs/Al$_x$Ga$_{1-x}$As Quantum Dots (see Fig. 6.3), resulting matrix that has to be diagonalized is extremely large, therefore, the in order to improve the convergency varied slightly the Luttinger parameters $\gamma_1$ and $\gamma_3$ and parameter $E_P$ (note that certain values of this parameter may lead to the existence of spurious solutions, see also Sec. 3.5). The obtained results are not affected by this variation (variation in the energy levels is much less than the error of the numerical approach.)

The excitonic states are calculated using the configuration interaction method. The two particle exciton Hamiltonian is expanded into a basis of antisymmetrized products of single particle wavefunctions, thus accounting for direct Coulomb interaction and exchange. Correlation effects are accounted for by expanding the basis to include excited-state-configurations (Rodt et al. 2005). The Coulomb and exchange matrix elements needed for the CI calculation are computed numerically from 6 electron and 6 hole single particle orbitals and are screened using different dielectric constants for GaAs and AlGaAs. Absorption is calculated by Fermi’s golden rule applied to CI states. It is important to stress that no adjustable parameters are present in this model.

Table 6.1: Material parameters used for the electronic structure calculations.

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<td>$1/(1 - x)/2.91 + x/1.57$</td>
</tr>
<tr>
<td>$\varepsilon_s$</td>
<td>[a]</td>
<td></td>
<td>13.18</td>
<td>10.06</td>
<td>linear</td>
</tr>
<tr>
<td>$\kappa$</td>
<td>[c]</td>
<td></td>
<td>1.28</td>
<td>0.12</td>
<td>linear</td>
</tr>
</tbody>
</table>


The experiments which we are referring to throughout this work were performed on an ensemble of QDs. When we compare our findings with the experimental results, we consider our model QD as representative for the ensemble of QDs. The latter is motivated by the unusual narrow photoluminescence broadening which points at an extremely homogeneous QD-ensemble.
6.3 Influence of interface intermixing on the electronic and optical properties

In a simplified picture, the carrier confinement in unstrained GaAs/Al_{1-x}Ga_xAs QDs is only a function of the dot size and shape, where the conduction and valence band offsets are taken from those of the bulk materials. However during the growth process Al-Ga intermixing at the interface is likely to occur leading to changes of the aluminium/gallium content at the heterojunctions. Recently obtained cross-section STM images of GaAs/Al_{1-x}Ga_xAs QDs support this assumption, suggesting that interfaces of GaAs and AlGaAs layers are not abrupt (Lenz et al. 2006, Hermann et al. 1991). Here, we investigate the consequences of such non-abrupt interfaces on the optical properties by treating the degree of interface intermixing as a variable.

In Fig. 6.4 a vertical scan through the center of the model structures is shown for different values of N, a quantity which parameterizes the degree of interface intermixing. The case N = 0 corresponds to the ideal case of abrupt interfaces and N = 9 to the case of strong interface intermixing. The corresponding local conduction and valence bands are shown in Fig. 6.5(a) and (c), and since they are extracted from the 3D rectangular grid they show step-like profile. The interface smoothing leads to the increase of the QDs’ effective size and to the decrease of the average Al content. As a consequence, the confinement of the electrons and holes is determined by the dominant effect of two present with increasing the N, first one, the effect of the increase of the QDs’ size or the second one the effect of the decrease of the average Al content.

Single-particle energies: The variation of the first three electron and hole energy levels with the number of intermixing steps is shown in Fig. 6.6. Two main results can be derived from this figure: First, Al-Ga intermixing leads to an increase of electron and hole energies and therefore to larger interband transition energies. Second, the electron intraband energies are almost unaffected by the interface roughening. The situation is similar for the hole intraband energies, except for N larger than 7, where the hole intraband energies slightly differ from those for smaller N. This behavior can be attributed to the increased confinement which leads to an enhanced valence band mixing. Fig. 6.5(b) and (d) show the lateral and vertical extent of the electron and hole groundstate for the abrupt interface (N=0) and for the largest considered degree of intermixing (N=9): In both cases the wave functions are stronger localized for N=9. It means that the although the effective size of the quantum dot increases with increasing N, the decrease of the Al content with N is the dominant effect and it leads to the increased confinement for the electrons and holes. This result is different from the one recently reported by Seguin et al. (2006) for small InAs/GaAs QDs, where it was found that both carrier types tend to increase their wave function extent upon annealing. The main reason for such a different behavior of electron/hole wave function extent as a function
of annealing can be found in the ratio between average QD size and electron/hole Bohr radius which is larger than one here and smaller than one in the case reported by Seguin et al. (2006).

**Excitonic properties:** As can be seen from Fig. 6.7 a change in the sharpness of the interfaces sensitively affects the exciton energy. The larger $N$ is, the larger is the exciton energy. We have already seen that the single particle energies increase (Fig. 6.6) for higher values of $N$. The increase of electron and hole localization for larger interface intermixing, shown in Figs. 6.5(b) and (d), is also reflected by an increase of the exciton binding energy as illustrated in the inset of Fig. 6.7. Therefore, the increase of exciton energy upon interface intermixing is a result of the increased single-particle energies resulting in a stronger wave-function localization. Fig. 6.8 finally shows the calculated excitonic absorption spectra as a function of $N$.

A comparison to measured single-QD PL spectra (Rastelli et al. 2004) yields a very good agreement for an intermixing value of $N=3$. With increasing $N$ a blue shift of the whole spectrum occurs (37 meV between $N=0$ and $N=9$) whereas the distances between the peaks remain approximately the same.
6.3. Influence of interface intermixing on the electronic and optical properties

Figure 6.5: Lateral (left) and vertical (right) scan through the local conduction (a) and valence (c) band edges of the model QD for \( N=0 \) (nonintermixed dashed) and \( N=9 \) (strongly intermixed). The respective square modulus of electron and hole wavefunctions are shown in (b) and (d), respectively. The lateral scans through the confinement potential and the probability density are taken at a vertical position 1nm below the GaAs Qwell (average of the z-component of the wave functions barycenter). Since the potential width is determined at this upper z-position it appears large compared to the lateral wave function extent. The step-like lateral confinement potential after annealing (a and c) originates from a combination of the terrace like QD structure and the applied smoothing procedure.
6.4 Effect of an applied magnetic field

6.4.1 The role of the magnetic field direction

Single-particle energies: An external magnetic field introduces an additional confinement in the plane perpendicular to the direction of the applied field and lifts the Kramers degeneracy. Its impact on the electronic properties depends on the strength of the already existing QD-confinement. It is large for small confinement and small for large confinement. Since our QDs are very flat the confinement is strong in growth direction ($e_z$) and small in lateral direction. Therefore we expect a large effect if the B-field is applied parallel to $e_z$ and a smaller one if applied perpendicular to $e_z$. The results of the corresponding calculations are shown in Figs. 6.9 and 6.10.
6.4. Effect of an applied magnetic field

For the case of a lateral magnetic field, the strong QD-confinement in the growth direction limits the influence of the magnetic field. In contrast, for the case of a magnetic field applied in the growth direction, electron and hole energy levels are strongly influenced by the magnetic field and a magnetic field induced band mixing of the hole energy levels is observed. This is a direct consequence of the relatively smaller lateral confinement in the dot (e.g. magnetic length for B=10T is about 8nm, while the base-length of the dot is about 35nm). Electron and hole wave functions for the two different magnetic field orientations are also shown in Fig. 6.10. One can clearly see the impact of the additional lateral confinement for \( B \parallel e_z \) on the lateral electron and hole wave function extent, which does not occur for \( B \parallel e_x \). An exchange of the character of the hole wave functions for magnetic field of B=20T applied in the growth direction is clearly observed. For \( B \approx 17T \) the \( h_0 \) energy level anticross with the \( h_1 \). As a consequence the hole wave function that corresponds to \( h_0 \) energy level before the anticrossing is \( h_1 \) wave function like after anticrossing (see hole wave functions in Fig. 6.10 whereas the wave function that corresponds to \( h_1 \) energy level before anticrossing is \( h_0 \)-wave function-like after the anticrossing.

Role of the interface roughness: In Sec. 6.3 we demonstrated that the interface intermixing increases the localization of the electrons and holes. As a consequence, it leads to the reduced influence of the magnetic field on the electrons and holes. It is therefore clear that the increase of the parameter N leads to an even smaller dependence of the electron and
6. Unstrained GaAs/Al\textsubscript{x}Ga\textsubscript{1−x}As Quantum Dots

Figure 6.8: Exciton absorption spectra as a function of N. A Blue shift in the transition energy with increasing interface intermixing is observed. The vertical red (dark grey) dotted line indicates the position of the measured PL peak.

hole energy levels with a magnetic field applied in the direction perpendicular to the growth direction, while for the magnetic field applied in the growth direction variation of the parameter N does not lead to the significant changes in the behavior of the electron and hole energy levels with the magnetic field. These conclusions are illustrated in Table 6.2 for both directions of the applied magnetic field.

The exciton-binding energy: The magnetic field dependence of the exciton-binding energy is analyzed by considering the quantity $\Delta E_{X−\text{bind}}$ defined as $\Delta E_{X−\text{bind}} = E_{X−\text{bind}}(B) - E_{X−\text{bind}}(B=0)$, with $E_{X−\text{bind}}(B)$ being the exciton binding energy at magnetic field B. In Fig. 6.11 $\Delta E_{X−\text{bind}}$ is shown as a function of the B-field and its orientation and the degree of interface intermixing. Since the magnetic field further enhances the wave function localization it is of no surprise that the exciton binding energies $E_{X−\text{bind}}$ increase upon an increase of the magnetic field (see Fig. 6.11(a)). This effect is much larger for $B \parallel e_z$ than for $B \parallel e_x$, since in the former case the increase of the wave function localization is much stronger than in the latter. As the interface smoothing leads to an additional carrier localization in our case $\Delta E_{X−\text{bind}}$ becomes smaller as a function of N (see Fig. 6.11(b)).
6.4. Effect of an applied magnetic field

Figure 6.9: Electron and hole energy levels (with respect to the GaAs conduction band) as a function of the magnetic field parallel to the growth direction (black lines) and perpendicular to the growth direction (light gray lines) are shown. The case of an ideal sharp interface (N=0) is shown.

The effect of smoothing of the interface on the excitonic absorption spectra for B=10T for magnetic field applied in the growth direction and in plane are given in Figs. 6.12(a) and 6.12(b) respectively. The exciton absorption spectra are richer for the case of magnetic field applied in the growth direction. The reason for this can be found in the larger spin splitting and the arising additional transitions in a magnetic field whose effect on the electronic structure is much larger than in the case of magnetic field applied in plane.

6.4.2 Comparison with experiment

The exciton absorption spectrum of our model QD for N=3 has already shown an excellent agreement with the measured single QD PL and PLE spectra (Rastelli et al. 2004) in the
Figure 6.10: Electron and hole wavefunction isosurfaces are plotted for 65% probability density for the magnetic field parallel to the growth direction and perpendicular to the growth direction. The case of an ideal sharp interface (N=0) is shown.

Table 6.2: The shift of the electron and hole energies as a function of N for two different magnetic field directions.

<table>
<thead>
<tr>
<th>N</th>
<th>( \Delta E_e (40T) ) (meV)</th>
<th>( \Delta E_h (40T) ) (meV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>15.26</td>
<td>6.22</td>
</tr>
<tr>
<td>1</td>
<td>-1.45</td>
<td>-0.69</td>
</tr>
<tr>
<td>2</td>
<td>5.13</td>
<td>3.85</td>
</tr>
<tr>
<td>3</td>
<td>-0.14</td>
<td>-0.4</td>
</tr>
<tr>
<td>5</td>
<td>5.19</td>
<td>3.16</td>
</tr>
</tbody>
</table>

Absence of an external magnetic field. Schildermans et al. (2005) have used the same samples to perform low and high excitation PL-measurements using high magnetic fields parallel and perpendicular to the growth direction. In Figs. 6.13 and 6.14 we compare the excitonic absorption spectra to the high exciton PL-results for both field directions. Although the latter case corresponds to a multiexcitonic decay rather than excitonic absorption, we can link the p-p channel absorption to the decay of 3- and 4-fold excitons and the d-d channel absorption...
6.4. Effect of an applied magnetic field

Figure 6.11: Exciton-binding energies as a function of magnetic field applied in the growth direction (a) and in the direction perpendicular to the growth direction (b) for different interface intermixing. $\Delta E_{\text{bind}}$ defined as $\Delta E_{\text{bind}} = E_{\text{bind}}(B) - E_{\text{bind}}(B = 0)$, where $E_{\text{bind}}(B)$ is the magnetic field dependent exciton-binding energy, and $E_{\text{bind}}(B = 0)$ is the exciton binding energy in the absence of a magnetic field.

to that of the 5- to 6-fold exciton, provided the renormalization effects, being linked to the presence of more than one exciton, are small. The agreement however is quite remarkable: Our calculations reproduce the experimental results qualitatively and to a large degree also quantitatively.

The splitting of the s-s channel peaks, caused by the Zeeman splitting of the single-particle orbitals is smaller than the experimental PL-broadening and therefore hidden in the experimental data. The smaller splitting of these peaks for lateral magnetic field is related
6. Unstrained GaAs/Al\textsubscript{1−x}Ga\textsubscript{x}As Quantum Dots

Exciton absorption spectra for $B=10\,\text{T}$ applied in the growth direction for three values of $N=3,7,9$.

Figure 6.12: Exciton absorption spectra for $B=10\,\text{T}$ applied in the growth direction for three values of $N=3,7,9$.

to the smaller Zeeman splitting of electron and hole orbitals, which in turn is related to the large $z$-confinement. An explanation for such a behavior of exciton absorption spectra as a function of the orientation of a magnetic field is given in Sec. IV A.

The exciton diamagnetic shift  More information about the interplay between the Coulomb interaction and the QD-confinement in the direction perpendicular to the direction of the applied magnetic field can be obtained by considering the exciton diamagnetic shift defined as $\Delta E_{\text{exc}} = E_{\text{exc}}(B) - E_{\text{exc}}(B=0\,\text{T})$. The transition energy of two lowest lying optically active excitons are averaged since their Zeeman-splitting is smaller than the broadening of the PL. The obtained data for the exciton diamagnetic shift are compared to the position of the PL peak taken from Ref. (Schildermans et al. 2005) for both orientations of the magnetic field and for different values $N$ of the interface intermixing. The best agreement for the diamagnetic shift for both magnetic field directions is achieved for a value of $N$ in between 0 and 3, whereas for the absolute values of $E_X$ (see inset of Figs. 6.15 and 6.16) the agreement is best for $N$-values in between 3 and 5.

Furthermore, as can be seen from Fig. 6.11 a smoother interface does not significantly affect the change of the X-binding energy with magnetic field. The changes in the curvature of the exciton diamagnetic shift with the variation of $N$ from 0 to 3 or from 3 to 5 are not significant when the magnetic field is applied in the growth direction, as shown in Fig. 6.15 and a good agreement with the experimental data is achieved for all these values of $N$ ($N=0,3,5$). A significant discrepancy to the experimental data is only observed if $N$ is larger than 5. For the lateral magnetic field case, one can see from Fig. 6.16 that the relative exciton diamagnetic shift for $N=5$ is quite close to the one for $N=9$. This is related to the
increased vertical QD-confinement for $N \geq 5$ which leads to a suppression of the magnetic field influence (magnetic length is larger than the height of the dot). The best overall agreement between our findings on the exciton energy and the exciton diamagnetic shift with the experimental data is obtained for our model QD with $N=3$. This result is in perfect agreement to the reported experimental results obtained from X-STM measurements on samples fabricated by using the same growth mode (Lenz et al. 2006).

A similar comparison has been recently performed (Sidor et al. 2006) using a single band model with additional terms for the conduction band non-parabolicity. A good agreement for the relative exciton shift was obtained whereas the absolute PL energies were not repro-
duced accurately. Note that the electron effective mass dominates in the exciton effective mass, and therefore any changes in the electron effective mass would be directly reflected through the changes in the relative exciton shift. Therefore, as the main stumble point in the work of Sidor et al. (2006) arises the question of the degree of the conduction band non-parabolicity actually present in the system. It was a priori assumed to increase, originating from coupling to higher conduction bands. Based on the eight-band $\mathbf{k} \cdot \mathbf{p}$ model we showed here that the inherent coupling of valence- and conduction band present in the eight-band $\mathbf{k} \cdot \mathbf{p}$ model is sufficient to explain the effects observed by Schildermans et al. (2005) without additional non-parabolicity induced by coupling to higher lying conduction bands.

**Figure 6.14:** The same as Fig. 6.13 but now for magnetic field applied in the lateral direction. The available experimental data are shown by open symbols.
6.5 Discussion

First, the interdiffusion of aluminium at the heterojunction during the growth significantly influences the transition energies (see Sec. 6.3). The ground state transition energies for the ideal case of no intermixing (\(N=0\) in Fig. 6.8) and the case of strong intermixing (\(N=9\) in Fig. 6.8) differ by about 37 meV while interlevel transitions are not affected significantly. For example, for \(N=0\) the distance between the transition 1 and transition 2 is 9.2 meV, while in the case of \(N=9\) this distance is 11.8 meV. The observed blue shift between PLE data and calculated absorption spectrum extracted from eight-band \(k\cdot p\) theory (Rastelli et al. 2004) can be explained by this intermixing. The effect of interface intermixing is far from being negligible and should be properly taken into account for potential applications of these systems in optoelectronic devices.

Second, the influence of an applied external magnetic field in the growth direction as well as in-plane on the electronic and optical properties was studied in Sec. 6.4. In general, since the dots are quite large in the lateral direction the effect of a magnetic field applied in the growth direction are more pronounced than if the magnetic field is applied perpendicular to the growth direction. For example, complicated behavior of hole energy levels as a function of magnetic field applied in the growth direction with strong anticrossing does not exist in...

Figure 6.15: The exciton diamagnetic shift as the function of a magnetic field for the optical active exciton states compared to the experimental results (symbols). An external magnetic field is applied in the growth direction. The inset depicts the absolute values for the calculated exciton energy vs. magnetic field together with the experimental data.
the case of magnetic field applied perpendicular to the growth direction which is a direct consequence of the competition between the confinement and magnetic field.

Two questions were raised in the work of Schildermans et al. (2005) related to tunnelling and the degree of non-parabolicity of the conduction band and its influence on the excitonic properties. In our model we explicitly include the mixing of the lowest lying conduction band and the three top most valence bands. Next, in the modelling of the excitonic properties the direct Coulomb interaction, exchange and correlation were included. From Figs. 6.5(b) and (d) it is clear that the penetration of the electron and hole wave functions into the barrier is small and that interface intermixing even increases the localization of the electrons and holes. Therefore, we conclude that the effect of tunnelling in these structures is negligible.

The non-parabolicity of the conduction band originates from the coupling to the valence band and to higher conduction bands. In our eight-band $k \cdot p$ model only the former is accounted for. The confinement and hence the localization of electron and hole states comes into play due to their resulting spread in $k$-space, and the accompanying contributions far from the $\Gamma$ point. This confinement in our case comes predominantly from the dot itself and is enhanced by the applied magnetic field.

From Figs. 6.13, 6.14, 6.15 and 6.16 it is clear that our fit-parameter free results agree very well with the experimental data. We conclude that the inherent coupling of valence- and conduction band present in the eight-band $k \cdot p$ model is sufficient to explain the effects observed by Schildermans et al. (2005) without additional non-parabolicity originating form

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**Figure 6.16:** The same as Fig. 6.15 but now for magnetic field applied in the lateral direction.
coupling to higher conduction bands.

6.6 Conclusions

We studied the electronic structure of unstrained GaAs/Al$_x$Ga$_{1-x}$As QDs in the presence of an external magnetic field. Interface roughness was observed to sensitively affect the transition energies, but hardly the intraband energies. For a magnetic field applied in the growth direction and in the direction perpendicular to the growth direction (where $B \leq 50$ T), we find good agreement between the exciton diamagnetic shift obtained from our calculations and the experimental data from Schildermans et al. (2005). We discussed the effect of tunnelling and the degree of non-parabolicity and found that tunnelling was negligible in our model and that inherent coupling of valence- and conduction band present in the eight-band $k \cdot p$ model is sufficient to explain the "non-parabolicity" effects.
To summarize, two main contributions were made to the field of modelling of the electronic and optical properties of quantum dot in this thesis:

I. We provided an elegant theoretical model for studying the electronic and optical properties of self-assembled QDs taking into account all the relevant effects, where dots can be grown on high index planes or exposed to an applied magnetic or electric field:

- First, different multi-band effective mass models applied to nanostructure were investigated. In the framework of the Burt-Foreman theory, we developed a nonsymmetrized eight-band effective-mass Hamiltonian for nanostructures in the presence of a magnetic field. We find that different treatments of the nanostructure-barrier interface result in the existence of nonphysical solutions for the hole energy levels of a nanostructure. To illustrate this effect, we have chosen a simple disk-like model quantum dot with parabolic in-plane confinement potential, and step-like potential in the growth direction placed in an external magnetic field, and compared the results extracted from our nonsymmetrized model with those obtained from the single-band and conventional multiband calculations, where ad-hoc symmetrization is used. In general, we found that the agreement between our nonsymmetrized model and the conventional models depends heavily on the difference in the structural parameters between the quantum dot and the barrier material. The model is tested on GaAs/Al$_{0.3}$Ga$_{0.7}$As, GaAs/AlAs, and InAs/GaAs quantum dots, where strain is not included in the model in order to resolve the influence of the boundary on the electronic structure. In structures with a large difference of Luttinger parameters between the constituent materials, such as InAs/GaAs quantum dots, the conventional multiband models lead to non-physical high magnetic field solutions which are substantially different from those obtained from the nonsymmetrized Hamiltonian and single-band model for
the ground state. A similar behavior is observed for the case of InAs/GaAs quantum wells, where energy levels as a function of \( k_t \) are analyzed. This discrepancy is attributed to an overestimation of band mixing in conventional models because of the unappropriate treatment of the boundary.

- By employing the nonsymmetrized Hamiltonian, as developed in Chapter 2, a numerical implementation of a full three-dimensional model for the electronic structure calculation of semiconductor QD and QDM grown on high index surfaces and/or placed in an external magnetic field is presented in Chapter 3. The strain distribution of the dots is calculated using continuum elasticity and single-particle states are extracted from the nonsymmetrized eight-band \( k \cdot p \) theory. The model properly takes into account the effects of different substrate orientation by rotation of the coordinate system in the way that one coordinate coincides with the growth direction, whereas the effects of a tilted external magnetic field are taken into account through the Zeeman effect and employing a gauge invariant scheme based on Wilson’s formulation of lattice gauge theory.

II. Second, we explain the diamagnetic shift of the exciton energy for different self-assembled QDs starting from the experimental information on size, shape and composition profile of the dot. The model consistently developed in Chapters 2 and 3 enable us to model electronic and optical properties of various QD systems making comparison with experiment or providing theoretical predictions. In this thesis we have chosen to investigate electronic and optical properties of QDs and QDMs grown on \([11k]\) substrates, where \( k = 1,2,3 \) (Chapter 4), type II InP/InGaP QDs and InAs QDs capped with GaAsSb (Chapter 5), and recently fabricated unstrained GaAs QDs in an external magnetic field (Chapter 6). In more detail:

- We predict variation of electronic and optical properties of InAs/GaAs quantum dots (QDs) and quantum dot molecules (QDM) with the substrate orientation. The QDs’ and QDMs’ transition energies are obtained for high index substrates \([11k]\), where \( k = 1,2,3 \) and are compared with \([001]\). We find that: (i) The QD size in the growth direction determines the degree of influence of the substrate orientation: the flatter the dots, the larger the difference from the reference \([001]\) case. (ii) We predicted the variation of the transition energies of QDM as a function of substrate orientation and inter-dot distances in the eight QD molecule showing that the variation of the inter-dot distance qualitative changes the transition energy dependence on the substrate orientation. For example, for \([111]\) grown molecules, changing the inter-dot distance varies the transition energies up to 50meV. (iii) We show that the size of the QD in the growth direction determines the influence of the (In,Ga)As capping layer on the optical properties of \([11k]\) grown InAs QDs, where \( k = 1,2,3 \). For flat dots, increase of In concentration in the capping layer
leads to a decrease of the transition energy, as is the case of [001] grown QDs, whereas for large dots an increase of the In concentration in the capping layer is followed by an increase of the transition energy up to a critical concentration of In, after which the optical transition energy starts to decrease. Furthermore, we point out the role of piezoelectricity for InAs/GaAs QDs grown on [11k], where $k = 1,2,3,4,5,7,9$ and for QDMs containing eight InAs/GaAs QDs grown on [11l], where $l = 1,2,3$.

- Two cases of type II QDs were discussed: InP/InGaP QDs and molecules in an external magnetic field, and InAs QDs capped with Ga(As,Sb). In both considered cases holes are placed outside the dot and in the former case are localized due to strain, and in the latter due to Coulomb interaction with electrons in the dot. For vertically coupled double and triple InP/In$_x$Ga$_{1-x}$P quantum dot molecules placed in an external magnetic field we show that the size of the dots and the inter-dot distance in the QDM determine which one of the exciton quartets will have the lowest energy and determine also which one of the individual states in the quartet will be the ground state in the presence of an external magnetic field. Competition between confinement, quantum mechanical coupling, and strain influence the exciton diamagnetic shift in double and triple QDM. We found that the available experimental data (Hayne et al. 2000, Hayne et al. 2001) were successfully described by one of the optically active exciton states of the lowest lying exciton quartet.

- The electronic and optical properties of unstrained self-assembled QDs with precisely known sizes and shape placed in an external magnetic field are investigated, and our findings were compared with the experimental results. We found that varying interface roughness sensitively affects the interband but hardly the intraband energies. For magnetic fields applied both in growth direction and perpendicular to it (for $B \leq 50$T), we found good agreement between our predicted exciton diamagnetic shift and recent experimental magneto-photoluminescence data (Schildermans et al. 2005). The inherent coupling of valence- and conduction band taken into account in the eight-band $k \cdot p$ model explains well the observed experimental results.
Samenvattend werden er in deze thesis twee belangrijke bijdragen geleverd in het gebied van de elektronische en optische eigenschappen van kwantumstippen:

I. We hebben gezorgd voor een duidelijk theoretisch model voor het bestuderen van de elektronische en optische eigenschappen van zelf-georganiseerde kwantumstippen door rekening te houden met alle belangrijke effecten, waar stippen kunnen worden gegroeid op hoge index vlakken of onderworpen aan een extern magnetisch of elektrisch veld:

- Eerst werden er verschillende theoretische modellen gebaseerd op meerbands-effectieve-massa benadering onderzocht en toegepast op nanostructuren. In het kader van de Burt-Foremantheorie ontwikkelden we een niet-gesymmetriseerde acht-bands-effectieve-massa Hamiltoniaan voor nanostructuren in de aanwezigheid van een magnetisch veld. We stelden vast dat verschillende behandelingen van het grensvlak van de nanostructuurbarrière leiden tot het ontstaan van niet-fysische oplossingen voor de gatenenergieniveaus van een nanostructuur. Om dit effect te illustreren hebben we gekozen voor een eenvoudige schijfvormige model kwantumstip met parabolische opsluitingspotentiaal in het vlak, en trapvormpotentiaal in de groeirichting geplaatst in een uitwendig magnetisch veld. De uit ons niet-gesymmetriseerd model afgeleide resultaten hebben we vergeleken met die van de enkelvoudige band en met de conventionele meerbandsberekeningen, waarbij gebruik wordt gemaakt van ad-hoc symmetrisering. In het algemeen hebben we vastgesteld dat de overeenstemming tussen ons niet-gesymmetriseerd model en de conventionele modellen erg afhangt van het verschil in structurele parameters tussen de kwantumstip en het materiaal van de barrière. Het model werd getest op GaAs/Al$_{0.3}$Ga$_{0.7}$As, GaAs/AlAs, en InAs/GaAs kwantumstippen, waarbij de spanning niet in rekening werd gebracht. In structuren met een groot verschil qua Luttinger parameters tussen de gebruikte materialen, zoals InAs/GaAs kwantum-
stippen, leiden de conventionele meerbandsmodellen tot *niet-fysische* resultaten bij hoge magnetische-veldwaarden die wezenlijk verschillen van de waarden die men krijgt bij de niet-gesymmetriseerd Hamiltoniaan en éénbandsmodel voor de grondtoestand. Gelijkaardig gedrag wordt waargenomen bij InAs/GaAs quantum putten, waarbij energieniveaus werden geanalyseerd in functie van \( k \). Deze discrepantie wordt toegeschreven aan een overschatting van band-mixing bij de conventionele modellen omwille van de onjuiste behandeling van de begrenzing.

- Voor de uitvoering van de niet-gesymmetriseerde Hamiltoniaan, zoals opgebouwd in Hoofdstuk 2 wordt in Hoofdstuk 3 een numerieke toepassing weergegeven van het volledige drie dimensioneel model voor de berekening van de elektronische structuur van halfgeleider kwantumstippen en kwantumstipmoleculen gegroeid op kristal vlakken met hoge index en/of in de aanwezigheid van een uitwendig magnetisch veld. De spanningsverdeling van de stippen werd berekend door gebruik te maken van continu¨um-elasticiteit en de toestand van de enkelvoudige deeltjes werden genomen van de niet-gesymmetriseerde acht-bands-\( \mathbf{k} \cdot \mathbf{p} \)-theorie. Het model houdt voldoende rekening met de gevolgen van verschillende substratorientaties door rotatie van het co¨ordinatensysteem zodat één co¨rdinaat samenvalt met de groeirichting. De effecten van een gekanteld uitwendig magnetisch veld werd in rekening gebracht via het Zeeman-effect en door toepassing van een ijkinvariantieschema gebaseerd op de formulering van Wilson.

II. Ten tweede werd de diamagnetische verschuiving van de energie van het exciton voor verschillende zelf-georganiseerde kwantumstippen berekend uitgaande van de experimentele gegevens betreffende grootte-, vorm- en samenstellingsprofiel van de kwantumstip. Het model dat in de Hoofdstukken 2 en 3 werd uitgewerkt maakt het mogelijk om de elektronische en optische eigenschappen van verschillende kwantumstipsystemen te vergelijken met experimenten of om theoretische voorspellingen te doen. We hebben er in deze thesis voor gekozen om de elektronische en optische eigenschappen van op [11k]-substraten gegroeide kwantumstippen en kwantumstipmoleculen te onderzoeken. Hierbij is \( k=1,2,3 \) (Hoofdstuk 4), type II InP/InGaP kwantumstippen en InAs kwantumstippen overkapt met GaAsSb (Hoofdstuk 5), en onlangs gefabriceerde GaAs kwantumstippen zonder spanningen in een uitwendig magnetisch veld (Hoofdstuk 6).

Meer gedetailleerd:

- Wij voorspellen een verandering van de elektronische en optische eigenschappen van InAs/GaAs kwantumstippen en kwantumstipmoleculen naargelang de substratorientatie. De overgangsenergiewaarden van kwantumstippen en kwantumstipmoleculen werden bekomen voor [11k]-substraten met hoge index, waarbij \( k = 1,2,3 \) en werden vergeleken met [001]. We stellen vast dat: (i) De grootte van de kwantumstip in de groeirichting de beïnvloedingsgraad van de substratorientatie
bepaalt: hoe egaler de stippen, hoe groter het verschil met het referentie oppervlak [001]. (ii) Wij voorspelden de verandering van de overgangsenergiewaarden van de kwantumstipmoleculen in functie van de substraatoriëntatie en de afstanden tussen de stippen in de achttien kwantumstipmoleculen, waaruit blijkt dat de verandering van de afstanden tussen de stippen de afhankelijkheid van de overgangsenergie ten opzichte van de substraatoriëntatie kwalitatief wijzigt. Bijvoorbeeld, voor op [111] gegroeide moleculen brengt een verandering van de afstand tussen de stippen een verandering van de overgangsenergiewaarden tot 50meV teweeg. (iii) We tonen aan dat de grootte van de kwantumstip in de groeirichting de invloed bepaalt van de (In,Ga)As overkappingslaag op de optische kenmerken van de op [11k] gegroeide InAs kwantumstippen, waarbij k=1,2,3. Voor platte stippen leidt een verhoging van de In-concentratie in de overkappingslaag tot afname van de overgangsenergie, zoals het geval ook is bij op [001] gegroeide kwantumstippen, terwijl voor grote stippen een verhoging van de In-concentratie in de overkappinglaag een toename van de overgangsenergie tot gevolg heeft tot de kritieke concentratie van In, waarna de optische overgangsenergie begint af te nemen. Verder wijzen we op de rol die piezo-elektriciteit speelt bij InAs/GaAs kwantumstippen gegroeide op [11k], waarbij k = 1,2,3,4,5,7,9, en bij kwantumstipmoleculen die acht op [11l] gegroeide InAs/GaAs kwantumstippen bevatten, waarbij l = 1,2,3.

Er werden twee gevallen van kwantumstippen van type II besproken: InP/InGaP kwantumstippen en moleculen in een uitwendig magnetisch veld, en InAs kwantumstippen overkapt met Ga(As,Sb). In beide gevallen bevinden er gaten ladingsdragers buiten de stip, in het eerste geval gelokaliseerd door middel van spanning en in het laatste geval door middel van Coulomb-wisselwerking met de elektro- nen in de stip. Voor verticaal gekoppeld dubbele en driedubbele InP/In$_{x}$Ga$_{1-x}$P kwantumstipmoleculen geplaatst in een uitwendig magnetisch veld tonen we aan dat de grootte van de stippen en de afstand tussen de stippen in de kwantumstipmoleculen bepalen welke van de exciton kwartetten de laagste energiewaarde zal hebben en ook bepalen welke van de individuele toestanden in het kwartet de grondtoestand zal zijn in de aanwezigheid van een uitwendig magnetisch veld. De onderlinge strijd tussen beperking van plaats, mechanische kwantumkoppeling en spanning beïnvloedt de diamagnetische excitonverschuiving in de dubbele en driedubbele kwantumstipmoleculen. We hebben vastgesteld dat de beschikbare experimentgegevens van Hayne et al. (2000) en Hayne et al. (2001) succesvol werden beschreven door één van de optisch actieve excitontoestanden van het laagstliggende exciton kwartet.

De elektronische en optische kenmerken van de zelf-georganiseerde kwantumstippen zonder spanning, met exact gekende grootten en vormen, en geplaatst
in een uitwendig magnetisch veld werden onderzocht en onze resultaten werden vergeleken met het experiment. We stelden vast dat de wisselende ruwheid van het grensvlak wel de intraband-, maar nauwelijks de intraband-energiewaarden beïnvloedt. Voor magnetische velden die zowel in de groeirichting als loodrecht op die richting worden toegepast (voor B ≤ 50T), stelden we een duidelijke overeenkomst vast tussen de door ons voorspelde diamagnetische excitonverschuiving en de recente experimentele magneto-fotoluminescentie data gegevens van Schildermans et al. (2005). De inherente koppeling van de valentie- en conductieband waarmee rekening wordt gehouden in het acht-bands-\( \mathbf{k} \cdot \mathbf{p} \)-model verklaart de waargenomen experimentele resultaten.
A multitude of current and potential applications of quantum dots requires a deeper understanding of the quantum dot electronic structure not only on the qualitative but also on the quantitative level. The presented model is sufficient to describe photoluminescence and magnetophotoluminescence measurements performed both, on an ensemble of QDs and on single QD. Currently, we are able to model self-assembled QDs made of III-V zinc-blende direct gap semiconductors, where minimum of the conduction band and maximum of the valence band (if we neglect linear k terms) are at the \( \Gamma \) point.

Further development of the model can go in two directions:

- Extension of the model to include a more precise description of the band diagram of the whole Brillouin zone as well as to calculate exciton complexes would give a powerful tool to cover a wide range of experiments in the area of semiconductor nanostructures.

- On the other hand, the present model can be modified by implementing a first principle envelope function that includes atomistic symmetry. Namely, the envelope functions are not formulated in terms of bulk compounds but atoms, whereas a traditional bulk-crystal description can be obtained from the atomistic formulation via linear transformation of variables (Foreman 2006a, Foreman 2005). It would also include a “real atomistic surface” in contrast to standard envelope function approach where the interface is “just a potential barrier”. Theoretically, such an implementation would be as good as atomistic approaches. The main drawback is the evaluation of a parameters set that has to be obtained for each new material under investigation.

Request for “complete” understanding of nanoworld with the aim to produce novel optoelectronic devices, enabling a breakthrough in modern photonic technologies is still pending. From the experimental point of view, fabrication of highly ordered QD configurations by exploiting a profound understanding of the formation processes is one of the primarily interests and in general would lead to the construction of QD superlattices, creating QD solids,
or integration of QD systems into photonic crystal cavities or in waveguides. Theoretical modeling should follow-up these experimental developments, providing an understanding of the electronic states in coupled QDMs, and QD superlattices, exciton and photon entanglement in those systems, identification of the results of optical spectroscopy performed on such QD systems, exciton complexes, and the effects of external electric and magnetic fields. Furthermore, investigation of the materials for solar energy still attracts a lot of attention, for example creation of seven excitons per single photon using semiconductor nanocrystals have been reported recently (Schaller and Klimov 2006).
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<td>0D</td>
<td>Zero dimensional</td>
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<tr>
<td>1D</td>
<td>One dimensional</td>
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<td>2D</td>
<td>Two dimensional</td>
</tr>
<tr>
<td>3D</td>
<td>Three dimensional</td>
</tr>
<tr>
<td>AFM</td>
<td>Atomic Force Microscopy</td>
</tr>
<tr>
<td>BF</td>
<td>Burt-Foreman</td>
</tr>
<tr>
<td>CB</td>
<td>Conduction band</td>
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<tr>
<td>CGM</td>
<td>Conjugate Gradient Method</td>
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<td>CI</td>
<td>Configuration interaction</td>
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<td>CL</td>
<td>Cathodoluminescence spectroscopy</td>
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<td>Continuum mechanical model</td>
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<td>DBR</td>
<td>Distributed Bragg Reflector</td>
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<td>DELL</td>
<td>Dot-in-a-Well structure</td>
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<td>Empirical Tight-Binding Model</td>
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<tr>
<td>FPA</td>
<td>Focal plane array</td>
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<td>First-Principle Envelope Function Theory</td>
</tr>
<tr>
<td>HRTEM</td>
<td>High-resolution transmission electron microscopy</td>
</tr>
<tr>
<td>IE</td>
<td>Isotropy Elasticity</td>
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<tr>
<td>JD</td>
<td>Jacobi-Davidson</td>
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<tr>
<td>JOCC</td>
<td>Jacobi’s orthogonal component correction</td>
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<td>LED</td>
<td>Light Emitting Diode</td>
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<tr>
<td>MOCVD</td>
<td>Metal-Organic-Chemical-Vapour-Deposition</td>
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<tr>
<td>MBE</td>
<td>Molecular Beam Epitaxy</td>
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<td>NSOM</td>
<td>Near-field Scanning Optical Microscopy</td>
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<td>Quantum Dot</td>
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<td>QDM</td>
<td>Quantum Dot Molecule</td>
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<tr>
<td>QMRS</td>
<td>Quasi Minimal Residual Simplified</td>
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<td>QW</td>
<td>Quantum Well</td>
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<tr>
<td>UHV</td>
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<td>X-STM</td>
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Research Expertise


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05/2002 First prize in the Serbian state competition in Laser Engineering, Belgrade, Yugoslavia


Research Publications

List of publications attached at the end.

Major Talks

06/03/2007 “Influence of the substrate orientation on the electronic and optical properties of InAs/GaAs quantum dots”, contributed talk, The 2007 American Physical Society Meeting, Denver, Colorado, USA.


03/04/2006 “Theoretical study of InAs/GaAs quantum dots grown on [11k] substrates in the presence of a magnetic field”, contributed talk, 6th International Workshop on Epitaxial Semiconductors on Patterned Substrates and Novel Index Surfaces, Nottingham, UK.

26/05/2005 “Quantum dot electronic states in a magnetic field”, invited talk, Institut für Festkörperphysik, Technische Universität Berlin, Berlin, Germany.

Technical skills

- Languages: C/C++, Fortran, Python
- Parallel programming: MPI
- PSE’s: Mathematica, MATLAB, Comsol

Language Proficiency

English (fluent), French (good), Dutch (basic), Serb-Croat (native)

Interests and Hobbies

Classical guitar, tennis, skiing
List of publications

Refereed journals

Conference proceedings


To be submitted


- V. Mlinar and F. M. Peeters, *Effective g-factor in self-assembled quantum dots grown on high-index surfaces*. 