dr Michał Zieliński
Uniwersytet Mikołaja Kopernika,
Instytut Fizyki, Wydział Fizyki, Astronomii i Informatyki Stosowanej
ul. Grudziądzka 5, 87-100 Toruń

Self-presentation

I. Name:

Michał Stanisław Zieliński

II. Education:

- 2002 r.: Nicolaus Copernicus University, Toruń, MSc degree, theoretical physics, supervisor – prof. dr hab. Włodzimierz Jaskólski, title of thesis: *Tight binding method: nanostructure and charge density visualization.*
- 2002 r.: Nicolaus Copernicus University, Toruń, MSc degree, microelectronics, supervisor – prof. dr hab. Wacław Bała, title of thesis: *Measurments of refraction index and thickness of thin organic layers (CuPc and PPV) by coupled-prism method.*
- 2006 r.: Nicolaus Copernicus University, Toruń, PhD in theoretical physics, supervisor – prof. dr hab. Włodzimierz Jaskólski, title of thesis: *Effects of strain on electronic and optical properties of quantum dots.*

III. Employment:

- 2006-2009: Research Officer, Quantum Theory Group, Institute for Microstructural Sciences, National Research Council of Canada – postdoctoral researcher
- 2009-: Assistant Professor, Quantum Mechanics Department, Institute of Physics, Nicolaus Copernicus University, Toruń

IV. Awards and scholarships

- 2014 r.: Second Class NCU Rector Award for individual scientific achievements
- 2013 r.: First Class NCU Rector Award for individual scientific achievements
- 2013-2016 r.: Scholarship for outstanding young scientists (Polish Ministry of Science and Higher Education)
- 2009 r.: Foundation for Polish Science and TPKN Grzegorz Białkowski Award (II place) for best PhD thesis in last three years: „Effects of strain on electronic and optical properties of quantum dots.”
- Award (II degree) of the Polish Physical Society for best Master's thesis: “Tight binding method for nanocrystals: density of probability and energy spectra.”
- 2000 and 2001 Scholarships of the Polish Ministry of Education

V. The scientific achievement:

a. Title of the scientific achievement

Series of publications: Atomistic calculations of excitonic effects in quantum dots.

b. Authors/authors, title, year of publication, name of the journal:

(More detailed contributions are given in separate Appendices, including statements of other coauthors)

[H1] M. Zieliński, M. Korkusiński, and P. Hawrylak,
Atomistic tight-binding theory of multi-exciton complexes in a self-assembled InAs quantum dot,
Phys. Rev. B **81**, 085301 (2010)
Impact factor (2010): 3.772
my contribution to the paper: 65%

[H2] M. Zieliński,
Including strain in atomistic tight-binding Hamiltonians: An application to self-assembled InAs/GaAs and InAs/InP quantum dots,
Phys. Rev. B **86**, 115424 (2012)
Impact factor (2012): 3.767
my contribution to the paper: 100%

[H3] M. Zieliński,
Multi-Scale Simulations of Semiconductor Nanostructures,
Acta. Phys. Pol. A **122**, 312 (2012) ,
(Proceedings of the WELCOME Scientific Meeting on Hybrid Nanostructures)
my contribution to the paper: 100%

[H4] M. Zieliński,
Valence band offset, strain and shape effects on confined states in self-assembled InAs/InP and InAs/GaAs quantum dots,
J. Phys. Condens. Matter **25**, 465301 (2013)
Impact factor (2013): 2.223
my contribution to the paper: 100%

[H5] M. Zieliński,
Influence of substrate orientation on exciton fine structure splitting of InAs/InP nanowire quantum dots,
Nanoscale Research Letters 2012, **7**:265
Impact factor (2012): 2.524
my contribution to the paper: 100%

[H6] M. Zieliński,
Excitonic fine structure of elongated InAs/InP quantum dots,
Phys. Rev. B **88**, 155319 (2013)
Impact factor (2013): 3.664
my contribution to the paper: 100%

[H7] M. Zieliński,
Fine structure of light-hole excitons in nanowire quantum dots,
Phys. Rev. B **88**, 115424 (2013)
Impact factor (2013): 3.664
my contribution to the paper: 100%

[H8] M. Zieliński, Y. Don, D. Gershoni,
Atomistic theory of dark excitons in self-assembled quantum dots of reduced symmetry,
Phys. Rev. B **91**, 085403 (2015)
Impact factor (2013/2014): 3.664
my contribution to the paper: 75%

[H9] M. Zieliński, K. Gołasa, M. R. Molas, M. Goryca, T. Kazimierczuk, T. Smoleński, A. Golnik, P. Kossacki, A.A.L. Nicolet, M. Potemski, Z. R. Wasilewski, and A. Babiński,
Excitonic complexes in natural InAs/GaAs quantum dots,
Phys. Rev. B **91**, 085303 (2015)
Impact factor (2013/2014): 3.664
my contribution to the paper: 40%
my contribution to the theory part of the paper: 100%

[H10] M. B. Bavinck, M. Zieliński, B. Witek, T. Zehender, E. Bakkers, V. Zwiller,
Controlling a Nanowire Quantum Dot Bandgap Using a Straining Dielectric Envelope,
Nano lett., **12**, 6202 (2012)
Impact factor (2012): 13.025
my contribution to the paper: 25%
my contribution to the theory part of the paper: 100%

[H11] P. Wojnar, M. Zieliński, E. Janik, W. Zaleszczyk, T. Wojciechowski, R. Wojnar, M. Szymura, Ł. Kłopotowski, L. T. Baczewski, A. Pietruchik, M. Wiater, S. Kret, G. Karczewski, T. Wojtowicz and J. Kossut,
Strain-induced energy gap variation in ZnTe/ZnMgTe core/shell nanowires,
Appl. Phys. Lett. **104**, 163111 (2014)
Impact factor (2013/2014): 3.515
my contribution to the paper: 25%
my contribution to the theory part of the paper: 90%

c. Discussion of scientific goals, results obtained, and their applications:

Introduction

Quantum dots are manmade semiconductor nanostructures [**1**]. Their small, nanoscopic, dimensions lead to a discreet electronic structure that resembles properties of single atoms. Unlike natural atoms, quantum dots spectra can be modified by a control of a growth process, that allows for control of size, shape and composition. Quantum dots have already fund numerous application as lasers or biological markers. There is an ongoing research aiming for quantum dot applications as efficient single photon [**2**,**3**] and entangled photon [**4**-**6**] sources. Apart from current and future applications, quantum dots allow for basic research on confined many-body systems.

The progress of experimental techniques in recent years lead to measurements of emission spectra of excitons (electron-hole pairs) confined in single quantum dots [**7**-**9**]. Emission spectra of typical self-organized quantum dot [**1**] consist of a doublet of optically inactive states – dark excitons – and energetically higher doublet of optically active states – bright excitons [**9**]. In the language of effective mass approximation the bright exciton states are given as electron-hole configurations of anti-parallel electron and hole (quasi-)spins alignment. On the other hand dark exciton states correspond to configurations of parallel spin alignment of both charge carriers. The energy difference between the dark and bright states is denoted as the electron-hole exchange splitting. Whereas the splitting of the bright doublet is known as the *bright exciton splitting* or the *fine structure splitting*. Traditionally this splitting is also known as the *anisotropic electron-hole exchange*. This splitting constitutes a fundamental limit [**6**] for potential quantum dot applications in so-called biexciton-exciton cascade [**4**] for the entangled photon generation. Understanding physics of the bright doublet splitting [**H4**-**H7**] and mechanisms that would allow for its reduction is thus essential for quantum dot applications in quantum telecommunication.

Recently conducted research shows perspectives for dark excitons studies in a fully optical experiments [**10**]. It is now possible to deterministically generate a dark exciton in a well-defined spin state using a single optical pulse [**P12**]. This research opens a route towards application of dark excitonic states as long-lived solid state qubits, whereas one of the key issues is the knowledge of dark excitons lifetimes and possible recombination channels [**H8**].

Novel experimental techniques allow for growth of semiconductor quantum dots embedded in quasi-one-dimensional nanowires [**11**]. This technology provide a large degree of quantum dot position, size and optical properties control [**12**,**13**]. Additionally, spectra of such nanostructures can be modified by a post-growth of lattice-mismatched nanowire shells [**H10,H11**]. This kind of approach could be called *strain engineering*, in analogy to similar techniques used in semiconductor industry.

The experimental research on nanostructures has been assisted by a progress in development of theoretical methods. Already in the 1990s there were an extensive theoretical studies on spectral properties of quantum dot confined excitons. The research was typically conducted based on the continuous medium approximation, utilizing tools such as the **kp** method or the effective mass approximation. These works allowed for understanding of main features of

excitonic spectra for quantum dot ensembles. Experimental research on single quantum dots, in particular research on excitonic fine structure, forced further development of computational methods such as the empirical pseudopotential method [**14**,**15**] and the empirical tight-binding method [**16-18**,**H1-H10**,**P1**,**P8**,**P9**].

In recent years, research based on atomistic approach, including my own work [**H1-H9**], has shown a fundamental influence of nanostructure and lattice symmetry, and substrate orientation on excitonic properties (including fine structure) of semiconductor quantum dots. It was also found that in realistic quantum dots effects of non-uniform composition profile and atomistic character of quantum dot-surrounding matrix interface play an important role. From the modelling point of view it lead to development of atomistic approaches going beyond the limits of methods derived from the continuous matter approximation.

The first goal of the discussed series of publications was to develop and implement theoretical methods allowing for atomistic description of excitonic properties of various nanostructures [**H1-H4**]. The next goal was an attempt to answer the key questions related to confined excitons spectra [**H5-H11**]. In particular the effort was focused on details of multiexcitons, and the bright, and the dark excitons fine structure. The research was conducted for self-assembled InAs/GaAs and InAs/InP quantum dots, nanowire InAs/InP quantum dots, and InP and ZnTe externally strained nanowires.

Theoretical methods

Self-assembled quantum dots are one of the most studied nanostructures. They are effectively grown as "islands" of a low-band gap semiconductor surrounded by a larger band gap material. Computational domains for this kind of nanosystems typically contain million atoms and more. For such a number of atoms the *ab-initio* type of calculation is beyond the reach of modern computers. As a practical solution, this complicated problem is solved by a series of computationally feasible, approximate computations [**15**,**19**]. General description of a method applied by myself and coworkers is shown below, whereas a detailed description can be found in the first paper of the series [**H1**].

Lattice mismatch effects

In the first step of the computational process, lattice mismatch of a quantum dot and a surrounding matrix is accounted for. From the numerical point of view this is done by an optimization procedure of positions of all atoms in the computational domain, that minimizes the total elastic energy of the system [**20**,**21**,**H1**,**P9-P11**]:

$$E_{TOT} = \frac{1}{2}\sum_{i=1}^{N}\sum_{j=1}^{nn} A_{ij}\left[\left(\vec{R}_i - \vec{R}_j\right)^2 - {d_{ij}}^2\right]^2 + \sum_{i=1}^{N}\sum_{j=1}^{nn}\sum_{k=j+1}^{nn} B_{ijk}\left[\left(\vec{R}_j - \vec{R}_i\right)\cdot\left(\vec{R}_k - \vec{R}_i\right) - \frac{1}{3}d_{ij}d_{ik}\right]^2.$$

Where in the above equation $\vec{R}_i$ describes the position *i*-th atom, d_{ij} is the bond length between *i* and *j* atoms in the ideal (unstrained) bulk crystal. A_{ij} and B_{ijk} are material parameters. The summation goes over all *N* atoms and *nn* nearest neighbors. The E_{TOT} minimum is found by a conjugate gradients method. The novelty of [**H1**] is e.g. the parallel implementation and the parallel computation on a computer-clusters that allowed for attacking nanosystems containing more than 100 million atoms. Paper [**H2**] describes in details the role of different parametrizations (A_{ij}, B_{ijk}), whereas paper [**H3**] studies the effect of a computational domain size and shape on the efficiency of the optimization process and the quality of results.

Single particle states

In the second step of calculation the single particle states are found by means of the empirical tight binding method. In this approach material related quantities (such as *onsite energies* and *hoppings*) are treated as empirical parameters. The tight binding wave function is given as a linear combination of atomic orbitals (*LCAO*):

$$\phi = \sum_{\vec{R}\alpha} c_{\vec{R}\alpha}\,|\,\vec{R}\alpha\rangle,$$

where the summation goes over all atomic positions $\vec{R}$, α is the (spin)orbital index, $c_{\vec{R}\alpha}$ is the expansion coefficient in the $\left|\vec{R}\alpha\right\rangle$ basis.

For a nanostructure consisting of *N* atoms, the tight binding Hamiltonian in the language of second quantization is given as:

$$\widehat{H}_{TB} = \sum_{i=1}^{N}\sum_{\alpha=1}^{20} \varepsilon_{i\alpha}\, c_{i\alpha}^{+} c_{i\alpha} + \sum_{i=1}^{N}\sum_{\alpha,\beta=1}^{20} \lambda_{i\alpha,i\beta}\, c_{i\alpha}^{+} c_{i\beta} + \sum_{i=1}^{N}\sum_{j=1}^{nn=4}\sum_{\alpha,\beta=1}^{20} t_{i\alpha,j\beta}\, c_{i\alpha}^{+} c_{j\beta},$$

where $c_{i\alpha}^{+}(c_{i\alpha})$ is the creation (annihilation) of electron on (spin)orbital α centered on *i*-th atom. $\varepsilon_{i\alpha}$ and $t_{i\alpha,j\beta}$ are correspondingly onsite (atomic) energies and offsite (hopping) parameters. Parameters $\lambda_{i\alpha,i\beta}$ account for the spin-orbit effect, whereas λ_a, λ_c are related to spin-orbit splitting for anion and cation *p* states (more details can be found in a work by D.J. Chadi [**22**]). In the current series of publications I have implemented $sp^3d^5s^*$ [**H2**,**23**] tight binding model, with 20 valence spin-orbitals per atom (including 10 *d* spin-orbitals).

In practical calculations [**H2**-**H10**] parameter set ε, λ, t was taken from [**23**] ([**H1**] used different parameter sets). These parameters were obtained by fitting semiconductor bulk band structures (energies and effective masses at high-symmetry points) to experimental data and *ab-initio* calculations.

Including strain in the tight binding Hamiltonian

A fundamental issue in a practical tight-binding calculations of nanostructure spectra is related to accounting for atomic positions modified in the optimization (strain calculation) step. These positions, and therefore bond lengths and angles are by definition different form the ideal bulk crystal, for which the tight binding parameters (ε, λ, t) where originally determined.

The above problem is thoroughly discussed and solved in paper [**H2**]. The solution is the functional dependency that relates the tight binding parameters with atomic positions. In particular, apart from the off-diagonal parameters t dependence on bond lengths and angles [**24**,**25**], it is necessary to account for the effect of strain on diagonal [**23**,**26**,**H2**] Hamiltonian matrix elements (ε). In [**H2**] a suitable diagonal correction was presented in a formula directly applicable for nanostructures. This correction was derived from its bulk biaxial strain counterpart. For example, the diagonal energy shift for d_{xy} orbital looks as following:

$$\varepsilon_{xy} = \varepsilon_d + \frac{2b_d}{nn}\sum_{i}^{nn}(d_i/d_i^o)\left[\frac{m_i}{m_i^0} - \frac{1}{2}\left(\frac{n_i}{n_i^0} + \frac{l_i}{l_i^0}\right)\right],$$

where ε_d is the unstrained on-site energy, *nn* – is the number of nearest neighbors, d_i is the bond length, n_i, l_i, m_i are direction cosines describing the bond orientation in space. Values with 0 index and without this index describe ideal and strained quantities correspondingly. The diagonal parameters correction is a necessary contribution to the $sp^3d^5s^*$ model, and thus must be used for both quantitative and qualitative agreement with the experiment. This is true

for the tight binding calculation of both strained bulk crystals (deformation potentials) and semiconductor nanostructures (effective energy gaps).

Numerical challenges

For one million atom quantum dot the tight binding Hamiltonian matrix ($\widehat{H}_{TB}$ in the $|\vec{R}\alpha\rangle$ basis) in the $sp^3d^5s^*$ parameterization has the size of 20×10^6 by 20×10^6. However due to nearest neighbors approximation it is a sparse matrix. This allows to store effectively only non-zero matrix elements, number of which is proportional to N rather than N^2.

A parallel version of Lanczos [**27**] algorithm is utilized for finding eigenvectors and eigenvalues for a selected energy window (close to bulk band gap). Moreover in the Lanczos algorithm (similarly to an analogous Arnoldi algorithm [**28**]) one needs not to operate on the matrix itself, but rather perform matrix-vector multiplications only. This enables on-flight non-zero matrix elements computation without storing them in computer's memory. Such an approach, utilized by the author in the entire series of publications, allows nowadays for the tight binding calculation on parallel computer clusters for numbers of atoms reaching 5×10^6 and Hamiltonian matrix dimensions reaching 10^8.

Excitons

The tight-binding calculation gives the electronic structure of single particles states. Quantum states of energies above the effective band gap are customarily denoted as electron states, whereas states below the gap are denoted as holes. The origin of these names can be traced back to bulk states description in terms of conduction (electrons) and valence (holes) bands.

In a typical quantum dot photoluminescence experiment one measures spectra coming from electron-hole (exciton) recombination. Apart from the single exciton emission it is possible to observe multi-excitonic complexes, both neutral (such as the biexciton) and charged (such as the trion X^-). The final step of the computational process involves therefore a many-body calculation. In the language of second quantization the Hamiltonian for an interacting electron-hole system is given as [**19**,**H1**]:

$$\widehat{H}_{ex} = \sum_i E_i^e \, e_i^+ e_i + \sum_i E_i^h \, h_i^+ h_i + \frac{1}{2}\sum_{ijkl} V_{ijkl}^{ee} \, e_i^+ e_j^+ e_k e_l + \frac{1}{2}\sum_{ijkl} V_{ijkl}^{hh} \, h_i^+ h_j^+ h_k h_l - \sum_{ijkl} V_{ijkl}^{eh,dir} \, e_i^+ h_j^+ h_k e_l + \sum_{ijkl} V_{ijkl}^{eh,exch} \, e_i^+ h_j^+ e_k h_l,$$

where i, j, k, l are indices of single particle states, E_i^e and E_i^h are electron and hole single particle energies taken from the tight binding calculation. V_{ijkl}^{ab} are Coulomb matrix elements (Coulomb and exchange integrals). A pair of upper indices describes the type of particles (e – electron, h – hole), and the type of the integral (*dir – Coulomb direct*, *exch – exchange*).

The multi-excitonic Hamiltonian is expanded in a basis electron-hole configurations formed from several lowest single particle electron and hole states. Depending on the research field such approach is known as the configuration interaction method or the exact diagonalization

approach. Typically in my calculations six (twelve with spin) lowest electron states and six (twelve with spin) lowest hole states are used. This corresponds to accounting for electron and hole *s*, *p,* and *d* shells of self-organized quantum dots. In this case the number of configurations for a single exciton is equal to 144, for trions is 792, for the biexciton 4356 and grows rapidly for other complexes.

In the basis of 12 electron and 12 hole states one needs to calculate 4×12^4 V_{ijkl} Coulomb matrix elements. These integrals are defined as:

$$V_{ijkl} = \int\int \phi_i^*(\vec{r}_1)\phi_j^*(\vec{r}_2)\frac{e^2}{\epsilon(\vec{r}_1,\vec{r}_2)|\vec{r}_1-\vec{r}_2|}\phi_k(\vec{r}_2)\phi_l(\vec{r}_1)dV_1dV_2,$$

where $\epsilon(\vec{r}_1,\vec{r}_2)$ is the dielectric screening, ϕ are the single particle (LCAO) states given by the tight binding method. The computational cost of V_{ijkl} calculation scales as $O(N^4)$, where N is the number of atoms. Utilizing a series of approximations [**H1**] it is however possible to reduce the computational cost to $O(N^2)$, moreover the calculation process can be efficiently parallelized. Nevertheless for large N (on the order of 10^6) the computation of V_{ijkl} constitutes the most time demanding stage of the calculation.

Once the many-body Hamiltonian matrix is established, the excitonic energy spectra is obtained by its diagonalization. This is followed by a calculation of the optical excitonic spectra. These spectra describe all possible routes for a recombination of N exciton to $N-1$ exciton system (or a vacuum state for a single exciton). Optical spectra are determined based on eigenvalues and eigenvectors of N and $N-1$ exciton cases and the Fermi's Golden Rule [**H1**].

Computational efficiency

Computational domains for typical self-assemble quantum dots involve a quantum dot and a certain volume of the quantum dot surrounding matrix. In the next paper of the series [**H3**] it has been shown that the amount of matrix material that needs to be accounted for, and therefore the size of the computational domain, depends significantly on the stage of the calculation. Multi-scale approach proposed in [**H3**] is based on the following observation: the long-range character of strain leads to domain sizes containing even up to 10^8 atoms, however that domain necessary for the converged tight-binding results can be significantly smaller, up to about 10^6 atoms. Importantly, in the following stage of the calculation, i.e. many-body part, one can even further reduce the computation domain size. This effectively leads to three, progressively smaller computational domains in all three stages of the calculation. It is even possible to further reduce the computational complexity, at the cost of reasonably small accuracy decrease: typically the domain size can be reduced in a lateral direction, whereas keeping the domain height (along the growth axis) unaltered.

Energy optical spectra of quantum dots

Electron and hole confining potentials in quantum dots depend on many factors, in particular quantum dot and surrounding material valence band offset. They also depend on quantum dot size, shape, and chemical composition. All these effects are studied in detail in the fourth paper of a series [**H4**].

Valence band offset

In has been shown [**H4**] that the valence band offset is one of the key semi-empirical parameters used in the electronic structure calculations of nanostructures. It has also been demonstrated that the character of the confining potential in quantum dots significantly depends on a non-trivial coupling between the valence band offset and the spatial strain distribution. In particular, it has been qualitatively shown that strain effects are responsible for a characteristic shell-like structure of disk-shaped self-assembled quantum dots. Strain effects cannot be neglected even for nominally low (3%) lattice mismatch and large valence band offset (e.g. 400 meV) systems such as InAs/InP quantum dots. On the other hand, effects of strain (and not the valence band offset) are responsible for the lack of the shell structure for lens-shaped InAs/GaAs quantum dots. The characteristic spectral structure of these systems is to a large degree result of atomistic character of InAs (dot) and GaAs (matrix) material junction. This specific single particle structure leads to a peculiar spectra of positively charged excitons [**29**] in lens-shaped InAs/GaAs quantum dots.

In [**H4**] it has also been shown that the effective depth of the confining potential has an important effect on such spectral features such as the binding energies of charged excitons and the biexciton, and the single exciton fine structure.

Substrate orientation

In the next paper of the series [**H5**] InAs/InP nanowire quantum dots has been studied as a function of nanowire substrate orientation. Nanowire quantum dots are effectively disk-shaped quantum dots, therefore they have high shape-symmetry. Whereas keeping the shape, the spatial orientation of the underlying crystal lattice was modified, which has a profound effect on the excitonic fine structure. In particular, for a typical crystal orientation such as [001] and [111] the bright exciton splitting vanishes on accounts of symmetry [**30**], whereas for [111] orientation the dark exciton splitting vanishes as well.

Further, in [**H5**] the theoretical study was performed for quantum dots “grown” with substrate orientation varying from [112] to [119]. It has been demonstrated, that the change of substrate orientation is assisted by a relatively small effective gap oscillations, while the structure of single particle states remains practically unaltered. On the other hand it has also been shown that, the excitonic fine structure for such unorthodox nanosystems is noticeably different from zero, despite high quantum dot shape symmetry. The bright exciton splitting depends significantly on the substrate orientation reaching maximal value for [112] case. It has been demonstrated that the effect can be attributed to presence of atomic thickness facets at the quantum dot-nanowire interface and the resulting biaxial strain distribution.

Elongated quantum dot spectra

In the following paper of the series [**H6**] both the self-assembled and nanowire InAs/InP quantum dots has been studied. For a disk-shaped self-assembled quantum dots the key role of the wetting layer has been shown. The wetting layer acts effectively as the source of the vertical asymmetry leading to a non-zero bright exciton splitting. It has been also demonstrated that the bright exciton splitting grows with the increasing self-assembled quantum dot height and is inversely proportional to quantum dot diameter. Additionally, in [**H6**] strain has been established as the key factor affecting qualitatively the value of the bright exciton splitting.

The other essential parts of [**H6**] include the extensive studies of the effect of nanostructure shape deformation on its excitonic spectra. The quantum dot base deformation from the cylindrical to ellipsoidal shape along [110] and [1$\underline{1}$0] non-equivalent crystal axes has been thoroughly studied. In simple phenomenological models the shape deformation was considered the key contribution to the bright exciton splitting [**9**]. Such models neglected the asymmetry of the underlying crystal lattice. In [**H6**] has been shown that both contributions are equally important, whereas for certain deformations that contribution due the shape cancels the contribution coming from the lattice asymmetry. In the unexpected (for continuous medium models) way this leads to a cancelation of the bright exciton splitting due to shape elongation. Additionally the degree of shape deformation, for which this reduction occurs, depends significantly on quantum dot size: the fine structure is more prone to deformation effects for a smaller diameter quantum dot.

Paper [**H6**] shows that due to lack of the wetting layer and high shape symmetry, nanowire quantum dot are good candidates for entangled photon generation, this research confirms therefore analogous results obtained by the empirical pseudopotential method [**30**]. However paper [**H6**] shows that effects of non-uniform composition, that exits in realistic $In_xAs_{1-x}P$/InP quantum dots, may present a fundamental obstacle to achieving this goal.

Bright exciton splitting calculations by the empirical tight binding method [**H6**] agree well with experimental data. On the other hand, results obtained by the empirical pseudopotential method [**30**] are typically an order of magnitude smaller than experimental results. Underestimation of the bright exciton splitting by the empirical pseudopotential method reveals certain limits of this approach. This key problem discussed in [**H6**] and has been already noticed and analyzed [**31**] by empirical pseudopotential method developers.

In summary, paper [**H6**] was devoted to a detailed studies of the excitonic fine structure of InAs/InP quantum dots. This work has been noticed and appreciated by *Physical Review B* editors as the „*Editors' choice*".

Light-hole ground state quantum dot

Research on nanowire InAs/InP quantum dots has been continued in the next paper of the series [**H7**]. This work has shown, that with an increasing quantum dot height the character of the ground hole state varies from heavy-hole to light-hole. In effect excitonic states, and in

particular the single exciton fine structure, are modified correspondingly. The ground state of a light-hole exciton is a dark state, the first and the second excited states are quasi-degenerate and are optically active. These states are several hundreds of μeV below the third excited state, which is also a bright state. Recombination from the first and the second excited states is polarized in quantum dot plane, similarly to a typical self-assembled quantum dot spectra. On the contrary, there is a strong emission form the third excited state, which is polarized in the perpendicular direction, i.e. along quantum dot growth axis.

Quantum dots with light-hole excitonic ground state could find important applications in quantum information and telecommunication [**32**,**33**]. My work [**H7**] has proposed growing such systems by a modification of a typical nanowire quantum dot growth. According to my knowledge this was the first paper, apart from the parallel work in Nature Physics [**34**], in the field of excitonic properties of light-hole excitons in quantum dots.

Dark excitons

The next paper of a series focused on the properties of dark excitons confined in InAs/GaAs quantum dots [**H8**]. Both high- and low-shape symmetry systems has been analyzed. As mentioned above, the overall quantum dot symmetry is a combination of a shape symmetry and the underlying lattice symmetry. In case of high shape symmetry systems, such as disk-, lens- or pyramid-shaped quantum dots the overall symmetry is C_{2v}. For these quantum dots the bright exciton spectra consists of two non-degenerate bright states. The light emitted from these states is polarized along [110] and [1$\underline{1}$0] crystal directions. Below the bright states there are two, also non-degenerate, dark states. In a simple effective mass model [**9**] the emission from the dark states by definition is exactly equal to zero. In more elaborate, multi-band models, such as the tight-binding method, effects of light-hole and heavy-hole mixing allow in principle for an optical recombination from one of the dark states. This transition manifests itself by a weak [001] polarized emission. The direction of polarization of this line is thus parallel to quantum dot growth axis. The intensity of [001] polarized dark exciton emission depends strongly on quantum dot shape and reaches about 10^{-7} of bright states intensity for cylindrical quantum dots. For pyramidal quantum dots this emission can reach up to 10^{-4} of bright states emission. The spectral structure of this sort is characteristic for all C_{2v} quantum dot with heavy-hole ground state.

Quantum dots obtained by an epitaxial growth extremely rarely have symmetry close to ideal. Paper [**H8**] studies effects of shape symmetry breaking, by introducing a facet between the quantum dots and the host material. The corresponding group symmetry is C_s. This symmetry reduction does not alter significantly spectral properties of bright states, however is has a fundamental effect on dark excitons spectra. In C_s quantum dots dark excitons gain pronounced non-zero oscillator strengths due to effects of mixing with bright excitons. Therefore both nominally “dark” states become unusually optically active and emit laterally polarized light (the same polarization as that of bright states). This emission is highly anisotropic, and the brighter of “dark” lines reaches even up to 1/1500 of bright excitons intensity. The theoretically calculated spectra and polarization properties are in a qualitative agreement with experimental data [**P12**]. The discussed results show that dark excitonic states

can be directly accessed in a fully optical experiment, with potential applications as a long-lived, charged neutral solid state qubits.

Chemical composition fluctuations

The chemical composition of realistic quantum dots differs significantly from an idealized model assumptions. This is particularly true for self-assembled $In_xGa_{1-x}As/GaAs$ quantum dots where the contribution from gallium atoms (surrounding matrix atoms) can reach more than fifty percent.

Effects of composition non-homogeneity were examined, in a different context, in several papers of the series discussed so far. In paper [**H6**] the effect of composition fluctuations on fine structure of elongated InAs/InP quantum dot was analyzed. This paper showed, that composition fluctuations reduce the bright exciton splitting for highly deformed quantum dots. On the other hand for cylindrical quantum dots, such a nanowire quantum dots, it is the composition non-homogeneity that gives the main contribution to the bright exciton splitting. In the following papers of the series [**H7**,**H8**] it has been shown that the light-hole exciton and the dark exciton spectra are to a certain degree resistant to charge fluctuations. Nevertheless, the effect of composition non-homogeneity profoundly affects the qualitative character of excitonic spectra, in particular energy splittings and line polarizations.

An extreme example of nanosystems which formation and spectral properties are driven by the composition non-homogeneity are natural quantum dots [**35**]. These dots are formed as composition fluctuations of the wetting layer. Paper [**H9**] presents theoretical and experimental studies of excitonic properties of such systems. This work shows that, whereas the main excitonic emission energy rather weakly depends on composition fluctuations, properties of other excitonic complexes are affected significantly. Composition non-homogeneity influences the biexciton and trions (X^-, X^+) binding energies profoundly. Paper [**H9**] shows experimental and theoretical studies of relations between binding energies of these complexes, what in certain cases can lead to a reversal of excitonic lines. Further, paper [**H9**] shows fundamental limits, due to composition fluctuations, for methods of quantum dot geometry prediction based on quantum dot spectral properties (so-called *inverse approaches* [**36**]).

SiO_2 shell

Similar to natural atoms, quantum dot („artificial atom") properties can be dynamically modified by a post-growth application of external electric and magnetic fields. Apart the above, manmade nanostructure properties can be altered by the application of external stress, resulting in an effective external strain field [**P8**,**P9**]. The externally applied deformation can be achieved by means of mechanical tools such as piezoelectric actuators. Another possible approach, available for nanowires and nanowire quantum dots, is based on a post-growth of an additional layer (shell) of a lattice mismatched material.

Paper [**H10**] presents experimentally measured spectra and theoretical calculations of optical properties of $InAs_xP_{1-x}$ nanowire quantum dots under external strain due to a post-growth of a

lattice mismatched SiO_2 shell. Apart from the external strain due to the outer shell, there exists an internal strain due to quantum dot and nanowire lattice mismatch. Most importantly these two kind of strains do not combine with each other in a simple, additive way.

In paper [**H10**] atomistic theory was successfully applied to estimate the character of combined internal/external strain and the magnitude of excitonic spectra shifts due to external strain. Atomistic calculation give a clue on how the post-growth process can lead to a nanosystem design of desired specifications. The SiO_2 layer allows for a large degree of controlled excitonic emission energy change from -116 meV to 55 meV [**H10**] with respect to a system without the SiO_2 shell. Moreover the shell growth is reversible: the shell can be removed by etching and the nanowire quantum dot spectral properties are virtually the same as before the shell growth. Such degree of control could be useful for potential quantum dots and nanowires applications in telecommunication and quantum cryptography.

ZnTe/ZnMgTe core-shell nanowires

Apart from typical III-V nanostructures, there is an ongoing investigation on II-VI materials based nanosystems. Polish scientific facilities are one of the leading in this research. The last paper of the discussed series [**H11**] was created in collaboration with experimental groups from the Institute of Physics Polish Academy of Sciences (IF PAN) and the Institute of Fundamental Technological Research (IPPT PAN).

Paper [**H11**] studies effects of $Zn_{1-x}Mg_xTe$ shell on excitonic emission from a ZnTe nanowire core. The theory applied in [**H11**] used similar methodology to that already developed during the earlier research [**H10**] on InP nanowires embedded in SiO_2 shell. Paper [**H11**] demonstrates, that both the increase of Mg shell content, and the increase of the shell thickness reduce the energy of the emission from the ZnTe nanowire core. The reduction of the effective core band gap is due to tensile strain, originating from the lattice mismatched shell. By a certain choice of growth parameters (and therefore the shell thickness and composition) it is possible to control the excitonic energy shift in a quasi-continuous way down to -120 meV as compared to the unstrained system. Results of theoretical calculations are in qualitative and quantitative agreement with the experiment.

Summary

The presented series of publications describes the development and implementation of theoretical atomistic approach aiming to calculate excitonic spectra for nanostructures containing million atoms and more [**H1**-**H11**]. Papers presented in the series were able to address numerous important questions related to semiconductor quantum dot excitonic spectra, including excitonic complexes binding energies, and the excitonic fine structure. These papers have demonstrated a fundamental role of shape, lattice symmetry, and chemical composition on the bright and dark exciton spectra. Understanding the physical background of the bright exciton splitting, and possible mechanisms of its reduction are of fundamental importance for potential quantum dot applications in the biexciton-exciton cascade scheme of entangled photon generation. On the other hand research on dark excitons may lead to their applications as qubits in quantum information.

In particular it has been shown that:

- Atomistic calculations for nanostructures involve a multi-stage [**H1**] and a multi-scale [**H3**] approach, and solving many crucial numerical issues. In the first stage of the calculation atomic positions are found by minimizing the total elastic energy of the system. In the second stage the single particle calculation is performed by means of the empirical tight binding method including the *d*-orbitals and the spin-orbit effect. In the final step, the many-body calculation gives energy and optical spectra of various excitonic complexes. All algorithms utilized in the series of publications were developed and implemented by the author himself. In particular, I have implemented a highly-efficient, parallelized versions of computer codes aiming for strain, the single particle and the many-body calculation.
- The diagonal parameters correction is a necessary contribution to the $sp^3d^5s^*$ tight binding model [**H2**] and must be accounted for the quantitative and qualitative agreement with the experiment. This applies to both strained bulk and nanostructures.
- The valence band offset has been established as one of the most important empirical parameters affecting the precision of nanostructure calculations [**H4**].
- Strain effects cannot be neglected even for systems with a small (3%) lattice mismatch. Strain effects are responsible for the characteristic shell-like structure of self-assembled InAs/InP disk-shaped and lens-shaped quantum dots [**H4**]. On the other hand, strain effects are responsible for the vanishing shell structure of hole states in InAs/GaAs lens-shaped quantum dots.
- Substrate orientation has an essential effect [**H5**] on excitonic spectra of InAs/InP nanowire quantum dots. This effect can be attributed to low underlying lattice symmetry and the atomic thickness facets at the quantum dot and nanowire interface. Such effects can be accounted for only by an atomistic approach such as that of [**H5**].
- The contribution to the bright exciton splitting coming from the quantum dot shape elongation can have opposite sign to the lattice asymmetry influence [**H6**]. This, in contrary to simple models, leads to the bright exciton splitting („anisotropic splitting") reduction due to shape deformation. The reduction of this splitting and utilization of

the biexciton-exciton cascade, allows in principle for the entangled photon generation. Paper [**H6**] has been awarded as *Physical Review B* „*Editors' choice*".

- High aspect ratio (tall) nanowire quantum dots can exhibit light-hole excitonic ground state [**H7**]. Paper [**H7**] was the first to discuss properties of light-hole excitons confined in semiconductor quantum dots. Nanostructures of such properties could find novel applications in information technology and telecommunications.
- Low quantum dot shape symmetry can lead to bright and dark excitons mixing [**H8**]. Dark excitons gain non-zero oscillator strengths due to mixing, whereas their emission is highly anisotropic and show strong laterally polarized component. These results has been recently confirmed by an experiment and could be considered as a stepping stone towards dark excitons manipulation by purely optical means. Dark excitons can effectively form a long-lived, charge neutral qubits with potential applications in quantum information.
- Nanowire quantum dots due to high symmetry, e.g. lack of the wetting layer and [111] (or [001]) substrate orientation, are good candidates for entangled photon generation [**H5**,**H6**]. On the other hand chemical composition fluctuations, which are always present in real quantum dots, effectively lower the overall quantum dot symmetry. The composition non-homogeneity therefore constitutes a lower limit for the bright exciton splitting [**H6**]. Results presented in my paper agree well with experimental data.
- Composition fluctuations have dominant effect on wetting layer quantum dots (natural quantum dots) excitonic spectra. In particular they affect, excitonic binding energies up to a point of excitonic lines reversal [**H9**]. Again, this effect has been confirmed by an experimental study.
- Lattice mismatched nanowire shells allow for a wide range control of excitonic emission energy of InAs/InP nanowire quantum dots [**H10**] and ZnTe nanowires [**H11**]. The key factor responsible for the modification of the emission energy is tensile strain due to the nanowire shell. Results of theoretical calculations are in qualitative and quantitative agreement with the experiment, whereas the large degree of strain control could be useful for quantum dots and nanowires applications in telecommunication or quantum cryptography.

The series consists of total 11 papers, including 6 single author papers, e.g. 3 *Physical Review B* papers, and one *J. Phys.: Condens. Matter* paper and one *Nanoscale Research Letters* paper. The first paper of the series (*Physical Review B*) was a result of my post-doctoral fellowship in the National Research Council of Canada. The rest of the papers where created in collaboration with experimental groups from both Poland (*Applied Physics Letters* and *Physical Review B* papers) and abroad. In particular the paper published in *Nano letters* was an effect of collaboration with a group of prof. Val Zwiller from Delft Technical University (*TU Delft*), whereas one of the *Physical Review B* papers was created in collaboration with prof. David Gershoni from *Israel Institute of Technology* (*Technion*).

Apart from the papers in the presented series, I find two papers particularly important. The first is the *Physical Review Letters* paper created in collaboration with a group of dr. G. W. Bryant from *NIST* (*National Institute of Standards and Technology*); the second is the recently published *Physical Review X* paper created in collaboration with the group of prof. David Gershoni from *Technion*. A more detail discussion of these and my other scientific achievements beyond the scope of the habilitation series can be found in the next chapter.

VI. Other scientific achievements

a. List of publications (after the PhD) not included in section V

[**P1**] W. Jaskólski, M. Zieliński, G.W. Bryant, J. Aizpurua
Strain effects on the electronic structure of strongly coupled self-assembled InAs/GaAs quantum dots: Tight-binding approach,
Phys. Rev. B **74**, 195339 (2006)
Impact factor in 2006: 3.107

[**P2**] J. Diaz, M. Zieliński, W. Jaskólski, G.W. Bryant
Tight-binding theory of ZnS/CdS nanoheterostructures. The role of strain and d orbitals,
Phys. Rev. B **74**, 205309 (2006)
Impact factor in 2006: 3.107

[**P3**] J.G. Diaz, W. Jaskolski, M. Zieliński, G.W. Bryant
Pressure-induced optoelectronic properties of InP nanocrystals: Tight-binding approach,
Phys. Stat. Solidi C **3**, 3832 (2006)
Impact factor in 2006: 1.221

[**P4**] J.G. Diaz, G.W. Bryant, W. Jaskolski, and M. Zieliński
Theory of InP nanocrystals under pressure,
Phys. Rev. B **75**, 245433 (2007)
Impact factor in 2007: 3.172

[**P5**] M. Chwastyk, P. Różański, and M. Zieliński,
Atomistic Calculation of Coulomb Interactions in Semiconductor Nanocrystals: Role of Surface Passivation and Composition Details,
Acta. Phys. Pol. A **122**, 324 (2012)
Impact factor in 2012: 0.531

[**P6**] E. S. Kadantsev, M. Zieliński, M. Korkusiński, and P. Hawrylak
Ab initio calculation of band edges modified by (001) biaxial strain in group IIIA-VA and group IIB-VIA semiconductors:
Application to quasi-particle energy levels of strained InAs/InP quantum dot,
J. Appl. Phys. **107**, 104315 (2010)
Impact factor in 2010: 2.064

[**P7**] E. S. Kadantsev, M. Zieliński, and P. Hawrylak,
Band engineering in nanowires: Ab initio model of band edges modified by (111) biaxial strain in group IIIA-VA semiconductors,
Phys. Rev. B **86**, 085411 (2012)
Impact factor in 2012: 3.767

[**P8**] G.W. Bryant, M. Zieliński, N. Malkova, J. Sims, W. Jaskolski, and J. Aizpurua
Effect of mechanical strain on the optical properties of quantum dots: Controlling exciton shape, orientation, and phase with a mechanical strain,
Phys. Rev. Letters **105**, 067404 (2010)
Impact factor in 2010: 7.621

[**P9]** G.W. Bryant, M. Zieliński, N. Malkova, J. Sims, W. Jaskólski, J. Aizpurua,
Controlling the optics of quantum dots with nanomechanical strain,
Phys. Rev. B **84**, 235412 (2011)
Impact factor in 2011: 3.691

[**P10]** M. Korkusiński, P. Hawrylak, M. Zieliński, W. Sheng, G. Klimeck
Building semiconductor nanostructures atom by atom,
Microelectronics Journal **39**, 318 (2008)
Impact factor in 2008: 0.787

[**P11**] M. Korkusiński, M. Zieliński, and P. Hawrylak,
Multiexciton complexes in InAs self-assembled quantum dots,
J. Appl. Phys. **105**, 122406 (2009)
Impact factor in 2009: 2.072

[**P12**] W. D. Sheng, M. Korkusinski, A. D. Guclu, M. Zieliński, P. Potasz, E. Kadantsev, O. Voznyy, and P. Hawrylak,
Electronic and optical properties of semiconductor and graphene quantum dots,
Frontiers of Physics **7**, 328 (2012)
Impact factor in 2012: 1.591

[**P13**] I. Schwartz, E. R. Schmidgall, L. Gantz, D. Cogan, E. Bordo, Y. Don, M. Zielinski, and D. Gershoni,
Deterministic Writing and Control of the Dark Exciton Spin Using Single Short Optical Pulses,
Phys. Rev. X **5**, 011009 (2015)
Impact factor: 8.463

[**P14**] F. Delgado, S. Loth, M. Zielinski, and J. Fernandez-Rossier,
The emergence of classical behaviour in magnetic adatoms,
EPL (Europhysics Letters), **109**, 57001 (2015)
Impact factor: 2.269

b. Discussion of other scientific achievements

Apart from publications discussed above, and papers published before my PhD, my remaining scientific activity was related to different aspects of nanostructure physics. These papers were excluded from the habilitation series because, they were conducted using methodology that I had developed during my PhD [**P1**,**P8**,**P9**], they were related to different subjects [**P2**-**P7**], they had mostly a review character [**P10**-**P12**] or they were devoted to studies of effects [**P13,P14**] different from the focus of the series.

Paper [**P1**] has concentrated on properties of double InAs/GaAs quantum dots ("quasi-molecules") as a function of inter-dot separation. This paper has shown e.g. the change of the "molecular" ground hole state character from the bonding to anti-bonding with decreasing quantum dots separation. This effect has been confirmed by an independent experimental and theoretical research (e.g. Nature Physics **4,** 291 from 2008); paper [**P1**] has now 48 citations (Web of Science). In the next paper [**P2**] the single particle properties of core-shell ZnS/CdS nanocrystals were analyzed, whereas [**P3**] and [**P4**] studied band gap evolution of InP nanocrystals under external pressure. The subject of nanocrystals was once again present in [**P5]**, where the role of surface passivation and chemical composition was studied in InAs and GaAs nanocrystals.

Another set of papers is a mini-series of two papers: [**P6**] and [**P7**] discussing results of *ab-initio* calculations of semiconductor bands under biaxial strain along [001] direction ([**P6**]; III-V and II-VI semiconductors) and along [111] direction ([**P7]**; III-V semiconductors). Both papers present calculations of deformation potentials and band edges evolution under bond length deformation. These dependencies were published in a functional form allowing for straight application in nanostructure calculations. In both papers the role of strain effects was illustrated on the example of quantum dots.

Another mini-series consists of [**P8**] and [**P9**] papers. These works (and also [**P1**-**P4**]) are a result of a long term collaboration with dr. G.W. Bryant from *NIST*. In these papers we have studied a single particle and many-body properties of quantum dots embedded in nanocantilevers. Among these articles I find [**P8**] published in the *Physical Review Letters* particularly important. This was the first paper showing the effect of external strain on the excitonic fine structure in InAs/GaAs quantum dots.

The next three papers [**P10**-**P12**] have mostly a review-like character. Paper [**P10**] discusses atomistic way of calculating strain effects. The research was illustrated on the example of several vertically stacked quantum dots. Paper [**P11**] discusses different results obtained for quantum dots obtained by the effective mass approximation, the **kp** method, and the atomistic approach. Finally [**P12**] is a review paper discussing numerous results and theoretical methods related to self-assembled and graphene quantum dots.

The next paper outside of the habilitation series is [**P13**] published recently in the *Physical Review X*. This mainly experimental work has demonstrated a possibility of deterministic dark exciton generation in a well-defined spin state by a single optical pulse. This effective qubit can be efficiently controlled by a series of short optical pulses (orders of magnitude shorter

than qubit lifetime and coherence time). Due to possibility of integration of quantum dots with photonic components, qubits based on dark excitons could have important application in quantum information.

The last of the papers **[P14]** was recently published in the EPL (Europhysics Letters). This work studies many-body properties of nanomagnets formed for by a chain of magnetic adatoms on a conducting surface. The paper deals in particular with a quantum state (zero magnetization) to a classical state (non-zero magnetization) transition. In particular effects of itinerant electrons or other localized spins have been thoroughly studied. Finally, it has been pointed out that magnetic adatoms chains are ideal systems to explore the quantum to classical transition experimentally.

Bibliometric data

ResearcherID: C-2587-2013

Total citations according to Web of Science (WoS): 240

Hirsch index (WoS): 9

Total number of publications: 32

Average citations per publication: 7.5

Bibliography

[**1**] L. Jacak, P. Hawrylak, and A. Wojs,
Quantum Dots,
Springer, Berlin (1998).

[**2**] P. Michler, A. Kiraz, C. Becher, W. V. Schoenfeld, P. M. Petroff, L. Zhang, E. Hu, A. Imamoglu,
A Quantum Dot Single-Photon Turnstile Device,
Science **290**, 2282 (2000)

[**3**] A. J. Shields,
Semiconductor quantum light sources,
Nature Photonics **1**, 215 (2007)

[**4**] O. Benson, C. Santori, M. Pelton, Y. Yamamoto,
Regulated and Entangled Photons from a Single Quantum Dot,
Phys. Rev. Lett. **84**, 2513 (2000)

[**5**] R. M. Stevenson, R. J.Young, P. Atkinson, K. Cooper, D. A. Ritchie, A. J. Shields,
A semiconductor source of triggered entangled photon pairs,
Nature **439**, 179 (2006)

[**6**] N. Akopian, N. H. Lindner, E. Poem, Y. Berlatzky, J. Avron, D. Gershoni, B. D. Gerardot, P. M. Petroff,
Entangled Photon Pairs from Semiconductor Quantum Dots,
Phys. Rev. Lett. **96**, 130501 (2006)

[**7**] J. Y. Marzin, J. M. Gérard, A. Izraël, D. Barrier, G. Bastard,
Photoluminescence of Single InAs Quantum Dots Obtained by Self-Organized Growth on GaAs,
Phys. Rev. Lett. **73**, 716 (1994)

[**8**] E. Dekel, D. Gershoni, E. Ehrenfreund, D. Spektor, J. M. Garcia, and P. M. Petroff,
Multiexciton Spectroscopy of a Single Self-Assembled Quantum Dot,
Phys. Rev. Lett. **80**, 4991 (1998)

[**9**] M. Bayer, G. Ortner, O. Stern, A. Kuther, A. A. Gorbunov, A. Forchel, P. Hawrylak, S. Fafard, K. Hinzer, T. L. Reinecke, S. N. Walck, J. P. Reithmaier, F. Klopf, and F. Schafer,
Fine structure of neutral and charged excitons in self-assembled In(Ga)As/(Al)GaAs quantum dots,
Phys. Rev. B **65**, 195315 (2002)

[**10**] E. Poem, Y. Kodriano, C. Tradonsky, N. H. Lindner, B. D. Gerardot, P. M. Petroff & D. Gershoni,
Accessing the dark exciton with light,
Nature Physics **6**, 993 (2010)

[**11**] M. H. M. van Weert, N. Akopian, U. Perinetti, M. P. van Kouwen, R. E. Algra, M. A. Verheijen, E. P. A. M. Bakkers, L. P. Kouwenhoven, V. Zwiller,
Selective Excitation and Detection of Spin States in a Single Nanowire Quantum Dot,
Nano Lett. **9**, 1989 (2009)

[**12**] Dan Dalacu, K. Mnaymneh, X. Wu, J. Lapointe, G. C. Aers, P. J. Poole, R. L. Williams,
Selective-area vapor-liquid-solid growth of tunable InAsP quantum dots in nanowires,
Appl. Phys. Lett. **98**, 251101 (2011)

[**13**] D. Dalacu, K. Mnaymneh, J. Lapointe, X. Wu, P. J. Poole, G. Bulgarini,V. Zwiller, M. E. Reimer,
Ultraclean Emission from InAsP Quantum Dots in Defect-Free Wurtzite InP Nanowires,
Nano Lett. **12**, 5919 (2012)

[**14**] L. W. Wang, A. Zunger,
Linear combination of bulk bands method for large-scale electronic structure calculations on strained nanostructures,
Phys. Rev. **B** 59, 15806 (1999)

[**15**] A. J. Williamson, L. W. Wang, A. Zunger,
Theoretical interpretation of the experimental electronic structure of lens-shaped self-assembled InAs/GaAs quantum dots,
Phys. Rev. B **62**, 12963 (2000)

[**16**] K. Leung, K. Whaley,
Electron-hole interactions in silicon nanocrystals,
Phys. Rev. B **56**, 7455 (1997)

[**17**] S. Lee, L. Jonsson, J. W. Wilkins, G. W. Bryant, and G. Klimeck,
Electron-hole correlations in semiconductor quantum dots with tight-binding wave functions,
Phys. Rev. B **63**, 195318 (2001)

[**18**] R. Santoprete, B. Koiller, R. B. Capaz, P. Kratzer, Q. K. K. Liu, M. Scheffler,
Electron-hole correlations in semiconductor quantum dots with tight-binding wave functions,
Phys. Rev. B **68**, 235311 (2003)

[**19**] W. Sheng, S.J. Cheng, P. Hawrylak,
Multiband theory of multi-exciton complexes in self-assembled quantum dots,
Phys. Rev. B **71**, 035316 (2005)

[**20**] P. N. Keating,
Effect of Invariance Requirements on the Elastic Strain Energy of Crystals with Application to the Diamond Structure,
Phys. Rev. **145**, 637 (1966)

[**21**] R. M. Martin,
Elastic Properties of ZnS Structure Semiconductors,
Phys. Rev. B **1**, 4005 (1970)

[**22**] D. J. Chadi,
Spin-orbit splitting in crystalline and compositionally disordered semiconductors,
Phys. Rev. B **16**, 790 (1977)

[**23**] J.M. Jancu, R. Scholz, F. Beltram, F. Bassani,
Empirical spds tight-binding calculation for cubic semiconductors: General method and material parameters*,
Phys. Rev. B **57**, 6493 (1998)

[**24**] W. A. Harrison,
Electronic Structure and the Properties of Solids,
(Freeman, New York, 1980).

[**25**] J. C. Slater, G. F. Koster,
Simplified LCAO Method for the Periodic Potential Problem,
Phys. Rev. **94**, 1498 (1954)

[**26**] T. B. Boykin, G. Klimeck, R. C. Bowen, F. Oyafuso,
Diagonal parameter shifts due to nearest-neighbor displacements in empirical tight-binding theory,
Phys. Rev. B **66**, 125207 (2002)

[**27**] Jane K. Cullum, Ralph A. Willoughby,
Lanczos Algorithms for Large Symmetric Eigenvalue Computations,
Progress in Scientific Computing (1985)

[**28**] W. E. Arnoldi,
The principle of minimized iterations in the solution of the matrix eigenvalue problem,
Quarterly of Applied Mathematics **9**, 17 (1951).

[**29**] M. Ediger, G. Bester, A. Badolato, P. M. Petroff, K. Karrai, A. Zunger, R. J. Warburton,
Peculiar many-body effects revealed in the spectroscopy of highly charged quantum dots,
Nature Physics **3**, 774 (2007)

[**30**] R. Singh, G. Bester,
Nanowire Quantum Dots as an Ideal Source of Entangled Photon Pairs,
Phys. Rev. Lett. **103**, 063601 (2009)

[**31**] R. Singh, G. Bester,
Effects of atomic ordering on the electronic and optical properties of self-assembled $In_xGa_{1-x}As/GaAs$ semiconductor quantum dots,
Phys. Rev. B **84**, 241402 (2011)

[**32**] D. Sleiter and W. F. Brinkman,
Using holes in GaAs as qubits: An estimate of the Rabi frequency in the presence of an external rf field,
Phys. Rev. B **74**, 153312 (2006)

[**33**] R. Vrijen, E. Yablonovitch,
A spin-coherent semiconductor photo-detector for quantum communication,
Physica E **10**, 569 (2001)

[**34**] Y. H. Huo, B. J. Witek, S. Kumar, J. R. Cardenas, J. X. Zhang, N. Akopian, R. Singh, E. Zallo, R. Grifone, D. Kriegner, R. Trotta, F. Ding, J. Stangl, V. Zwiller, G. Bester, A. Rastelli, O. G. Schmidt,
A light-hole exciton in a quantum dot,
Nature Physics **10**, 46 (2014)

[**35**] A. Babiński, J. Borysiuk, S. Kret, M. Czyż, A. Golnik, S. Raymond, Z. R. Wasilewski,
Natural quantum dots in the InAs/GaAs wetting layer,
Appl. Phys. Lett. **92**, 171104 (2008)

[**36**] V. Mlinar, M. Bozkurt, J. M. Ulloa, M. Ediger, G. Bester, A. Badolato, P. M. Koenraad, R. J. Warburton, and A. Zunger,
Structure of quantum dots as seen by excitonic spectroscopy versus structural characterization: Using theory to close the loop,
Phys. Rev. B **80**, 165425 (2009)