VILNIUS UNIVERSITY
State Research Institute of Center for Physical
Sciences and Technology

Andrius
RIMKUS

Optical properties of InGaAs heterostructures

SUMMARY OF DOCTORAL DISSERTATION

Natural Sciences
Physics **N 002**

VILNIUS 2019

This dissertation was written between 2014 and 2018 in Center for Physical Sciences and Technology, Vilnius, Lithuania.

Academic supervisor – dr. Bronislovas Čechavičius (Center for Physical Sciences and Technology, Natural Sciences, Physics **N002**).

Academic consultant – dr. Ramūnas Nedzinskas (Center for Physical Sciences and Technology, Natural Sciences, Physics **N002**).

This doctoral dissertation will be defended in a public meeting of the Dissertation Defence Panel:
Chairman – **prof. dr. Nerija Žurauskienė** (Center for Physical Sciences and Technology, Natural Sciences, Physics **N002**).
Members: (members listed in alphabetical order of surnames)
dr. Ramūnas Adomavičius (Center for Physical Sciences and Technology, Natural Sciences, Physics **N002**);
dr. Aurimas Čerškus (Center for Physical Sciences and Technology, Natural Sciences, Physics **N002**);
dr. Renata Karpič (Center for Physical Sciences and Technology, Natural Sciences, Physics **N002**);
prof. dr. Saulius Marcinkevičius (KTH Royal Institute of Technology, Natural Sciences, Physics **N002**).

The dissertation shall be defended at a public meeting of the Dissertation Defence Panel at 14 hour on 31st of may 2019 in the Conference hall of Semiconductor Physics Institute (Center for Physical Sciences and Technology), Saulėtekio ave. 3, LT-10257 Vilnius, Lithuania.

The text of this dissertation can be accessed at the libraries of of Vilnius University and Center for Physical Sciences, as well as on the website of Vilnius University: *https://www.vu.lt/naujienos/ivykiu-kalendorius*

VILNIAUS UNIVERSITETAS

VALSTYBINIS MOKSLINIŲ TYRIMŲ

INSTITUTAS
FIZINIŲ IR TECHNOLOGIJOS MOKSLŲ
CENTRAS

Andrius
RIMKUS

Gamtos mokslai, fizika (N002)

Vilnius, 2019

Disertacija rengta 2014–2018 metais VMTI Fizinių ir technologijos mokslų centre.

Mokslinis vadovas:
Dr. Bronislovas Čechavičius (VMTI Fizinių ir technologijos mokslų centras, gamtos mokslai, fizika – N002).

Mokslinis konsultantas:
Dr. Ramūnas Nedzinskas (VMTI Fizinių ir technologijos mokslų centro, gamtos mokslai, fizika – N002).

Disertacija ginama viešame gynimo tarybos posėdyje.

Pirmininkė:
Prof. dr. Nerija Žurauskienė (VMTI Fizinių ir technologijos mokslų centras, gamtos mokslai, fizika – N002).

Nariai:
Dr. Ramūnas Adomavičius (VMTI Fizinių ir technologijos mokslų centras, gamtos mokslai, fizika – N002),

Dr. Aurimas Čerškus (VMTI Fizinių ir technologijos mokslų centras, gamtos mokslai, fizika – N002),

Dr. Renata Karpič (VMTI Fizinių ir technologijos mokslų centras, gamtos mokslai, fizika – N002),

Dr. Saulius Marcinkevičius (KTH Karališkasis technologijos institutas, gamtos mokslai, fizika – N002).

Turinys

RELEVANCE OF THE THESIS

The emission of InAs/GaAs quantum dots (QD) is usually registered around 1.1 µm. In order to adapt these quantum structures to the needs of modern solar cells [1, 2] and telecommunication [3], they can be capped with InGaAs layer to reduce surface tension field and to redshift the energy of the quantum dots optical transitions. This architecture of quantum structures makes it possible to change their material composition and optical response by choosing parameters of the quantum well (QW) surrounding the dots. In this dissertation, temperature dependent spectroscopic study of InAs quantum dots grown without and with strain relieving InGaAs layer is presented.

Semiconductor quantum dots as aforementioned are great candidates for various applications in optoelectronics. Innovative epitaxial nanostructures — quantum rods (QRods) consists of vertically elongated InGaAs quantum dots immersed in InGaAs/GaAs QW. The combined carrier confinement in QRods leads to a variety of novel properties with respect to regular QDs, showing features of the pure QD, pure QW and mixed electronic states. Besides, the bound states could also appear in region of QW continuum states [5]. As a consequence, interesting optical and carrier dynamics properties could arise in QRods [4], both for the fundamental physics and for potential applications in optoelectronics. Keeping this in mind, QRods have been grown in a molecular beam epitaxial reactor using an As_2 or As_4 flux. Thus quantum structures were grown comprised of In-rich InGaAs QRods surrounded by a 2-D InGaAs QW. In this work the influence of As source and QRods height on interband optical transitions in InGaAs quantum rod and the surrounding InGaAs QW was investigated using temperature dependent spectroscopic techniques. Used spectroscopic techniques was supported by the numerical calculations, and has given an insight into the physical and optical properties of the structures studied.

Self-forming semiconductor structures, due to their shape engineering by controlling their electronic and optical properties, attract a lot of scientists. In this work we discuss another morphology of such structures — quantum rings (QRings). These structures have already been investigated experimentally and theoretically, but for the proper nanostructure to be grown it is crucial to understand the influence of their growing conditions and the effects of capping layer on the structure itself. In this work, optical properties of InAs QRings were studied. Several ring structures grown with different thickness of GaAs capping layer were tested to reveal size-dependent optical response of the QRings. Also hybrid nanostructure containing InAs QRings and InAs/InGaAs quantum dots capped with InGaAs layer was examined and characterized.

Work objective

To investigate the nature of optical transitions and electronic structure in InGaAs quantum dot systems of different design and to detect optical, quantum mechanical, etc. parameters relevant to the theoretical modeling of these quantum structures and technological optimization by using modulation spectroscopy, photoluminescence, and photoluminescence excitation techniques. This work includes:

- characterization and investigation of interband optical transitions of different morphology structures in a wide temperature range using modulated reflectance, photoluminescence, and photoluminescence excitation spectroscopy techniques;
- analysis of morphology and surrounding environment effects on electronic states and optical properties of studied nanostructures.

Scientific novelty of the thesis

In this work, a complex study of quantum dots, quantum rods and quantum rings using sensitive modulation spectroscopy and photoluminescence techniques in the 3–300 K temperature range was performed with different excitation energy and power. Analysis of InAs/InGaAs/GaAs quantum dot heterostructures revealed:

- photoreflectance mechanisms, determining peculiarities of quantum dot electronic states and internal electric field modulation, as well as reflectance spectra associated with reflection from GaAs substrate.
- influence of InGaAs capping layer on the InAs quantum dots electron energy spectrum, light emission efficiency and structural morphology.

Study of InGaAs quantum rods of various morphology surrounded by InGaAs quantum well and embedded in GaAs revealed:

- influence of As source used (As_2 and As_4) during the epitaxial growth on electronic structure and optical properties of quantum rods;
- effects of quantum rod height on optical properties and carrier confinement.

InAs GaAs quantum rings revealed:

- influence of InAs and GaAs layer thickness on optical properties and morphology of quantum rings.
- Also unique hybrid InAs/InGaAs/GaAs quantum dot and InAs/GaAs quantum ring structure was investigated and characterized.

Nanostructures studied

In this work, several quantum structures for THz region detectors and optical amplifiers were designed:

1. InAs quantum dots with and without InGaAs capping layer surrounded by GaAs/AlAs quantum wells (QDs);
2. InGaAs quantum rods embedded in InGaAs/GaAs quantum wells (QRods);
3. InAs quantum Rings in GaAs matrix (QRings) and hybrid quantum ring and quantum dot structures.

Practical value

Spectroscopy and calculations of quantum states reveal optical properties, absorption and emission spectrum dependencies of these quantum systems, reflecting features of the electronic structure and its relationship with morphology of nanostructure. It is critical information for the optimization, cultivation and development of new modern optoelectronic technologies and devices.

There are following statements presented for defense:

Quantum dots

I. Changes in the electronic structure of InAs quantum dots covered with InGaAs are caused by a decrease in tension, a lower material decomposition and an increase in dot height.

II. The insertion of the InGaAs capping layer reduces the spectral linewidth of the InAs quantum dots and eliminates the Stokes shift, so the quantum dots in the InAs/InGaAs/GaAs heterostructure are more homogeneous.

Quantum rods

I. Using As_4 molecular flux results in deeper quantum well than using As_2.

II. Transition between zero-dimensional and one-dimensional structures is achieved by forming higher quantum rod nanostructures.

Quantum rings

I. InAs quantum rings grown by partly covering quantum dots with 2 nm GaAs layer have a lower energy difference between bound state and continuum compared to quantum dots or quantum rod structures.

List of Publications

The dissertation is based on the following publications:

1. R. Nedzinskas, B. Čechavičius, A. Rimkus, J. Kavaliauskas, G. Valušis, L. Li, E. H. Linfield, *Optical Features of InAs Quantum Dots-in-a-well Structures*, Lith. J. Phys. **54**, 54–57 (2014).

2. R. Nedzinskas, B. Čechavičius, A. Rimkus, E. Pozingytė, J. Kavaliauskas, G. Valušis, L. Li, E. H. Linfield, *Temperature-dependent modulated reflectance of InAs/InGaAs/GaAs quantum dots-in-a-well infrared photodetectors,* J. Appl. Phys, **117**, 144304 (2015).

3. A. Rimkus, E. Pozingytė, R. Nedzinskas, B. Čechavičius, J. Kavaliauskas, G. Valušis, L. Li, E. H. Linfield, *Temperature-dependent modulated reflectance and photoluminescence of InAs–GaAs and InAs–InGaAs–GaA quantum dot heterostructures*, Opt. Quantum Electron., **48**(3) (2016).

The scientific results were reported in the following conferences:

1. E. Pozingytė, **A. Rimkus**, R. Nedzinskas, *Temperature dependent photoreflectance spectroscopy of*

InAs dots-in-a-well structures, Open Readings 2015, 2015.03.24-27, Vilnius, Lietuva.

2. **A. Rimkus**, E. Pozingytė, R. Nedzinskas, B. Čechavičius, *Comparative spectroscopic study of InAs dots-in-a-well quantum structures with / without ingaas cap layer*, Open Readings, 2015.03.24-27, Vilnius, Lietuva.

3. **A. Rimkus**, E. Pozingytė, R. Nedzinskas, B. Čechavičius, *Temperature dependent spectroscopic study of InAs dots-in-a-well structures with InGaAs capping layer*, OPTO2015 OPTO-Meeting for Young Researchers & 10th Anniversary International SPIE Student Chapter Meeting, 2015.05.27-30, Wroclaw, Poland.

4. E. Pozingytė, **A. Rimkus**, R. Nedzinskas, V. Karpus, B. Čechavičius, J. Kavaliauskas, G. Valušis, *Analitinis modelis cilindrinių kvantinių taškų ir kvantinių strypelių elektronų energijos spektro skaičiavimams*, 41-oji Lietuvos Nacionalinė Fizikos Konferencija, 2015.06.17-19, Vilnius, Lietuva.

5. **A. Rimkus**, E. Pozingytė, R. Nedzinskas, B. Čechavičius, J. Kavaliauskas, G. Valušis, L. Li, E. H. Linfield, *Epitaksinių InAs kvantinių žiedų fotoatspindžio ir fotoliuminescencijos tyrimas*, 41-oji Lietuvos Nacionalinė Fizikos Konferencija, 2015.06.17-19, Vilnius, Lietuva.

6. R. Nedzinskas, **A. Rimkus**, E. Pozingytė, B. Čechavičius, J. Kavaliauskas, G. Valušis, L. Li, E. H. Lin-

field, *Tarpjuostiniai elektroniniai šuoliai InAs/InGaAs kvantinių taškų heterodariniuose*, 41-oji Lietuvos Nacionalinė Fizikos Konferencija, 2015.06.17-19, Vilnius, Lietuva.

7. E. Pozingytė, **A. Rimkus**, R. Nedzinskas, B. Čechavičius, J. Kavaliauskas, G. Valušis, L. H. Li, E. H. Linfield, *Temperature dependent Photomodulated Reflectance of InAs InGaAs Dots-in-a-Well Quantum Structures*, 44th Jaszowiec International School and Conference on the Physics of Semiconductors, 2015.07.20-25, Wisla, Poland.
8. **A. Rimkus**, E. Pozingytė, R. Nedzinskas, B. Čechavičius, J. Kavaliauskas, *Teperature dependent spectroscopic study of InAs InGaAs dots-in-a-well structures*, 17th International Conference-School Advanced Materials and Technologies, 2015.08.27-31, Palanga, Lietuva.
9. R. Nedzinskas, **A. Rimkus**, E. Pozingytė, B. Čechavičius, J. Kavaliauskas, G. Valušis, L. Li, E. H. Linfield, *Photoreflectance study of InAs-InGaAs dots-in-a-well heterostructures*, 5th International School and Conference on Photonics, 2015.08.24-28, Belgrad, Serbia.
10. S. Paurazaitė, E. Pozingytė, S. Miasojedovas, **A. Rimkus**, P. Ščajev, S. Tumėnas, R. Nedzinskas, L. Trinkler, B. Berzina, V. Korsaks, T. Yan, C. L. Chen, L. Chang, M. M. C. Chou, *Optical investigation of epitaxial InGaAs quantum rods,* 42-oji Lie-

tuvos Nacionalinė Fizikos Konferencija, 2015.10.04-06, Vilnius, Lietuva.

11. **A. Rimkus**, E. Pozingytė, R. Nedzinskas, B. Čechavičius, J. Kavaliauskas, G. Valušis, L. Li, E. H. Linfield, *Optical study of vertically elongated InGaAs quantum dots*, Open Readings 2016, 59th International Conference for Students of Physics and Natural Sciences, 2016.03.15-18, Vilnius, Lietuva.

12. R. Nedzinskas, **A. Rimkus**, E. Pozingytė, B. Čechavičius, J. Kavaliauskas, L. Li, E. H. Linfield,, *Photoluminescence and photoreflectance of epitaxial InAs quantum rings*, OPTO2016 OPTO-Meeting for Young Researchers & International SPIE Student Chapter Meeting, 2016.07.06-09, Gdansk, Poland.

13. **A. Rimkus**, E. Pozingytė, R. Nedzinskas, B. Čechavičius, J. Kavaliauskas, *Photoluminescence and photoreflectance of selfassembling InAs quantum rings*, 18th International Conference-School Advanced Materials and Technologies, 2016.08.27-31, Palanga, Lietuva.

14. **A. Rimkus**, E. Pozingytė, R. Nedzinskas, B. Čechavičius, J. Kavaliauskas, G. Valušis, L. Li, E. H. Linfield, *Optical study of vertically elongated InGaAs/GaAs quantum dots grown using* As_2 *and* As_4 *sources*, 46th Jaszowiec International School and Conference on the Physics of Semiconductors 2017.06.17-23, Szczyrk, Poland.

15. **A. Rimkus**, E. Pozingytė, R. Nedzinskas, B. Čechavičius, J. Kavaliauskas, *Optical investigation of InGaAs/GaAs quantum rods grown using* As_2 *and* As_4 *sources*, 19th International Conference-School Advanced Materials and Technologies, 2017.08.27-31, Palanga, Lietuva.
16. R. Nedzinskas, **A. Rimkus**, E. Pozingytė, B. Čechavičius, J. Kavaliauskas, L. Li, E. H. Linfield, *Optical investigation of elongated InGaAs quantum dots*, OPTO2018, 2018.07.04-07, Gdansk, Poland.
17. **A. Rimkus**, E. Pozingytė, R. Nedzinskas, B. Čechavičius, J. Kavaliauskas, L. Li and E. H. Linfield, *Influence of the temperature and the excitation power on the optical properties of InGaAs quantum rods*, Apropos16, 2018.10.10-12, Vilnius, Lietuva.

OVERVIEW OF DISSERTATION

Introduction

The introductory chapter substantiates the relevance of the dissertation, formulates the aim and tasks of the work, discusses novelty and practical value of the dissertation. This chapter also presents controversial statements and dissertation topic. List of scientific publications together with international and national conferences is also provided.

1 Overview of the literature

This chapter is meant to introduce the modulation spectroscopy and methods of measurements successfully applied in the laboratory of Semiconductor optics located in Center for physical sciences and technology. The following is a description of molecular fibers used for self-forming quantum dot structures and the mechanisms of the morphological formation of points during growth. The section concludes with a literature review, which provides a wider introduction to quantum dot nanostructures and topical issues. An overview of the literature indicates which specific studies were not conducted at the time, thus emphasizing the contribution and value of our research and the research described in the dissertation.

2 Samples and experiment

In this work studied quantum structures consists of:

- InAs quantum dots with and without InGaAs capping layers embedded within GaAs/AlAs QWs,
- InGaAs quantum rods, embedded within InGaAs QW, and sandwiched between GaAs barrier layers.
- InAs quantum rings embedded in GaAs matrix. Also hybrid QRings and QD quantum structure.

Prior to presenting the results of spectroscopic researches, their theoretical analysis, achievements and problems, the work of other scientific groups that is relevant to this work is overviewed. The literature review will look at similar (or even the same) structures as described in this paper in order to highlight the novelty of this work, as well as the influence of different methodologies on the results of the experiment and on the conclusions to be made, which will be further discussed in the following chapters of the dissertation.

Diagram of nanostructures, growth protocols, micrographs of TEM structural analysis, etc. are also presented in this chapter.

The second part of the chapter in detail describes the experimental setup used in this work. The peculiarities of modulated reflection — photoreflectance and contactless electroreflectance together with photoluminescence and photoluminescence excitation spectroscopy and application for specific spectral groups of optical properties are presented. Modulation spectroscopy has a high sensitivity to measurements ($\Delta R/R = 10^{-4}$– 10^{-5}), even at room temperature, which makes it possible to efficiently investigate optical properties of quantum structures and thereby reveal a comprehensive electronic structure of samples measured.

3 Optical properties of semiconductor InGaAs/GaAs heterostructures

This chapter is the first original part of the dissertation.

In this part of the dissertation, separate sections and subsection investigated energy spectra of InAs/GaAs/AlAs different quantum heterostructures: InAs quantum dots (with and without InGaAs capping layer), InGaAs quantum rods and InAs quantum rings are presented and discussed. The numerical calculations required for interpretation of the modulated reflection spectra were performed using *nextnano*^{3} software [1].

During the simulation, these parameters are selected: shape, dimensions and material composition of heterostructure. Then, the distribution of stresses at the surface of quantum dot and in its environment is determined.

---

[1] Web page: `http://www.nextnano.de/`

Finally, by solving three-dimensional Schrödinger and Poisson equations, the quantum dot's electronic states are calculated. We calculated the spectrum of energy levels by applying a strain-dependent 8-band $k \cdot p$ model, and taking the material parameters from the source [6].

3.1 Optical properties of InGaAs quantum dots

Experimental studies of nanostructures consisting of InAs quantum dots (QDs) with and without InGaAs capping strain-relieving layer and embedded in GaAs/AlAs were performed using temperature-depended modulated reflection, photoluminescence and photoluminescence excitation spectroscopy.

Reflective effects from the substrate of the sample

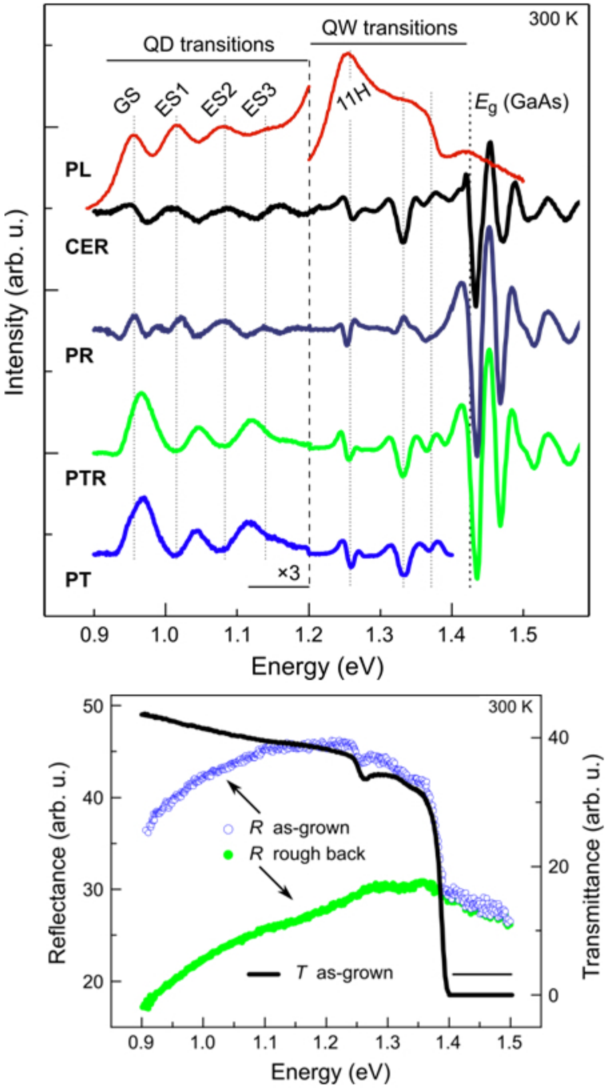
Prior to the spectroscopic measurements of InAs QDs embedded in InGaAs/GaAs quantum well the effects of reflection from the rear wall of the sample substrate on the photoreflectance spectra was investigated. In this work, the DWELL structures studied were grown on a polished (gloss) semi-insulating GaAs substrate, which is why it is important to discuss this phenomenon in more detail.

The phenomenon of reflection from the rear wall of the sample was investigated for bulk GaAs [7], but not for quantum dot structures. The probability of the absorption process in the QD layer is defined by the absorption cross section $\sigma = \alpha/N$ where α - absorption coefficient, and the N - QD density. It is important to mention that due to the low number QD layers, penetration depth of exciting radiation $(1/\alpha)$ exceeds the total thickness of quantum dot in the quantum well structure (with photon energy lower than the fundamental GaAs absorption edge $\eta\omega = E_g^{GaAs}$). Therefore reflection from the rear end of the sample grown on the reflecting pad could be significant.

In order to directly compare the PR spectra with and without reflection from the rear wall of the sample, the DWELL structure was divided into two parts, of witch one was coarsen. The effect of the reflection from the rear walls was experimentally investigated by measuring PR (Fig. 1a), not modulated reflection (R), and transmission (T) spectra (Figure 1b) of both samples at room temperature. Also PT, CER and high excitation power PL spectra are also shown for comparison.

Photomodulated absorption effect

In the view of results mentioned above, it can be assumed that a photomodulated signal from a sample with a glossy substrate consists of two components. The first one relates to a light-modulated internal electric field and is known as photo-reflection. This component has been isolated from the rectangular sample substrate (Fig. 1a). The second component, known as the photomodulated transmission (PT) in a reflection geometry



1 Fig. The energy spectra of QDs structure at room temperature: (a) comparative spectra of high excitation power photoluminescence (PL), contactless electroreflectance (CER), photoreflectance (PR), phototransmittence in reflectance geometry (PTR) and phototransmittance (PT); (b) R and T spectra of a sample with a glossy and coarsen substrate

(PTR), is associated with a partial reflection from the sample substrate (Fig. 1b). As in the traditional PT experiment, PTR component occurs due to photomodulated absorption (PA) effect. Indeed, the spectrum of PT is very similar to the PTR spectrum in the small photon energy region, where optical transitions between the ground and excited states of the QD are observed (shown in Fig. 1a). One of the main features is that the effect of PA dominates in PTR spectrum of the sample with glossy substrate. The dominance of the PA effect in InAs quantum dot modulated reflection spectra is further analyzed by examining the propagation of the probed light in a sample with a glossy tray in the dissertation.

Photomodulation mechanisms

Looking at the Fig 1a line-shaped symmetry in the QD optical transition energy we see that the PT and PTR spectra clearly differ from the PR spectrum. A positive signal is clearly seen in the PT and PTR spectra between the QD fundamental states of energy, while the derivative curve is observed in the CER and PR spectra. In this chapter of the dissertation mentioned before phenomena is analyzed by taking into account two distinguished modulation mechanisms:

- Filling of the states (Pauli's filling);
- Quantum-Confined Stark Effect (QCSE).

Influence of stress-reducing InGaAs layer

In this part performed comparative studies of InAs quantum dots inserted in the GaAs/AlAs quantum well with (sample L442) and without (sample L444) strain-reducing InGaAs layer PR and PL spectra are discussed.

The experimental results of DWELL heterostructures have been interpreted using analytical calculations. The following types of optical transitions have been identified from the comparative spectral analysis (the number indicates the spectral position of the optical characteristic in Fig. 2):

(1) InAs QD ground state (both structures);

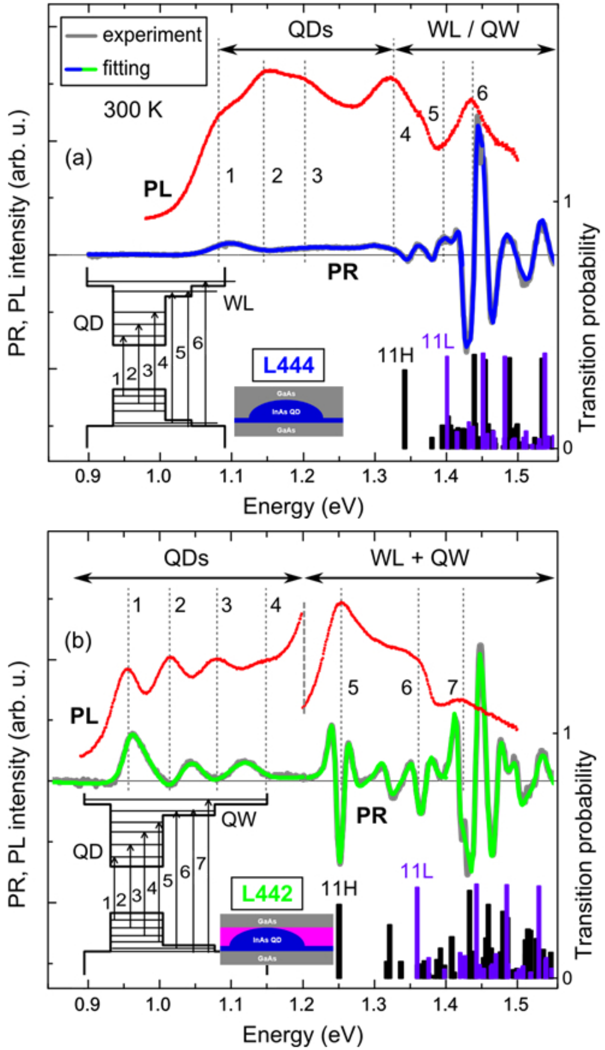
(2-4) InAs QD excited states (both structures);

(4*) strain induced QD state (L444 only);

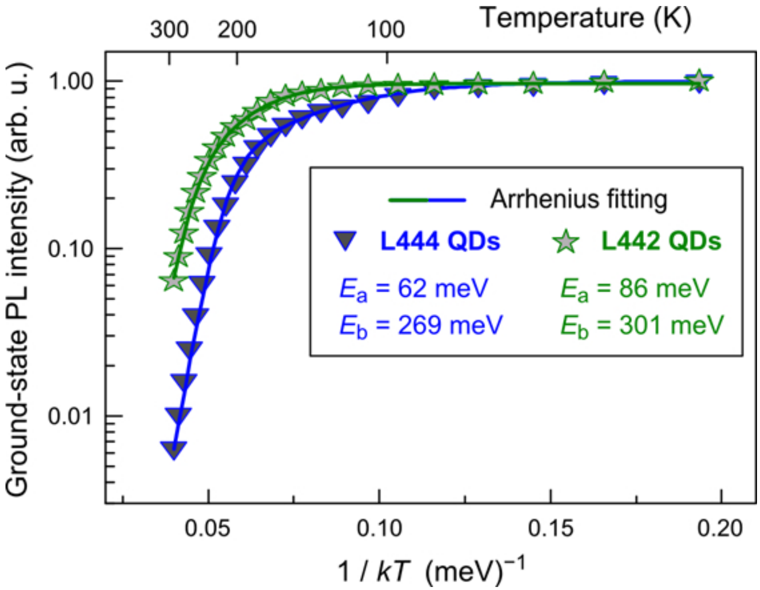
(5) States of InAs/InGaAs binary QW for L442 structure or quantum states of InAs wetting layer and the ground state of GaAs quantum well system (WL-QW) for L444 structure;

(6,7) Exited quantum states of InAs/InGaAs binary QW (L442) or InAs WL-QW (L444).

To test the assumption that observed redshift of InAs QD GS optical transitions were caused by increase of QD physical size and shape variation, analytical calculations for InAs/InGaAs structures were performed. In this method the In concentration of the strain relieving $In_xGa_{1-x}As$ layer and dimensions of average quantum



2 Fig. PL and PR spectra of DWELL structures at room temperature (a) QD structure without InGaAs capping layer (b) structure capped by InGaAs layer. The results of digital calculations are represented by vertical columns: Spectral position indicates optical transition energy, amplitude - intensity. Inset shows band diagram and numbered experimentally identified interband transitions.



3 Fig. The integrated intensity of the InAs QD ground state PL emission band (normalized) as a function of the temperature for the DWELL structure without (L444; triangles) and with (sample L442; stars) InGaAs cap layer. The least-square method fitting was performed using the Arrhenius-type function.

dots were calculated. The modeling results for analytical calculation of InGaAs/GaAs QW are shown in Fig. 2 by vertical bars. The comparison of the experimental results with the computational data, shows lower In concentration than indicated in the growth protocol. This is thought to be due to the redistribution of In between the quantum dots and capping layer. Thus, the redshift of transitions between the basic states can be explained by an effective increase in QD dimensions.

The temperature dependent intensity of photoluminescence for the interband quantum dot GS transitions is given in Fig. 3. In this figure it is displayed that quenching of the PL intensity for both samples (with and without InGaAs layer) shows two distinct slopes: in intermediate and in high temperatures. This thermal decay was evaluated in using, standard for this analysis, Arrhenius-type expression:

$$I = \frac{I_0}{1 + C_1 \exp(-{}^{E_a}\!/_{kT}) + C_2 \exp(-{}^{E_b}\!/_{kT})} \qquad (1)$$

Also, the dependence of the jumps between the main states on the intensity of temperature on the structures investigated was determined from the spectra of PL. Immediately it can be seen that the intensity of the capped structure is greater in a wide area. We will notice that the activation energy of the capped structure is higher than the main state and continuity. This is associated with a smaller distance between the ground state and the continuum due to the larger quantum dots of the capped structure.

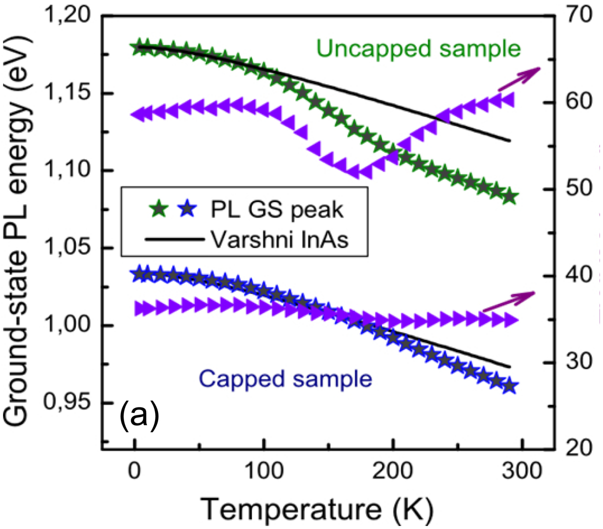
Statement for defense

> Changes in the electronic structure of InAs quantum dots covered with InGaAs are caused by a decrease in tension, a lower material decomposition and an increase in dot height.

Temperature-dependent measurements

In order to get a deeper understanding of thermal processes, PL measurements were carried out in the temperature range of 10–300 K. Energies of QD optical transitions together with full width at half maximum (FWHM) as a function of temperature for the QD structures with and without InGaAs capping layer is shown in Figure 4a. In the picture samples are compared with each other as well as with numerical calculations that were fitted using a Varshni type expression. We see that the GS peak energy and the FWHM parameter have different temperature dependency. The anomalous FWHM parameter reduction at 170 K, coupled with a decrease in optical transitions energy between ground quantum dot states, can be explained by the rearrangement of thermally activated carriers between quantum dots of different physical dimensions. In other words, at low temperatures, in quantum dots the carriers are distributed randomly. Carriers can be thermally activated from small QD to WL as the temperature rises and then fall into larger QDs whose activation energy is higher (Fig. 4b). Due to this process, the narrowing of the PL spectrum and the thermal redshift are noticeable. In other words, for the structure with InGaAs capping layer (L442) FWHM parameter stays almost constant with the increase of temperature and GS energy peak is quite accurately represented by the curve obtained from the semi-empirical Varshni equation.

Peak energy and the full width at half maximum

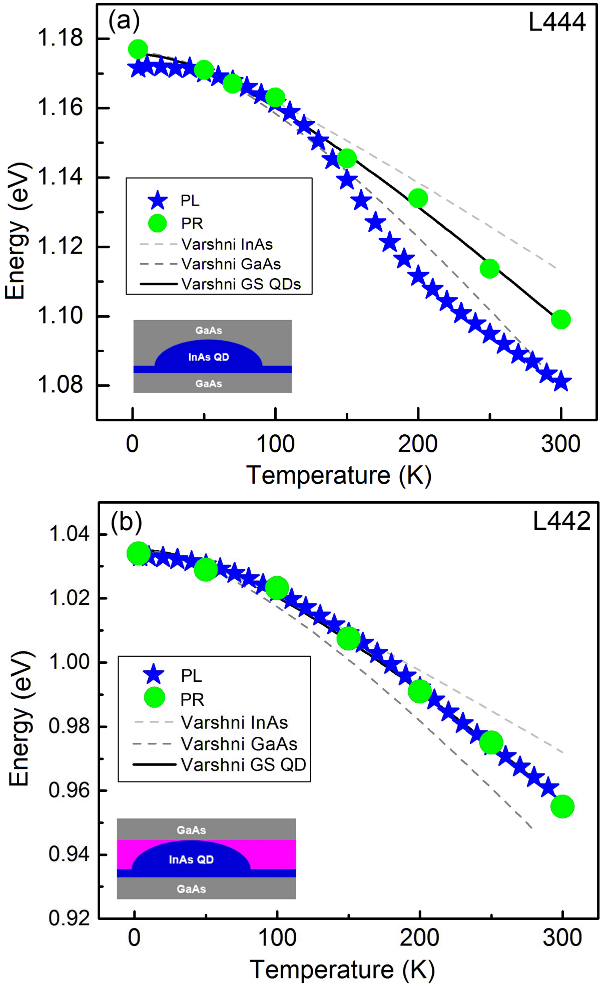


4 Fig. Peak energy and FWHM of the ground-state PL Gaussian line versus temperature for DWELL structure without (sample L444; top) and with (sample L442; bottom) InGaAs cap layer. The temperature dependence of GS peak energy for both DWELL structures is compared to the energy gap shift of bulk InAs (for convenience Varshni curves are shifted along y axis).

(FWHM) parameter of the InAs QD ground-state PL Gaussian line illustrate exceptionally distinctive temperature behavior for Dwell structures with and without strain relieving InGaAs cap layer (Fig. 4a). For uncapped sample L444, the atypical decrease of FWHM parameter at ~170 K is accompanied by a red-shift of the emission peak, which is ordinary for InAs/GaAs QDs. Usually, this behavior can be explained by thermally activated carrier escape and retrapping in QDs of different sizes. When the temperature exceeds 200 K, electron–phonon scattering becomes important, thus a strong increase in FWHM is observed. The shift of PL band in a high temperature region becomes similar to that of a bulk InAs bandgap (Varshni curves, Fig. 4a).

The situation is different with the InGaAs-capped sample L442, where FWHM parameter exhibits almost no reduction as the temperature rises and the peak position of PL shifts without any obvious kink almost following the bandgap shift of bulk InAs. This suggests that the use of the InGaAs cap layer yields much smaller QD size dispersion, which is manifested by indistinct thermal transferring and redistribution of carriers in the 3–300 K temperature range.

In Figure 5 we see the temperature dependence of interband optical transitions between QDs ground states for the structures without (a) and with (b) InGaAs capping layer. Experimental data was obtained from the photoluminescence and modulated reflection spectra. In the low temperature range up to T ~ 100 K in the uncapped structure (Fig. 5a), both the absorption (PR)



5 Fig. Peak energy of the ground-state PR and PL Gaussian line versus temperature for DWELL structure without (a) and with (b) InGaAs cap layer.

and the emission (PL) lines follow curve calculated by Varshni equation:

$$E(T) = E_0 - \frac{\alpha T^2}{T + \beta}, \tag{2}$$

where E_0, α and β are fitting parameters. In this case, the quantum dots are randomly filled with carriers and the PL spectrum corresponds with the density of the quantum states. In the temperature range T $>$ 100 the emission (PL) energy of peak position deviates from absorption curve (PR) and Varshni's law (Stokes shift being observed). This shift is visible because at higher (T $>$ 100) temperatures the energy is sufficient to redistribute the carriers between the quantum dots of different sizes. When comparing the thermal characteristics of the uncapped and capped with InGaAs layer structures, we notice that in the case of the capped structure PL and PR peak energy functions accurately follow Varshni curve, which indicates a more homogeneous quantum dot size distribution.

Statement for defense

> The insertion of the InGaAs capping layer reduces the spectral linewidth of the InAs quantum dots and eliminates the Stokes shift, so the quantum dots in the InAs/InGaAs/GaAs heterostructure are more homogeneous.

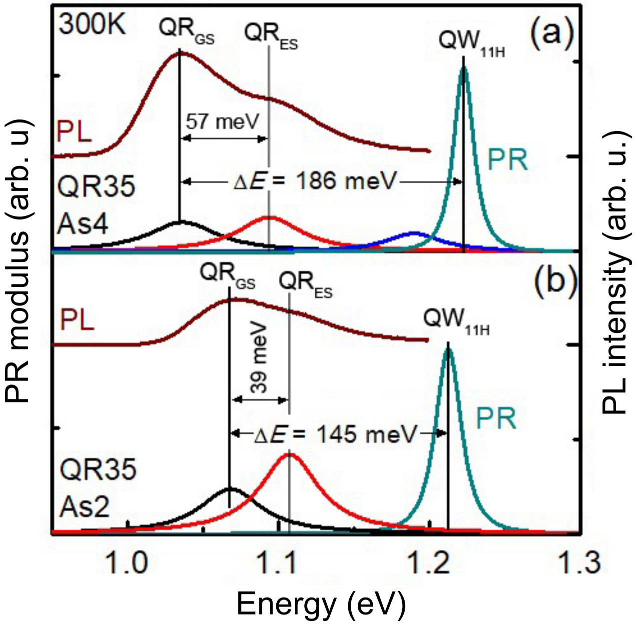
3.2 Optical properties of InGaAs quantum rods

Quantum rods (QRods) are intermediate structures between 0-D and 1-D. Optical studies of such quantum structures are important for the advancement of modern quantum engineering and for the development of new generation optoelectronic devices. In this part of dissertation, we present investigative results of InGaAs quantum rods grown using As_2 or As_4 molecular beam, their optical properties and the electronic structure analyzed by photo-reflection (PR) spectroscopy and photoluminescence (PL) techniques. Quantum rods QR(N) with different number of superlattice periods N (different height) were investigated:

- QR10, QR20 and QR35 alongside with quantum dot structures for comparison.

Morphology of InGaAs quantum rods

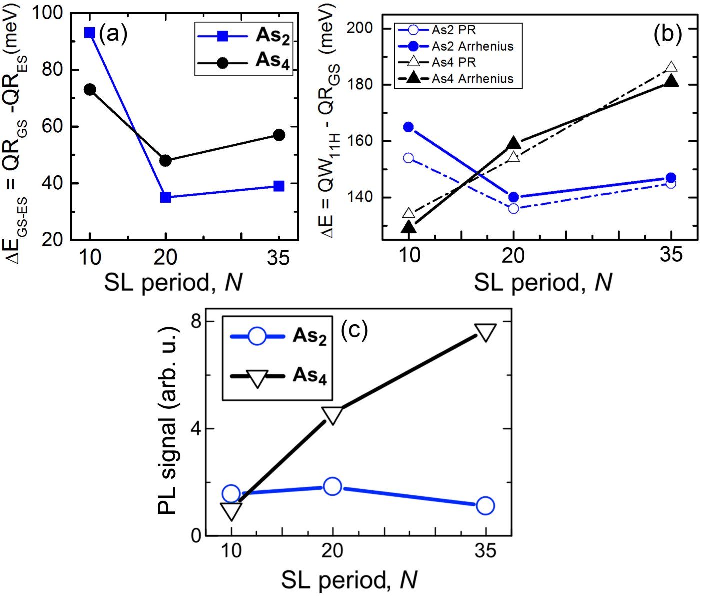
PR and PL spectra for InGaAs QRods grown using As_2 and As_4 sources, is shown in Fig. 6. Heterostructure grown using As_4 shows significantly higher energy difference between QRods ground and excited states and between ground states of QRods and InGaAs QW. These energy differences suggest that using As_4 results in higher dimensional quantization energy compared to structures grown using As_2 flux. In order to understand in greater detail the influence of As source and quantum rod height on PL intensity and electronic structure, we



6 Fig. PL spectra and PR modulus for QR35 samples consisting of a 35 period SL grown using As4 (a) and As2 (b) sources. The PR modulus data are normalized to that of 11H optical transitions of the InGaAs QW. ΔE denote energy separation between the InGaAs QR-related ground-state (GS) and QW-related 11H transitions.

investigated the dependence of the abovementioned energy differences and PL intensity on the number of SL periods (QRods height).

In Fig. 7a-b it is visible that the energy difference ΔE is larger for As_4 grown taller ($N \geq 20$) rods. It is important to note that the intensity of light emission increases significantly with the use of the As_4 source (Fig. 7c). This increase is well correlated with ΔE, i.e. with the improvement of carrier localization. Thus, the spec-



7 Fig. Dependencies of the energy separation E between the InGaAs QRods related ground and exited state (a), QRods and QW related ground state transitions (b) and intensity of PL emission (c) on SL period number for samples grown using As_4 and As_2 sources.

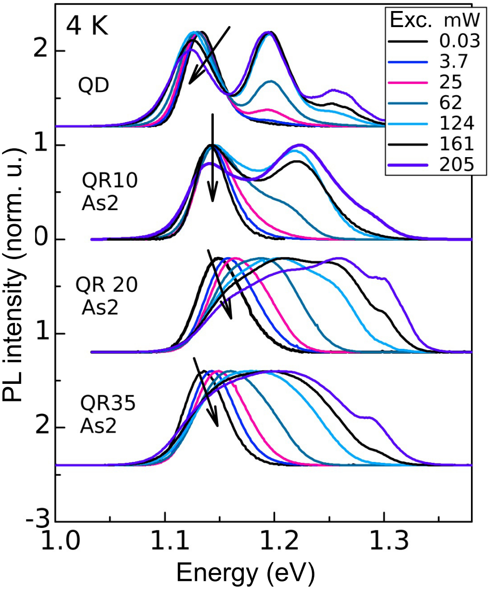
tral changes of InGaAs quantum rod structures grown with different As sources in QRods and QW spectral region are related to the In content change in InGaAs layers. It was assumed that a higher indium contrast between InGaAs QRods and InGaAs QWs is achieved using the As_4 source.

Statement for defense

Using As_4 molecular flux results in deeper quantum well than using As_2.

Impact of excitation power

The influence of excitation power on the PL spectra of quantum rods was also studied. PL at 4 K of quantum rods with different heights (QR10, QR20 and QR35) grown under As_2 flux are shown in 8 Figure. For comparison, PL spectra of quantum dots measured under the same conditions is provided. The results show that the increase of excitation power has different effect on quantum dots and quantum rods optical emission spectra.



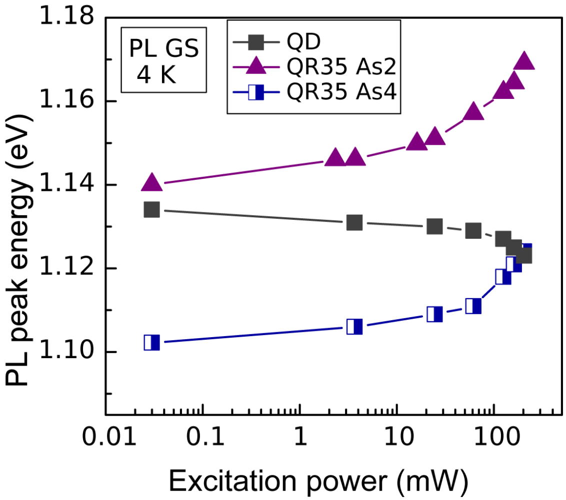
8 Fig. PL spectra, measured at 4 K using different excitation power, of InGaAs QRods grown using As_2 flux. For comparison, shown quantum dot (QD) spectra, measured under the same conditions.

In the case of quantum dots (Figure 8, upper curves), the following effects are observed with increasing excitation power:

1. gradual saturation of the lowest energy PL transitions, which is accompanied by redshift of PL energy and increase of linewidth;
2. appearance of emission peaks caused by optical transitions between discrete excited states.

These phenomenon, observed under high excitation conditions, reveal the discrete (quasi-atomic) energy levels of the investigated quantum dots with the δ-function type density of states, as well as filling effects of QD electronic states.

Different effects are shown in QRods PL dependence on excitation power spectra (Fig. 8, lower curves). As the excitation power increases, the asymmetric propagation of the PL curve and its continuous shift on the high energy side are observed. These effects are more pronounced on high rods (QR20, QR35 structures). A strong change in PL energy growth begins when excitation power transcend value of 10 mW order (Fig. 9). The analysis of the results showed that these spectral changes can be caused by factors such as shielding of internal piezoelectric fields with excited carriers and phase space filling effects. The experimental observation of the continuous propagation of the PL band on the side of large energies should be associated with the filling of the energy states. It is a sign of a change in the density of the electronic states when transitioning from the quantum dot to the quantum rod . The results show



9 Fig. PL peak energy of As_2/As_4 grown InGaAs quantum rods (QR35) as a function of the excitation power at 4 K. For comparison, shown quantum dot (QD) spectra, measured under the same conditions.

that quantum rods with higher number of SL periods $N \geq 20$ should be described as quasi-one-dimensional rather than zero-dimensional nanostructure.

Statement for defense

Transition between 0-D and 1-D structures is achieved by forming higher quantum rod nanostructures.

3.3 Optical properties of InAs quantum rings

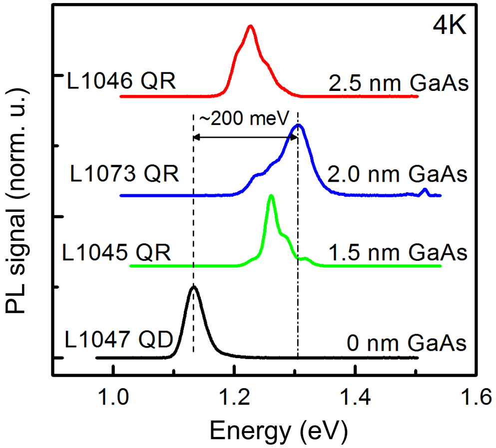
In this section we will discuss PL and PR spectra of InAs quantum rings (QRings) measured in the temperature range of 3 to 300 K. QRings were grown on InAs quantum dots by partly covering them with 1.5–2.5 nm

GaAs layer and annealing in an As_2 atmosphere. Two sets of samples were grown at Leeds University (Great Britain):

- 1st series of QRings samples consisting of samples L1041 (2.1 ML InAs + 2.0 nm GaAs), L1042 (2.4 ML InAs + 2 nm GaAs), L1045 (2.4 ML InAs + 1.5 nm GaAs) and L1046 (2.4 ML InAs + 2.5 nm GaAs). These series was grown by applying a different thickness layer of GaAs on InAs quantum dots, that way changing their dimensions and morphology.
- The 2nd QRings series - quantum ring sensor structures - samples L1073, L1081 and hybrid quantum dots and quantum rings structure - L1072.

Effects of GaAs capping layer thickness

In order to investigate the influence of GaAs capping layer thickness on the formation of quantum rings, InAs QRings structures were formed by nearly covering the layer of InAs quantum dots with a 1.5 nm, 2 nm and 2.5 nm thickness GaAs. Low temperature PL spectra for the first group of samples, are shown in Fig. 10. Looking at the PL spectra, we can see that InGaAs QRings spectra in contrast to the InAs QD spectrum (Fig. 10, L1047 QD) shows a significant blueshift of 200 meV. This increase in optical transition energy can be interpreted by small dimensions of quantum rings (compared to QDs). Indeed, the reduction of the electron localization leads to the shift of the ground state energy towards continuum. With this in mind, it can be



10 Fig. Normalized PL spectra of InAs quantum rings (QR) at 4 K. The thickness of GaAs cap layers used to grow rings is marked on the spectra. The quantum dot spectrum (QD) is given for comparison.

said that such QRings structures are useful for designing long-range infrared photovoltaic sensors utilizing optical transitions between localized electron energy states and continuum.

Statement for defense

> InAs quantum rings grown by partly covering quantum dots with 2 nm GaAs layer have a lower energy difference between bound state and continuum compared to quantum dots or quantum rod structures.

4 Main results and conclusions

DWELL

The nature of temperature dependent optical transitions in InAs quantum dots embedded into InGaAs/GaAs/AlAs quantum wells as well as the peculiarities of their photomodulation were investigated by photoreflection and photoluminescence techniques in the range of 3 to 300 K. Furthermore, the impact of the strain-reducing InGaAs layer on the optical properties and electronic structure of quantum dots was revealed.

1. After comparing photoreflection and phototransmission spectra, a phototransmission component was observed in the photoreflectance spectrum witch influences the shape and the intensity of the spectra. In this regard, the substrate of the sample was roughened to more accurately assess the spectral peculiarities of the reflectance by eliminating the phototransmission component and to provide evidence for the influence of back-surface reflection effects on the photoreflection spectra of DWELL structures.

2. The red shift of interband transitions between ground states of quantum dots alongside with greater number of exited states in InGaAs capped structure can be explained by the decrease in stress and the increase in the effective dot size due to the redistribution of In between the quantum dots and the

capping layer during growth.

3. Increased peak intensity and reduced linewidth of photoluminescence spectrum for the InGaAs capped structure indicate a more homogeneous distribution of InAs quantum dot sizes.
4. The composition of InGaAs capped InAs quantum dots is partially altered due to Ga/In interdiffusion.
5. Thermal quenching of emission from InAs quantum dots at medium temperatures is associated with a lower carrier flow to quantum dots, and at high temperatures with QD exciton ionization.
6. The photomodulation mechanism of quantum dots depends on temperature: quantum confined Stark effect dominates at high temperatures and filling of quantum dot electron states — at low temperatures.
7. The absence of Stokes shift and the agreement of experimental results with calculations done using Varshni type expression indicate that InAs quantum dots are more homogeneous in the structure that was capped with the strain-releasing InGaAs layer.

QRods

Optical transitions and electronic structure of InGaAs quantum rods embedded in InGaAs quantum well were studied in detail by applying temperature dependent photoreflection, photoluminescence and photoluminescence excitation techniques.

1. When compared grown using As_4 and As_2 flux, it was revealed that the As_4 grown structures exhibit higher intensity of photoluminescence, redshift of emission wavelength and greater energy distance between the ground and the first excited state.

2. It was revealed that As_4 grown heterostructure also has a greater energy distance between the ground state of the quantum rods and the lowest energy state associated with the quantum well. This could indicate better localization of the charge carriers due to greater contrast of In between the InGaAs quantum rod and the InGaAs quantum well.

3. After temperature dependent photoluminescence measurements anomalous peak energy dependence and thermal growth of linewidth parameter with rising temperatures were revealed. The observed phenomenon is associated with the thermally activated carrier redistribution between different sized quantum rods through the electronic states of the quantum well and a GaAs barrier.

4. It has been found that the thermal quenching of emission intensity is associated with thermal activation of excitons between ground states of the InGaAs quantum rods and the InGaAs quantum well at medium temperatures and with carrier ther-

mal activation from the ground to the exited states in InGaAs quantum rods at high temperatures.

5. Photoluminescence spectra dependence from excitation power studies reveal that high quantum rods (number of periods $N \geq 20$) would be more suitable to describe as quasi-one-dimensional rather than zero-dimensional nanostructures.

QRings

InGaAs quantum ring photoreflectance, photoluminescence and photoluminescence excitation measurements were performed at 3–300 K temperature.

1. Low-temperature photoluminescence measurements of structures show that the energy of interband transitions between ground states of InAs quantum rings is up to 200 meV higher compared with InAs quantum dots. This indicates smaller dimensions of quantum ring structures and lesser quantum localization of charge carriers.

2. The anomalous temperature dependent energy shift of ground state transitions and linewidth of InAs quantum ring structures can be explained by nonhomogeneity of quantum ring sizes and redistribution of thermally activated carriers among them.

3. It has been shown that the photoreflectance technique is suitable for spectroscopically characterizing of InAs quantum ring heterostructures. Low

energy spectral features observed in the combined InAs/InGaAs/GaAs quantum dot and InAs/GaAs quantum rings heterostructure are related to interband optical transitions between ground and excited states of quantum dots and quantum rings. In higher energy region observed spectral features are associated with InGaAs QW and wetting layer of QRings.

4. The photoluminescence excitation spectra of hybrid heterostructure revealed that there is no carrier transfer between the quantum rings and the quantum dots.

References

[1] A. Luque, A. Marti, A. J. Nozik, Advanced Inorganic Materials for Photovoltaics, **32**(3), 236–241 (2007).

[2] P. Yua, J. Wub, L. Gaoc, H. Liub, Z. Wanga, Solar Energy Materials and Solar Cells, **161,** 377–381 (2017).

[3] F. Olbrich, J. Höschele, M. Müller, J. Kettler, S. L. Portalupi, M. Paul, M. Jetter, P. Michler, Appl. Phys. Lett. **111**, 133106 (2017).

[4] R. Nedzinskas, B. Čechavičius, A. Rimkus, E. Pozingytė, J. Kavaliauskas, G. Valušis, L.H. Li and E.H. Linfield, J. Appl. Phys. **117**, 144304 (2015).

[5] N. Prodanovic, N. Vukmirovic, D. Indjin, Z. Ikonic, P. Harrison, J. Appl. Phys. **111**, 073110 (2012).

[6] I. Vurgaftman, J. R. Meyer, and L. R. Ram–Mohan, J. Appl. Phys. **89**, 5815 (2001).

[7] R. L. Tober and J. D. Bruno, J. Appl. Phys. 68, 6388 (1990). 20H. Roppischer, N. Stein, U. Behn, and A. B. Novikov, J. Appl. Phys. **76**, 4340 (1994).

Summary

In this work temperature dependent photoluminescence and photoreflectance spectroscopy techniques are employed in order to study epitaxial InAs/InGaAs quantum heterostructures. A detailed analysis of the optical transitions, taking place in quantum structures is necessary in order to characterize their optimal growth parameters.

Part I of this doctoral thesis presents an comprehensive literature overview, in regard to studied InAs/InGaAs nanostructures. **Part II** specifies investigated quantum structures and experimental set-up of used spectroscopic techniques. Also brief principles of modulation spectroscopy, photoluminescence, molecular beam epitaxy and Stranski–Krastanow growth mode are detailed in this part. **Part III** presents original experimental results of the doctoral thesis. It consists of the empirical and theoretical analysis of material composition, electronic structure and optical properties of InAs/InGaAs quantum dots, InGaAs quantum rods and InAs quantum rings structures. Also summarized results and conclusions are given at the end of this part.

About the author

Andrius Rimkus was born in 1989. May 20 in Vilnius. In 2008 graduated from Vilnius Salomėjos Nėries gymnasium. In the same year, he enlisted in Faculty of Physics at Vilnius University, where 2008–2012 he studied Physics of modern technology and management program. Prepared the bachelor's final thesis "Investigation of Stable Cell Phase Pulse Generation in the Middle Infrared Spectrum", supervisor prof. habil. dr. Audrius Dubietis. After that he started master studies at the Faculty of Physics of Vilnius University, where in 2012–2014 studied a program of Laser physics and optical technology. Prepared Master's Thesis in "Optical spectroscopy of semiconductor heterostructures", supervisor dr. Ramūnas Nedzinskas. From 2012 he worked as a Engineer and later as a junior researcher in Semiconductor Optics Laboratory at the Center for Physical Sciences and Technology. He has studied PhD at the Center for Physical Sciences and Technology since 2014.

"– Why have you decided to work as scientist?"

It is a really complex question. Strongly influenced by Isaac Asimov, I believe that you should not stop learning at a given age. And while you can grasp only a tiny portion of all this knowledge, it would be "(...) a tragedy just to pass through and get nothing out of it".

UŽRAŠAMS

UŽRAŠAMS

Vilniaus universiteto leidykla

Universiteto g. 1, LT-01513 Vilnius

El. p. info@leidykla.vu.lt,

www.leidykla.vu.lt

Tiražas 30 egz.