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Review for the award of the doctoral degree to Paweł Holewa, M.Sc. in the field of natural and exact sciences, in the discipline of physical sciences

The PhD thesis of MSc Paweł Holewa entitled "InAs/InP quantum dots for telecom quantum photonics" has been prepared under the supervision of Dr Elizaveta Semenova (Technical University of Denmark) and Dr hab. inż. Marcin Syperek, prof. PWr (Wrocław University of Science and Technology). It addresses issues at the intersection of solid state physics, photonics, quantum optics and semiconductor technology. It arises as a response to the growing demand for efficient and robust sources of nonclassical light for telecommunication, coming from both, fundamental research and industry.

In his dissertation, the Author studies InAs/InP-type quantum dots as an optimal source of quantum light in the third telecommunication window. He aims at:

1. Elaboration of method production of InAs/InP-type quantum dots, so that they exhibit low surface density and high planar symmetry and act as bright single photon sources
2. Development of photonic structures ensuring efficient collecting of emission from individual InAs/InP-type quantum dots

According to the results of research presented by the M.Sc. Holewa in the thesis, the response to the first issue is fabrication of InAs/InP or InAs/(In,Al,GaAs)/InP quantum dots by epitaxial methods with additional steps introduced to the growth procedure such as growth ripening or employing so called droplet epitaxy. The second technique is particularly attractive thanks to high planar symmetry of fabricated quantum dots. The response to the second issue is modification of the photonic environment of the emitter: introducing Al mirror beneath the quantum dot layer and deterministic fabrication of circular Bragg Reflectors deposited on the sample surface at individual quantum dots positions. The Author proves usefulness of the such coupled quantum dot-cavity system for applications in quantum optics by demonstrating quantum dot emission in the third telecommunication window characterized by extraction efficiency of above 16%, single-photon purity of around 99.68% and two-photon interference visibility above 19%.

The first part of the thesis provides a comprehensive, detailed, yet concise introduction to the subject of the study. The second part is a core part of the thesis and it takes a form of series of five scientific articles of M.Sc. Paweł Holewa.

I will first refer to the first part of the dissertation.

In the first chapter, a motivation and objectives of the thesis are presented in a clear and convincing way. Along with them basic concepts relevant for the thesis subject are introduced. In particular, the Author discuses specificity of the third telecommunication window photonics, properties of InP-based quantum dots, and methods for increasing photon collection efficiency from structures with quantum dots.

The second chapter starts with a detailed discussion of the physics governing the sample growth by metalorganic vapour-phase epitaxy. A complexity of processes taking place during the sample growth is highlighted. Next, mechanisms standing behind formation of epitaxially grown quantum dots are presented. The subsequent part is devoted to electronic structure of quantum dots and resulting emission properties. In particular, it contains a description of eight-band $k{\cdot}p$ theory utilized by the Author for calculation of energy levels of quantum dot confined single carriers and, upon application of Hartree-Fock approach, determination of energy of confined excitons. Quantities such as exchange interaction, fine structure, optical selection rules and dynamics of radiative recombination of confined excitonic complexes are then discussed. Next sections are devoted to quantities characterizing non-classical emission and physics of interaction between an emitter and an optical micro-cavity. In particular, weak- and strong light-matter coupling limits are introduced. I should underline here that the discussion of practically all concepts in the Chapter 2 is not just qualitative, but it is supported by calculations or formulas describing them quantitatively.

Chapter 3 is devoted to experimental aspects of the research conducted in a frame of the thesis. The Author describes fabrication procedure of quantum dots, with particular attention paid to factors that govern the epitaxial growth of quantum dots by metalorganic vapour-phase epitaxy, design of experimental setups utilized and a procedure of deterministic fabrication of cavities coupled to QDs. In addition, a procedure for determination of the photon extraction efficiency and single photon purity is described.

Chapter 4 contains a brief summary of the results presented in the second part of the thesis. JSuff

A bibliography is the final chapter of the first part of the thesis. The choice of the literature references is well done, and the bibliography is exhausting. I must mention that I like very much recalling of the first works devoted to Hanbury-Brown and Twiss type interferometer. It is very rare, as usually only a canonical paper, published in Nature in 1956 is mentioned.

Before I pass to the second part of the thesis, let me express that I highly appreciate that the Author contained the first, introductory part, despite no formal requirement for doing so. It shows M.Sc. Holewa has a very good command of physics standing behind research he performs. As a side remark, in my opinion, one gains an opportunity to learn quite a lot when collecting pieces of knowledge to be contained in their thesis.

The second part of the thesis contains a collection of four articles published in such journals as ACS Photonics, Nanophotonics, Physical Review B, Physical Review Applied, and one being a preprint. In all of these articles, P. Holewa is the first Author, and, as results from the statements provided, he has a predominant contribution to all these works. As I expressed above, the purpose of the thesis is from one side – search for optimal quantum emitters, that is bright InAs/InP-based QD operating the third telecommunication window, with low surface density and negligible fine structure splitting of the exciton. From the other side, it is the development and engineering of a photonic environment ensuring the highest efficiency of collection of the light emitted by quantum dots.

The first three articles address the issue of fabrication and properties of InAs/InP-based quantum dots emitting in the third telecommunication window:

- P. Holewa *et al.*, *Optical and electronic properties of low-density InAs/InP quantum dotlike structures devoted to single-photon emitters at telecom wavelengths*, Physical Review B 101, 19 (2020).

The article presents a joined experimental and theoretical study of morphological and optical properties of low-density InAs/InP QDs fabricated by metal-organic vapor phase epitaxy in the Stranski-Krastanov growth mode. The main finding is that the quantum dots emission spectrum takes the form of well-resolved spectrally peaks, all characterized by comparable radiative lifetime. This can be traced back to structural properties of quantum dots with families of different heights varying from a single to a few monolayers of InAs and effects of weak confinement of electron and exciton confinement dependent on quantum dot height. The Author was able to resolve emission excitonic complexes confined in individual quantum dots (fine structure splitting of the neutral exciton of the order of tens of μeV) and characterize their properties in experiment and by the calculations.

- P. Holewa *et al.*, *Optical and Electronic Properties of Symmetric InAs/(In,Al,Ga)As/InP Quantum Dots Formed by Ripening in Molecular Beam Epitaxy: A Potential System for Broad-Range Single-Photon Telecom Emitters*, Physical Review Applied 14, 6 (2020).

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The article reports on results of an optical study performed on an ensemble of InAs/(In,Al,Ga)As/InP quantum dots produced in this case by ripening-assisted Molecular Beam Epitaxy. Again, distinct peaks are observed in the emission spectra. Theoretical modelling indicates that they can be attributed to sub-groups of quantum dots differing in height by 2 monolayers of the material. Negligible polarization of the emission suggests that quantum dots are not affected by a possible strain related to a crystallographic structure. Temperature-dependent measurements reveal that emission quenching mechanism results mostly from the escape of holes to the barrier layer.

- P. Holewa *et al.*, *Droplet epitaxy symmetric InAs/InP quantum dots for quantum emission in the third telecom window: morphology, optical and electronic properties*, Nanophotonics 11, 8 (2022).

The works is devoted to the optical and electronic properties of the ensemble of and individual InAs/InP quantum dots produced by metal-organic vapor phase epitaxy in the droplet epitaxy growth mode. Such quantum dots were expected to exhibit a higher in-plane symmetry than those grown in Stranski-Krastanov mode. They form in pits etched around them in the growth process. This effect is explained by a proposed kinetic model. Modelling in a frame of the eight-band $k{\cdot}p$ method in the configuration–interaction approach enabled interpretation of observed optical properties of quantum dots. A single photon emission with a purity on the level as high as 92.5% was reported.

The next two articles aim at photonic enhancement of the collection efficiency of the signal emitted from InAs/InP-based quantum dots:

- P. Holewa *et al.*, *Bright Quantum Dot Single-Photon Emitters at Telecom Bands Heterogeneously Integrated with Si*, ACS Photonics 9, 7 (2022).

This work proposes theoretically and demonstrates experimentally a design, where a structure with InAs/InP quantum dot layer is deposited on an aluminium mirror integrated with a silicon substrate. The design ensures a significant increase of photon extraction efficiency (to around 11%) in a broad spectral range. This, in turn, enables spatial positioning of quantum dots by emission imaging. Single photon emission of charged exciton was obtained with a purity on the level of 97%.

- P. Holewa *et al.*, *Scalable quantum photonic devices emitting indistinguishable photons in the telecom C-band*, arXiv:2304.02515

This work adds new important elements to the knowledge and technology developed by the precedent ones in the series. Namely, photon extraction efficiency is enhanced in a deterministic way. A layer with low-spatial density InAs/InP quantum dots produced by metal-organic vapor phase epitaxy in the Stransky-Kastanov mode is deposited on an aluminium mirror on Si substrate. Photoluminescence imaging allows for the determination of the position of quantum dots emitting at a desired wavelength. Next, circular Bragg grating

(CBG) cavities are fabricated on the sample surface at positions of these preselected quantum dots (with a very good yield of 30%). The cavities enable an increase in photon extraction efficiency to the level of 16.6%. A very high purity (around 99.68%) of single photons emission from a quantum dot at C-band wavelengths and unprecedented photon indistinguishability above 19% is reported.

The second part of the thesis contains also Supplementary materials to two of the articles contained, list of publications and conference presentations, a useful list of acronyms, list of symbols, and list of figures and tables. List of conference presentations indicates that the M.Sc. Holewa has gained already a very good experience in communicating his research (in particular, he delivered 11 contributed talks and presented 11 posters on international scientific conferences).

All the presented works present exhibit a high level of novelty and originality. Those four of them that have been already published, collected 40 citations. Overall number of scientific papers of M.Sc. Holewa is 16, they were citated 154 times and his Hirsch index is 7 (according to Scopus database). These are excellent results taking into account a relatively early stage of his scientific carrier.

In general, one should appreciate the comprehensive skills of the P. Holewa as a researcher. He has a command of such variety of optical spectroscopy methods as polarization-resolved time-integrated and time resolved micro-photoluminescence, single photon correlations and Hang-Ou Mandel type measurements as well as structural methods such as atomic force microscopy. In addition, he is able to conduct calculations of the electronic structure of solids within 8-band $k{\cdot}p$ method implemented in the Nextnano software. It is worth pointing out that P. Holewa was involved not only in the work of strictly research type, but he also contributed to the demanding and time-consuming elaboration of new technologies. These included growth of quantum dots of desired optical properties, preparation of masks for electrolithography, production of hybrid structures with Al mirrors and Bragg Circular Cavities. All of this is quite unique in this day and age, when specialization is deep, but limited to a selected narrow field.

My overall assessment of the dissertation is very high. I appreciate its outstanding scientific level, high level of innovation of presented results, the comprehensive knowledge contained in the introductory part, as well as the excellent editorial quality of the dissertation. Anyway, out of my duty as a reviewer, I would have a few minor comments or questions. I list them in the order of relevancy below.

Minor, but still relevant issues:

Page 39 – The Author states “the CX state is degenerate, and its emission is circularly polarized”. I do not agree with the statement that charged exciton in the absence of magnetic field emits light in circular polarization. (The same concerns neutral exciton emission confined in a perfectly symmetric quantum dot.) Any superposition of energy degenerate |+1> and |-1> exciton states is also an eigenstate of the exciton. This means that the emission of charged exciton or symmetric neutral exciton can occur in any polarization, depending on the actual combination of states it is formed from at the moment of recombination. As a result, the charged exciton emission is, in typical case, just unpolarized.

Page 53 – how factor V_0 encoding cavity mode volume should be understood in the formulas 2.99 and 2.100, that is in the discussion of a spontaneous emission of an emitter in free space?

Page 103, but also a general remark: I do not see a clear justification for etching of mesas, in principle facilitating optical spectroscopy studies of individual quantum dots. The mesas are of a few μm diameter, while the diameter of the excitation spot is comparable or even smaller, of around 2 μm. The surface density of the studied quantum dots is low, of around of $10^8/cm^2$, which means a separation of around 1.5 μm between quantum dots (which are, in addition, dispersed in emission energy, so do not overlap in the spectrum). At the same time, I would expect that the presence of a surface on the edges of the mesa can provide a non-radiative recombination channel, and lead to undesired increase of the magnitude of charging effects and fluctuations of the electric field in the vicinity of a studied quantum dot. Indeed, the Author states that charged exciton recombination dominates the emission spectra (Phys. Rev. B 101, 195304 (2020).) or that the exciton lifetimes are prolonged in mesas due to the presence of the electric field (Nanophotonics, 11 1515 (2022).). In which way are the mesas helpful?

Page 105 – In the discussion of temperature induced broadening: how linewidth of transitions of individual quantum dots was determined? Typically one observes a transition in the form of a narrow, zero-phonon line and phonon sidebands of much larger width. The Author mentions only one linewidth and does define the curve that is fitted.

Page 157 - Would the Author expect that neutral exciton (instead of the studied charged exciton) emission could ensure higher single photon purity and a larger degree of indistinguishability than the one observed?

Minor, less relevant issues:

Page 8 – The Author states that "simultaneous coupling [to the optical mode] of both transitions is required to boost the generation of entangled photon pairs from the XX-X cascade", but neglects in the following discussion of cavity designs utilized for generation of entangled photon pairs, the design involving two coupled micropillars (Dousse *et al.*, Nature'2010 [Ref. 122]). This design ensures both, exciton and biexciton lines coupled to the optical modes, without compromising mode quality factor.

Page 11 – The reference 169 is mentioned as an example of work, where quantum dots positions are determined by photoluminescence imaging. In fact, in this work, photoluminescence scanning in-situ has been utilized.

Page 38 – When discussing energy order and binding energies of excitonic complexes confined in semiconductor quantum dot it would be worth to emphasize that these quantities strongly depend on quantum dot geometry. In particular, the energy order of excitonic complexes might be different than the one discussed and shown in Figure 2.10 (see, e. g., Few-particle energies versus geometry and composition of InxGa1_{-x}*As/GaAs self-organized quantum dots*, A. Schliwa *et al.*, PRB 79, 075443 (2009).)

Page 41 – When presenting a rate equation model for simulation of quantum dot emission intensity it would be worth to point out that any charged states of the quantum dot are neglected and that the quasiresonant excitation is assumed (in the case of non-resonant excitation carriers are captured to the dot separately and charged states are unavoidable).

Page 51 – the “>>” and “<<” should be changed to ”>” and “<”, when defining weak and strong coupling limits, as there is no intermediate limit of coupling when relevant quantities are comparable.

Page 54 - A minor detail: I would add “emitter oscillator strength” to the Box 6, summarizing factors decisive for the light-matter coupling strength.

Page 75 - text and Nanophotonics 11, 8 (2022). How the statement “the FSS of (50±5) μeV determined for an X line was related to the post-processing-induced imperfections and not the intrinsic property of the QD itself.” should be understood? Does the Author expect that post-processing introduced any electric field to the structure?

Page 104 – Could the non-negligible linear polarization of the charged exciton emission be related to a non-negligible transfer of linear polarization of the excitation? Is it excluded that the excitation was of a quasi-resonant type?

Page 135 – A short-lived contribution from higher-excited states can also result in shorter lifetimes of QD ensemble emission in comparison to a few-particle exciton complexes.

Typo errors/technical issues:

Page 57 – migration of adatom migration -> adatom migration

Page 114 – The expression “micro-PL” instead of “high-resolution PL” would be more unequivocal.

Page 118, Figure 6c – Both arrows in the case of the G-band should be directed to the left.

Page 134 – Uncertainty of the provided decay time constant of 10 ps seems too small, as overall temporal resolution of the setup is 80 ps.

The above comments and questions do not undermine in any way the exceptional scientific quality of the work nor my very positive reception of it as a reviewer. From my reading of the presented thesis, the Author appears as a well-skilled, mature researcher, who is able to conduct scientific research and to communicate its results.

In conclusion, the dissertation submitted for review by M.Sc. Paweł Holewa provides an original solution to a scientific problem of elaboration of efficient quantum light sources for the third telecommunication window. It confirms a broad theoretical knowledge of the Author in the discipline of physical sciences and his ability to conduct scientific work as an independent researcher. In my opinion, the dissertation more than satisfies the customary requirements and formal requirements for doctoral dissertations set out in Article 187 of the Act of 20 July 2018. "Law on Higher Education and Science" (Dz.U.2020, item 85 as amended). In view of the above, **I request that MSc Paweł Holewa be admitted to the further stages of the procedure for the award of the degree of Doctor of Physical Sciences**. Taking into account the excellent level and quality of the presented research, the high originality and importance of the results, and the significant value of the research to the development of the field (which is, in particular, the production and study of efficient quantum emitters based on InP/InAs operating in the third telecommunication spectrum), **I request with a full conviction that the dissertation be distinguished**.

Jan Suffczyński