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Optimizing the InGaAs/GaAs Quantum Dots for 1.3 μm Emission

A. Maryński[a], P. Mrowiński[a], K. Ryczko[a], P. Podemski[a], K. Gawarecki[b], A. Musiał[a], J. Misiewicz[a], D. Quandt[c], A. Strittmatter[c,†], S. Rodt[c], S. Reitzenstein[c] and G. Sęk[a,*]

[a]Laboratory for Optical Spectroscopy of Nanostructures, Department of Experimental Physics, Faculty of Fundamental Problems of Technology, Wrocław University of Science and Technology, Wybrzeże Wyspiańskiego 27, 50-370 Wrocław, Poland

[b]Department of Theoretical Physics, Faculty of Fundamental Problems of Technology, Wrocław University of Science and Technology, Wybrzeże Wyspiańskiego 27, 50-370 Wrocław, Poland

[c]Institute of Solid State Physics, Technische Universität Berlin, Hardenbergstr. 36, D-10623 Berlin, Germany

Hereby we present comprehensive experimental and theoretical study on fundamental optical properties and electronic structure of GaAs-based quantum dots grown using metalorganic chemical vapor deposition technique. Substantial redshift of emission to the second telecommunication window of 1.3 μm is obtained via strain engineering utilizing additional capping layer of $In_{0.2}Ga_{0.8}As$ in this context referred to as strain reducing layer. The quantum dot structure has been experimentally characterized by means of photoreflectance spectroscopy and power-dependent photoluminescence revealing 3 transitions originating from hybrid states confined in an asymmetric double quantum well formed of the wetting layer and strain reducing layer, as well as higher states of the quantum dots themselves with the first excited state transition separated by 67 meV from the ground state transition. Origin of the observed transitions was confirmed in theoretical modelling using 1-band single-particle approach for the quantum well part, and excitonic quantum dot spectrum was obtained within 8 band $\boldsymbol{k}\cdot\boldsymbol{p}$ formalism followed by configuration interaction calculations, respectively. Additionally, photoluminescence excitation spectroscopy measurements allowed identifying a spectral range for efficient quasi-resonant excitation of the investigated quantum dots into the 2D density of states to be in the range of 835–905 nm.

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1. Introduction

Telecommunication and quantum communication applications require light sources operating in the near infrared range where the spectral windows for standard silica fibers of 1.3 μm and 1.55 μm can be identified as featuring no dispersion and the lowest optical losses, respectively. The main advantage of using solid state systems with quantum dots (QDs) as an active region of optoelectronic devices is their flexibility and as a result possibility of tailoring their properties for a specific application. For more advanced non-classical light sources (e.g., of single photons, entangled photon pairs, etc.) indispensable for secure quantum communication and local quantum networks, QDs are the structures of choice due to their discrete energy levels and Fock-state-like statistics of emission. To take full advantage of their application potential a thorough understanding of the optical properties and underlying electronic structure as well as their determinants is necessary for each new group of structures. For practical implementations of short-range quantum communication networks and protocols, spectrally well-isolated and time-stable transitions in the 1.3 μm range and deep confining potential are beneficial. For application as an active material of single-photon sources the emission of single QDs at cryogenic temperatures has to be explored. One of the approaches to target these challenges is to use GaAs-based QDs and redshift of their emission (typically centered below 1 μm at 10 K) towards telecommunication range. These structures combine the advantage of mature fabrication and material processing technology with compatibility and straightforward integration. One of efficient methods to reach emission at 1.3 μm is to engineer the strain in InAs/GaAs QDs: usage of InGaAs strain reducing layer [1–10], bilayer of different-size QDs was the first layer acting as a seeding layer and modifying the strain conditions for the second one [11–13], increase of QD height by growth up to second critical thickness [14], or nitrization of InAs/GaAs QDs [15].

In this work, the approach with additional strain reducing layer introduced during epitaxial growth of InGaAs/GaAs QDs was applied to shift the QD emission to 1.3 μm. The optical properties were determined and the electronic structure was calculated by $\boldsymbol{k}\cdot\boldsymbol{p}$ method. This enabled for full description of the QD structure and iden-

*corresponding author; e-mail: Grzegorz.Sek@pwr.edu.pl

†Present address: Institute of Experimental Physics, Otto von Guericke University Magdeburg, D-39106 Magdeburg, Germany

tification of both possible efficient excitation and carrier escape channels. This is the first characterization step of fundamental optical and electronic properties of such nanostructures aiming at applications as an active region in non-classical light sources, e.g., for quantum key distribution within local quantum networks which could be further realized based on these structures at cryogenic temperatures.

2. Experimental

The sample was grown by metalorganic chemical vapor deposition (MOCVD) on (001) GaAs substrate (see Fig. 1c with the layer structure). The active region consists of a single layer of self-assembled $In_{0.75}Ga_{0.25}As$ QDs grown in the Stranski–Krastanov mode on wetting layer (WL) capped with nominally 4 nm thick $In_{0.2}Ga_{0.8}As$ strain reducing layer (SRL) [16] and is sandwiched between GaAs layers and AlGaAs barriers. The size of the QDs determined based on the cross-sectional transmission electron microscopy (TEM) study (Fig. 1b) is 6–8 nm in the growth direction and around 30 nm in base size with strongly inhomogeneous indium distribution within the QD volume.

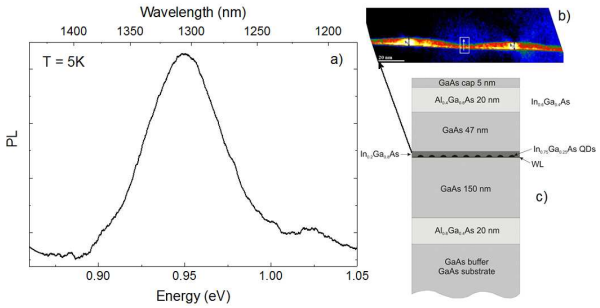

Fig. 1. (a) Low-excitation and low-temperature (5 K) photoluminescence spectrum of the inhomogeneously-broadened ensemble of InGaAs/GaAs QDs under non-resonant excitation at 660 nm; (b) cross-sectional TEM image of the InGaAs/GaAs QDs (courtesy of AG Lehmann, TU Berlin), (c) sample layer design of investigated QD structure.

For optical characterization three experimental techniques were utilized: photoreflectance (PR), photoluminescence (PL) and photoluminescence excitation spectroscopy (PLE). All studies were performed at cryogenic temperatures and the optical response of the whole QD ensemble was analyzed. For the photoreflectance measurements the sample was mounted in a He refrigerator providing the lowest base temperature of 10 K. The experiment was carried out in a bright configuration in which light from a halogen lamp is reflected from the sample surface, whereas the modulation at 278 Hz (mechano-optical modulator) is provided by 660 nm laser. The reflection signal is further spectrally resolved using a monochromator and detected with InGaAs single-channel photodiode using phase-sensitive detection with lock-in amplifier. Photoluminescence study was performed in a standard experimental configuration with 0.4 numerical aperture microscope objective used for both, focusing the non-resonant excitation laser (660 nm) on the sample surface (down to approximately 2 μm spot size) and collecting the emission, which is then imagined on an entrance slit of a spectrometer equipped with liquid nitrogen-cooled InGaAs linear array detector. For that as well as for photoluminescence excitation spectroscopy experiments the sample was mounted in the He continuous-flow cryostat providing the sample temperature of 5 K. In the case of PLE experiment a Ti:sapphire laser operated in a 160 fs mode with 76 MHz repetition rate with 100 μW average power was used as a tunable excitation source providing the spectral resolution of about 1 nm.

3. Results and discussion

In order to confirm the expected influence of the strain reducing layer on the ground state emission energy of the InGaAs/GaAs QDs a photoluminescence spectrum at low-excitation and low-temperature was measured (Fig. 1a). The maximum intensity of the QD ensemble emission is at 1305 nm (5 K) in agreement with the structure design. The investigated QDs form a rather homogeneous ensemble with the inhomogeneous broadening of only 35 meV ensuring many of single emitters with their ground state transition close to the target wavelength of 1.3 μm. The observed broadening is comparable to results for state-of-the-art QD structures of similar approaches grown using molecular beam epitaxy (MBE) technique. Due to the high homogeneity, the QD density will need to be further reduced or sample surface needs to be patterned in order to resolve well single emission lines originating from individual optical transitions, which is however beyond the scope of this work.

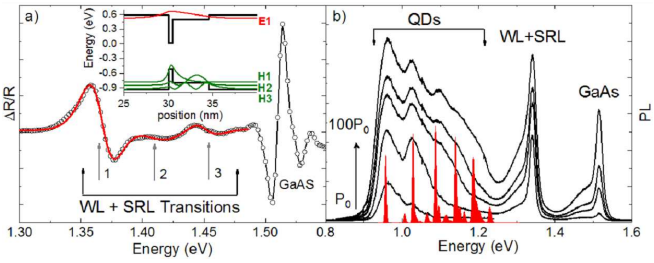

Fig. 2. (a) Low-temperature (10 K) photoreflectance spectrum of the InGaAs/GaAs QD structure in the range of transitions in InGaAs WL and SRL as well as GaAs substrate; experimental data marked in gray (open symbols and solid line) and respective fit as red solid line; inset: band edge diagram (black solid lines) with the envelopes of the wave functions of the first E1 electron (red solid line) and three lowest H1, H2, H3 hole single particle levels calculated within 1-band $\boldsymbol{k}\cdot\boldsymbol{p}$ model, (b) low-temperature (5 K) excitation power-dependent photoluminescence spectra (black solid lines) overlaid with an excitonic spectrum calculated within the 8 band $\boldsymbol{k}\cdot\boldsymbol{p}$ approach combined with configuration-interaction method for realistic QD parameters (red curves).

To identify the excited state transitions in the QDs high-excitation photoluminescence experiment was carried out (Fig. 2b). The excitation power-dependent emission spectra revealed at least 2 clearly-resolved excited states transitions in the QDs as well as additional peaks of even higher-order transitions. It was possible to observe them due to the common state filling effects at higher excitation powers. The first excited state (which can be called "p-shell" in contrast to the ground state referred to as "s-shell") is well-separated from the ground state transition — the so-called s–p splitting equals to 67 meV which is still relatively large taking into account expected energy level separation for this kind of dots.

Based on the QD parameters determined from structural characterization and nominal (designed) parameters of the adjacent layers the excitonic spectrum of the QD was modelled and compared with the experiment (red spectrum in Fig. 2b). First, single particle states were calculated within the 8-band $\boldsymbol{k}\cdot\boldsymbol{p}$ model with realistic structure parameters including inhomogeneous (Gaussian assumed) indium composition in the QD, influence of strain within the continuous elasticity approach and piezoelectricity up to the second order on the band structure [17]. Excitonic spectrum was further calculated using configuration-interaction method with basis containing 40 electron and 40 hole single-particle states. The theoretically obtained excitonic spectrum corresponded well to the results of high-excitation PL measurements — see the comparison in Fig. 2b, where the calculated spectrum is shown in red (artificially broaden by a constant value to make it more realistic). The calculations additionally revealed that the energy splitting is shared in the ratio approximately 2:1 between the conduction and valence band single particle states.

For high excitation, states in the InGaAs quantum wells (WL and SRL) as well as emission from the substrate are also visible in the spectrum (marked in Fig. 2b), but PL spectra are insufficient for a detailed analysis of the optical response in this spectral range. Therefore, to be able to describe and understand the electronic structure in the WL and SRL range an absorption-like experimental technique was used, i.e. modulation spectroscopy in a form of photoreflectance. Low-temperature (10 K) PR spectrum in the relevant spectral range is presented in Fig. 2a. At 1.52 eV, a clear resonance from the GaAs substrate can be identified. At lower energies in the range of 1.35–1.48 eV three other transitions can be resolved — supported by the fitting curve shown in red in Fig. 2a. To understand their origin, calculations of the single-particle energy levels in the WL-SRL quantum wells were conducted within the 1-band $\boldsymbol{k}\cdot\boldsymbol{p}$ model including strain and assuming nominal structure parameters. The obtained wave functions envelope for the lowest electron (red) and hole (green) states confined within the WL-SRL potential are presented in the inset of Fig. 2a. First of all, these results show that there is only 1 electron confined level in the conduction band and 3 hole levels confined in the valence band. The related transitions lead to the resonances observed experimentally in the PR spectrum. For nominal system parameters, the energy difference between the experimentally and theoretically determined transition energies is in the range of 20 meV. To get more exact quantitative agreement one would need to tune the system parameters slightly, which is however not necessary for proper interpretation of the origin of observed transitions, which was the primary goal. The second important observation is that the WL and SRL states cannot be treated separately — observed states origin from a coupled, double QW system.

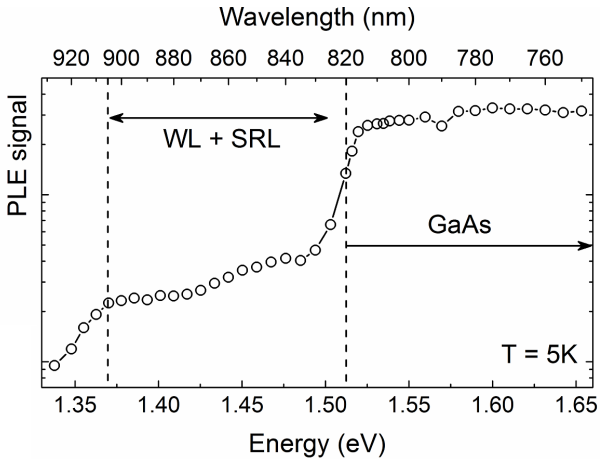

Fig. 3. Low-temperature (5 K) photoluminescence excitation spectrum of InGaAs/GaAs QDs. The excitation wavelength (Ti:sapphire laser with 160 fs pulses at 76 MHz repetition rate and average excitation power of 100 μW) was varied in the range of InGaAs WL and SRL and GaAs substrate and the intensity maximum of the QD ensemble emission was evaluated at each excitation wavelength. A GaAs and WL + SRL absorption edge is clearly visible at 823 nm and 905 nm, respectively.

Their nature has been further confirmed in a PLE experiment (Fig. 3). In that case the excitation wavelength has been varied in the range where the GaAs- and WL-SRL-related transitions were observed in previous experiments, and the maximum of emission intensity of the QD ensemble at the ground state has been monitored. At around 820 nm the absorption edge of the GaAs can be identified. Due to change in the density of states from 3D to 2D and a difference in thickness of the absorbing layers, the signal drops substantially when going to lower energies (by about 1 order of magnitude). This relatively sharp transition is followed by a slow, gradual decrease in the PLE signal (with some modulations overimposed), and finally by a second edge at around 905 nm corresponding to the absorption edge of the WL-SRL part of the structure. Temperature-dependent PLE study up to room temperature (not shown here) revealed low energy tail spanning down to 1.26 eV beyond this second absorption edge, most probably related to localized states

in the WL. The performed PLE measurements allowed identifying a spectral range below the GaAs absorption edge at which QDs can be efficiently excited to be 835–905 nm. Such a quasi-resonant excitation could be very beneficial as an excitation scheme to obtain higher single-photon purity and indistinguishability as well as to diminish dynamical broadening (spectral jitter) of the single QD emission lines. This is mainly due to the fact that less carriers with high-energy are created in the structure therefore less processes are involved in relaxation to the QD ground state. It lowers the time jitter for consecutive emission processes. Identification of efficient quasi-resonant excitation channels is crucial for future study on the level of single QDs and evaluating potential of these InGaAs/GaAs QDs for more advanced nanophotonic and quantum optics applications.

4. Conclusions

Hereby, we have shown that applying an $In_{0.2}Ga_{0.8}As$ strain reducing layer in MOCVD-grown GaAs-based QDs allowed redshifting their emission towards application relevant spectral range within the 2nd telecommunication band at low temperature. In total, 3 optical transitions in the adjacent quantum wells, namely wetting layer and strain reducing layer, were identified in the photoreflectance experiment and their coupled QW character was proven using single-particle 1-band $\boldsymbol{k}\cdot\boldsymbol{p}$ modelling. The overall absorption edge related to these QW states was determined via photoluminescence excitation spectroscopy to be at 905 nm specifying the range for efficient quasi-resonant QD excitation. The QDs emission at low temperature corresponded well to the designed wavelength (1.3 μm) with an inhomogeneous broadening of the QD ensemble of 35 meV indicating on a high ensemble homogeneity comparable to state-of-the-art of MBE-grown QDs. High-excitation photoluminescence allowed to identify higher energy transitions in the QDs with the first excited state (p-shell) well-separated from the ground state transition by 67 meV ensuring good isolation of the s-shell transition in the QD important for further study on the single QD level and advanced nanophotonic applications and possibility of operation at elevated temperatures. The experimental findings are in agreement with results of theoretical calculations of excitonic spectrum within 8 band $\boldsymbol{k}\cdot\boldsymbol{p}$ and configuration-interaction formalism, indicating additionally twice larger contribution of electron energy (in comparison to the hole state) to this splitting.

Acknowledgments

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