Quantum confinement in MOVPE-grown structures with self-assembled InAs/GaAs quantum dots

K Kuldová1,2,4, M Molas2, J Borysiuk2,3, A Babiński2, Z Výborný1, J Pangrác1, J Oswald1

1 Institute of Physics of the AS CR, v. v. i., Cukrovarnická 10, CZ-162 00 Praha 6, Czech Republic
2 Institute of Experimental Physics, University of Warsaw, Hoża 69, PL-00-681 Warszawa, Poland
3 Institute of Physics PAS, Al. Lotników 32/46, PL-02-668 Warszawa, Poland

E-mail: kuldova@fzu.cz

Abstract. In this communication we report on low-temperature, micro-photoluminescence study of quantum confinement in MOVPE-grown structures with InAs/GaAs quantum dots (QDs) with GaAs and/or strain reducing InGaAs/GaAs capping. We focus our attention on sharp emission lines, which appear in both structures at energies up to 80 meV below the wetting line emission. Power-dependent measurements confirmed their attribution to single excitons as well as biexcitons. Negative binding energy of biexcitons with systematic dependence on their energy was observed. It has been proposed that the investigated emission lines result from radiative recombination in flat non-fully developed QDs in the investigated structure. The attribution is confirmed by transmission electron microscopic analysis of investigated structures.

1. Introduction

Scientific interest in self-assembled InAs/GaAs quantum dots (QDs) is driven by interesting fundamental physics and numerous potential applications. One of those applications is the area of inexpensive, low threshold and high output lasers emitting in the 1.55 \( \mu \text{m} \) communication band. Tuning the emission energy within the infrared band is a challenge for technology. Standard approach, namely covering the QDs with a GaAs capping layer (CL), leads to a rapid shrinkage of the QD heights, which results in a blue shift of the QDs luminescence at least to \( \sim 1.2 \, \mu \text{m} \). A promising way to avoid the blue-shift of the QD emission is the application of a thin (5 nm) InGaAs strain reducing layer (SRL), which covers QDs [1, 2]. The understanding of processes, which are at play during formation of such QDs with SRL is of prime importance from technological point of view. Also important is the knowledge of possible confinement potentials, which can trap photoexcited carriers, potentially affecting the performance of optoelectronic devices. The study of carrier localizations in the energy region of the wetting layer (WL) is the first step of such study [3, 4].

4 To whom any correspondence should be addressed.
2. Experimental

Structures with InAs/GaAs QDs and ternary SRLs (23% of In) were prepared by low pressure metalorganic vapor phase epitaxy (LP MOVPE) in AIXTRON200 on Si(100) GaAs substrates using Stranski–Krastanow growth mode. The InAs QDs were covered by 30 nm of GaAs CL or by 4 nm of In$_{0.23}$Ga$_{0.77}$As SRL and 30 nm of GaAs CL. Reflectance anisotropy spectroscopy in-situ measurement (EpiRAS 200TT–LayTec) was used to monitor the formation and development of InAs QDs, as well as the growth of InGaAs and GaAs layers [5]. Mesa-patterning on the samples limits the number of photoexcited dots and allows to reproducibly locate investigated dots. Samples for optical measurements were mounted in a He-flow cryostat and measured at temperature 4.2 K. The 532 nm laser line was used for non-resonant optical excitation. The excitation light was delivered and the photoluminescence (PL) collected via a microscope objective (spot size ~1 µm), dispersed using a 0.5 m monochromator and detected using a liquid-nitrogen-cooled Si charged coupled device camera. The macro PL was detected by a Ge detector using standard lock-in technique.

3. Results and discussion

The PL spectrum from the investigated structure is composed of the emission from the GaAs barrier, self-assembled QDs, and the emission from the WL (figure 1). Note the red shift of PL from the QDs and the WL due to the SRL capping. Micro-PL spectra from both samples in the WL energy region measured on a 5×5 µm mesa are presented in figure 2. The PL band at the 1.36 eV is attributed to radiative recombination to the doubly ionized Cu$_{Ga}$ acceptor, which corresponds to the 0.156 eV acceptor level [6]. In the case of the SRL sample the Cu$_{Ga}$– related band co-exists with the WL emission. The Cu$_{Ga}$– related band originates from the GaAs substrate and can be bound up with Cu contamination of polished and etched surface of SI GaAs wafers [7].

We focus our attention on sharp emission lines, which appear in both structures at energies up to 80 meV below the WL emission.

To classify these emission lines, power-dependent PL spectra were measured. In figures 3 and 4 are represented µ-PL spectra of excitonic lines for increasing excitation power density $P_{excit}$ of both samples respectively. The power dependence of each line for low excitation is characterized by $(P_{excit})^n$, the exponents $n$ are presented for each line in figures. In insets are shown examples of corresponding fits.

In both structures pairs of sharp lines (B, C and D, E for the sample without the SRL and J, K for the sample with the SRL) can be distinguished.
The emission lines have similar power-dependence: lower (higher) energy line from each pair emerges with linear (quadratic) excitation power density dependence. Limited number of lines observed in the energy region 1.3 - 1.4 eV for the sample without the SRL allows us to attribute lines B and D to two different excitons and lines C and D to their respective biexcitons in QDs. Binding energies of those biexcitons are equal to $E_{B,C} = -1.59$ meV and $E_{D,E} = -2.51$ meV. There are two types of features in the spectra of the SRL sample: lines J, K and L are sharp with the FHWM comparable to that of figure 3; the linewidth of H, M and N lines is at least 5 times larger. The J, K and L lines have linear, quadratic and sublinear excitation power density dependence. We attribute these lines to the exciton, biexciton and a charged trion respectively. Binding energies of the biexciton is equal to $E_{J,K} = -1$ meV. The line I has a complex structure composed of at least three closely spaced peaks. In our opinion this line is composed of the neutral exciton, biexciton and charged exciton from a single QD with a very low binding energy. In agreement with [8], the biexciton binding energies in both samples decrease with increasing transition energy and their values follow a linear dependence (see figure 5).

The order of appearance of few-particle states (exciton X, biexciton XX and charged excitons, i.e., positive and negative trions $X^+$, $X^-$) are discussed in [9] for a large number of QD structures of varying size, shape, and composition. In [8] the binding energy for flat InAs/GaAs MOVCD QDs decreases with increasing transition energy changing its sign at about 1.24 eV. The binding to antibinding transition is attributed to three-dimensional confinement, quenching correlation, and exchange and causing local charge separation.

The results of optical measurements can be confronted with an analysis of the sample morphology. Cross-sectional transmission electron microscopy (TEM) reveals that self-assembled InAs QDs in sample without the SRL are flat, with the height of 2.5 nm and base diameter ~25 nm (figure 6). This confirms results of atomic force microscopy of an uncapped sample QDs of height 2.5 - 3.5 nm and diameter 15 - 25 nm (not shown here). The QDs in SRL samples (figure 6, bottom) are higher (5 nm)
and thinner with diameter ~20 nm as predicted in [1]. Beyond fully developed self-assembled InAs QDs the presence of lower objects (~2 × WL height) have been observed on both samples with significantly lower density. Their diameter is comparable with this of fully developed QDs. In our opinion these not-fully developed QDs are the features confining excitons investigated in our work. Although their density seems to be much lower than the density of self-assembled QDs, they may influence the performance of possible devices relaying on transport of excitons in the WL. They also seem to be interesting from fundamental point of view, as the biexciton binding energy may vanish in those objects.

Figure 6. The TEM images of the sample without (top) and with the SRL (bottom). The InAs WL for the GaAs CL sample, In$_{0.23}$Ga$_{0.77}$As SRL in contact with WL for the SRL sample, self-assembled InAs QDs (SQD), and non-fully developed InAs QDs (NQD) are identified.

4. Conclusions
In conclusion, we have performed microluminescence studies of MOVPE-grown structures with InAs/GaAs QDs with GaAs capping layer and InGaAs strain reducing layer. We have observed excitonic and biexcitonic emission lines, which have been attributed to the not-fully developed QDs in those structures. The biexciton binding energy was observed to linearly depend on the energy of exciton, vanishing below 1.30 eV. The spectroscopic results were confirmed by transmission electron microscopy, which revealed the presence of those objects.

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References
[1] Hazdra P, Oswald J, Komarnitskyy V, Kuldová K, Hospodková A, Hulicius E and Pangrác J 2009 Superlattices and Microstructures 46 324
[2] Hospodková A, Hulicius E, Pangráč J, Oswald J, Vyskočil J, Kuldová K, Šimeček T, Hazdra P and Caha O 2010 J. Crystal Growth 312 1383
[3] Babinski A, Jasinski J 2002 Thin Solid Films 412 84
[4] Babiński A, Borysiuk J, Kret S, Czyż M, Golnik A, Raymond S and Wasilewski Z R 2008 Appl. Phys. Lett. 92 171104
[5] Hulicius E, Oswald E, Pangráč J, Vyskočil J, Hospodková A, Kuldová K, Melichar K and Šimeček T 2008 J. Crystal Growth 310 2229
[6] Wang Z G, Gislason H P, and Monemar B 1985 J. Appl. Phys. 58 230
[7] Fang Z Q, Look D C, and Jones R L 1997 J. Electronic Materials 26 L29
[8] Rodt S, Heitz R, Schliwa A, Sellin R L, Guffarth F, and Bimberg D 2003 Phys. Rev. B 68 035331
[9] Schliwa A, Winkelinkemper M, and Bimberg D 2009 Phys. Rev. B 79 075443