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Photoluminescence study of InGaAs/GaAs quantum dots with bimodal inhomogeneous broadening

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Abstract. The comparative studies of optical and structural properties of InGaAs/GaAs quantum dots grown in Stranski–Krastanov growth mode by molecular-beam epitaxy and metal-organic chemical vapor deposition is presented. An analysis of the photoluminescence at ultralow pump levels resulted that the quantum dots ensemble grown by metal-organic chemical vapor deposition exhibits photoluminescence corresponding to quantum dots ground state and at the same time ensemble of quantum dots grown by molecular-beam epitaxy demonstrates the bimodal behavior which can be explained by the presence of two characteristic ensembles of InGaAs/GaAs quantum dots with different sizes and different peaks of photoluminescence. The results on InGaAs/GaAs quantum dots studying by transmission electron microscopy are presented and discussed as well.

Keywords: molecular-beam epitaxy, metal-organic chemical vapor deposition, gallium arsenide, InGaAs, Stranski–Krastanov growth mode

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Исследование фотолюминесценции квантовых точек InGaAs/GaAs с бимодальным неоднородным уширением

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Аннотация. Представлены сравнительные исследования оптических и структурных свойств квантовых точек InGaAs/GaAs, выращенных методом Странски-Крастанова с использованием молекулярно-пучковой и газофазной эпитаксии. Анализ спектров фотолюминесценции при сверхнизких уровнях накачки показал, что массив квантовых точек, выращенных методом газофазной эпитаксии, проявляет фотолюминесценцию, соответствующую переходам через основные состояния, а массив квантовых точек, выращенных методом молекулярно-лучевой эпитаксии, проявляет бимодальное поведение - наличие двух характерных ансамблей квантовых точек с разными размерами, соответствующих двум пикам фотолюминесценции. Представлены и обсуждены результаты исследования квантовых точек InGaAs/GaAs методом просвечивающей электронной микроскопии.

Ключевые слова: молекулярно-лучевая эпитаксия, газофазная эпитаксия, арсенид галлия, InGaAs, механизм Странски-Крастанова

Финансирование: Авторы из ФТИ им. А.Ф. Иоффе выражают благодарность за поддержку проекту РНФ № 22-19-00221, https://rscf.ru/project/22-19-00221/ в части разработки структуры, эпитаксии методом молекулярно-пучковой эпитаксии и исследования спектров фотолюминесценции структур, выращенных методом молекулярно-пучковой эпитаксии. А.А. Надточий и Н.В. Крыжановская благодарят проект FSRM-2023-0010 Министерства науки и высшего образования Российской Федерации за поддержку в части исследований спектров фотолюминесценции структур, выращенных методом газофазной эпитаксии. Авторы из Университета ИТМО выражают благодарность за поддержку федеральный проект «Передовые инженерные школы» в части исследований методом просвечивающей электронной микроскопии структур, выращенных методом газофазной эпитаксии.

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Introduction

The usage of optically pumped quantum dots (QDs) micropillar lasers with vertical microcavities is preferable to realize the optical reservoir computing (RC) scheme [1–4] due to low power consumption of photonic RC, high spectral homogeneity, small pitch and diameters of microlasers in relation with vertical-cavity surface-emitting lasers (VCSELs) case [5]. Due to possibility of the precise control of microlasers diameter, it is possible to realize a dense array of spectrally homogeneous microlasers emitting in a frequency-range of about 50 GHz (~200 μeV) required for RC [3].

This paper presents the results on study of the optical properties of 980 nm range InGaAs QDs grown by molecular-beam epitaxy (MBE) according to the Stranski-Krastanov growth mode and a comparison with the results for InGaAs QDs grown by metal-organic chemical vapor deposition (MOCVD).

Materials and Methods

For the first structure InGaAs QDs were formed by deposition of $In_{0.63}Ga_{0.37}As$ layer with thickness of 2.6 monolayers (ML) using the Stranski-Krastanov mode and MBE. A 300 nm thick GaAs matrix layer was located between two 35 nm thick $Al_{0.23}Ga_{0.77}As$ barriers. The QD layers were placed in the middle of the matrix layer. The growth temperature of the barrier layers, as well as the matrix layer, was 580 °C. Epitaxy of the QDs layers was carried out at 490 °C. The growth rate was a 0.55 E/s. The exposure time in arsenic flow before the deposition of the 5 nm thick GaAs capping layer was 30 seconds. After the capping layer deposition, the temperature was increased up to 580 °C, and the samples were annealed for 240 seconds to reduce the inhomogeneous broadening of QDs in the ensemble. Arsine, tertiary butylarsine, trimethylgallium, trimethylaluminum, and trimethylindium were used as precursors for MOCVD growth of QDs (structure 2). A 320 nm thick GaAs matrix layer was located between two 35 nm thick $Al_{0.42}Ga_{0.58}As$ barriers. The QD layers were placed in the middle of the matrix layer. The growth temperature of the barrier layers, as well as the matrix layer, was 700 °C. Epitaxy of the QDs layers was carried out at low temperature (580 °C), and the growth rate was a 0.68 E/s. The thickness of the GaAs capping layer was fixed at 2 nm. The exposure time before the deposition of the capping layer was 30 seconds. After the capping layer deposition, the temperature of the substrate holder increased up to 615 °C aimed with realize the QDs high-temperature annealing. The photoluminescence (PL) spectra were studied at 13 K in a wide range of excitation power densities. Optical pumping was carried out by a Yag:Nd laser at a wavelength of 532 nm with a 150 mW CW output power, which corresponds to the excitation power density ~ 5 kW/cm^2. The range of laser power attenuation using neutral filters was $(1-3\cdot10^{-6})$. Temperature studies of the PL spectra were also carried out in the temperature range of 13–325 K at a moderate excitation power of 4.5 mW.

Transmission electron microscopy (TEM) studies were carried out using a JEM2100F electron microscope (Jeol) at an accelerating voltage of 200 kV. Samples were prepared in cross-sectional geometry according to a conventional technique, including thinning by precision lapping and sputtering with argon ions at the final stage until perforation.

Results and Discussion

The results of TEM studies are shown in Fig. 1. In both cases, the estimated QDs surface density is about $7\cdot10^{9}$ cm^{-2}. In structure 1 grown by the MBE, one can see the QDs with 2.8–3.9 nm range height. The QD outer boundary has a thin darker contrast than the surrounding GaAs

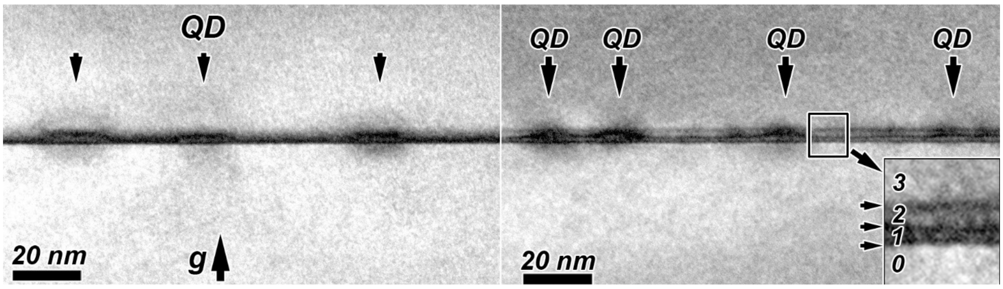


Fig. 1. TEM image of structure 1 (left panel) and structure 2 (right panel)

matrix and the inner part of the QD. It can be assumed that the wetting layer (WL) composition estimated using a TEM contrast [6] has mole fraction of indium about 0.2 ($In_{0.2}Ga_{0.8}As$). The thickness of the WL is no more than 0.8 nm. The central part of QD has a less contrast, which corresponds to a higher indium mole fraction (more than 40%).

In structure 2 grown by the MOCVD, the InGaAs (layer 1, cf. Fig. 1, right panel) is visible on the GaAs layer (layer 0). This InGaAs layer has pyramid thickening, both contrast intensity corresponds to about 40% in mole fraction. The estimated thickness of planar part is about 1.5 nm, while the QDs height limited by the boundary between layers 1 and 3 and is about 4.1 nm. The small capping layer (layer 2) thickness (estimated to about 2.6 nm) result the evaporation of large QDs with thickness exceeded this value at high-temperature annealing.

The results of low-temperature PL studies are shown in Fig. 2. The study of PL spectra at low temperatures makes it possible to avoid the thermal escape of charge carriers from QDs and their subsequent redistribution over the ensemble. In this case, a decrease in the pump level leads to a weakening of the contribution of excited states in the PL spectra (cf. Fig. 2). Thus, the PL spectrum at ultralow excitation densities should be a superposition of emission through the ground states of the QD ensemble [7]. For the structure 1 grown by MBE, the presence of additional lines (shoulder) in the short-wavelength spectral region was observed, which remain unchanged when the pumping is reduced below a certain level (cf. Fig. 2). This behavior was discussed earlier [8, 9] and can be associated with the presence of two ensembles of QDs with different sizes. For the structure 2 grown by MOCVD, a decrease in the half-width of the PL spectrum was observed, and the shape of the spectra was more symmetrical in comparison with the results for structure 1 under identical pumping conditions, which indicates on more homogeneous size distribution of QDs (absence of bimodality).

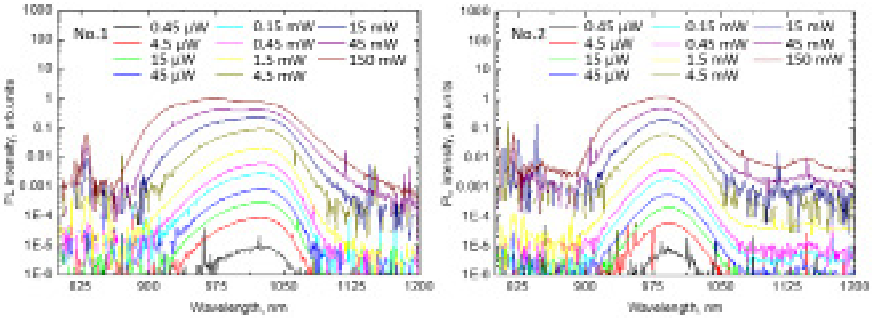
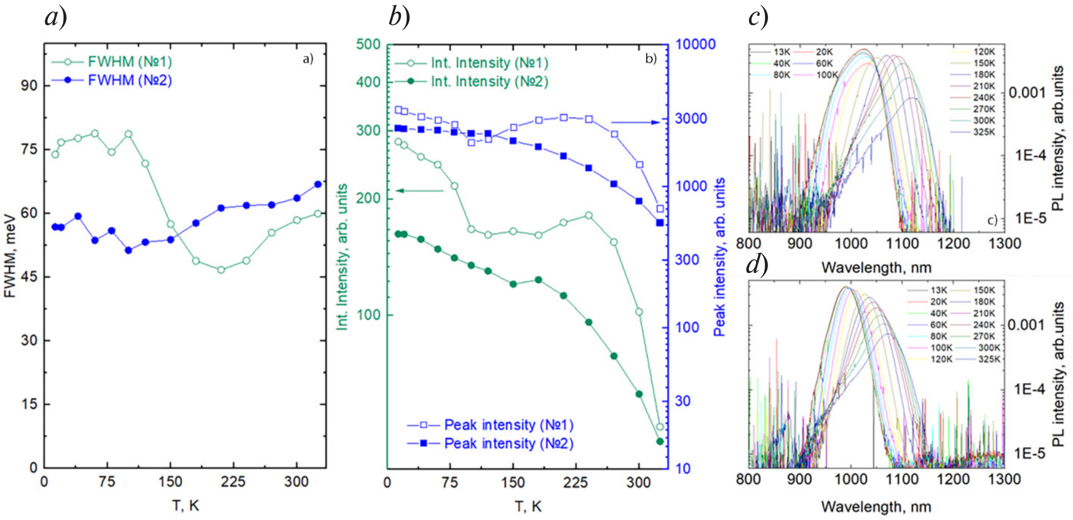


Fig. 2. PL spectra of structure 1 (left panel) and structure 2 (right panel), measured at 13 K



a) *b*) *c*) *d*)

Fig. 3. FWHM versus temperature (*a*) and integrated/peak intensity versus temperature (*b*). PL spectra of structure 1 (*c*) and structure 2 (*d*), measured at different temperatures

Temperature studies of the PL spectra behavior for both structures were carried out at a moderate excitation power (4.5 mW) as well when there is no contribution from the excited states. Temperature studies of the PL spectra (the behavior of the full width at half maximum, FWHM, value, as well as the integral intensity/amplitude of the PL peak versus temperature, cf. Fig. 3) also have confirmed the previously mentioned assumption about the presence of two characteristic ensembles of QDs in the structure 1 grown by the MBE. At low temperatures, in structure 1, a nonequilibrium distribution of charge carriers over QD states is observed and is determined by QDs density. The peak intensity ratio allows one to assume the present of two different QDs ensembles. As the temperature increases, it becomes possible the escape of the carriers from an ensemble with smaller QDs (with weak carrier localization), followed by their capture through a wetting layer into an ensemble with larger QDs, which was previously observed both for the case of bimodal InAs [10, 11] and InGaAs [12] QDs. This process is accompanied by increasing in the peak intensity with temperature for the long-wavelength peak (in studied structure, in the temperature range of 100–210 K, cf. Fig. 3,*b*). Simultaneously, this process is accompanied by a decrease in the intensity of the short-wavelength shoulder, which leads to decreasing in the width of the PL maximum in the given temperature range (100–210 K) and its shifting to lower photon energies [13]. The subsequent rise in temperature leads to suppression of the recapture mechanism.

As a result, an increase in the maximum width of the PL spectrum is observed in the given temperature range. As the temperature increases in structure 2 grown by the MOCVD, a weak temperature dependence of the FWHM values is demonstrated, which indicates a homogeneous distribution of QDs. Fig. 3,*b* also does not demonstrate the growth of the peak PL intensity with increasing temperature, which is typical for the case of bimodal QDs. At low temperatures (up to 120 K), the peak intensity remains constant. Further temperature growth results to a drop in peak intensity, which is typical for thermal escape and further nonradiative recombination of the charge carriers outside the QDs [13].

Conclusion

Low-temperature studies of the PL spectra in a wide range of excitation power densities have been carried out. It was shown that the structure with QDs grown by the MOCVD demonstrates PL through the ground states in the ensemble of QDs. It is shown that the structure with QDs grown by the MOCVD contains one ensemble of QDs. In turn, for the structure with QDs grown by the MBE, the presence of additional lines in the short-wavelength spectral region at a low temperature and pumping was observed, which can be associated with the presence of two ensembles of QDs. Although the MOCVD technique makes it possible to realize a single ensemble of QDs, the low wafer thickness uniformity (about 2% [3]) limits to use this method in the fabrication of highly homogenous QDs micropillar arrays suitable for optical RC. In opposite, the MBE technique demonstrate better wafer thickness uniformity. Further studies aimed at creating the single ensemble of QDs grown by MBE are related to use the submonolayer deposition mode.

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