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Time-resolved photoluminescence study of InGaAs/GaAs quantum well-dots with upconversion method

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Abstract. For the first time we show time-resolved photoluminescence dependencies with 0.2 ps resolution for the novel type of InGaAs/GaAs quantum-sized heterostructures, referred to as quantum well-dots (QWDs). Photoluminescence upconversion method, that allows achieving time resolution up to 0.2 ps, was used to obtain time-resolved spectra for light (*lh*) and heavy hole (*hh*) optical transitions of QWDs. We concluded that the capture of charge carriers to the *lh* and *hh* states of QWDs occurs simultaneously in the time range of ~ 10 ps and is probably limited by carrier diffusion in the matrix. The characteristic time of photoluminescence decay for the *hh* state (3 ns) was found to be greater than that of *lh* one (2 ns).

Keywords: time-resolved photoluminecsence, InGaAs heterostructures, quantum well-dots

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Материалы конференции
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Исследование фотолюминесценции InGaAs/GaAs квантовых яма-точек с временным разрешением методом АП-конверсии

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Аннотация. Впервые показана зависимость сигнала фотолюминесценции (ФЛ) от времени для нового типа квантоворазмерных гетероструктур InGaAs/GaAs, называемых квантовыми яма-точками (КЯТ). Был использован метод ап-конверсии фотолюминесценции, позволивший достичь временного разрешения до 0,2 пс и получить временные зависимости сигнала ФЛ для оптических переходов легких (*lh*) и тяжелых

дырок (*hh*) в КЯТ. Было показано, что захват носителей заряда в *lh* и *hh* состояния КЯТ происходит одновременно в диапазоне времени ~ 10 пс, и, вероятно, ограничивается диффузией носителей в матрице. Характерное время затухания фотолюминесценции для состояния *hh* (3 нс) оказалось больше, чем для *lh* (2 нс).

Ключевые слова: время-разрешенная фотолюминесценция, InGaAs гетероструктуры, квантовые яма-точки

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Introduction

A lot of modern optoelectronic devices, such as microlasers [1] or solar cells [2], widely use an active area based on quantum wells (QWs) or quantum dots (QDs). Both types of quantum-sized heterostructures have their advantages and disadvantages determined by their structural and optical properties. For example, QD structures are less affected by the structural defects during growth process, have comparatively wide range of emission wavelength and suppressed lateral carrier diffusion. QWs, on the other hand, have much higher optical gain/absorption than QDs, but due to long diffusion length of charge carriers in lateral plane its application in microdevices is limited. The need to further optimize the properties of modern devices, as well as the need to develop new applications, leads researchers to look for a new configuration of the quantum-sized active region.

Recently developed quantum well-dots (QWDs) are attractive, because they combine properties of both QD and QW structures. QWDs can be described as a dense array of small QDs or QW with quasi regular variation in its composition and width. Such structures have suppressed formation of defects due to partial relaxation of elastic strain and allow achieving maximum material gain at $1.1 \cdot 10^4$ cm^{-1} [3]. InGaAs/GaAs QWDs used as an active medium in microlasers with a diameter of 20–50 μm made it possible to obtain continuous wave (CW) lasing with a wavelength of ~ 1.1 μm at room temperature and at temperatures as high as 110 °C [4].

However, the studies of processes of relaxation and recombination of charge carriers in QWDs remain relevant. In this work we studied optical properties of InGaAs/GaAs QWDs with time-resolved photoluminescence to provide a deeper understanding of charge carriers' kinetics in the QWDs. The results obtained are in demand for the realization of ultrahigh-frequency electrooptical devices based on QWDs.

Experimental

The InGaAs/GaAs heterostructure with QWDs was grown by MOVPE epitaxy on a slightly misoriented GaAs (100) substrate. After deposition of the GaAs buffer layer, the 200 nm thick $Al_{0.39}Ga_{0.61}As$ layer was grown to prevent carrier leakage from quantum-confined active region to the substrate. The active region was formed by a 1 layer of $In_{0.4}Ga_{0.6}As$ with deposited thickness of 8 monolayers and placed in the middle of a 600 nm thick GaAs layer [5]. The structure was terminated with a 50 nm thick $Al_{0.39}Ga_{0.61}As$ layer and 5 nm GaAs cap.

Photoconductivity spectrum was measured using a conventional lock-in technique with a halogen lamp in a combination with a monochromator as a light source in the geometry of normal light incidence. Indium contacts were soldered onto the surface of the structure as described in [6]. Photoluminescence spectrum in CW regime was recorded using 532 nm YAG:Nd laser and cooled Ge photodiode in combination with monochromator.

Time-resolved photoluminescence (TRPL) spectra of the InGaAs/GaAs heterostructure were investigated at room temperature with subpicosecond time resolution using the upconversion method. The source of femtosecond radiation was the mode-locked Ti:Sapphire "Coherent Mira 900" laser with a pulse duration of ~ 130 fs, repetition rate of 76 MHz, and tunable wavelength from 700 to 980 nm. PL signal was registered with FOG-100 spectrometer (CDP Systems). The laser pulse with wavelength of 780 nm was divided into two beams using a beamsplitter. First of them was passed through the delay line, while the second one was used to excite the PL in the sample. PL signal was collected by the optical system and mixed with the first beam on a nonlinear optical BBO crystal. Resulted upconverted signal was registered using a monochromator and a photomultiplier tube. This setup configuration provided TRPL spectra in the wavelength range of 850...1200 nm with optical resolution of 10–15 nm and up to 8 ns of time delay with time resolution of ~ 0.2 ps.

Results and Discussion

In Fig. 1 we demonstrate the scheme of the grown structure and transition electron microscopy (TEM) cross-section of QWD layer as insert. The formation of "islands" with variable In content is caused by misoriented substrate orientation (from 2° to 15° tilt degree). Atomic steps, caused by vicinal surface, facilitate creation of such islands of variable In content. Misoriented substrate also allows growing bigger QD (~ 20–30 nm) that cover many atomic steps [7].

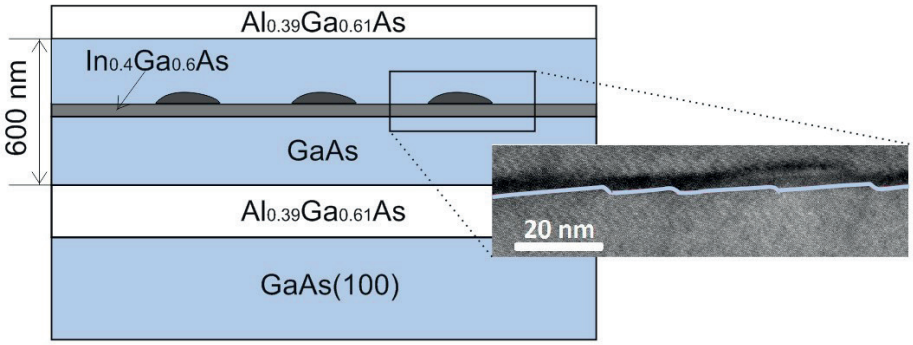

Fig. 1. Scheme of grown structure with QWD region. Inset shows a TEM cross-section of misoriented sample with QWDs

Photoluminescence (PL) and photoconductivity (PC) spectra are shown in Fig. 2. We observe PC plateau below ~ 870 nm, which corresponds to the GaAs matrix, and two peaks at 980 nm and 1040 nm. According to previous discussions [8, 9], these two peaks are attributed to radiative transitions between electron level *e*1 and two hole levels: heavy and light holes (*e1-hh*1 and *e1-lh*1). In contrast to PC, PL spectrum in Fig. 2 demonstrates Stokes-shifted peaks for GaAs matrix and *e1-hh*1 transition, and only a shoulder for *e1-lh*1. This could be explained by step-shaped density of states for QWDs, leading to overlay of PL signal of *e1-lh*1 with continuance of *e1-hh*1.

Fig. 3 demonstrates TRPL spectra taken at different delays. One can see pronounced GaAs peak decaying with time and one broad QWD peak at ~ 1020 nm, remaining its intensity during the period of 2 ns. Even though TRPL spectra do not evidence different QWD states, we found it reasonable to analyze in details kinetics for wavelength corresponding to light and heavy hole states, as well as GaAs.

PL kinetics of QWD structure taken at room temperature and different wavelength of GaAs matrix (~ 875 nm), *e1-lh*1 (~ 980 nm), and *e1-hh*1 transitions of QWD (~ 1040 nm) are shown in Fig. 4. Pumping power was on an average level of 85 mW, which corresponds to fluence ~ 10 μJ/cm^2. GaAs curve demonstrates picosecond risetime, relatively fast decay at initial part (Fig. 4, *b*) and after that monoexponential decay (Fig. 4, *a*) with τ ~ 1.4 ns. We attribute fast decay in GaAs peak to the effect of filling of QWD states with carriers resulting in relatively fast decrease in carrier concentration in GaAs.

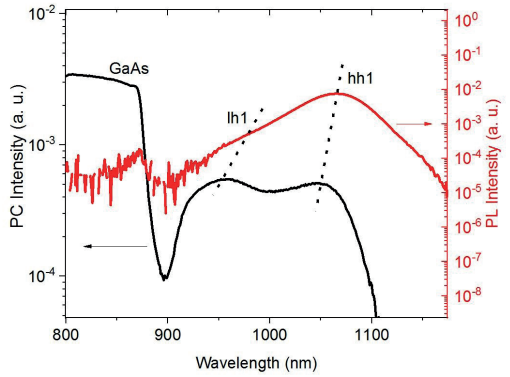

Fig. 2. PL and PC spectra of the sample, obtained in CW regime, room temperature

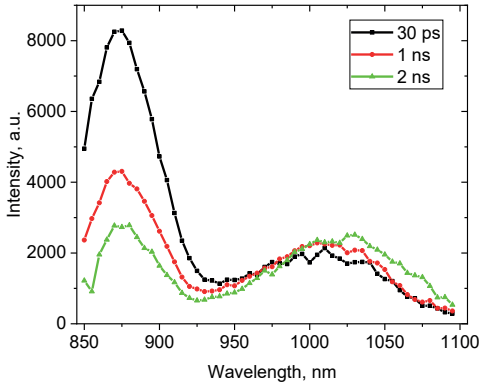

Fig. 3. Evolution of TRPL spectrum with time after excitation, room temperature

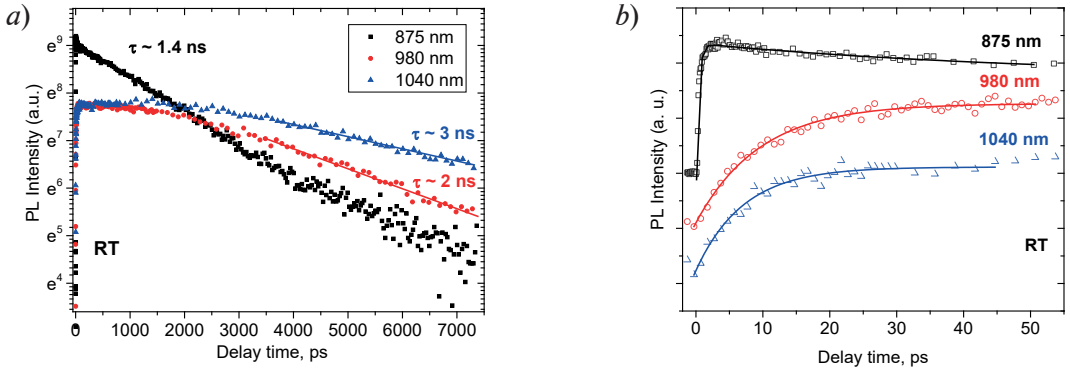

Fig. 4. PL kinetics taken at different wavelengths at room temperature, full time range (*a*), risetime range (*b*). The curves are normalized and shifted on (*b*) for clarity of presentation. Symbols show experimental data, lines — fitting

Initial part of GaAs curve may be fitted well with empirical equation (given $\tau_{rise} << \tau_{decay}$):

$$I_{PL}(t) = A\left(e^{-t/\tau_{decay}} - e^{-t/\tau_{rise}} + C\right),$$

giving characteristic risetime, τ_{rise} ~ 0.4 ±0.02 ps, and decay one, τ_{decay} ~ 40 ±20 ps.

At the same time initial parts of QWDs' curves show typical 1st order step response and fitted well with:

$$I_{PL}(t) = A\left(1 - e^{-t/\tau_{rise}}\right),$$

resulting in τ_{rise} ~ 9.5 ps and ~ 7 ps for 980 nm and 1040 nm, correspondingly. It seems clear, that both *hh* and *lh* states are filling with charge carriers simultaneously due to their comparable risetime, and there is no cascade relaxation in QWDs over theirs states. However, comparison to results from Aleshkin *et al.* [10] shows, that risetime values of QWD peaks several times higher than that of InGaAs QW of similar optical range (7–9 ps vs 1–3 ps). Such difference can be caused by longer diffusion time of charge carriers in larger GaAs reservoir to single QWD layer comparing to the much smaller GaAs regions in case in [10]. Also note comparable or even longer decay of GaAs peak at initial part than risetime of QWD peaks (~ 40 ps vs ~ 10 ps). This fact may indicate predominant exciton diffusion and capture in QWDs, rather than separate electron or hole capture, because otherwise, one must see decay in GaAs faster than increase in QWDs.

Further time kinetics of QWD PL could be characterized by several sequential processes. After the initial filling of states with charge carriers, *hh* and *lh* transitions remain the same intensity for ~ 1 ns (Fig. 4, *a*). As long as GaAs matrix remains filled with charge carriers, they migrate to QWDs and provide stable source of carriers' reproduction instead of those which have recombined. When carrier density in GaAs decreases, we observe beginning of PL decaying of *lh* state. Depletion of *lh* state starts at delay ~ 1 ns, while *hh* state stays filled until 2.5–3 ns. This probably illustrates general dynamic of charge carrier concentration of QWDs (Fig. 5).

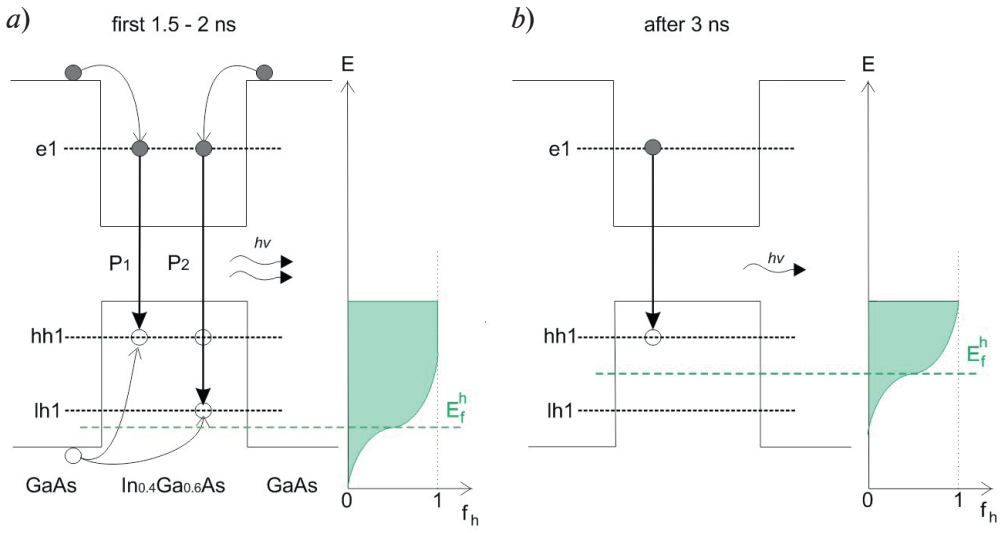

Fig. 5. Schematic presentation of QWD band structure and probable time evolution of carrier dynamics during (*a*) first 2 ns of laser excitation and (*b*) after first 1.5–2 ns with quasi Fermi functions

According to Aleshkin *et al.* [10] carrier relaxation rate in InGaAs QW, which is close in properties to QWDs, reaches 100 meV/ps at room temperature. This means that during the time range of approximately 5 ps charge carriers are thermalized and distributed over QWD states according to Fermi-Dirac statistic with quasi-Fermi levels (Fig. 5). As the rate of carrier recombination in QWDs is much slower than nanoseconds, according to Fig. 4, *b* all QWD states may be considered as the united system (Fig. 5), meaning that eliminating or adding carrier to any QWD state influences the others. As concentration of charge carriers is decreasing in QWDs and quasi-Fermi level in the valence band shifts upward (is opposite for electrons), PL signal of lh state (Fig. 5, *a*), first, and then hh one (Fig. 5, *b*) is starting to decay.

According to above, relatively fast decay of PL for lh state (τ_{decay} ~ 2 ns at delays 4–7 ns (Fig. 4, *a*) reflects high rate of carrier loss in heavily filled QWDs, when the carriers recombine both from *lh* and *hh* states (Fig. 5, *a*). Switching to more moderate QWDs' carrier filling (Fig. 5, *b*) results in a weak PL of *lh* state and, correspondingly, slower PL decay of *hh* state (τ_{decay} ~ 3 ns), as the carriers recombine primarily through *hh* state.

Overall PL lifetime at a level of e^{-1} for *hh* state reaches 6–8 ns (Fig. 4, *a*) and correlates well with the previous results [5], proving that non-radiative recombination rate is relatively low due to high quality of the sample.

Conclusion

The results obtained allow us to conclude that the capture of charge carriers from the GaAs matrix to the QWD *lh* and *hh* states occurs simultaneously and relatively slow (~ 10 ps). Long capture time can be explained by a long time of carrier diffusion through large GaAs reservoir. According to the GaAs fast decay time, we assume that different charge carriers (electrons and holes) diffuse and capture in QWDs simultaneously. Charge carriers form inside QWDs united system of thermalized carriers with Fermi-Dirac distribution and quasi-Fermi level. The characteristic time PL decay for the *hh* state (3 ns) was found to be greater than that of *lh* one (2 ns). The values of rise and decay times possess great importance for the design of high-speed devices based on QWDs and the estimation of their potentially achievable characteristics.

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