Investigation of multimodality effect in quantum dots
InGaAs/GaAs grown by MOVPE

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Abstract. Self-assembled InGaAs quantum dots with In content 80\% grown on vicinal GaAs substrates by MOVPE method have been investigated by photoluminescence technique in a wide range of temperatures and excitation densities. Multimodality of distribution of size and (or) shape in array of vertically stacked quantum dots synthesized under different growth conditions as well as in a single layer structure was found on the ground of appearance of a set of peaks in photoluminescence spectra at lowered temperatures. Influence of growth conditions on density and radiation wavelength of quantum dot modes has been analyzed. Investigation of photoluminescence of samples as a function of temperatures has allowed determining activation energies of three channels of the losses that were attributed to different modes of quantum dots.

1. Introduction
A lot of modern semiconductor optoelectronic devices, such as lasers or solar cells often contain quantum dots (QDs) in the active region. The unique properties of QDs (delta-like energy spectrum, low surface density) determine their leading position as the active media in low-threshold lasers and single photon sources. Besides, the effect of self-organization and consequent relaxation of elastic stress during pseudomorphic growth allows one to obtain multilayered structure with QDs (vertically stacked QDs) having high volume density, which is crucial for realization of quantum dots based solar cells [1]. Due to selforganization there is a probability of forming QDs of different size and/or shape. Each type of QDs generates photons with specific wavelength and, as a result, array of QDs has broadened radiation spectrum. On the other hand this feature may be used in positive way in comb-lasers [2], microdisk resonators [3,4] and tunable lasers, which require broad gain spectrum. In this paper we address to optical properties of the QDs grown by metal-organic vapor-phase epitaxy (MOVPE) and having multimodality in distribution of size and (or) shape [5].

2. Experimental
Experimental structures have been grown by using the MOVPE technique on vicinal GaAs (100) substrates with 6\° of misorientation. The structures contained single layer or vertical array (stack) of five layers of self-assembled In\textsubscript{0.8}Ga\textsubscript{0.2}As/GaAs QDs. The growth of QDs was carried out at the temperature of 520 \textdegree C while the other layers were grown at 700 \textdegree C. To avoid the degradation of
deposited QDs during substrate heating we used GaAs cap layer grown at 520 °C and covering QDs (Fig. 1). QD layers in stacked structures were separated by 40-nm GaAs spacer. Al_{0.3}Ga_{0.7}As barriers were used to prevent escape of carriers from QD area. Structures studied varied in the thickness of deposited InGaAs layer forming QDs and the thickness of GaAs cap layer. The structures with stacked QDs differed by the quantity of deposited In_{0.8}Ga_{0.2}As material and thickness of cap-layer, which were following: 3 monolayers of In_{0.8}Ga_{0.2}As and 8.5 nm of cap layer (C8I3), 2 monolayers of In_{0.8}Ga_{0.2}As and 8.5 nm of cap layer (C8I2), and 2 monolayers of In_{0.8}Ga_{0.2}As and 5 nm of cap layer (C5I2). Forth structure contained one layer of QDs similar to C8I3 and was called CS8.

Optical properties of structures grown were investigated by photoluminescence (PL). PL spectra were excited by second harmonic (532 nm) of Nd:YAG laser with maximum excitation density about 5kW/cm^2. Variation of excitation density was achieved by using several neutral optical filters. PL spectra were registered by cooled Ge diode using conventional lock-in technique. Closed-cycle helium cryostat was used to study PL spectra at temperature range of 10-320 K.

3. Results and discussion

3.1. Multimodality in InGaAs/GaAs quantum dots grown by MOVPE

Evolution of PL spectrum of CS8 structure with excitation density at 300 K (a) and 20 K (b) is shown on Fig. 2.

At room temperature conditions increase of excitation modifies spectrum from single-peak shape (QD1) to the shape with a number of pronounced peaks (QD2 and QD3) demonstrating saturation-like
behaviour (Fig. 2, a). This observation may remind behaviour of conventional MBE-grown quantum dots (see, for instance [6,7]), where peaks QD2 and QD3 are attributed to the first and the second excited states.

But this idea is refuted while analysing low temperature PL spectrum (Fig. 2, b). At low temperature conditions transport of carriers from one quantum dot to another is suppressed. Carriers captured by a certain quantum dot, cannot escape from it, and, as a result, they take part in optical recombination in this QD. This results in random population of QDs by carriers. So, appearance of a set of the peaks at low temperature and low excitation density points to a multimodal distribution of QD while intensity of PL peak being proportional to modal density of QD. One can see on Fig. 2, b that the multipeak shape of the PL spectrum doesn’t change through the wide range of excitation density. So, it can be stated, that observed PL peaks resulted from radiative transitions from ground states of different QD modes (QD1, QD2, QD3), and excited states has no influence on PL spectrum, unlike classic quantum dots [8]. Similar behaviour was observed for all investigated samples, and figure 2 shows PL spectra for one-layer structure because of the fact that optical effects due to QD multimodality are most clearly seen for this structure.

In this letter we investigate mostly optical properties of QD arrays caused by multimodality while structural properties and growth aspects of self-assemble of such structures are the subjects of further research.

Evolution of PL spectrum of structure CS8 with temperature is shown on Fig. 3, a.

Redistribution of intensity between PL peaks with increase of temperature is clearly seen. This observation is in agreement with model [9], schematically presented on Fig. 3, b, which describes carrier transport between QDs in terms of thermal escape of carriers into matrix and consequent recapture. At temperatures below 100K transport of carriers is suppressed. Photoinduced carriers are populating QDs randomly (black colour process in Fig. 3, b), realizing so-called nonequilibrium carrier distribution. PL spectrum at this stage demonstrates only ground states for each of the modes of QD array. This situation is continued till the temperatures about 100K. Increasing of temperature up to 150K activates thermal escape of carriers from predominantly small QDs (type QD3 or QD2) into
matrix followed by recapture into big QDs (QD1). Due to such unidirectional transport of carriers between QDs, distribution deviates from nonequilibrium one (blue colour process in Fig. 3, b). Intensity of QD3 peak degrades dramatically relative to QD2 and QD1, and the PL spectrum is narrowed. Further temperature increase (above 240K) opens possibility of carrier escape into matrix from big QDs - QD1 (red colour process in Fig. 3, b). Carrier transport is becoming random and filling of QDs is occurring according to Fermi-Dirac statistics, i.e. equilibrium distribution. At this stage weak QD3 peak is emerging in PL spectrum. So, we can conclude that temperature modification of PL spectrum of multimodal QDs has a complex character.

3.2. Influence of growth parameters on optical properties of multimodal QD array

Fig. 4 shows PL spectra for structures C8I3, C8I2, and C5I2. At temperatures about 20 K PL peaks of each type of quantum dots are clearly seen. At temperature of about 320 K one can see only peaks of long wavelength types of QDs (QD1 and QD2) due to mechanism of filling QDs described above. Note, that the width at half maximum of PL spectrum, which is composed from ground states of different types of QDs, reaches 140 nm for structure C5I2 at 20K.

![Figure 4. PL spectra (shifted for clarity) of structures C8I3, C8I2, and C5I2 taken at 20 K (a) and 320 K (b).](image)

The results of analysis of PL spectra on Fig.4 are shown on Figure 5. Accurate position of PL peaks for each type of QDs was determined via analysing of differentiated PL spectrum.
Analysis of Fig. 4 and 5 is giving a number of observations. Firstly, increasing of the thickness of GaAs cap layer (Fig. 1) leads to slight increase of the size of big QDs (QD1) and decrease of their density, as following from redshift of QD1 peak and reduce its intensity. So, one can conclude that size of big QDs (QD1) is limited to the thickness of cap layer. At the same time increasing of the thickness of cap layer has a little effect on mode QD3. Secondly, increasing of the effective thickness of deposited InGaAs material leads to significant increase of density of small QDs (QD3) while remaining their size (peak position has small variations only). Besides, deposition of thicker InGaAs layer results in remarkable increase of density of big QDs (QD1) with insignificant decrease of the size of the dots.

3.3. Temperature dependence of integrated photoluminescence
Temperature dependences of integrated PL intensity for investigated structures are shown on Fig. 6.

Experimental data (symbols) were approximated (solid line) with the model [10] which includes three channels of losses. Activation energies have been determined for each of the channels. Fitting of the
experimental data with models with less than three of channels of losses has given unsatisfactory results. These channels of losses of carriers were associated with thermally induced carrier escape from QDs of three types (QD1, QD2, QD3) into matrix and consequent non-radiative recombination. For all structures investigated determined activation energies have fall into three groups with characteristic energies about 10 meV, 30-80 meV, and 100-400 meV, which were attributed to QD3, QD2, and QD1, correspondingly. However, there is no strong correlation between activation and localization energies (determined on the ground of position of the peaks of ground states for each types of QDs), which is the subject of further research.

4. Conclusions
Heterostructures containing self-assembled In$_{0.8}$Ga$_{0.2}$As/GaAs quantum dots grown by metal-organic chemical vapor epitaxy were investigated by means of photoluminescence (PL). All investigated structures have demonstrated multimodality of distribution of size and(or) shape of QD array. Multimodality of QDs resulted in complex evolution of the shape of PL spectra with temperature increase. To protect QDs from degradation during growth process GaAs cap layer deposited at lowered temperature was used. It was found that cap layer thickness has significant influence on the size of QDs of long wavelength type. Density of QDs of short wavelength type was found to be strongly sensitive to effective thickness of deposited InGaAs layer. Fitting of temperature dependencies of integrated PL intensity has allowed determining activation energies of three channels of the losses that were attributed to different modes of QDs.

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