Photoluminescence excitation spectroscopy of InAs/InGaAs/GaAs quantum dot arrays in a wide temperature range

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Abstract. Detailed photoluminescence and photoluminescence excitation (PLE) studying in temperature range 10-300 K with variation of detection energy of self-organized InAs/InGaAs/GaAs quantum dots is presented for the first time. Abnormal quantum well-like behavior of quantum dot arrays in PLE experiment at raised temperatures is observed. Changing in properties of quantum dots with temperature is attributed to modification of the type of the mechanism of dot population by charge carriers from non-equilibrium one to equilibrium one. Activation of thermally assisted carrier transport between quantum dots is found to destroy dot selection mechanism by detection energy in PLE experiment.

1. Introduction
In(Ga)As/GaAs semiconductor quantum dots (QDs) are the one of the most promising materials of modern photo- and nanoelectronics. Fundamental property of QDs is a discrete spectrum of energy states due to theirs zero dimension. This feature distinguishes QDs from other quantum-sized structures, namely quantum wells and quantum wires having continuous spectrum of energy states. QDs are used in a wide range of devices, especially in lasers, light emitting diodes, photodiodes, and solar cells. Unique properties of QDs allow one to achieve record characteristics of these semiconductor devices [1, 2].

Key parameters of QD based devices considerably depend on energy structure, charge carrier transport and relaxation processes taking place in employed QDs. These properties depend on a growth conditions in a complex manner, which makes a theoretical modeling insufficiently accurate and reliable evaluation method. For example, QD shape, composition and elastic stress, which are hard to take into account, are able to influence both on energy spectra and charge carrier transport and relaxation processes.

One of the most informative experimental methods to investigate energy spectra of quantum-sized structures is a photoluminescence excitation spectroscopy (PLE). This is a nondestructive method, applicable to structures with nontransparent substrates and resistant to influence of absorbing centers out of an investigated area, which is an advantage over absorption spectroscopy methods. Furthermore, it doesn’t require a contact soldering, which is an advantage over photoconductivity spectroscopy and CV methods, what makes PLE an ideal for express analysis of quantum-sized heterostructures.
The peculiarity of application of PLE to structures with QD array is the possibility of obtaining spectra from dots of different sizes, which gives an additional instrument to get insight in physics of QDs [3]. PLE analysis of QDs is usually carried out at lowered temperatures [3-4], and in [4] QD PLE spectra were obtained in temperature range (100-200 K) but without variation of detection energy.

In this work we investigate in details evolution of QD PLE spectra in temperature range (20-200 K) and liken these data with temperature dependent photoluminescence.

2. Experimental setup
Two samples containing 1 and 3 layers of InAs/InGaAs/GaAs self-organized QDs were grown by molecular-beam epitaxy on GaAs substrates (100). For the first sample, QDs were formed by depositing of 2.75 monolayers of InAs and capping by 5 nm of In$_{0.15}$Ga$_{0.85}$As quantum well. For the second sample, QDs were formed by depositing of three sheets of QDs formed by depositing of 2.5 monolayers of InAs and capping by 5 nm of In$_{0.13}$Ga$_{0.87}$As quantum well. In both cases QDs were grown in conventional growth conditions (see, for example, [5]) at substrate temperature below 500° C and InAs growth rate in the range 0.15-0.3 Å/s. Spacer between QD sheets comprises 30 nm of GaAs. QDs were synthesized in the middle of a 200 nm-thick GaAs region surrounded by Al$_{0.3}$Ga$_{0.7}$As barriers. Photoluminescence (PL) spectra were recorded using MDR-23 monochromator and a cooled germanium photodiode. Second harmonic of YAG:Nd laser with the wavelength of 532 nm was used as photoluminescence excitation source. MDR-12 monochromator and an incandescent lamp were used as excitation source for PLE. A closed cycle helium cryostat was used to provide measurements in the temperature range 10-325 K. PL and PLE spectra were recorded using standard lock-in technique.

PLE spectra result from PL emission following absorption by heterostructure of the exciting radiation with varying wavelength. Therefore, analysis of PLE spectra allows one to obtain information on the energy states of the heterostructure under study. However, such analysis is complicated by the presence of LO phonon replicas in PLE spectra [3].

In the context of QD arrays, PLE method has an advantage due to the capability of QD size selection. Usually, arrays of QDs have a relatively broad PL spectrum due to inhomogeneous broadening. Given sufficient spectral resolution of the experimental setup, it is possible to isolate the PLE spectra of QD of different size (shape), emitting at different wavelength (Fig. 1).

![Figure 1](image-url)
3. Results and discussion

Fig. 2 shows a low temperature PL spectrum and a set of PLE spectra taken at different detection energies across the main PL peak. The PL spectrum shows strong main peak which we associate with the ground state of QDs and a high energy shoulder which we attribute to excited state. On PLE spectra one can see a set of peaks. The peak appearing on PLE spectra at the same position we attribute to quantum well state. Peaks which position depends somehow on detection energy were numbered as 1-3 and analyzed below.

(a) [Graph showing PL and PLE spectra for a single layer structure at 20 K]

(b) [Graph showing PL and PLE spectra for a three layer structure at 20 K]

Figure 2. PL spectrum at excitation density of 200 Wcm\(^{-2}\) (left axis) and two PLE spectra (right axis, shifted for clarity) of structure with single QD layer (a) and three QD layers (b), 20 K. Detection energies for PLE spectra are marked by arrows.

Energy distances of PLE peaks 1-3 (see Fig. 2) to detection energy as a functions of detection energy are plotted in Fig. 3. Peak #2 demonstrates noticeable positive slope. This means that increase in the detection energy (and corresponding decrease in QD size) results in increase of peak energy distance to the ground state of selected QDs. Peaks #1 and #3 are nearly independent of detection energy. These allows us to identify peaks #1 and #3 as LO phonon replicas, and to associate peak #2 with excited state of QDs. PLE peaks for the sample with three QD layers at 20 K behave in nearly similar way.

(a) [Graph showing energy distance of peak 1 as a function of QD size]

(b) [Graph showing energy distance of peak 2 as a function of detection energy]

(c) [Graph showing energy distance of peak 3 as a function of detection energy]

Figure 3. The energy distances (\(\Delta E\)) of PLE peaks #1 (a), #2 (b), and #3 (c) to detection energy as a functions of detection energy.
PLE experimental data similar to that shown in Fig. 3 were collected and linearly fitted for different temperatures for both samples. Peaks #1 and #3 have showed the same static behavior as at 20 K, but not peak #2. Figure 4 shows the temperature evolution of the dependence of excited to ground state energy distance ($\Delta E_{ES-GS}$ – the energy distance of PLE peak #2 to detection energy) vs detection energy for both structures.

![Figure 4](image-url)

**Figure 4.** The temperature evolution of excited to ground state energy distance ($\Delta E_{ES-GS}$ – the energy distance of PLE peak #2 to detection energy) for sample with single QD layer (a) and three QD layers (b). The dependences were shifted along horizontal axis on the value of PL maximum ($E_{PPL}$) for better comparison.

In the range 20-50 K the dependence in Fig. 4, a and b preserve its slope, while further temperature increase results in gradual rotation of the dependence down to reverse slope at temperatures around 200 K. At higher temperatures PLE peaks are hard to analyze as their intensity decreases rapidly with temperature.

Figure 5, a summarizes these data and shows the slope of the dependences on Fig. 4 as a function of temperature.

It is seen that in the temperature range 20-110 K the slope varies insignificantly for both samples. However, at higher temperatures the slope begins to decrease sharply and becomes negative at temperatures above 140 and 160 K for sample with single QDs layer and three QDs layers, respectively.

![Figure 5](image-url)

**Figure 5.** The slope of the dependence of excited to ground state energy distance ($\Delta E_{ES-GS}$) on detection energy vs temperature (a); and temperature dependence of the RMS of PL peak (b) for sample with single QDs layer (blue solid) and three QDs layers (red dashed).
Found dependences of $\Delta E_{ES-GS}$ on detection energy at raised temperatures do not coincide with the expected behavior of excited state in PLE experiments described above. For instance in temperature range 140-160 K peak #2 has a zero slope of the dependence $\Delta E_{ES-GS}$ on detection energy what is similar to behavior of peaks 1 and 3 (i.e. phonon replica). Note, that there is no report on such dependences at raised temperatures. In [4] PLE spectra of QD samples at raised temperatures were shown, however no information on investigation of PLE spectra with variation of detection energy was presented.

Interpretation of found abnormal behavior of QD excited state in PLE experiments at raised temperatures may be based on the well-known phenomenon of switching of the type of QD filling with charge carriers from non-equilibrium one at lowered temperature to equilibrium one at raised temperature. At low temperature due to suppressing of thermally assisted transport of carriers between QDs population of dot array is random or non-equilibrium [2]. Increase of temperature results in activation of thermal escape and transport of carriers between QDs (primarily from shallow ones to deeper ones). The ground state PL peak is correspondingly narrowing as in emission participate primarily deep QDs rather than whole array. Temperature dependences of RMS (effective width) of PL peak for both samples are shown in Figure 5, b. Minimum is clearly seen on the dependences at 160 and 180 K for sample with single QD layer and three QD layers, respectively. This temperature range correlates well with the temperature point at which sharp decreasing of the slope occurs in Fig. 5, a.

Thus, we can conclude that in temperature range 140-160 K thermally assisted carrier transport between QDs in the array is activated. This leads to liberalization of QD selection by detection energy in PLE experiment as carriers no more limited to the dot where they were born (as it happened at lowered temperature). In contrast, carriers can be emitted through other dots in array (primarily deeper ones). Complete absence of QD selection should result in quantum well-like behavior in PLE, i.e. position of the peaks is governed by excited states of QDs with average (most expectable) size in array, and is insensitive to detection energy. In such a case the slope of the dependence of $\Delta E_{ES-GS}$ on detection energy on Fig. 4 will be close to -1. The tendency of lowering of the slope with the temperature in Fig. 5, an expected to lead to this value in perspective. In other words, QD array in PLE experiments at raised temperatures behave itself as a quantum well.

According to the temperature dependences of both RMS and the slope of the dependence of $\Delta E_{ES-GS}$ on detection energy, the temperature of transition of QD population from non-equilibrium to equilibrium type occurs on 20 K later for sample with three QD layers. This implies higher activation energy for thermally assisted carrier transport, which looks strange taking into account position of QD ground state peak (three layer QD structure demonstrates 16 meV-blueshift of PL peak compared to single layer one, see Fig. 2). However, including of QW peak into analysis clarifies the situation. Three layer structure has higher energy of QW state due to smaller In concentration in it (13%) as compared to single layer structure (15%). As a result, energy distance between QD ground state and QW state for three layer structure is higher by ~9 meV (Fig. 2), which is in agreement with dependences on Fig. 5. Thus, one can conclude that key role for thermally assisted carrier transport between QDs is played by QW rather than matrix.

Conclusion

Heterostructures with InAs/InGaAs/GaAs quantum-dots-in-a-well were synthesized by molecular beam epitaxy and studied by photoluminescence (PL) and photoluminescence excitation (PLE) spectroscopy methods in temperature range 20-300 K. Analysis of PLE spectra with variation of detection energy allowed us to identify PLE peaks as LO phonon replica or as an energy state in quantum confined structure. For the first time, PLE spectra with varying of detection energy were recorded and analyzed for quantum dot structures at temperatures up to 200 K. It is found, that array of quantum dots at temperatures, higher than the temperature of establishing of equilibrium carrier population, starts to modify its behavior in PLE experiments as a like a quantum well. Changing in behavior of quantum dots with temperature is attributed to activation of thermally assisted carrier transport between dots at raised temperatures and consequent violation of QD selection by detection
energy in PLE experiment. It is also found that thermally assisted carrier transport between QDs occurs via QW rather than matrix.

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