Micro-photoluminescence of InP/GaInP₂ quantum dots structures for topological quantum gates

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Abstract. Self-organized quantum dots (QD) InP/GaInP₂ are natural electronic molecules (EM). They are promising objects for the study of the physics of strongly correlated electronic systems and the creation of quantum logic elements. In this work, we used micro-photoluminescence technique to study the effect of molecular oxygen on emission properties of these QDs and possible reducing their surface density.

1. Introduction

Self-organized quantum dots (QD) InP/GaInP₂ are natural electronic molecules (EM). They are promising subjects for the study of the physics of strongly correlated electronic systems and the creation of quantum logic elements [1-3]. The control of the EM states in the QD is carried out by photoluminescence spectrum (PL) measurements and until now has been carried out mainly by near-field scanning optical microscopy methods, due to the relatively high surface density of the QD (~10⁸ cm⁻²).

The practical use of EM in quantum logic elements requires the creation of structures with isolated QD up to 10 micrometers, i.e. a surface density up to ~10⁶ cm⁻², which, will allow to control of EM states by a simpler method of micro-PL.

It is worth noting that it has already been possible to reduce the surface density of the InP/GaInP₂ QDs through the treatment of the sample with ozone plasma [2] or through selective area epitaxy using SiO₂ masks [4]. The disadvantage of the ozone plasma approach is that the PL of QD at the processing time (~5 sec) has completely disappeared. Because of the rapid extinction, it is almost impossible to obtain a QD with a predetermined surface density. Probably this behavior because the plasma is a source of highly energetic atoms that fit in the crystalline lattice of the QD, thereby extinguishing them.

Using SiO₂ mask has also problems, since QDs obtained do not show any luminescence even at low temperature, which probably related to the oxygen reacting with QDs during epitaxy. Thus, using of excited oxygen treatment of QD structures, producing less severe radiation, can be alternative way to reduce surface density and it also can be used to study mechanism of PL quenching.
2. Experiment

The sample was grown by metal organic chemical vapor on the GaAs(001) substrate 2 degrees misoriented towards [111]. The InP QDs are flat lens with a thickness of ~15 nm, a lateral size of ~150 nm and a density of ~10^8 cm^-2[5].

The test sample was excited oxygen treatment using radical etching (RE) technology at the Secon500 facility. During the RE processes, the treated sample was located in the area separated from oxygen plasma by a perforated metal screen. This separation of the region of discharge plasma and the region where the sample is located prevents the electrons and ions from reaching the surface of the sample, and at the same time creates practically no barriers for excited molecular oxygen. Once the oxygen reaches the surface of the sample, it has a chemical reaction with it, forming volatile compounds, which are removed from the reaction zone as a result of continuous pumping.

This paper presents the results of PL spectra measurements in the temperature range (10-300 K) and at different time of oxygen treatment and time of recombination of fundamental transition GaInP and QD InP, spatial intensity distribution (map) of PL bands.

![Figure 1](Image)

**Figure 1.** PL spectra of the sample after treatment excited oxygen 5, 10, 20, 30, 50 minutes at 300K and recombination time of QD and GaInP before and after oxygen treatment 50 minutes

The spectra of micro-PL at 300K and 77K were measured at a facility equipped with a 50X micro-objective and a continuous Nd:YAG laser (532nm), power of radiation incident the sample was 10 microwatt, (Figure 1, Figure 3). The confocal map was made using a continuous laser Nd:YAG (532nm), optics 100X, and the radiation intensity of sample 10 microwatt (Figure 2). Recombination time was measure at the Picoharp 300 facility using method of Time-Correlated Single Photon Counting.
3. Results

In the obtained PL spectra (Figure 1) of the sample with QD InP, measured at 300K, there are three main radiation bands: GaInP (640-650nm), QD (700-800nm) and GaAs (860nm). This spectra is characteristic for the QD InP in the GaInP matrix grown in an metal organic chemical installation. For handling time of 5-30 minutes in these spectra, the intensity decline of GaInP and the intensity does not decline of QD. When the material is handling for 50 minutes, the intensity drops sharply of the QD and GaInP. The micro-PL at 300K indicates that the surface density has dropped of the QD.

The data obtained through confocal microscopy (Figure 2) show that the surface density has not changed as was expect from micro-PL at 300K. The spectra (Figure 2) show that the intensity of QD and GaInP is equivalent to intensity of GaAs.

Figure 3 shows the spectrum of the micro-PL sample with QD InP measured at 10K and 80K. There are peaks in the spectra at 10K corresponding to the emission of single QD of different sizes. The larger QD are the bigger the radiation wavelength of QD is. The peaks are close to each other. That indicates that the surface density has not decreased. At a temperature of 80K, the peaks advising the radiation GaInP (650nm) and GaAs (860nm) disappear, and the radiation intensity before and after processing the sample with oxygen at the same. This spectrum is characteristic of QD InP in the GaInP matrix grown in a metal organic chemical at 10K and 80K.

The graphs in figure 1 show that the time of the fundamental QD transition has not changed, and the time of the fundamental transition of GaInP changed to time of ~0.3 ns.

4. Discussion

Probably, the drop of intensity at 300K is caused by the embedding of oxygen into the crystalline lattice of layer GaInP covering QD. In the GaInP layer, defective energy levels occur due to the built-in oxygen. They cause the emergence of radiant-free recombination centers. In GaInP, the photons excite the charge carriers up to 2.3 eV and then they relax to the energy level of 1.9 eV, then some charge carriers are captured by the QD InP and relax within to the level of 1.6-1.8 eV, afterwards they recombine radiating photons. Other charge carriers that did not get into the QD transfer from the conductivity zone to the valence zone GaInP emitting a photon with energy of 1.9 eV. The GaAs substrate will be reached by the photons that have not excited the charge carriers in GaInP, therefore the radiation intensity of the
substrate does not depend on the defects of the outer layer. When a defective level appears (it remains below 1.9 eV), the charge carriers can relax to it radiationlessly, and then recombine to valence zone emitting a phonon. This transition is more probable than GaInP radiation or the capture of the charge carrier by QD, so the intensity of the GaInP layer and QD radiation decreases. The time of recombination of GaInP has decreased, that also indicates new energy levels appeared in the forbidden area of the structure of GaInP (Figure 1).

At a temperature of 80K the intensity of the PL before and after oxygen treatment remains. This is so because at a temperature of 300K the thermal energy (kT) is enough to free the defective level from charge carriers. Because of this, vacant places of defective level are free for charge carriers to relax on and recombine without radiating a photon. At 80K, this level is being filled with charge carriers, so kT-energy is not enough to move into the conductivity zone, so when charge carriers relax to 1.9 eV from 2.3 eV, they cannot move further to the defective level, as it is fully filled. That is the reason the intensity of the QD remains the same.

Figure 3. PL spectrum at 10K after oxygen treatment (blue) and 80K before oxygen treatment (black) and after oxygen treatment 50 minutes (red).

5. Conclusion
As the result of these studies we found that the oxygen molecules are embedded into the surface layer of GaInP, forming defective energy levels that capture charge carriers, thereby the intensity of GaInP and QD reduce significantly. This show, that one need to use high quality SiO₂ layers and reduced growth temperatures in selective epitaxy to prevent oxygen interaction with QDs.
References

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