The Effect of Exposure to Arsenic Flow on the Optical Properties of Quantum Dot Arrays in the InAs/GaAs(100) System


Abstract—The optical properties of quantum dot arrays in the MBE-grown InAs/GaAs(100) epitaxial system with an effective InAs layer thickness of 1.9 monolayers were studied in samples exposed to the beam of As$_2$ for various times after switching off the In beam. The results of photoluminescence measurements showed that the emission wavelength increased with the exposure time within certain limits. This behavior agrees with predictions of the kinetic model of the initial stage of quantum dot formation.

In recent years, there has been rapid growth in the number of publications devoted to the influence of heteroepitaxial growth conditions on the structural and optical properties of quantum dot (QD) arrays. This is related to the need for developing methods for obtaining QD arrays with the properties required in particular applications. For example, the size of QDs in the heterostructures for optoelectronic devices has to correspond to the working wavelength, while the QD density must remain unchanged and the parameters of epitaxial growth conditions on the structural and optical properties of QD arrays with the properties required in particular applications. This is related to the need for developing methods for obtaining QD arrays with the properties required in particular applications. For example, the size of QDs in the heterostructures for optoelectronic devices has to correspond to the working wavelength, while the QD density must remain unchanged and the parameters of epitaxial growth conditions on the structural and optical properties of QD arrays with the properties required in particular applications.

This study continues systematic quantitative investigation of the effect of controllable technological parameters of epitaxial growth on the properties of QD arrays in the InAs/GaAs(100) system. We present the results of photoluminescence (PL) measurements for InAs quantum dots grown by molecular beam epitaxy (MBE) on single crystal GaAs(100) substrates. The effective thickness of deposited InAs, the growth temperature, and the rate of InAs deposition were fixed, while the time of the final exposure to the As$_2$ beam was varied.

According to the results of a theoretical analysis based on the kinetic model of QD formation under the action of elastic stresses, the kinetics of the average lateral island size variation with time is described by the following equation in dimensionless variables:

$$\frac{t - t_c}{t_R} = \ln \left( \frac{1 + l + l^2}{1 - l} \right) + \sqrt{3} \arctan \left( \frac{2l + 1}{\sqrt{3}} \right) + \frac{\pi}{2\sqrt{3}} \equiv U(t).$$

Here, $t = t_0 + t_{exp}$, $t_0 = H_0/V$ is the time required for depositing $H_0$ monolayers (ML) of InAs at a given deposition rate $V$ [ML/s]; $t_{exp} = H_c/V$ is the time required for obtaining a wetting layer of the critical thickness $H_c$ [1] (for the system under consideration, $H_c \sim 1.7$ ML); $L = L/L_R$ is the characteristic relaxation time. It should be noted that the values of $L_R$ and $t_R$ (determined by expressions derived in [5]) depend on the system energetics, lattice mismatch, island geometry, effective thickness $H_0$, substrate surface temperature $T$, and growth rate $V$. Assuming that the island shape in the initial growth stage remains unchanged and the parameters $H_0$, $T$, and $V$ are maintained constant, the values of $L_R$ and $t_R$ can also be considered as constants determined by these parameters.

Since the right-hand part of Eq. (1) does not contain any model parameters, the dependence of $l$ on $(t - t_c)/t_R$ (determined through inversion of Eq. (1)) has a universal form depicted in Fig. 1. For the given values of the effective thickness, temperature, and growth rate, the theory predicts a monotonic increase in the average island size with the exposure time. For large times...
The island size exhibits saturation and approaches the quasi-stationary size \( L = L_R \). In the quasi-stationary state, all substance present in the wetting layer in excess of the amount corresponding to the equilibrium layer thickness (determined by the balance of contact and elastic forces in accordance to the Müller–Kern criterion [6]) is distributed within the islands. The adopted theoretical model [4] also predicts that the relative dispersion of the distribution of islands with respect to the lateral size decreases with increasing exposure time.

The experiments were performed in an MBE setup of the EP1203 type using semi-insulating single crystal GaAs(100) substrates. In a series of four samples, the active region was formed by depositing an InAs layer with an effective thickness of 1.9 ML at a deposition rate of \( V = 0.03 \text{ ML/s} \), followed by exposure to the As\(_4\) beam for \( t_{\text{exp}} = 0, 7.5, 15, \) and 22.5 s. The substrate temperature during QD layer formation, exposure to arsenic, and GaAs coating was \( T = 485^\circ\text{C} \) (a 5-nm-thick GaAs layer was deposited in order to avoid indium loss by evaporation from QDs). The process of InAs QD deposition was monitored by reflection high-energy electron diffraction (RHEED). The observed RHEED patterns showed that a sharp transition from linear to point diffraction (characteristic of the three-dimensional island growth) in all cases took place after deposition of the InAs layer with a thickness of about 1.7 ML.

In order to eliminate the transport of nonequilibrium carriers into a near-surface region and into the substrate in the course of optical measurements, the active region was bounded from both sides by short-period superlattices Al\(_{0.25}\)Ga\(_{0.75}\)As/GaAs (5 pairs, 25 Å/25 Å). Finally, a 5-nm-thick protective GaAs layer was deposited. The buffer layer, superlattices, and the upper GaAs layer were deposited at a temperature of 600\(^\circ\text{C}\). The partial pressure of As\(_4\) in the growth chamber did not exceed \( 1.5 \times 10^{-6} \text{ Pa} \). The PL spectra were excited by an Ar\(^+\) laser (\( \lambda = 514.5 \text{ nm} \); beam power density, \( \sim 100 \text{ W/cm}^2 \)). The emission was measured with a cooled Ge photodiode.

Figure 2 shows the room-temperature PL spectra of two samples, for which the exposures to arsenic after QD layer growth were \( t_{\text{exp}} = 0 \) and 7.5 s. The inset in Fig. 2 presents experimental data showing variation of the emission wavelength depending on the exposure time. As can be seen from the PL spectra, an increase in \( t_{\text{exp}} \) leads to narrowing (from \( \sim 80 \) to \( \sim 30 \text{ meV} \)) of the emission band corresponding to radiative recombination via the ground state in QDs. This is evidence of a more homogeneous distribution of the QD size. Obtaining QD arrays with homogeneous QD dimensions is important in many applications. In particular, this is a necessary requirement for the formation of “quantum molecules” [7].

As the exposure time increases to 15 s, the position of the PL band maximum on the wavelength scale shifts toward higher values, but further increase in \( t_{\text{exp}} \) leads to a shift to shorter wavelengths. The initial increase in the emission wavelength is explained by the QD size growing in the course of exposure to arsenic as a result of the absorption of substance from the wetting layer. Thus, the experimental data and theoretical predictions qualitatively agree for exposure times up to \( \sim 15 \) s. Then, since the amount of substance in the wetting layer is limited, the QD size and, hence, the emission wave-