08

Peculiarities of growth of InAs quantum dot arrays with low surface density by molecular beam epitaxy

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Received October 10, 2022
Revised November 1, 2022
Accepted November 1, 2022

The influence of the substrate temperature and the flux of In adatoms on the structural and optical characteristics of InAs quantum dots with a low surface density is experimentally studied. An increase in the substrate temperature under conditions of a high flux of In adatoms promotes an increase in their surface migration and a certain decrease in the density of the array of quantum dots (down to $\sim (1-2) \cdot 10^{10}\,\mathrm{cm}^{-2}$), however, in this case a significant short-wavelength shift of the photoluminescence spectrum is observed despite an increase in lateral sizes of dots. A decrease in the incident flux of In adatoms at optimal substrate temperatures makes it possible to reduce the dot density more efficiently (down to $\sim (1-2) \cdot 10^{9}\,\mathrm{cm}^{-2}$).

Keywords: molecular-beam epitaxy, quantum dots, surface density.

DOI: 10.21883/TPL.2022.12.54955.19389

Development of non-classical light sources necessary for state of the art optical quantum technologies, including quantum metrology, quantum cryptography and linear quantum computation, is an extremely complex technological problem [1]. Self organized semiconductor quantum dots (QDs) in the InGaAs/AlGaAs system open broad perspectives for creating single-photon emitters [2]. To realize efficient single-photon sources, it is necessary to not only ensure high efficiency of photon output and emission directionality but also to decrease surface density of the InAs QD array to the level of $1 \cdot 10^{9}\,\mathrm{cm}^{-2}$ and lower. A possible approach to decreasing the InAs QD surface density consists in depositing a thin InAs layer whose thickness is close to the critical thickness of transition from the layer-by-layer growth mode to the island one (hereinafter referred to as the 2D → 3D transformation) [3]. However, in the framework of this approach it is extremely difficult to obtain single-photon emission in the spectral ranges of 1.3 and 1.55 μm that are most interesting in view of creating telecommunication systems providing absolutely safe data exchange via the quantum cryptography protocols. Realization of single-photon emission in the telecommunication ranges needs QDs of larger sizes. In the framework of the MBE technique, the possible solution is associated with decreasing the flux of in adatoms towards the growing layer surface and enhancing surface migration of In adatoms, since, for the purpose of reducing mechanical stresses and surface energy, in this growth mode the process of insertion of In adatoms into already existing three-dimensional objects (QDs) prevails over formation of new objects. These epitaxial conditions may be realized by increasing the substrate temperature [5,6], decreasing the QD growth rate [7,8], growth interruption [9], formation of the flux gradient when the substrate holder stops rotating [10] or inclines [11], formation of stress fields in the underlying epitaxial layer [12]; combining these approaches is also possible. However, specific modes of growing low-density InAs QD arrays significantly depend on the growth chamber configuration, arrangement of the materialsśources, and designs of the effusion cells and substrate holder.

This paper presents the results of comprehensive studies aimed at searching for such modes of the InAs QD array epitaxial growth on GaAs substrates, which are able to provide a controllable decrease in the QD surface density. The influence of the substrate temperature and the decrease in the flux of in adatoms on the InAs QD structural characteristics has been studied.

All the studied structures were grown on the GaAs (001) substrates by molecular beam epitaxy at the Riber 21 Compact setup having a solid-state arsenic source. The structures consist of one InAs QD layer formed in the Stranski–Krastanov growth mode by depositing an InAs layer of a certain effective thickness at the arsenic As_4 effective pressure of $2 \cdot 10^{-7}$ Torr. In the process of depositing the InAs QDs, the substrate temperature was varied in the range of 460–540°C, while the remaining epitaxial layers were grown at 600°C. As for the structures to be studied by atomic force microscopy (AFM), their epitaxial growth was stopped once the QD array was formed, and fast cooling of the substrate was performed in the flux of arsenic. In the structures to be studied by transmission electron microscopy (TEM), the InAs QD

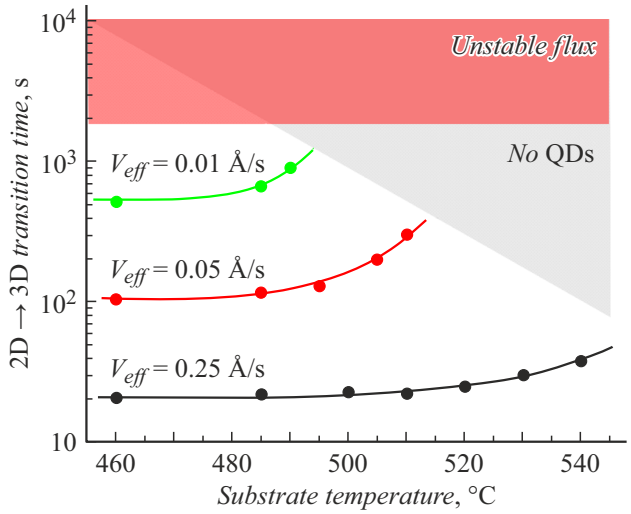

Figure 1. Dependences of the 2D → 3D transformation time on the substrate temperature during the InAs deposition at the fluxes of In adatoms corresponding to the growth rates of InAs V_{eff} = 0.25, 0.05 and 0.01 Å/s.

array was capped with a GaAs layer 30 nm thick; the first 10 nm of the layer were deposited at the QD growth temperature.

The InAs growth rate may be quantitatively determined based on the time of the reflection high-energy electron diffraction pattern transformation from that characteristic of the layer-by-layer (two-dimensional) growth to that characteristic of the growth of coherently stressed islands (three-dimensional) (hereinafter referred to as the 2D → 3D transformation time). It is commonly accepted that the 2D → 3D transformation is observed when an InAs layer of the critical thickness (∼ 1.6−1.7 ML) is deposited on the GaAs surface. However, the InAs growth rate depends not only on the incident flux of In adatoms but also on the indium sticking coefficient. With increasing substrate temperature, desorption of the In adatoms from the growing surface increases and their sticking coefficient decreases, while the flux of In adatoms incident on the substrate is defined by the effusion cell temperature and is independent of the substrate temperature. Therefore, the effective InAs growth rate (V_{eff}) was determined based on the 2D → 3D transformation time at the substrate temperature of 460°C, when desorption of the In adatoms may be ignored and the sticking coefficient of in adatoms is close to unity.

Fig. 1 presents the dependences of the 2D → 3D transformation time on the substrate temperature at different flux of In adatoms corresponding to V_{eff} = 0.25, 0.05 and 0.01 Å/s. At lower substrate temperatures (below 485°C), a proportional increase in the 2D → 3D transformation time is observed with decreasing flux of In adatoms towards the growing layer surface. However, the 2D → 3D transformation increases times with increasing substrate temperature, which is caused by not only enhancement of the In adatom desorption, but also by enhancement of the effect of the deposited InAs intermixing with underlying GaAs, which leads to an increase in the InAs critical thickness [13]. Finally, the In adatom desorption begins prevailing over adsorption starting from a certain critical temperature, and formation of QDs is no longer observed.

Fig. 2 presents the results of analyzing the surface density of InAs QDs obtained by depositing an InAs layer ∼ 2.2 ML thick (controlled as exceedance of the 2D → 3D transformation time by 30%, since the critical thickness depends on the substrate temperature and flux of In adatoms) at different substrate temperatures and flux of In adatoms. The AFM data shows that, when the flux of In adatoms is high (V_{eff} ∼ 0.25 Å/s), the substrate temperature increase from 485 to 510°C causes a decrease in surface density of the QD array from $4 \cdot 10^{10}$ to $1.8 \cdot 10^{10}$ cm^{-2} and an essential increase in the QD lateral size; this correlates well with earlier published data [5,14]. When the substrate temperature continues increasing, there appears a tendency for the emergence of large dislocated QD clusters and/or conglomerates partially evaporating during subsequent QD regrowth depending on thickness of the GaAs capping layer [15]. Notice that, under the conditions of high desorption rate and enhanced surface migration of In adatoms, the sample cooling mode strongly affects the morphology of non capped QDs; therefore, in these cases estimation of the QD surface density based on AFM data is, more likely, of the qualitative character, while the quantitative estimate may be obtained from TEM results.

At the same time, the decrease in the flux of In adatoms allows more efficient control of the QD surface density due to a longer duration of surface migration (since the 2D → 3D transformation time increases) [7]. When the substrate temperature is 485°C and the flux of In adatoms corresponds to V_{eff} ∼ 0.01 Å/s, a reduction of the QD surface density down to $3 \cdot 10^9$ cm^{-2} is observed. Though work [7] has demonstrated the in principle possibility of the QD density reduction to $2 \cdot 10^8$ cm^{-2} with continuously decreasing incident flux of In adatoms, stable operation of the effusion cell in this mode can hardly be ensured technologically. The existence of a considerable desorption at reduced flux of In adatomses restricts selection of the limiting temperature of the InAs QD formation to the values not higher than 510 and 495°C at V_{eff} ∼ 0.05 and ∼ 0.01 Å/s, respectively. As a result, the QD array surface density may be controllably reduced to ∼ $(1-2) \cdot 10^9$ cm^{-2} by using substrate temperatures of 505 and 490°C in choosing rates V_{eff} ∼ 0.05 and ∼ 0.01 Å/s, respectively. Generally, the attained values of the QD surface density correlate well with the [6,7] results for comparable flux of In adatoms despite a significant difference in the growth temperatures.

Notice that efficient implementation of single-photon sources based on InAs QD arrays with a lower density (below $1 \cdot 10^9$ cm^{-2}) needs application of either *in situ* electron-beam lithography combined with cathode-luminescence spectroscopy [15,16] or *in*

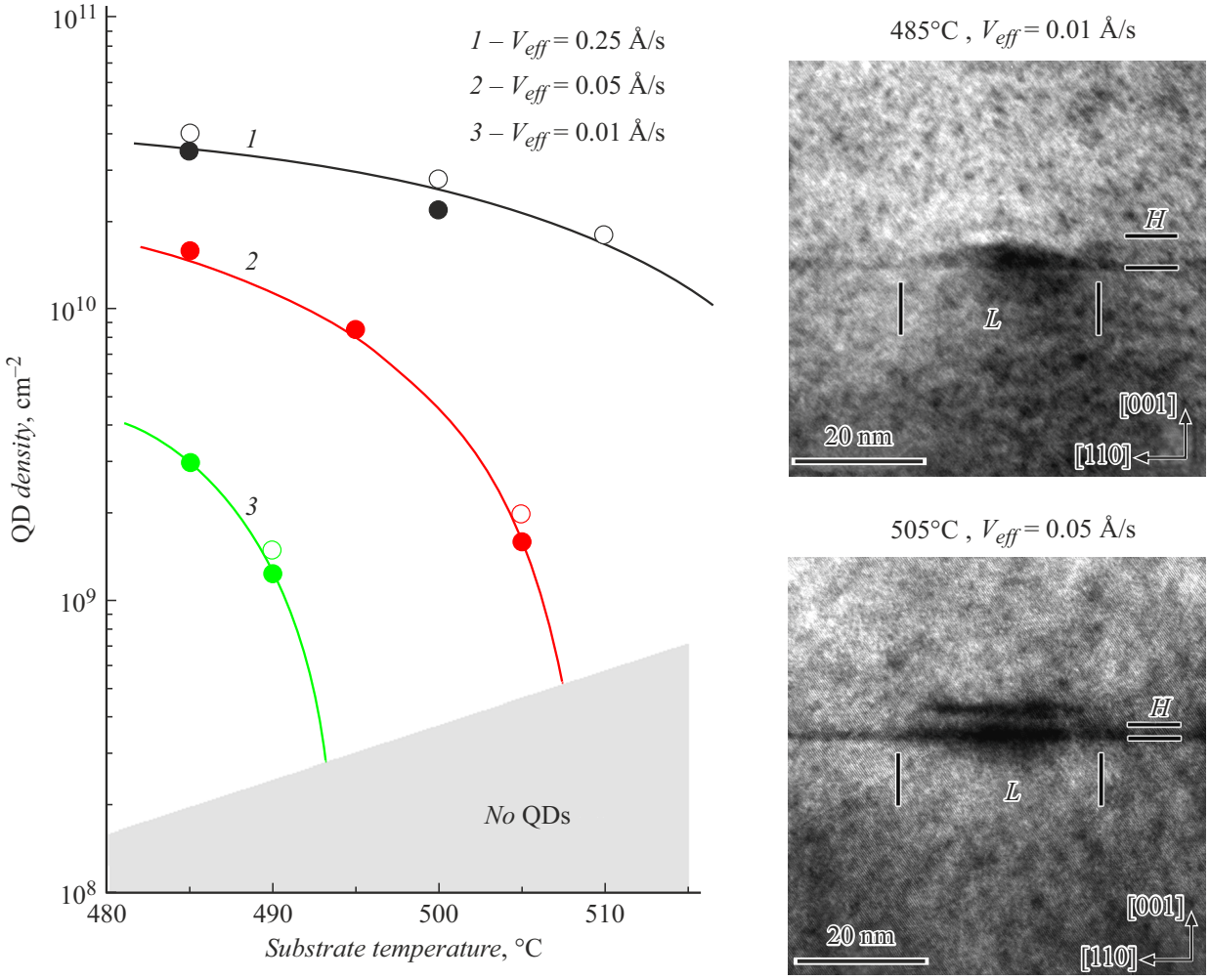

Figure 2. Dependences of mean surface density of the InAs QD array (2.2 ML) on the substrate temperature at the fluxes of In adatoms corresponding to the InAs layer growth rates V_{eff} = 0.25, 0.05 and 0.01 Å/s. Solid circles represent the QD density determined by AFM, open circles represent the QD density determined by TEM. The region free of QD formation is colored grey. The insets present the typical TEM images of the $(1\bar{1}0)$ cross-sections of individual InAs QDs formed in different growth modes.

situ cryo-photolithography combined with the micro-photoluminescence system [17–19] to ensure prechoosing and spatial selection of single QDs with proper spectral characteristics at cryogenic temperatures.

Fig. 3 presents the photoluminescence (PL) spectra for the InAs QDs (2.2 ML) grown in different modes; in measuring the spectra, cryogenic temperatures and low pump density were used in order to adequately estimate the extent of non-uniform broadening of the QD array. On the one hand, the substrate temperature increase at a high flux of In adatoms ($V_{eff} > 0.25$ Å/s) promotes formation of the InAs QD array whose PL spectrum peak has an almost Gaussian shape, which may be interpreted as a more symmetric but broad QD size distribution. In addition, in this case a short-wavelength shift of the QD PL peak is observed (to ~ 1000 nm). Taking into account the observed increase in the lateral size, this can evidence both a significant decrease in the QD height (and, hence, a decrease in the QD volume) and enhancement of the effect of deposited InAs intermixing with underlying GaAs [13]. However, in the case of InAs QDs grown at the substrate temperatures above 520°C an additional sharp decrease in the PL intensity is observed; which, apparently, can be associated with formation of dislocations in QDs exceeding a certain critical volume and/or formation of large dislocated InAs clusters. On the other hand, reduction of the flux of In adatoms leads to a considerable long-wavelength shift of the QD PL spectrum (from ~ 1102 to ~ 1195 nm), which correlates with the [7] results. TEM investigations revealed an increase in the QD average lateral size (up to 27 ± 2 nm) and changing of the QD shape from pyramidal to a flatter one (see the upper inset of Fig. 2). In addition, a decrease in the peak half-width of the QD ground state is observed, which may be interpreted as formation of a more uniform QD array. When the flux of In adatoms are low, the PL spectra exhibit an essential contribution of the wetting

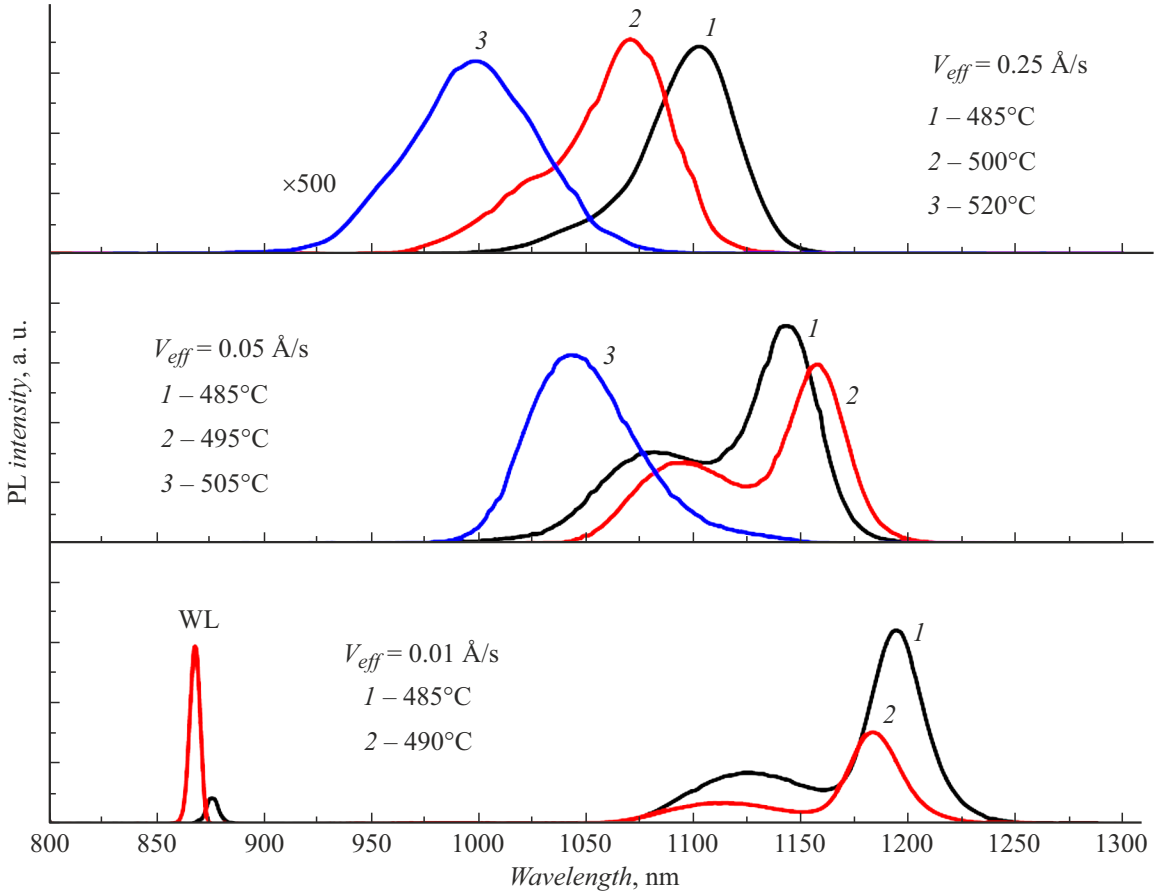

Figure 3. Photoluminescence spectra of InAs QDs (2.2 ML) grown at different substrate temperatures and fluxes of In adatoms corresponding the InAs layer growth rates of $V_{eff} = 0.25$, 0.05 and 0.01 Å/s. Optical pumping was performed using laser YAG:Nd (532 nm), excitation density was 30 W/cm^2, measurement temperature was 10 K.

layer (WL), which evidences a decrease in the QD array density.

Notice that PL spectra of samples grown at an elevated substrate temperature under a lower flux of In adatoms have features distinct from those of similar samples grown at a high flux of In adatoms ($V_{eff} > 0.25$ Å/s). For instance, at an intermediate flux of In adatoms ($V_{eff} \sim 0.05$ Å/s), first an additional long-wavelength shift of the QD ground state peak (from ~ 1143 to ~ 1158 nm) is observed with increasing substrate temperature (from 485 to 495°C); this evidences further increase in the QD volume. However, the situation changes drastically at the substrate temperature of 505°C, and there is observed a tendency similar to the case of InAs QDs grown at a high flux of In adatoms and elevated temperature, namely, further increase in the QD lateral size (up to 31 ± 2 nm) and change in the QD shape (see the Fig. 2 bottom inset) accompanied by formation of dislocations in larger QDs and, hence, by a short-wavelength shift of the QD PL peak (to ~ 1043 nm). Notice that formation of QDs does not occur at higher substrate temperatures.

In case the flux of In adatoms is low ($V_{eff} \sim 0.01$ Å/s), even a small increase in the substrate temperature (from 485 to 490°C) leads to a short-wavelength shift of the QD ground state (from ~ 1195 to ~ 1183 nm); this indicates the impossibility of further increase in the InAs QD volume within the considered approach. The observed increase in the wetting layer peak intensity and decrease in the QD emission intensity may be interpreted as a reduction of the QD array surface density [7].

Thus, minimizing the flux of In adatoms ($V_{eff} < 0.01$ Å/s) to the growing layer surface in order to enhance the In adatom surface migration enables efficient reduction of the InAs QD array density to $\sim (1-3) \cdot 10^9$ cm^{-2} with a simultaneous shift of the PL peak corresponding to the QD ground state to 1.3 μm at room temperature. However, this method is efficient only in a narrow temperature range ($\sim 5-10$°C).

Acknowledgements

The QD structural parameters were analyzed at CCU „Material Science and Technologically Advanced Diagnostics“ (Ioffe Institute).

Financial support

The study was supported by the Russian Foundation for Basic Research and DFG (project № 20-52-12006). A.M. Nadtochiy is grateful to the HSE NRU Fundamental Research Program.

Conflict of interests

The authors declare that they have no conflict of interests.

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