

Regular arrays of triple InAs/GaAs quantum dots in a triangular arrangement

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The study presents a novel approach for the epitaxial growth of ordered arrays of triple quantum dots arranged in a triangular configuration. The proposed method involves preliminary patterning of GaAs(111)*B* substrates using a combination of focused ion beams and local droplet etching followed by depositing InAs via molecular beam epitaxy. Optimization of the technological parameters allows for high selectivity and yield of triple quantum dots within the array, as well as for control over the inter-dot distances.

Keywords: molecular beam epitaxy, focused ion beams, nanopatterning, quantum dots.

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Recently, triple quantum dots (QDs) in various configurations have attracted increasing interest due to their unique physical properties and significant potential for application in promising devices in nano- and optoelectronics, plasmonics, and quantum technologies [1–3]. One of the most common configurations is a chain of linearly arranged triple QDs allowing, for instance, the control of three individual spins and coherent electron transport between two remote QDs [4,5]. However, theoretical [1,3,6] and experimental [7,8] studies demonstrate that the triangular arrangement of triple QDs provides a number of additional advantages related to the possibility of realizing charge frustration [1,8] and increasing the reliability of quantum teleportation [6], as well as the degree of qubit entanglement [3].

The most widely used technique for fabricating QD complexes with controllable configuration is lithographic formation of metal gates above a semiconductor layer with two-dimensional electron gas [4,7,8]. However, this approach has a number of significant disadvantages, such as complexity and high cost, as well as QD susceptibility to charge noise and fluctuations in the energy level positions [9]. At the same time, epitaxial QDs are practically ideal nanophotonic elements with the required optical properties and possibility of arbitrarily reducing their sizes without significant complication of the manufacturing process. Molecular beam epitaxy (MBE) ensures the highest quality and purity of the material, as well as sharp heterointerfaces. However, issues related to developing scalable approaches to epitaxial QD formation at preset positions still remain open.

Available publications pay special attention to InAs-based systems because of their high potential for photonics in the range of telecommunication wavelengths, low effective mass of charge carriers, strong spin-orbit interaction, and other advantages [10,11]. One of the options for experimental implementation of linear triple QDs is the use of

InAs nanowires fed with voltage via transversely arranged elongated gates [12]. Quantum-dot InGaAs molecules were also formed using a combination of local Ga-droplet etching (LDE) and subsequent deposition of InAs [13]. However, as far as we know, epitaxial growth of triangular-arranged triple QDs has not yet been implemented. Moreover, there is no evidence to demonstrate techniques for scalable fabrication of ordered arrays of such QDs.

This paper presents an approach to fabricating ordered arrays of epitaxial triple InAs QDs in the triangular configuration. Such QDs are fabricated using GaAs(111)*B* substrates subjected to modification by combined methods of focused ion beams (FIBs) and LDE, which leads to creation of regular arrays of pyramidal pits. In the process of InAs deposition, nucleation of islands takes place predominantly in the pyramid base vertices, which ensures formation of triangular QD complexes.

The study was performed by using epi-ready (111)*B* GaAs substrates. The surface modification was performed in scanning electron microscope (SEM) Nova NanoLab 600 equipped with a FIB column with a source of Ga⁺ ions. FIB-modification points were located at the distance of 0.5 μm apart from each other within a square matrix 4 × 4 μm in area. The ion beam acceleration voltage (*U*) was 10 and 30 kV. The ion irradiation dose was controlled by varying number *N* of the ion beam passages. Upon the FIB-modification, the substrates were placed in the growth chamber of the MBE SemiTEq STE 35 system and annealed in the flux of As₄ at 600 °C. In the next stage, QDs were formed by depositing an InAs layer 2.5 monolayer thick on the patterned surface at the temperature of 500 °C. The samples' morphology was studied by atomic force microscopy and SEM.

Immediately upon the FIB treatment, regular arrays of uniformly shaped and sized pits get formed on the

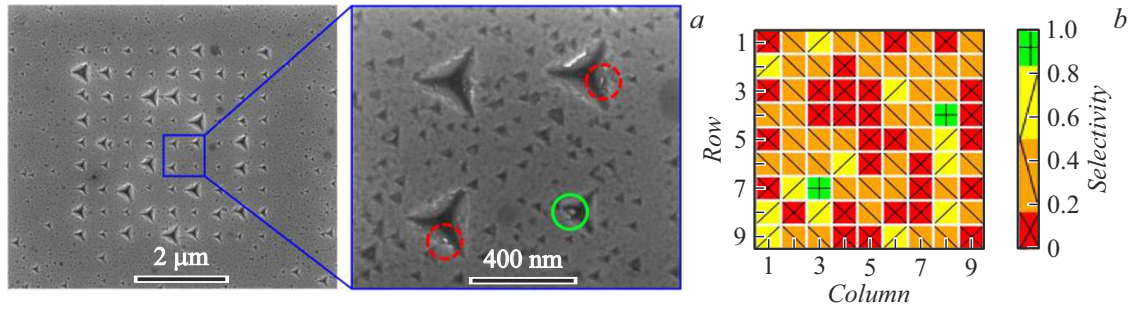


Figure 1. SEM images (*a*) and selectivity map of triple QDs (*b*) in the array of pits obtained after FIB-modification of the substrate at $U = 10$ kV and $N = 1$. In the map, red cells with diagonal cross-hatching correspond to zero selectivity ($\theta = 0$) when QDs do not emerge at lateral vertices of pyramidal pits; orange and yellow cells with left and right diagonal hatching represent pits with one and two QDs in lateral vertices ($\theta = 0.33$ and 0.67); green cells with vertical-horizontal hatching represent the pits where exactly three QDs have been formed in lateral vertices ($\theta = 1$). The color versions of figures are presented in the electronic version of the article.

GaAs(111)*B* surface. As the number of FIB passes increases from 1 to 60, the pit diameter increases monotonously from ~ 50 to ~ 120 nm. The depth of the modification sections varies in a similar way and reaches 30 nm after 60 FIB passes. Significant variations in the pit size and shape occur after annealing FIB-modified substrates, which results in local gallium-droplet etching of sites exposed to FIB; the etching mechanisms were discussed in detail in our previous study [14]. The pits formed at the FIB-modification points have got the pronounced shape of pyramids (tetrahedra) with high symmetry and increased size: when the number of FIB passes varies from 5 to 60, the diameter increases monotonously from ~ 190 to ~ 290 nm. Such a significant etching-out of the surface is caused by a high concentration of gallium embedded at the points exposed to FIB.

The pits formed at small N exhibit a large spread in size: at $N = 1$, the pit diameter standard deviation is 150 nm, whereas at higher values (5 to 60) it varies from 8 to 21 nm. In addition, at small N arrays may contain pits of both small and very large size (up to ~ 410 nm in diameter), which is unattainable at high N (Fig. 1, *a*). As N increases, the LDE process becomes more uniform because etching is stabilized by the competing process of the etching material (gallium) crystallization in the arsenic flux inevitably existing in the growth chamber.

Deposition of the 2.5 InAs monolayer led to formation of clearly distinguishable islands at the vertices of pyramidal pit bases (marked with green solid circles in Figs. 1, *a* and 2, *a*). The InAs islands may also emerge at other positions: in the pit center, along the joints of pyramid faces or on the faces themselves (marked with red dashed circles in Fig. 1, *a*). However, in the context of this study whose goal was formation of triple QDs in the triangular configuration, such nucleation is considered undesirable.

Fig. 1, *a* demonstrates that, despite the fact that pits in the array formed at $N = 1$ are distinguished by a significant size spread, they also have the shape of full or truncated pyramids and contain InAs nanostructures predominantly at the vertices of their bases (lateral vertices). A higher

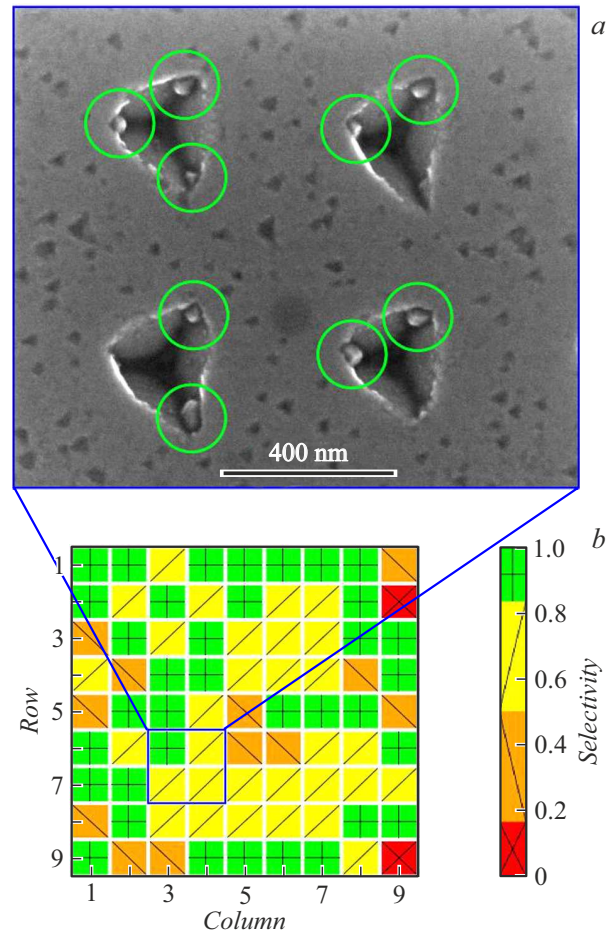


Figure 2. SEM images (*a*) and triple QD selectivity map (*b*) in the array of pits obtained after the substrate FIB-modification at $U = 10$ kV and $N = 20$.

probability of QD nucleation at the pyramid base vertices is explained by the presence of protruding faces preventing the surface diffusion of atoms from the pit.

To quantify the selectivity degree θ of triple QDs in the pits, this study uses relation $\theta = n / \max(N_{tot}, 3)$ where n

is the number of QDs in lateral vertices, N_{tot} is the total number of QDs in a pit, $\max(N_{tot}, 3) = 3$ if $N_{tot} \leq 3$, and N_{tot} if $N_{tot} > 3$. According to this relation, the selectivity is maximal (equal to unity) when exactly three QDs get formed in the vertices of the pyramidal pit base; in other cases it decreases due to either the absence of QDs in the required positions or the presence of parasitic QDs outside the lateral vertices.

Selectivity of the array QDs may be conveniently visualized by using special maps where the cells' color and hatching correspond to the value of QD selectivity. The triple QD selectivity map for the sample with $N = 1$ (Fig. 1, *b*) shows that only 2% of pits in the array contain triple QDs ($\theta = 1$), while the remaining pits contain no more than two QDs. Hereinafter, metric characterizing the triple QD share in the array is referred to as triple QD yield. As shown in Fig. 3, *a*, the maximum average selectivity and yield of triple QDs at 10 kV are 0.76 and 49%, respectively. The selectivity map for $N = 20$ is shown in Fig. 2, *b* and also indicates that triple QDs are formed predominantly along the array perimeter; this is associated with accumulation of material deposited beyond the FIB-modified region in outermost pits and filling with islands of all the most preferable nucleation centers. When the number of passages ranges from 10 to 60, average selectivity of triple QDs at 10 kV varies from 0.72 to 0.76 and decreases to 0.62 in the case of five FIB passes ($N = 5$) and to 0.29 in the case of

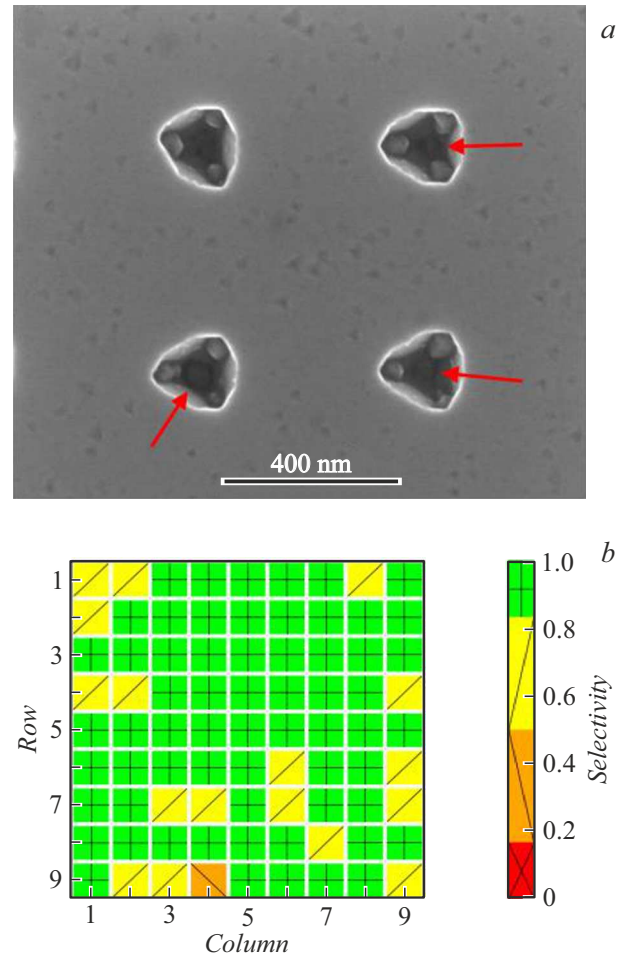


Figure 4. SEM images (*a*) and triple QD selectivity map (*b*) in the array of pits obtained after the substrate FIB-modification at $U = 30$ kV and $N = 30$ (*a*) and 20 (*b*). Arrows indicate parasitic QDs nucleating in the pit centers.

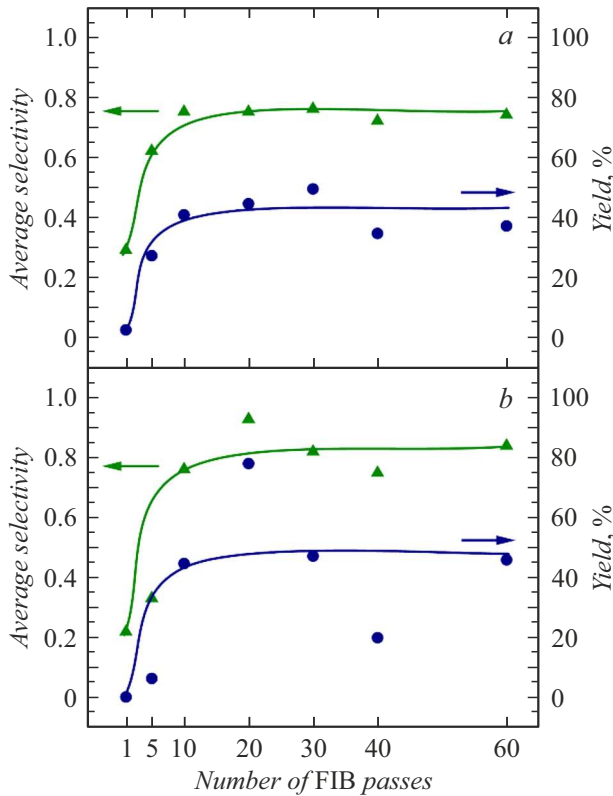


Figure 3. Average selectivity and yield of triple QDs versus the number of FIB passes at $U = 10$ (*a*) and 30 kV (*b*). The curves are drawn as visual guides.

one passage ($N = 1$) (Fig. 3, *a*). The yield of pits with triple QDs is 2% in the case of one FIB pass and ranges from 27 to 49% in the remaining range of N .

The dependences in Fig. 3, *b* demonstrate that an increase in the FIB acceleration voltage to 30 kV leads to a significant increase in both the triple QD average selectivity and yield which reach 0.93 and 78%, respectively, for the FIB pass number $N = 20$ (Fig. 4, *b*). As the number of FIB passes increases, the probability of island nucleation at the pit lateral vertices increases; however, the increase in both considered parameters is limited by nucleation in the pit centers (islands marked with arrows in Fig. 4, *a*). In the context of this study, such QDs are then considered parasitic and reduce the selectivity from 1 to 0.75 also decreasing the triple QD yield in the array. The QD nucleation in pit centers explains the significant yield decrease after 40 FIB passes (Fig. 3, *b*). After 60 passages, the QD yield stabilizes again, which is associated with emergence of a crater in the pyramidal pit center caused by large ion implantation

depth at 30 kV and related deep Ga-droplet etching of the substrate.

The minimum average selectivity of triple QDs in the case of 30 kV is 0.22 (at $N = 1$), whereas that at N of 10 to 60 varies from 0.75 to 0.93 (Fig. 3, *b*). At the same time, while at $N = 1$ the triple QD yield is zero, that at $N = 20$ reaches 78 %. Note also that the fraction of pits each containing at least one QD in a lateral vertex is 96–100 % for 10 kV and 98–100 % for 30 kV at N of 10 to 60.

In addition to an increase in selectivity of triple QDs in pits, an important consequence of increasing the FIB voltage is the possibility of reducing the average inter-QD distance in the triangular complex. While at 10 kV the average inter-QD distance at $N = 30$ is ~ 210 nm, that at 30 kV decreases to ~ 100 nm.

Thus, the paper demonstrates an approach to fabricating ordered arrays of triple InAs QDs in a triangular configuration on a patterned GaAs(111)*B* surface. The obtained triple QD complexes may be used as an object of fundamental research and also in fabricating promising devices for nanoelectronics and quantum technologies.

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Conflict of interests

The author declares that he has no conflict of interests.

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