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Low-density arrays of complexes of triple InAs/GaAs(111)B quantum dots

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An approach for epitaxial formation of ordered complexes of InAs/GaAs triple quantum dots in a triangular configuration with low surface density ($0.25-1\,\mu m^{-2}$) is presented. Quantum dots are formed at the vertices of the bases of pyramidal holes on GaAs(111)B substrates patterned via a technique based on a combination of focused ion beams and local droplet etching. The distance between the dots in the complex is determined by the patterning topology and increases with increasing beam treatment period. Optimization of the patterning parameters ensures high selectivity of triple quantum dots in the holes. The photoluminescence spectra of quantum dots exhibit emission in the range of 900−1150 nm with an intensity that rises with an increase in the ion dose and a decrease in the beam treatment period.

Keywords: quantum dots, InAs, GaAs(111)B, epitaxy, focused ion beam.

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1. Introduction

The current stage of the technological development is characterized by the active search for and transition to the fundamentally new systems of data transmission, processing, and storage, such as quantum cryptography, spintronics, optical and quantum calculations. One of the most promising objects of the study in this respect are the semiconductor quantum dots (QD), which is due to their unique properties, arising from the marked effect of dimensional quantization. The atomiclike energy spectrum of QD makes it possible to absolutely precisely control the charge and spin states, and also to control the tunnel processes, which serves as a physical basis for the functioning of qubits and elements of nanoelectronics. Besides, the semiconductor QDs based on $A^{III}B^{V}$ compounds perfectly integrate into the photonic platform for the implementation of the systems of optical quantum computing and communications [1], which is related to their high quantum efficiency, ability of electric pumping and precision control of optical properties, and also the mature technology for the manufacture of heterostructures on their basis.

The study of the complexes of triple QDs in various configurations is of specific interest for the purposes of implementation of nano- and optoelectronics [2–4], plasmonics [5] and quantum technologies [6–8] devices on their basis. Thus, in paper [9] based on a linear chain of three QDs, the effect of superexchange was experimentally demonstrated — the mediated link between the remote spins through the third intermediate electron, which opens the alternative path for the implementation of the quantum gates between the remote qubits [9]. Due to the precise tuning of energy levels in the linear triple QDs, the direct coherent transfer of electron was achieved between the remote dots, which opens the prospects of implementation of the quantum bus for the exchange of information between the qubits at large distances [10]. Triple QDs in a triangular configuration are of special interest, too, which is related to their outstanding thermoelectric properties [11], the ability to implement the charge frustration [4,12], to control a chiral current [13], and also to increase the degree of entanglement of qubits [6] and the reliability of quantum teleportation [14].

To develop the full-scale highly efficient systems of quantum computing, nanoelectronics and photonics, apart from the achievement of high quality of QDs, it is also necessary to provide for their low surface density (as a rule, not more than $1\,\mu m^{-2}$), making it possible to isolate certain QDs and their complexes for the development of the functional devices on their basis. For these objectives, the method is actively used nowadays to localize the formation of QDs in the holes obtained by local droplet etching (LDE) [15,16], which allows varying the QD density in the wide range of the values. The LDE effect has been long known in the literature and is used to create the holes of symmetrical pyramidal shape, where the QD material is further deposited [17]. Apart from the ability to decrease the surface density of the QD, such method provides for the pyramidal shape of the holes, making it possible to achieve high symmetry of QDs and, accordingly, the minimum values of the fine-structure splitting in the cascade biexciton–exciton decomposition to increase the degree of entanglement of photons [18]. However, the main drawback of the LDE is the absence of control over the position of the developed holes on the surface and, consequently, QDs formed therein in virtue of the random nature of the processes of formation of metal drops that etch the substrate. This is one of the key obstacles on the path of the development of the scalable technology for the

manufacturing of the devices on the basis of low density QDs and their complexes, including triple ones.

One of the options for the solution to the specified problem is the development of the QD complexes with the controlled configuration using the lithographic formation of the system of metal gates above the heterostructure with 2D electron gas (2DEG). However, this approach has some significant limitations. First, the lithographic manufacturing of nanostructures with the specific dimensions of 20–30 nm is technologically a very complicated and expensive process. Second, QDs induced by gates in 2DEG are highly sensitive to charge noise, being one of the common reasons for the decoherence of qubits. Finally, such QDs are not stable, isolated fragments of the materials, which makes them susceptible to the fluctuations of the energy levels. As a result, they demonstrate unsatisfactory values of quantum efficiency, emission linewidth and spectral diffusion. At the same time the epitaxial QDs represent practically ideal objects of photonics, having the required optical properties and the ability to control the QD dimensions in a wide range in process of growth. Besides, the epitaxial technology provides for high quality and purity of the material, and also the stability of the positions of the energy levels due to sharp heteroboundaries between the QDs and the surrounding matrix. Nevertheless, the issues related to the development of the scalable approaches to the epitaxial formation of QDs in the strictly specified positions still remain open.

The high significance of the precision positioning of QDs and their complexes stimulates the development of the methods to create arrays of preferential nucleation sites on the surface. To structure the surface, various types of lithography are used: optical, electron beam, nanoimprint, nanosphere, laser interference etc.[19]. However, their major drawback is the need to use the etching agents leaving impurities and pollutants in the active region of the heterostructure, which inevitably deteriorates its electron and optical characteristics. The alternative solution is the method of focused ion beams (FIB) [20], which makes it possible to set the centers of QD nucleation with the required surface density and specified geometric parameters without the use of masks, and also provides for a wide range of control over the degree of damage to the surface layer [21]. Using the combination of FIB and molecular beam epitaxy (MBE), highly ordered arrays of QDs In(Ga)As were obtained previously on substrates GaAs(001) [22–24]. However, according to the literature data, the epitaxial formation of low-density arrays of the complexes of triple QDs with high selectivity has not yet been demonstrated.

This paper presents the approach to the manufacture of ordered arrays of epitaxial complexes of triple QDs InAs, formed in the triangular configuration, with low surface density. To manufacture such QDs, the features of crystallography of GaAs(111)B substrates are used, which were exposed to modification using the method we proposed previously on the basis of the combination of FIB and LDE [24,25], causing the formation of regular arrays of pyramidal holes. Subsequent deposition of InAs causes preferential nucleation of islands at the vertices of the pyramid base, providing for the formation of equilateral triangles from three QDs. Variation of the structuring modes at the stage of FIB-processing makes it possible to manage the size of the holes and accordingly the distance between the QDs in a triple complex, and also the selectivity and the yield of triple QDs in the array. Photoluminescence (PL) spectra demonstrate that such QDs radiate in the range of 900–1150 nm with the intensity that depends both on the number of passages, and on the distance between the points of exposure of FIB.

2. Experiment method

The ordered arrays of QDs were formed on GaAs(111)B class „epi-ready“ substrates. The first stage included the development of topographic defects by FIB method in a two-beam electron microscope Nova NanoLab 600 with the Ga^+ ion source. The points of FIB-processing were arranged with a period of (L) 1 and 2 μm within the square areas with the surface area of $4 \times 4\,\mu m^2$. Accelerating FIB voltages (U) 10 and 30 kV and various ion radiation doses controlled by changing the number of passages N (number of single exposures) of the ion beam were used for modification. After modification of the surface, the samples were discharged from the microscope and exposed to natural oxidation in the laboratory box atmosphere at room temperature for 24 hrs. At the next stage the samples were loaded into the growth chamber of MBE SemiTEq STE 35 system, where they were annealed in the flux of As_4 at temperature of 600 °C for 60 min., implementing the LDE procedure with the formation of pyramidal holes in accordance with the mechanism described in paper [25]. Then InAs with thickness of 2.5 monolayers was deposited on the surface structured in this manner at temperature of 500 °C for the formation of QDs. For optical measurements, QDs were covered with a coating layer of GaAs with thickness of 50 nm. The relief and the geometric parameters of the obtained structures were controlled by the method of scanning electron microscopy (SEM). The optical properties of the covered QDs were studied by the method of photoluminescence (PL) spectroscopy at 77 K. The pumping was carried out with an Nd:YAG-laser operating in continuous mode ($\lambda = 532$ nm), with the laser spot diameter $\sim 2\,\mu$m and density of excitation power 1 kW/cm^2.

3. Findings and discussion

As a result of GaAs(111)B substrate processing using the above procedure (FIB-modification with the subsequent LDE), holes were formed on the surface in the form of inverted pyramids (tetrahedrons). The diameter of the holes relative to the FIB-modified areas at the same time increased more than twofold, which is related to the intense etching of the surface with gallium, introduced in the points of

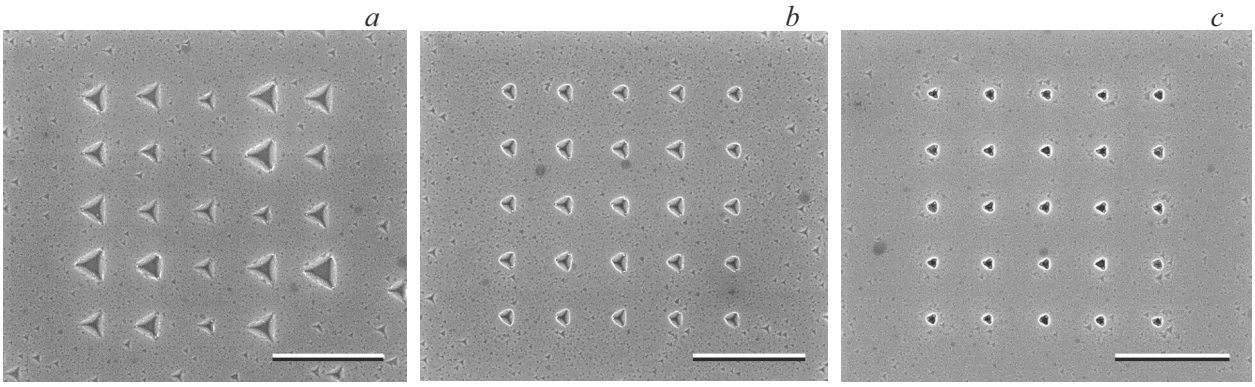


Figure 1. SEM-images of arrays of holes with QDs obtained after FIB-modification of the substrate at $L = 1\,\mu$m: $U = 10$ kV, $N = 1$ (a); $U = 10$ kV, $N = 60$ (b) and $U = 30$ kV, $N = 40$ (c). Scale length — $2\,\mu$m.

exposure to the ion beam. If the number of passages is low, FIB holes have a significant spread in dimensions. At $U = 10$ kV and $L = 1\,\mu$m in the range of N from 1 to 10 passages the standard deviation of the diameter varies from 19 to 31 %, whereas at higher values — from 4 to 6 %. Besides, the minimum diameter of the hole in the array at $N = 1$ is ~ 170 nm, whereas the largest diameter reaches ~ 660 nm (Figure 1, a). This phenomenon is explained by the fact that at low doses the etching process prevails over the competing process of recrystallization of FIB-modified regions of the substrate, enriched with gallium, in the background flux of arsenic present in the growth chamber. At the same time, the influence of random factors increases, such as uneven distribution of ions in FIB-processing and thermal dissociation of the GaAs oxide layer with a complex composition, destabilizing the process of droplet etching and thus increasing the spread of dimensions and the morphology of the holes in the array. As the number of FIB passes increases, the homogeneity of the holes by dimensions in the arrays rises (Figure 1, b), which is due to the minimization of the above random factors and achievement of the equilibrium between the competing processes of LDE and gallium crystallization.

At $U = 30$ kV the trend to decrease the spread of hole dimensions with the increased number of FIB passes persists, but also a dramatic transition occurs from the large holes to smaller holes, which is explained by the decrease in the diameter of the ion beam and minimization of the supporting ion scattering processes. At $N = 20$ the array contains holes of both types (large to ~ 560 nm and small to ~ 200 nm), however, as N further increases, the holes have approximately the same average diameter ~ 210 nm (Figure 1, c).

Another important feature of the considered method for substrate structuring is the reverse dependence of the roughness values in the surface regions between the holes on the number of FIB passes, which is due to growing importance of the role of redeposition of the sputtered material. Being deposited above the film of its own oxide on the surface of the FIB-modified substrate, it serves as a sacrificial layer, which reacts with it in process of heating, forming volatile products of chemical reaction and facilitating the removal of surface oxides prior to the start of accumulation of surplus gallium conglomerates under them, which etch, as in the above cases of LDE, the substrate and cause the development of the surface relief [26].

The target configuration of the QDs within the considered paper consists of the triple complexes of QDs formed in the tops of the pyramidal hole base (hereinafter referred to as in — lateral ones). Higher probability of QD nucleation in the lateral tops of the hole is explained by the presence of the faces protruding above such tops and preventing surface diffusion of atoms from the hole. Nucleation at the bottom of the hole is related to the higher energy of the atom bond in the point of crossing of pyramid faces. Besides, QDs may also be formed spontaneously in small holes beyond the points of FIB-modification, being actually parasitic structures from the point of view of the regular array. Therefore, to decrease the unwanted nucleation, the preferable is the use of FIB-modification modes, minimizing the roughness between the points of exposure, i. e. with higher number of passages.

The case of nucleation of exactly three QDs in three lateral tops of the hole corresponds to the maximum selectivity of triple QDs. The absence of the QDs in the lateral top or nucleation of QD beyond its limits decreases the selectivity of triple QDs in accordance with formula $\theta = n/\max(N_{\text{tot}}, 3)$, where n — number of QDs in lateral tops, N_{tot} — total number of QDs in the hole, $\max(\mathrm{N_{tot}}, 3) = 3$, if $N_{\text{tot}} \leq 3$, and N_{tot}, if $N_{\text{tot}} > 3$. Figure2, a shows the dependences of the average selectivity of triple QDs in the array on the number of FIB passes at 10 and 30 kV. The curve demonstrates that in case of $L = 1\,\mu$m at 10 kV the selectivity varies within the narrow limits (0.63–0.73) almost in the entire range of numbers of FIB passes (from 5 to 60, with the deviation to the minimum value of 0.47 at 1 passage), and at 30 kV the spread of values is within wider limits, but the highest average value of selectivity increases to 0.9 (at $N = 30$). Reduction in the selectivity of triple QDs at higher values

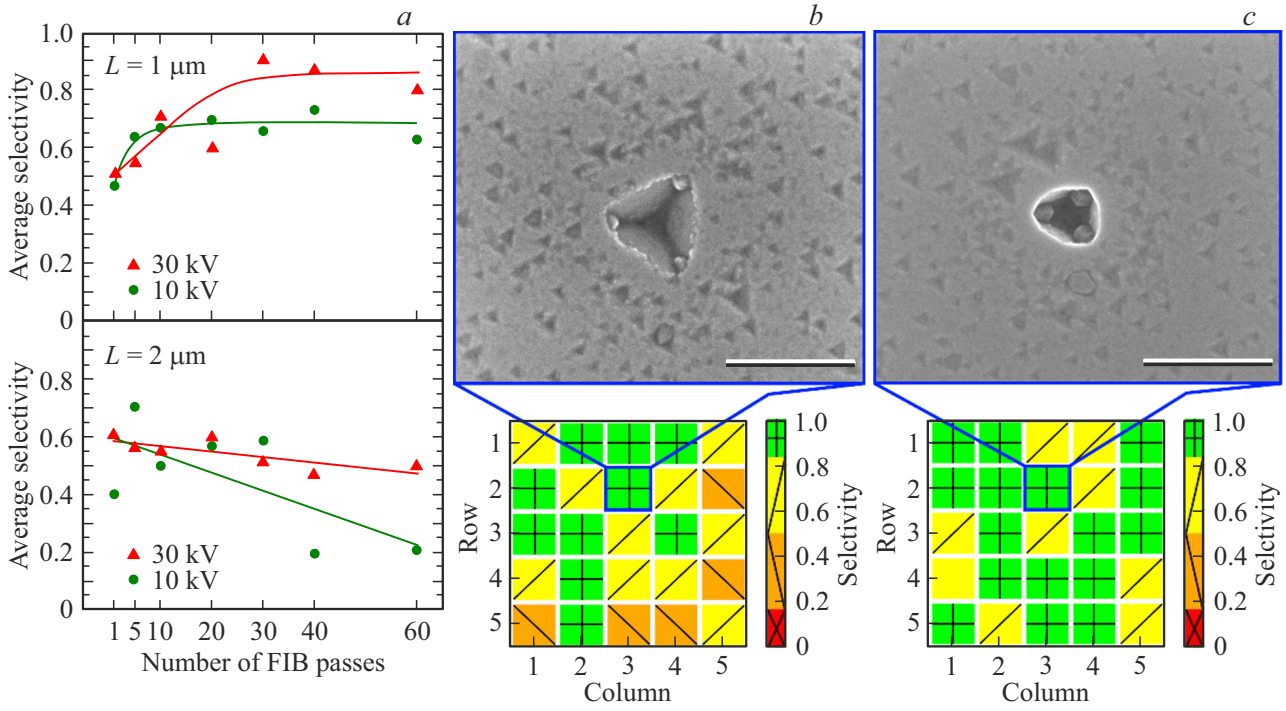


Figure 2. Dependences of the average selectivity of triple QDs (a) in the arrays of holes produced after FIB-modification of the substrate at $U = 10$ and 30 kV, on the number of FIB passes and also SEM images (top row) and chart of selectivity of triple QDs (bottom row) at $U = 10$ kV, $N = 40$ (b) and $U = 30$ kV, $N = 30$ (c). The length of the scale in SEM-images is 400 nm.

of N is explained by the QD nucleation in the center of the pyramidal hole to form the complexes of quadruple QDs, which in the context of the considered paper is unwanted.

SEM-images in Figure 2, b and c demonstrate the typical holes with triple QDs with the number of passages corresponding to the maximum average selectivity at 10 and 30 kV. In both cases triple QDs in the form of an equilateral triangle are formed in the holes, and their share (yield) in the array is 40 % for 10 kV and 64 % for 30 kV, which is visualized on the charts, the hatching and color of cells on which correspond to the selectivity of triple QDs in the hole (at the bottom in Figure 2, b and c). The red color and cross diagonal hatching correspond to the zero selectivity ($\theta = 0$) — absence of QDs in the tops of the pyramid base; orange (left) and yellow (right diagonal hatching) colors correspond to one and two QDs in the tops ($\theta = 0.33$ and 0.67) accordingly; the green color (cross vertical-horizontal hatching) indicates that the hole contains exactly three QDs, each is present in the lateral vertices of the holes ($\theta = 1$). It is clear on the charts of selectivity of triple QDs that the average selectivity and yield of triple QDs increase significantly with the significant increase of the accelerating voltage of FIB. It is also important to note that all holes in the considered arrays contain at least one QD.

The increase in the pitch between the holes up to 2 μm causes a significant increase in the size of holes (Figure 3), related to the decrease in the influence of the redeposition effect: the material of the substrate (GaAs), sputtered with an ion beam and entering adjacent holes, prevents intense etching of the surface due to the reduction of the gallium fraction in the point of exposure to FIB. As a result of gallium diffusion from the area of exposure to the surface, a wide hole is formed in the form of an inverted pyramid with a truncated shape due to the deficit of the etching agent. The influence of the redeposition effect on the size of the holes is also confirmed with a SEM-image in Figure 1, a, according to which even at minimum number of FIB passes ($N = 1$), holes of large diameter may appear on the surface, despite a small dose of ion radiation. The QD material in the holes of such large size is accumulated not only in the lateral tops of pyramids, and also on their faces and in the coupling areas of side faces and the bottom of the hole due to the limitation of the diffusion length of adatoms (Figure 3). In the second case a solid triangular chain of QDs is formed on the bottom of the hole, and this chain may be interpreted as a triangular nanowire.

Despite the presence of parasitic material on the walls of the holes, rounded islands are clearly distinguishable at their lateral vertices: 2 islands at 10 kV (Figure 3, a) and 3 — at 30 kV (Figure 3, b). The average distance between such QDs at the same time increases from ~ 250 nm at $L = 1\,\mu$m to ~ 710 nm at $L = 2\,\mu$m at 10 kV, decreasing to ~ 110 and ~ 640 nm accordingly at 30 kV (Figure 3, c). The average selectivity of triple QDs at $L = 2\,\mu$m decreases significantly and has a decreasing trend with the growth of the number of FIB passes (Figure 2, a), which is due to the damage to the shape and symmetry of pyramidal holes. Therefore, increasing the distance between the points

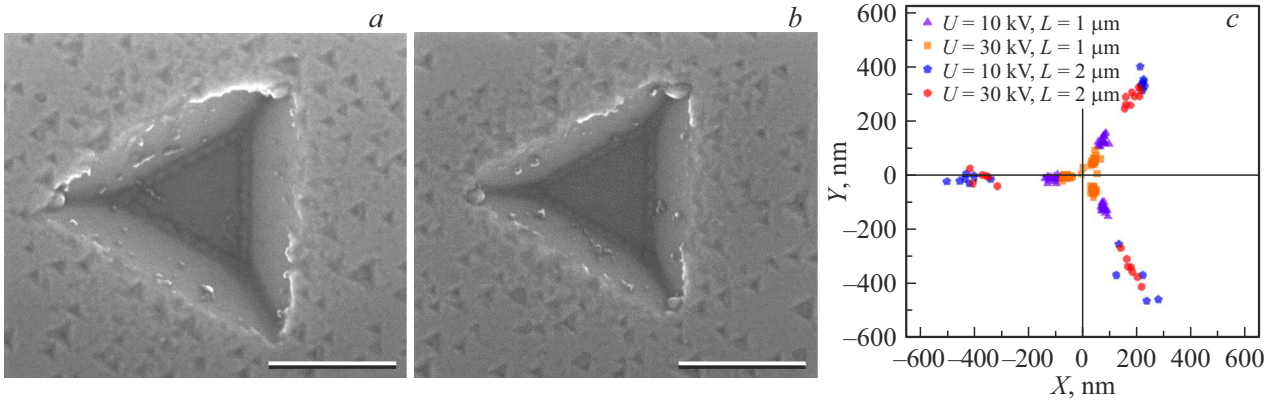


Figure 3. SEM-images of holes with QDs obtained after FIB-modification of the substrate at $L = 2\,\mu$m and $N = 10$: $U = 10$ kV (a), $U = 30$ kV (b) (scale bar: 400 nm), and also the scatter plots of the positions of triple QDs in the tops of the bases of pyramidal holes.

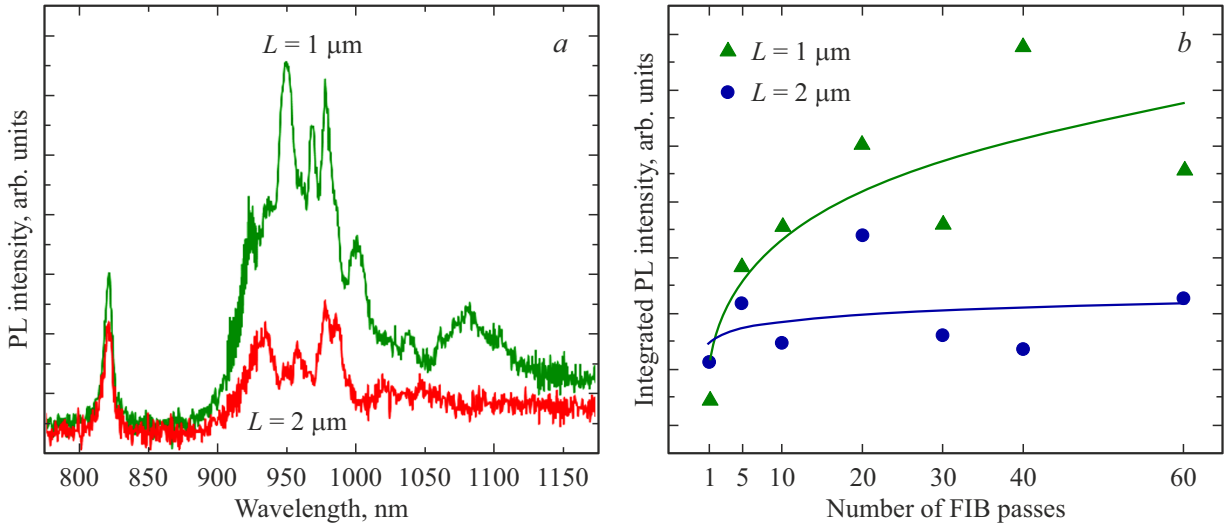


Figure 4. Spectra (a) and dependences of integral intensity of PL in samples with triple QDs on the number of FIB passes (b).

of FIB exposure, you may achieve the reduction in the surface density of complexes of triple QDs to $0.25\,\mu\text{m}^{-2}$ and increase of the distance between the QDs within a single complex. Besides, certain problems remain unsolved, which require further studies and optimization of the modes of preparation of the growth surface: decrease in dimensions of the holes with the preservation of their low surface density (increase in the distance between them), and also simultaneous increase of homogeneity of dimensions of the holes and decrease in the roughness of the surface between them, which will allow providing a small distance between triple QDs in the complex and decreasing the probability of parasitic nucleation at the bottom of the holes.

Samples with covered QDs demonstrate PL radiation in the range of 900–1150 nm at 77 K (Figure 4, a). The PL spectra for both L show several pronounced peaks, presumably, corresponding to various dimensional groups of QDs. Taking into account the diameter of the laser spot $\sim 2\,\mu$m and surface area of ion beam exposure $4 \times 4\,\mu\text{m}^2$, you can assume that in this case QDs are excited approximately from a quarter of the surface area of the modified region, which includes ~ 6 and ~ 2 holes at $L = 1$ and $2\,\mu$m accordingly, each containing ~ 3 QDs. Besides, you cannot exclude the contribution to the PL spectra of parasitic QDs formed between the holes and QDs out of the laser excitation area, which radiate as a result of recombination of charge carriers excited in the area of the laser spot. Therefore, in case of $L = 1\,\mu$m the PL spectrum shows emission from several dozen QDs, most probably forming several dimension groups with the centers of radiation at wavelengths ~ 950, ~ 970, ~ 980, ~ 1000 and ~ 1080 nm. At the same time in case of $L = 2\,\mu$m the number of QDs with the radiation within the PL spectrum is considerably lower, which is reflected in a smaller number of PL peaks: ~ 935, ~ 960 and ~ 980 nm.

The integral intensity of PL in samples with QDs increases as the distance decreases between the exposure poitns (Figure 4, b), which may be due to the increase in

the degree of redeposition, causing smoother surface and lower concentration of defects, which adversely impact the optical properties of QDs. It should be noted that in case of $L = 2\,\mu$m the growth of PL intensity with the number of FIB passes is much less pronounced with the maximum at $N = 20$, which may also be explained by the effect of redeposition: at long distance between the points of FIB exposure its effect is minimized, which causes the absence of strong dependence of PL intensity on the number of FIB passes.

4. Conclusion

Therefore, the paper studied the processes of epitaxial growth of InAs QD using the MBE on the FIB-structured surface of GaAs(111)B, making it possible to form the ordered arrays of triangular triple QDs with low surface density. It is shown that at a low dose of ion radiation, a high spread of holes is observed by size, as well as low selectivity of triple QDs, whereas as it increases, the holes of homogeneous size are produced, which provide for the same distance between the QDs in a triple complex. Higher voltage of FIB makes it possible to improve the selectivity and yield of triple QDs in the holes, and to decrease their size. Maximum selectivity of triple QDs is achieved at the voltage of 30 kV, the distance between the holes of $1\,\mu$m and at 30 FIB passes and reaches 0.9. At the distance of $2\,\mu$m, large holes are formed on the surface in the form of inverted truncated pyramids, in the tops of the base of which single QDs nucleate, with their solid chains — on the bottom. PL spectra of samples with QDs demonstrate radiation in the range of 900–1150 nm with the maximum intensity at 40 FIB passes at the distance of $1\,\mu$m between the holes.

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Conflict of interest

The authors state that they have no conflict of interest.

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