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Geometry of InSb quantum dots grown on a surface of the In(As,Sb) matrix layer

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Arrays of InSb quantum dots were obtained on matrix layers of solid solutions in the InAs–InSb system by liquid-phase epitaxy. The effect of an additional element introduced into the crystal lattice of the matrix layer on the surface density, geometric parameters and structural properties of the grown nanoheterostructures was shown. The presence of a common element in the $InAs_{1-y}Sb_y$ matrix layer and InSb nanoislands made it possible to increase the surface density of quantum dots to $1.8 \cdot 10^{10}\,cm^{-2}$. A transformation of the shape of nanoislands (from a convex lens to a truncated pyramid) was observed depending on the matrix layer in the composition range $0 < y < 0.09$. A model of the shape of self-organizing InSb quantum dots deposited on the surface of a ternary solid solution in the InAs–InSb system was proposed.

Keywords: Quantum dots, solid solutions, InAs, InSb, atomic force microscopy, nanoheterostructures.

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1. Introduction

Arrays of InSb quantum dots (QD) in the InSb/InAs system can be produced by different technological methods [1–3]. A quite simple and inexpensive method of liquid-phase epitaxy (LPE) was taken as an example to demonstrate that it was possible to produce the arrays of the InSb quantum dots on InAs substrates with a surface density of up to $2 \cdot 10^{10}\,cm^{-2}$ [4]. These quantum dots were grown directly on a surface of the single-crystal InAs substrate with a working surface oriented in the plane (001) in the temperature range $T = 420–450\,^{\circ}C$. A bimodal nature of distribution of the InSb quantum dots on the surface of the InAs binary compound was detected, which was manifested by presence of large-size nanoobjects (10 nm in height and 56 nm in diameter) with the surface density that is smaller by an order of magnitude, along with small quantum dots (3 nm in height and 26 nm in diameter) [5]. According to results of the studies performed by means of a transmission electron microscope, the small quantum dots were almost homogeneous in composition without an arsenic impurity, whereas a moire period for the large quantum dots could be related to the penetration of the As atoms into the In(As, Sb) quantum dot (up to the concentration of about 24 %) and a respective change of the lattice parameter of the nanoobject. At the same time, an assumed form of the produced nanoobjects was close to a hemi-sphere form with an oval base due to light asymmetry depending on growth along the directions of the type [110] or $[1\bar{1}0]$ on the lattice plane (001).

It is known that variation of a molar composition of the multi-component solid solution makes it possible to control a width of a band gap and an energy diagram of the epitaxial layer produced on its base [6]. Moreover, using the multi-component solid solution as a matrix layer enables predefining parameters of a lattice constant of the matrix and initial mismatch between contacting semiconductors and, therefore, controlling a strain value in nanoheterostructures that contain the quantum dots. It was shown in the study [7] that with variation of the composition of the InAsSbP matrix layer, on which the quantum dots were directly deposited from the InSb alloy, due to incorporation of additional elements (for example, phosphorus and antimony), a size distribution of the deposited nanoobjects was noticeably changed. The present study is aimed at a possibility of showing the influence of the additional element to be incorporated into the crystal lattice of the matrix on the surface density, geometrical parameters and structural properties of the quantum dots grown on it.

2. Experiment and techniques of investigating the nanoobjects in the InSb/InAs system

The heterostructures with the InSb quantum dots were grown by using a liquid-phase epitaxy set-up based on a diffusion furnace SDO-125/4. The arrays of the quantum dots were epitaxially grown in a horizontal quartz reactor using a graphite cassette of the „case“ type in a hydrogen flux at the fixed temperature $T_{gr} = 425\,^{\circ}C$. The arrays of the InSb quantum dots were produced in a single technological process with growing of the matrix layer at the system's cooling rate $\upsilon_{gr} = 0.3\,^{\circ}C/min$. The matrix layers were

epitaxial layers of the solid solutions in the In−As−Sb system, which were grown on the InAs(001) single-crystal substrates.

The surface relief was studied using the atomic force microscope (AFM) Solver P47H and the probe laboratory „Integra Aura“ that are manufactured by the Russian company NT-MDT. The measurements were performed in a semi-contact mode using silicon consoles (cantilevers) NSG01 with a typical resonance frequency of about 150 kHz and typical rigidity of 5 N/m. The amplitude of free resonance oscillations of the very tip of the cantilever, where the AFM is fixed, was 60 nm. The semi-contact mode is sometimes referred to as a discontinuous-contact mode, since the probe contacts the sample surface for a small part of the oscillation period. When studying the samples with the InSb quantum dots that are separately arranged on the open InAs surface, the amplitude of forced oscillations of the cantilever was selected to be approximately in two times less than the amplitude of the free oscillations. The AFM studies were carried out under atmospheric conditions, wherein an average force of interaction of the AFM probe with the sample surface turned out to be still quite small at a level of about 0.5 nN, which in turn did not result in strain both of the relief of the quantum dots and of the pointed tip of the probe itself.

The experimental data obtained from the AFM images of the surfaces of the arrays of the unclosed InSb quantum dots were analyzed by means of the SPM Image Magic statistical-processing software that is specially designed for automatically determining coordinates, a height and a diameter of near-spherical isolated objects. The most reliable determined parameter of the quantum dots is their height. The automatically determined diameter of the object can be slightly distorted due to a hardware effect of convolution with the shape of the pointed tip of the probe. At our disposal we had the probe laboratory „Integra Aura“ equipped with feedback sensors for all the three coordinates, therefore, the coordinates of the quantum dots in a plane were also determined with a high degree of reliability, whose measurement accuracy affects another important parameter — an average surface density of the objects. A Z-scanner was calibrated by means of a certified test grid TGZ01 with a step height of 18.5 nm.

3. Arrays of the InSb quantum dots on the surface of the epitaxial layers in the In−As−Sb system of the solid solutions

The arrays of the coherent quantum dots were produced during epitaxial deposition of the InSb binary compound to the surface of the multi-component matrix layers based on the $InAs_{1-y}Sb_y$ solid solutions within a composition range $0 < y < 0.09$ (Figure 1). We have detected the bimodal nature of distribution of the produced InSb quantum dots both when growing to the surface of the InAs binary

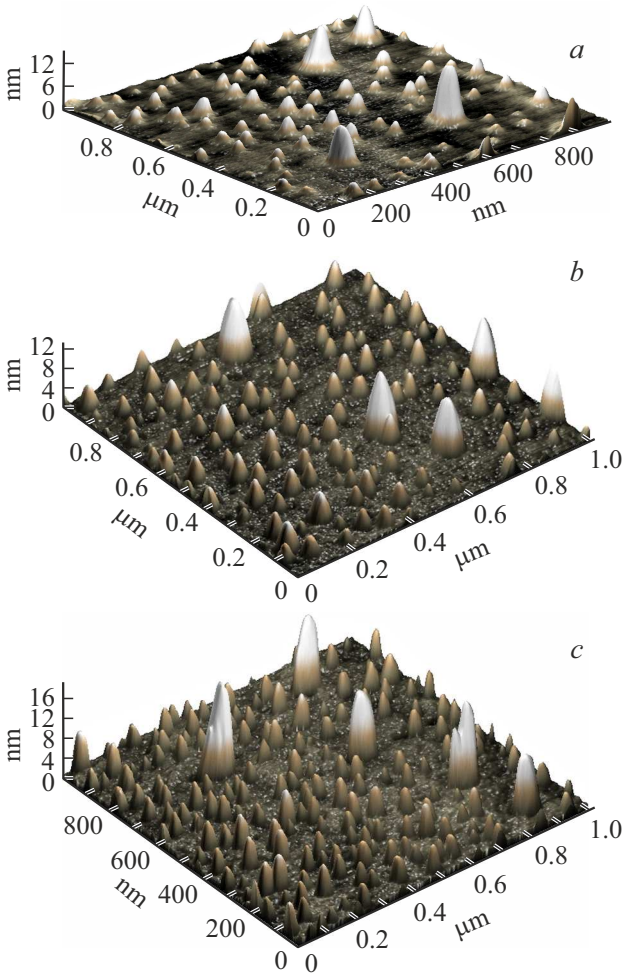


Figure 1. 3D-topographies of the arrays of the quantum dots on the surface of the epitaxial layer InAs (a), $InAs_{0.94}Sb_{0.06}$ (b) and $InAs_{0.91}Sb_{0.09}$ (c).

compound as well as to the surface of the epitaxial layer of the $InAs_{1-y}Sb_y$ ternary solid solution. As can be seen from the figure, the concentration of the large quantum dots was almost unchanged (the surface density was preliminary estimated to be about $6 \cdot 10^8\,cm^{-2}$), whereas the total surface density of the InSb quantum dots produced on the matrix layers of the InAsSb ternary solid solution increased.

Statistical histograms (Figure 2) obtained by processing the AFM images show that the main part of the arrays of the quantum dots is made up by coherent elastically-stressed nanoislands [5]. When the statistical histograms of the studied arrays of the quantum dots were analyzed, we found an increase of the average height (h) and a simultaneous decrease of the average lateral size (D) of the small nanoislands when the antimony content in the matrix layer increased (see Table). At the same time, we observed a linear dependence of the geometrical parameters of the quantum dots on the composition of the $InAs_{1-y}Sb_y$ solid solution (Figure 3, a). Depending on the content of the antimony atoms in the epitaxial layer of the $InAs_{1-y}Sb_y$

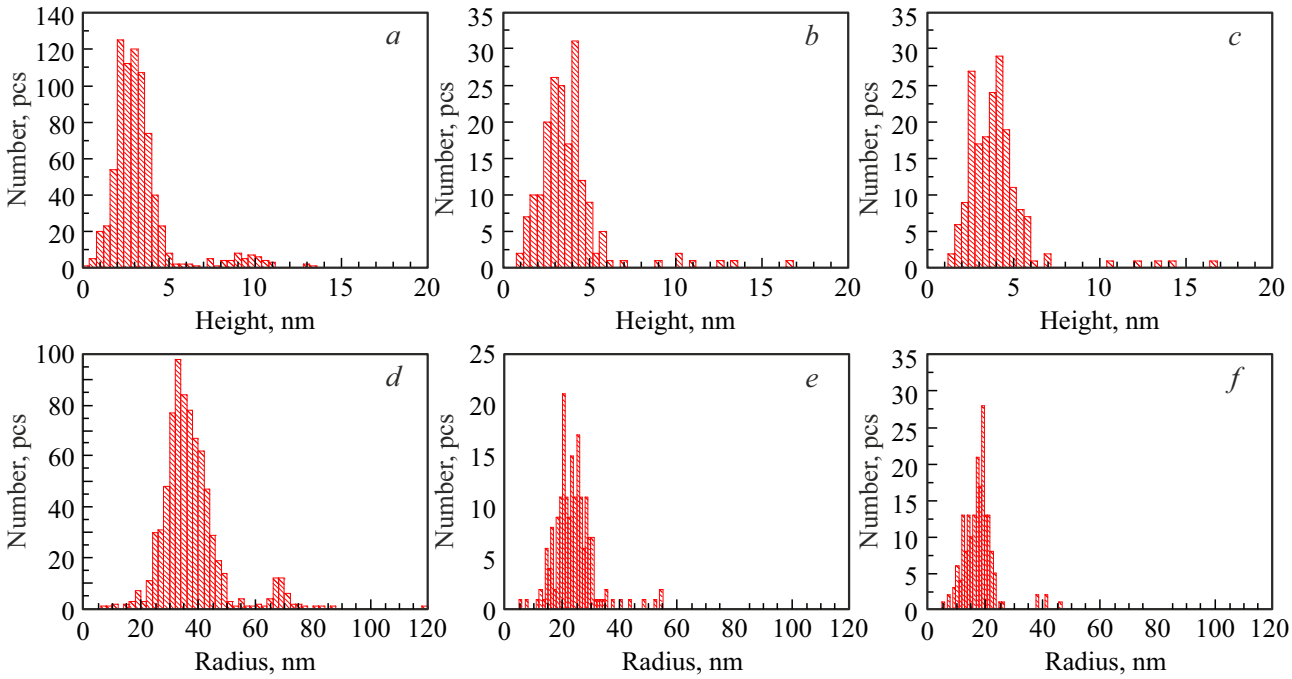


Figure 2. Statistical data of distribution along the height and the lateral parameter for the InSb quantum dots produced on the surface of the InAs binary compound (a, d) and the three-component solid solutions $InAs_{0.94}Sb_{0.06}$ (b, e) and $InAs_{0.91}Sb_{0.09}$ (c, f).

Surface density and the geometrical parameters of the quantum dots produced on the matrix layers based on the $InAs_{1-y}Sb_y$ solid solutions

Sb content in the matrix layer, y	Surface density of the QDs, 10^{10} cm^{-2}	Height of the small QDs, nm	Lateral size of the small QDs, nm	Actual parameter, h/D	Mismatch of the solid solution relative to the InAs substrate
0	0.8	2.5	34	0.07	0
0.06	1.25	3.2	22	0.145	$+4.2 \cdot 10^3$
0.09	1.8	4.0	18	0.22	$+6.3 \cdot 10^3$

ternary solid solution we observed transformation of the shape of the deposited InSb quantum dots: a flat lens-like drop with the actual parameter $h/D = 0.07$ at $y = 0$ was transformed into a relief dome-like nanoisland with $h/D = 0.22$ at $y = 0.09$. As a result we obtain the increase of the actual parameter (h/D), which can affect an energy spectrum of the eigenstates of the quantum dot itself.

Consequently, the shape of the InSb quantum dots produced on the epitaxial layers of the InAsSb ternary solid solution directly depended on the concentration of the mutual (both for the matrix and the quantum dot) element of the V group in the crystal lattice of the matrix layer. We assume that the chemical composition of the surface of the matrix layer affects adhesion of contacting compounds. Presence of the mutual element (Sb) in the matrix layer and the quantum dots can contribute to better interaction between the nanoobject and the matrix due to mutual chemical bonds, which in turn can result not only to a change of the nanoobject shape, but to closer packing of the produced arrays of the quantum dots as well.

Indeed, the surface density of the small quantum dots sharply increased with the increase of the antimony portion in the matrix layer. The observed increase is quite well described by a nonlinear dependence (Figure 3, *b*), which can be presented as a power function

$$F(y) = n_0 + (Ay)^3,$$

where n_0 — the value of the surface density of the quantum dots during deposition to the binary surface $(y = 0)$, A — the coefficient that corresponds to the composition of the solid solution of the matrix layer. It is obvious that the antimony atoms in the layer of the solid solution can act as additional centers of formation of the nanoobjects. Thus, incorporation of the chemical element that is also present in the quantum dots into the matrix layer activates the additional channel of formation of potential crystallization centers, which is manifested in the exponential function of the surface density of the coherent nanoislands on the antimony content in the ternary solid solution, i.e. the quite

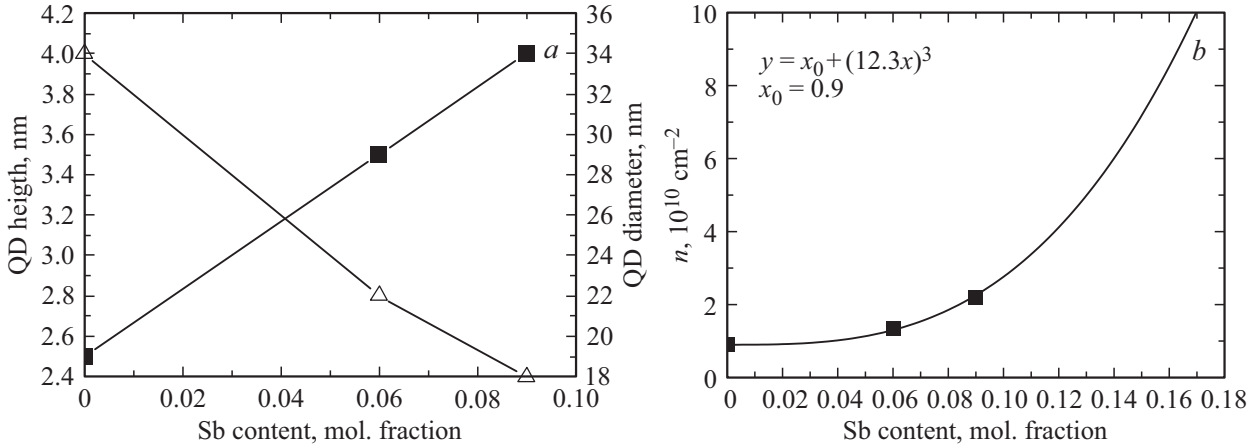


Figure 3. Dependences of the structural characteristics of the deposited arrays of the InSb quantum dots on the composition of the InAsSb matrix layer: *a* — the parameters of the nanoobject, *b* — the surface density of the quantum dots.

small increase of antimony results in the significant increase of the concentration of the grown quantum dots.

However, further increase of the antimony content in the grown matrix layer can be limited by stoichiometry of the ternary solid solution. The matter is that incorporation of the additional element of the V group (Sb) as a replacement atom for As into the crystal lattice of the binary compound (InAs) results in the increase of the lattice parameter of the epitaxial layer of the ternary solid solution ($InAs_{1-y}Sb_y$) relative to the indium arsenide binary substrate [8]. This in turn results in disruption of a morphology of the deposited layer and appearance of structural defects on its surface (longitudinal dislocations, etc.), thereby indicating a transition into a deposition phase of the relaxed compound.

One should also take into account the influence of the chemical composition of the surface of the matrix layer on a configuration of the quantum dots. It is known that the ordered arrays of the three-dimensional coherently-stressed nanoislands are formed due to an effect of self-organization as a result of relaxation of a stress that arises due to a difference in values of the parameters of the crystal lattice of the semiconductor being grown and the semiconductor matrix, to which deposition is done [9]. As said above, presence of the additional element of the V group (Sb) in the crystal lattice of the ternary solid solution ($InAs_{1-y}Sb_y$) results in the increase of the lattice parameter of the epitaxial layer relative to the binary substrate (InAs). In turn, the mismatch of the matrix layer relative to the InSb binary compound decreases. Consequently, it is expected that the internal stress is reduced in the InAsSb/InSb epitaxial heterostructure as compared to the structure based on the binary compounds InAs and InSb. Then it is possible to increase the height of the quantum dots by growing additional indium antimonide monolayers due to an increase of a limit of a critical thickness of the stressed layer.

In addition to the change of the average characteristic parameters of the quantum dots produced by us, there was variation of the configuration of the deposited nanoobjects. Figure 4 shows 3D-images of the nanoislands, which are representative for the quantum dots that are considered in the present study. The indium antimonide quantum dots produced on the indium arsenide binary matrix layers were shaped as a lens with an ellipsoid-like base. It was shown in the study [5] that the coherently-stressed nanoobjects retained an smooth upper surface without any abrupt transitions and singularities. It should be also noted that the shape of the nanoislands remained the same irrespective of a method of preparation of the matrix surface: chemically treated or epitaxially grown (see Figure 4, *a* and 4, *b*). It is believed that an external surface of the deposited epitaxial layer has better planarity as compared to the working surface of the substrate, which is prepared by means of wet chemical etching [10]. Nevertheless, the values of the surface densities of the arrays of the quantum dots for both the surfaces were also the same. Thus, it can be assumed that it is the chemical composition of the matrix surface that has a decisive effect on the shape and the density of the deposited quantum dots.

During the experiment, we changed the chemical composition of the matrix surface by incorporating the additional chemical element into the crystal structure of the InAs binary compound. As a result, when the InSb quantum dots were deposited to the surface of the $InAs_{1-y}Sb_y$ ternary solid solution, we observed appearance of facetedly-shaped nanoislands. In our case, degradation of the morphology of the matrix layer in mismatch with the InAs substrate resulted in formation of additional centers of singularity, which was manifested in transformation of the shape of the nanoislands. With small amounts of antimony in the epitaxial layer of the ternary solid solution, the external surface of the quantum dots exhibit many small facets (Figure 4, *c*). A polygon formed by facets is originated in the base of the nanoobject and these facets are parallel to lattice planes. With further increase of the antimony content, the number of the facets decreases (Figure 4, *d*), wherein the facets that are parallel to certain lattice planes are more

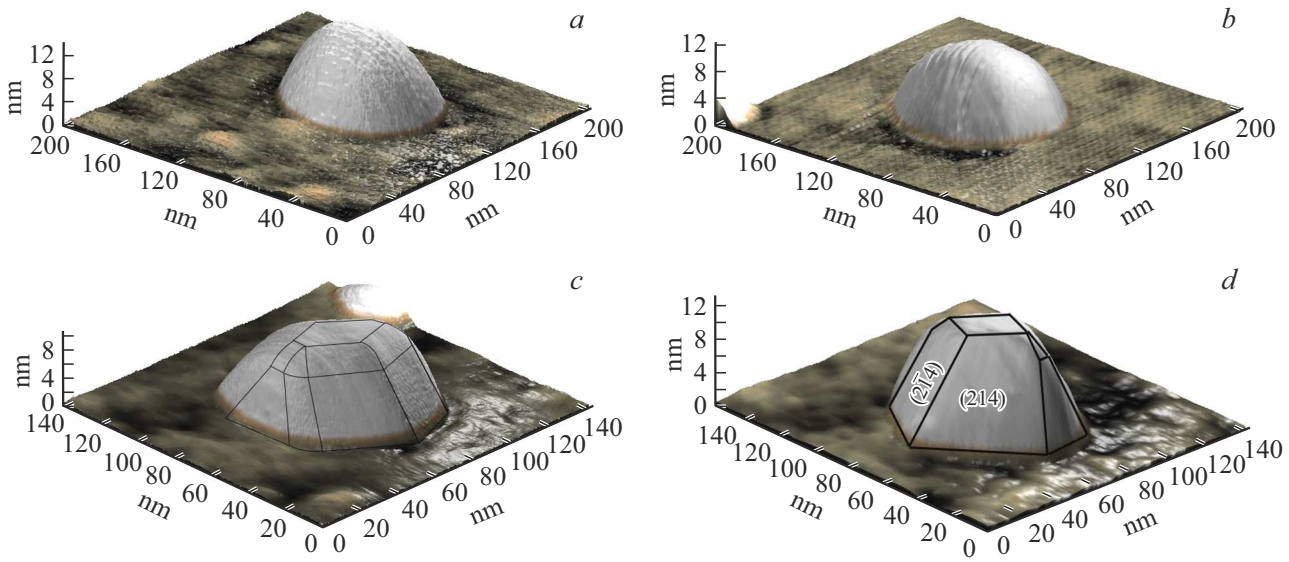


Figure 4. InSb single quantum dot of the surface of the InAs binary compound (a, b) and the InAsSb three-component compound (c, d).

pronounced. The other facets are resolved not very well and do not exhibit clear edges. The base of the nanoobject is transformed into an octagon or even a hexagon and the produced quantum dots can be presented as a truncated pyramid.

For the $InAs_{0.91}Sb_{0.09}$/InSb heterostructure, a flat vertex of the quantum dot can be described as a rhomb-like plateau with the angles 120° and 60°, respectively. Values of the angles were experimentally obtained from horizontal 2D projection of the AFM image of the nanoobject. As can be seen in Figure 4, *d*, two well-defined facets that determine the shape of the quantum dot can be discerned at the right and left side of the image. An angle of inclination between these facets and the matrix surface (001) was obtained from a profile of the section perpendicular to these facets and was about $29 \pm 2°$. It should be underlined that an interval of the allowable angles was obtained by averaging of measurements of several quantum dots. We believe that the quantum dots with sides limited by the lattice planes of the family (214) can be grown on the matrix surface of the $InAs_{0.91}Sb_{0.09}$ ternary solid solution. Miller indices for designation of the produced facets were determined by taking into account angles of the rhomb-like plateau in the base of the quantum dot and angles of inclination of facets that limit the sides of the quantum dot. We note that determination of the Miller indices of these facets is a quite difficult task, since an error of the measurements is still too high and the declared values can be considered as estimates. Based on the obtained experimental data, we propose a model of the shape of the self-organized InSb quantum dots that are deposited to the surface of the ternary solid solution in the InAs–InSb system, which are limited by at least four facets that are oriented parallel to the planes $\{nm4\}$, where $n = 2; -2$ and $m = 1; -1$. We believe that there is a certain critical size of the quantum dot, which is determined by a size of the facets {214}. The facets {214} can be stable up to a certain size, when an increment of the free energy due to relaxation of strain inside the three-dimensional island does not exceed the surface energy of the nanoobject [11]. Consequently, the resultant surface energies can be responsible for sharp size distribution of the self-organized InAs quantum dots and, therefore, shall be considered in further theoretical studies.

4. Conclusion

Thus, it was found that the bimodal nature of the InSb quantum dots on the surface of the matrix layer produced based on the compounds in the InAs–InSb system did not depend on the antimony content in the ternary solid solution. It was shown that the change of the composition of the matrix layer by incorporating the chemical element that is common both for the crystal lattice of the matrix and that of the quantum dot, made it possilbe to increase not only the surface density of the deposited nanoobjects, but their actual parameter as well. The presence of the common element in the matrix layer and the quantum dot facilitates multiplication of the mutual chemical bonds, which in turn helps to improve interaction between the nanoobject and the matrix layer. However, degradation of the morphology of the matrix layer in mismatch relative to the substrate due the presence of the additional chemical element in the crystal lattice, results in the change of the internal stress inside the nanoheterostructure, thereby causing transformation of the shape of the quantum dot. The obtained data confirm the fact that the composition of the matrix surface determines the shape of the deposited nanoobject. The experimentally-observed transformation of the shape of the nanoislands from the convex lens to the truncated pyramid shall be taken into account when

calculating the energy spectrum of the eigenstates of the InSb quantum dots in the $InAs_{1-y}Sb_y$/InSb heterostructure

Conflict of interest

The authors declare that they have no conflict of interest.

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