Effect of morphology features of patterned surface on the nucleation processes of In/GaAs nanostructures during droplet epitaxy

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Abstract. We present the results of theoretical studies of the self-organization processes of nanoscale metal In droplets on GaAs(001) substrates with artificial structural heterogeneities of various types – with a rectangular, trapezoidal, and triangular shapes. The study showed that to improve the accuracy of nanostructure positioning and homogeneity, it is necessary to use patterns with triangular grooves. In order to ensure the full groove filling by the material and to suppress the undesirable structure formation outside modified areas, it is necessary to provide sufficient diffusion length of adatoms taking into account the peculiarities of the patterned surface morphology.

1. Introduction
Modern semiconductor technology is inexorably approaching the threshold of miniaturization and, consequently, an increase in its efficiency. This necessitates a transition to fundamentally new computing and telecommunication platform architectures. From this point of view, the creation of solid-state schemes for the implementation of such platforms based on the use of A3B5 nanoheterostructures with self-organizing structures, the creation of which requires the implementation of effective self-organization processes, looks most promising [1-17]. In turn, the droplet epitaxy technology has a high flexibility in the formation of self-organizing nanostructures [18-24]. Studying the physics and mechanisms of the processes underlying droplet epitaxy allows significant progress in the implementation of the technology for controlling the epitaxial synthesis and self-organization of A3B5 nanostructures, which ensures effective control of the properties of the structures obtained and their positioning. This opens up broad prospects in improving the efficiency of photodetectors and photovoltaic converters based on A3B5 heterostructures due to surface plasmon effects [25-27], creating sources of single [28, 29] and entangled photons [30], functional blocks of cellular automata [31] and quantum computing [32], etc. One of the promising approaches to ensure the management of processes of self-organization, including with droplet epitaxy, is the use of structured substrates [33, 34].

The purpose of this work is theoretical studies of the self-organization processes of nanoscale In droplets on the GaAs growth surfaces with complex morphology, taking into account the main governing parameters of the droplet epitaxy technique. In this case, we used the previously developed
hybrid mathematical model of droplet epitaxy processes, a feature of which is the use of a combination of calculations using the Monte Carlo kinetic method and analytical calculations according to the thermodynamic theory of nucleation [35, 36]. For the study of growth processes, we used GaAs(001) structured substrates with different shapes, sizes and densities of specified inhomogeneities.

2. Results and discussion

The calculation results showed that on substrates with complex morphology, the probability of island nucleation is distributed non-uniformly and depends on the surface area. On GaAs substrates with rectangular holes, nucleation can occur on flat areas, bottom or walls of the hole or on the edges of the faces. We have shown that on the As-stabilized GaAs substrate with rectangular holes, adatoms linger mainly on (001) faces, which are parallel to the planes substrate terraces, while the walls of the holes remain unfilled due to enhanced diffusion from faces (110). Due to the increase in the amount of material around the walls of the holes due to diffusion from the (110) faces, as well as an increase in the binding energy of the adatom with the substrate in the corner of the modified region, nucleation can also occur in the vicinity of the intersection of the hole bottom with its walls (Figure 1). Therefore, to achieve the best homogeneity of the geometric characteristics of nanostructures, the diameter of the rectangular holes should be reduced, adjusting the height so as to achieve the required effective volume and aspect ratio.

Figure 1. The morphology of In/GaAs(001) nanostructures array after deposition of 3 ML In on an As-stabilized substrate with rectangular holes at \( T = 150 \, ^\circ\text{C} \) and \( v = 0.1 \, \text{ML/s} \). The length of the simulated area is 100 nm, the diameter and height of the hole are 20 and 10 nm, respectively, the distance between the hole centers is 50 nm.

The intermediate shape between the holes of a rectangular and triangular shape are trapezoidal ones. On substrates with this modification, the positions for the most likely nucleation will be determined based on the combination of technological regimes and diffusion parameters, which have a higher intensity on the (111) faces than on the (001) ones. The study showed that with a large ratio of trapezoid bases, the In droplets forms approximately in the center of the hole, whereas with a small ratio and relatively large sizes of hole, the preferred position for nucleation shifts towards one of the corners as is the case with the straight shape (Figure 2).

Figure 2. The morphology of In/GaAs(001) nanostructures array after deposition of 4 ML In on an As-stabilized substrate with trapezoid holes with bases of 20 and 13 nm at \( T = 150 \, ^\circ\text{C} \) and \( v = 0.1 \, \text{ML/s} \). The length of the simulated area is 100 nm, the height of the recess is 5 nm, the distance between the hole centers is 50 nm.

An analysis of the results of theoretical studies has shown that the use of a shape close to a triangular one is distinguished by more pronounced positions for nucleation, which makes it possible to increase the uniformity in size and accuracy of positioning of nanostructures. On the such surface, it is possible to achieve full or partial stabilization of gallium or arsenic (Figure 3). As can be seen from the Figure 3, in all cases, the preferred position for the island nucleation is the hole bottom due to the
increase in the number of substrate atoms surrounding the adatom under consideration, and, consequently, the increase in its binding energy with the current environment.

**Figure 3.** The morphology of In/GaAs(001) nanostructures array after deposition of 2 ML In on the substrate with triangular holes with Ga- (left) and As-stabilization (right) at $T = 150 \degree C$ and $v = 0.1 \text{ ML/s}$ The length of the simulated area is 50 nm, the diameter and height of the recess are 5 and 5 nm, respectively, the distance between the centers of the recesses is 25 nm.

This phenomenon extends to a triangular holes with any angle between the walls. At the same time, as the angle between the walls increases, the probability of removing the adatom from the most preferable position increases, which can lead to nucleation on the wall or outside the structural inhomogeneity (Figure 4).

**Figure 4.** The morphology of In/GaAs(001) nanostructures array after deposition of 3 ML In on an As-stabilized substrate with triangular holes at $T = 150 \degree C$ and $v = 0.1 \text{ ML/s}$. The length of the simulated area is 100 nm, the diameter and height of the recess are 10 and 5 nm, respectively, the distance between the centers of the recesses is 50 nm.

In order to ensure 100% filling of the holes and suppress the formation of undesirable structures outside the modified areas, it is necessary to ensure sufficient diffusion length of adatoms taking into account morphological features. The probability of island nucleation outside the hole increases with decreasing temperature and increasing growth rate, which is associated with a decrease in the probability of adatoms to take the most energetically advantageous positions as a result of a decrease in the intensity of the diffusion surface and as a result of a decrease in the average diffusion length of adatoms, respectively. At temperature $T = 150 \degree C$ and growth rate $v = 0.1 \text{ ML/s}$, the distance between the triangular shape holes with a diameter and depth of 20 nm, at which 100% ordering of the nanostructure array is reached (in the calculation of the ratio of the number of complexes "1 nanostructure in 1 hole" to the number of holes) is 50 nm. With an increase in the distance between the holes, nucleation is observed outside the predetermined centers. But increasing the temperature to 250 $\degree C$ and above allows to eliminate unwanted nucleation at distances between the holes exceeding 100 nm.

**Figure 5.** The morphology of In/GaAs(001) nanostructures array after deposition of 5 ML In on an As-stabilized substrate with triangular holes with a diameter and depth of 20 nm at $T = 400 \degree C$ and $v = 0.1 \text{ ML/s}$. The length of the simulated area is 200 nm, the distance between the hole centers is 100 nm.

Thus, the use of modified surfaces allows to adjust the density of nanostructures, providing the possibility of obtaining, including laterally coupled nanostructures. The transition to the incomplete
condensation mode can further reduce the size of the structures while maintaining their density. So, an increase in temperature from 400 to 500 °C leads to a decrease in the size of nanostructures from 19 to 16 nm, all other things being equal (Figure 5).

Analysis and systematization of the obtained theoretical results showed the ability to control the size of nanostructures while maintaining the specified array configuration by reducing the thickness of the deposition material. Compared with a decrease in droplet size on a flat surface (limited due to the presence of a non-zero critical thickness of droplet formation), a significantly smaller nanostructure can be achieved on a modified substrate, which is associated with a decrease in the nucleation activation barrier and an increased probability of droplet nucleation in the region of structural inhomogeneities and, therefore, a decrease in the deposition thickness at which nucleation begins. Thereby, due to this effect, a reduction in the wetting layer thickness is achieved, which has a significant impact on the electronic and optical characteristics of the devices, as well as a reduction in the size of nanostructures.

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