Analytical–Monte Carlo model of the growth of In nanostructures during droplet epitaxy on the triangle-patterned GaAs substrates

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Abstract. Kinetic Monte Carlo simulations combined with the analytical nucleation theory are used to study droplet epitaxy of indium on the GaAs substrates patterned with triangular-shaped nanoholes. The simulation results demonstrate that the growth on the Ga-stabilized surface provides much better selectivity than the formation of nanostructures on the As-stabilized surface because of the stronger bonding with As atoms of the substrate. We estimate technological parameters which allow obtaining required characteristics of nanostructures with precise positioning and complete filling of holes. The distance between nanostructures is considered to be controlled by the growth temperature at an appropriate interhole distance. At the same time, dimensions of nanostructures are outlined by the geometrical parameters of holes and can be adjusted by the deposition thickness.

1. Introduction
Control over the interaction between active elements of nanodevices is an outstanding problem in quantum science and engineering. Great prospects for use in novel devices are associated with nanostructures based on GaAs and related materials [1-8]. Semiconductor quantum dots fabricated by droplet epitaxy are particularly attractive for the realization of quantum computers and photonic integrated circuits, as they can be controllably positioned and varied in geometrical characteristics. During the Stranski-Krastanov growth on patterned GaAs substrates, InAs quantum dots can nucleate outside the holes or form a multiple quantum dot in a single hole [9,10]. At the same time, the droplet epitaxy technique allows formation of a single quantum dot with controllable parameters within each hole [11,12]. However, a detailed investigation of the nucleation and growth processes of nanostructures during droplet epitaxy on patterned substrates is still necessary. To fabricate quantum dot arrays with required geometrical parameters in given positions, appropriate technological parameters should be chosen. Since it is inexpedient to carry out a large number of experiments, it is appropriate to use simulations.

In this work, kinetic Monte Carlo model based on the analytical nucleation theory is used to describe the processes of droplet epitaxy of indium on the nanopatterned GaAs substrates. Previously, we validated the model using the experimental data in a wide range of growth temperatures. The studies demonstrated nonmonotonic dynamics of the adatom supersaturation and island critical size as well as formation of the wetting layer with thickness depending on the substrate temperature [13].
Productivity of the analytical approach and versatility of the Monte Carlo method make it possible to consider complicated phenomena during nucleation and growth of islands [14-19].

2. Description of the model
The model is based on the kinetic Monte Carlo simulations which use the results of analytical calculations within the thermodynamic theory of nucleation. The key points of the model agree with those for the flat substrate which were used previously to describe the processes of nucleation and growth during In/GaAs(001) droplet epitaxy [13]. In this study, we develop the model to discuss the growth mechanisms on patterned substrates. Critical regions are formed throughout the simulation area in such a way that the area of the region above the substrate remains the same. Thus, nucleation may occur in any point and the nucleation probability is determined on microscopic level. Lennard–Jones potential is used to calculate the binding energy between atoms of the crystal. A type and an order of events are defined by waiting times which are taken from calculations by the Arrhenius equation.

3. Results and discussion
The results of our simulations on patterned substrates demonstrated a difference between growth processes on the Ga and As-stabilized surfaces. Due to the fact that the bonds of metal atoms (In and Ga) with each other is weaker than those with As atoms, the mobility of adatoms on the Ga-stabilized surface is higher [17]. At the same time, adatoms on the As-stabilized surface stick to the substrate much stronger because they are bound directly to the As atoms of the substrate. As a result, the wetting layer on the As-stabilized surface consists in several monolayers (ML) (Fig. 1b) whereas it is about 1 ML on the Ga-stabilized surface at the growth temperature $T = 100^\circ$C (Fig. 1a). Our previous study demonstrated that the formation of the wetting layer on the Ga-stabilized surface may be even prevented completely when $T = 150^\circ$C [17].

This behavior gives an opportunity of better control of the growth characteristics with the technological parameters. The As-stabilized surface can be used when the wetting layer is required for a device structure. However, we consider a situation when the formation of the wetting layer should be suppressed. As Fig. 1 shows, the Ga-stabilized surface is appropriate for this condition. In this case, the deposited material is located in the holes rather than on the flat areas of the substrate (Fig. 1a).

![Figure 1. Morphology of the array of In/GaAs(001) nanostructures after deposition of 5 ML of indium on the a) Ga-stabilized and b) As-stabilized nanopatterned substrate with $d = 10$ nm, $h = 6$ nm, $r = 20$ nm at $T = 100^\circ$C (length of the simulation area $L = 100$ nm).](image)

The diffusion length of In adatoms on the GaAs substrate is quite large as compared with the diffusion length of Ga adatoms [20-21]. Consequently, a sufficient distance between holes should be provided to prevent nucleation and growth of nanostructures outside the holes. The results of our simulations show that the deposited material can be collected only within the triangular holes (Fig. 2a, 3a, 3b). To ensure the best selectivity of the growth, an appropriate combination of the parameters of
patterning and the growth conditions should be selected. When the substrate temperature \( T = 100 \, ^\circ\text{C} \), the interhole distance \( r = 75 \, \text{nm} \) is sufficient to localize In nanostructures within the holes (Fig. 2a). The deposition of indium with thickness \( H = 5 \, \text{ML} \) allows partial filling of the holes. In certain cases, the deposited material exceeds the borders of the hole.

![Figure 2](image1)

**Figure 2.** Morphology of the array of In/GaAs(001) nanostructures after deposition of 5 ML of indium on the Ga-stabilized nanopatterned substrate with \( d = 16 \, \text{nm}, h = 12 \, \text{nm} \) and a) \( r = 50 \, \text{nm} \), b) \( r = 100 \, \text{nm} \) at \( T = 100\, ^\circ\text{C} \) (\( L = 200 \, \text{nm} \)).

As the interhole distance increases up to 150 nm, the nucleation and growth of droplets on the flat part of the substrate is probable (Fig. 2b). Although the diffusion length of In adatoms on the GaAs surface is large, the growth at a low temperatures \( T = 100\, ^\circ\text{C} \) may result in undesirable formation of nanostructures outside the prepared nucleation centers if they are located wide apart. Fig. 3a shows that the growth temperature should be increased up to 200\(^\circ\text{C}\) to achieve better selectivity on the substrate with the same parameters of patterning. In this case, the deposited material is distributed between the holes whereas it is consumed by droplets formed on the flat surface at a lower temperature (Fig. 2b).

![Figure 3](image2)

**Figure 3.** Morphology of the array of In/GaAs(001) nanostructures after deposition of a) 5 ML, b) 4 ML of indium on the Ga-stabilized nanopatterned substrate with \( d = 16 \, \text{nm}, h = 12 \, \text{nm}, r = 100 \, \text{nm} \) at \( T = 200\, ^\circ\text{C} \) (\( L = 200 \, \text{nm} \)).

One of the most advantages of the droplet epitaxy method is a possibility to control the size of nanostructures independently of their surface density [22-26]. Growth on patterned substrates makes it possible to control positions of nanostructures as well as their geometrical characteristics. As Fig. 3b demonstrates, preventing of exceeding the level of the substrate by nanostructures can be achieved along with complete filling of holes. A decrease of the deposition thickness from 5 to 4 ML at \( T = 200\, ^\circ\text{C} \) leads to a decrease in the height of nanostructures relative to the hole bottom from 17 nm to the hole depth \( h \) which is equal to 12 nm. It should be noted that the maximum diameter of a nanostructure remains the same and equal to the hole diameter \( d = 16 \, \text{nm} \) as well as the surface density of nanostructures does not change.
4. Conclusion
Thus, the precise selective growth of nanostructures can be realized during droplet epitaxy of In on triangle-patterned GaAs substrates. However, appropriate technological parameters at the stages of both substrate preparation and indium deposition must be chosen to accomplish the growth with required characteristics. The distance between nanostructures can be controlled by the growth temperature on the substrate patterned with appropriate interhole distance. The deposition thickness allows controlling the degree of filling of holes and dimensions of nanostructures within the holes of appropriate depth and diameter.

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