Kinetic Monte Carlo simulation of the growth of In nanostructures by droplet epitaxy on AlGaAs nanopatterned surfaces

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Abstract. In this paper, the results of the simulation of the In/AlGaAs growth on nanopatterned surfaces using modified analytical–Monte Carlo model are presented. The surface density of nanostructures is demonstrated to slightly decrease with increasing Al content. A decrease of an interhole distance leads to the occupation of a small part of a hole which can be prevented by a decrease of the hole volume. The best localization of In nanostructures on AlGaAs surfaces nanopatterned with holes at a distance of more than 125 nm can be achieved at a temperature of 300°C or higher. A decrease in temperature requires a sufficient decrease of an interhole distance to avoid nucleation beyond predefined positions.

1. Introduction
Unique atom-like properties of semiconductor quantum dots attract particular attention due to their potential application in promising optoelectronic and nanophotonic devices [1-3]. Currently, the most common technique for A3B5 quantum dot fabrication involves strain-driven self-assembly by molecular beam epitaxy known as the Stranski-Krastanov growth mode [4-7]. However, this growth mode involves strain-driven self-assembly and has the stochastic nature which results in random quantum dot nucleation and non-uniform distribution in their size and shape. Furthermore, the formation of a residual wetting layer leads to electronic coupling amongst neighboring quantum dots which presents an obstacle for many applications [8-11].

In order to fabricate quantum dot arrays with required characteristics in given positions, appropriate techniques and technological parameters should be chosen. Better control of quantum dot shape and size can be enabled by selective quantum dot placement as surface kinetics can then be designed to produce a desired quantum dot characteristic. The selectivity can be achieved by nanoscale patterning of the substrate by different methods [12-17]. A new method of droplet epitaxy enables formation of nanostructures with independent control of their size, surface density and chemical composition as well as possibility to exclude the wetting layer formation [18-22].

In this paper, kinetic Monte Carlo-based simulation of the growth of In nanostructures on AlGaAs nanopatterned surfaces by droplet epitaxy is presented. Versatility of the Monte Carlo approach combined with the productivity of analytical calculations makes it possible to consider complex processes during nucleation and growth of islands on patterned surfaces with different chemical composition [23-29].
2. Description of the model
Previously developed hybrid analytical–Monte Carlo model [30, 31] was upgraded to take into account the growth on patterned surfaces with different Al content. The model considering In/GaAs growth initially allows incorporation of atoms of other sorts, such as Al atoms in the substrate or Ga atoms as deposited species, and modification of the substrate profile. In this study, we include into consideration epitaxial surfaces with various Al content and complex substrate morphology, such as triangular and rectangular patterning.

3. Results and discussion
Using the developed and optimized mathematical model, we carried out theoretical study of the processes of the self-assembly of nanoscale In metallic droplets on complex AlGaAs growth surfaces under different technological conditions.

In order to establish regularities in the formation of nanostructures on surfaces with complex morphology, we carried out simulations of the growth on GaAs, Al$_{0.5}$Ga$_{0.5}$As and AlAs surfaces at various substrate temperatures in a range from 150 to 300°C and various deposition thicknesses in a range from 1.0 to 3.0 monolayers (ML). We found that the difference in the geometrical parameters of nanostructures have a slight dependence on the presence of Al in the surface layer. In this connection, the results of the study are mainly shown for the Al$_{0.5}$Ga$_{0.5}$As surface.

In a low-temperature range ($T=150^\circ$C) at an insufficiently small distance between holes ($r\geq125$ nm) nucleation on a flat part of the surface is possible. Undesirable formation of nanostructures on the intervals between substrate modifications is caused by the appearance of additional nucleation centers (Figure 1a).

![Figure 1](image-url)

**Figure 1.** Morphology of nanostructure arrays after deposition of 5 ML of In at a temperature $T=150^\circ$C on different epitaxial surfaces: a) Al$_{0.5}$Ga$_{0.5}$As, $T=150^\circ$C, b) AlAs, $T=150^\circ$C, c) Al$_{0.5}$Ga$_{0.5}$As, $T=300^\circ$C, d) Al$_{0.5}$Ga$_{0.5}$As, $T=200^\circ$C. The width of the simulation area is 250 nm.

In some cases, nucleation outside the holes is suppressed, which is more likely on the AlAs surface (Figure 1b). The study also demonstrated that the best localization of nanostructures on the surfaces of Al$_{0.5}$Ga$_{0.5}$As and AlAs is achieved by increasing the temperature up to 300°C (Figure 1c), which is due to an increase in the intensity of surface diffusion of adatoms. The formation of nanostructures with maximum selectivity is also observed on the Al$_{0.5}$Ga$_{0.5}$As surface with square holes with a diameter of 14 nm, approximately equal in volume to triangular holes with a diameter of 20 nm, at a temperature of 200°C (Figure 1d).

A decrease of the distance between modified areas from 125 nm to 50 nm leads to a uniform distribution of In deposited material between holes (Figure 2a). However, in this case droplets occupy a small part of a hole. In order to achieve the best hole filling, a hole volume should be decreased or
deposition amount increased. A decrease of the hole diameter from 20 nm to 10 nm allows reaching better coverage of the hole under given technological conditions (Figure 2b).

![Figure 2. Morphology of nanostructure arrays after deposition of 5 ML of In on the epitaxial surface of Al$_{0.5}$Ga$_{0.5}$As at a temperature $T = 200^\circ$C: a) $d = 20$ nm, b) $d = 10$ nm. The width of the simulation area is 250 nm.](image)

Thus, the best localization of nanostructures is achieved at a high temperature ($T = 300^\circ$C) or with decreasing interhole distances. Perfect localization of In droplets was also experimentally observed after deposition on nanopatterned GaAs surface at the same temperature of 300°C [21].

According to the well-known regularities [32-34], an increase of the substrate temperature leads to an increase of an average diameter of In nanostructures for each of the surfaces: GaAs, Al$_{0.5}$Ga$_{0.5}$As and AlAs (Figure 3). The diameter-temperature curves are not monotone increasing and have dips at certain temperature values. This irregularity is caused by the fluctuation of the shape of nanostructures which may lead to a change both in the height and diameter of nanostructures.

![Figure 3. Temperature dependences of the average diameter of In nanostructures after deposition of 5 ML of In on triangle-patterned Al$_x$Ga$_{1-x}$As surfaces with holes having a diameter of 20 nm and an interhole distance of 100 nm.](image)

As one can see in Figure 3, an average diameter of In nanostructures formed on triangle-nanopatterned surfaces increases with rising temperature and then reaches saturation. This is attributed to the fact that a temperature increase leads to the enhancement of the compliance of nanostructure and hole parameters, namely an increase of percentage of nanostructures formed in certain positions relative to their total quantity. This property is technologically valuable for precise site-controlled growth.

4. Conclusions

Thus, an increase in the Al content in the epitaxial surface leads to a decrease in the surface density of In nanostructures, as well as to an increase in their average size, which should be taken into account when switching to the droplet epitaxial growth on modified surfaces. Reducing the distance between the holes allows the growth of high-density arrays of nanostructures at higher temperatures. In this
case, an increase in temperature during growth on the surface with any Al content leads to the saturation of the average diameter of nanostructures.

The best localization of In nanostructures on AlGaAs surfaces nanopatterned with holes at a distance of more than 125 nm can be achieved at a temperature of 300°C or higher. A decrease in temperature requires a sufficient decrease of an interhole distance to avoid nucleation beyond predefined positions.

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