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Droplet epitaxy of site-controlled In/GaAs(001) nanostructures with a variable distance: experiments and simulations

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Abstract. This paper presents a complex experimental and theoretical study of the droplet epitaxial growth of In/GaAs(001) nanostructures on patterned surfaces. We observe that holes formed after GaAs overgrowth of surfaces treated with a focused ion beam are the preferred centers for the nucleation of In droplets at any temperature in a range from 250 °C to 350 °C. Good selectivity and localization of droplets are achieved along a square perimeter of holes located at a distance from 0.5 to 4.2 μm apart. However, lower temperatures are required to provide filling of more holes and formation of an ordered array of droplet pairs. Using kinetic Monte Carlo simulations, we demonstrate growth conditions which allow filling of all holes located at variable distances in a range from 20 to 340 nm and avoiding unnecessary nucleation beyond the holes.

Keywords: droplet epitaxy, focused ion beams, patterned surfaces, Monte Carlo method

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Материалы конференции
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Капельная эпитаксия селективно-позиционированных наноструктур In/GaAs(001) с переменным дистанцированием: эксперимент и моделирование

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Аннотация. В работе представлено комплексное экспериментальное и теоретическое исследование процессов роста наноструктур In/GaAs(001) методом капельной эпитаксии на структурированных поверхностях. Обнаружено, что углубления, формируемые после заращивания поверхностей, обработанных фокусированным ионным пучком, слоем GaAs, являются предпочтительными центрами зарождения капель In при любой температуре в диапазоне от 250 °C до 350 °C. Достигнута высокая степень селективности и локализации капель в углублениях, расположенных вдоль периметра квадратного массива на расстоянии от 0.5 до 4.2 мкм. Однако для достижения большей степени заполнения углублений и формирования упорядоченных массивов пар капель требуются

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пониженные температуры. С помощью моделирования кинетическим методом Монте-Карло продемонстрированы технологические режимы, при которых достигается заполнение всех углублений, расположенных на переменном расстоянии друг от друга в диапазоне от 20 до 340 нм, с подавлением нежелательной нуклеации за пределами предзаданных центров.

Ключевые слова: капельная эпитаксия, фокусированные ионные пучки, структурированные поверхности, метод Монте-Карло

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Introduction

High optical quality and emission wavelength range of InAs/GaAs quantum dots (QDs) make them very attractive for use in advanced optoelectronic and quantum photonic devices [1–3]. QD properties are largely determined by their geometrical characteristics, such as size, shape and surface density. One more important parameter of a QD array which enables prediction of QD position is their spatial arrangement. Prediction of QD sites facilitates post-growth treatment of a QD-based heterostructure and improves a yield of devices based on single QDs, such as quantum light emitters.

In order to obtain regular arrays of InAs/GaAs QDs with a high level of selectivity (implied as a percentage of QDs located in required positions), various methods were applied to pattern the substrate, such as local anodic oxidation [4], focused ion beams (FIB) [5], nanoimprint [6], electron-beam [7], etc. However, site-controlled formation of QDs located at a variable distance from each other is also significant. For instance, lasers operating on whispering gallery modes require a circular arrangement of QDs with suppression of nucleation within the inner cavity region [8]. QDs in quantum information devices must be located at variable positions to control interaction between QD spins [9]. So-called QD molecules are also in high demand during the last years both in fundamental [10] and applied aspects as an active element in nanoelectronic and quantum computation systems [11]. Nevertheless, QD formation in strictly desired positions is difficult because of the stochastic nature of their epitaxial growth. Technological conditions may lead to the formation of either nucleation beyond predefined sites, or absence of nucleation within these sites because of a large adatom diffusion length. Thus, specific growth regimes are necessary to achieve a unity selectivity and 100% localization of QDs defined here as a percentage of holes filled with QDs.

In this paper, we present experimental and theoretical studies of the droplet epitaxial growth of In nanostructures on FIB-patterned surfaces with a variable distance between nanoholes. A method of droplet epitaxy allows formation of In droplets with a specified surface density and a size that can be altered depending on the amount of deposited material and arsenic flux parameters [11–13]. Then, In droplets can be transformed into InAs QDs with a procedure of high-pressure arsenization.

Materials and Methods

Epi-ready GaAs(001) substrates patterned by FIB in Nova Nanolab 600 scanning electron microscope (SEM) with Ga^+ ion source were used in the experimental studies. Two types of FIB arrays with area 5×5 μm^2 were formed with a distance of 0.5 and 1.0 μm between point of FIB treatment, an accelerating voltage of 30 kV and various ion doses: from 1 to 300 ion beam passes

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where 1 beam pass corresponds to an ion dose of 17.4 fC/µm^2. Then, the samples were transferred into SemiTEq STE35 molecular beam epitaxy equipment to carry out droplet epitaxy. After a standard procedure of the oxide removal, 15 nm of GaAs was grown as a buffer layer at a substrate temperature of 500 °C and 45 nm of GaAs at 580 °C.

At the next stage, the arsenic flux was blocked in order to reduce the background pressure in the growth chamber to ~ $1\cdot10^7$ Pa. Next, In atoms were deposited on the substrate surface at a nominal rate of 0.05 and 0.25 monolayers (ML) per second in an amount of 3 equivalent ML. The substrate temperature was varied from 250 °C to 350 °C during the deposition process. After the deposition had been finished, the samples were cooled and placed in the SEM chamber to measure geometric parameters.

Theoretical studies of the formation of In/GaAs(001) nanostructures by droplet epitaxy on patterned surfaces were carried out using a previously developed mathematical model based on a combination of analytical expressions of classical nucleation theory and kinetic Monte Carlo method [12, 13]. The model has a 1+1 dimensionality that simulates the material system under consideration in a section. The universal principles of atom interaction specified in the model allow taking into account the structural inhomogeneities of the surface of different shapes and sizes. In this paper, triangular-shaped holes with (111) faces, similar to the experimentally observed pyramidal holes, were specified as modification sites.

Results and Discussion

At the first stage, the parameters of arrays of In/GaAs(001) nanostructures formed at different substrate temperatures on a flat surface were analyzed. It was found that increasing the substrate temperature from 250 °C to 350 °C leads to a decrease in the surface density of droplets, estimated as a ratio of their number in the SEM image to the scan area, from $3.1\cdot10^8$ to $2.4\cdot10^7$ cm^{-2}, which corresponds to the average distance between droplets, 0.57, 1.26 and 2.04 µm, respectively. SEM images of the samples obtained after overgrowth of the FIB-modified surface with a GaAs buffer layer and subsequent growth by droplet epitaxy are shown in Fig. 1.

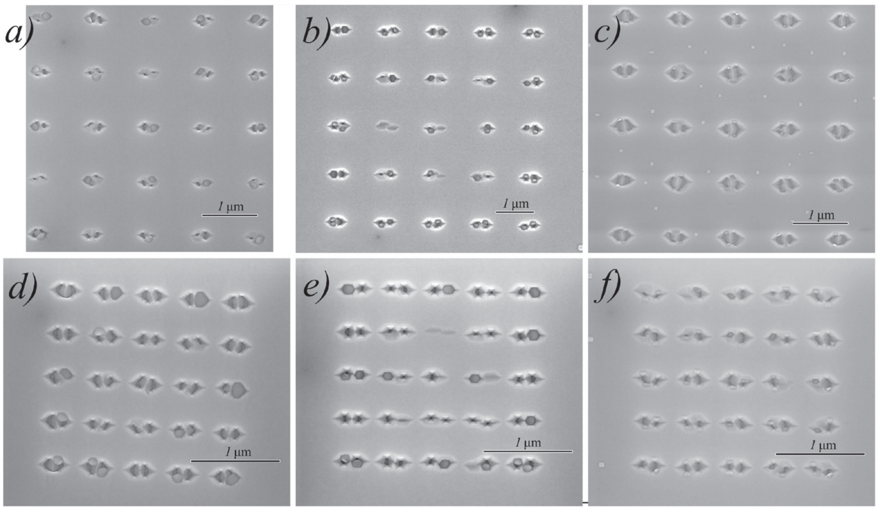

Fig. 1. SEM images of droplet arrays obtained on FIB-patterned surfaces with a distance between FIB treatment points 1 µm (*a* – *c*) and 0.5 µm (*d* – *f*) at various deposition temperatures: 350 °C (*a*, *d*), 300 °C (*b*, *e*) and 250 °C (*c*, *f*)

It follows from the presented images that the obtained holes are the preferred centers of nucleation and subsequent growth of In droplets in them, as evidenced by the absence of nanostructures outside the areas of modification, except for the case of growth on the surface with holes located 1 µm apart at 250 °C (Fig. 1, *c*). However, it should be noted that the droplet material is not evenly distributed between all holes in cases where atoms have an increased diffusion length, which is characteristic of growth at elevated temperatures. This behavior is related to the presence of a critical size of a stable island, below which the probability of its decay increases significantly.

When droplets were formed at 300 °C on the surface with holes located 0.5 µm apart, site-controlled droplet formation was found in almost every hole along the array perimeter with no nucleation in the inner region (Fig. 2, *a*). This allows to conclude that it is possible to localize nanostructures in an array with a variable distance between nucleation centers in the range from 0.5 to 4.2 µm.

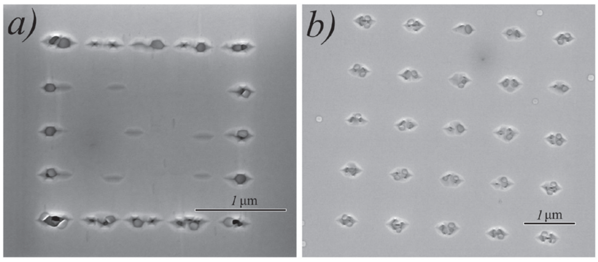

Fig. 2. SEM images of droplet arrays obtained on FIB-patterned surfaces with a distance between FIB treatment points 1 µm (*a*) and 0.5 µm (*b*) at various deposition rates: 0.05 ML/s (*a*) and 0.25 ML/s (*b*)

It was also found that it is possible to create arrays of In droplet pairs with close-to-unity filling of holes located at a distance of 1 µm (Fig. 2, *b*). This configuration, obtained at 300 °C with a deposition rate increased to 0.25 ML/s, also demonstrates an almost complete absence of undesirable nucleation beyond the modification sites.

Analysis of the theoretical studies showed that a decrease in the substrate temperature from 350 °C to 250 °C, as in the case of the experimental results, leads to a significant decrease in the diffusion length of In adatoms and a corresponding increase in the surface density of islands. Fig. 3, *a* demonstrates a typical model morphology of the system with In/GaAs droplets formed at 300 °C on a surface with pairs of 100 nm diameter holes spaced 500 nm apart. Fig. 3, *b* shows the morphology of the droplets obtained on a similar surface under the same growth conditions.

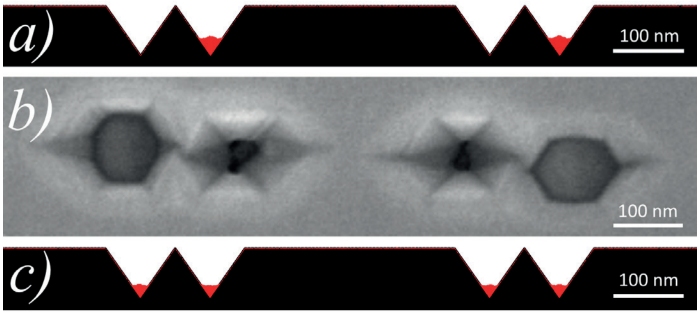

Fig. 3. Morphology of In droplet arrays obtained on patterned surfaces at 300 °C by means of the simulations (*a*) and SEM (*b*) and at 200 °C (simulations)

In the temperature range of 250 °C–350 °C, the computational experiments reveal holes in which there are no droplets, due to both the low probability of critical cluster formation and the high probability of their decay under the influence of high substrate temperature. However, lowering the temperature to 200°C, as seen in Fig. 3, *c*, leads to achieving 100% filling of the holes with the absence of droplets outside of them.

To evaluate a possibility of selective formation of droplets on structured surfaces with holes located at different distances from each other, we simulated In/GaAs droplet epitaxy processes on surfaces with holes of smaller size (20 nm). Fig. 4 shows the morphology of the droplet arrays formed on the surfaces with variable patterns.

At a temperature of 250 °C, the surface density of holes when simulated on a flat surface is $4\cdot10^{8}$ cm^{-2}, which corresponds to an average distance of 500 nm between the holes. Although this diffusion length of adatoms is sufficient to minimize nucleation outside the holes located at a comparable or smaller distance from each other, the temperature is too high to fill all holes, including those located close to each other, with droplets (Fig. 4, *a*).

A reduction in the deposition temperature to 200 °C makes it possible to provide 100% selectivity, expressed as the ratio of the number of droplets located in holes to the total number of droplets and holes divided by two (Fig. 4, *b*). A further decrease in the temperature to 150 °C leads to undesirable nucleation outside the holes, which is due to a decrease in the intensity of surface diffusion and an increase in the probability of nucleation (Fig. 4, *c*).

Fig. 4. Morphology of In droplet arrays obtained on patterned surfaces by means of the simulations at various temperatures: 250 °C (*a*), 200 °C (*b*) and 150 °C (*c*)

Conclusion

Thus, it was found that the holes formed after GaAs overgrowth of of FIB-patterned surfaces are the preferred centers of nucleation of In droplets at any temperature in the range from 250 °C to 350 °C. However, at higher temperatures, the degree of filling of the holes decreases, which is associated with an increased probability of decay of subcritical islands. By means of Monte-Carlo simulation, technological regimes are established at which it is possible to achieve 100% localization of In droplets on a patterned surface with holes located at a variable distance from each other in the range from 20 to 340 nm.

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