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# Study of InAs/GaAs quantum dots formation in subcritical growth modes on patterned substrates

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**Abstract.** In this work we present the results of experimental studies of the InAs/GaAs quantum dot formation in subcritical growth modes on nanopatterned substrates. For this purpose, we used two ways for surface patterning: local droplet etching and modified oxide desorption technique. We have experimentally shown that both methods allow in situ formation of nanosized pits (or nanoholes) on the surface, but their shape and density is quite different. We also have shown that the using of growing surface nanopatterning allows both to obtain self-assembled nanostructures (including QD) at subcritical deposition thicknesses and to localize its formation in nanoholes with high selectivity and suppressing a wetting layer formation. In addition, our results have also shown that the nanohole character on a structured surface (shape, size, density) has a key effect on both the processes of nanostructure nucleation and growth and their structural and optical properties, which should also be taken into account when developing methods for creating heterostructures with regular arrays of quantum dots.

**Keywords:** quantum dot, A3B5, wetting layer, nanopatterning, molecular beam epitaxy, nanostructure, self-organization

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# Исследование формирования квантовых точек InAs/GaAs в докритических режимах роста на структурированных подложках

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**Аннотация.** В данной работе представлены результаты экспериментальных исследований формирования квантовых точек InAs/GaAs при осаждении докритических толщин (для механизма Странского-Крастанова) на подложках с развитой (структурированной) поверхностью, полученной с использованием двух различных способов: локального капельного травления и модифицированного удаления собственного окисла. Показано, что оба способа позволяют in situ формировать на поверхности наноразмерные углубления, но с существенной разницей в их форме и плотности. Также показана возможность формирования самоорганизующихся наноструктур на структурированных поверхностях в докритических режимах с высокой степенью локализации и без сопутствующего образования смачивающего слоя. Кроме того, показано, что характер углублений на структурированной поверхности (форма, размер, плотность) оказывает ключевое влияние как на процессы нуклеации и роста наноструктур, так и на их оптические свойства, что также необходимо учитывать при разработке методик создания гетероструктур с регулярными массивами квантовых точек.

**Ключевые слова:** квантовые точки, A3B5, смачивающий слой, капельное травление, наноструктурирование, молекулярно-лучевая эпитаксия, наноструктуры, самоорганизация

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## Introduction

The unique properties of semiconductor quantum dots (QD) make them promising objects for creating micro- and nanoscale light sources on their basis, including non-classical light source, which act as active elements of integrated nanophononics and systems of quantum communications and quantum computing [1, 2]. In this regard, there is a need to develop methods for precise control of QD parameters, such as size, shape, chemical composition, etc. From this point of view, the most promising approach seems to be based on the pre-growth structuring (or nanopatterning) of the substrate surface, i.e. controlled formation of nanoholes on the surface, which later act as centers for self-organizing nanostructure nucleation. This approach, as well as its variations, is actively used to localize the formation and obtain ordered arrays of various types of self-organizing nanostructures such as nanowires [3], quantum dots [4], droplet nanostructures [5], etc. At the same time, today there are no systematic studies of the influence of created nanoholes morphological features on the structural and functional characteristics of selectivity grown nanostructures. The aim of our work is to study the influence of the nanopatterning techniques, as well as produced nanohole sizes and shapes, on the InAs/GaAs nanostructure formation in them, including their morphological and optical properties.

## Materials and Methods

For experimental study we used two ways to nanopattern GaAs substrates: local droplet etching (LDE) and so-called modified oxide desorption technique. Both methods allow in situ formation of nanosized pit (or nanoholes) on the surface, but their shape is quite different. As it can be seen from (Fig. 1, *a*) during LDE processing in the range of studying regimes, bowl-shaped nanoholes with a diameter of about 100 nm, a depth of several nanometers and a density of about $10^8$ cm$^{-2}$ are typically formed on the GaAs surface. In the case of patterning via modified oxide desorption technique, there is formed an array of faceted nanoholes with diameter up to 50 nm, at least 5 nm in depth (according to AFM data) and about $10^9$ cm$^{-2}$ in density on the surface (Fig. 1, *b*). After surface nanopatterning stage InAs layer with thickness varied from 0.1 to 1.5 monolayers (ML) was deposited using molecular beam epitaxy. Also, we repeated the same heterostructure with InAs layer (1.5 ML) on the atomically flat surface without nanopatterning which we used as a reference sample. For optical properties study by photoluminescence (PL) we repeated the same structures placed them in the central part of AlGaAs/GaAs/AlGaAs heterostructure.

All samples were grown by molecular beam epitaxy. PL studies were carried out at temperature 4 K. YLF:$Nd^{+3}$ laser operating in the cw mode ($\lambda$ = 527 nm) was used to excite PL in the analyzed samples. Excitation power density was about 1 kW/$cm^{-2}$. Each series of samples (with LDE and with modified oxide desorption), together with the reference one, was studied in a separate cycle under the same conditions.

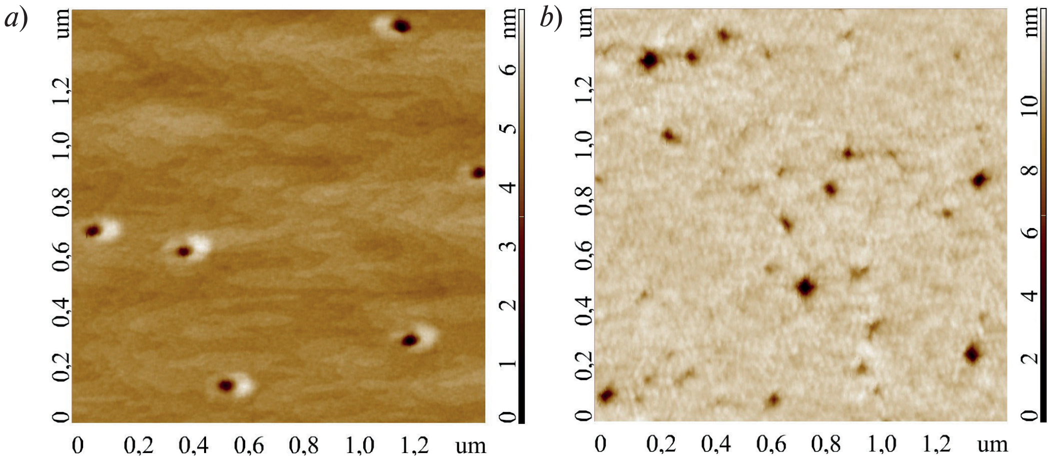


Fig. 1. AFM images of GaAs surfaces patterned by LDE (*a*) and modified oxide desorption (*b*) technique

## Results and Discussion

The results of experimental studies have shown that the shape and size of the nanoholes, depending on the method and mode of their preparation, has a key effect not only on the selectivity of nucleation processes, but also on the character of growing nanostructures, as well as their optical properties. As it can be seen from Fig. 2, *a*, in the case of LDE, oval nanostructures of small height (1–2 nm) are formed on the surface, the lateral size of which is much larger than the size of the initial nanoholes, 150–200 nm. In this case, a decrease in the InAs deposition thickness even to 0.1 ML leads to the decomposition of such structures into two substructures, each of which, nevertheless, remains rather large (about 50 nm in diameter). In the case of patterning via modified oxide desorption technique, pronounced QDs are formed on the surface, rising up to 3 nm above the surface level, the size of which is non-linearly correlated with the size of the original nanoholes (Fig. 2, *b*). At the same time, a large number of small QDs are also present on the surface. We attribute the formation of small QD to a high surface roughness due to the features of the oxide removal process.

An analysis of the PL spectra shows that the InAs nanostructures on the LDE-patterned substrates are optically inactive, which may indicate their defectiveness due to their too large sizes (Fig. 2, *c*). It is clearly seen that, at large deposition thicknesses, the spectra of such samples are like the spectrum of the reference sample with 1.5 ML of InAs on flat surface – in both cases, the spectra show an intense line of the wetting layer at a wavelength of 860 nm and a long-wavelength shoulder in the range of 870–890 nm. This shoulder is due to the contribution of the so-called platelets [6], which are fluctuations in wetting layer thickness and are a transitional form between quantum well (wetting layer) and quantum dots [6]. The main difference between the reference and LDE-patterned samples (Fig.2, *c*), black and blue spectra respectively) is the absence of quantum dot lines (above 900 nm) in the spectrum which may be due to the deposited material consumption by the InAs nanostructures formed in the nanoholes. A decrease in InAs deposition thickness leads to the disappearance of the long-wavelength shoulder, and then to a decrease in the WL intensity (840–860 nm). We attribute this to the fact that most of the deposited material accumulates in the nanostructures in nanoholes, as we said earlier, even at 0.1 ML. And wetting layer fragments can dissolve due to the indium segregation during capping by GaAs layer.

As it can be seen from Fig. 2, *d*, in the case of patterning via modified oxide desorption technique, the PL spectra are significantly different. At large deposition thicknesses (1.5 ML),

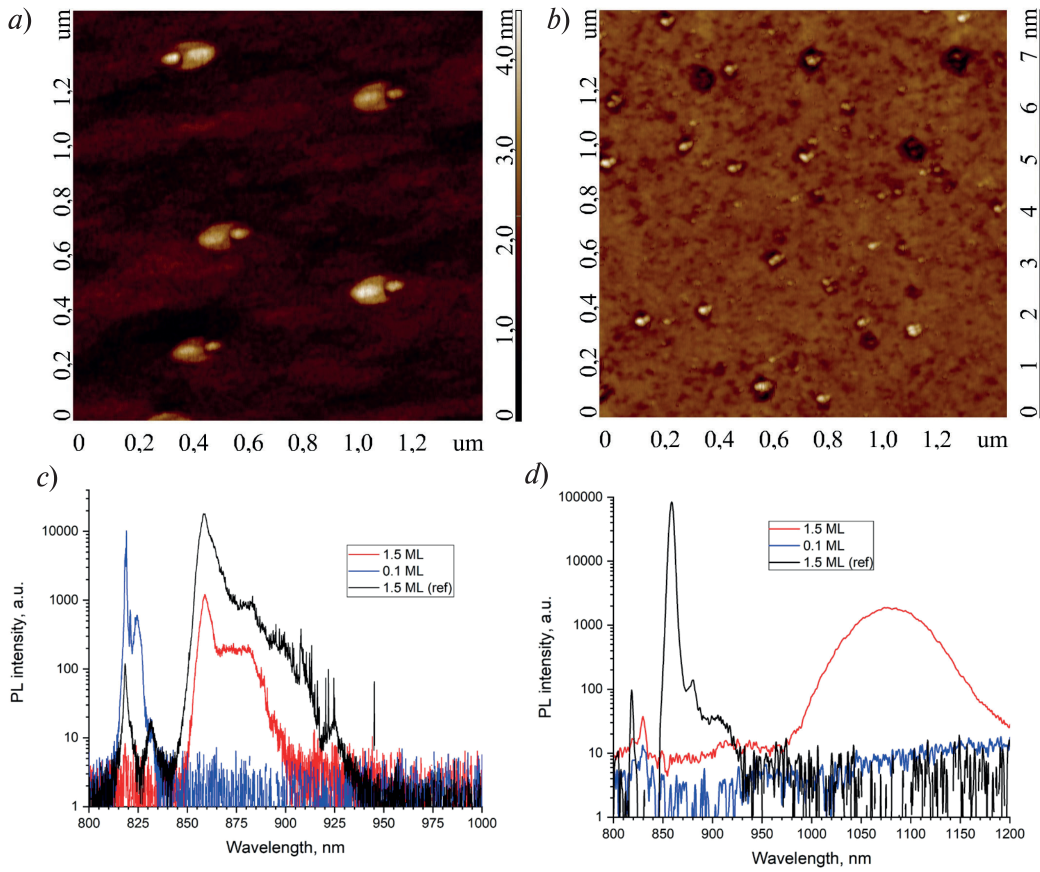


Fig. 2. AFM and PL characterization of InAs nanostructures on substrates patterned by LDE (*a*, *c*) and by modified oxide desorption technique (*b*, *d*)

there is a broad peak in the spectrum in the range of 1000–1200 nm, which is due to the contribution of inhomogeneous QDs. At the same time, the lines associated with wetting layer and platelets are completely absent. We assume that, due to the high surface roughness, all the deposited material accumulates in the nanoholes and forms InAs QDs. In this case, the formation of the wetting layer is completely suppressed. A decrease in the deposition thickness below 1.5 ML leads to the disappearance of all lines that could be associated with InAs structures. This is unexpected, because according to the AFM data, quantum dots are formed in nanoholes. This effect requires further study. However, we assume that the absence of QD lines in the PL spectrum after heterostructure formation can also be caused by their dissolution during the deposition of GaAs cover layer due to the strong segregation of In atoms and QD material intermixing with GaAs [7].

## Conclusion

Thus, in our work we have shown that the use of a nanostructured surface makes it possible to obtain quantum nanostructures in the InAs/GaAs system in subcritical growth modes and to suppress the wetting layer formation. We have also shown that the use of LDE modes for surface modification, which leads to the formation of bowl-shaped nanoholes, ensure a high selectivity of the nanostructure growth process, but does not lead to the QD formation. Instead, large InAs structures are formed, which probably contain defects and therefore do not radiate even at low temperatures. At the same time, the use of surfaces with faceted nanoholes makes it possible to form QDs in a wide range of conditions with complete suppression of wetting layer formation. However, our results show that the QDs observed on the surface in this case can disappear during GaAs overgrowing due to the effects of segregation and intermixing. Thus, it can be assumed that for the selective formation of quantum dots of optical quality on structured surfaces, it is important to use not only faceted nanoholes, but also to study the specifics of the formation of a cover layer and its interaction with QD in such systems.

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