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Influence of annealing conditions on the characteristics of nanoholes formed by focused ion beams on the GaAs(111) surface

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Abstract. In this paper, we study the effect of annealing of GaAs(111) substrates under various conditions on the morphological characteristics of nanoholes formed by focused ion beams. In the absence of annealing and when annealing in the absence of the arsenic flux, the depth and lateral size of nanoholes increase with the number of ion beam passes. In the case of annealing of the substrates in the arsenic flux, the dependences of the hole depth and lateral size on the number of beam passes is non-monotonic, which is attributed to the competition of the processes of surface etching by gallium droplets during thermal oxide removal and droplet crystallization in the arsenic flux. We demonstrate technological conditions enabling formation of highly symmetric nanoholes in the form of triangular pyramids.

Keywords: focused ion beams, annealing, local droplet etching, GaAs(111)

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Материалы конференции
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Влияние условий отжига на характеристики наноуглублений, формируемых фокусированными ионными пучками на поверхности GaAs(111)

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Аннотация. В работе представлены результаты исследования влияния отжига подложек GaAs(111) при различных условиях на морфологические характеристики наноуглублений, формируемых фокусированными ионными пучками. В отсутствие отжига и при отжиге в отсутствие потока мышьяка глубина и латеральный размер углублений возрастают с числом проходов ионного пучка. В случае отжига подложек в потоке мышьяка зависимости глубины и латерального размера углублений от числа проходов немонотонны, что связано с конкуренцией процессов травления поверхности каплями галлия во время термического сгона окисла и процессов кристаллизации капель в потоке мышьяка. Продемонстрированы технологические режимы, позволяющие формировать высокосимметричные наноуглубления в форме треугольных пирамид.

Ключевые слова: фокусированные ионные пучки, отжиг, локальное капельное травление, GaAs(111)

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Introduction

Emitters of single and entangled photons have recently attracted increased interest due to the possibility of their use in quantum information devices [1, 2]. A good candidate for these emitters is an epitaxially grown InAs quantum dot (QD) formed at a specified site of the GaAs surface [3]. QDs with C3v symmetry are particularly important and can be grown on surfaces with (111) orientation [4]. Because of their symmetry, such QDs demonstrate vanishing fine structure splitting, which favors a high degree of entanglement of photon pairs [5]. Despite the increased interest in this topic, the results of studies of the formation of nanoholes on the GaAs(111) surface – QD nucleation centers – by focused ion beams (FIB) followed by annealing in a molecular beam epitaxy (MBE) chamber to restore the disturbed crystalline regions have not been presented so far.

In this paper, the influence of FIB and MBE technological conditions on the characteristics of nanoholes formed on the GaAs(111) surface are investigated and formation of nanoholes with high C_{3v} symmetry is achieved. The results of the study of the dependence of the shape, depth and lateral size of nanoholes obtained directly after FIB treatment and after subsequent annealing under different conditions on the number of ion beam passes are also presented.

Materials and Methods

FIB treatment of the GaAs(111) surfaces was carried out on a FEI Nova NanoLab 600 scanning electron microscope (SEM) equipped with a FIB column (with Ga^+ ion source) at an accelerating voltage of 5 kV. FIB treatment point arrays of 4×4 μm in size with 2 μm spacing between the points and the number of ion beam passes (N) from 1 to 60 were used. In the next step, the FIB treated substrates were placed in a SemiTEq STE 35 MBE unit for annealing under different conditions to transform the nanoholes. Sample 1 was not annealed in the MBE chamber, samples 2 and 3 were annealed until the oxide removal at 600 °C without and with the arsenic flux, respectively, and then both samples were additionally annealed in the arsenic flux for 60 min.

The obtained samples were examined using a FEI Nova NanoLab 600 SEM and an NT-MDT Ntegra atomic force microscope (AFM) in a semi-contact mode. The average depth and lateral size of the nanoholes were estimated using the built-in tool of the Image Analysis software package designed for AFM image processing.

Thermodynamic analysis of chemical reactions in the Ga-As-O system was carried out in the FactSage 6.3 software package (using EquiSage, Reactions and PhaseSage modules). The package allows estimating the probability of chemical reactions in the considered system on the basis of the dependence of the Gibbs free energy change on the temperature.

Results and Discussion

AFM images of nanoholes formed by FIB without subsequent annealing demonstrate that FIB treatment leads to the formation of a shallow hole surrounded by a halo of swelled and redeposited substrate material (Fig.1, *a*, *b*). An increase in the number of FIB passes leads to deepening of nanoholes at their almost constant lateral size (diameter) and a decrease in the height of surrounding halos (Fig. 1, *c*).

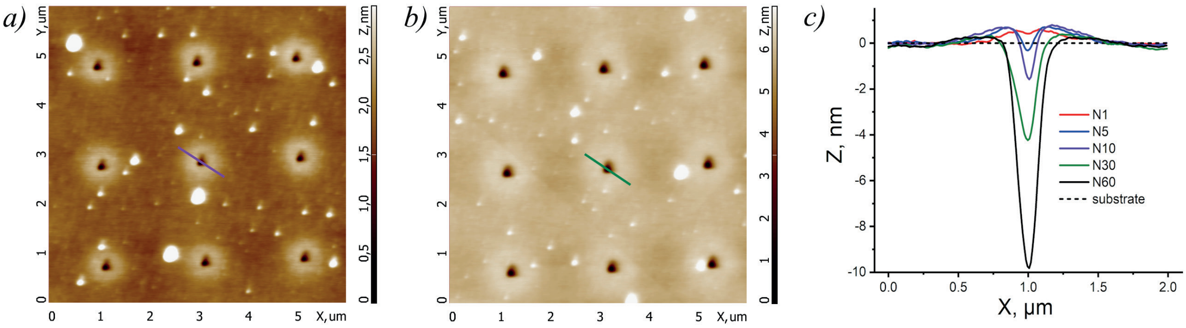

Fig. 1. AFM images of arrays of nanoholes formed after FIB treatment of the surface with various number of ion beam passes: 10 (*a*), 30 (*b*) and AFM cross sections of nanoholes located in the array centers (*c*)

An increase in the number of FIB passes leads to a monotonous increase both in the depth and diameter of nanoholes (Fig. 2, *a*). Annealing of the FIB-modified substrate results in the local droplet etching (LDE) of the GaAs(111) surface by Ga droplets [6, 7] and the formation of nanoholes with a shape of close-to-regular triangular pyramids (Fig. 3, *a*, *c*). The effect of LDE is particularly pronounced on the nanoholes obtained after 1 FIB pass and annealing under the arsenic flux: the average nanohole depth and lateral size, which are 0.2 and 70 nm immediately after the FIB treatment (Fig. 2, *a*), increase to 156 and 367 nm, respectively (Fig. 2, *b*). In the absence of arsenic flux, the surface is etched much less intensively (Fig. 3, *b*). However, an increase in the number of FIB passes to 10 and subsequent annealing without As results in the formation of an ordered array of uniformly sized nanoholes in the form of complete triangular pyramids (Fig. 3, *c*), as opposed to truncated ones in Figure 3, *a*. A further increase in the number of FIB passes up to 60 leads to an increase in the nanohole depth and reappearance of a flat base on its bottom in the case of As-free annealing and to the formation of complete triangular pyramids after the As-supplied annealing.

While both the depth and lateral size of nanoholes increases almost monotonically with the number of FIB passes for substrates with annealing in the As-free atmosphere (Fig. 2, *c*), the presence of arsenic leads to rises and dips in the dependences of hole size on the number of FIB

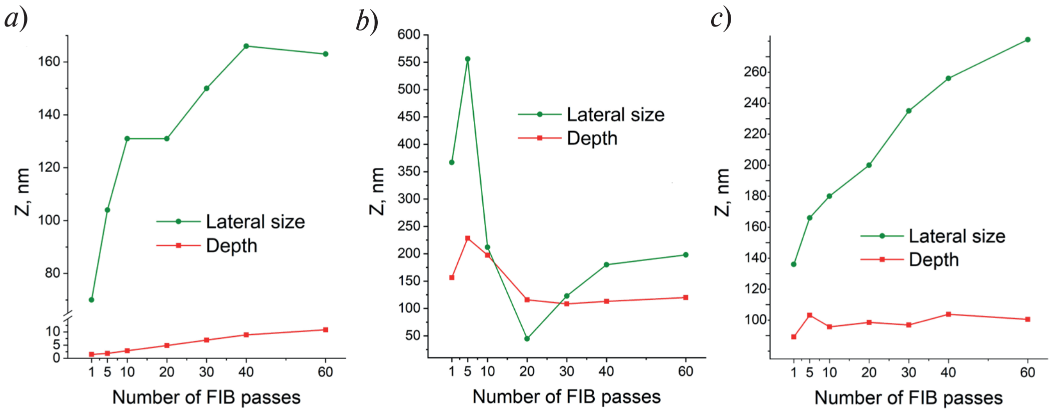

Fig. 2. Dependences of the lateral size and depth of nanoholes on the number of FIB passes in the FIB-treated areas before annealing (*a*), after annealing in the As flux (*b*) and without the As flux (*c*)

passes (Fig. 2, *b*). This behavior can be explained by the competition of the processes of surface etching by gallium droplets and droplet crystallization in the arsenic flux.

Although a standard procedure of the oxide removal from GaAs substrates is carried out in the absence of arsenic flux, the results of our studies demonstrate that its presence leads, first, to an increase in the size of nanoholes on the surface areas not treated by FIB and second, to an increase in the size and degree of symmetry of nanoholes at a small number of FIB passes (Fig. 3, *a*, *b*). When the number of passes is increased, the difference between the sizes of nanoholes obtained after annealing in the arsenic flux and in its absence becomes less noticeable.

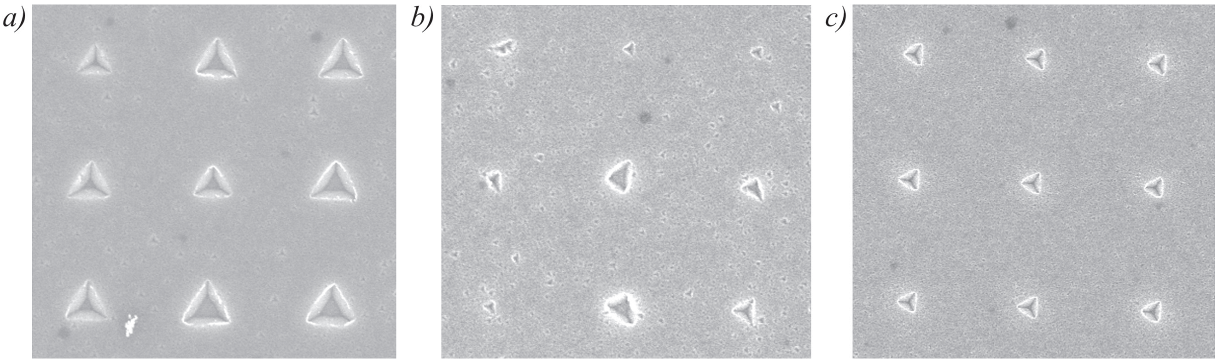

Fig. 3. SEM images of arrays of nanoholes formed after FIB treatment of the surface with various number of ion beam passes: 1 (*a*, *b*), 10 (*c*) – after annealing under different conditions: in the As flux (*a*), without the As flux (*b*, *c*)

Thermodynamic analysis of chemical reactions occurring in the Ga-As-O system (interaction of arsenic and gallium oxides with the underlying GaAs layer, as well as decomposition of GaAs and arsenic oxides) showed that gallium oxide Ga_2O_3 remains on the surface during heating longer than As_2O_3 and As_2O_5 arsenic oxides and serves as a kind of mask protecting gallium accumulating under it from crystallization in the arsenic flux. At the same time, arsenic stabilizing the GaAs surface serves as a drain for gallium atoms under the surface areas covered with stable Ga_2O_3-containing oxides. In this regard, an increase in the arsenic flux leads to enhancement of LDE, resulting in the formation of large nanoholes with pronounced crystal faceting. In turn, in the absence of the arsenic flux, gallium predominantly remains under the oxide and crystallizes after its removal, preventing etching of the substrate and the formation of pyramidal nanoholes.

Conclusion

Thus, the arsenic flux has a significant effect on the transformation of nanoholes formed by the FIB method, leading to an increase in their size at a small number of FIB passes and to its decrease at a large number of FIB passes. Highly symmetrical and uniformly sized nanoholes in the form of triangular pyramids were obtained after As-supplied annealing of the GaAs(111) substrate with arrays of FIB impact points formed after 60 ion beam passes and after 10 ion beam passes in the case of annealing in the As-free atmosphere.

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