Droplet epitaxy of In/AlGaAs nanostructures on the As-stabilized surface

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Abstract. The article presents the results of experimental studies of the regimes of formation of self-organizing In/AlGaAs nanostructures by the method of droplet epitaxy under As-stabilization conditions at different Al content in the surface layer. Dependences of the influence of the growth temperature, surface composition, and deposition thickness on the geometric characteristics of the In nanodroplet arrays such as density, size and dispersion. The possibility of controlling the parameters of nanostructures array by changing the Al content in the surface layer. An unusual dynamics of the change in the critical thickness of the formation of In nanodroplets is revealed with a change in the composition of the surface.

1. Introduction

The existing methods for the formation of A3B5 quantum dots by molecular beam epitaxy (MBE) are based on the Stransky-Krastanov mechanism – the elastic relaxation of mechanical stresses in lattice-mismatched systems, the growth of which is accomplished by the simultaneous deposition of components of III and V groups [1–3]. The disadvantages of this approach are the need for mismatch of crystal lattices, as well as the interdependence between the density and size of quantum dots formed by such a mechanism [1]. Moreover, the mismatch magnitude should lie within a certain range of values [1]. The combination of these factors dramatically narrows the range of combinations of materials and the ability to control the geometric characteristics of quantum dots [4, 5].

At the same time, droplet epitaxy [6, 7], based on the separate deposition of the components, lacks these drawbacks. Droplet epitaxy allows not only independently control the density and size of quantum dots [8, 9], but also to form quantum dots in any A3B5 systems [7, 10-13]. Moreover, the method is promising for the integration of low-dimensional A3B5 systems with silicon technology, as well as for the creation of different types of hybrid systems of great interest for optoelectronics and THz devices [14-20].

The use of multi-stage droplet epitaxy techniques [21-23] allows to effectively minimize the influence of negative factors associated with pre-surface surface treatment in the case of using modified substrates [24–29], which also favorably distinguishes this method from other A3B5 quantum dot synthesis techniques.

However, today there is no unambiguous representation not only of the microscopic processes and behavior of individual atoms with growth in the drip epitaxy regime in multicomponent systems, but there are also practically no studies on growth on surfaces of variable composition, which is important for controlling the parameters of the quantum dots formed.
In the present work, we carry out experimental studies of the formation of the self-assembled In nanostructures on the planar surfaces of AlGaAs epitaxial layers on the surfaces of epitaxial layers of variable composition.

2. Experiment
Experimental studies of the droplet epitaxy of In/AlGaAs nanostructures were carried out using the SemiTEq STE 35 MBE system. GaAs(001) wafers of the epi-ready class were used as substrates. After the procedure for thermal desorption of the oxide and smoothing of the GaAs surface in the As flux, a 400 nm thick GaAs buffer was grown at standard conditions [30, 31]: substrate temperature \( T = 580^\circ \text{C} \), effective growth rate \( V = 1 \text{ ML/s} \) and \( V/\text{III} \) flux ratio \( J = 4 \). Then, under the same conditions, an AlGaAs layer 10 nm thick was grown, the mole fraction of Al in which varied depending on the sample stepwise and amounted to 0.25, 0.5, and 1. Then all the sources were simultaneously closed, and the substrate temperature was lowered to the values at which the studies of self-organizing nanostructures formation were carried out. The cooling modes were selected in such a way as to ensure that the surface has the As-stabilized structure, observed during the formation of the functional layer by RHEED.

After stabilizing the temperature of the sample at a predetermined value, In deposition was carried out. The substrate temperature \( T \) in this case varied from 150 to 300°C in order to avoid desorption of adatoms and activation of exchange processes that made it difficult to interpret the experimental data. The growth rate varied in the range 0.25-0.5 ML/s. The deposition thickness of In \( (H) \) varied in the range 0.5-3 ML with step 0.25 ML. After completion of growth, the substrate was rapidly cooled in order to suppress undesirable material redistribution over the surface due to surface diffusion.

3. Results and discussion
The results of experimental studies of the In/AlGaAs system showed a significant difference from the results obtained for Ga/AlGaAs [32], which is due to much greater mobility and chemical activity of In adatoms on the surface. Because of the particular features of the system under consideration, the greatest interest is in the range of small effective thicknesses, which makes it possible in the future to ensure the formation of quantum dots with sizes providing acceptable structural perfection and energy characteristics. The high mobility of the In adatoms results in a substantial increase in the dimensions of the nanostructures in comparison with Ga. Thus, at an equivalent deposition thickness of 3 ML, the structure diameter was 3.67 nm for Ga and 20.21 nm for In at \( T = 150^\circ \text{C} \). Increasing the temperature to \( T = 300^\circ \text{C} \), the structure size increased to 6.7 nm for Ga and up to 98.41 nm for In. It should be especially noted that the dispersion of the sizes of the nanostructures In is much lower than for the Ga – on average, it decreases from 30-40% at \( T = 150^\circ \text{C} \) to 5-15% at \( T = 300^\circ \text{C} \), which is also due to higher values of the surface diffusion of adatoms [33–37].

Figures 1 and 2a show the dependence of the average size and density, respectively, for the two extreme cases – for In/GaAs and In/AlAs. As it follows from an analysis of the dependences obtained, an increase in the Al content in the epitaxial layer significantly changes the kinetics of the growth processes. At growth temperatures up to 200°C, an increase in the Al fraction leads to an increase in the density In droplets from \( 2.2 \cdot 10^7 \text{ cm}^{-2} \) to \( 2.1 \cdot 10^8 \text{ cm}^{-2} \) while approximately preserving their sizes about 20 nm. Increase in growth temperature to 300°C leads to an increase in the size of the structures with an increase in the Al mole fraction, while maintaining the difference in density by an order of magnitude. Thus, at deposition thickness of 3 ML the average In droplet diameter increases from 98 nm for GaAs surface to 177 nm for AlAs surface. And at the same time the droplet density decreases from \( 1.8 \cdot 10^8 \text{ cm}^{-2} \) до \( 3.7 \cdot 10^7 \text{ cm}^{-2} \), respectively.

In general, the range of variation in the density of droplet arrays also tends to expand as the Al content in the surface epitaxial layer increases.
Particular attention should be paid to the nature of the change in the critical thickness and, as a consequence, the thickness of the wetting layer with increasing Al content in the surface layer. In a case of GaAs surface (Al fraction is 0%) it has a pronounced tendency to decrease until it completely disappears when going into the incomplete condensation mode as it show at Figure 2b. At the same time in a case of AlAs surface (Al fraction is 100%) the thickness of the wetting layer is practically unchanged and is 1.0-1.25 ML over the entire temperature range under consideration. This unusual behavior of the system is due, apparently, to an increase in the chemical interaction between In adatoms and the surface when the composition of the functional layer changes. To give an unambiguous explanation of the processes and mechanisms underlying these phenomena will allow their further study with the use of the theoretical approaches developed by us [38, 39].

Figure 1. Dependence of the average size of In/GaAs (a) and In/AlAs (b) droplets on the effective deposition thickness at different growth temperatures.

Figure 2. The temperature dependences of the density (a) and the critical thickness of the formation (b) of an ensemble of In/(Ga,Al)As nanostructures.
Statistical processing of the experimental data obtained by us showed that ensembles of self-organizing nanostructures significantly change their structural characteristics during the transition of formation from the subcritical thickness of deposition to supercritical (see Figure 3).

Figure 3. Histograms of the size distribution of an ensemble of In/AlAs nanostructures formed in different deposition modes: a) subcritical \((T = 150°C, H = 1.5 \text{ ML})\) and b) supercritical \((T = 150°C, H = 3.0 \text{ ML})\).

As can be seen from the Figure 3, in the subcritical region, the geometric parameters of the ensemble have a pronounced bimodal distribution, which, as we move to the supercritical region, changes to a unimodal distribution with a sequential decrease in the dispersion to the previously designated 5-15% as the substrate temperature increases.

It should be separately noted that an increase in the exposure time of structures, subject to the preservation of thermodynamic conditions on the surface, does not significantly change the nature of the distribution.

4. Conclusion
In summary, we carried out the experimental studies of the formation of the self-assembled In nanodroplets on the planar surfaces of AlGaAs epitaxial layers under As-stabilization conditions at different Al content in the surface layer. It is shown that an increase in the Al content in the epitaxial layer significantly changes the kinetics of the processes on the surface. This leads to an expansion of the range of possible densities of the obtained In nanodroplets without a significant change in their sizes. In addition, it was shown that with the increase of the Al fraction on the surface, the critical thickness of the formation of the In nanostructures is reduced and stabilized at a value of 1 ML. Also shown is the change in the distribution of the geometric characteristics of the nanostructures during a transition from subcritical to supercritical deposition modes.

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