Effect of the Al content in the substrate on the In nanodroplets growth by droplet epitaxy

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Abstract. The paper presents the results of an experimental study of the effect of Al content in the epitaxial surface layer on the growth of In nanostructures formed by droplet epitaxy. We have shown that an increase in the Al content leads to a decrease in the droplet density and an increase in their size. We also showed that the influence of the Al content on the droplet characteristics is much less significant than that in case of the Stranski-Krastanov growth. The increase in the critical thickness of droplet formation on Al-containing surfaces when the temperature decreases is not significant. It allows us to control quantum dot emission wavelength almost independently of their geometric characteristics.

1. Introduction
One of the promising ways for the further development of microelectronics is the transition to the creation of elements and devices based on quantum-dimensional structures, since horizontal and vertical nanowires, quantum dots, etc [1-10]. Among them, it is especially possible to distinguish quantum dots already actively used in optoelectronics and nanophotonics devices [11-13]. An important feature of quantum dots, as well as whole nanoscale systems, is the strong dependence of their functional characteristics on their structural and geometric parameters. This requires the development of technological methods and approaches that provide not only the ability to reproducibly obtain homogeneous arrays of such structures, but also precision control over the size, shape, composition of structures and their relative position in the array [1, 4, 14].

The traditional way of obtaining quantum dots for epitaxial technologies is the growth in the Stransky-Krastanov growth mode. The nanostructures obtained in this way have high material quality and good optical characteristics. At the same time, one of the significant drawbacks of this method is the need for a crystal lattice mismatch in the system, as well as the relationship between the density and size of structures in the array. This not only narrows the range of materials combinations in which quantum dots can be obtained, but also the possibility of varying their parameters, and hence the functional characteristics (band structure, optical range, etc.) [4, 15].

An alternative way to obtain quantum dots is the technology of droplet epitaxy [15-18]. This method is not only free from the drawbacks inherent in the Stransky-Krastanov mechanism, but also allows one to form a wide range of structures (quantum dots, rings, complexes based on them, etc.) in practically any binary systems and does not depend on the presence of lattice mismatch. Despite the advantages, droplet epitaxy is not well understood due to the large number of control parameters, as well as the high sensitivity of the growth to their change [19-21].
Since the first stage of droplet epitaxy (the formation of metal nanodroplets) largely determines the parameters of the final structures, the aim of this work was to study the effect of surface composition on the formation of droplet nanostructures in the In/AlGaAs system, as the most demanded for device application.

2. Experimental procedure

Experimental samples were grown using Semiteq STE35 MBE system with solid-state sources. After removal of native oxide, GaAs buffer layer 250 nm thick was grown on GaAs(001) substrate. Then, an AlGaAs epitaxial layer was grown with a thickness of 10 nm and an aluminum content of 0, 50, and 100%, respectively. Then all sources were closed, and the sample was cooled to a growth temperature. After reaching the growth temperature and vacuum less than $2 \times 10^{-7}$ Pa, indium was deposited with an equivalent rate of 0.25 ML/s and a thickness of 0.75 – 3.0 ML. The substrate temperature in this case varied in the range of 150 – 450°C. After the formation of an array of droplet nanostructures, the samples were studied by SEM and AFM methods.

3. Results and discussion

Since, by analogy with traditional epitaxial techniques, it was expected that an increase in the aluminum content would lead to a decrease in the diffusion length of adatoms and, as a consequence, to an increase in the density of nanostructures and their size, the results we obtained are ambiguous. An increase in the aluminum content in the case of droplet epitaxy led to an increase in the sizes of indium droplet nanostructures and a decrease in their density (Figure 1 (a, b)).

![Figure 1 (a, b). Surface aluminum mole fraction dependences of the (a) average diameter and (b) surface density of indium nanostructures after deposition of 3 ML indium at the substrate temperature 150 C, 200 C, 300 C and 450 C.](image)

Moreover, as can be seen from the constructed dependences, this effect intensifies with increasing substrate temperature. At low growth temperatures (less than 300 C), the influence of the substrate composition is relatively small: the average diameter of the droplet structures increases from 16 to 21 nm at 150 C and from 35 to 41 nm at 200 C with an increase in the aluminum fraction from 0 to 1. The density decreases by about 1.5 – 2 times: from $3 \times 10^{10}$ cm$^{-2}$ to $2 \times 10^{10}$ cm$^{-2}$ at 150 C and from $3 \times 10^{9}$ cm$^{-2}$ to $1.7 \times 10^{9}$ cm$^{-2}$ at 200 C.

In the region of relatively high growth temperatures (for droplet epitaxy of indium), the effect of the aluminum content is more pronounced. The density of indium droplet structures decreases by an order of
magnitude: from $1.8\times10^8$ cm$^{-2}$ to $1.4\times10^7$ cm$^{-2}$ at substrate temperature of 300 C and from $8.5\times10^6$ cm$^{-2}$ to $7\times10^5$ cm$^{-2}$ at 450 C. The average size of droplet nanostructures in this case increases by two times: from 98 to 176 nm at 300 C and from 230 to 437 nm at 450 C.

An important parameter of the first stage of droplet epitaxy technique is the critical thickness – the thickness of the deposition, after which three-dimensional structures (droplets) begin to form [22, 23]. Earlier, we showed that the critical thickness in the case of droplet epitaxy is determined by the initial state of the surface and the specifics of the interaction of adatoms with the surface, which largely depends on its temperature [22, 23]. Figure 2 shows the dependences of the critical thickness obtained by us at various temperatures on the surface composition.

Figure 2. Surface aluminum mole fraction dependences of the critical thickness of indium droplet formation at the different substrate temperature 150 C, 200 C, 300 C and 450 C.

As can be seen from Figure 2, the dependence of the critical thickness is complex, largely determined by the growth temperature. At low temperatures (less than 300 C), an increase in the aluminum content leads to a sharp decrease in the critical thickness from 3.0 and 2.25 ML to 1.25 and 1.0 ML at a substrate temperature of 150 C and 200 C, respectively.

At high temperatures, the type of the dependence changes. So, if at 300°C the surface composition practically does not affect (the critical thickness is about 1.0 ML for all surface type), then already at 450°C the critical thickness slightly increases from 0.75 to 1.0 ML. It is also important to note that, as follows from the presented dependences, the difference between the samples with the Al$_{0.5}$Ga$_{0.5}$As and AlAs surfaces is insignificant for most temperature conditions.

We suppose that the effects we discovered fit well with our previously [22, 23] proposed concept of the wetting layer formation in a droplet epitaxy, adjusted for the higher chemical activity of aluminum atoms in the surface layer. It should be noted that the small value of critical thickness for Al-containing surfaces and its sharp difference from the clean GaAs surface in the same growth conditions indirectly indicate that the mechanism of interaction of the adatoms with the substrate is responsible for the thickness changes, and not the adsorption of arsenic from residual atmosphere. We suppose that due to the high activity of aluminum, the interaction of adatoms and atoms of the first monolayer of the wetting layer with the surface is much stronger for Al-containing surfaces. This complicates their exchange with other adatoms, which leads, on the one hand, to an increase in their mobility, and, on the other hand, to a weak sensitivity of the wetting layer to temperature.
4. Conclusions
Thus, we conducted experimental studies of the effect of the aluminum content in the composition of the surface epitaxial layer on the formation of In droplet nanostructures using the droplet epitaxy technology. We found that, in contrast to traditional epitaxy, in our case an increase in the mobility of adatoms is observed on Al-containing surfaces, which is expressed in a decrease in the density of structures and an increase in their size. Moreover, we showed that the value of the critical thickness on Al-containing surfaces is practically temperature independent and is approximately equal to 1.0 ML in the entire range of the considered modes. We believe that this behavior is due to the features of the interaction of adatoms with surfaces of various compositions.

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