Arsenic background pressure effect on In droplet morphology

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Abstract. The paper presents the results of an experimental study of the effect of arsenic background pressure on the characteristics of droplet nanostructures formed by droplet epitaxy. We have shown that with an increase in the initial size of the droplets, the sensitivity of the system to the background pressure decreases. It is shown that the residual pressure of arsenic can be used as an additional control parameter of the droplet epitaxy technique.

1. Introduction

Geometric characteristics of self-assembled A3B5 semiconductor nanostructures largely determine their functional properties [1-14]. The size, shape, relative position of the structures in the array directly affect their band structure. Together with the chemical composition of the nanostructures themselves, the composition of the surrounding matrix, and crystal perfection, this determines the totality of the functional properties of the systems based on them – the electrophysical and optical characteristics, etc. In this regard, the task of precision control of the parameters of self-organizing A3B5 nanostructures is relevant [15-16]. Since it largely predetermines the functional characteristics of devices based on them, the task of controlling the parameters of self-organizing nanostructures looks especially relevant [17-21].

In this regard, droplet epitaxy looks extremely promising technologies, allowing, in contrast to the Stranski-Krastanov method, not only to form a wide range of nanostructures (from metal nanoclusters to quantum dots and different types of complexes based on them) in a wide range of compositions and material systems (including lattice matched), but also significantly expand the range of control parameters of nanostructures due to the significant number of process control parameters [22-28]. Nevertheless, despite active research in this area, the influence of only the main parameters of the droplet epitaxy process (deposition rate and thickness, growth temperature, crystallization parameters) is relatively well studied [29-33]. At the same time, the influence of secondary parameters of this technique is still not well understood [34].

The purpose of this work is to experimentally study the influence of such a secondary parameter as the As residual (background) pressure of the working environment on the morphology of metallic In nanodroplets formed on GaAs(001) substrates by the droplet epitaxy.

2. Experiment details

Experimental studies were carried out on the SemiTEq STE 35 installation equipped with solid-state molecular sources of group III elements and a valve source of arsenic. For epitaxial growth we used...
GaAs(001) epi-ready substrates. After thermal removal of the GaAs native oxide with the subsequent annealing of the substrate in the As flux, a GaAs buffer layer 250 nm thick was deposited at a temperature $T = 580^\circ$C. Then growth was interrupted by cut-off growth component fluxes, and the substrate was cooled to the temperature of droplet nanostructure formation. It should be noted that the cooling parameters were selected in such a way that in the As flux absence on the surface the (2×4) reconstruction is maintained. Initially, a series of samples was formed at various temperatures $T = 150…300^\circ$C and deposition thicknesses $H = 1…3$ ML and a fixed As residual pressure of $1\times10^{-7}$ Pa. In the second part of the experiment the time and rate of substrate cooling was varied so that the deposition of droplets occurred at different values of the As residual pressure. We grew two series of samples at the same growth temperature $T = 300^\circ$C, effective deposition rate of In (InGaAs ternary growth calibrated) $v = 0.25$ ML/s and different equivalent deposition thicknesses 1.5 and 3.0 ML, respectively. The exposure time of droplet structures in different series was also the same. In addition, in each series, the As background pressure was also changed, at which the deposition was carried out. Then the samples were investigated by the methods of AFM and SEM.

3. Results and discussion

Fig. 1 presents the results of experimental studies of the effect of growth temperature on the critical thickness of droplet formation and the minimum diameter of nanostructures observed in this case, which is achievable with the given parameters of the droplet epitaxy. As can be seen from the Fig. 1, an increase of deposition temperature $T$ from $150 \ ^\circ$C to $300 \ ^\circ$C leads to an increase in the minimum diameter from 15 to 65 nm, respectively. At the same time, a decrease in the deposition thickness below the critical value to reduce the droplet size leads to the suppression of the formation of droplet nanostructures.

![Figure 1](image.png)

**Figure 1.** Temperature dependences of critical thickness and minimum achievable size during In/GaAs(001) droplet epitaxy at equivalent deposition rate $v = 0.25$ ML/s.

Analysis of the data obtained during the second part of the experiment showed that the As background pressure can have a significant impact on the final size of the droplet nanostructures. So, as we can see from Fig. 2, when the thickness of the deposition of 1.5 ML increase in background pressure from $8.7\times10^{-8}$ Pa to $1.3\times10^{-7}$ Pa results in decreasing In droplet size by more than 200% – from 65 nm to 30 nm. As follows from comparison with the results presented above, using only basic control parameters of droplet epitaxy, obtaining nanodroplets with such dimensions is not achievable in principle.
Figure 2(a, b, c). SEM images of In nanodroplets obtained at growth temperature $T = 300^\circ$C, deposition thickness $H = 1.5$ ML and different values of background pressure $P$: (a) $8.7 \times 10^{-8}$ Pa; (b) $1.0 \times 10^{-7}$ Pa; (c) $1.3 \times 10^{-7}$ Pa.

At the same time, as follows from Fig. 3 and Fig. 4, with an increase in the initial size of the nanostructures, the sensitivity of the system to the background pressure decreases. So, at deposition thickness of 3 ML In, the droplet size changes by only 50% – from 160 nm to 110 nm, while the background pressure of As varied over a significantly wider range of values – $(1.0 - 6.3) \times 10^{-7}$ Pa.

Figure 3(a, b, c). SEM images of In nanodroplets obtained at growth temperature $T = 300^\circ$C, deposition thickness $H = 3.0$ ML and different values of background pressure $P$: (a) $1.0 \times 10^{-7}$ Pa; (b) $3.0 \times 10^{-7}$ Pa; (c) $6.3 \times 10^{-7}$ Pa.

Figure 4. Dependences of In droplet size on As background pressure obtained at growth temperature of $300^\circ$C and different deposition thicknesses: 1.5 ML (blue) and 3 ML (red). Symbols – experimental data, lines – approximation curves.
This behavior of the system is due to a sharp increase in the volume of droplet nanostructures with an increase in the deposition thickness with a constant exposure time of the formed structures and a slight change in the As flux on the surface from the residual atmosphere. The decrease in the size of droplet structures with an increase in the As flux on the surface during exposure is due to In diffusion from the droplet in order to bind with arsenic adsorbed on the surface.

Thus, we have shown that in order to preserve the reproducibility of the results in the process of droplet epitaxy of In(As)/GaAs nanostructures, it is necessary to carefully control the residual pressure of arsenic. In addition, we have shown that the background pressure can potentially be used as an additional control parameter in order to modify the dimensions of the original structures.

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