Semiconductors, 2024, Vol. 58, No. 11

Influence of structured substrate surface properties on the GaAs planar nanowire morphology (Monte Carlo simulation)

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Received December 5, 2024
Revised December 6, 2024
Accepted December 6, 2024

This work is devoted to the Monte Carlo simulation of the self-catalyzed growth of GaAs planar nanowires according to the vapor-liquid-solid mechanism on GaAs substrates covered with a structured film-mask. A structured film-mask is defined as a layer with the silicon oxide properties and with geometry in the form of grooves of different depths, widths, and distances between them. The influence of the film-mask properties and the structured surface geometry on the GaAs planar nanowire morphology was analyzed. A range of flux intensities for stable GaAs planar nanowires growth at the chosen temperature was found. The conditions for transition from vapor-liquid-solid growth to selective growth (without a drop) of planar nanowire were found, which makes it possible to obtain defect-free nanowires along the groove.

Keywords: planar nanowire, GaAs, structured surface, simulation, Monte Carlo.

DOI: 10.61011/SC.2024.11.59956.20S

1. Introduction

The interest in planar nanowires is due to their compatibility with the standard planar technology for making nanoelectronic devices [1–3]. Field-effect transistors based on GaAs planar nanowires have already been developed [4,5]. In [6], the growth of GaAs planar nanowires on GaAs substrates doped with *p*- and *n*-type impurities showing transistor characteristics is reported. The growth of planar nanowires with ZnSe/ZnTe radial heterojunctions on sapphire substrates demonstrating rectifying characteristics and photoluminescence under illumination is reported in [7]. Recently, planar nanowire matrices as the basis for quantum computers are being more widely used [8].

One of the ways to form nanowires is the vapor-liquid solid (VLS) mechanism using catalyst droplets. Gold is mainly used as a catalyst for the growth of semiconductor nanowires using the VLS mechanism [2,4,6]. However, Au atoms can be embedded in the growing crystal, which causes deterioration of the performance of optoelectronic devices based on such nanowires [9]. Therefore, the self-catalytic growth of $A^{III}B^{V}$ nanowires was proposed, where a metal included in the growing wire is used as a growth catalyst [10]. When the growth conditions of vertical nanowires were optimized, the appearance of planar nanowires was observed, which was attributed to unsuccessful growth modes [11]. Currently, a special search for the conditions of $A^{III}B^{V}$ planar nanowires formation is required. The possible directions of planar nanowires growth are given by the crystallographic orientation of the substrate surface [12], with the choice among the possible growth directions being determined by the arrival of the deposited material in the catalyst droplet [13]. In particular, there is uncertainty in the choosing the growth direction of the planar nanowires along the substrate. This is a problem because the manufacture of a number of planar nanowires-based devices involves a directional array of such crystals. The direction of planar nanowires growth can be controlled by pre-determined substrate surface geometry, i. e., planar nanowires growth on structured surfaces [14].

To integrate $A^{III}B^{V}$-based planar nanowires into silicon technology, one must overcome the problem of the mismatch between the lattice constants of most $A^{III}B^{V}$ and Si semiconductors. This issue can be resolved by organizing the GaAs planar nanowires growth on a dielectric structured mask on a Si substrate [15]. To date, silicon dioxide layers have been used as a film-mask for the self-catalytic growth of vertical GaAs nanowires. A number of studies have shown the influence of silicon oxide thickness on the orientation and morphology of GaAs planar nanowires grown both on silicon [16,17] and on GaAs [10] substrate. The value of the contact angle of Ga droplets on the substrate surface, which depends on the surface wettability, determines the nature of the subsequent growth of nanocrystals: planar or non-planar. It has been shown experimentally that the surface wettability of a gallium droplet is affected by the SiO_x layer composition and surface roughness, which depend on the thickness of the film-mask [16]. The oxide thickness affects not only the contact angle of the droplet with the surface, but also the droplet density. Thin silicon oxide films exhibit greater roughness, which should lead to a decrease in the diffusion length of Ga adatoms across the SiO_x surface and hence an increase in droplet density. On SiO_2 films, the droplet density decreases (compared to SiO_x) due to the active desorption of gallium adatoms from the surface [18]. However, work investigating the effect of silicon oxide thickness on nanowire growth has focused on vertical growth. There are practically no data on the self-catalytic growth of $A^{III}B^{V}$ planar nanowires by the VLS mechanism in literature, which, apparently, is due to the high requirements to the substrate surface properties and the narrow range of growth conditions required to form such structures. The dependence of the composition and morphology of the silicon oxide surface on its thickness may allow using structured oxide layers to organize the planar nanowire growth.

Various experimental techniques for the growth of semiconductor nanowires are currently used and many methods are available to control their morphology and structure. However, for a complete understanding of the processes leading to a particular nanowire morphology, it is necessary to consider the growing of such structures theoretically. Thus, the reasons for the planar and non-planar growth of GaAs nanowires on graphite nanoplates partially covering a thermally oxidized Si(111) substrate were considered using a two-dimensional analytical model [15]. Theoretical consideration of the kinetic characteristics of growing ZnSe and ZnTe planar nanowires on sapphire substrates has shown that the growth of ZnSe and ZnTe planar nanowires is determined by the surface diffusion of the material across the substrate, in contrast to the growth of vertical nanowires, when the surface diffusion along the lateral surface of the nanowires is the determining factor [19].

A suitable method for theoretical consideration of the kinetics and details of planar nanowire growth is Monte Carlo simulation. This study aims to find the optimal conditions for the self-catalytic growth of a unidirectional GaAs planar nanowire array on substrates coated with a structured silicon oxide layer by Monte Carlo simulation.

2. Monte Carlo model

The self-catalytic growth of GaAs planar nanowires was simulated using the SilSim3D software package based on a three-dimensional kinetic lattice Monte Carlo model [20]. Previously, using SilSim3D, we examined the self-catalytic growth of GaAs nanowires on singular and vicinal substrates uniformly covered by a thin film-mask with through-cylindrical holes in which gallium droplets were placed [13,21]. The present study is a continuation of these works. GaAs was chosen as the substrate material, but covered with a structured film-mask without through-holes. The focus is on investigating the influence of the properties and geometry of the structured layer on the morphology of GaAs planar nanowires. A schematic representation of the model substrate is shown in Figure 1. The structured film-mask consisted of alternating thick and thin layers with different properties. Structured mask films with constant values of groove width $W_t = 4\,\text{nm}$ and thin mask layer thickness $h_1 = 0.6\,\text{nm}$, varying values of groove spacing W and groove depth h_2 were considered in the simulations. The groove width W_t determines the maximum diameter of the planar nanowires, and the selected value of the mask thickness at the bottom of the groove h_1 corresponds to the thickness of the silicon oxide layer that provides good surface wettability of the gallium drop. The depth of the grooves h_2 varied in the range of 1–4 nm. The simulations showed that the poor wettability of the M_2 material by the Ga drop leads to the independence of the planar nanowire morphology from the groove height.

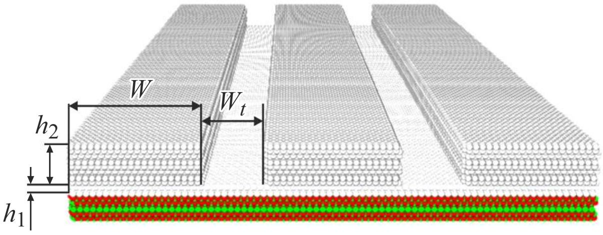

Figure 1. Schematic representation of the model substrate: W_t — width, h_2 — depth of the grooves, W — distance between them, h_1 — thickness of the thin layer at the bottom of the groove. Ga(s) is shown in red, As — green, the bottom layer of the mask film — white, and the top layer of the mask film — light grey. In the following, similar designations are used for the figures. (A color version of the figure is provided in the online version of the paper).

The Monte Carlo model is based on the following assumptions that occur during the self-catalytic growth of GaAs planar nanowires on a substrate covered with a structured mask film. The model takes into account that the properties of the mask film change with its thickness similarly to the silicon oxide layer [16]. The properties of the thin layer of the mask film at the bottom of the grooves with respect to gallium diffusion differ from those of the thick layer. The diffusion of atoms on the surface of the thick layer is much higher than on the surface of the thin layer at the bottom of the grooves. The diffusion characteristics of the thin layer of the mask film decreased due to both the change in the chemical composition of the film and the microroughness of the surface. Therefore, during the gallium deposition, the droplets are formed at the bottom of the grooves. The formed gallium droplets interact with the mask film, dissolving it to form a volatile component, after which the droplets are in contact with the GaAs substrate. Only after this the formation of GaAs nanocrystals at the drop-substrate interface and the subsequent nanowires growth during the deposition of gallium and arsenic are possible. In this case, the Ga drop is the growth catalyst, and the GaAs substrate under the drop plays the role of a seed crystal.

In simulation the self-catalytic growth of GaAs planar nanowires using structured films, we considered a 7-component system consisting of arsenic in atomic and molecular form (As, As_2), gallium in solid and liquid states (Ga(s), Ga(l)), particles of thin (M_1) and thick (M_2) layer of the mask film, and particles of the volatile component of the mask film (M_v). It is known that the interaction of Ga with SiO_2 results in the formation of volatile components such as Ga_2O and SiO [22]. In order to simplify, only one type of component responsible for the volatile compounds formed was introduced into the model. The following elementary events are taken into account in the model: adsorption of Ga(s) and As_2; diffusion of the components across the surface; decomposition and formation of As_2 by reversible reaction $As + As \leftrightarrow As_2$; dissolution of the mask film material by the gallium drop to form a volatile component ($M_i + Ga(l) \rightarrow M_v + Ga(l)$, where $i = 1, 2$) and its diffusion through the drop ($M_v + Ga(l) \rightarrow Ga(l) + M_v$); dissolution of GaAs and diffusion of As in liquid gallium through reactions $Ga(s) + Ga(l) \rightarrow Ga(l) + Ga(l)$ and $As + Ga(l) \rightarrow Ga(l) + As$; crystallisation of GaAs by interaction of liquid gallium with arsenic ($Ga(l) + As \rightarrow Ga(s) + As$); desorption of Ga, As_2 and the volatile component M_v. Certain activation energies correspond to each event.

The model takes into account the change in the properties of the mask film with its thickness. To reconcile the model and experimental [16] dependences of Ga droplet density on silicon oxide thickness, the properties of the thin M_1 and thick M_2 mask film were different. The differences include different binding energies of liquid gallium to the M_1 and M_2 mask film particles in the configurations responsible for surface wetting by the droplets. By varying the binding energies of liquid gallium to the surface of the mask film along the perimeter of the droplet, the contact angle of the droplet on the surface can be changed. Contact angles of 76, 100, and 116° were obtained, corresponding to the experimental values of the contact angles of gallium droplets on silicon oxide films with thicknesses of 0.6, 1.2, and 1.6 nm [16]. The binding energy of gallium atoms to the material M_1, which determines the diffusion of Ga adatoms across the thin layer of the mask film, was chosen equal to $E(Ga - M_1) = 0.8\,eV$, and the surface of this layer was set rough in order to increase the droplet density. Thin films of thermal oxide SiO_x, which are similar in composition to $x = 1$ have such properties [16]. Since the droplet density should be much lower on the M_2 material (a thick layer with SiO_2 properties) than on M_1, the binding energy of gallium atoms to the film was reduced to $E(Ga - M_2) = 0.3\,eV$. Taking into account the more active desorption of Ga from the surface of the thick film [18], the barrier to desorption from the surface of the M_2 material decreased compared to the M_1 material.

No data on the etching rate of SiO_x films with liquid gallium were found in the literature, so the activation energy of the film-mask dissolution reaction is a variable parameter. The calculations, the results of which are presented below, were performed at dissolution activation energies of 1.4 and 2 eV for M_1 and M_2 materials, respectively. The model energy parameters responsible for GaAs dissolution in the gallium drop and determining the surface tension were chosen as in [23].

3. Simulation results

The simulation of GaAs planar nanowire growth was carried out at a temperature of 890 K. This temperature is the optimum temperature for planar nanowire growth on GaAs(111)A substrates [24]. During simulation, the planar nanowire growth with Ga droplet pre-deposition was considered. In this case, gallium was pre-deposited on the substrate covered with a structured mask film to form droplets in the grooves, and the arsenic flux for planar nanowire growth was switched on after the droplets etched the film underneath. In this case, the required droplet density in the grooves could be achieved regardless of the growth conditions, ensuring stable growth of planar nanowires. The droplet density in the grooves is determined by the surface properties of the thin film-mask layer, the distance between the grooves, the substrate temperature, and the rate of gallium deposition. The maximum droplet spacing in the model is limited by the groove length, which was taken to be 70 nm. In this study, all calculations were performed at the same temperature — 890 K. As the gallium flux intensity F_{Ga} increases, the droplet spacing decreases due to a decrease in the diffusion length λ_{Ga}. To evaluate the effect of F_{Ga} on the diffusion length of gallium over the structured surface of the mask film, λ_{Ga} was previously evaluated on the unstructured surface of the M_1: at $F_{Ga} = 1$ ML/s – $\lambda_{Ga} \sim 25$ nm and at $F_{Ga} = 1.2$ ML/s – $\lambda_{Ga} \sim 20$ nm. In our case, the flux intensity F is measured in ML/s. On a surface with (111) orientation, 1 monolayer (ML) characterizes the number of surface atoms and corresponds to $7.8 \cdot 10^{14}$ atom/cm^2. When gallium is deposited on a structured substrate, the droplet density in the grooves is influenced by the additional inflow of gallium from the surface of the thick layer M_2 between the grooves. It is found that the droplet density in the groove at a fixed Ga flux increases with increasing width of the thick layer between the grooves W. Thus, at $F_{Ga} = 1$ ML/s for $W = 8$ nm, one droplet is formed in the groove, and for $W = 32$ nm — 2 droplets each. This means that when changing the geometry, i.e., changing the periodicity in the surface structure of the mask film (different W) it is necessary to vary the Ga flux intensity to obtain the same droplet density in the grooves. The Ga flux intensities to obtain a single droplet in each groove during pre-deposition are $F_{Ga} = 1.1$, 0.6, 0.4 ML/s for thick layers between grooves with widths of $W = 8$, 32, 68 nm, respectively. It should be noted that reducing the groove spacing may result in droplets not nucleating in every groove. The Ga pre-deposition step not only allows to ensure a given droplet density, but also to obtain droplets of a given size due to the duration of the pre-deposition. At optimal choice of flux intensity and duration of deposition, Ga droplets will not only fill the entire width of the groove,

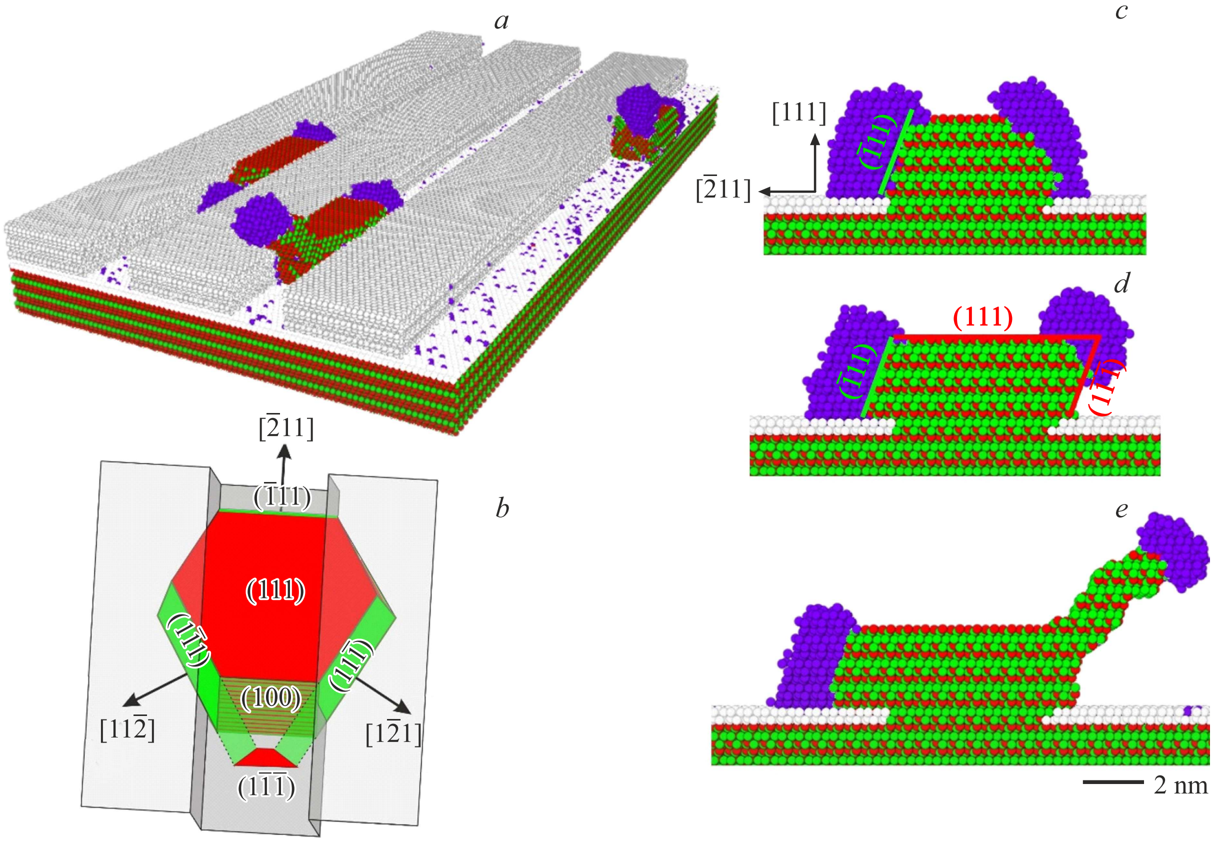

Figure 2. Kinetics of planar nanowire growth disruption. Structured substrate with GaAs nanowires in three grooves at $W = 8$ nm (*a*); schematic representation of the crystal under the drop in groove (*b*); cross sections of nanowires in the second groove at times $t = 0.11$ s (*c*), 0.15 s (*d*), 0.2 s (*e*) after the onset of Ga deposition and As_2 at $F_{Ga} = 1.1$ ML/s, $F_{As_2} = 11$ ML/s, $T = 890$ K. The (111)A facets are marked in red and (111)B — in green. Ga(l) is marked in purple.

but will also have time to etch a thin layer of the mask film at the bottom of the groove. Only after the contact of the droplet with the crystalline substrate does the growth of nanocrystals begin when the flux As_2 is switched on.

Stable growth of planar nanowires depends not only on the optimal ratio of gallium and arsenic fluxes, but also on their absolute values. At non-optimal choice of gallium and arsenic flux intensities, a disruption of planar nanowire growth to inclined growth is observed. Figure 2, *a* shows a model substrate on which one nanowire grows planar, and the other two nanowires are disrupted from planar to inclined growth. Let us consider the mechanism of planar nanowire growth disruption in more detail. At the initial stages of growth, a crystal is formed under the drop, the shape of which is determined by the crystallography of the underlying substrate. A schematic representation of the crystal under the drop on the surface of GaAs(111)A substrate is shown in Figure 2, *b*. On the structured mask film, crystal growth is not possible in all directions allowed by crystallography due to limitations by the groove walls, which are indicated in grey in Figure 2, *b*. Therefore, the crystal under the drop expands in opposite directions, tearing the drop into two parts. Figure 2, *c*–*e* shows the cross sections of the nanowire at different time points along the [$\bar{2}11$] direction. During the initial stages of growth in the cross section, the crystal under the droplets takes the shape shown in Figure 2, *c*. The left side of the crystal is a ($\bar{1}11$)B facet, and the right side is the intersection of two ($1\bar{1}1$)B and ($11\bar{1}$)B facets. The outgrowth of the left ($\bar{1}11$)B facet (Figure 2, *d* and *e*) displaces the droplet in the [$\bar{2}11$] direction, allowing the growth of the planar nanowire. Free growth of the two remaining (111)B facets is impossible because of the limitation by the walls of the groove in the mask film. These facets gradually grow only in the groove region. Eventually, an additional ($1\bar{1}\bar{1}$)A facet, emerges from the substrate side, limiting the growth of ($1\bar{1}1$)B and ($11\bar{1}$)B facets in the [$21\bar{1}$] direction (Figure 2, *b* and *d*). Since the crystal growth is determined by the (111)B facets, the drop detaches from the mask film on the right side of the crystal (Figure 2, *d*) and, as a consequence, the growth of the inclined nanowire occurs (Figure 2, *e*). It should be noted that under similar deposition conditions, but on GaAs(001) substrates, a more stable growth of planar nanowire in the groove because of the complete symmetry of the facets of the nanocrystal under the drop is observed.

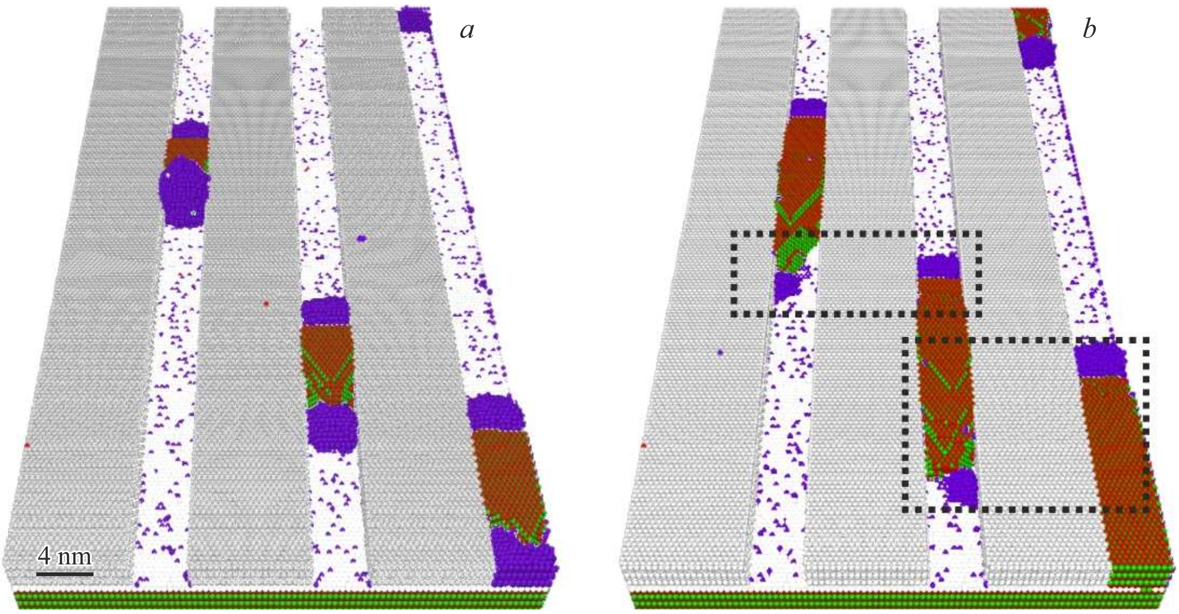

Figure 3. View of model substrate with $W = 8$ nm at different duration of arsenic and gallium deposition: $t = 0.3$ s (*a*), $t = 0.8$ s (*b*); droplet predeposition was carried out at $F_{\mathrm{Ga}} = 1.1$ ML/s; planar nanowire growth — at $F_{\mathrm{Ga}} = 0.6$ ML/s, $F_{\mathrm{As}_2} = 6$ ML/s.

To prevent the failure of planar growth on GaAs(111)A substrate, the crystal growth rate under the drop should be reduced so that the drop continues to wet the surface of the oxide film on both sides of the crystal. To do this, the F_{Ga} and F_{As_2} flux intensities must be properly selected. The growth rate of the crystal is determined by the arsenic flow rate. If the $F_{\mathrm{As}_2}/F_{\mathrm{Ga}}$ ratio is too high, it leads to a high crystal growth rate and a high probability of failure of planar nanowire growth. In addition, an excessive amount of arsenic compared to gallium can lead to a decrease in the size of the Ga droplet and its disappearance. If the $F_{\mathrm{As}_2}/F_{\mathrm{Ga}}$ ratio is small, the crystal growth is accompanied by an increase in the Ga drop size. The simulation allowed us to determine the optimal ratio of Ga and As_2 fluxes for stable planar nanowires growth on the structured surface. For all options of the considered structured surfaces, the optimal ratio of $F_{\mathrm{As}_2}/F_{\mathrm{Ga}}$ fluxes is equal to 10. A decrease in the absolute values of the fluxes while maintaining their ratio increases the stability of planar nanowires growth, i.e. reduces the probability of planar nanowire growth disruption.

The stability of planar nanowire growth is also affected by the distance between the grooves. If the distance between the grooves is small, the effect of changing the size of droplets in neighbouring grooves as they get closer together was observed during the growth process. Figure 3 shows how the sizes of initially identical gallium droplets change as they move during the growth process. It can be seen that the lower drop from the first groove has decreased compared to the upper drop in the second groove, and the lower drop from the second groove has decreased compared to the drop from the third groove (the areas of interacting drops are highlighted with a dotted line). This is explained by the rather small groove spacing ($W = 8$ nm) compared to twice the diffusion length of arsenic across the surface of the M_2 layer, resulting in droplet competition for arsenic. Estimates of the As_2 diffusion length before evaporation at 890 K over the surface of the top layer of the mask film gave a value of ~ 10 nm. It should be noted that at the chosen growth temperature in the considered flux range, the gallium diffusion length across the mask layer M_2 between the grooves is much larger than the maximum model value W. This implies that different crystal growth rates under the droplets, determined by the arsenic influx, are responsible for the variation in the size of closely spaced droplets. The exchange of atoms from neighbouring grooves was also observed experimentally during the selective growth of GaAs nanowires on the structured surface [25].

It is possible to avoid the effect of droplet size variation by increasing the groove spacing. When W exceeds twice the diffusion length of arsenic before evaporation, the diffusive collection of arsenic from the surface will be sufficient for the planar nanowire growth from both competing droplets. The simulation as carried out for a set of W values from 8 to 68 nm. Figure 4 shows the stable planar nanowire growth on structured substrates with $W = 32$ and 68 nm. It should be noted that for any width of thick mask layer between the grooves, the growth conditions need to be changed from pre-deposition conditions for stable planar nanowire growth. For different groove spacing, the Ga and As_2 flux ratios are the same, but the absolute values are different.

Since several droplets can nucleate in one groove, and the planar nanowire growth from each droplet occurs in opposite directions, a planar growth disruption occurs when

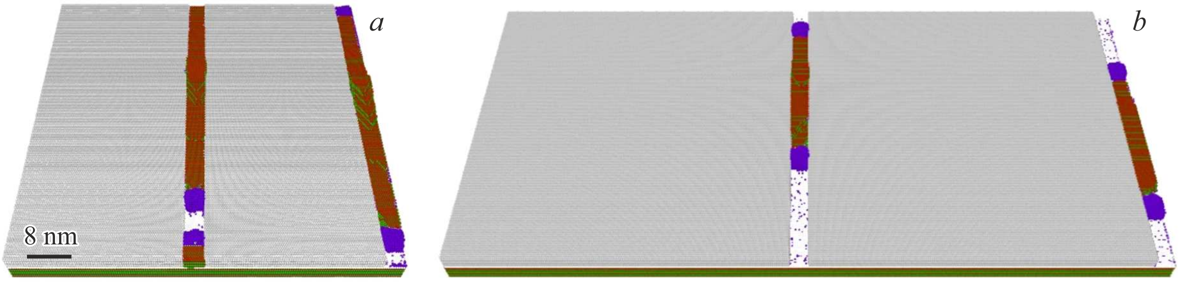

Figure 4. 3D view of model substrates with different groove spacing with stable growth of planar nanowires during 2 s: *a* — $W = 32$ nm, the Ga pre-deposition at $F_{\text{Ga}} = 0.6$ ML/s; planar nanowire growth — $F_{\text{Ga}} = 0.5$ ML/s, $F_{\text{As}_2} = 5$ ML/s; *b* — $W = 68$ nm: the Ga pre-deposition at $F_{\text{Ga}} = 0.4$ ML/s; planar nanowire growth — $F_{\text{Ga}} = 0.2$ ML/s, $F_{\text{As}_2} = 2$ ML/s.

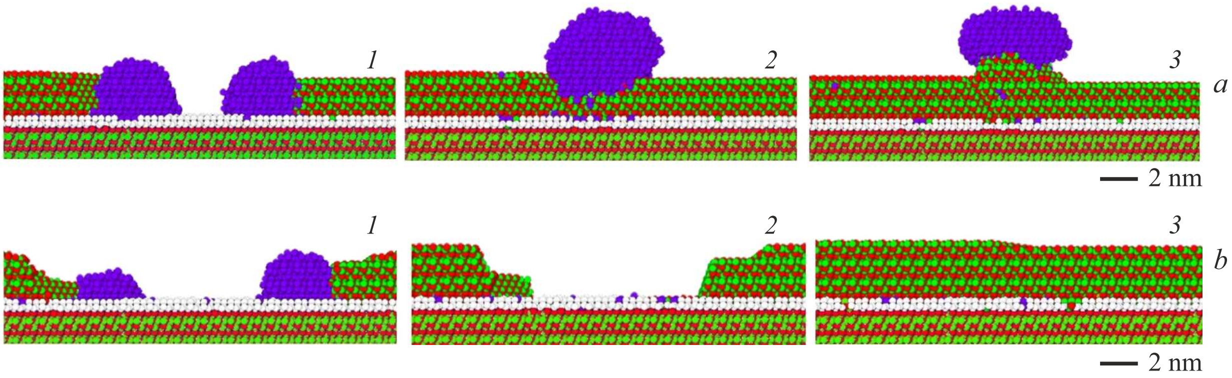

Figure 5. Fragments of the model surface cross-sections with two planar nanowires growing in opposite directions. *a* — droplet merge during the VLS growth of planar nanowires with subsequent formation of an additional vertical nanowire ($F_{\text{Ga}} = 0.5$ ML/s, $F_{\text{As}_2} = 5$ ML/s). *b* — formation of defect-free planar nanowires when the growth mode is changed — transition from VLS to selective growth (*1* — $F_{\text{Ga}} = 0.5$ ML/s, $F_{\text{As}_2} = 5$ ML/s, *2* — $F_{\text{As}_2} = 5$ ML/s, *3* — $F_{\text{Ga}} = 0.3$ ML/s, $F_{\text{As}_2} = 20$ ML/s).

two oppositely moving droplets meet — vertical growth is observed (Figure 5, *a*). The appearance of non-planar nanowires on the planar nanowire surface can be avoided by changing the growth conditions for the absorption of gallium droplets and growing planar nanowires without droplets. To do this, before the droplets merge, the gallium flux was switched off while the arsenic flux was maintained (Figure 5, *b–1*). After the droplets disappeared, the growth conditions were changed again. To prevent the repeated formation of Ga droplets, further GaAs growth was carried out at an overestimated ratio of Ga and As_2 fluxes: $F_{\text{Ga}} = 0.3$ ML/s, $F_{\text{As}_2} = 20$ ML/s. Thus, at the final stage of growth, we switched from VLS growth to planar nanowire growth in the absence of a droplet. Unlike standard selective growth, in this case the nanowire continues to grow on the surface of the mask film.

4. Conclusion

The conditions of self-catalytic growth of planar GaAs nanowires on GaAs substrates covered with a structured mask film have been analyzed using Monte Carlo simulations. The influence of the characteristics of the mask film in terms of properties approximating silicon oxide layers and the geometry of the structured surface on the growth character and morphology of GaAs planar nanowires was investigated. Taking into account the influence of silicon oxide thickness on the wettability of its surface by gallium drops has shown the possibility of using structured silicon oxide layers to organize planar nanowire growth. The structured film morphology was an alternation of thin and thick SiO_x layers forming a system of parallel grooves. The thin layer defining the groove bottom properties corresponded to x close to 1, and the thick layer defining the properties of the sidewalls and terraces between the grooves corresponded to $x = 2$. The groove spacing of the structured layer was shown to affect the diffusive collection of arsenic into droplets, leading to different growth rates of planar nanowires in neighbouring grooves. To ensure the stable growth of planar nanowires, the groove spacing should exceed twice the arsenic diffusion length across the top layer of the mask film. At a given temperature, a range of intensities of gallium and arsenic fluxes was found in which stable growth of GaAs planar nanowires was observed. At the optimum ratio of gallium and arsenic flux intensities $F_{\text{As}_2}/F_{\text{Ga}} = 10$ a correction of their absolute values by varying the groove spacing is required for stable planar nanowire growth. The transition from VLS growth to drop-free planar nanowires growth allowed defect-free nanowires to be obtained along the entire groove length.

Funding

The study was supported by the Ministry of Science and Higher Education of the Russian Federation.

Conflict of interest

The authors declare that they have no conflict of interest.

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Translated by J.Savelyeva