Monte Carlo investigation of the MBE growth of GaAs on the surfaces with different crystallographic orientations

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Abstract. MBE growth of GaAs on the surfaces with different orientations is studied by kinetic Monte Carlo simulations. The model allows taking into account the most relevant processes and reconstructions on the surfaces. The simulations show that islands grown on the (111)A surface have a chaotic shape whereas those on the (001) and (110) surfaces have more ordered structure due to the features of the surface structure. The diffusion length of Ga adatoms is observed to be dependent on the growth rate and V/III flux ratio. This influence is stronger for the (111) and (110) surfaces demonstrating that diffusion is weaker on the (001) surface. However, the diffusion length reduces with increasing growth rate and As₂/Ga flux ratio tending to become equal for all surfaces under consideration.

1. Introduction
The formation of A\textsuperscript{III}B\textsuperscript{V} nanostructures is realized by a number of techniques. Many advantages make molecular beam epitaxy (MBE) one of the most promising methods for the fabrication of nanostructure devices for nanophotonics, opto- and nanoelectronics [1].

In the growth process of A\textsuperscript{III}B\textsuperscript{V} materials, the most attention is paid to the characteristics of grown structures which are controlled by the substrate temperature. However, most studies do not consider the V/III flux ratio and growth rate dependences, although these parameters have a huge influence on the characteristics of growing films [2–6].

Despite the fact that studies of the epitaxial growth of semiconductor materials based on GaAs have been carried out for a long time, the growth mechanisms have been mainly studied on a substrate with the (001) orientation. The substrates with other orientations, such as (110) and (111) A, which have other properties and characteristics in comparison with (001), have become no less significant in recent times.

The study of the (111)A orientation of GaAs is important due to the possibility to realize laterally symmetric quantum dots belonging to the C\textsubscript{3v} point symmetry group of the (111) surfaces. At such symmetric points, effective spin orientation through optical pumping and minimal multiplet splitting is observed. This makes it possible to efficiently generate pairs of entangled photons [7,8]. In addition, the surface of GaAs (111) A is subject to increased attention due to its unique technological properties: silicon-doped GaAs layers grown on such a surface by the MBE method exhibit either n- or p-type conductivity depending on the growth conditions [9]. A\textsuperscript{III}B\textsuperscript{V} semiconductors with zinc blende structure give almost atomically clean cleavage surfaces (110) in view of their charge neutrality, which are used as mirror planes of optical cavities.

The relevance of studying epitaxial growth on surfaces with different orientations is also due to the possibility of forming low-dimensional structures on pre-structured substrates [10–13]. In such cases,
the initial surface is a system, in most cases, of low-index crystallographic planes. For the controlled formation of arrays of nanostructures and heterostructures based on them, precise control of the growth process and a deep understanding of elementary growth processes are needed, including adsorption-desorption, surface diffusion and two-dimensional nucleation. Growth by this method is very sensitive to technological parameters and can be accompanied by the formation of undesirable defects. In connection with this, it is necessary to study the mechanisms of the formation of epitaxial layers at an atomic scale on surfaces with different crystallography.

In this paper, we investigate the influence of the growth rate and the V/III flux ratio on the diffusion characteristics of Ga adatoms with the growth of GaAs on surfaces with different crystallographic orientations. To implement this study, we used the kinetic Monte Carlo method which takes into account such important factors as multicomponent growth, the surface structure, molecular form of species, modelling in a wide range of growth temperatures and growth rates, taking into account the growth time corresponding to the experimental growth, and also features of the growth system, which is not feasible with the help of analytical models.

2. Description of the model

2.1. Growth system

Within the framework of the model under consideration, it is assumed that growth occurs by the "vapor-solid" mechanism. The initial simulated GaAs (001) surface has the crystalline structure of zinc blende and the reconstruction of β(2×4), and the GaAs (111) A and GaAs (110) have a reconstruction of (2×2) and (1×1), respectively.

2.2. Algorithm

The algorithm for the growth simulation is based on the kinetic Monte Carlo method. The minimal step of the model is a simple event specified randomly in the timeline, the probability of which is determined by the activation barrier for an event, each of which is related to the type of microscopic process, the location of the particle on the reconstructed surface and its closest surroundings in the interaction radius of two interatomic distances for the surface with orientation (001), and for (111)A and (110) the interaction radius is one. The values of the activation energies for the main events were chosen on the basis of both experimental and earlier theoretical studies [2–18]. For intermediate values, the activation energy was modified by adding binding energies of the conglomerates obtained as a result of calculations in the density functional theory [18] to the activation barrier for the corresponding jump of a free atom. The frequency of an event is determined by the Arrhenius equation [19–23]:

\[ p_i = v_0 \exp\left(\frac{E_i}{kT}\right) \]

where \( v_0 \) is the vibration frequency of the lattice atoms estimated at \( 10^{13} \, \text{s}^{-1} \) [24], \( E_i \) is the activation energy of a microscopic process, and \( k \) is the Boltzmann constant.

2.3. Microscopic processes

The following microscopic processes are taken into account in the present simulation.

2.3.1. Adsorption and desorption of Ga. Ga atoms are deposited at random lattice sites from a flux determined by the growth rate and equal to:

\[ J_{Ga} = 0.5 \frac{v}{\sigma} \]

where \( \sigma \) is the area occupied by one lattice atom. Ga atoms are adsorbed with a single sticking coefficient, desorption is neglected [25,26].

2.3.2. Surface diffusion of Ga. Migration of a Ga adatom is implemented as successive diffusion jumps to neighboring lattice sites. The surface diffusion of Ga is anisotropic for GaAs (001), and for
(110) and (111)A it is isotropic. Ga adatom can diffuse from any state, but if it is in a strongly bound state [22], then the probability of its jump becomes close to zero, and the adatom can be considered as incorporated into the crystal.

2.3.3. Physisorption and surface diffusion $\text{As}_2$. Molecules of $\text{As}_2$ can be adsorbed without dissociation in the form of a dimer [14].

Therefore, modelling in this paper assumes that arsenic dimers precipitate into random lattice sites. $\text{As}_2$ molecules are chemically adsorbed (chemisorbed) only on adatoms of Ga, whereas the interaction of $\text{As}_2$ with an uncovered surface occurs through weak molecular adsorption (physisorption) in several places [14]. Diffusion of As takes place in the form of dimer molecules in adjacent rows. However, this process competes with the desorption and chemisorption processes of $\text{As}_2$, which determines the necessity of introducing an activation barrier for the diffusion [16]. The mobility of arsenic on the surface is mainly due to the presence of the physisorbed state of $\text{As}_2$. Any contribution of atomic As on the surface is neglected in connection with the absence of dissociative chemisorption $\text{As}_2$ [22].

2.3.4. Chemisorption of $\text{As}_2$. Chemosorption of $\text{As}_2$ on one or more gallium atoms is possible. The activation energy of this process also depends on the location and environment of the arsenic dimer, depending on the binding energy of the complex of atoms [14].

2.3.5. Desorption of $\text{As}_2$. In the model, it is accepted that desorption of arsenic is possible only from the physisorbed state [17,23].

3. Results and discussion

The morphology of the surface at various technological parameters is shown in Figure 1. The essential difference between the morphology on various surfaces is obvious. This is primarily due to the difference in the surface structure. While the surface with orientations (110) and (111)A have isotropic reconstructions (1×1) and (2×2), respectively, the surface (001) is (2×4)-reconstructed. This is appeared in the presence of alternating arsenic dimericrows and missing trenches. As a result, the migration of adatoms is limited in the direction transverse to the dimer rows which leads to anisotropy in the shape of islands [2]. According to the results of experimental studies [27,28], the diffusion on the (111)A surface is isotropic, while on the (001) surface the diffusion intensity essentially depends on its direction, which is confirmed by the results of the simulation.

Also in Figure 1, there is a significant difference in the geometric characteristics of islands on the (001) surface and on the (110) and (111)A surfaces. In addition to the fact that the dimer rows limits the surface diffusion of adatoms, the density of packing of the crystal atoms differs on the surfaces under consideration. In the case of (001), the interatomic distance is larger, as a result of which the energy barrier for diffusion increases. In connection with this, nucleation processes prevail over the growth processes of two-dimensional islands and the island density increases with a corresponding decrease in their average size [29].
Figure 1. Morphology of the GaAs surfaces in a simulation area of 160 Å × 200 Å after deposition of 0.1 ML GaAs (○ – Ga atoms, ● – As atoms): a) GaAs(001): $T = 580^\circ$C, $R_\text{As/Ga} = 6$, $v = 0.1$ ML/s, b) GaAs(111): $T = 500^\circ$C, $R_\text{As/Ga} = 6$, $v = 0.1$ ML/s, c) GaAs(110): $T = 500^\circ$C, $R_\text{As/Ga} = 6$, $v = 0.1$ ML/s.

The surfaces (110) and (111) A demonstrate a similar morphology and close in size and surface density of islands. However, the islands on the (110) surface have sharper facets, while on the (111) they are chaotic in shape, as evidenced by the results of STM studies of two-dimensional islands[15]. This phenomenon can be explained by the features of the crystal structure of the surface.

Using the developed model, the diffusion characteristics of Ga adatoms were also calculated as a function of the kinetic parameters of the MBE method (Figures 2 and 3). It is shown that on the (001) surface the diffusion length is really much smaller than on the other surfaces under consideration (Figures 2c and 3c) which is confirmed in [27,30].

Figure 2. Growth rate dependences of the a) lifetime, b) diffusion coefficient, c) diffusion length of Ga adatoms on the GaAs(001), GaAs(110) and GaAs(111) surfaces at $R_\text{As/Ga} = 6$.

The simulation also shows that the diffusion of Ga adatoms exhibits a different behavior when the V/III flux ratio changes. Both the growth rate and the V/III flux ratio have a significant effect on the diffusion parameters of adatoms, but adatoms on the (110) and (111)A surfaces are most influenced, in good agreement with the works [2,15]. Although the diffusion length on the GaAs(001) is less dependent on the technological parameters, both the growth rate and V/III flux ratio have an influence on the diffusion characteristics of Ga adatoms which was shown in previous works [2,31]. It can be seen from the results that the diffusion length and lifetime (Figures 2a and 3a) of Ga adatoms on the (110) surface are significantly larger than those of (001) and (111)A in connection with the lower binding energy of adatoms with the surface. The diffusion characteristics on the (110) and (111)A surfaces are similar, but diffusion is more intense at the (110) surface. Despite the weak dependence of the diffusion length and lifetime of Ga adatoms on the (001) surface, the behaviour of these characteristics with changing technological parameters are the same.
With an increase in the growth rate from 0.01 to 1 ML/s, the diffusion length and lifetime of Ga adatoms decrease for all surfaces, and it is most significant when changing at low growth rates. A similar dependence is observed when the V/III flux ratio is increased from 3 to 40, but the effect of this quantity is greater than the growth rate. This regularity is also observed in [11,30].

An important feature of the diffusion on the (001) surface is the reverse dependence of the diffusion coefficient on the V/III flux ratio compared to other surfaces (Figure 2b). With an increase in the flux ratio, the diffusion coefficient on the (001) surface increases, while the diffusion length and lifetime decrease. This indicates that a change in the lifetime of adatoms before incorporation does not lead to a sufficiently significant change in the diffusion length. A similar result was obtained in [20] for a temperature dependence in which the lifetime of Ga adatoms before incorporation increased with temperature, while the diffusion coefficient exhibited the opposite dependence.

4. Conclusion
In summary, the influence of the crystallographic orientation of GaAs on the MBE growth kinetics was investigated using the kinetic Monte Carlo method. The island morphology was shown to be strongly dependent on the orientation due to the difference between the surface reconstructions. The diffusion of Ga adatoms on the GaAs(001) surface was observed to be anisotropic and more suppressed in comparison with the (110) and (111)A surfaces. It was found that the diffusion length is the largest on the GaAs(110) surface, and it is the most strongly influenced by the growth rate and As$_2$/Ga flux ratio. The surface lifetime and diffusion length are weakly dependent on these parameters but decrease with increasing growth rate and V/III flux ratio as well.

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