# Monte Carlo simulation of V/III flux ratio influence on GaAs island nucleation during MBE

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**Abstract.** The kinetic Monte Carlo simulation of GaAs/GaAs(001) molecular beam epitaxial growth considering V/III flux ratio influence on nucleating island characteristics is presented. It is shown that the island density increases with the surface coverage increase and reaches saturation after deposition of ~0.1 monolayer of GaAs. The increase of V/III flux ratio from 3 to 40 leads to the increase of the island density from $1.9 \cdot 10^{12}$ to $2.6 \cdot 10^{12}$ cm$^{-2}$. At the same time the average size decreases from 4.4 to 4.1 nm. The island size distribution function narrows with V/III flux ratio increase. This is attributed to the shortage of gallium atoms in comparison with deposited arsenic molecules that prevents large island formation and leads to the dramatic growth of little island concentration. The simulation demonstrates good agreement with experimental results.

## 1. Introduction

Molecular beam epitaxy (MBE) is one of the most promising fabrication method for nano- and optoelectronic, photonic and quantum processing device structures. A more accurate understanding of the fundamental kinetic processes during epitaxial growth including submonolayer regime would aid the manufacturing process.

During MBE of III-V semiconductor structures their parameters are primarily controlled by substrate temperature and growth rate. At the same time V/III flux ratio is not usually considered to alter growing structure characteristics although experiments show that it has a significant influence on nucleating island size and surface density [1-3].

By now, many experimental and theoretical techniques are developed to reveal growth mechanisms during epitaxy of semiconductors. These methods have permitted to scrutinize a lot of parameters of growth process, such as activation energy [4-6], migration rate [7], diffusion coefficient [1,4,8], diffusion length [7,8]. However, very little attention is focused on macroscopic characteristics which are average island size, form, surface density and size distribution function.

In this paper, we report kinetic Monte Carlo simulation of GaAs epitaxial growth on the GaAs(001) surface considering V/III flux ratio effect on the growing island geometrical characteristics. Monte Carlo method enables calculations in a wide range of technological parameters and at large time and length scales what is unattainable with *ab initio* calculations.

## 2. Description of the model

In this simulation, the GaAs(001) surface is assumed to have zinc blende crystal structure with $\beta2(2\times4)$ reconstruction. This means that there are arsenic dimer rows alternating with missing trenches on the surface [9]. We consider the growth from the fluxes of two principally different particles [10,11] on the surface with complicated structure, so it is necessary to take into account a lot of microscopic processes. Each process is implemented as a set of simple events depending on a particle type, location and environment. A basic parameter of an event $i$ is an activation energy barrier $E_i$ which defines the frequency $f_i$ of occurrence of this event by the Arrhenius law:

$$f_i = \nu_0 \exp(E_i / kT), \tag{1}$$

where $\nu_0$ is the attempt frequency set at $10^{13}$ s$^{-1}$ [12]; $k$ is the Boltzmann constant; and $T$ is the growth temperature.

The following microscopic processes are involved in the simulation and predefine more than 40 elementary events:

- Ga adsorption and desorption. Ga atoms are deposited into random gallium sublattice sites with unity sticking probability and do not desorp any longer;
- Ga surface diffusion. Ga adatom migration is implemented as a sequence of site-by-site diffusion hops. Ga diffusion is anisotropic so that the activation energy of free Ga atom diffusion is 1.3 eV along the [1$\bar{1}$0] direction [13] and 1.5 eV along the [110] direction [1];
- As$_2$ physisorption and surface diffusion. Arsenic is deposited on the surface from the As$_2$ molecule flux and occupy random doubled sites in the arsenic sublattice. As$_2$ molecules can only adsorb chemically (chemisorb) on Ga adatoms whereas interaction of As$_2$ with uncovered surface is via weakly bound (physisorbed) state [4]. The presence of this state enhances As$_2$ molecule migration. Since one arsenic molecule performs $\sim10^9$ hops during one gallium hop we assume that one diffusion step of As$_2$ molecule has random distribution:

$$l = -\lambda_{As} \ln \delta, \tag{2}$$

where $\lambda_{As}$ is arsenic molecule diffusion length depending on technological parameters and varying from 1 to 30 μm [14-16], $\delta$ is a random number having uniform distribution in the range (0,1]. Since site-by-site migration of arsenic molecules is neglected, the calculations have been accelerated essentially.

Arsenic diffusion activation energy is 0.2 eV [17]. The contribution of atomic As is neglected since As$_2$ dimers do not dissociate under typical growth conditions [18].

- As$_2$ desorption. We assume that arsenic can re-evaporate from the surface only from the physisorbed state, and the activation barrier of this process is equal to 0.37 eV [19,20].
- As$_2$ chemisorption. As$_2$ molecules can only chemisorb on Ga adatoms. However, the activation energy of this process varies from 0.25 eV for the chemisorption in a trench site on two gallium dimers to 0.55 eV in case of the chemisorption on a dimer row [19].

The reverse transition of a chemisorbed As$_2$ molecule to the physisorbed state is probable. The activation energy also depends on the location and environment of an arsenic dimer and varies from 1.7 to 2.6 eV [4].

As opposed to the traditional kinetic Monte Carlo method, the characteristic time interval in our simulation is deterministic, but the number of events $n_{ev}$ occurring during one time step is distributed randomly:

$$n_{ev} = -n_0 \ln \delta, \tag{3}$$

where $n_0$ is a nominal number of events.

## 3. Results and discussion

We reveal that islands preferentially nucleate in the trenches and have elongated form along the $[1\bar{1}0]$ direction (figure 1) what is connected with the diffusion anisotropy [1]. The (2×4) reconstruction remains on the surface during the entire submonolayer growth. The morphology of islands formed under the same technological parameters is in good agreement with the scanning tunnelling microscopy images [13] and other theoretical models [1,18].
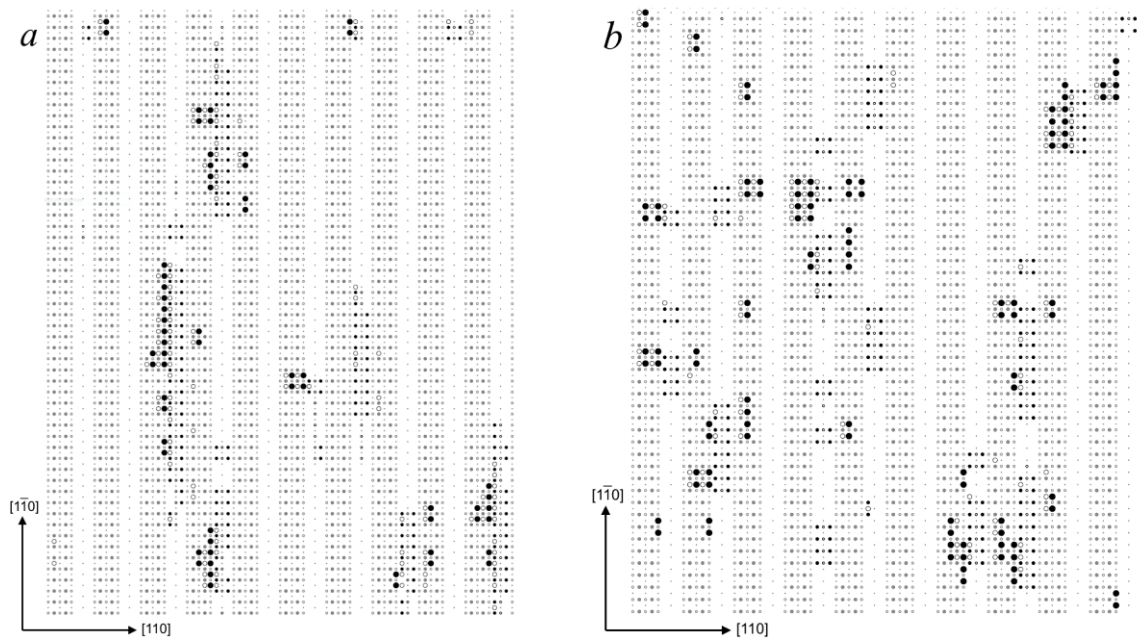


**Figure 1.** Island morphology in a simulation area of 160 Å × 200 Å after deposition of 0.1 ML GaAs at $T = 580°C$, $v = 0.1$ nm/s: *a*) $J_{As/Ga} = 3$ *b*) $J_{As/Ga} = 40$. The open circles represent Ga atoms and the filled circles As atoms. The substrate is marked in grey and deposited material in black.

Figure 1 shows that V/III flux ratio $J_{As/Ga}$ increase leads to larger surface scattering of islands and smaller anisotropy of their form. At higher V/III flux ratio we observe a large number of conglomerates consisting of several atoms and being nucleation centers of big islands.

In order to estimate gallium arsenide growing film characteristics quantitatively the island surface density (figure 2), average size (figure 3) and size distribution function (figure 4) have been calculated.

The island density rises with the coverage increase and reaches saturation after deposition of ~0.1 monolayer (ML) of GaAs (figure 2), in consistence with experiments [21]. The increase of V/III flux ratio leads to the island density growth. This is attributed to the suppression of Ga adatom diffusion caused by $As_2$ molecule concentration increase. Consequently, Ga adatoms become nuclei of new clusters rather than incorporate into stable islands. The saturation value of the island density at the substrate temperature $T = 580°C$, growth rate $v = 0.1$ nm/s and $J_{As/Ga} = 10$ is $2.2 \cdot 10^{12}$ cm$^{-2}$, in good agreement with experiments [22].
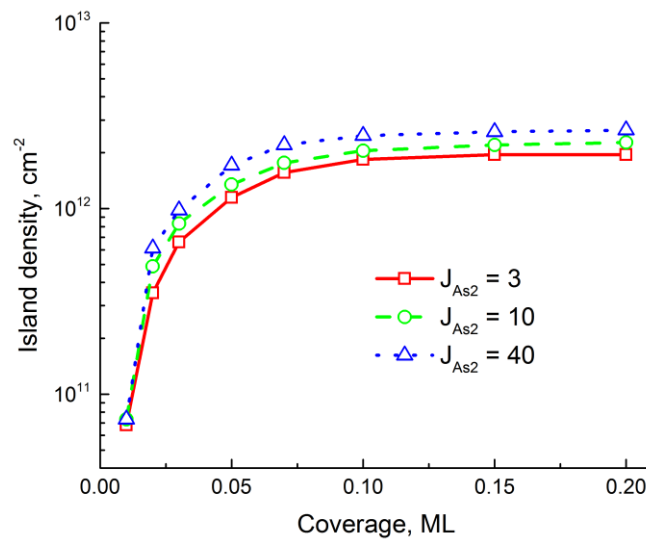
**Figure 2.** Island surface density as a function of coverage at $T = 580°C$, $v = 0.1$ nm/s and different V/III flux ratios.
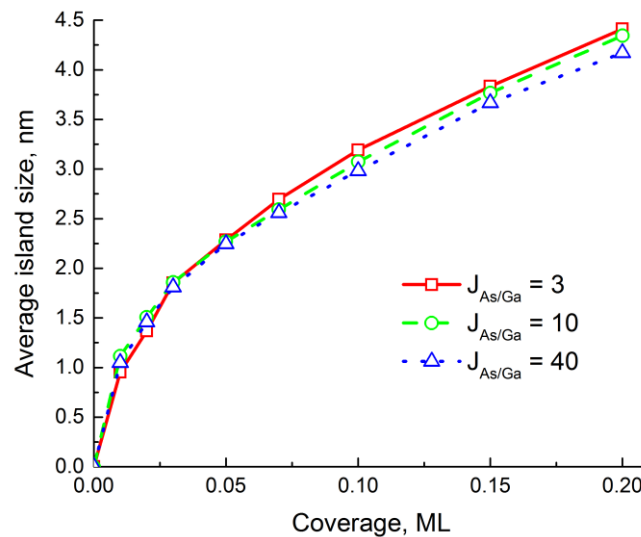


**Figure 3.** Island average size as a function of coverage at $T = 580°C$, $v = 0.1$ nm/s and different V/III flux ratios.

The average size of islands increases monotonically with the coverage increase (figure 3). However, V/III flux ratio increase leads to the slight decrease of island size due to above reasons. The alteration of the size is less significant than that of the density. It is connected with the consumption of deposited material for the island growth at any V/III flux ratio whereas the nuclei concentration remains the same value after a certain moment. The average island size at $T = 580°C$, $v = 0.1$ nm/s and coverage $\theta = 0.2$ ML is equal to ~4 nm that corresponds to experimental data [22].
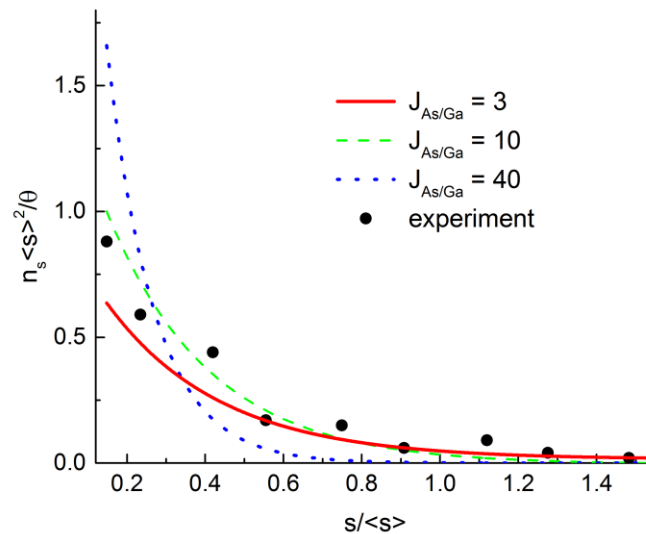
**Figure 4.** Island size distribution function at $T = 580°C$, $v = 0.1$ nm/s
and different V/III flux ratios. Experimental data are taken from [13].

The simulation size distribution function is based on the concept [23] that the number density $n_s$ of islands consisting of $s$ atoms can be stated as:

$$n_{\mathrm{s}} = \frac{\theta}{<s>^2} f(s/<s>), \qquad (4)$$

where $<s>$ is the average size of islands formed at $J_{As/Ga} = 10$; $f$ is a scaling function. The form of the distribution for the GaAs(001) surface shows that islands have a critical size of one atom and grow by gallium dimer capture [21] in contrast with (110) and (111)A surfaces [24]. The simulation size distribution is in good quantitative agreement with the experimental dependence for $J_{As/Ga} = 10$ [13]. Figure 4 shows that V/III flux ratio increase narrows the distribution with its shift to the small-size region. It is mainly due to the increase of arsenic molecule surface concentration and respective formation of small unstable clusters. At the same time islands are distributed quite uniformly along the size axis at small flux ratio $J_{As/Ga} = 3$ as adatoms can form quite many big islands along with little clusters.

## 4. Conclusion
In summary, the present simulation makes it possible to investigate GaAs epitaxial growth mechanisms thoroughly and take into account V/III flux ratio influence on the nucleating island characteristics. The flux ratio alteration does not affect the average island size significantly but can change the nucleation mechanisms and island morphology in whole. At large V/III flux ratios the density of little clusters increases whereas there is more homogeneous island size distribution on the surface at small V/III flux ratios. Being able to control nucleating island characteristics, V/III flux ratio is very important technological parameter of molecular beam epitaxy method.

## 5. References

[1]  LaBella V P, Bullock D W, Ding Z, Emery C, Harter W G and Thibado P M 2000 *J. Vac. Sci. Tech.* A **18** 1526
[2]  Ledentsov N N *et al.* 1996 *Sol. St. Electron.* **40** 785
[3]  Riel B J, Hinzer K, Moisa S, Fraser J, Finnie P, Piercy P, Fafard S and Wasilewski Z R 2002 *J. Cryst. Growth* **236** 145
[4]  Morgan C G, Kratzer P and Scheffler M 1999 *Phys. Rev. Lett.* **82** 4886
[5]  Murdick D A, Wadley H N G and Zhou X W 2007 *Phys. Rev.* B **75** 125318
[6]  Shiraishi K and Ito T 1998 *Phys. Rev.* B **57** 6301
[7]  Amrani A, Djafari Rouhani M and Mraoufel A 2011 *Appl. Nanosci.* **1** 59
[8]  Kangawa Y, Ito T, Taguchi A, Shiraishi K, Irisawa T and Ohachi T 2002 *Appl. Surf. Sci.* **190** 517
[9]  Daweritz L and Ploog K 1994 *Semicond. Sci. Tech.* **9** 123
[10] Foxon C T and Joyce B A 1977 *Surf. Sci.* **64** 293
[11] Tok E S, Neave J H, Zhang J, Joyce B A and Jones T S 1997 *Surf. Sci.* **374** 397
[12] Kley A, Ruggerone P and Scheffler M 1997 *Phys. Rev. Lett.* **79** 5278
[13] Avery A R, Dobbs H T, Holmes D M, Joyce B A and Vvedensky D D 1997 *Phys. Rev. Lett.* **79** 3938
[14] Nishinaga T and Shen X Q 1994 *Appl. Surf. Sci.* **82-83** 141
[15] Nishinaga T 2004 *Prog. Cryst. Growth Charact. Mater.* **48-49** 104
[16] Higuchi Y, Uemura M, Masui Y, Kitada T, Shimomura S, Hiyamizu S 2003 *J. Cryst. Growth* **251** 80
[17] Tok E S, Neave J H, Allegretti F E, Zhang J, Jones T S and Joyce B A 1997 *Surf. Sci.* **371** 277
[18] Kratzer P and Scheffler M 2002 *Phys. Rev. Lett.* **88** 036102
[19] Garcia J C, Neri C and Massies J 1989 *J. Cryst. Growth* **98** 511
[20] Foxon C T and Joyce B A 1975 *Surf. Sci.* **50** 434
[21] Itoh M, Bell G R, Joyce B A and Vvedensky D D 200 *Surf. Sci.* **464** 200
[22] Itoh M, Bell G R, Avery A R, Jones T S, Joyce B A and Vvedensky D D 1998 *Phys. Rev. Lett.* **81** 633
[23] Evans J W and Bartelt M C 1994 *J. Vac. Sci. Technol.* A **12** 1800
[24] Ratsch C, Smilauer P, Zangwill A, Vvedensky D D 1995 *Surf. Sci.* **329** L599