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Kinetics of epitaxial formation of nanostructures by Frank–van der Merwe, Volmer–Weber and Stranski–Krastanow growth modes

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ABSTRACT

Nowadays, two-dimensional crystals (2D materials) and structures with quantum dots (0D materials) are considered as one of the most promising materials for electronics and photonics of the next generation. The basic method of synthesis of heterostructures with 2D and 0D structures is their self-induced formation during the molecular beam epitaxy. Three epitaxial growth modes are distinguished: Frank–van der Merwe, Volmer–Weber, and Stranski–Krastanow, that allow one to obtain multi-layered structures with 2D materials, quantum wells and quantum dots. In this work generalized kinetic model of epitaxial growth of nanostructures by all three mechanisms is presented. Comparison of various growth modes is conducted and their peculiarities are pointed out. Ways to control the properties of obtained 2D and 0D nanostructures are proposed with the help of the established model.

1. Introduction

Nowadays, two-dimensional crystals (2D materials) and structures with quantum dots (0D materials) are considered as one of the most promising materials for electronics and photonics of the next generation [1]. Unique properties of 2D materials make it possible to create on their basis devices of a new generation: topological transistors, high-sensitive gas sensors, energy-intensive sources of power, thermoelectric generators, and quantum computers [2,3]. Meanwhile, nanoheterostructures with quantum dots can be used to create highly efficient photodetectors, solar cells and light-emitting devices [4]. Moreover, these structures are actively used for surface modification and creation of various coatings [5]. They may be adopted, for example, for creation of textured surfaces, anti-reflection coatings or realization of photonic crystals and microresonators [6,7].

The basic method of synthesis of heterostructures with 2D and 0D structures nowadays is their self-induced formation during the molecular beam epitaxy. For various applications in modern industries it is essential to create heterostructures with 2D layers and 0D quantum dots with different properties that are defined by such parameters as thickness and roughness of 2D layers, elastic strain distribution, presence or absence of defects, average size and surface density of quantum dots [1,2,5].

Three epitaxial growth modes are distinguished: so called Frank–van der Merwe, Volmer–Weber, and Stranski–Krastanow [8,9].

The Frank–van der Merwe growth mechanism is observed in auto-epitaxial systems and heteroepitaxial systems with very low lattice mismatch between the deposited material and the substrate. In this case a film grows by two-dimensional mechanism either by emergence of two-dimensional islands, or due to formation of monoatomic steps [9]. This mode is used for epitaxial synthesis of 2D materials (for instance, silicene, germanene or stanene) or multi-layered structures with quantum wells [1,2,5].

In heteroepitaxial systems with non-zero lattice mismatch layer-by-layer growth occurs at the initial stages. However, thicker deposited layer has greater elastic energy. Then a tendency appears that aimed to reduce elastic strain by virtue of nucleation of three-dimensional islands. In these systems the Stranski–Krastanow growth mode is realized. As a result, 3D clusters form on the substrate covered by thin wetting layer [9,10].

Finally, the Volmer–Weber mechanism is typical for highly-mismatched systems, and island growth takes place directly on the substrate without formation of the wetting layer [10].

These three mechanisms allow researchers to obtain all the spectrum of low-dimensional structures: multi-layered structures with 2D materials, quantum wells and quantum dots.

In spite of the fact that the processes of the epitaxial formation of two-dimensional layers and quantum dots have been actively studied

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for a long time, there are still many blank spots in this issue, especially concerning the theoretical description of the processes occurring at different stages of the growth of two-dimensional structures and nanoislands. New experimental works are constantly appearing that reveal unexpected effects during epitaxy in various modes [11–13].

Nevertheless, for the successful application of structures with 2D materials and quantum dots, it is necessary to be able to predict the dependences of the thickness and roughness of two-dimensional layers, elastic strain distribution, surface density, average size, and size distribution function of islands in an array of nanoislands on the conditions of their synthesis by molecular beam epitaxy, such as temperature, growth rate, and amount of deposited material.

In this work generalized kinetic model of epitaxial growth of nanostructures by all three mechanisms is presented. Comparison of various growth modes is conducted and their peculiarities are pointed out. Ways to control the properties of obtained 2D and 0D nanostructures for the purposes of surface modification, deposition of coatings and creation of device-oriented structures are proposed with the help of the established model.

2. Theory

We will consider the Volmer–Weber mechanism as a limiting case of the Stranski–Krastanow growth when the critical thickness of the wetting layer tends to zero, and growth according to the Frank–van der Merwe mechanism as a limiting case of the Stranski–Krastanow growth with an unlimited increase in the critical thickness of transition from two-dimensional to three-dimensional growth. The nucleation of islands practically without the formation of an intermediate two-dimensional layer occurs with an increase in the mismatch between the lattice constants of the deposited material and the substrate. And the transition to pure two-dimensional growth by the Frank–van der Merwe mechanism is realized at close values of the lattice constants. Thus, by varying the value of the mismatch parameter, we can sequentially consider all possible modes of epitaxial growth.

Similar to considering the formation of islands on a wetting layer in the Stranski–Krastanow mode [14–16] for establishment of theoretical models of growth of two-dimensional layers and quantum dots by the Frank–van der Merwe and Volmer–Weber mechanisms we will find, first of all, change in free energy of the system during the formation of an island.

For simplicity we will consider islands with the shape of a spherical cap with the base diameter L and height H (besides that, this shape is observed experimentally, for example, in the Ge/SiO_2/Si system [17–19]).

The expression for the change in free energy upon nucleation of an island containing i atoms may be written in the following form:

$$\Delta F(i) = \Delta F_{surf}(i) + \Delta F_{elas}(i) + \Delta F_{attr}(i). \tag{1}$$

This expression takes into account three competing factors that influence island formation: the change in surface energy ΔF_{surf}, elastic strain relaxation ΔF_{elas}, and decrease in attraction of atoms to the substrate ΔF_{attr}.

The change in surface energy ΔF_{surf} is equalto the difference between the energy of external surface of an island and the energy of a 2D layer with the area equal to the area of an island base [9,16], and may be written as a function of a number of atoms in an island i:

$$\Delta F_{surf}(i) = \frac{\pi}{4}[\gamma(4\kappa^2+1) - \gamma_0]\alpha^2 l_0^2 i^{2/3}, \tag{2}$$

where γ_0 and γ are specific surface energies of the base and free surface of an island, $\kappa = H / L$ is the island height to diameter ratio, l_0 is the distance between atoms on the surface of the substrate, d_0 is the height of one monolayer (ML) of deposited material, and α is the geometrical factor, predetermined by the shape of an island:

$$\alpha = \left[\frac{24 d_0}{\pi\kappa l_0(4\kappa^2+3)}\right]^{1/3}. \tag{3}$$

The second term ΔF_{elas} in expression (1) is equal to the difference in elastic energies of i atoms in 2D layer and in an island with respect to the relaxation of elastic strain [9,16]:

$$\Delta F_{elas}(i) = -(1-Z)\lambda\varepsilon_0^2 l_0^2 d_0 i, \tag{4}$$

where Z is the Ratsch–Zangwill elastic strain relaxation coefficient, showing the decrease in elastic energy of an island with given geometry [20,21], λ is the elastic modulus of the deposited material, and ε_0 is the lattice mismatch between deposited material and the substrate.

The change in the wetting energy ΔF_{attr} amounts to the difference of energies of attraction to the substrate of atoms in 2D layer and in an island and is defined as

$$\Delta F_{attr}(i) = \frac{\Psi_0}{d_0}\exp\left(-\frac{h}{k_0 d_0}\right) l_0^2 d_0 i, \tag{5}$$

where Ψ_0 is the wetting energy density, h is the thickness of two-dimensional layer, and k_0 is the relaxation coefficient (of order of unity), characterizing exponential decrease in the attraction energy with the thickness typical for semiconductor materials [10,11]. Dimensionless value $\zeta = (h / h_{eq} - 1)$ is called 2D layer superstress and characterizes the level of meta-stability of the system. Here h_{eq} is the equilibrium thickness of two-dimensional layer [9,22]:

$$h_{eq} = k_0 d_0 \ln\left(\frac{\Psi_0}{d_0(1-Z)\lambda\varepsilon_0^2}\right). \tag{6}$$

Thus, the dependence of free energy function ΔF on the number of atoms in an island, expressed in the thermal units $k_B T$ (where k_B is the Boltzmann's constant, T is the substrate temperature), may be written in the following form:

$$\Delta F(i) = Ai^{2/3} - B\zeta i, \tag{7}$$

where

$$A = \frac{\pi}{4}[\gamma(4\kappa^2+1) - \gamma_0]\frac{\alpha^2 l_0^2}{k_B T}, \tag{8}$$

$$B = \frac{(1-Z)\lambda\varepsilon_0^2 l_0^2 d_0}{k_B T}\ln\left\{\frac{\Psi_0}{d_0(1-Z)\lambda\varepsilon_0^2}\right\}. \tag{9}$$

From the expression (7) the critical number of atoms in an island i_c may be found:

$$i_c = \left(\frac{2A}{3B\zeta}\right)^3. \tag{10}$$

For $i = i_c$ the function $\Delta F(i)$ reaches its maximum. After that activation barrier of nucleation $\Delta F(i_c)$ is calculated. Then, using the Zeldovich formula, the rate of islands nucleation is evaluated [23,24]:

$$I = \frac{W^+(i_c)}{l_0^2}\sqrt{\frac{\Delta F''(i_c)}{2\pi}}\, e^{-\Delta F(i_c)}, \tag{11}$$

where $W^+(i_c)$ is the rate of atoms incorporation into an island of critical size.

To find $W^+(i)$ for islands with the shape of a spherical cap with base perimeter πL, it is necessary to use the following expression:

$$W^+(i) = \frac{\pi D}{l_0^2}\frac{\alpha B(\zeta+1)}{\nu} i^{1/3}, \tag{12}$$

where D is the coefficient of atoms diffusion on the surface [9,25], and ν is the cut-off parameter of the elastic strain field that shows how far the elastic strain field propagates from the border of an island and has typical values of order of 10 [23]. Hereafter, for islands nucleation rate as a function of the superstress the following formula will be obtained:

$$I(\zeta) = \frac{a}{\tau l_0^2}\zeta(\zeta+1)e^{-\Delta F(i_c)}, \tag{13}$$

where

$$a = \frac{3B}{4\sqrt{\pi A}}, \tag{14}$$

$$\tau = \frac{3l_0^2\nu}{2\pi\alpha BD}. \tag{15}$$

Critical thickness of transition from 2D to 3D growth h_c is found by solving the following equation for the critical superstress $\zeta_c = (h_c / h_{eq} - 1)$ [9,16]:

$$\frac{4}{3\sqrt{\pi}}\frac{h_{eq}}{d_0}\frac{\zeta_c}{2a(\zeta_c+1)F(\zeta_c)}\left[\frac{2F(\zeta_c)}{\zeta_c^2}\frac{\tau}{t_{eq}}\right]^{\frac{5}{2}}\exp[F(\zeta_c)] = 1, \tag{16}$$

where $t_{eq} = h_{eq} / V$ is the time of growth of a 2D layer with equilibrium thickness, and V is the deposition rate.

All further calculations of the kinetics of the formation of quantum dots by the Volmer–Weber mechanism are carried out similarly to the case of growth according to the Stranski–Krastanow mode [9,26], but with parameters recalculated according to the above formulas.

3. Results and discussion

When modeling the dependences of the parameters of two-dimensional layers and quantum dot array on the lattice mismatch between the deposited material and the substrate ε_0 we used the following values for the parameters of the theoretical model: $l_0 = 0.4$ nm, $d_0 = 0.15$ nm, $\lambda = 1.5 \cdot 10^{12}$ dyn/cm^2, $\gamma = \gamma_0 = 1000$ erg/cm^2, $\Psi_0 = 500$ erg/cm^2, $\kappa = 0.1$, $k_0 = 1$, $\nu = 10$, $Z = 0.7$, $D(T) = 10^{-4}\cdot\exp(-1.2 / k_BT)$ cm^2/s. These values approximately correspond to the parameters for the case of germanium deposition on a silicon surface [16,23,26].

Fig. 1 shows the dependence of the critical size of an island calculated with the help of expression (10) on the value of lattice mismatch for the growth temperature $T = 450$ °C.

It is seen from Fig. 1 that with increase in ε_0 the critical size of an island i_c decreases, that means the lowering of activation barrier of nucleation $\Delta F(i_c)$ and gradual shift from the growth of two-dimensional layers (the Frank–van der Merwe mechanism) to the nucleation of islands directly on the surface of the substrate (the Volmer–Weber mechanism). Areas corresponding to all three possible growth modes are formally outlined in Fig. 1.

The critical thickness of the two-dimensional layer was estimated by solving Eq. (16). Calculated values of the critical thickness of the transition from 2D to 3D growth as function of lattice mismatch between the deposited material and the substrate are shown in Fig. 2.

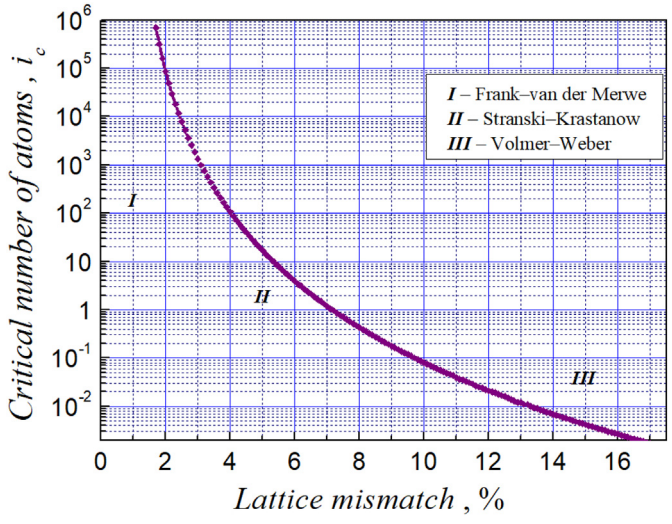

Fig. 1. Dependence of the critical number of atoms in an island on the value of lattice mismatch for the growth temperature $T = 450$ °C.

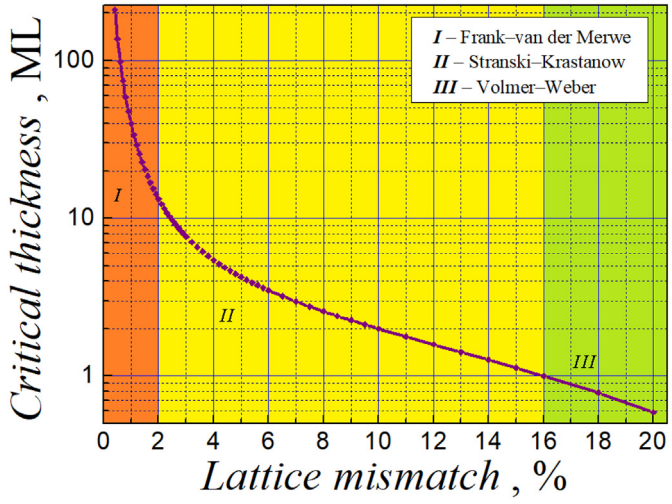

Fig. 2. Dependence of the critical thickness of the transition from 2D to 3D growth on the value of lattice mismatch for the growth temperature $T = 450$ °C.

For the thicknesses of the two-dimensional layer $h < h_c$ a layer-by-layer growth of one material on the surface of another is realized. With the increase in the deposited material thickness $h > h_c$ a transition from layer to island growth is observed, which reduces the energy of the system due to the relaxation of elastic strain [9,23]. In accordance with Fig. 2, the higher the value of lattice mismatch, the earlier a nucleation of 3D islands begins.

Using Fig. 2, the borders of ranges of ε_0 corresponding to three modes of epitaxial growth may be determined more sharply. The range of values $\varepsilon_0 < 2\%$ is traditionally referred as the Frank–van der Merwe growth mode in low-mismatched epitaxial systems. The range of values of mismatch parameter ε_0 where $h_c < 1$ ML corresponds to the growth by the Volmer–Weber mechanism. In this case nucleation of 3D islands takes place nearly immediately after the start of deposition, during formation of the first monolayer. Finally, the range of intermediate values of mismatch parameter ε_0 is characterized by growth in the Stranski–Krastanow mode, when the nucleation of 3D islands begins after reaching by 2D wetting layer of certain critical thickness.

From the point of view of device applications, the most interesting are the dependences of the parameters of the formed structures on the conditions of their synthesis. Fig. 3 shows the calculated dependences of the average size and surface density of islands on the lattice

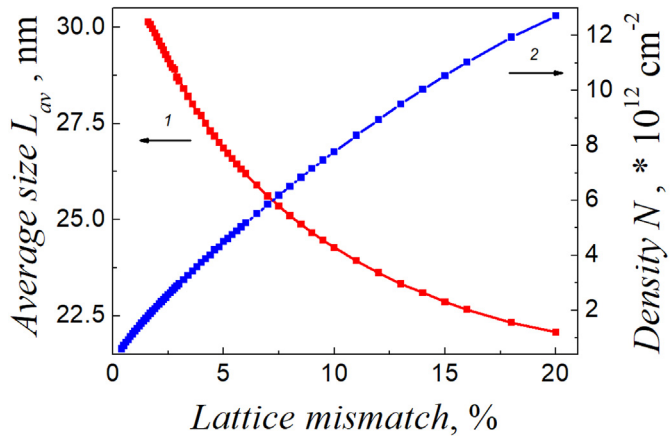

Fig. 3. Dependences of average size (*1*) and surface density (*2*) of islands on the value of lattice mismatch for the growth temperature $T = 450$ °C.

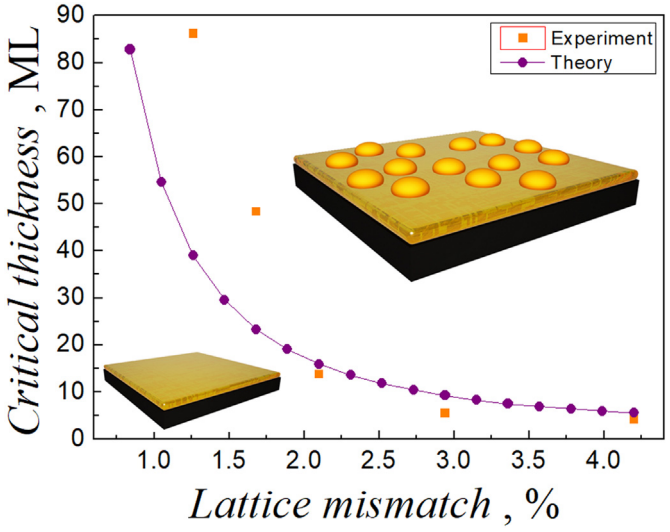

Fig. 4. Comparison of the experimental [32] and calculated values of the critical thickness of transition to 3D growth for the substrate temperature $T = 400$ °C and various mismatch parameters (contents x) in the Ge_xSi_{1-x}/Si system.

mismatch for the growth temperature $T = 450$ °C and growth rate $V = 0.1$ ML/s.

In accordance with Fig. 3 the average size of nanoclusters decreases with an increase in the mismatch parameter, while the surface density of islands, on the contrary, increases. Therefore, to achieve the highest densities of quantum dots of the minimum size, the Volmer–Weber growth mode should be used, which is implemented in highly mismatched systems.

As a real material system for comparing the results of numerical simulations and experimental data, we will consider the processes of the formation of two-dimensional layers and quantum dots during molecular beam epitaxy in the germanium-silicon system. The Ge/Si material system is currently one of the most promising for the development of semiconductor nanoelectronics and nanophotonics. Structures with quantum dots of germanium in silicon are of interest from the point of view of creating high-speed transistors, photonic crystals, photodetectors, and solar cells [7,27]. The main method to create nanoislands is their self-organization during molecular beam epitaxy [5].

Historically, the first to use for germanium epitaxy were clean silicon surfaces with the crystallographic orientations (001) and (111) [28,29]. The lattice mismatch for germanium on a clean silicon surface is 4.2%. Therefore, these systems are characterized by the Stranski–Krastanow growth mechanism, when a two-dimensional wetting layer of the deposited material is formed at the first stage, and then 3D islands nucleate [5,30,31].

In the case of deposition of layers of Ge_xSi_{1-x} solid solution on a clean silicon surface, for the Ge contents from $x = 1$ to $x = 0$ the lattice mismatch linearly decreases from 4.2% to zero [32,33]. So, this system is convenient for observation of gradual increase in the critical thickness of the transition to 3D growth and moving from the Stranski–Krastanow mechanism to the Frank–van der Merwe growth mode.

Dependence of the critical thickness of the transition from 2D to 3D growth on the value of mismatch parameter (that is on the content x) in the Ge_xSi_{1-x}/Si system for the temperature $T = 400$ °C wascalculated (Fig. 4). Results of calculations show good agreement with the experimental data [32].

And finally, for the germanium/silica system, the lattice mismatch is 15.2%, which determines the realization of the Volmer–Weber growth mode in this system [34]. This system is of particular interest to researchers, since they often face the task of minimizing the size of the synthesized islands and increasing their density. It is in this system that islands with an extremely high density of up to 10^{12}–10^{13} cm^{-2} and sizes < 10 nm were obtained [17–19].

Calculations with the presented model give for the Ge/SiO_2 system at the growth temperature $T = 400$ °C the surface density of quantum dots $N \approx 1.5 \cdot 10^{12}$ cm^{-2} and the average size of quantum dots $L_{av} \approx 9$ nm. These values of the surface density and average size of islands are in good agreement with the experimental data on the growth of germanium islands on the oxidized silicon surface [17–19].

Thus, varying the value of the mismatch parameter, we can sequentially examine all possible modes of epitaxial growth, and the approximation of the consideration of the Frank–van der Merwe and Volmer–Weber mechanisms as the limiting cases of the growth according to the Stranski–Krastanow mode is reasonable.

4. Conclusions

As a result, in the presented work the main features of the epitaxial nucleation and growth of two-dimensional layers and quantum dots by the Frank–van der Merwe, Stranski–Krastanow, and Volmer–Weber mechanisms are described. The generalized kinetic model of the growth of 2D structures and 3D islands by various mechanisms is developed. Comparison of different growth modes is conducted and their peculiarities are pointed out. Ways to control the properties of obtained 2D and 0D nanostructures for the purposes of surface modification, coatings deposition and creation of device-oriented structures are proposed with the help of the established model. The dependences of the average size and surface density of quantum dots on the parameters of their synthesis and the value of the lattice mismatch between the deposited material and the substrate are obtained. Good agreement between the calculated values and experimental results confirms the applicability of the proposed model.

Author contributions

Kirill A. Lozovoy – Methodology, Writing - Original Draft, Funding acquisition.

Alexander G. Korotaev – Resources, Writing - Review & Editing.

Andrey P. Kokhanenko – Supervision, Formal analysis.

Vladimir V. Dirko – Software, Visualization.

Alexander V. Voitsekhovskii – Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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