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Generalized Muller–Kern formula for equilibrium thickness of wetting layer with respect to the dependence of surface energy of island facets on the thickness of 2D layer

Kirill A. Lozovoy*, Andrey P. Kokhanenko, Alexander V. Voitsekhovskii

National Research Tomsk State University, Department of Radiophysics, 36 Lenin av.,
Tomsk 634050, Russian Federation

*e-mail: lozovoymailbox@gmail.com

Abstract: Experimental results indicate particular importance of such value as equilibrium thickness of wetting layer during epitaxial growth according to the Stranski–Krastanow mechanism in systems with lattice mismatch. In this paper the change in free energy during transition of atoms from wetting layer to the island in such systems is considered. Recent experimental results also show that surface energy of island's facets depends on the thickness of deposited material. So, in this paper the equilibrium thickness of wetting layer, at which transition from 2D to 3D growth becomes energetically favorable, is calculated in the assumption of the dependence of specific energy of island's facets on the wetting layer thickness. In this approximation new generalized Muller–Kern formula is obtained. As an illustration of proposed method, an example of numerical calculation according to the new formula for the material system of germanium on silicon (001) surface is given. The result for the found equilibrium thickness of wetting layer is rather unexpected since it differs from the value obtained in the bounds of traditional Muller–Kern model.

Keywords: Molecular beam epitaxy; Self-organization; Stranski–Krastanow growth; Free energy; Equilibrium thickness of wetting layer

Introduction. Heterostructures with quantum dots have firmly occupied their place as one of the basic materials for creation of optoelectronic devices such as solar cells and photodetectors of visible and infra-red range.[1–3]

One of the main methods of such heterostructures creation is their spontaneous formation during molecular beam epitaxy. Self-organization of quantum dots is possible only when there is a lattice constants' mismatch between deposited material and substrate. In this case quantum dots grow in the so called Stranski–Krastanow mode. In this mechanism initially layer-by-layer growth is realized. Then, after the thickness of deposited material reaches certain critical value, the transition from 2D to 3D growth occurs. The moment of this transition and conditions for its realization are of the paramount importance for the whole Stranski–Krastanow growth.[4]

Results of theoretical and experimental investigations of recent years shows significance of such value as equilibrium thickness of transition from 2D to 3D growth during epitaxy of thin semiconductor layers in the Stranski–Krastanow growth mode.[5–9]

At the same time equilibrium and critical thicknesses of wetting layer should be distinguished as there is a fundamental difference between them. The former value determines the thickness of wetting layer at which phase transition from 2D to 3D growth becomes energetically feasible. It is specified by energetic parameters of materials system. The latter one characterizes the wetting layer thickness at which nucleation of 3D islands progresses most intensively. It may be registered experimentally by change in picture of high energy electron diffraction. It also may be found from the equation of kinetic balance of processes of atoms' deposition on the substrate and consumption of wetting layer atoms by the growing islands. It depends, in particular, on the material's deposition rate. Thus, the equilibrium thickness of wetting layer is purely thermodynamic value, while the critical thickness of transition from 2D to 3D growth is determined also by kinetic parameters of growth. However, there is a deep connection between them and calculations of critical thickness are impossible without knowing the equilibrium one.

The transition from 2D to 3D growth during Stranski–Krastanow mode as a classical question has been investigated by some methods in the past few years. For example, Li et al.[10] have established a general thermodynamic model to address the self-assembly of islands by taking into account the size-dependent surface and interface energies and the interactions between islands and developed this approach in further papers.[11, 12] The proposed model explains the physical mechanism of quantum dots formation and predicts two critical sizes in the quantum dots growth: critical sizes of quantum dots formation and of their stable array. This models also suggests the critical volume of quantum dots when the transition from pyramids to dome-shaped islands takes place. The method based on chemical potential has also been used to study the critical thickness of wetting layer during strain-induced growth on surfaces with nanoscale curvature.[13] A review paper[14] for the thermodynamic theory of growth of nanostructures has also discussed the growth of islands in Stranski–Krastanow mode.

Theoretical consideration of just equilibrium thickness is promising for explanation of subcritical quantum dots formation (in the range of wetting layer thicknesses between equilibrium and critical ones) and for interpretation of appearance of elongated quantum dots with a large length to width ratio, observed in the recent experiments after continuous low-temperature exposition of the germanium layer of subcritical thickness on a silicon surface.[5–7] In addition, examination of equilibrium thickness and factors that determine its value is necessary for deep insight in processes occurring during formation of semiconductor nanoislands in the Stranski–Krastanow mechanism and for estimation of kinetic values that characterize these processes.

Thus, in this paper a new look on the known theory and a new approach to the problem are proposed. It allows one to make more accurate assessment of the equilibrium thickness.

Theory. During theoretical description of kinetics of transition from 2D to 3D growth, first of all, change of free energy during transition of atoms from wetting layer to island is considered. For this purpose, for example, the Muller–Kern model may be used.[15, 16] We will consider growth of island, containing i atoms and situated on the surface of wetting layer with the thickness h. In this case competing factors are change of free energy due to increase of surface energy ΔF_{surf}, elastic strain relaxation ΔF_{elas} and reduction in the attraction of atoms to substrate ΔF_{attr}.

The change of free energy during transition of atoms from wetting layer to island is therefore may be written as a sum of three summands:[17]

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

In this paper we will not consider the change of free energy due to formation of additional edges of island[18] as it is not essential for our analysis.

Change in surface energy during transition of atoms from wetting layer to island represents the difference between surface energy of island's facets and surface energy of wetting layer with the area equal to one of island's base:[19]

$$\Delta F_{surf}(i) = \frac{(\gamma(\varphi)/\cos\varphi - \gamma(0))}{k_B T}\alpha^2 l_0^{\,2} i^{2/3}, \tag{2}$$

where $\gamma(0)$ and $\gamma(\varphi)$ are the specific surface energies of base and lateral faces of the pyramid, φ is the angle between the lateral face and the base of the island, l_0 is the mean distance between atoms on the surface, k_B is Boltzmann's constant, T is substrate temperature, and α is the geometrical factor, which depends on the island's shape and is given in case of pyramidal quantum dots with square base as:

$$\alpha = \left(\frac{6 d_0 ctg\varphi}{l_0}\right)^{1/3}, \tag{3}$$

where d_0 is the height of the monolayer (ML) of deposited material.

The second term in (1) equals to the difference of elastic energies of i atoms in island and wetting layer with respect to the relaxation of elastic strain:[19]

$$\Delta F_{elas}(i) = -[1 - Z(\varphi)]\lambda\varepsilon_0^2 \frac{l_0^{\,2} d_0}{k_B T} i, \tag{4}$$

where $Z(\varphi)$ is the coefficient of elastic energy relaxation,[20–21] λ is the material's modulus of elasticity, ε_0 is the lattices mismatch.

Change in the wetting energy in (1) equals to the difference between energies of atoms' attraction to substrate at the top of wetting layer and in the island. Following Muller and Kern it may be found as[19]

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

where Ψ_0 is the wetting energy density on the surface of the substrate, k_0 is the relaxation coefficient.

Then the dependence of free energy function ΔF on the number of atoms in island without taking into account additional energy of edges is defined as:[19]

$$\Delta F(i) = \frac{(\gamma(\varphi)/\cos\varphi - \gamma(0))}{k_B T}\alpha^2 l_0^{\,2} i^{2/3} - \left\{[1 - Z(\varphi)]\lambda\varepsilon_0^2 - \frac{\Psi_0}{d_0}\exp\left(-\frac{h}{k_0 d_0}\right)\right\}\frac{l_0^{\,2} d_0}{k_B T} i. \tag{6}$$

The expression in braces in equation (6) equals to the difference of change of free energy due to elastic strain relaxation and due to the difference in the attraction energies according to the Muller–Kern model. This expression equals to zero at certain value of wetting layer thickness $h = h_{eq}$, which is called equilibrium thickness. At $h = h_{eq}$ formation of islands becomes energetically advantageous. Equating the expression in braces in equation (6) to zero

$$[1 - Z(\varphi)]\lambda\varepsilon_0^2 - \frac{\Psi_0}{d_0}\exp\left(-\frac{h_{eq}}{k_0 d_0}\right) = 0 \tag{7}$$

and expressing h one can obtain so called Muller–Kern formula for the equilibrium thickness of wetting layer:

$$h_{eq} = d_0 k_0 \ln\left\{\frac{\Psi_0}{d_0[1 - Z(\varphi)]\lambda\varepsilon_0^2}\right\}. \tag{8}$$

The wetting energy density on the surface of the substrate Ψ_0 is determined by the balance of surface energy of substrate and deposited material:[16, 22]

$$\Psi_0 = \gamma_s - \gamma_d - \gamma_{s-d}\,, \tag{9}$$

where γ_s is the specific surface energy of the substrate, $\gamma_d \equiv \gamma(0)$ is the specific surface energy of deposited material, and $\gamma_{s\text{-}d}$ is the specific surface energy of the interface between them.[23, 24] Usually, $\gamma_{s\text{-}d}$ is significantly smaller than γ_s and γ_d[22, 25], so we will neglect this small contribution in our further speculations.

Traditionally it was considered that specific surface energy of facets has a constant value. However, recent theoretical investigations[26–30] have shown that surface energy (in particular, for silicon-germanium system) depends on the thickness of deposited material. The value of surface energy of facets decreases from its maxima at h = 0 ML and reaches saturation with the wetting layer thickness increase. In our paper we are taking into account this effect.

According to work[30] expressions for dependence of specific surface energies of island's base and facet may be written as following:

$$\gamma(0,h) = \left[\gamma(0,0) - \gamma(0,\infty)\right] e^{-B_0 \frac{h}{d_0}} + \gamma(0,\infty)\,, \tag{10}$$

$$\gamma(\varphi,h) = \left[\gamma(\varphi,0) - \gamma(\varphi,\infty)\right] e^{-B_\varphi \frac{h}{d_0}} + \gamma(\varphi,\infty)\,, \tag{11}$$

where $\gamma(\varphi, 0)$ and $\gamma(\varphi, \infty)$ are the specific surface energy of facet with the contact angle φ on the surface of pure silicon (without wetting layer) and on the surface of pure strained germanium (infinite wetting layer thickness) respectively, B_0 and B_φ are dimensionless parameters that characterize the rate of specific surface energy change with the deposited material thickness.

According to equation (9) with the specific surface energies the wetting energy density Ψ_0 will be depend on the thickness of wetting layer too:

$$\Psi_0(h) = \gamma_s - \left[\gamma(0,0) - \gamma(0,\infty)\right] e^{-B_0 \frac{h}{d_0}} - \gamma(0,\infty)\,. \tag{12}$$

Hence, it is necessary to revise formula (6), so it takes into account the dependence of specific surface energy on the wetting layer thickness h:

$$\Delta F(i,h) = \frac{\left(\gamma(\varphi,h)/\cos\varphi - \gamma(0,h)\right)}{k_B T}\alpha^2 l_0^2 i^{2/3} - \left\{\left[1 - Z(\varphi)\right]\lambda\varepsilon_0^2 - \frac{\Psi_0(h)}{d_0}\exp\left(-\frac{h}{k_0 d_0}\right)\right\}\frac{l_0^2 d_0}{k_B T} i\,. \tag{13}$$

Since the wetting energy density Ψ_0 now depends on the wetting layer thickness, the Muller–Kern formula (8) will no longer be correct. To find equilibrium thickness h_{eq} it necessary now to represent the expression in braces in (13) in implicit form and to equal it to zero:

$$\left[1 - Z(\varphi)\right]\lambda\varepsilon_0^2 - \frac{1}{d_0}\left\{\gamma_s - \left[\gamma_0(0) - \gamma_\infty(0)\right] e^{-B_0 \frac{h_{eq}}{d_0}} - \gamma_\infty(0)\right\}\exp\left(-\frac{h_{eq}}{k_0 d_0}\right) = 0\,. \tag{14}$$

Thus, in order to find the equilibrium thickness of wetting layer it is necessary to solve transcendental equation (14) instead of equation (7).

Two more important parameters of classical nucleation theory are the critical number of atoms i_c in the island at which the function of free energy (13) reaches its maximum and the nucleation's activation barrier $\Delta F(i_c)$ (that is the value of this maximum). In order to define i_c, it is necessary to find the derivative for the free energy function (13) and equate it to zero:

$$\left.\frac{\partial \Delta F(i,h)}{\partial i}\right|_{h=const} = \\ = \frac{2}{3}\frac{\left(\gamma(\varphi,h)/\cos\varphi-\gamma(0,h)\right)}{k_B T}\alpha^2 l_0{}^2 i^{-1/3} - \left\{[1-Z(\varphi)]\lambda\varepsilon_0^2 - \frac{\Psi_0(h)}{d_0}\exp\left(-\frac{h}{k_0 d_0}\right)\right\}\frac{l_0{}^2 d_0}{k_B T} = 0 . \tag{15}$$

Solving equation (15) we will obtain the dependence of critical size of nucleus on the wetting layer thickness:

$$i_c(h) = \left(\frac{2\left(\gamma(\varphi,h)/\cos\varphi-\gamma(0,h)\right)\alpha^2}{3\left\{[1-Z(\varphi)]\lambda\varepsilon_0^2 - \frac{\Psi_0(h)}{d_0}\exp\left(-\frac{h}{k_0 d_0}\right)\right\}d_0}\right)^3 . \tag{16}$$

Results and discussion. We will consider as an example growth of quantum dots of germanium on silicon (001) surface. For this material system required thermodynamic parameters have the following values:[17, 19, 30] $l_0 = 0.395$ nm, $d_0 = 0.145$ nm, $\lambda = 1.27\cdot10^{12}$ dyn/cm^2, $k_0 = 0.8$, $\varepsilon_0 = 0.042$, $\varphi = 20°$, $Z(\varphi) = 0.7$, $\gamma_s = 1260$ erg/cm^2, $\gamma(0, 0) = 1450$ erg/cm^2, $\gamma(0, \infty) = 1000$ erg/cm^2, $\gamma(\varphi, 0) = 1440$ erg/cm^2, $\gamma(\varphi, \infty) = 920$ erg/cm^2, $B_0 = 1.02$, $B_\varphi = 0.85$.

Equation (14) has been solved numerically for material system Ge/Si(001) in order to find equilibrium wetting layer thickness h_{eq} for Stranski–Krastanow growth of quantum dots in this system. According to the Muller–Kern criterion[16] at wetting layer thicknessess $h < h_{eq}$ layer-by-layer growth takes place. If $h < h_{eq}$ a 2D to 3D transition is observed, which results in a reduction of free energy.[17] As a result for the equilibrium wetting layer thickness with the use of listed above parameters the value $h_{eq} = 2.5$ ML was obtained. This value corresponds to the results of a number experiments on growth of quantum dots in Ge/Si(001) system.[9, 24, 31–32] This value is also approved by appearance of elongated quantum dots with a large length to width ratio, observed in the experiments after continuous low-temperature exposition of germanium layer with the thickness of about 3 ML on a silicon surface.[5–8]

The obtained value significantly differs from the traditional value for equilibrium thickness of wetting layer of 3.0 ML that was obtained by the traditional Muller–Kern formula (8) and was widely used so far.[17–19, 33] For example, simplified thermodynamic and kinetic models were used to calculate free energy of stress-driven formation of quantum dots growth in Ge/Si(001) system and to calculate islands' nucleation rate and surface density.[17] But in order to facilitate calculations abstract cuboid-shaped quantum dots were considered (while quantum dots in this system have a shape of pyramids). More complex thermodynamic and kinetic models considering real shapes of nanoislands were developed in further works[18, 19, 33] and adopted to estimate free energy function, nucleation rate, surface density and size distribution function of quantum dots in the ensemble. However these models have not taken into account the thickness-dependent surface energy.

The found value of equilibrium thickness of wetting layer allows us to make a supposition that taking into account the dependence of surface energy of island's facets on the thickness of 2D layer will lead to considerable change in theoretical predictions concerning the dependencies of such parameters of quantum dots array as their surface density and size distribution function on growth conditions.

Figure 1 shows dependencies of free energy function (13) on number of atoms in the island for growth temperature $T = 470$ °C at various thicknesses of deposited germanium.

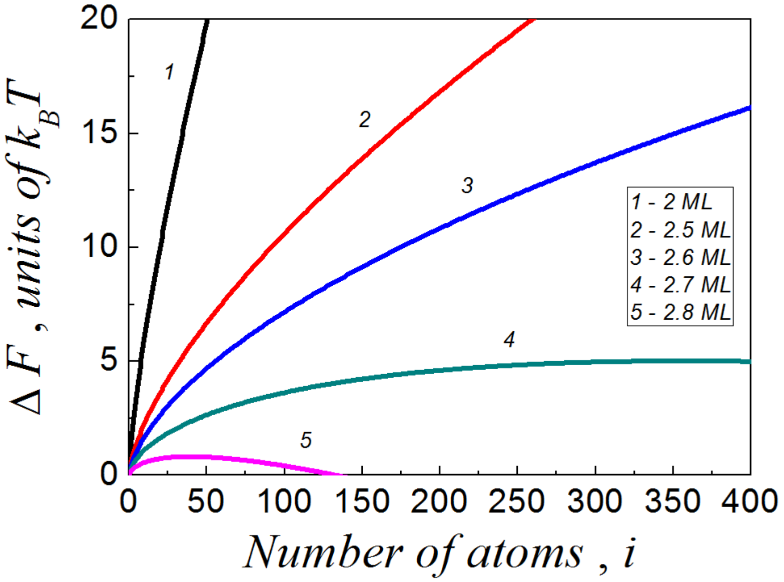

Fig. 1 Function of free energy at growth temperature $T = 470$ °C for various thicknesses h of Ge wetting layer.

For wetting layer thicknesses $h = 2.0$ ML and $h = 2.5$ ML (curves *1* and *2* in figure 1) free energy tends to infinity with the number of atoms in island increasing. It means that it is thermodynamically non-favorable to incorporate new atoms in the nanoclusters. On the contrary, for wetting layer thicknesses more than 2.5 ML (curves *3–5* in figure 1) function of free energy becomes bounded. There is a critical number of atoms in the island i_c after which free energy of the island decreases with the new atoms adding to the island.

Figure 2 represents the dependence of critical number of atoms in the island i_c on the Ge wetting layer thickness. In addition, figure 2 shows in more details the free energy as a function of number of atoms in the island for Ge wetting layer thicknesses $h = 2.7$ ML and $h = 2.8$ ML at growth temperature $T = 470$ °C.

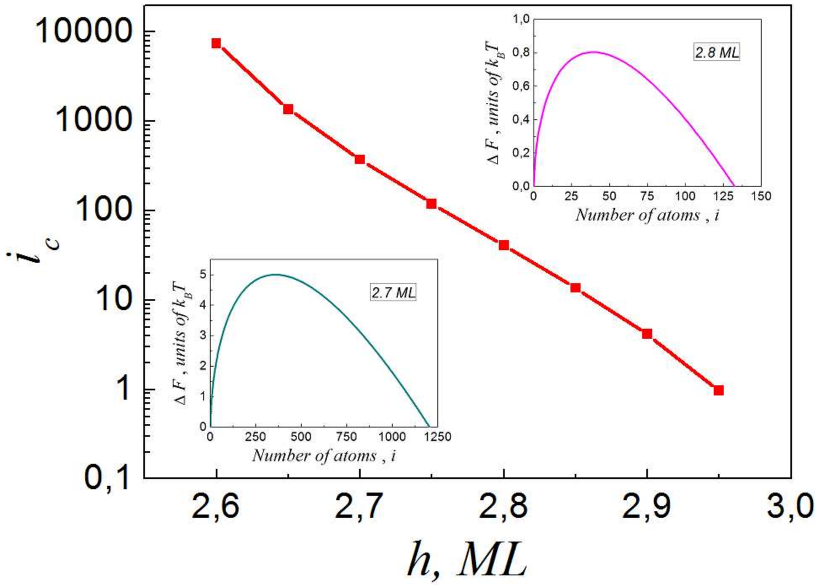

Fig. 2 Critical number i_c of atoms in the island as a function of Ge wetting layer thickness h at growth temperature $T = 470$ °C. Insets: the function of free energy at growth temperature $T = 470$ °C for Ge wetting layer thicknesses $h = 2.7$ ML and $h = 2.8$ ML.

Such non-monotonous character of free energy function is explained by the increasing of the surface energy of islands facets and consequent decrease in the elastic energy. So, as it was repeatedly stated in literature,[1, 2, 4, 11–12] the physical mechanism of the 2D to 3D transition is the balance between the thickness-dependent surface energy and the relaxation of the elastic strain caused by lattice parameters mismatch. After reaching a certain thickness of wetting layer the possibility of quantum dots formation on the surface of wetting layer realizes rather than keeping layer-by-layer 2D growth.[12]

Thereby, for small thicknesses of deposited germanium ($h < 2.5$ ML) free energy grows unrestrictedly. For wetting layer thicknesses $h > 2.5$ ML the maximum appears on the curves. This maximum corresponds to the potential barrier of nucleation. It is necessary to overcome this barrier for the subsequent 3D growth of the island. For example, at wetting layer thickness $h = 2.7$ ML the maximum is reached for the critical number of atoms in the nucleus $i_c \approx 373$, and its value is equal to $\Delta F(i_c) \approx 5\ k_BT$. For thickness $h = 2.8$ ML the critical number of atoms is $i_c \approx 41$, and nucleation activation barrier is $\Delta F(i_c) \approx 1\ k_BT$ (Figures 1, 2). And it is more probable to overcome the thermodynamic barrier of nucleation with the wetting layer thickness increasing. So, at the wetting layer thicknesses larger than equilibrium $h > h_{eq} = 2.5$ ML the transition from 2D to 3D growth becomes thermodynamically feasible.

Conclusions. Thus, in this paper new generalized Muller–Kern equation is obtained for calculation of equilibrium thickness of wetting layer during growth of semiconductors in the Stranski–Krastanow mode that takes into account the dependence of specific surface energy of facets on the wetting layer thickness. This equation is solved numerically for the model system of germanium quantum dots on silicon (001) surface. For this system an unexpected result for the value of equilibrium thickness is obtained, which significantly differs from the previously used value. In whole, this approach allows one to predict more accurately the growth conditions for synthesizing quantum dots arrays with desirable characteristics, important for creation of device-oriented nanoheterostructures. And although for the whole picture of growth process the main characteristics are kinetic values and, in particular, critical thickness of transition, such thermodynamic values as equilibrium thickness of wetting layer serve as the basis for its finding and consequently demand rigorous calculation.

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