Elongated Quantum Dots of Ge on Si Growth Kinetics Modeling with Respect to the Additional Energy of Edges

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Abstract. In this paper refining of mathematical model for calculation of parameters of self-organised quantum dots (QDs) of Ge on Si grown by the method of molecular beam epitaxy (MBE) is done. Calculations of pyramidal and wedge-like clusters formation energy were conducted with respect to contributions of surface energy, additional edge energy, elastic strain relaxation, and decrease in the atoms attraction to substrate. With the help of well-known model based on the generalization of classical nucleation theory it was shown that elongated islands emerge later than pyramidal clusters. Calculations of QDs surface density and size distribution function for wedge-like clusters with different length to width ratio were performed. The absence of special geometry of islands for which surface density and average size of islands reach points of extremum that was predicted earlier by the model not taking into account energy of edges was revealed when considering the additional contribution of edge formation energy.

1. Introduction

Semiconductor materials containing nanosized Ge clusters are popular for application in optoelectronics since the 1990s. They are used in development of photodetectors, solar cells and light emitting devices that are quite competitive with traditional optoelectronic materials such as AIII-BV compounds.

The one of the most promising Si/Ge heterostructures production methods is molecular-beam epitaxy. The morphology of Ge islands during the growth process can be controlled by changing the temperature of substrate, Ge deposition rate, and the total thickness of Ge layer.

It is known that ensemble of nanoislands of Ge on Si(100) surface consists of pyramidal (hut-) and elongated (wedge-) clusters, which are pyramids with square and rectangle base, respectively. But the differences in the kinetics of formation of these two types of clusters in quantum dots arrays are still studied poorly [1, 2].

In [3, 4] an attempt was made to study kinetics of wedge-like quantum dots formation with the help of the model, based on the classical nucleation theory and proposed in [5]. This model relies on the expression for change of free energy of atoms during transition from the wetting layer to the island taking into account change in free energy due to formation of additional facet surface, elastic strain relaxation, and lessening of atoms attraction to substrate [5]. Meanwhile, the contribution in change of the free energy due to formation of additional edges in the island is not taking into account as it is thought to be negligibly small. But at the same time results obtained in [6, 7] show that taking into account the contribution of edge energy is essential for explanation of some recently discovered

effects during growth of quantum dots of germanium on silicon. In [8] an attempt is made to develop kinetic model accounting for the contribution into free energy of island formation due to edge energy.

The aim of this paper is to compare growth kinetics of pyramidal and wedge-like clusters with various length to width ratios with respect to the contribution of additional edge energy into change in free energy during formation of islands.

2. Theoretical model, results and discussion

For calculations we used the kinetic model of Ge QDs on Si growth developed in [8]. This model allows us to define temperature dependencies of QDs surface density N and size distribution function for different growth rates V.

The free energy of pyramidal islands with a rectangular base and constant contact angle φ is a function of the number of atoms i in the island and is represented by

$$\Delta F(i) = Ai^{2/3} - B\zeta i + Ci^{1/3}, \tag{1}$$

where $\zeta = h / h_{eq} - 1$ is the wetting layer superstress, h is the Ge wetting layer thickness, h_{eq} is its equilibrium thickness. In this equation, free energy is expressed in $k_B T$ units, where T is the substrate temperature and k_B is the Boltzmann constant.

With respect to different geometrical shapes of QDs parameters A, B and C will be defined as

$$A = \frac{r[\gamma(\varphi)/\cos\varphi - \gamma(0)]\alpha l_0^2}{k_B T}, \tag{2}$$

$$B = \frac{[1 - Z(\varphi)]\lambda\varepsilon_0^2 l_0^2 d_0}{k_B T} \ln\left\{\frac{\Psi_0}{d_0[1 - Z(\varphi)]\lambda\varepsilon_0^2}\right\}, \tag{3}$$

$$C = \frac{\beta[2 + (r-1)\cos\varphi]\alpha l_0}{k_B T \cos\varphi}, \tag{4}$$

where r is the island's length to width ratio, $\gamma(0)$ and $\gamma(\varphi)$ are the specific surface energies of the base and the lateral faces of 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, $Z(\varphi)$ is the coefficient of elastic relaxation [9, 10], λ is the material's modulus of elasticity, ε_0 is the lattices' mismatch, d_0 is the height of one monolayer (ML), Ψ_0 is the wetting energy density on the substrate surface, β is the specific energy of edge formation, and α is the geometrical factor depending on the island's shape:

$$\alpha = \left(\frac{12 d_0 ctg\varphi}{(3r-1) l_0}\right)^{1/3}. \tag{5}$$

Parameter α defines the relationship between the lateral width of the island L and the number of atoms in it:

$$i = \left(\frac{L}{\alpha l_0}\right)^3. \tag{6}$$

Using the Zeldovich formula [11], for the nucleation rate elongated islands we will obtain the following dependence on the wetting layer superstress

$$I(\zeta) = \frac{a}{\tau l_0^2}\zeta(\zeta+1)e^{-\Delta F(i_c)}, \tag{7}$$

where i_c is the critical size at which the free energy (1) is at its maximum:

$$i_c = \left(\frac{C}{\sqrt{A^2 + 3CB\zeta} - A} \right)^3, \tag{8}$$

τ is the characteristic time of atoms incorporation processes:

$$\tau = \frac{3l_0^2 \nu}{4(r+1)\alpha BD}, \tag{9}$$

D is the coefficient of diffusion of atoms from the wetting layer to the islands, ν is the cut-off parameter of the elastic strain field [9].

In case of taking into account energy of edges parameter a is defined as

$$a = \frac{3}{2}\sqrt{\frac{|\Delta F^{"}(i_c)|}{2\pi}}\,\frac{i_c^{1/3}}{\zeta}. \tag{10}$$

Calculations of parameters of pyramidal and wedge-like clusters with different length to width ratio r were carried out. For calculations the following material values were selected [5, 7–9, 12]: $\lambda = 1.27 \cdot 10^{12}$ dyn/cm^2, $\varepsilon_0 = 0.042$, $d_0 = 0.145$ nm, $l_0 = 0.395$ nm, $\Psi_0 = 450$ erg/cm^2, $\gamma(0) \approx \gamma(\varphi) = 800$ erg/cm^2, $\varphi = 20°$, $h_{eq} = 3$ ML, $\nu = 10$, $D = 10^{-4}$*exp(–1.21 / $k_B T$) cm^2/s, $\beta = 1.5*10^{-6}$ erg/cm.

From comparison of the nucleation rates for islands with square and rectangle base it is shown that elongated islands with higher r emerge later but their nucleation doesn't progress more intensively as it was predicted earlier [3, 4] (see figure 1).

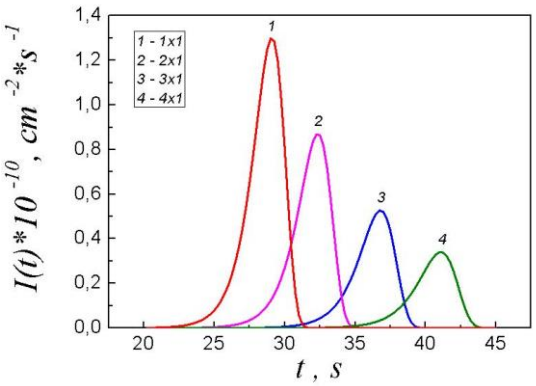

Figure 1. Time dependence of nucleation rate for elongated islands with $r = 1..4$ in Ge/Si(001) system at substrate temperature $T = 470$ °C and deposition rate $V = 0.1$ ML/s.

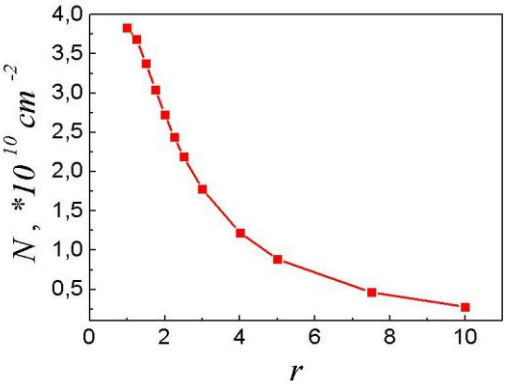

Figure 2. Surface density of wedge-like clusters of Ge on Si(001) surface as a function of r at substrate temperature $T = 470$ °C and deposition rate $V = 0.1$ ML/s.

Modelling of QDs growth dynamics allows us to estimate surface density and size distribution function for the different shaped islands. Results of numerical experiments show that for the same growth conditions average size of elongated islands with small length to width ratio r is larger than for pyramidal clusters when taking into account additional energy of edges (see figures 2 and 3). Their size variation and surface density are smaller.

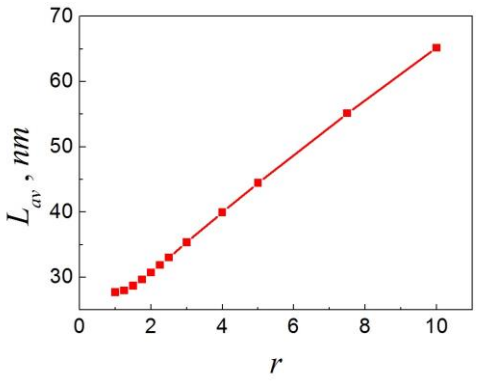

Figure 3. Average size of wedge-like clusters of Ge on Si(001) surface as a function of r at substrate temperature $T = 470$ °C and deposition rate $V = 0.1$ ML/s.

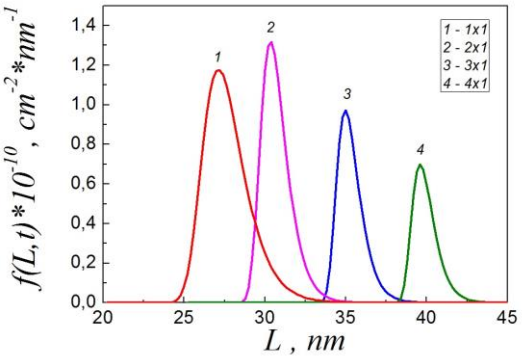

Figure 4. Size distribution function for wedge-clusters of Ge on Si(001) with $r = 1..4$ at substrate temperature $T = 470$ °C and deposition rate $V = 0.1$ ML/s.

It must be noticed, however, that surface density and average size of the elongated islands change monotonically with the ratio increasing as opposed to the results of previous modelling [4]. There is no singular geometry of QDs with $r = 2$ for which average size of the dot reaches its minimum and surface density inversely has its maximum value (for the given growth conditions). With the increase of the length to width ratio from 1 to 10 surface density decreases gradually and the average size increases, both reaching their extrema at $r = 1$ that correspond to pyramidal clusters with square base.

3. Conclusions

Thus, in this paper an attempt is done to apply different shaped QDs of Ge on Si(100) the kinetic model of initial stages of growth of QDs accounting for additional energy of edges. Estimations with the help of this model show that average size and surface density of clusters monotonically depends on length to width ratio r and there is no special geometry of QDs as it was predicted before.

Acknowledgments

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