

OSTWALD RIPENING IN HETEROSTRUCTURES WITH QUANTUM DOTS UNDER DISLOCATION-SURFACE DIFFUSION

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The size distribution function of islands for semiconductor heterostructures is calculated within the framework of the mechanism of Ostwald ripening (OR) under the condition that islands grow at the expense of dislocation-surface diffusion. It is shown that, as regards the root-mean-square deviations of the calculated curves, the proposed mechanism of island growth corresponds to a number of the experimentally determined arrays of quantum dots (QDs) in a Ge/Si(001) heterostructure.

In its classical version, the formation of a new phase includes three stages: the appearance of centers (nuclei, clusters) of a new phase, their independent growth, and a further development of these centers that interact with one another through the solid solution of a diluted substance. The latter stage of the new phase formation (also called the late stage), in which small particles dissolve while larger ones grow at their expense, which is conditioned by the Gibbs–Thomson effect (the dependence of the pressure or concentration at the boundary of a particle on the curvature of its surface) is called the Ostwald ripening or, rarely, coalescence.

As concerns the island phase that appears during the heteroepitaxial growth of semiconductor systems in the Stranski–Krastanow mode [1] in the form of 3D-islands, it is considered that OR represents a decisive factor in the formation of the size distribution of islands. In this case, the OR process is accompanied with the variations of both the size and form of islands. They acquire a regular faceting, by turning into high-perfect quantum-size nanocrystals which are free of dislocations and are coherently coupled to the substrate. They are usually called quantum dots. Changing the form and size of QDs, one can control their energy spectrum, which is important for practice.

A coherent conjugation of islands with a substrate means that there are no misfit dislocations (MD) in the boundary region. The absence of MD results in the fact that, due to the inconsistency of the crystal lattices of materials of an island and the substrate, there arise elastic deformations in the periphery regions of islands and the substrate which induce stresses in

islands. That's why an important feature of QDs is their internal stress. For example, in the most widespread Ge/Si(001) heterosystem, the difference in the lattice parameters amounts to 4%. It is just elastic deformations in heteroepitaxial systems that represent the key factor of their further evolution.

Lately, a special attention in the literature is paid to the theory of OR, namely to the LSW (Lifshits–Slezov–Wagner) theory [2–4] modified for a surface [5–7]. In particular, it is used to explain the evolution of clusters on semiconductor surfaces on the basis of the results of observations performed with a modern high-resolution technique [8–12].

It's also worth noting another viewpoint, according to which elastic deformations distort the adjacent region between the substrate and an island by changing the regularities of the attachment of adatoms to the latter. This results in the deceleration of the growth rate of islands in the oversaturated “sea” of adatoms, as well as in the formation of narrower size distributions than it is predicted by the LSW theory in the framework of the QD model [13–19].

However, at present, there doesn't exist another theory allowing one to calculate these narrower distributions with regard for elastic deformations [20–21]. In addition, the narrower size distributions can be also obtained in the framework of the LSW theory in the case where, in addition to surface diffusion, one also allows for the possibility of diffusion of another kind, namely, the dislocation diffusion [22–23].

At first sight, it seems to be strange, as QD are considered just as dislocation-free nanocrystals. However, considering the dislocation diffusion, we intend matrix dislocations (rather than MD) that can arise in the wetting or substrate layer upon its plastic deformation during the “planar film — island film” transition in the process of self-organization by the mechanism described in [1]. A further elastic interaction between the deformation fields of islands and dislocations results in the capture of the latter and their

fixation near the base of islands, along their perimeters, radially to the surface [23]. Internal stresses in islands don't allow dislocations to penetrate inside islands. To say it figuratively, they are glued to the perimeters of islands. Due to the intersection of dislocations with the substrate surface, there arise dislocation grooves, along which the diffusion of adatoms takes place.

The dislocation mechanism of diffusion in the OR process is possible under the condition that the flow of adatoms, which is conditioned by dislocation diffusion, towards an island is much higher than that arising due to the surface diffusion. That is, we have

$$D_s^{(d)} Z d \left(\frac{dC}{dR} \right)_{R=r} \gg D_s 2\pi r \left(\frac{dC}{dR} \right)_{R=r}, \quad (1)$$

where $D_s^{(d)}$ stands for the coefficient of diffusion in parallel to dislocation grooves, D_s is the surface diffusion coefficient, $(dC/dR)_{R=r}$ is the concentration gradient on the island surface, Z is the number of dislocations fixed near the base of the island of radius r ($Z = \text{const}$), d is the width of a dislocation groove, $d = 2\sqrt{\frac{2q}{\pi}}$, $b^2 \leq q \leq 60b^2$, where q stands for the cross section of a dislocation groove, and b is the Burgers vector. Let's consider the islands to be disk-like and have a constant height h [22]. The general case for variable h and r is considered in [23].

Relation (1) imposes restrictions on the dimensions of islands, whose growth takes place at the expense of the dislocation diffusion, namely,

$$r \ll \frac{Z d D_s^{(d)}}{2\pi D_s}. \quad (2)$$

If condition (2) is violated, considering the total flow of adatoms, one should also allow for the component appearing due to the surface diffusion in addition to that arising at the expense of dislocation one.

The present paper is devoted exactly to the calculation of the size distribution function of islands under the condition of dislocation-surface diffusion, where none of the flows can be neglected. To our mind, it is also important because, just as the stability of a crystal lattice of a substance is provided with the simultaneous action of different types of bonds, the crystal growth occurs under the condition of a mixed diffusion, where only one type of diffusion can prevail (it can be the matrix, surface, or dislocation diffusion, grain boundary diffusion, etc.).

In order to determine the size distribution function of islands $f(r, t)$, it is necessary to know the rate of growth

(dissolution) of a separate island. It can be derived from the equation

$$\frac{d}{dt} (\pi r^2 h) = j v_m, \quad (3)$$

where v_m is the adatom volume, j is the total flow. Under the conditions of dislocation-surface diffusion,

$$j = j_d + j_s, \quad (4)$$

where j_d is the flow running to the island due to the diffusion along dislocations, while j_s appears at the expense of the surface diffusion; their values are specified by the left and right parts of inequality (1), respectively.

After the substitution of (4) into (3) with regard for the values of j_d and j_s from (1), as well as the concentration gradient at the boundary of an island of radius r , $(\frac{dC}{dR})_{R=r} = \frac{\sigma v_m}{kT \ln l} C_\infty \frac{1}{r^2} \left(\frac{r}{r_k} - 1 \right)$ [7], we obtain

$$\frac{dr}{dt} = \frac{\sigma v_m^2 C_\infty}{2\pi h k T \ln l} \frac{1}{r^2} \left(D_s^{(d)} Z d \frac{1}{r} + 2\pi D_s \right) \left(\frac{r}{r_k} - 1 \right), \quad (5)$$

where C_∞ is the equilibrium concentration of adatoms at the plane boundary, σ is the specific surface energy, r_k is the critical radius, and k is the Boltzmann constant.

By x and $(1 - x)$, we denote, respectively, the parts of j_s and j_d in the total flow j :

$$x = \frac{j_s}{j}, \quad 1 - x = \frac{j_d}{j}, \quad \frac{j_d}{j_s} = \frac{1 - x}{x}. \quad (6)$$

In order to express the growth rate (5) in terms of the partial flows j_s and j_d , we take $D_s^{(d)} Z d \frac{1}{r}$ out of the brackets and multiply the numerator and the denominator of the second term by $r_g (\frac{dC}{dR})_{R=r_g}$. As a result, we obtain

$$\frac{dr}{dt} = \frac{\sigma v_m^2 C_\infty D_s^{(d)} Z d}{\pi h k T \ln l} \frac{1}{r^3} \times \left(\frac{2\pi r_g D_s (\frac{dC}{dR})_{R=r_g}}{D_s^{(d)} Z d (\frac{dC}{dR})_{R=r_g}} \frac{r}{r_g} + 1 \right) \left(\frac{r}{r_k} - 1 \right), \quad (7)$$

where r_g is the maximal size of islands, and $(\frac{dC}{dR})_{R=r_g}$ is the concentration gradient at the boundary of an island of radius r_g .

The ratio $2\pi r_g D_s (\frac{dC}{dR})_{R=r_g} / (Z d D_s^{(d)} (\frac{dC}{dR})_{R=r_g})$ is equal to the ratio of the flows $\frac{j_s}{j_d}$ for a particle of the maximal size r_g . According to (6), it can be replaced by $\frac{x}{1-x}$, because relation (6) doesn't contain any restrictions

on the sizes of particles. Therefore, (7) can be rewritten as

$$\frac{dr}{dt} = \frac{\sigma v_m^2 C_\infty D_s^{(d)} Z d}{\pi h k T \ln l} \frac{1}{r^3} \left(\frac{x}{1-x} \frac{r}{r_g} + 1 \right) \left(\frac{r}{r_k} - 1 \right). \quad (8)$$

At $x = 0$, we obtain the first limiting case, where the growth of islands is restricted by the diffusion along dislocations. The corresponding distribution function has a form [7]

$$g(u) = \frac{u^3 \exp \left[-\frac{1}{3(1-u)} \right] \exp \left[-\frac{1}{9\sqrt{2}} \arctg \frac{u+1}{\sqrt{2}} \right]}{(1-u)^{25/9} (u^2 + 2u + 3)^{29/18}}. \quad (9)$$

Now, taking $2\pi D_s$ out of the brackets in formula (5) and once more multiplying the numerator and denominator of the first term by $r_g \left(\frac{dC}{dR} \right)_{R=r_g}$, we get

$$\frac{dr}{dt} = \frac{\sigma v_m^2 C_\infty D_s}{h k T \ln l} \frac{1}{r^2} \left(\frac{1-x}{x} \frac{r_g}{r} + 1 \right) \left(\frac{r}{r_k} - 1 \right). \quad (10)$$

Considering (10) at $x = 1$, we obtain another limiting case where the growth rate is restricted only by the surface diffusion. In this case, the size distribution function of islands is determined by the Lifshits–Slezov distribution [2–3] modified for surfaces [7]:

$$g(u) = u^2 (1-u)^{-28/9} (u+2)^{-17/9} \exp \left(-\frac{2/3}{1-u} \right), \quad (11)$$

where $u = \frac{r}{r_g}$.

Prior to pass to the determination of the size distribution between the above-mentioned limiting cases, we find the ratio of the maximal radius of islands r_g to the critical one r_k , $\frac{r_g}{r_k}$. Within the framework of the LSW theory, the critical radius of islands r_k coincides with their average radius $\langle r \rangle$: $r_k \equiv \langle r \rangle$. The less this ratio, the closer is the average radius of islands to the maximal one. This means that, in the first approximation, the ratio r_g/r_k can be a measure of uniformity of the size distribution.

The ratio r_g/r_k can be derived from the equation [24]

$$\left. \frac{d}{dr} \left(\frac{\dot{r}}{r} \right) \right|_{r=r_g} = 0, \quad (12)$$

where $\dot{r} \equiv dr/dt$.

Equation (12) with regard for Eq. (8) or (10) yields

$$\frac{r_g}{r_k} = \frac{4-x}{3-x}. \quad (13)$$

In the limiting cases, we get $\frac{r_g}{r_k} = \frac{4}{3}$ (dislocation diffusion) at $x = 0$ and $\frac{r_g}{r_k} = \frac{3}{2}$ (surface diffusion) at $x = 1$.

Relation (13) allows us to determine the time dependences for r_g and r_k . If we substitute r_g into (8) instead of r and replace $\frac{r_g}{r_k}$ by its value from (13), we get

$$\frac{dr_g}{dt} = \frac{A}{r_g^3} \frac{1}{(1-x)(3-x)}, \quad (14)$$

where $A = \frac{\sigma v_m^2 C_\infty D_s^{(d)} Z d}{\pi h k T \ln l}$. After the integration of (14), we obtain

$$r_g^4 = \frac{4A}{(1-x)(3-x)} t \quad (15)$$

or, with regard for (13),

$$r_k^4 = \frac{4A(3-x)^3}{(1-x)(4-x)^4} t. \quad (16)$$

By analogy, Eq. (10) becomes

$$\frac{dr_g}{dt} = \frac{B}{r_g^2} \frac{1}{x(3-x)}. \quad (17)$$

After the integration, we get

$$r_g^3 = \frac{3B}{x(3-x)} t \quad (18)$$

or

$$r_k^3 = \frac{3B(3-x)^2}{x(4-x)^3} t, \quad (19)$$

where $B = \frac{\sigma v_m^2 C_\infty D_s}{h k T \ln l}$.

It's easy to see that, after grouping Eqs. (15) and (18) as well as (16) and (19), r_g and r_k satisfy the equations

$$\frac{r_g^4(1-x)}{4A} + \frac{r_g^3 x}{3B} = \frac{2t}{(3-x)}, \quad (20)$$

$$\frac{r_k^4(1-x)}{4A} + \frac{r_k^3 x}{3B} = 2 \frac{(3-x)^2}{(4-x)^3} t. \quad (21)$$

According to [21], in order to determine $f(r, t)$, it should be presented in the form of a product of two functions

$$f(r, t) = \varphi(r_g) \cdot g(u), \quad (22)$$

where $g(u)$ is the distribution of relative sizes $u = r/r_g$ of islands. Using the conservation law of mass M of the island film, we can derive $\varphi(r_g)$. Indeed, we get

$$M = K \int_0^{r_g} r^2 f(r, t) dr, \quad (23)$$

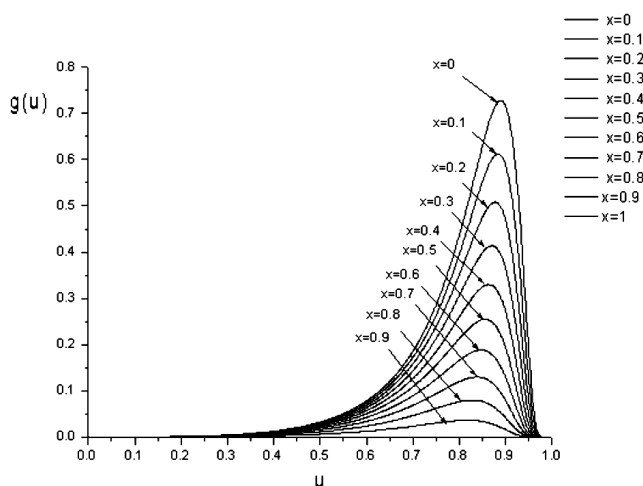


Fig.1. Size distributions calculated in the interval $\Delta x = 0.1$

where $K = \pi h \rho$, and ρ is the density of the substance of islands. After the substitution of (22) into (23), we obtain

$$\varphi(r_g) = \frac{Q}{r_g^3}, \tag{24}$$

where $Q = M/K \int_0^1 u^2 g(u) du$.

The relative size distribution function $g(u)$ can be derived from the continuity equation

$$\frac{\partial}{\partial t}(f(r, t)) + \frac{\partial}{\partial t}(f(r, t)\dot{r}) = 0. \tag{25}$$

Replacing $f(r, t)$ and \dot{r} in (25) by their values from (8) and (22) and then passing from the differentiation with respect to r and t to the differentiation with respect to u , we can separate the variables in (25) and obtain the equation for the determination of $g(u)$

$$\frac{dg(u)}{g(u)} = -\frac{3v_g + 2\frac{v}{u^3} - \frac{1}{u^2}\frac{dv}{du}}{uv_g - \frac{v}{u^2}} du, \tag{26}$$

where $v_g = \frac{dr_g}{dt} \frac{r_g^2}{A}$, $v = \left(\frac{x}{1-x}u + 1\right) \left(\frac{4-x}{3-x}u - 1\right)$.

After the substitution of v and $v_g (v_g = v|_{u=1})$ into Eq.(26), we obtain

$$\begin{aligned} \frac{dg(u)}{g(u)} = & -\{3u^4 - (x^2 - 4x)u^2 + (x^2 - 4x + 2)4u^2 - \\ & -3x^2 + 12x - 9\} / \{u[u^4 + (x^2 - 4x)u^2 - \\ & -(x^2 - 4x + 2)2u + x^2 - 4x + 3]\}. \end{aligned} \tag{27}$$

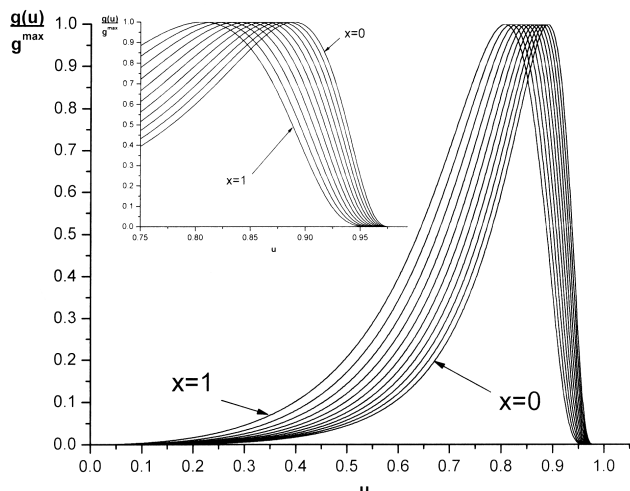


Fig. 2. Size distributions calculated in the interval $\Delta x = 0.1$ and normalized to their maxima (the inset shows the enlarged scale)

In order to integrate (27), we must perform the prime factorization of the quadric polynomial in the denominator. After that, Eq.(27) takes the form

$$\frac{dg(u)}{g(u)} = -du[3u^4 - (x^2 - 4x)u^2 + (x^2 - 4x + 2)4u^2 - 3x^2 + 12x - 9] / [u(u - 1)^2(u^2 + bu + c)], \tag{28}$$

where $b = 2$, and $c = x^2 - 4x + 3$.

The integration of (28) gives the relative size distribution function of islands under the dislocation-surface diffusion as

$$\begin{aligned} g(u) = & \frac{u^3(u^2 + bu + c)^{\frac{D}{2}}}{(u - 1)^K} \exp\left(\frac{F}{u - 1}\right) \times \\ & \times \exp\left(\frac{(E - Db)/2}{\sqrt{c - b^2/4}} \operatorname{arctg} \frac{u + b/2}{\sqrt{c - b^2/4}}\right), \end{aligned} \tag{29}$$

where

$$\begin{aligned} D = & [3c^2 + (x^2 - 4x + 6b - 6)c + 6b^2 + \\ & + (4x^2 - 16x + 14)b + 7x^2 - 28x + 19] / \\ & / [c^2 + (b + 1)2c + b^2 + 2b + 1], \\ E = & \frac{(3 - D)c + (D - 3)b^2 + (2b + 1)D + x^2 - 4x - 3}{2 + b}, \\ F = & D(b + 1) - 3b - E, \quad K = 6 - D. \end{aligned} \tag{30}$$

Figure 1 demonstrates the graphic form of the obtained distributions. The curves are calculated according to formula (29) in the interval $\Delta x = 0.1$ between zero and unity, which corresponds to two

Characteristics of the relative size distribution of islands under dislocation-surface diffusion

| | S | Φ | D | σ | S_k | E | $\langle u \rangle$ | u' |
|-----------|------------------------|------------------------|----------|----------|--------|-------|---------------------|------------------------|
| $x = 0$ | 1.158 | 0.322 | 0.012739 | 0.113 | -1.498 | 2.861 | 0.75 | 0.41379 |
| $x = 0.1$ | 0.992 | 0.274 | 0.013137 | 0.115 | -1.457 | 2.662 | 0.744 | 0.72786 |
| $x = 0.2$ | 0.837 | 0.23 | 0.013564 | 0.116 | -1.417 | 2.453 | 0.738 | 0.61233 |
| $x = 0.3$ | 0.693 | 0.189 | 0.014025 | 0.118 | -1.377 | 2.284 | 0.731 | 0.50785 |
| $x = 0.4$ | 0.561 | 0.152 | 0.014529 | 0.121 | -1.336 | 2.102 | 0.724 | 0.32954 |
| $x = 0.5$ | 0.44 | 0.118 | 0.015086 | 0.123 | -1.287 | 1.924 | 0.716 | 0.25452 |
| $x = 0.6$ | 0.331 | 0.088 | 0.015715 | 0.125 | -1.244 | 2.905 | 0.708 | 0.18819 |
| $x = 0.7$ | 0.232 | 0.061 | 0.016441 | 0.128 | -1.2 | 1.571 | 0.698 | 0.13003 |
| $x = 0.8$ | 0.144 | 0.037 | 0.01731 | 0.132 | -1.156 | 1.402 | 0.688 | 0.07959 |
| $x = 0.9$ | 0.067 | 0.017 | 0.018415 | 0.136 | -1.115 | 1.243 | 0.681 | 0.03648 |
| $x = 1$ | 6.454×10^{-4} | 1.629×10^{-4} | 0.02 | 0.141 | -1.086 | 1.132 | 0.667 | 3.469×10^{-4} |

limiting cases of diffusion, the dislocation diffusion and the surface one (Fig. 1). One can see that, with increase in x , the maxima of the distributions $g^{\max} \equiv g(u')$ decrease, while the values of u' , at which these maxima are reached, are shifted towards lower values of u . It is demonstrated by Fig. 2, in which we present the same distributions normalized to their maxima. The very values of u' are determined from the equation

$$3u^4 - (x^2 - 4x)u^2 + (x^2 - 4x + 2)4u^2 - 3x^2 + 12x - 9 \Big|_{u=u'} = 0. \quad (31)$$

For the curves depicted in the figure, we have calculated the most widely used numerical characteristics of the distributions that are given in the Table: S and Φ stand for the area and volume of the island phase per unit area of the substrate, S_k is the asymmetry coefficient, E is the excess, D is the dispersion, σ' is the root-mean-square deviation, and $\langle u \rangle = \frac{r_k}{r_g}$ is the average relative size of islands. The inverse problem (the reconstruction of a size distribution on the basis of the moments determined experimentally) is also often solved.

But in practice, the principal parameter whose value allows one to judge about the uniformity of the array of quantum dots of a heterostructure is the root-mean-square deviation $\sigma' = \sqrt{D}$, where $D = \langle u^2 \rangle - \langle u \rangle^2$. From this viewpoint, the best size distributions were obtained for germanium islands in the Ge/Si(001) heterosystem. In this case, $\sigma' < 10\%$ [25]. The theoretical distributions that correspond to such values of the dispersion D or σ' are obtained in [22–23] under the condition that the principal factor which determines the size distribution form is OR.

At the same time, there exists a number of distributions of Ge islands of the dome type with an average size of 50–100 nm in the considered Ge/Si(001) system, for which the values of σ' fluctuate within the range 10–30% [12]. According to the data given in the Table, the value of σ' for the calculated distributions

varies from 11 to 14%. This means that a part of the given examples can be explained in the framework of the proposed mechanism of formation of QDs in heterostructures.

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ОСТВАЛЬДІВСЬКЕ ДОЗРІВАННЯ ГЕТЕРОСТРУКТУР
З КВАНТОВИМИ ТОЧКАМИ В ПРОЦЕСІ
ДИСЛОКАЦІЙНО-ПОВЕРХНЕВОЇ
ДИФУЗІЇ

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Резюме

У рамках механізму оствальдівського дозрівання розрахована функція розподілу острівців за розмірами для напівпровідникових гетероструктур за умови, що ріст острівців відбувається шляхом дислокаційно-поверхневої дифузії. Показано, що, судячи за значеннями середньоквадратичних відхилень розрахованих кривих, запропонований механізм росту острівців відповідає ряду експериментальних масивів квантових точок в гетеросистемі Ge/Si(001).