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Kinetic model of coherent island formation in the case of self-limiting growth

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The stage of nucleation and growth of stressed coherent islands is investigated for the Stranski–Krastanow growth mode. Both elastic deformations and the attraction of atoms by the wetting layer due to a wetting potential are taken into account. It is shown that these two effects do not change the functional dependence of the free energy of island formation on the size of the island and result in the renormalization of the surface tension of islands and the equilibrium concentration of adatoms. Analytical expressions for the rate of self-limiting growth and cluster-size distribution of coherent islands are obtained. The evolution equation of supersaturation is deduced and the time dependences of all the main features of the deposition process such as supersaturation, nucleation rate, surface concentration of islands, and their size distribution are calculated. Self-limiting growth of islands is shown to result in a secondary nucleation process.

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I. INTRODUCTION

The self-assembled growth of dislocation-free coherent islands is currently used to produce dense ordered structures of nanoscale quantum dots.[1,2] The increasing interest in such systems is caused by the prospect of their practical use in optoelectronics due to the carrier confinement in quantum dots.[3] For this reason a small size and narrow cluster-size distribution are required. In order to control the morphology of growing islands, it is necessary to find out whether it is determined by the growth kinetics or by thermodynamics.[2] Recently, the thermodynamic model of the free energy of a strained island ensemble has been developed by Shchukin *et al.*[4] It includes the surface energy, elastic deformation energy, and the energy of elastic relaxation on edges. In this work it has been shown that for certain conditions the free energy of a coherent island ensemble may have a minimum that corresponds to the most favorable size of islands.[1,4] This minimum may result in a narrow size distribution.[5,6]

An alternative explanation of narrowing of the size distribution during growth is based on the kinetic control of self-limiting growth.[7–9] This effect takes place when larger islands grow more slowly than smaller ones because the elastic deformation leads to an energetic barrier for the embedding of adatoms into islands[7] and the growth on island facets.[9] As a result, small islands are able to catch up in size with larger ones. This fact also results in a narrowing of the size distribution. This type of self-limiting growth of islands was recently observed in the Ge/Si(100) system.[9] The authors explain this effect by an increasing nucleation barrier for each repeated growth of a new atomic layer on an island facet.

In the present work the mechanism of self-limiting growth provides the basis for the theoretical description of the processes of nucleation and growth of quantum dots in the case of the Stranski–Krastanow growth mode. The time dependence of the main characteristics of growth such as supersaturation, nucleation rate, surface concentration of islands, and their size-distribution function are calculated.

II. THEORY

A. Free energy and nucleation rate of islands

The starting point of any nucleation theory is the calculation of the free energy of island formation F as a function of the number of atoms i in an island. For the case of 3D coherent islands growing by the Stranski–Krastanow mode it may be presented in the form

$$F(i)=E_{\mathrm{surf}}-i\Delta\mu+E_{\mathrm{el}}-E_{\mathrm{att}}\ . \tag{1}$$

Here E_{surf} is the surface energy term, $\Delta\mu$ is the difference between the chemical potential of the 2D gas of adsorbed atoms on the wetting layer and of atoms in the new-phase island, E_{att} is the energy of attraction of island atoms to the substrate, which always takes place in Stranski–Krastanow growth,[10] and E_{el} is the elastic energy of an island. We assume for simplicity that each new-phase island has the permanent shape of a hemispherical segment with a contact angle θ and a base radius R (see Fig. 1), which is situated on a wetting layer of the same material. Notice that this angle is distinguished from a wetting angle that is equal to zero because the surface energy of an island is equal to the surface energy of the wetting layer. But the mismatch of film and substrate materials results in a positive value of the contact angle of dislocation free islands to decrease the stress elastic energy.[12] Let k be the number of monolayers in the wetting layer and h_0 the height of one monolayer. Then $h=kh_0$ is the height of the complete wetting layer (Fig. 1). For this island shape the aspect ratio is equal to $\beta=(1-\cos\theta)/(2\sin\theta)$. For example, a value of $\theta=30°$ conforms with $\beta=0.134$, which

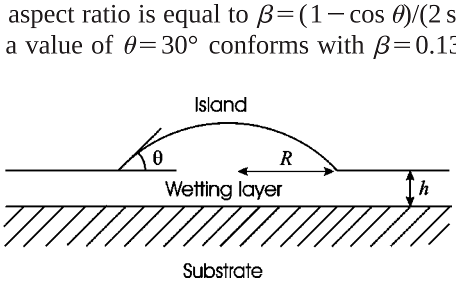

FIG. 1. Schematic representation of an island of hemispherical shape with contact angle θ and base radius R being situated on a wetting layer of the same material with height h.

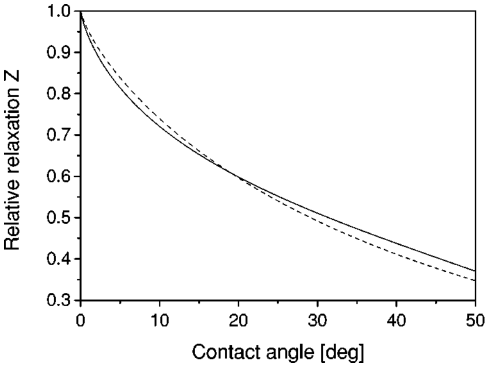

FIG. 2. The dependence of the relative relaxation of elastic deformation stress in an island on the contact angle; solid line, the Ratsch–Zangwill approximation (Ref. 13); dashed line, finite element calculations (Ref. 11) averaged over island volume.

approximately corresponds to the case of Ge islands on Si(100).[12] The energy of formation of an island surface equals the product of the specific energy of the island-gas interface σ_{isl} with the difference between the initial surface area of the wetting layer and newly formed surface area $S_{\text{surf}}=2\pi R^2(1-\cos\theta)/\sin^2\theta-\pi R^2=4\pi\beta^2R^2$,

$$E_{\text{surf}}=4\pi\beta^2\sigma_{\text{isl}}R^2. \tag{2}$$

The difference in the chemical potential is of the usual form

$$\Delta\mu=k_BT\ln\frac{n_1}{n_e}, \tag{3}$$

where k_B is the Boltzmann constant, T is the temperature, n_1 is the surface concentration of adatoms, and n_e is the equilibrium concentration of adatoms. The elastic energy of an island due to the lattice misfit can be presented in the form

$$E_{\text{el}}=\lambda\varepsilon_0^2\Omega iZ(\beta), \tag{4}$$

where $\lambda=(c_{11}+2c_{12})(c_{11}-c_{12})c_{11}^{-1}$ is the elastic modulus, c_{11} and c_{12} are the elastic moduli in the standard Voigt notation,[1] $\varepsilon_0=(a-b)/b$ is the misfit parameter, b is the lattice parameter of a substrate, a is the lattice parameter of the film material, Ω is the volume per atom in the film, and $Z(\beta)$ is the relative relaxation of the island stress. For a given island shape this function was found numerically by the method of finite elements.[11] Moreover, the dependence of Z on the aspect ratio or contact angle can be estimated by the Ratsch–Zangwill approximation.[13] Results of calculations performed within the framework of these estimations for a hemispherical segment are given in Fig. 2. It can be seen that the Ratsch–Zangwill approximation is in good agreement with finite element calculations. According to the Müller–Kern theory,[10] the energy of attraction of island atoms to a substrate per unit surface is determined by the wetting energy per unit surface $\Phi_\infty=\sigma_s-\sigma_f-\sigma_{f-s}$, where σ_{f-s} is the specific interface energy of the boundary between the film and the substrate, and σ_s, σ_f are the surface tensions of the substrate and the film, respectively. Φ_∞ represents the energy of attraction of the nearest atomic layer of the film to the substrate. The following atomic layers have a weaker attraction to the substrate. In the case of semiconductor materials this decay can be considered to be exponential:[10] $\Phi_m=\Phi_\infty\exp(-m/k_0)$, where m is the number of monoatomic layers separating a given film layer from the substrate, and k_0 is the dimensionless parameter of relaxation.[10] Therefore,

$$E_{\text{att}}=\Phi_\infty n_0^{-1}\exp\left(-\frac{k}{k_0}\right)\sum_{m=0}^{L}i_m\exp\left(-\frac{m}{k_0}\right), \tag{5}$$

where k is the number of monolayers in the wetting layer, L is the number of monolayers in the island, n_0 is the surface concentration of atoms in a monolayer of the film, and i_m is the number of atoms in the mth layer of an island. The value of k is determined by the balance of the stress energy and the energy of attraction to the substrate.[10] For large islands whose height is much larger than k_0 (normally, $k_0\simeq1$) it is possible to approximate the sum by

$$E_{\text{att}}=\Phi_\infty\exp\left(-\frac{k}{k_0}\right)(1+k_0)\pi R^2. \tag{6}$$

Thus, in the framework of these assumptions the free energy of coherent island formation on a wetting layer is equal to

$$F=4\pi\beta^2\sigma_{\text{isl}}R^2-\Phi_\infty\exp\left(-\frac{k}{k_0}\right)(1+k_0)\pi R^2 -ik_BT\ln\frac{n_1}{n_e}+i\lambda\varepsilon_0^2\Omega Z(\beta). \tag{7}$$

The first and last terms of this expression contain the aspect ratio β. Moreover, σ_{isl} depends on the shape of the islands according to the Wulf–Kaishew theorem. Therefore, minimizing the sum of these terms gives the equilibrium aspect ratio and shape of the islands. This minimization procedure was performed in Ref. 14 and it was shown that the equilibrium aspect ratio slightly increases with cluster sizes up to the dislocation formation process. An even more complicated problem was solved in Ref. 6, where the elastic energy of edges and the repulsive interaction between islands were taken into account to describe the pyramid-to-dome transition under near-equilibrium conditions. In our case, when strong supersaturation takes place, we neglect these effects and consider $\beta=\text{const}$ in order to describe the kinetic aspects of the evolution of an island ensemble.

It is convenient to rewrite this function in terms of renormalized parameters

$$F=4\pi\beta^2\sigma_{\text{eff}}R^2-ik_BT\ln\frac{n_1}{n_{\text{eff}}}. \tag{8}$$

Here

$$\sigma_{\text{eff}}=\sigma_{\text{isl}}-\Phi_\infty\exp(-k/k_0)(1+k_0)/(4\beta^2) \tag{9}$$

is the effective surface tension of an island,

$$n_{\text{eff}}=n_e\exp(\kappa Z(\beta)) \tag{10}$$

is the effective equilibrium concentration of adatoms, and

$$\kappa=\lambda\varepsilon_0^2\Omega/(k_BT) \tag{11}$$

is a dimensionless constant that characterizes the ratio between elastic and thermal energies. Let us estimate these values for Ge islands on a Si(100) surface at $T=550\,°\mathrm{C}$. In this case $k_0\sim 1^{10}$, $\sigma_{\mathrm{isl}}\approx\sigma_f=800$ erg/cm^2, $\sigma_s=1260$ erg/cm^2, and $\sigma_{f-s}=10$ erg/cm^2, which yields $\Phi_\infty=450$ erg/cm^2. For $k=3$ and $\beta=0.134$ we obtain $\sigma_{\mathrm{eff}}=490$ erg/cm$^2\approx 0.6\sigma_{\mathrm{isl}}$. If we choose, for example, $k=2$, then σ_{eff} will be negative. This fact means that it is more advantageous for germanium to spread over the substrate surface than to form islands. In order to decrease the substrate attraction it is necessary to add one more monolayer to the wetting layer.

Since for Ge/Si(100) $\varepsilon_0=0.042$, $c_{11}=1.14\times 10^{12}$ dyn/cm^2, $c_{12}=0.38\times 10^{12}$ dyn/cm^2, and $\Omega=2.27\times 10^{-23}$ cm^3, we obtain $\lambda=1.27\times 10^{12}$ dyn/cm^2 and $\kappa=0.45$. The relative relaxation of the elastic stress in an island corresponding to $\theta=30°$ amounts to approximately 0.5 (see Fig. 2). Therefore, $n_{\mathrm{eff}}=1.25n_e$, i.e., the elastic stress increases the equilibrium concentration of adatoms by 25%.

For the further analysis it is convenient to rewrite the free energy (8) in the form

$$F(i)=\left[\frac{3}{2}(\sqrt{2ci})^{2/3}-i\ln(\xi+1)\right]k_BT, \tag{12}$$

where $\xi=n_1/n_{\mathrm{eff}}-1$ is the effective supersaturation and

$$c=\frac{\pi}{3}\frac{\sigma_{\mathrm{eff}}^3\Omega^2}{(k_BT)^3}\left(\frac{1-\cos\theta}{1+\dfrac{\cos\theta}{2}}\right)^2. \tag{13}$$

The estimation of the dimensionless parameter c at the same conditions yields $c=0.38$. The function (12) reaches its maximum at $i=i_c$, where the number of atoms in the critical nuclei is equal to

$$i_c=\frac{2c}{\ln^3(\xi+1)}. \tag{14}$$

The value of this maximum or the relative nucleation barrier height is equal to

$$H=\frac{F(i_c)}{k_BT}=\frac{c}{\ln^2(\xi+1)}. \tag{15}$$

For instance, for the case of Ge growth on Si(100) at $\xi=0.17$ these parameters have the values $H\approx 15$ and $i_c\approx 200$, which corresponds to $R(i_c)\approx 2.2$ nm.

If $F(i)$ is known, the nucleation rate can be found by the Zeldovich formula[15,16]

$$I=n_1\sqrt{-\frac{F''(i_c)}{2\pi}}W(i_c)e^{-H}. \tag{16}$$

Here $W(i_c)=2\pi R(i_c)n_1D/l_0$ is the rate of attachment of adatoms to a critical nuclei, D is the diffusion coefficient over the wetting layer, and $l_0=n_0^{-1/2}$ is the average distance between nearest atoms in a monolayer. Substitution of Eqs. (12) and (15) into Eq. (16) gives the dependence of the coherent island nucleation rate on supersaturation

$$I(\xi)=Dn_{\mathrm{eff}}^2(1+\xi)^2\ln(1+\xi)\sqrt{\frac{k_BTn_0}{\sigma}}\frac{1}{2\beta}\times\exp\left(-\frac{c}{\ln^2(1+\xi)}\right). \tag{17}$$

B. Rate of self-limiting growth of islands

In order to describe the evolution process of a system of coherent islands, the growth rate of an island must be found. For this purpose we use the results of Refs. 7–9, in which it was stated that the elastic deformation energy results in an energy barrier $G(R)$ for the attachment of adatoms into an island. In the absence of this barrier the attachment flux per unit length of an island boundary would be equal to $(n_R-n_{\mathrm{eff}})D/l_0$, where n_R is the concentration of adatoms near an island with the base radius R. If a barrier exists this flux decreases exponentially

$$j=(n_R-n_{\mathrm{eff}})(D/l_0)\exp(-G(R)/k_BT). \tag{18}$$

The function $G(R)$ was found numerically in Ref. 9. This dependence can be approximated by a power function[7]

$$G(R)\sim k_BT_0(R/\sqrt{D\tau})^p, \tag{19}$$

where τ is the lifetime of adatoms, $\sqrt{D\tau}$ is their diffusion length, and $T_0\sim\varepsilon_0^2$ is the characteristic temperature of self-limiting growth. Then the diffusion flux per unit length of an island boundary is equal to $j=(n_R-n_{\mathrm{eff}})(D/l_0)\exp(-\alpha(R/\sqrt{D\tau})^p)$, where $\alpha=T_0/T$ is a dimensionless parameter which is proportional to ε_0^2 and depends on the temperature T as $(D\tau)^{p/2}/T$. Thus, in order to find this flux it is necessary to solve the diffusion equation for the adatom concentration with proper boundary conditions

$$D\left(n''+\frac{1}{r}n'\right)-\frac{n}{\tau}=0,$$

$$-Dn'|_{r=R}=(n_R-n_{\mathrm{eff}})(D/l_0)\exp(-\alpha(R/\sqrt{D\tau})^p),$$

$$n(\infty)=n_1. \tag{20}$$

Here $n(r)$ is the adatom concentration at a distance r from the center of an island and n_1 is the adatom concentration at infinite distance from an island. Having solved this equation it is easy to find the growth rate of coherent islands

$$v_i=\frac{di}{dt}=2\pi Rj=2\pi Dn_{\mathrm{eff}}\xi f\left(\frac{R}{\sqrt{D\tau}}\right), \tag{21}$$

where the growth function f is given by

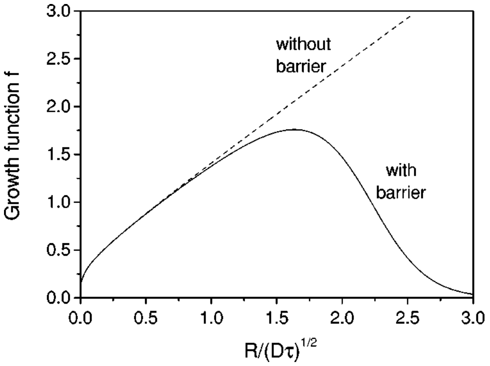

FIG. 3. The dependence of the self-limiting growth rate of an island on the normalized radius $R/\sqrt{D\tau}$; solid line, with an energy barrier; dashed line, without an energy barrier.

$$f(x)=\frac{x\dfrac{K_1(x)}{K_0(x)}e^{-\alpha x^p}}{\dfrac{l_0}{\sqrt{D\tau}}\dfrac{K_1(x)}{K_0(x)}+e^{-\alpha x^p}} \tag{22}$$

and K_0 and K_1 are the MacDonald functions. If the energy barrier for the adatom incorporation into an island is negligible, i.e., $(l_0/\sqrt{D\tau})K_1/K_0 \ll \exp(-\alpha x^p)$, ordinary growth by diffusion takes place with the rate xK_1/K_0. Otherwise, if the energy barrier is significant, i.e., $(l_0/\sqrt{D\tau})K_1/K_0 \gg \exp(-\alpha x^p)$, it limits the growth of coherent islands. This phenomenon was observed experimentally for sufficiently large islands.[9,17,18] In Fig. 3 the growth function (22) is shown for $l_0/\sqrt{D\tau}=0.01$, $\alpha=1$, $p=2$ (the values α and p were chosen on the basis of a rough approximation of the numerical results given in Ref. 9). It can be seen that the influence of the energy barrier for adatom attachment starts at approximately $R>1.7\sqrt{D\tau}$. Starting from this point the size-distribution function begins to be narrowed. We continue to describe this process quantitatively.

C. Evolution of supersaturation and size distribution of islands

We consider the early stage of deposition when the nucleation process takes place. In this case the surface coverage of islands is much less than unity and we can neglect both diffusion and elastic interaction of islands. Under these conditions coherent dislocation free islands are formed, whereas small island spacings result in the process of dislocation formation.[12] Moreover, we neglect the coalescence process of islands, which takes place at the late stage of deposition. Thus, at the early stage of deposition all islands grow independently and the distribution function $g(i,t)$ of coherent islands in terms of the number of atoms i at time t is determined by the continuity equation in size space[16]

$$\frac{\partial g}{\partial t}+\frac{\partial(v_i g)}{\partial i}=0. \tag{23}$$

Following the general technique described in Ref. 19 we turn from the variable i to the new variable ρ

$$\rho(i)=\int_0^i \frac{di'}{f(R(i')/\sqrt{D\tau})}, \tag{24}$$

where $R(i)=(3i\Omega/2\pi)^{1/3}[1+(1/2)\cos\theta]^{-1/3}(1-\cos\theta)^{-2/3}\sin\theta$ is the dependence of the base radius of an island on the number of atoms. In this case the evolution equation (23) is simplified considerably

$$\frac{\partial g}{\partial t}+\frac{\xi(t)}{t_g}\frac{\partial g}{\partial \rho}=0, \tag{25}$$

where $t_g=1/(2\pi D n_{\mathrm{eff}})$ is the characteristic time for coherent island growth. The initial condition of Eq. (25) is obviously

$$g(\rho,0)=0. \tag{26}$$

The boundary condition of Eq. (25) should be set at the boundary between the near-critical and overcritical regions.[19] However, this condition may be transferred to the point $\rho=0$

$$\frac{\xi(t)}{t_g}g(0,t)=I(\xi(t)), \tag{27}$$

because near-critical islands are so small compared to their actual size that they are of little importance in the matter balance of the wetting layer surface.[19] This condition means that the flux I of nuclei proceeds through the point $\rho=0$ with the rate ξ/t_g. Equation (25) with the conditions (26) and (27) can be solved analytically. After doing this we go back from the variable ρ to the variable i,

$$g(i,t)=\begin{cases}\dfrac{I(\xi(t(y(t)-\rho(i))))\xi(t(y(t)-\rho(i)))}{t_g f(R(i)/\sqrt{D\tau})}, & \rho(i)\leq y(t),\\ 0, \quad \rho(i)>y(t). & \end{cases} \tag{28}$$

Here, $t(y)$ is the inverse function to $y(t)=t_g^{-1}\int_0^t \xi(t')dt'$. Since $g(i,t)\sim 1/f(R(i)/\sqrt{D\tau})$ the function g increases and becomes narrower in the region where f is small, i.e., at large $R/\sqrt{D\tau}$ values (see Fig. 3).

The dependence of supersaturation on time $\xi(t)$ and thus $y(t)$ and $t(y)$ may be found from the conservation of matter on the wetting layer. In the case of small coverage of the wetting layer by coherent islands it can be written in the form

$$\frac{dn_1}{dt}=J(t)-\frac{n_1}{\tau}-\int_0^\infty v_i g(i,t)\,di, \tag{29}$$

where $J(t)$ is the arrival rate of adatoms on the wetting layer. The integral on the right-hand side of Eq. (29) describes the decrease of adatoms due to their embedding into islands. Hereafter we use the simplest model for the arrival rate J,

$$J(t)=J_0(1-e^{-t/t_s}), \tag{30}$$

where J_0 is the maximum flux and t_s is the characteristic time for reaching a constant flux. Substitution of Eqs. (21) and (30) into Eq. (29) gives the required evolution equation for supersaturation

$$\frac{d\xi}{dt}=\frac{\xi_0-\xi}{\tau}-\frac{1+\xi_0}{\tau}e^{-t/t_s}-2\pi D\xi\int_0^{\infty}f\left(\frac{R(i)}{\sqrt{D\tau}}\right)v_i g(i,t)di. \tag{31}$$

Here $\xi_0=J_0\tau/n_{\mathrm{eff}}$ is the highest effective supersaturation possible. Substitution of the obtained size-distribution function (28) into Eq. (31) gives an integro-differential equation in the effective supersaturation. Solving it and substituting the solution into Eqs. (17) and (28) one obtains the time dependence of the nucleation rate and the size-distribution function.

III. NUMERICAL RESULTS AND DISCUSSION

The integro-differential equation (31) was solved numerically by taking into account Eqs. (17) and (28), and the initial condition $\xi(0)=0$. For the calculation of the diffusion coefficient and the equilibrium concentration of adatoms the following Arrhenius equations are used

$$D=\frac{\nu}{4n_0}e^{-E_D/k_BT}, \tag{32}$$

$$\tau=\nu^{-1}e^{E_d/k_BT}, \tag{33}$$

$$n_e=n_0e^{-E_a/k_BT}, \tag{34}$$

where ν is the frequency of lattice oscillations of atoms, E_D is the activation energy for diffusion, E_d is the activation energy for desorption, and E_a is the energy of adatom formation. For example, at $T=550\,^{\circ}\mathrm{C}$, $\nu=10^{12}\ \mathrm{s}^{-1}$, $E_D=0.65$ eV, $E_d=1.3$ eV, and $E_a=0.6$ eV, which approximately corresponds to the system Ge/Si(100), we obtain $D=4.7\times10^{-8}\ \mathrm{cm}^2\,\mathrm{s}^{-1}$, $\tau=0.9\times10^{-4}$ s, $\sqrt{D\tau}=21$ nm, and $n_e=1.2\times10^{11}\ \mathrm{cm}^{-2}=2.1\times10^{-4}n_0$. The solution of Eq. (31) for $\xi_0=0.18$ and $t_s=30$ s with these constants and the values stated above that approximately correspond to Ge growth on Si(100) is shown in Fig. 4(a). Substitution of $\xi(t)$ into Eq. (17) gives the time dependence of the nucleation rate that is presented in Fig. 4(b). Integration of the nucleation rate over time results in the surface concentration of new-phase islands,

$$N(t)=\int_0^t I(\xi(t'))dt', \tag{35}$$

presented in Fig. 4(c). The size-distribution function of coherent islands is calculated according to Eq. (28). It is shown in Fig. 5 at different times. The characteristic behavior of the system becomes quite clear from these pictures. At the beginning the effective supersaturation increases under the influence of the adatom flux. Nucleation starts and progresses and growing islands act as sinks for adatoms. At a certain time these sinks become more effective than the external flux in creating adatoms. This means that the supersaturation goes down. The equilibrium situation corresponds to the maximum supersaturation and nucleation rate. For the parameter values given the relevant quantities are $\xi_{\max}=0.172\approx0.95\xi_0$ and $I_{\max}=6.5\times10^7\ \mathrm{cm}^{-2}\,\mathrm{s}^{-1}$. The higher $I_{\max}$, the shorter the nucleation time and the narrower the size distribution.[20] This conclusion is supported by experiments.[21] However, in the case of too large an arrival rate the coalescence of islands begins quickly[22] (in this work the coalescence process is not considered), which results in a broadened and bimodal size-distribution function. After some time the growing islands reach the region of self-limitation, where the nuclei absorb fewer and fewer adatoms. Thus the effective supersaturation goes up again, resulting in the process of secondary nucleation. This phenomenon has been frequently observed in different systems with coherent islands.[23] Due to

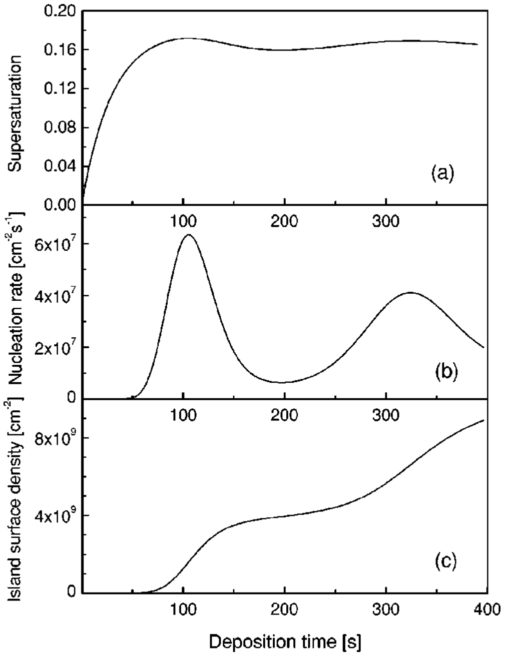

FIG. 4. The time dependences of the main characteristics of nucleation processes using the following constants: $\xi_0=0.18$, $T=550\,^{\circ}\mathrm{C}$, $\tau=10^{-4}$ s, $t_s=30$ s, $D=4\times10^{-8}\ \mathrm{cm}^2\,\mathrm{s}^{-1}$, $n_{\mathrm{eff}}=1.5\times10^{11}\ \mathrm{cm}^{-2}$, $\alpha=1$, $p=2$, $l_0=0.42$ nm, $\theta=30°$, $c=0.38$; (a) effective supersaturation, (b) nucleation rate, (c) surface density of islands.

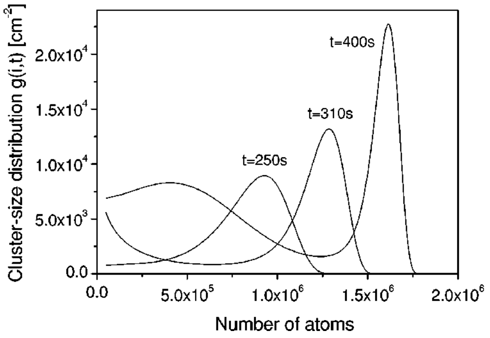

FIG. 5. Cluster-size distributions for three different growth times: $t=250$ s, $t=310$ s, and $t=400$ s. The conditions are the same as in Fig. 4.

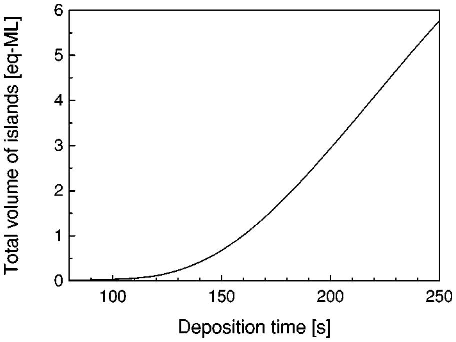

FIG. 6. Time dependence of the total volume of islands at the same conditions as in Fig. 4.

secondary nucleation the cluster-size distribution can become bimodal (see Fig. 5). If the cluster-size distribution is known one can easily calculate the total volume of islands and express it in equivalent monolayers

$$M(t)=n_0^{-1}\Omega\int_0^\infty ig(i,t)di. \qquad (36)$$

This function is presented in Fig. 6 up to the onset of secondary nucleation. It is easy to see that for large t the integrated growth rate $U=dM/dt$ tends to a constant value. Therefore, a one-to-one correspondence between the integrated growth rate and ξ_0 is observed. Since the integrated growth rate can be measured experimentally, it is convenient to use this quantity as a variable to describe the adatom flux onto the wetting layer. The dependence of the island density (up to secondary nucleation) on the integrated growth rate U is plotted in Fig. 7 for two different temperatures: $T=550\,°\mathrm{C}$ and $T=740\,°\mathrm{C}$. It is easy to see that the curves are very close to the power law $N=N_0U^p$ (for $T=550\,°\mathrm{C}$, $p\approx0.65$, and for $T=740\,°\mathrm{C}$, $p\approx0.71$). This theoretical conclusion conforms with experimental results for molecular beam epitaxy (MBE) of Ge on Si(100) at $T=740\,°\mathrm{C}$.[24] Similar results were obtained for Ge/Si(111) as well.[25]

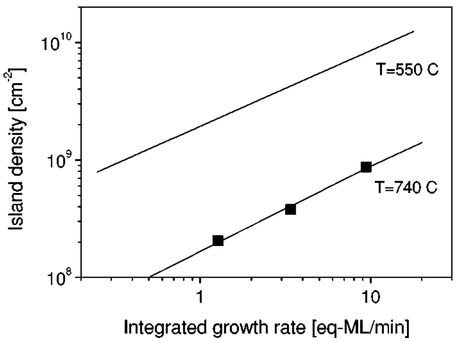

FIG. 7. Dependence of the surface density of islands on the integrated growth rate at the same conditions as in Fig. 4 at $T=550\,°\mathrm{C}$ and $T=740\,°\mathrm{C}$. Black squares are the experimental results of Ref. 23 replotted on a logarithmic scale.

In summary, a kinetic model of nucleation and growth of coherent islands has been developed for the case of Stranski–Krastanow growth. It describes the effect of stress energy on nucleation and self-limiting growth of coherent islands. The evolution equation is deduced as a function of the effective supersaturation. The time dependence of the main characteristics of the deposition process, such as the nucleation rate, the surface concentration of islands, and their size-distribution function are calculated. Self-limiting growth of islands is shown to be followed by a secondary nucleation process. The higher the integrated growth rate the sooner this process begins.

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