Size calibration of strained epitaxial islands due to dipole–monopole interaction

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Abstract. Irreversible growth of strained epitaxial nanoislands has been studied with the use of the kinetic Monte Carlo (KMC) technique. It has been shown that the strain-inducing size misfit between the substrate and the overlayer produces long range dipole–monopole (d–m) interaction between the mobile adatoms and the islands. To simplify the account of the long range interactions in the KMC simulations, use has been made of a modified square island model. An analytic formula for the interaction between the point surface monopole and the dipole forces has been derived and used to obtain a simple expression for the interaction between the mobile adatom and the rectangular island. The d–m interaction was found to be longer ranged than the conventional dipole–dipole potential. The narrowing of the island size distributions (ISDs) observed in the simulations was shown to be a consequence of a weaker repulsion of adatoms from small islands than from large ones which led to the preferential growth of the former. Furthermore, similar to the unstrained case, the power-law behavior of the average island size and of the island density on the coverage has been found. In contrast to the unstrained case, the value of the scaling exponent was not universal but strongly dependent on the strength of the long range interactions. Qualitative agreement of the simulation results with some previously unexplained behaviors of experimental ISDs in the growth of semiconductor quantum dots was observed.

Keywords: irreversible aggregation phenomena (theory), heteroepitaxy (theory), molecular beam epitaxy (theory), thin film deposition (theory)

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1. Introduction

The phenomenon of self-assembly of size-calibrated coherent nanoislands taking place in some heteroepitaxial systems during strained epitaxy has been extensively studied for more than two decades because of its prospective use in microelectronics [1, 2]. It seems to be well established that the phenomenon is governed by the elastic strain in the overlayer caused by the lattice size misfit with the substrate [1, 2]. Usually, it is assumed that the main role in the size calibration (SC) play the long-range forces propagated via the elastic strain in the substrate [3–5]. However, explicit growth simulations with the use of the kinetic Monte Carlo (KMC) technique within the models accounting for such forces in [6–8], did not show any narrowing of the island size distributions (ISDs). Moreover, in [6] even some broadening of the ISD was seen. Notably, the broad ISDs obtained in the simulations were very similar to those seen during irreversible growth [9]. So by all evidence the growth in [6–8] was controlled by kinetics; this is further supported by the fact that in thermodynamically controlled strained epitaxy in [10, 11] ISDs narrowing was observed. These results seems to suggest that the SC is implausible under conditions of kinetically controlled growth. Such growth, however, usually takes place at smaller temperatures and at faster deposition rates than the thermodynamically limited growth [10] which

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Size calibration due to dipole–monopole interaction presents some important practical advantages. For example, smaller substrate-deposit interdiffusion and so better control in heteroepitaxial growth [12]. Therefore, the question of whether the SC may be achieved under conditions of kinetically controlled growth is of considerable practical interest.

The aim of the present paper is to suggest a new mechanism of the SC during irreversible growth underlain by the repulsive long-range forces induced by the misfit strain. In contrast to the thermodynamically controlled growth when the conventional dipole–dipole (d–d) interatomic interactions [3,4] are sufficient to ensure the SC [10], the d–d forces are too weak to assure SC in the kinetically controlled case, as the explicit simulations in [6–8] and our arguments in section 3 below show. However, it is known that besides the dipole forces at the strained surfaces there also exist monopole forces that are of longer range than the d–d interactions and in the case of the step-bounded surface structures, such as steps, islands, and pits play a dominant role in their energetics and kinetics [5, 13–18].

In the present paper we will show that the monopole forces interacting with the force dipoles induced by the mobile adatoms play a similarly important role in the growth kinetics. In particular, they provide a mechanism of the SC during irreversible growth. As will be shown in section 3, in the presence of d–m interactions the strength of the repulsion between the island and the mobile atom may grow with the island size in such a way that the atoms will avoid larger islands by preferentially attaching to smaller ones, thus making the island sizes more homogeneous. The explicit confirmation of this mechanism in explicit KMC simulations will be provided in section 4. But because the main difficulty in studying the strained epitaxy with the KMC technique is the necessity of accounting for the long-range interactions acting between all atoms in the simulated system, essential simplifications and approximations are necessary to make the simulations feasible. The approximations used in KMC simulations in [6–8, 10] with the d–d interactions are not suitable for the d–m case. Therefore, in section 2 we will introduce a simple model of strained islands, in section 3 we will calculate the strain-induced interactions between the adatoms and the islands in the planar approximation, and in section 4 we will explain the approximate description of the capture of the mobile adatoms by the square islands. In this section we discuss the applicability of our results to the explanation of experimental data on the SC of the quantum dots in the Ge/Si(0 0 1) system studied in [19,20].

In the final section we present our conclusions.

2. The model of strained islands

The main problem in KMC simulation of many-body systems with long-range interactions is that to simulate the change in the position of a single atom, the energy of its interaction with all other atoms in the system need be calculated first. And in the case of the Metropolis algorithm [21] which is quite appropriate in this case (at sufficiently high temperatures, at least), the calculation needs to be performed twice: for the initial and the final positions of the moving atom. And in the end the move can be rejected by the algorithm.

Because one is usually interested in the thermodynamic limit, the simulated system should be reasonably large to mitigate the finite-size effects. For example, in our
Size calibration due to dipole–monopole interaction simulations we used, following [10], the system consisting of 250 $\times$ 250 sites on a square substrate lattice. With maximum coverage $\theta = 0.2$ chosen to remain in the precoalescence growth regime [9] our system contained up to 12,500 atoms. In order to be able to calculate the interaction with all of them at each KMC step we had to simplify the task by adopting several simplifying assumptions. In particular, we assumed that due to the fast intra-island diffusion the islands acquire simple quasiequilibrium symmetric shapes that can be found with the use of the Wulff construction [22]. This approach has been widely used in simulations of unstrained epitaxial growth [6, 23–27] so below we adapt it to our needs. In particular, this will allow us to make comparison of our results with experiments on the growth of three-dimensional quantum dots (QDs) because crucial to our SC mechanism will be only the island-substrate interface and the shear strain propagated by it. So, though in our simulations we use monolayer-high islands, if the base layer of a QD is size calibrated, the height (hence, the volume) that can be found via the Wulff construction will be subject to the SC [22]. In the present paper, however, we will restrict our simulations to the simplest case of monolayer-high (or submonolayer [28]) islands.

Submonolayer islands on the square substrate lattice at low temperature will strive to acquire rectangular shapes [29]. In the harmonic approximation the elastic forces in orthogonal directions decouple. This allows one to treat the elastic relaxation independently within each linear atomic chain that composes the island [11,30]. Another simplification is to neglect the displacements of the surface atoms in the direction perpendicular to the surface, as suggested in [3,4,31]. This leaves us with one-dimensional (1D) chains of atoms lying at the rigid substrate [11,30].

Unfortunately, it is difficult to reliably calculate numerical values of the interactions between surface adatoms. In the cases where both ab initio calculations and experimental data are available they may disagree several times [32]. Therefore, for qualitative discussion below we will invoke model interatomic pair potentials and the harmonic Frenkel–Kontorova (F–K) model which roughly agrees with ab initio calculations at least in some cases [3,30,33].

In discussing interatomic interactions between adatoms we will separate them into direct interactions via the pair potentials that are strong but of short range and indirect interactions that are propagated via elastic deformation of the substrate. The latter interactions are typically much weaker but are long-ranged. The direct interactions dominate interatomic binding within the islands. Typically their strength is in the range of electron-Volts for the nearest neighbor (NN) atoms [34] and is two to three orders of magnitude larger than the indirect interactions at the same distance [6,7]. Therefore, in discussing intra-island interactions we will neglect the indirect contributions.

In the F–K model the atoms are harmonically bound to the deposition sites at the substrate, so the monopole forces in the in-plane directions are proportional to the atomic displacements and to the stiffness constant of the harmonic coupling $k$.

The displacements $u_j$ of atoms $j = 1, 2, \ldots, l$ within a chain consisting of $l$ atoms can be calculated as [30]

$$u_j = f \sinh[\phi(2j - l - 1)]/[\sqrt{\alpha} \cosh(\phi l)], \quad (1)$$

where $f$ is the size misfit between the substrate and the overlayer,

$$\alpha = k/k_{\text{NN}} \quad (2)$$

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is the ratio of the rigidities of the elastic springs binding the atom to the substrate $k$ and to the nearest neighbor atom $k_{\text{NN}}$ and

$$\phi = \ln(\sqrt{1 + \alpha/4} + \sqrt{\alpha/2}).$$

(3)

Unless explicitly stated otherwise, in (1) and everywhere in the paper all lengths are measured in the substrate lattice units (l.u.), so that, e.g., the numerical value of the misfit parameter $f$ coincides with the relative misfit.

The parameters of the F–K model in (1)–(3) can be fixed through their fit to more accurate descriptions [30,33]. We will assess them via the fit to a model pair potential [30]. Assuming that interatomic interaction can be described by the Lennard-Jones potential $U_{\text{LJ}}(r)$, we can assess the interatomic spring constant as

$$k_{\text{NN}}(f) = \frac{d^2U_{\text{LJ}}(r)}{dr^2}|_{r=r_{\text{NN}}},$$

where we explicitly pointed out the dependence of $k_{\text{NN}}$ on the misfit. When the misfit is positive, the adatoms are compressed and $k_{\text{NN}}$ grows with $f$. The atoms became more rigid and approach the incompressible hard-core atom model where the displacements within the chain can be found from purely geometric considerations [11] $u_j = f(2j - l - 1)/2$, (5)

i.e. the atomic displacements depend linearly on the distance from the middle of the chain. The behavior in (5) is obtained from (1) as $\alpha \to 0$ which corresponds to the case of infinitely rigid atoms ($k_{\text{NN}} \to \infty$). The rigidity of real atoms, however, is finite, though it can be large, which means small $\alpha$. To assess the influence of different misfits and of finite rigidity on the island relaxation, let us assume that the growth of the islands proceeds via the Stranski–Krastanov mode typical for the QD growth [1]. In this mode the islands grow on the wetting layer consisting of the same kind of atoms as the island. Therefore, the elastic binding to the substrate characterized by $k$ can be assumed to be approximately the same for all values of the misfit. The change in the misfit does not influence the binding significantly because the adatoms are free to move in the vertical direction. The elastic interaction between the NN adatoms characterized by $k_{\text{NN}}$, on the other hand, will be strongly misfit-dependent, as the distance dependence of the model pair potentials shows [30,34]. Thus, the misfit dependence of $\alpha$ in (2) is mainly due to $k_{\text{NN}}$ in (4). The second derivative can reach arbitrary large values at positive misfit because $U(r - f)$ is very steep at the small values of the argument. For negative values of $f$, however, the second derivative quickly diminishes and even became negative. Thus, there exists a value of misfit at which the relaxation of end atoms in large islands in (1) reaches its maximum values. This is important because the monopole forces are proportional to the atomic displacements.

We illustrated this behavior in figure 1 with the parameters chosen to simulate the Ge/Si(001) system with $f = 0.042$ (the relative misfit in the system is 4.2%). Assuming that germanium was deposited on a substrate with negative misfit, we found that the maximum displacements occur at $f = -0.055$ and are about twice smaller than in the case of the positive misfit. This correlates with the fact that the size calibration in QDs occurs much more often in systems with positive misfit [11,35]. In connection with the negative misfit data shown in figure 1 it should be noted that irrespective of any possible mechanism of the SC based on the island size dependence of the adatom-island interaction, the very fact that the atomic displacements saturate at small island sizes makes the efficiency of
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Figure 1. Absolute values of the atomic displacements calculated according to (1). Only one half of the displacements are shown with the other half having the same values but the opposite sign. Black symbols: calculations with $\alpha = 0.025$ and $f = 0.042$ for the chains consisting of (from left to right) 10, 20, 50 and 80 atoms. Red symbols: $\alpha = 0.22$, $f = -0.055$ and $l = 20$. The solid lines show the displacements of the end 40 and 10 atoms of an infinitely long chain with the parameters corresponding to the symbols of the respective color. For further explanations see the text.

such a mechanism rather poor because a strong size dependence of the displacements can be expected only in very small islands. But when the number of atoms in the island is small, purely statistical fluctuations significantly broaden the ISDs, as shown in [36]. Furthermore, in section 4.2 below we present arguments why the existence of such a mechanism is highly improbable.

As will be show in the next section, the SC may take place in islands where the atomic displacements and, as a consequence, the monopole forces grow with the island size. It is this type of island that we will use in our KMC simulations in section 4. But it is obvious that in reality the atoms are not absolutely rigid and the compressibility $(1/k_{NN})$ is finite, though it can be small. From figure 1 it is seen that the displacements follow the linear behavior (5) only in sufficiently small islands. In large islands the displacements and the monopole forces saturate and phenomenologically can be described by constant monopole force density at the island edge, as in [5, 13–18]. In this case the small and large islands repulse mobile monomers with similar strength so our SC mechanism ceases to operate.

The discrete strain can be calculated as (assuming the chain is oriented in the $x$ direction)

$$ \varepsilon_{xx} = \frac{\Delta u}{\Delta x} = u_{i+1} - u_i $$

because the $x$-coordinate difference is equal to 1 l.u. As can be seen from figure 1, the average strain in small islands has appreciable value which diminishes with growing island size. In large islands strain remains only in the ends of the chain, so the average strain will tend to zero as $1/l$. It is exactly the behavior found in square Co islands on the Cu(0 0 1) surface in ab initio molecular dynamics simulations in [37]. The authors assess that the saturation starts in islands of $\sim 2$ nm size. Size-calibrated metallic islands of similar sizes are needed to produce efficient catalysts [38], so our SC mechanism can be of practical importance in this field.

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3. Long-range elastic interactions on the surface

In this section we derive expressions for the substrate-propagated elastic interactions to be used in KMC simulations in section 4. Due to the long-range nature of the interactions [39], the hopping adatom interacts with all other atoms on the surface. In order to calculate the interaction with the large number of atoms at each atomic hop, computationally efficient expressions for the interaction energy are needed.

As was explained earlier, such expressions can be derived for epitaxial islands of simple geometry. Thus, following [6, 23–27] we assume that at low temperature the islands on the square substrate lattice acquire rectangular shapes and will derive the expressions for the interaction of the mobile monomers with such islands. To simplify calculations we will treat elasticity in the continuum approximation assuming the substrate is an isotropic elastic medium bounded by a plane [3, 4, 39]. The energy of interaction of an adatom with an island can be found as the work performed by the force field \( F_d(r) \) induced in the substrate by the adatom along the displacement field \( u(r) \) due to the elastic deformation of the substrate by the island [4]

\[
V(r) = - \int F_d(r - r') \cdot u(r') \, dr'. \tag{7}
\]

To find the interaction potential \( V \) we first need to calculate the two fields entering (7).

3.1. The force field of isolated adatom

Let us consider a substrate with all atoms being fixed at their equilibrium positions. An atom deposited at low temperature at the substrate will occupy some minimum energy position. The adatom will exert forces \( F^d_i \) at the substrate atoms which in response will relax to their new minimum energy positions. The displacements of the substrate atoms will be different in the presence and in the absence of an island on the surface. The difference in the relaxation energies in these two cases in the continuum approximation (7) gives the potential of interaction of the adatom with the island.

The forces \( F^d_i \) strongly depend on the properties of both the adatom and the substrate. For example, when there is an appreciable charge transfer between the substrate and the adatom, long-range electrostatic field will arise. Its interaction with the substrate atoms will depend on whether they are ionized or not as well as on their polarization properties. In the case of the metallic substrate long-range forces due to the Friedel oscillations will be present [40, 41]. Additional complications appear when the substrate undergoes reconstruction [34]. Thus, in general, the adatom-substrate interactions should be rather complex and judging from the complexity of indirect adatom–adatom interactions will require many parameters for their model description [32, 40]. Because, as we mentioned earlier, their \textit{ab initio} estimate is unreliable, a fit to experimental data would be necessary.

To minimize the number of fitting parameters we, following [4], will restrict ourselves to the simplest case of the short range dipole forces that in the isotropic continuum approximation can be characterized by only one parameter:

\[
F^d(r - r') = A \nabla_r \delta(r - r'), \tag{8}
\]

where \( r' \) is the adatom position on the surface and \( A \) is a constant. Despite its simplicity, distribution (8) can approximately describe the important case of the SC of semiconductor.

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QDs that has been extensively investigated recently. For example, in the Ge/Si system because of the absence of the charge transfer between Ge and Si, long-range interactions are absent. Therefore, to simplify the calculations of interatomic interactions model potentials are usually set to zero beyond the maximum distance of the order of 1–3 interatomic spacings [34,42]. In the continuum approximation the forces of such a short extent can be modelled with the use of the delta-function (8).

To understand physical meaning and to assess the value of the parameter $A$, let us first ‘calculate’ it from (8) as follows

$$A = -\frac{1}{2} \int \mathbf{r} \cdot \mathbf{F}(\mathbf{r}) d\mathbf{r}. \tag{9}$$

It is natural to assume that in microscopic (atomistic) picture this expression should read

$$A = -\frac{1}{2} \sum_i \mathbf{r}_i \cdot \mathbf{F}_i, \tag{10}$$

where the sum is over the substrate atoms within the topmost layer. An additional argument in support of (10) will be given in section 3.2 below.

To assess the numerical value of $A$ it would be natural to use a model potential and restrict the sum in (10) to a few most important terms, e.g. to the substrate atoms nearest to the adatom. In this approximation, however, the equilibrium position of the isolated adatom will be at the interatomic potentials minima where the forces (the potential gradients) vanish. Thus, $A$ is defined by the adatom-substrate interactions at longer than NN distances.

Because even ab initio calculations cannot provide a reliable quantitative description of adatom interactions, [32], in a model approach we may hope to only assess the orders of magnitude of the quantities of interest. In our calculations we will use a simplified and slightly modified treatment of the problem suggested in [3]. It was observed that the repulsive part of model potentials is very short-ranged in comparison with the attractive part [3]. Therefore, when interactions with further than NN atoms are of interest, to a good precision only the attractive part may be used in calculations. For example, in the case of the Lennard-Jones potential which we will use in our estimates the attractive part reads

$$U_{LJ}^a(R) = -\frac{C_2}{R^6}, \tag{11}$$

where $R = |\mathbf{R}|$ is a three-dimensional (3D) radius vector. We will need to perform some calculations in 3D space because the adatom does not belong to the surface and thus its position cannot be described with the use of the 2D vectors that we use in the majority of our calculations. 3D radius-vectors will be denoted by the uppercase $\mathbf{R}$.

To proceed further we first note that in mechanical equilibrium the forces exerted by the adatom on the substrate atoms sum to zero. In particular, assuming that the normal to the surface is directed along $z$-axis and the adatom is placed at the 3D coordinates origin we have

$$\sum_i \partial U(R_i)/\partial R_i^z = 0, \tag{12}$$

where $\mathbf{R}_i$ is a 3D vector connecting the adatom and the substrate atom $i$. Now if we further assume that the surface is flat and all atoms have the same $z$ coordinate, then
by multiplying (12) by $R_i^2/2$ and adding it to (10) with $F^d_i$ replaced by the 2D gradient of $U(R_i)$ with the minus sign, we can express $A$ through the virial of the interatomic potential

$$A = \frac{1}{2} \sum_i R_i \cdot \nabla_{R_i} U(R_i).$$

(13)

Finally, substituting in this equality the Lennard-Jones potential and applying the Euler’s homogeneous function theorem to (11) we arrive at

$$A \approx 3 \sum_i C_2 R_i^6,$$

(14)

where the prime recalls that the substrate atoms nearest to the adatom are excluded from the summation. Because $C_2$ in (11) is positive, parameter $A$ should be positive in all cases where pair model potential gives a dominant contribution to the adatom-substrate interaction, i.e. at least in the case of physisorption [3].

Let us estimate $A$ in (14) for the classic QD system Ge/Si(001) with the size calibration due to the misfit strain [1, 2]. For the purposes of crude estimates we may assume an unreconstructed Si(001) surface with a flat topmost layer and Si atoms forming the square lattice with the lattice constant $c = 3.84\text{Å}$. As is known, germanium and silicon atoms are very similar in their properties, in particular, the parameters characterizing their model potential interactions have close values differing by $\sim 20\%$ at most [42]. Therefore, we will calculate $A$ in (14) with Si as the adatom which will spare us the necessity of calculating the parameters of the Ge–Si interaction [42].

Thus, with Si adatom in the bridge position at the distance $R_{NN} = 2.35\text{Å}$ from two NN substrate atoms, with $C_2 \approx 700\text{eVÅ}^6$, and the cutoff radius for the pair potential $R_c = 7.3\text{Å}$ [34] one gets

$$A_{\text{Si}} \approx 1.2\text{eV}.$$  

(15)

3.2. Point dipole–monopole and dipole–dipole interactions for individual adatoms

If an in-plane monopole force $F$ is applied at the coordinate origin, the 2D displacement field $u(r)$ in (7) produced by it can be calculated in the isotropic case with the use of the linear elasticity theory [39]. So substituting expressions (8.19) for $u(r)$ from [39] together with (8) into (7) one gets after some algebra

$$V_{d-m}(r) = \frac{A(1-\nu)}{2\pi \mu} \frac{F \cdot r}{r^3},$$

(16)

where $\nu$ is the Poisson ratio and $\mu$ the shear modulus of the substrate. Everywhere in the present study we, following [3, 4], will restrict elasticity to 2D in-plane forces and displacements, so the component of the monopole force in the direction perpendicular to the surface is set to zero in expressions (8.19) of [39]. In more sophisticated models of strain in epitaxial islands (see, e.g. [43]) this component is non-vanishing and should be included in (16) along the above line of derivation.

With the use of (16) the interaction energy of two adatoms can be derived by means of integration of (16) with $F$ replaced by the point dipole force distribution (8) to give [4]

$$V_{d-d}(r) = \frac{1-\nu}{2\pi \mu} \frac{A^2}{(cr)^3} = \frac{\gamma}{r^3},$$

(17)
where $\gamma > 0$ is the interaction strength and $r$ is measured in the lattice units, so $r$ is dimensionless and the closest atoms are separated by the distance $c = 11 \text{u}$. We note that the dipole–dipole interaction (17) was derived in [3] in the microscopic approach. So by comparing (17) with the corresponding expression from [3] we obtain more rigorous argument in favor of (10).

Substituting into (17) $A_{\text{Si}}$ (15), $c_{\text{Si}} = 3.84 \text{Å}$, $\nu_{\text{Si}} = 0.17$, and $\mu_{\text{Si}} \approx 1 \text{eV}$ one obtains $\gamma_{\text{Si}} \approx 3 \text{meV}$ which favorably agrees with $\gamma = 5 \text{meV}$ for the Ge/Si(0 0 1) system suggested in [7].

3.3. Interaction of mobile adatoms with rectangular islands

3.3.1. The dipole–dipole forces. Assuming that the atoms within the island form a rectangle with sides $a$ and $b$ along the $x$ and $y$ directions, respectively, and summing contributions of the form of (17) over all atoms within the island in the continuum approximation one gets:

$$V_{ab}^{d-d}(x, y) = \gamma \left[ \frac{\vec{r}}{\bar{x}\bar{y}} \right] \bar{x}_+ \bar{y}_+,$$

(18)

Here, two-dimensional radius vector $\vec{r} = (x, y)$ points from the island center which we placed for simplicity at the coordinate origin $(0, 0)$ to the position of the mobile atom; $\bar{x}_\pm = x \pm a/2$, $\bar{y}_\pm = x \pm b/2$, and $\bar{r} = (\bar{x}^2 + \bar{y}^2)^{1/2}$. The square brackets in (18) denote the substitution of four possible combinations of $\bar{x}_\pm$, $\bar{y}_\pm$ for $\bar{x}$ and $\bar{y}$ with necessary signs.

3.3.2. The monopole–dipole forces. The dipole–dipole interactions correspond to the situation where each adatom is positioned in the geometric center of the substrate lattice unit cell, so all atomic displacements in the substrate are symmetric and the resulting force distribution corresponds to the force dipole. Such a behavior would describe rather a non-strained situation because the presence of a positive misfit $f$, for example, means that the adatoms are too large to fit into the substrate cells without pushing their neighbors. This situation can be described within the model of the rigid-core adatoms [11] which should be adequate for situations where $f > 0$ and the atomic relaxation in the island base layer is small. Thus, it is easy to see from (5) that if the diameter of the rigid core is $1 + f$, then the atom with the coordinates $\vec{r} = (x, y)$ inside the island centered at the origin will be displaced by its neighbors from the center of the lattice cell it belongs to, in the direction $\vec{r}$ as

$$\Delta \vec{r} = f \vec{r}.$$

(19)

[11]. But the adatoms are bound to the substrate cell centers by an effective harmonic spring with the spring constant $k$. So the displaced adatom at position $\vec{r}$ within the island will exert on the substrate a shear force in the $x$–$y$ plane with the density

$$\mathbf{F}^m(\vec{r}') = k \Delta \mathbf{r}\delta(\vec{r} - \vec{r}') = k \mathbf{f}\delta(\vec{r} - \vec{r}'),$$

(20)

where for simplicity we again resorted to the continuum approximation. We supplied the force density with the superscript ‘$m$’ to stress its monopole character. Of course, there always exists an atom at $\vec{r} = -\vec{r}$ with the opposite force density so that at large distances from the island the potential will have the dipole–dipole asymptotic of the type of (17). In large islands, however, the distance $2r$ between the atoms can be large so an adatom approaching the island boundary will experience effectively the monopole forces.
So substituting (20) into (16) and integrating over the rectangular island \(a \times b\) centered at the origin \((0, 0)\) one gets the potential of interaction with the adatom placed at the point \((x, y)\) external to the island as

\[
V_{ab}^{d-m}(x, y) = C \left[ (x + \bar{x}) \ln(\bar{y} + \bar{r}) + (y + \bar{y}) \ln(\bar{x} + \bar{r}) \right]_{\bar{x}, \bar{y}}^{ar{x}_+ \bar{y}_+},
\]

(21)

where

\[
C = Akf (1 - \nu) / 2\pi \mu
\]

(22)

and other notation is the same as in (18).

To further simplify the KMC simulations, we will assume that the islands are of square shape [23–25] because in the case of weak elastic forces and small islands we are going to study, the aspect ratios of the rectangular islands are known to be close to unity [6, 18].

4. KMC simulations of the growth of the square islands

To assess the efficiency of the proposed mechanism, kinetic Monte Carlo (KMC) simulations were carried out with the use of a variant of the square-island model developed in [23–25]. As explained in the previous section, to simplify the calculation of the elastic forces we slightly modified the model by applying the approach of [26, 27] to the square islands instead of the circular ones. Namely, we assumed that the side length of the island \(a = \sqrt{s}\), where \(s\) is the island size. Then the square capture zone [9] with the side length \(a + 2\) is formed by surrounding the island with a strip of width 1. Any atom that enters the capture zone either by direct impingement or via the hopping diffusion is irreversibly caught by the island whose size becomes \(s + 1\).

4.1. Growth in the absence of elastic interactions

To validate our KMC setup we first carried out the simulations without the long-range interactions. Two points could arouse concern in connection with our approach. First, because the capture of the monomers by the islands is different from the conventional square island model [23–25], the question arises as to whether the physics of the growth remains qualitatively the same. Second, because of the difficulties with accounting for the interaction of the diffusing monomer with all atoms in the system, the size of the simulated lattice was chosen to be \(250 \times 250\) sites which is smaller than typically used for the simulations of the growth without long-range interactions.

The diffusion constant was calculated according to the standard expression

\[
D = \nu_{at} \exp(-E_d/\kappa_B T)
\]

with typical values for the attempt frequency \(\nu_{at} = 1\) THz, the diffusion barrier \(E_d = 0.7\) eV [11], and the deposition rate \(1.4\) ML min\(^{-1}\) [19]. To gather good statistics simulations were repeated from 160 to 480 times so that the statistical errors in our data are very small usually not exceeding the sizes of the symbols used to plot the data.

We first checked the soundness of our approximations by simulating the growth without elastic forces and comparing the results with simulations on similar models of
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Figure 2. Simulated ISD for irreversible growth at 400 °C in the absence of misfit strain in our model (histogram); for comparison are shown simulation data from [6] (filled circles) and [25] (crosses).

compact islands. The data shown in figure 2 are plotted in the scaling variables [9], as is conventional in the precoalescence regime at coverage $\theta$ not exceeding $\sim 0.2$:

$$N_s = \theta g(s/s_{av})/s_{av}^2,$$

where $N_s$ is the density of islands of size $s$, $s_{av}$ is the average island size, and $g$ a universal scaling function. As can be seen, the agreement is very good; most importantly, no ISD narrowing is seen in our data which means that the SC obtained in the simulations shown on figure 3 is due to the strain and not because of the approximations made.

4.2. The influence of repulsive interactions on the growth kinetics

The broadness of the ISDs of the kind shown in figure 2 can be understood within the framework of the following qualitative picture of the irreversible growth [9]. After the initial phase of the island nucleation the aggregation growth regime follows when the majority of the deposited atoms get caught by the existing islands. The island that a deposited atom meets and attaches to is the one that is closest to the deposition site. Because at the ballistic deposition regime that we use in our KMC simulations the atoms are deposited on all surface sites with equal probability, an island growth rate (hence, its size) is defined by the number of sites on the surface that are closer to this island than to any other one. In this way the substrate gets divided into the so-called capture zones (CZs) and it is the broad distribution of the sizes of these CZs that define the width of the ISD up to the broadening due to the statistical fluctuations in the number of atoms that are important only in small islands [36].

Now let us discuss how this picture can be influenced by the adatom-island interactions. We first note that, as follows from (21) and (22), the dipole–monopole interaction can be attractive when $Af$ is negative (the dipole–dipole interaction (18) is always repulsive). For example, in the physisorption case when $A > 0$ this may happen under negative misfit $f < 0$. But the attractive interaction cannot essentially change the aggregation picture described above. The adatom within the given CZ will only diffuse faster to the island. This may diminish the island nucleation rate because of the shorter...
time spent by the adatom in the mobile state. But the nucleation at this stage is already suppressed. Also, at the nucleation stage the influence of the adatom-island interaction is negligible because the islands are yet very small and are far apart. Thus, the ISD width in the case of attractive interactions should be very similar to the non-interacting case, so the SC is implausible.

Adatom-island repulsion, in contrast, can significantly influence the growth at the aggregation stage. When the repulsion is sufficiently strong, it is difficult for the diffusing adatom to get attached to the island. Thus, having being repulsed by the island during several attempts to approach it, the adatom may eventually leave the current CZ and diffuse into another one. This may be repeated many times before the adatom will be caught by one of the islands. In this process the densities of adatoms at different sites will approach the equilibrium distribution

\[ N_1(i) \propto \exp\left[-E(i)/k_B T\right], \tag{24} \]

where \( E(i) \) is the energy of the adatom at site \( i \). This can be used to assess the growth rate of the islands of different sizes. In [44] we showed that the rate of the island growth is proportional to the density of mobile atoms at the sites situated one hopping step away from the island. They may be called the island’s nearest neighbors. Thus, the possibility of SC can be judged on the basis of values of \( E(i_{NN}) \) in the vicinity of the islands. In the precoalescence regime that we are dealing with in this study, the islands are usually well separated, so the adatom energy \( E \) can be well approximated by the potential of interaction of the adatom with the nearest island.

First, we show that the conventional dipole forces (17) [3, 4] are not sufficiently strong for our SC mechanism to be operative. Let us for definiteness consider the point \( i_{NN} = (a/2 + 1, 0) \), at 1 l.u. distance from the middle of the edge of length \( a \) of the island centered at the coordinate origin \((0,0)\). From (18) it is easy to see that at large \( a,b \)

\[ E(i_{NN}) \approx V_{ab}^{d-d}(a/2 + 1,0) \]

saturates to a constant value. Thus, the mobile atoms can...
reach both large and small islands with equal ease, so the d–d repulsion does not cause SC during irreversible growth. This is confirmed by the KMC simulations accounting for the dipole–dipole interactions reported in [6] where rectangular islands were grown but no narrowing of the ISDs was found.

In the case of the dipole–monopole forces the potential (21) at the nearest-neighbor distance from the square island boundary is

\[ E(i_{NN}) \approx V_{aa}^{d-m}(a/2 + 1, 0)|_{a \to \infty} \sim Ca \ln a \]

which means that in contrast to the dipole–dipole interaction, the potential based on the dipole–monopole forces does differentiate between mobile adatom capture by large and small islands, as can be seen from (24) (we recall that \( C > 0 \) in the repulsive case),–thus providing a mechanism for kinetically controlled SC.

4.3. KMC simulation of strained epitaxy

To simulate the growth with realistic strain in our model we need to chose the value of the constant \( C \) in (21). For consistency we chose it in such a way that islands with \( a = 1 \) corresponding to isolated adatoms asymptotically reproduced the dipole–dipole potential (17). After some algebra the asymptotic of (21) was found to be \( C/(12r^3) \), so \( C = 12\gamma \) would ensure that at distances larger than 11.0. the interaction of an island consisting and of an adatom will have the same strength as the adatom–adatom interaction (17).

The range of numerical values of \( \gamma \) used in the simulations was chosen according to our estimate made in section 3.2 (\( \gamma \approx 3 \) meV) and in [7,45] (5 and 15 meV, respectively). Because the estimated values vary in a rather broad range and \textit{ab initio} estimates of non-local interactions are known to be unreliable [32], the simulations were carried out for five values of \( \gamma = 0, 2.5, 5, 10 \) and 20 meV.

The main results of our KMC simulations are shown in figures 4 and 5. Two conclusions can be drawn from the data. The value of index \( \omega \) introduced in [46] from the power-law dependence

\[ s_{av} \propto \theta^{-\omega} \]

(26)
can be found from our data on \( d_{av} \approx \sqrt{s_{av}} \propto \theta^{-\omega/2} \). This index is convenient for experimental measurement because it is directly connected to the index characterizing the total island density

\[ N = \theta/s_{av} \propto \theta^{1+\omega} \]

(27)

Our first conclusion is that the index strongly depends on the strength of the elastic forces, especially at small \( \gamma \), as can be seen from table 1. Experimentally, the smaller values of \( \omega \) may look as the growth saturation, as seems to be the case in [47]. The second conclusion is that in the presence of strain the dispersion \( \sigma \) of the island diameter distribution (IDD) exhibits a saturated behavior, as can be seen from figure 4(b). In combination with monotonous growth of \( d_{av} \) the dispersion to mean diameter ratio diminishes with coverage in qualitative agreement with experimental data [47].

4.4. Discussion

The dispersion to mean diameter ratios in figures 4(c) and 5(b) are not as small as in some experimental data that exhibit the best cases of SC. One of the reasons is that in
Figure 4. Log–log plots of the coverage dependences of the mean island diameters (a), of the diameter distributions dispersions (b), and of the dispersion to the diameter ratios (c) for five values of the interaction parameter $\gamma$: 0 (+), 2.5 meV (●), 5 meV (○), 10 meV (×), and 20 meV (△). Solid lines are linear fit to the data; dashed lines are guides to the eye. Note that only $\gamma = 0$ data exhibit the scaling behavior.

Figure 5. Temperature dependences of the mean island diameter $d_{\text{av}}$ (a) and of the ratio of the diameter distribution dispersion $\sigma$ to $d_{\text{av}}$ (b) at coverage $\theta = 0.2$ for different strength of the elastic interaction; notation is the same as in figure 4.
Table 1. Dependence of the scaling exponent $\omega$ as defined in (26–27) on the strength of the long-range interaction parameter $\gamma$ (17).

| $\gamma$ (meV) | $\omega$  |
|---------------|-----------|
| 0             | −0.82     |
| 2.5           | −0.59     |
| 5.0           | −0.48     |
| 10            | −0.39     |
| 20            | −0.34     |

In our calculations we used the standard formulas of statistics and took into account all available IDD data from the smallest to the largest island sizes. Because they contain a tail in the IDD curves at small diameters (see figure 3), the values of $d_{av}$ are smaller than diameter values at the IDD maxima. This augments both $\sigma$ and $\sigma/d_{av}$ values. Here, it is pertinent to note that similarly asymmetric IDDs are rather commonly observed experimentally (see, e.g. [19,20,47,48]). But because experimental data as a rule contain more different structures than exist in our simple model, size distributions for islands of different morphologies are usually studied separately. Therefore, usually only the data in the vicinity of the maximum density of islands of a given morphology are taken into account in the processing of experimental data. Thus, for example, the value of the diameter at the IDD maximum for a given kind of island is taken for $d_{av}$ and the IDDs widths are calculated only in its vicinity [19, 20, 47]. In [47], for example, the relative FWHM with respect to $d_{av}$ taken to be equal to $d$ at the IDD maximum was found to be quite small (15%). But as can be seen from figure 3, in our case it has similar value $\sim 14\%$. It is common to fit experimental IDD with the Gaussian curve. But for the latter the FWHM $\approx 2.35\sigma$ so the effective value of $(\sigma/d_{av})_{eff}$ with such data processing can be as small as 0.06. Thus, the SC mechanism proposed may underly even the best cases of SC seen experimentally.

5. Conclusion

To conclude, in this paper we suggested a kinetic mechanism of the SC in strained epitaxy. It differs from the kinetic mechanisms in the presence of the Ostwald ripening [47, 48] in that there is no need for atomic detachments. This means that the mechanism can be operative at smaller temperatures which may have important practical advantages. But because of similar growth behavior, the mechanism can contribute to the phenomena attributed to the ripening. Furthermore, it may underly the unusual narrowing of IDD with temperature observed in the QDs growth in [19,20] (see figure 5). Such behavior is qualitatively different from that observed both in thermodynamically controlled growth and in the kinetically controlled growth in the absence of strain.

The proposed mechanism of SC heavily relies on the island size dependent monopole forces due to the misfit shear strain in the substrate, so the strength and spatial extent of the forces are of crucial importance. The simple rigid-core model [11] we used to illustrate our mechanism is presumably good for small islands simulated in the present paper and for those grown experimentally in [12]. For very large islands, however, saturation toward
the constant monopole density similar to that on the surface steps should be expected. Calculations in [49] revealed the shear strain that linearly varies across the QD/substrate interface in capped InAs/GaAs pyramids of at least 12 nm in diameter. Experimentally large interface shear strain of considerable spatial extent caused by the Ge/Si(001) QDs of an order of magnitude larger diameter was observed in [50]. This may mean that the mechanism described in the present paper contributes to the SC of QDs of all sizes. But even restricted to islands a few nanometer in diameter, the mechanism would still be of considerable practical interest. The maximal catalytic efficiency is achieved in size-calibrated metallic islands of small diameters [38]. In semiconductor heteroepitaxy the quantum size effect that allows for variation of the QD photoluminescence wavelength is operative only in small QDs. The model of monolayer–high islands that we studied in the present paper can be taken as a starting point for modelling the growth of the submonolayer QDs [28]. Their small spatial dimensions may allow for the fabrication of the most compact QD devices due to dense QD packing.

Finally, in the course of our study we derived a simple expression for the dipole–monopole interaction for in-plane displacements and forces that can be generalized to 3D in case of necessity. This expression can be used in other studies of nucleation and growth unrelated to SC. For example, in studying the influence on the growth of QDs and QD molecules of the surface steps, islands and pits that are usually present in strained epilayers [51].

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