A Level Set Simulation for the Strain-Driven Sharpening of the Island Size Distribution during Heteroepitaxy

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We use an island dynamics model for heteroepitaxial growth to study the narrowing and sharpening of the island size distribution as a function of the strain in the submonolayer growth regime. Our island dynamics model is coupled to an elastic model that is based on atomistic harmonic interactions. The elastic equations are solved self-consistently at every timestep during the simulation for the entire system. This is possible because the numerical timesteps in the island dynamics model that is based on the level set technique are significantly larger than the timestep of a typical atomistic event such as adatom diffusion and detachment, while we still retain all the relevant physics that are associated with adatom diffusion and detachment.

Semiconductor quantum dots (QDs) are the focus of many current research efforts, and hold great promise for many technological applications such as next generation opto-electronic devices [1]. A well established fabrication process is to grow QDs epitaxially via molecular beam epitaxy (MBE). For optimal performance of these QDs and arrays of QDs it is neccessary for the size distribution of the dots to be rather uniform. This requires very precise control of the growth and fabrication of QDs, and it is thus of paramount importance to have a fundamental understanding of the processes during the growth of QDs, and how to control them.

Typical semiconductor systems used for QDs have a lattice mismatch that leads to strain in the system. It is 4% for Ge/Si, and up to 7% for $In_xGa_{1-x}As$/GaAs (depending on the composition parameter x). It has been established almost 20 years ago that this strain facilitates the formation and self-organization of QDs [2, 3]. However, the exact effect of strain on the driving forces for the formation of QDs is still not completely understood. For example, is the formation of QDs ultimately a thermodynamic effect, or does strain mainly influence the kinetics during growth? To answer such questions, we need models that faithfully include the effects of strain.

It has been notoriously difficult to properly include strain in a full 3-dimensional simulation of epitaxial growth for systems of reasonable (and meaningful) size. The reason is that solving the elastic equations is rather expensive. It is almost prohibitively expensive to solve the elastic equations at every timestep in an atomistic simulation, such as a kinetic Monte Carlo (KMC) simulation, where a typical numerical timestep is the inverse of the diffusion constant, and is often $O(10^{-6}$s) (or smaller). We note that recent progress has made it possible to to such KMC simulations [4, 5], but the system sizes studied are rather small, and at present do not allow any statistical analysis.

One way out of this dilemma is to not solve the elastic equations globally at every timestep, but to only solve them after a certain number of timesteps, and/or to solve them only locally (whereever the last event took place). A global update is then done periodically [6]. Clearly, this approach has to be done rather carefully. Some insight can also be gained by focusing on 2-dimensional models, where it is a lot faster to solve the elastic equations [7, 8], or to study continuum-type models [9] that typically stress thermodynamic arguments, but do not include the detailed kinetics. An alternative approach that we will decribe below is to build a model where the numerical timestep is significantly larger, but where the model still properly accounts for the relevant atomistic events. We also note a number of models where the main effect of strain is effectively accounted for by assuming an island size and/or height dependent detachment rate [10–12].

In this communication, we present an island dynamics model that employs the level set technique [13–15] for the strain driven regularization of islands during heteroepitaxy. A virtue and feature of this method is that we can solve the elastic equations for the entire system at every timestep during the simulation. This is possible because a typical numerical timestep is $O(10^{-2}$ - 10^{-3} s), which is orders of magnitude larger than the timescale of microscopic events such as diffusion. Nevertheless, all the atomistic processes are included within this method. We show that strain leads to a regularization of island sizes in the submonolayer growth regime, as is evident from the narrowing and sharpening of the island size distribution (ISD). This result will be compared to experimental data for $In_xGa_{1-x}As$ on GaAs(100) [16].

In our model for epitaxial growth, islands are described by a level set function, and the growth of the islands is described by the time evolution of the level set function [13–15]. The velocity of the island boundaries is then obtained from solving the following diffusion equation for the adatom concentration $\rho(\mathbf{x}, t)$:

$$\frac{\partial \rho}{\partial t} = F + \nabla \cdot (\mathbf{D} \nabla \rho) - 2\frac{dN}{dt} + \nabla \cdot \left(\frac{\rho}{k_B T} \mathbf{D} (\nabla E_{\mathrm{ad}}) \right) \quad . \tag{1}$$

In Eq. (1), $\mathbf{D}$ is a diffusion tensor where the diagonal entries are $D^{(i)}(\mathbf{x})$ and $D^{(j)}(\mathbf{x})$. F is the deposition flux, dN/dt is the nucleation rate, and the last term is the thermodynamic drift, where k_B is the Boltzmann constant, and T is the temperature. The nucleation rate is given by

$$dN/dt = \sigma_1 \langle [(D^{(i)}(\mathbf{x}) + D^{(j)}(\mathbf{x}))/2] \rho^2(\mathbf{x}) \rangle \quad . \qquad (2)$$

where σ_1 is a capture number [17, 18], and the average $\langle \cdot \rangle$ is taken over all lattice sites.

Stochastic elements are required to properly describe island nucleation and the thermal dissociation of small islands. In particular, the rate of nucleation is deterministic, as described by Eq. (2), but the spatial position of a newly nucleated island is chosen with the probability that is weighted by the local value of $\rho^2(\mathbf{x})$ [19]. Similarly, island breakup is correlated with the local detachment rate $D_{\mathrm{det}}(\mathbf{x})$ and the probability to shrink below the size of a dimer [13]. Once an island has been broken up, we assume fast diffusion and for simplicity distribute the mass of the island uniformly over the entire lattice. We note that the detachment rate used in our model is an effective detachment rate, that is the average rate for an atom to detach from a boundary (regardless of coordination), *and* to subsequently diffuse out of the capture area of the islands. In other words, we allow for detachment, but do not resolve every detachment and subsequent reattachment event [13]. We also note that a coordination (or island shape) dependent detachment rate could be incorporated, but this would not affect the basic message of this communication.

For the solution of Eq. (1) we enforce the boundary condition

$$\rho_0(\mathbf{x}) = \rho_{\mathrm{eq}}(D_{\mathrm{edge}}(\mathbf{x}), D_{\mathrm{det}}(\mathbf{x}), \mathbf{x}) \quad , \qquad (3)$$

where $D_{\mathrm{edge}}(\mathbf{x})$ is the spatially varying microscopic rate for edge diffusion [20]. We highlight in particular the dependence on D_{det}: the higher the detachment rate of adatoms, the higher the value of ρ_0 at the island boundary.

Our elastic model is essentially an atomistic cubic model that includes harmonic nearest and next nearest neighbor terms. We assume that nearest neighbor springs are twice as strong as next nearest neighbor springs. We define a discrete profile from the level set function as follows

$$h_{ij} = [\phi_{ij}]$$

where $[\cdot]$ denotes the integer part. At each grid point in the film there is a displacement field defined, $\mathbf{u}_{ijk}$. These displacements satisfy a large linear system that is given in Ref [5]. The total elastic energy can be written as

$$W = \frac{\lambda}{2} \sum_{\mathrm{allatoms}} E_{\mathrm{strain}}(\mathbf{x})$$

where $E_{\mathrm{strain}}(\mathbf{x})$ is the energy contained in all the springs connected to the atom located at position $\mathbf{x}$. The expression we use is an extension to three dimension from the two dimensional formula found in Ref. [21]. The scale factor λ is chosen so that the strain energy per atom in a fully strained system is approx. 0.04 eV for a system with 1% misfit, and 0.6 eV for a system with 4% misfit. These numbers are plausible for typical semiconductor systems, but we note that all our results can easily be re-scaled (i.e., when we quote below results for 1% and 5% misfit, this might be a slightly different misfit for a particular system). Moreover, the main purpose of this communication is to report a qualitative trend, as strain increases.

For each time step we demand the the film is mechanical equilibrium. This entails solving a large linear system. In addition, we incorporate the effect of a semi-infinite substrate by using an artificial boundary condition. The resulting system is solved with a multigrid-Fourier method. For details see Ref. [22].

In principle, all of the microscopic parameters $D^{(i)}(\mathbf{x})$, $D^{(j)}(\mathbf{x})$, $D_{\mathrm{det}}(\mathbf{x})$, and $D_{\mathrm{edge}}(\mathbf{x})$ are affected by strain. We find that the effect on $D_{\mathrm{edge}}(\mathbf{x})$ is essentially irrelevant (since islands are always rather compact within the model), and leave $D_{\mathrm{edge}}(\mathbf{x}) = 0$ (a higher value does essentially have no effect). We parameterize the adatom diffusion $D^{(i)}(\mathbf{x})$ and $D^{(j)}(\mathbf{x})$ according to density-functional theory (DFT) results for a typical semiconductor system [23, 24], but also find that the effect of a strain dependent diffusion is almost irrelevant for the strain driven regularization of the ISD discussed below. This is different from our work on stacked quantum dots [24], where we found that the strain-driven variation of $D^{(i)}(\mathbf{x})$ and $D^{(j)}(\mathbf{x})$ is crucial for the *placement* of islands.

The main effect of strain is the dependence of D_{det} on it. We are not aware of any DFT results or other systematic study of the dependence of D_{det} on strain. But we believe that it is quite plausible that detachment is enhanced upon both, compressive and tensile strain, and preliminary (unpublished) DFT calculations by us support this assumption. More precisely, we assume that detachment is enhanced according to

$$D_{\mathrm{det}}(\mathbf{x}) = D^0_{\mathrm{det}}(\mathbf{x}) \exp((E_{\mathrm{strain}}(\mathbf{x})/k_B T) \quad , \qquad (4)$$

where $D^0_{\mathrm{det}}(\mathbf{x})$ is the detachment rate for the unstrained system.

The effect of strain on our model is illustrated in more detail in Fig. 1. We show a typical island morphology [(a) and (e)] the resulting strain energy [(b) and (f)], and its effect on the detachment rate [(c) and (g)] and boundary value $\rho_0(\mathbf{x})$ [(d) and (g)] for a system with small strain (left) and high strain (right). It is evident that the strain energy in the middle of the islands as well as around the island edges is significantly higher for the system with larger strain (f). As a result, the detachment rate as

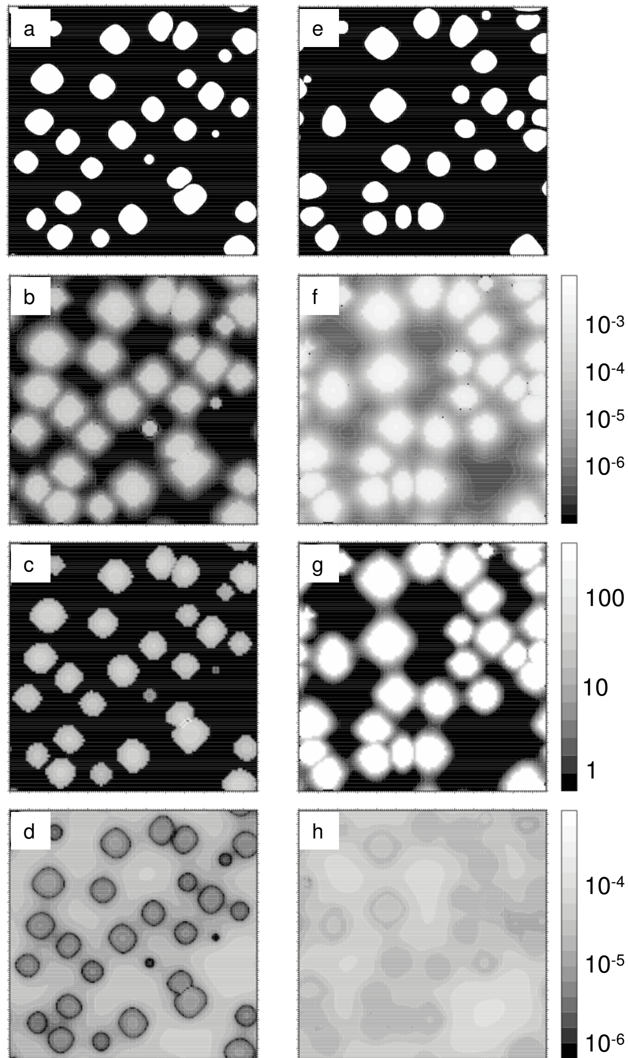

FIG. 1: The effect of strain in our model. We show results for 1% strain (left column), and 5% strain (right column). Shown are typical morphologies after a coverage of 20% (a) and (e), the corresponding elastic energy on the surface (b) and (f), the resulting detachment rates (c) and (g), and adatom concentrations (d) and (h). The units of the elastic energy are in terms of the spring constants that are O(1). The units of the adatoms concentration are adatoms per lattice sites. The units of the detachment rate are the number of detachment events per second. Note that the detachment rates are effective numbers that are smaller than in an atomistic (KMC) simulation; we only consider detachment events where the adatoms do not re-attach to the island.

calculated according to Eq. (4) is significantly enhanced (g), which leads to a higher value for ρ_0 (h). We note that $D_{\rm det}$ and ρ_0 are defined for the entire system [as can be seen in Figs. 1(c), (d), (g), and (h)], but they only have physical meaning around the island boudaries.

The increased value for ρ_0 at the island boundary implies that the gradients of ρ are less steep at the boundaries of islands with higher strain, so that islands with more strain grow slower than less strained islands. Since smaller islands are less strained than larger islands, strain slows down growth of the larger islands more than growth of the smaller islands, which leads to a regularization of the island sizes. This can be seen by comparing Figs. 1(a) and (e), where it appears that there are more really small islands in (a), and also a few rather large ones.

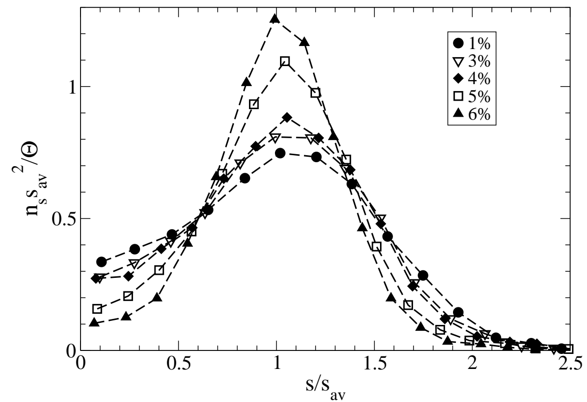

FIG. 2: The scaled island size distribution after the deposition of 20% of a monolayer for different values of strain. n_s is the number of islands of size s, $s_{\rm av}$ is the average island size, and Θ is the coverage.

More convincing and quantitative evidence of the regularization of the island sizes can be seen in Fig. 2, where we show the scaled ISD after the deposition of 20% of a monolayer, as a function of strain. The scaled ISD for the system with more strain is clearly narrower and sharper, which is equivalent to the statement that the ISD is more regular. An additional contribution to the regularization of the islands is the fact that for systems with large strain, small islands (i.e., dimers) are less stable against break-up, because even for islands as small as a dimer, strain enhances the detachment rate (which is the same as the break-up rate for a dimer). It is well known that enhanced break-up for dimers also leads to a sharper ISD. The reason is that small islands that have nucleated in an unfavorable spot (close to existing, bigger islands) have an enhanced chance to break-up, and to subsequently re-nucleate in a more favorable spot. If the islands are located further apart from each other, they compete less for additional adatoms, and are also less strained. Therefore, they can grow to islands with a more regular ISD. This is the reason why our results differ from the ones reported by Nandipati and Amar [12], who presented results for the effects of strain on the ISD for irreversible growth.

Our results agree well with experimental data for $In_xGa_{1-x}As$ on GaAs(100) by Leonard *et al.* [16]. In their experiment, the authors of Ref. (16) varied the composition parameter x, without changing any other growth condition, and reported that the ISD is getting sharper

and narrower.

The results presented in this communication illustrate that an island dynamics model that is coupled to the level set method is well suited to model epitaxial growth of strained systems. We have focused in this communication on the submonolayer growth regime. As a next steps, we will show that this method can also be extended to model multilayer growth, and the formation of full QDs. But some challenges still remain for this extension. In particular, the boundary condition (3) needs to be changed to the more general mixed Robin boundary condition $\partial\rho/\partial n + \alpha\rho = \beta$, to account for an additional step-edge barrier for interlayer mass transport, and its dependence on strain. This requires new numerical schemes to solve the diffusion equation (1), and is work that is currently in progress.

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