A Level-Set Method for Self-Organized Pattern Formation during Heteroepitaxial Growth

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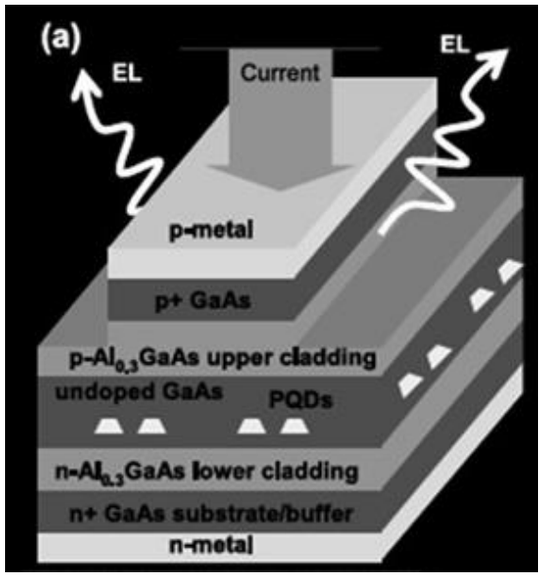
- Russ Caflisch

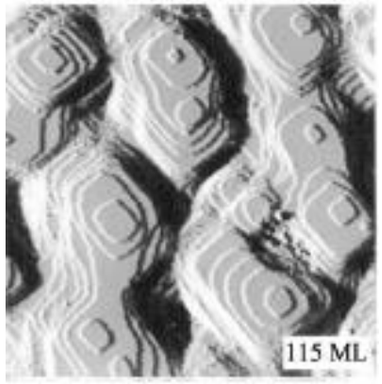
Motivation

- Many technologically important devices are grown via epitaxy.
- Often, the dimension in the x-y-plane is large (continuous), while an atomic height resolution is required.
- Strain leads to ordering

Embedded Quantum Dots in AlGaAs system for LEDs

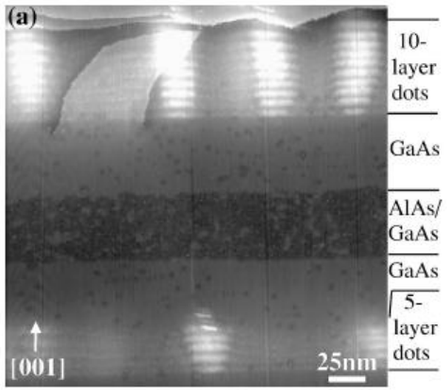


Cu on Cu(100)



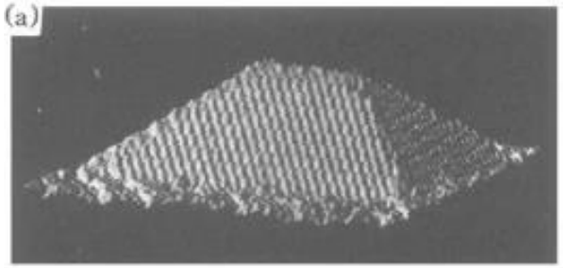
Zuo et al., PRL 78 (1997)

$Al_xGa_{1-x}As$ system



B. Lita et al., APL **74**, (1999)

Self-Organized hut formation Ge/Si(001)



Y.-W. Mo et al., PRL **65** (1990)

We want a model for epitaxial growth that is efficient, has atomic (height) resolution, and provides a natural framework to include strain.

The Challenge for Including Strain in a Growth Model

Strain arises when the material that is deposited has a different lattice constant than the substrate. For example, Ge is 4% bigger than Si.

- Strain calculations for a system of typical size in 2+1 dimensions are expensive (at least seconds, maybe minutes).
- A typical timestep in an atomistic simulation is of order 10^{-6} seconds (which is the inverse of a typical diffusion constant $D=10^{6}$).
- Need of the order of 1 million timesteps (or more) to simulate 1 second.

Possible solutions to this challenge

1) Don't solve global elastic field at every timestep. Instead, solve it only locally (maybe not even at every timestep), and do occasional global updates.

2) Develop a model where the simulation timestep can be taken much larger, but where still all the microscopic dynamics are retained.

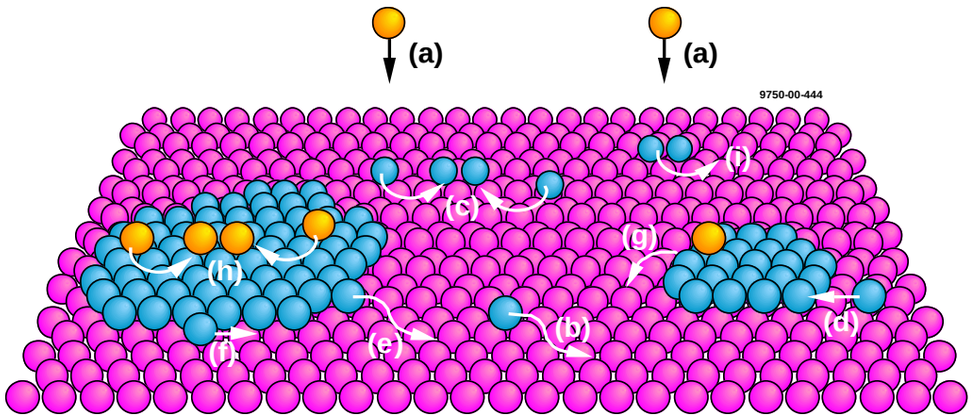
- We have developed a level set method.
- Typical timestep in the simulation is of order 10^{-2} seconds.

Outline

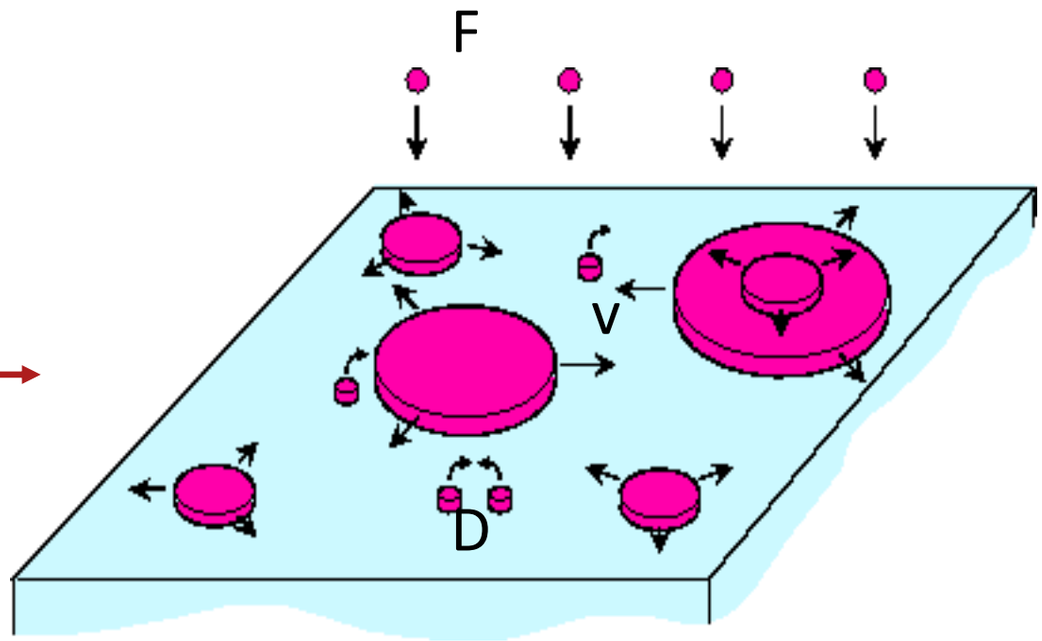
- Introduction
- The level-set method for epitaxial growth
- Our elastic model
- Strain-induced ordering in the submonolayer growth regime
- Ordering of stacked quantum dots
- Inclusion of a step-edge barrier to model mound formation

The Island Dynamics Model

Atomistic picture
(i.e., kinetic Monte Carlo)



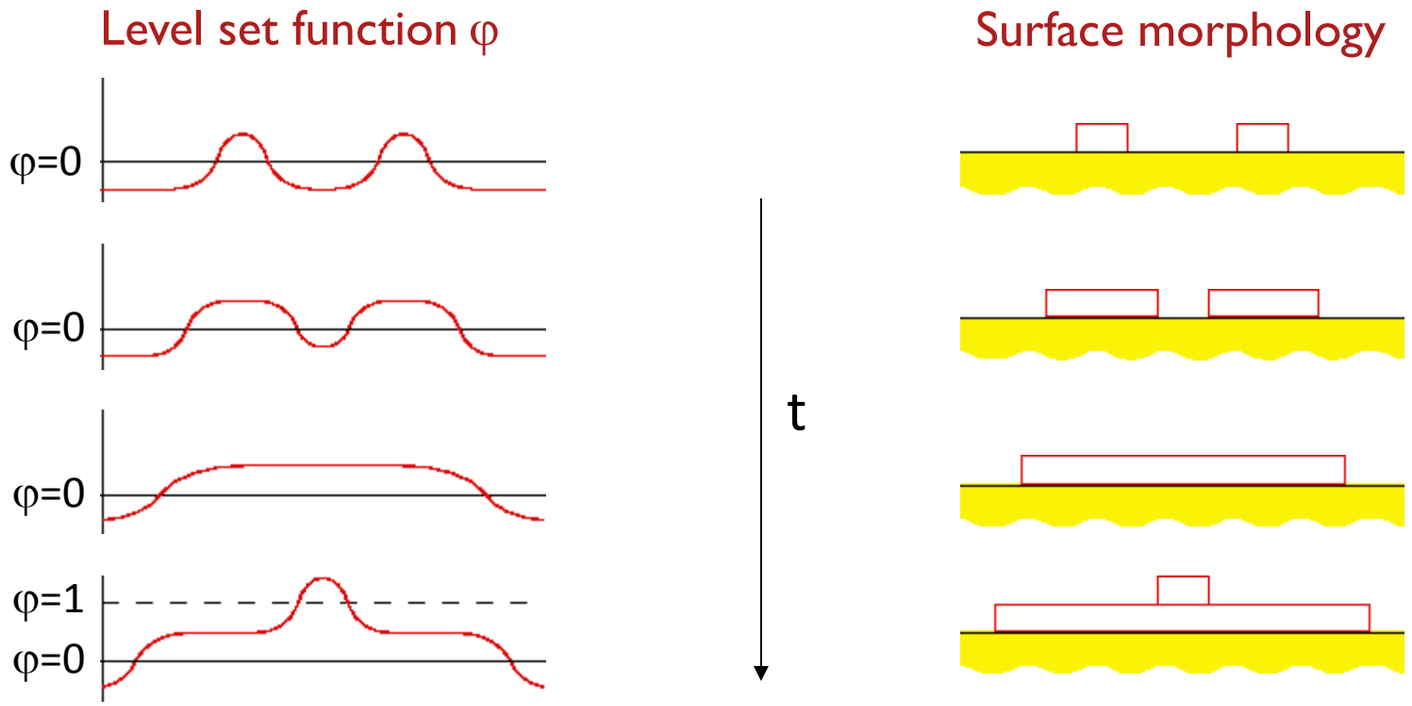
Island dynamics



- Identify all possible processes for all atoms.
- Assign a rate for each of these processes.
- Run a kinetic Monte Carlo simulation.

- Treat Islands as continuum in the plane
- Resolve individual atomic layer
- Evolve island boundaries with levelset method
- Treat adatoms as a mean-field quantity (and solve diffusion equation)

The Level Set Method: Schematic



- Level set function is continuous in plane, but has discrete height resolution
- Adatoms are treated in a mean field picture
- Governing Equation:

$$\frac{\partial \varphi}{\partial t} + v_n \mid \nabla \varphi \mid = 0$$

The Level Set Method: Formalism (Red Terms are Strain Dependent)

Diffusion equation for adatom concentration: $\frac{\partial \rho}{\partial t} = F + \nabla \bullet \mathbf{D}(\nabla \rho) - 2\frac{dN}{dt} + \text{drift}$

Nucleation rate ~ $D\rho(\mathbf{x},t)^2$

$\mathbf{D} = \mathbf{D}(\mathbf{x}) = \begin{pmatrix} D_{xx}(\mathbf{x}) & 0 \\ 0 & D_{yy}(\mathbf{x}) \end{pmatrix}$ is diffusion matrix.

Diffusion in x-direction Diffusion in y-direction

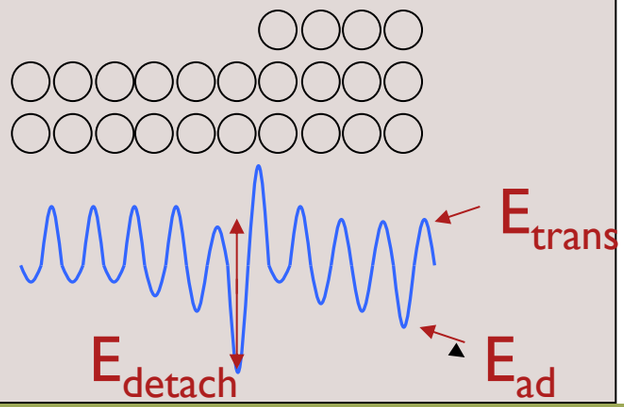
Boundary condition: $\rho = \rho_{eq}(\mathbf{D}_{det}, \mathbf{x})$

From Caflisch et al., PRE, 1999:

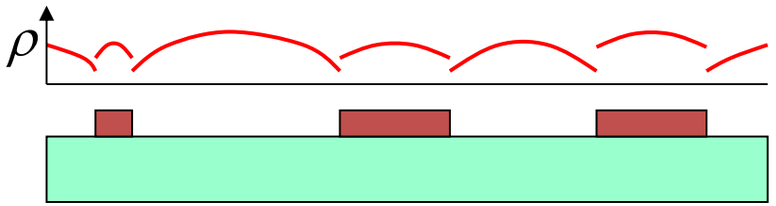
$$\rho_{eq} = \frac{D_{det}}{D_T}\frac{16}{3}\left(\frac{16}{15}\frac{LF}{D_E}\right)^{2/3}$$

$$D_{ii}(\mathbf{x}) \approx \exp(-(E_{trans}(\mathbf{x}) - E_{ad}(\mathbf{x}))/kT)$$

A typical potential energy surface



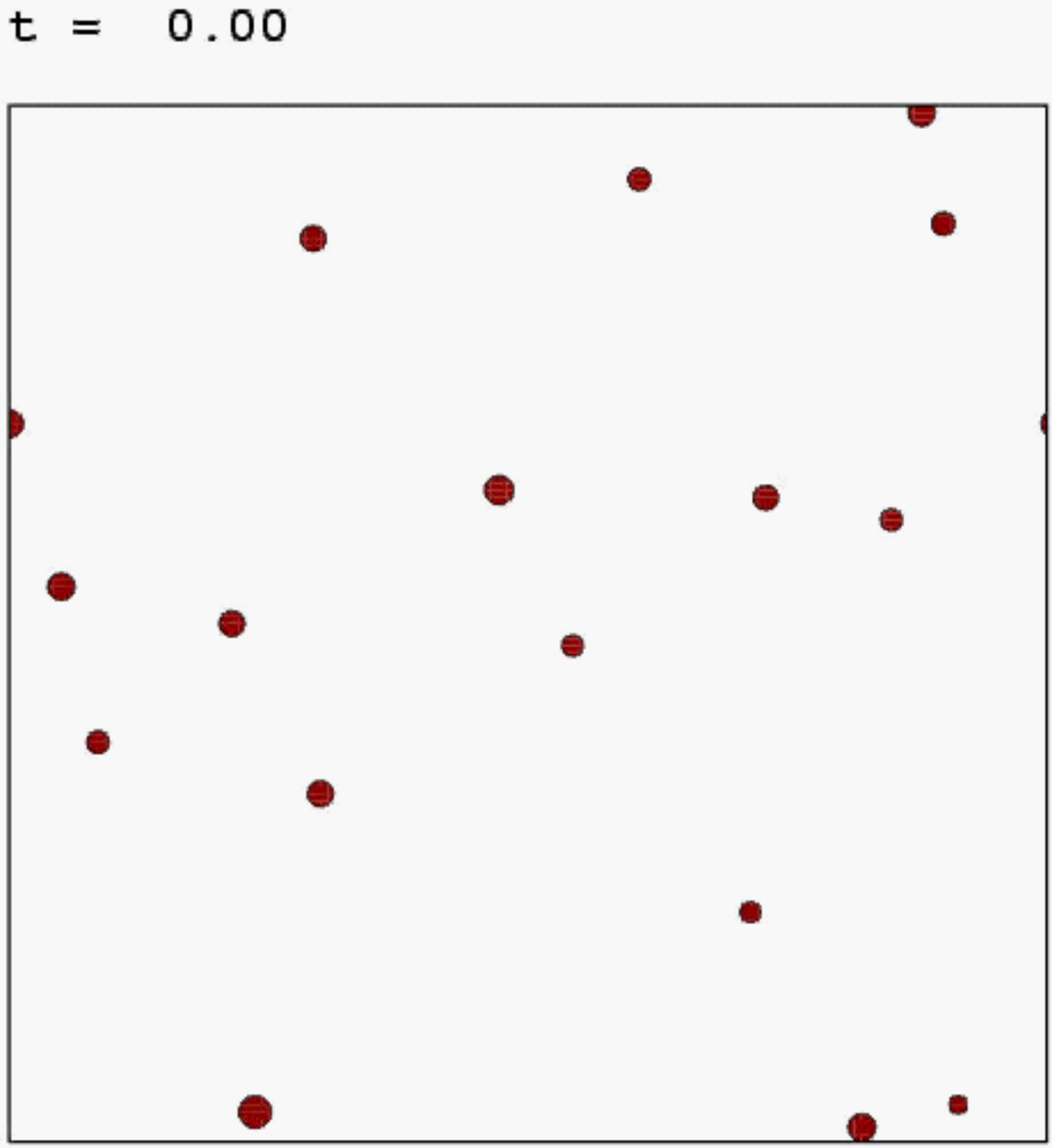
Typical solution of diffusion equation:



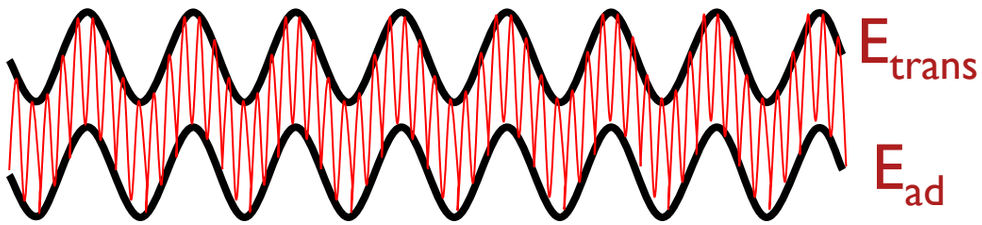
Velocity: $v_n = \mathbf{n} \bullet \mathbf{D}(\nabla \rho)^- - \mathbf{n} \bullet \mathbf{D}(\nabla \rho^+)$

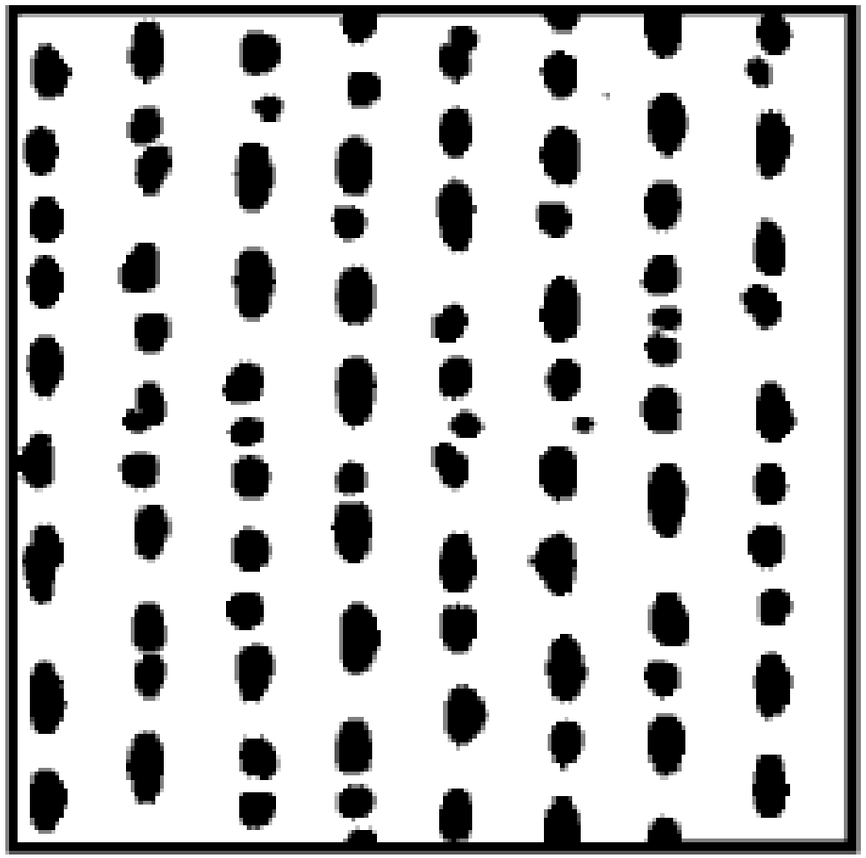
- Stochastic element needed for nucleation and dissociation of islands

A typical level set simulation

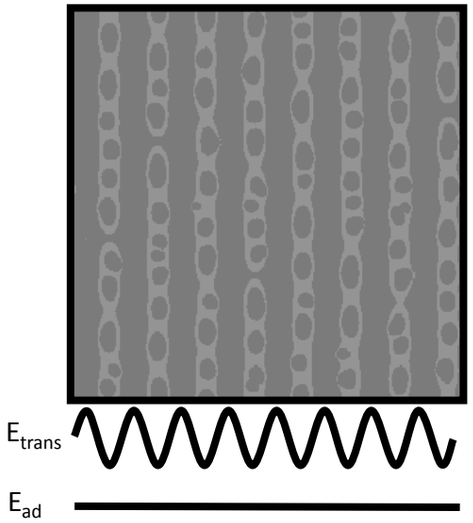


Variations of the Potential Energy Surface



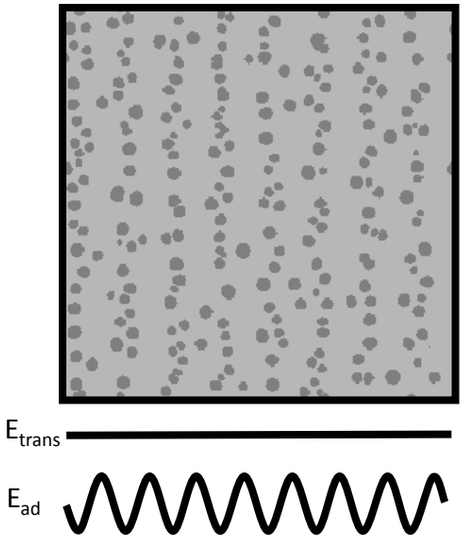


Kinetic limit



Nucleation in region of fast diffusion

Thermodynamic limit



Nucleation in region of slow diffusion (but high adatom concentration), dominated by drift

Nucleation rate ~ $D\rho(\mathbf{x},t)^2$

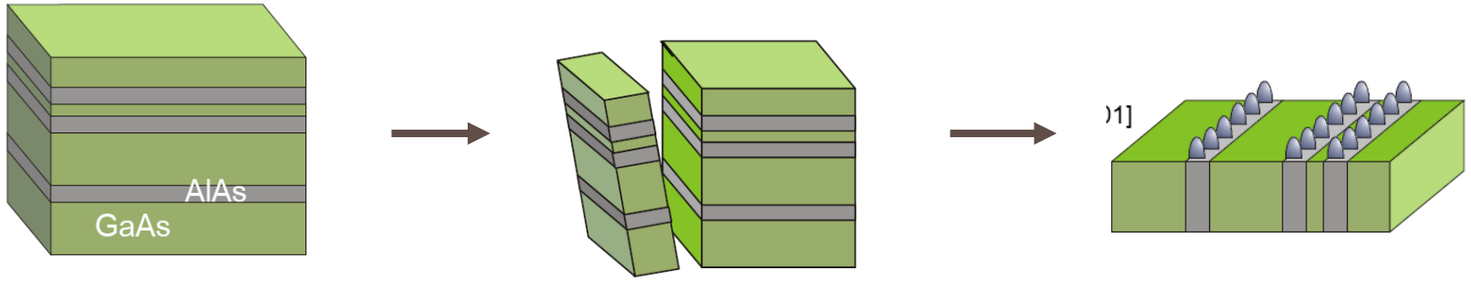
X. Niu, R. Vardavas, R.E. Caflisch, and C. Ratsch, Phys. Rev. B 74 (2006).

Ordering by Cleaved Edge Overgrowth

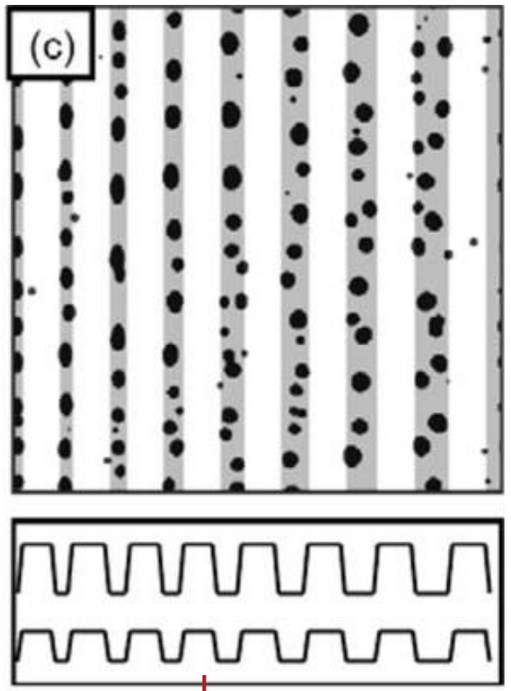
Grow AlAs/GaAs superlattice

Cleave and rotate

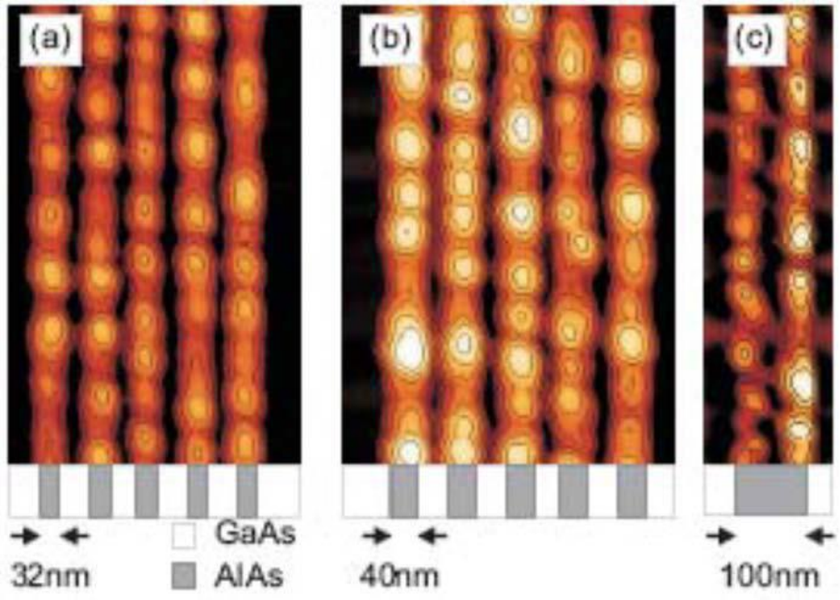
Grow InAs quantum dots



Simulations

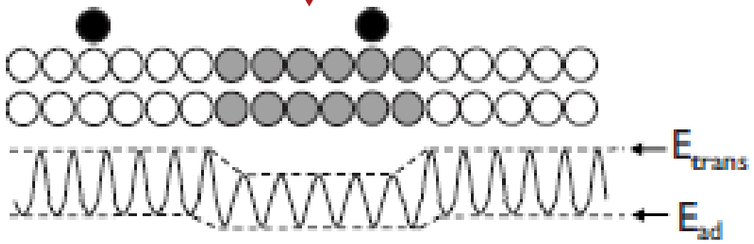


Quantum dots grow on top of the AlAs stripes



Hypothesis

- Variations of the PES between AlAs and GaAs surface lead to ordering
- We can test this with simulations

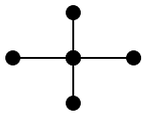


Work of E. Uccelli, A. Fontcuberta I Morral, G. Abstreiter, et al.

X. B. Niu, E. Uccelli, A. Fontcuberta i Morral, and C. Ratsch, APL 95 (2009)

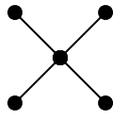
Include Strain: Calculate Elastic Field at Every Timestep

- Our Model: Harmonic ball-and-spring model that includes



Nearest neighbor springs

$$E = k(S_{xx}^{\ 2} + S_{yy}^{\ 2})$$



Diagonal springs

$$E = k_{diag}(S_{xx} + 2S_{xy} + S_{yy})^2 + k_{diag}(S_{xx} - 2S_{xy} + S_{yy})^2$$

- This can be related to (and interpreted as) a continuum energy density

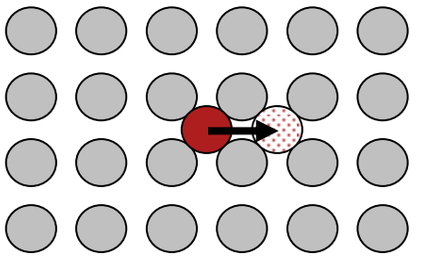
$$E = \alpha(S_{xx}^{\ 2} + S_{yy}^{\ 2}) + \beta S_{xy}^{\ 2} + \gamma S_{xx} S_{yy}$$

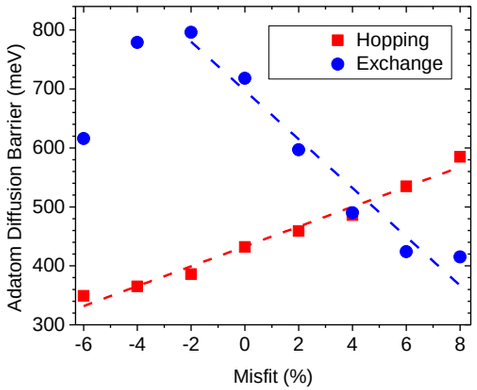
- Minimize energy with respect to all displacements: ∂_u E [u] = 0, to obtain strain at every lattice site.

- The relevant microscopic parameters at every grid point are then varied as a function of the local strain (or local displacement), according to the strain dependence calculated by DFT.

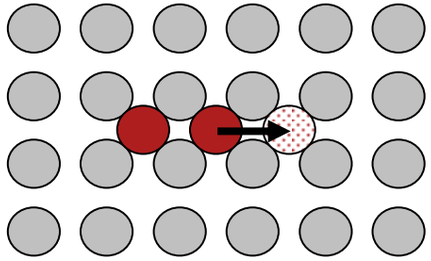
DFT Results for Strain Dependence for Microscopic Parameters for Ag on Ag(100)

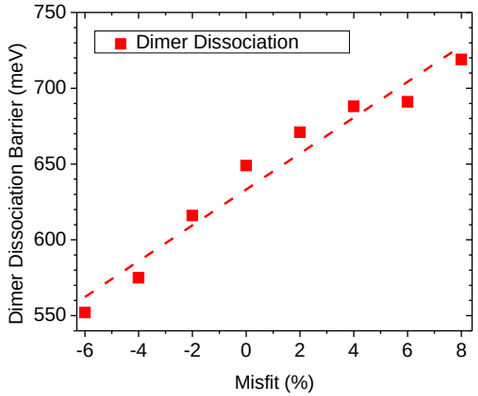
Adatom Diffusion



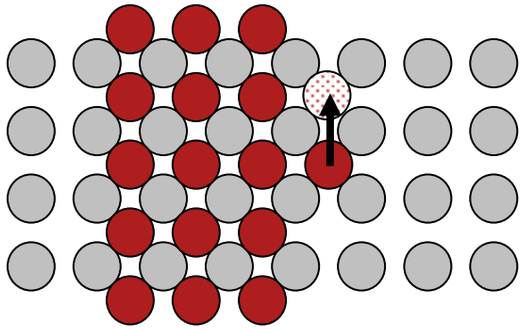


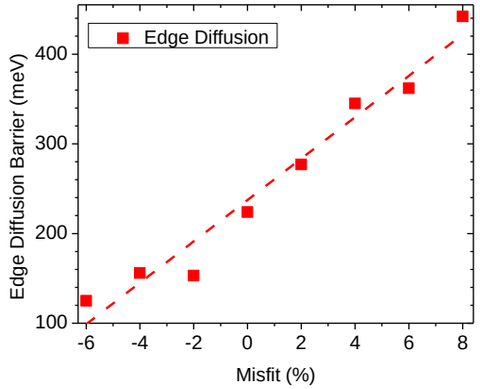
Dimer Dissociation



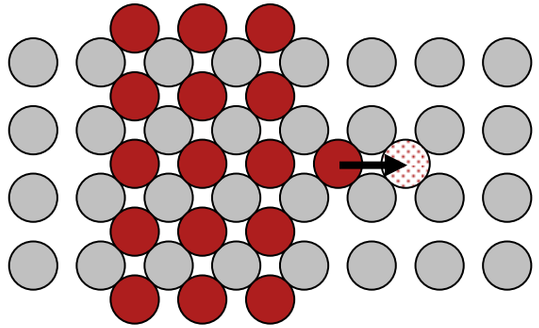


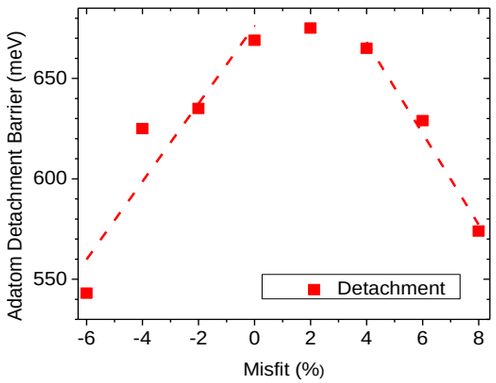
Edge Diffusion

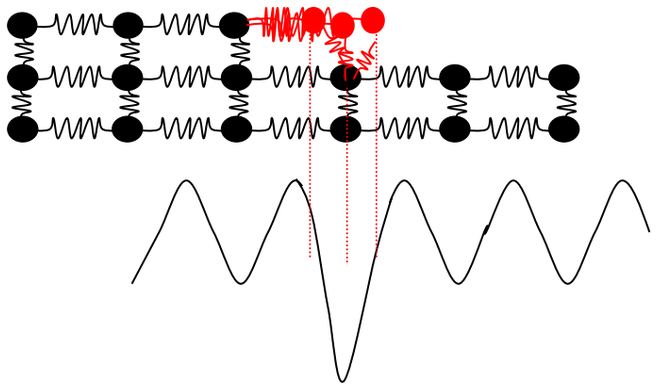




Detachment



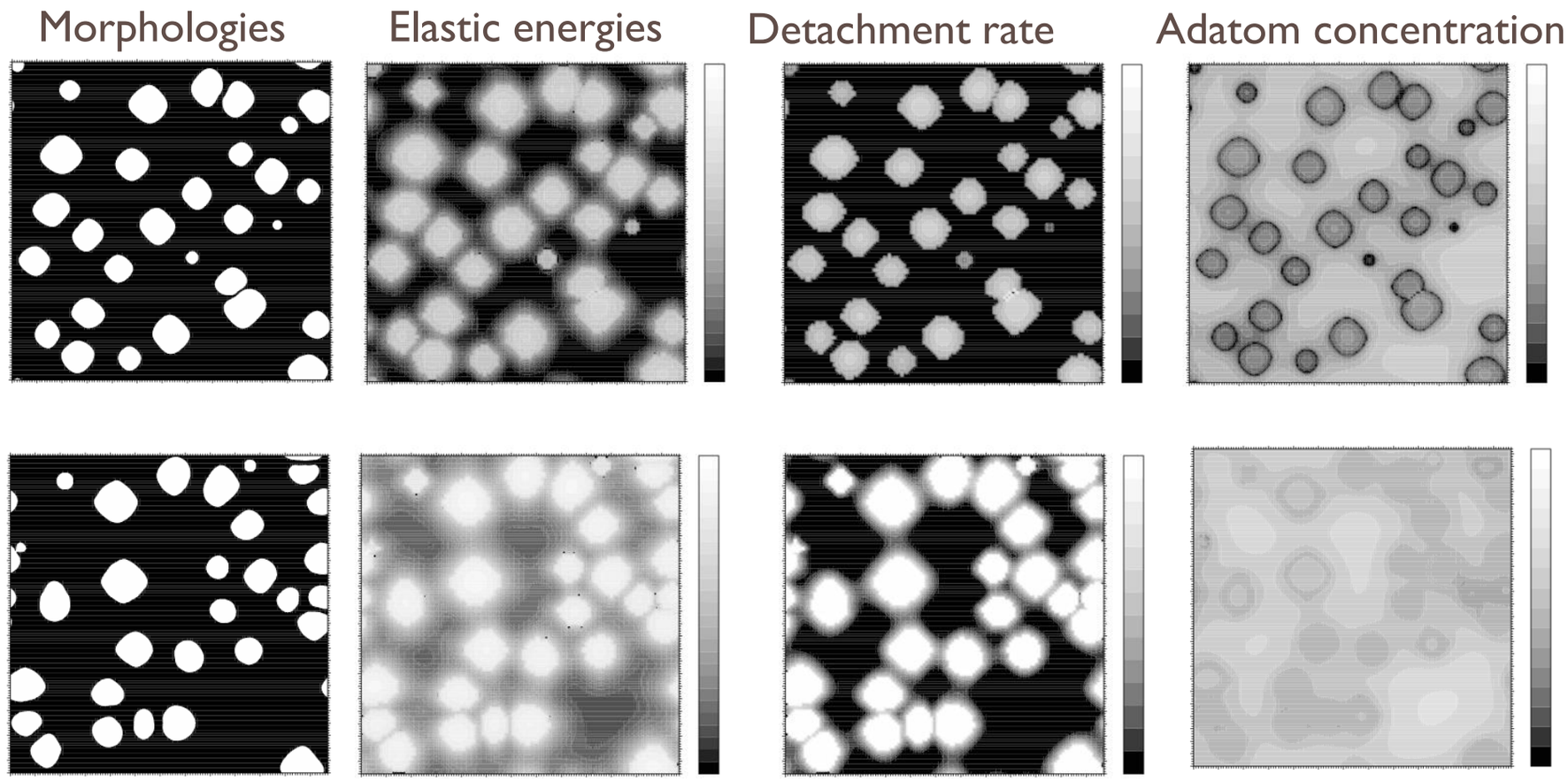




- DFT calculations where done with FHI-AIMS code.

C. Ratsch, Phys. Rev. B 83, 153406 (2011).

Effect of Strain in the Simulation

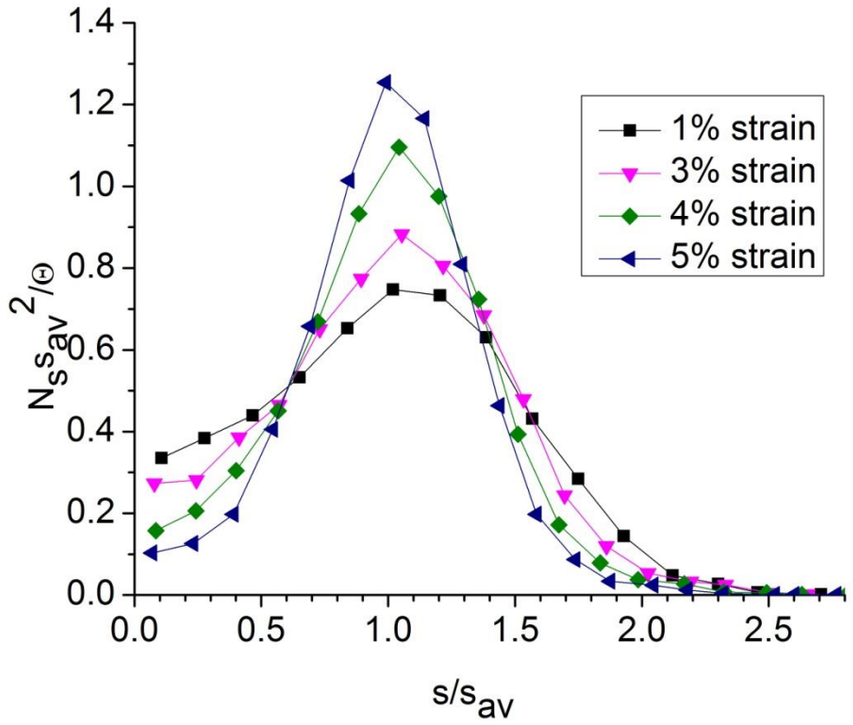


With increasing compressive strain, islands become more regular, because

- growth of large islands slows down, and
- small islands are more likely to break up.

Sharpening of the Scaled Island Size Distribution during Submonolayer Epitaxy upon Compressive Strain

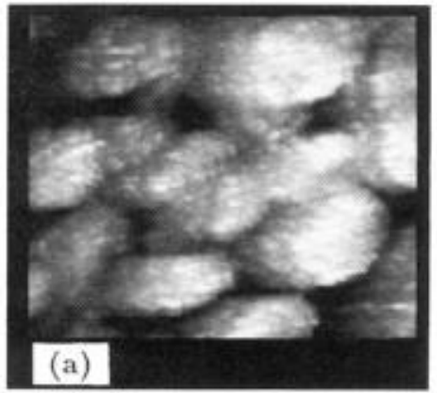
Level-set Simulation



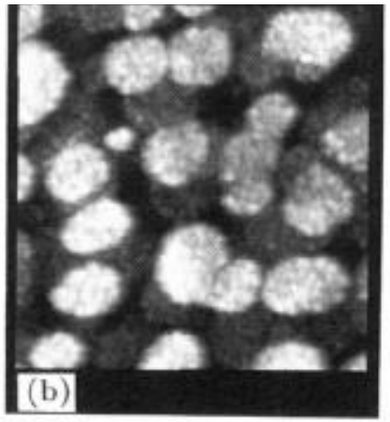
C. Ratsch, J. DeVita, and P. Smereka,
Phys. Rev. B **80**, 155309 (2009).

Experiment: $In_xGa_{1-x}As/GaAs(001)$

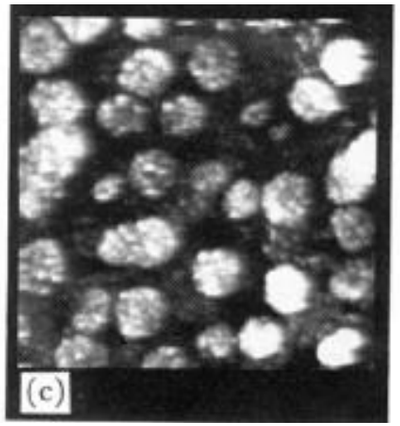
2.2% misfit



2.9% misfit



3.4% misfit



C.W. Snyder et al., Phys. Rev. Lett. 66, 3032 (1991).

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- Our elastic model
- Strain-induced ordering in the submonolayer growth regime
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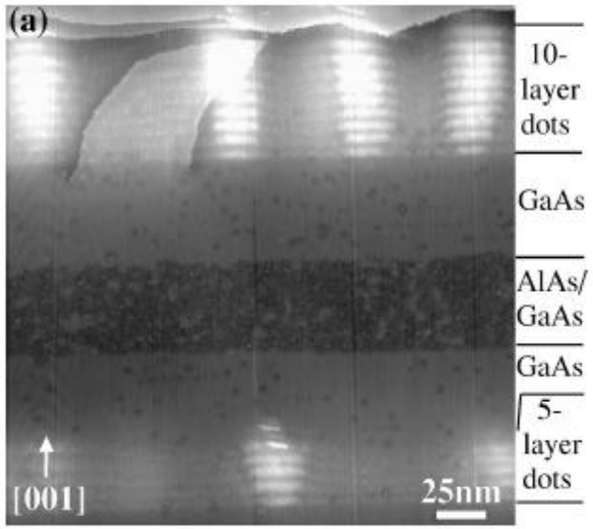
Simulation of Stacked Quantum Dots

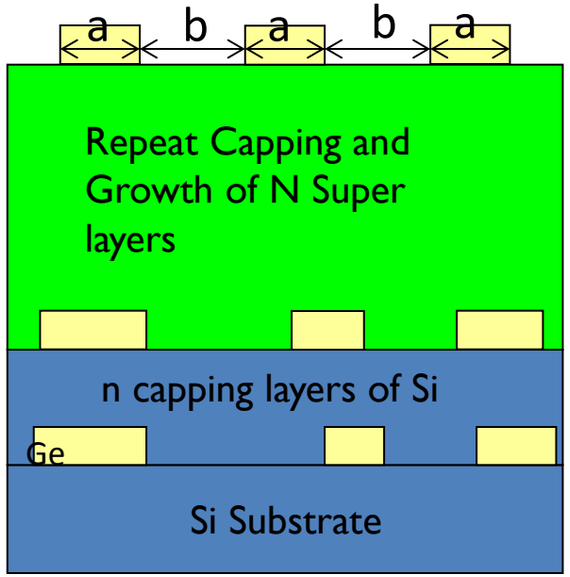
Experimental observation: Stacked quantum dots align under certain conditions

$Al_xGa_{1-x}As$ system



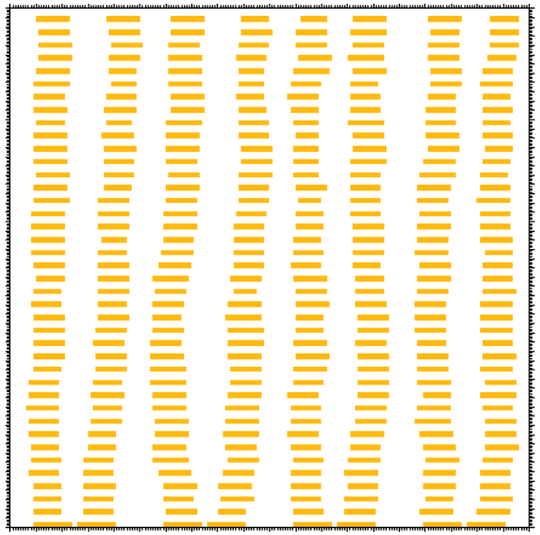
B. Lita et al., APL **74**, (1999).

Question/goal: can we understand and model this, and make some predictions and suggestions?

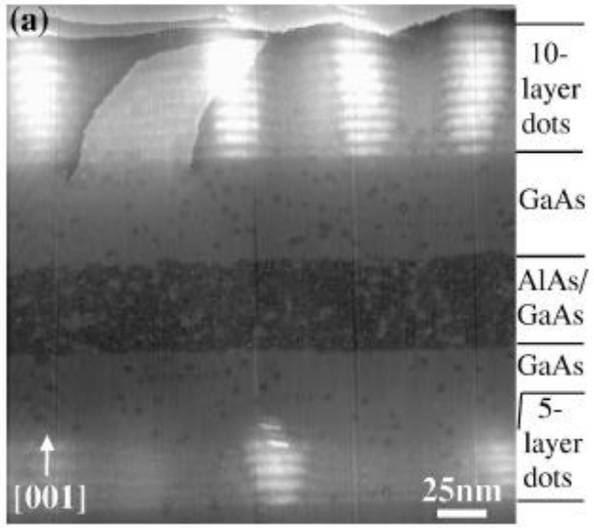


- Growth of islands on substrate without strain (constant diffusion and detachment)
- Fill in capping layer "by hand"
- Calculate strain on top of smooth capping layer
- Modify microscopic parameters for diffusion and detachment) according to strain
- Run growth model
- Repeat procedure

Ordering of Stacked Quantum Dots



$Al_xGa_{1-x}As$ system

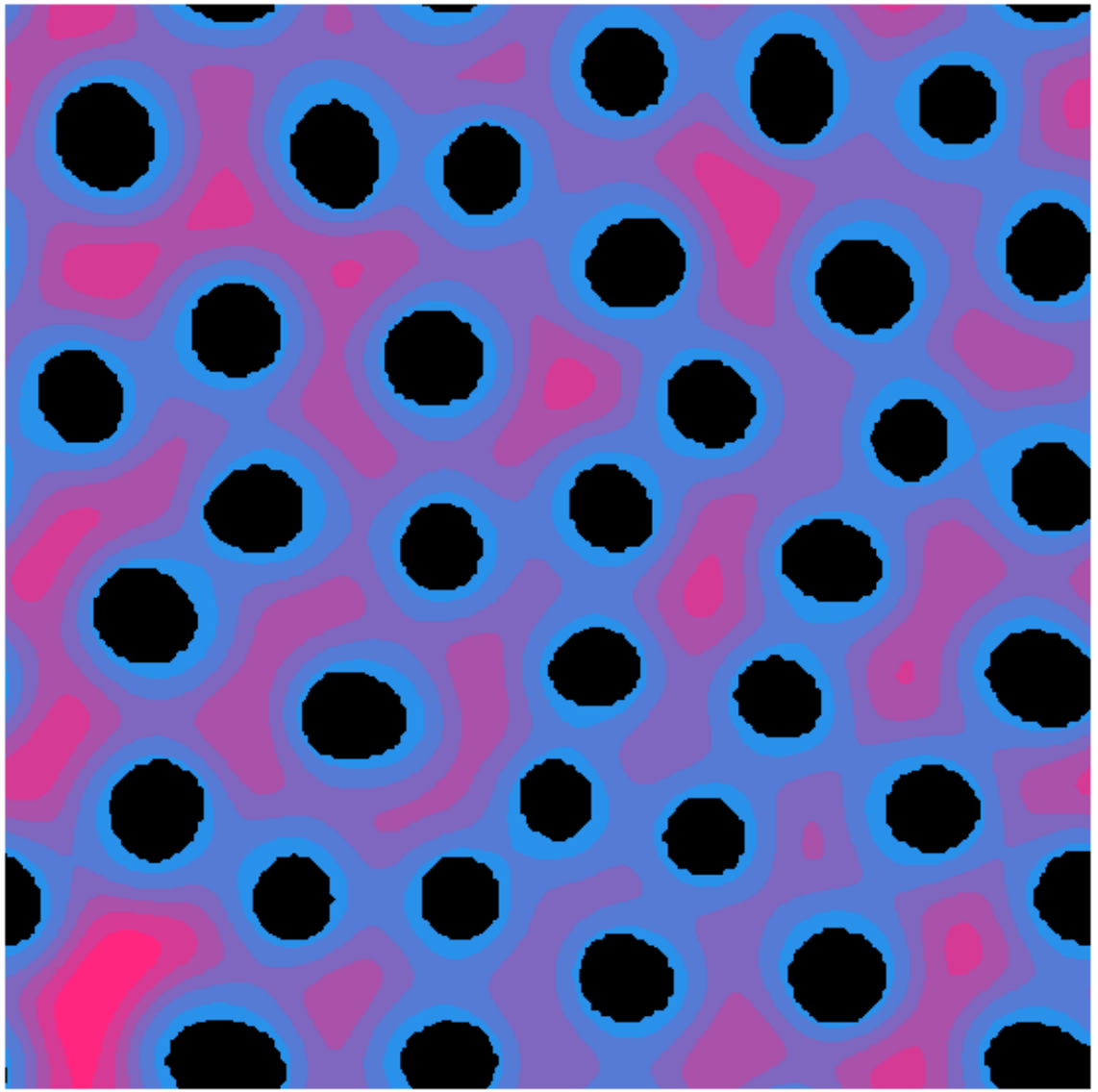


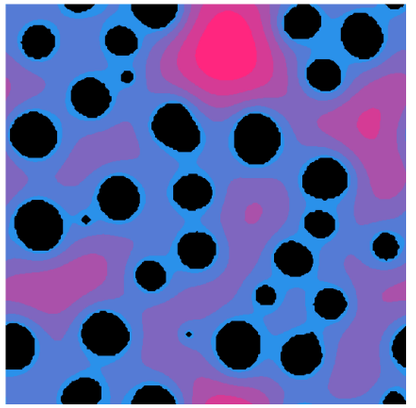
B. Lita et al., APL **74**, (1999)

Spacing and size of stacked dots becomes more regular

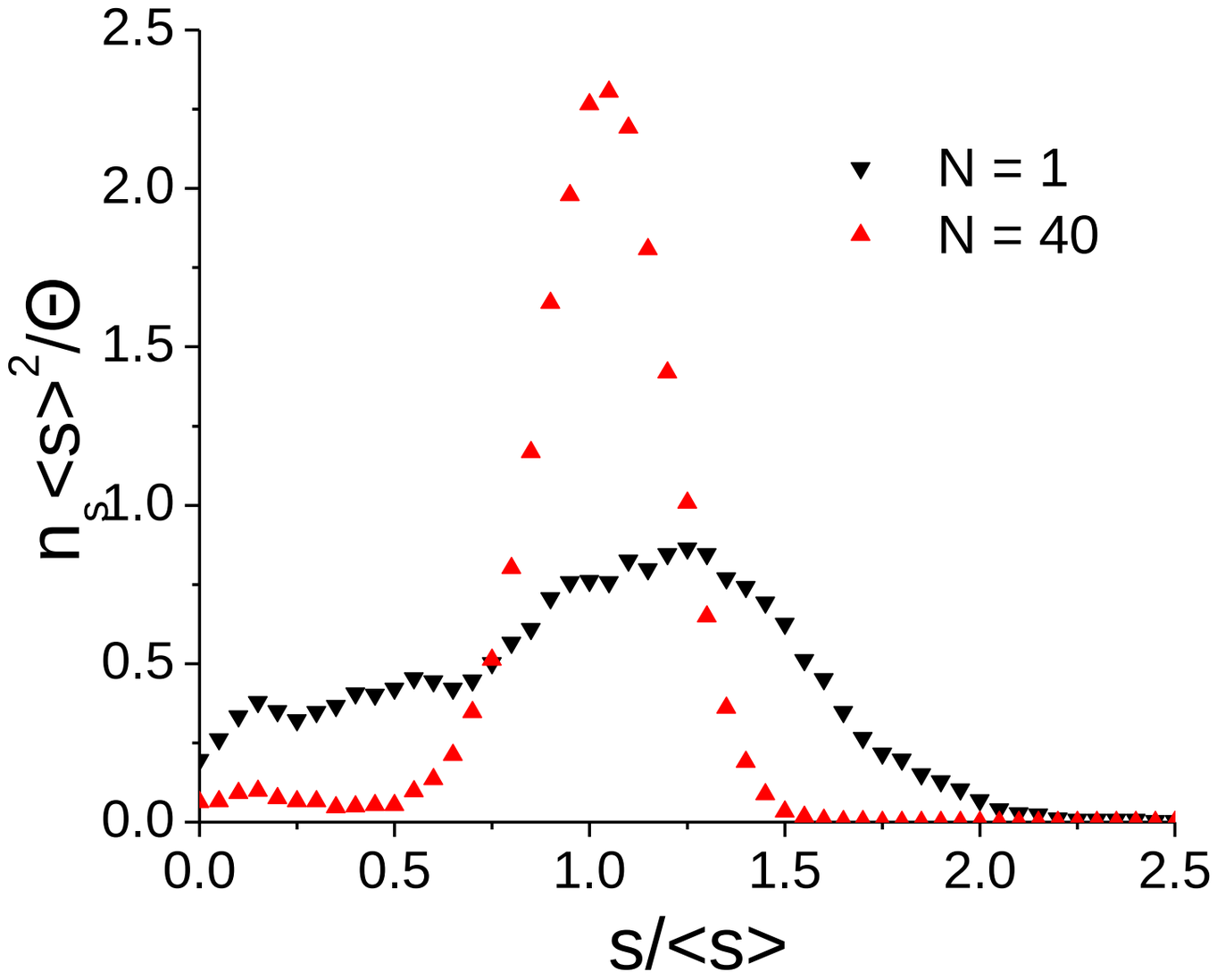
X. Niu, Y.-J. Lee, R.E. Caflisch, and C. Ratsch, Phys. Rev. Lett. 101, 086103 (2008).

Simulation of Growth of 20 Superlayers





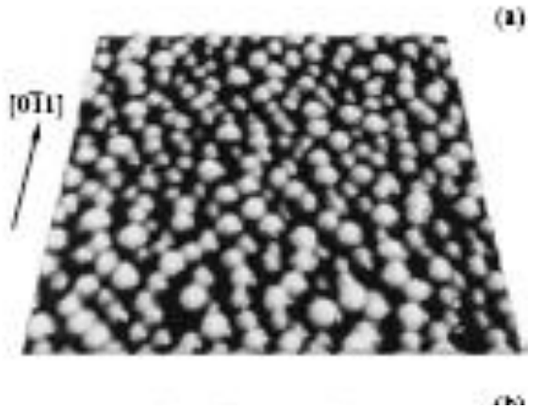
Regularization of Dot Size



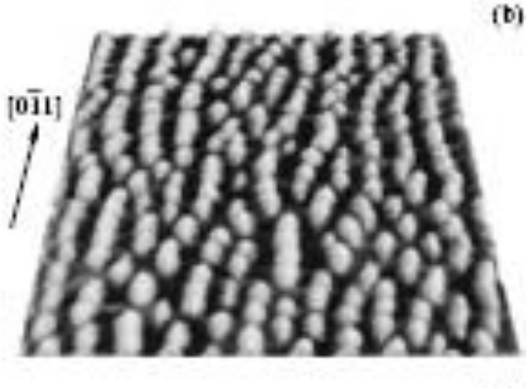
Sample Slide

Growth of stacked quantum dots of $In_{0.5}Ga_{0.5}As/GaAs(100)$

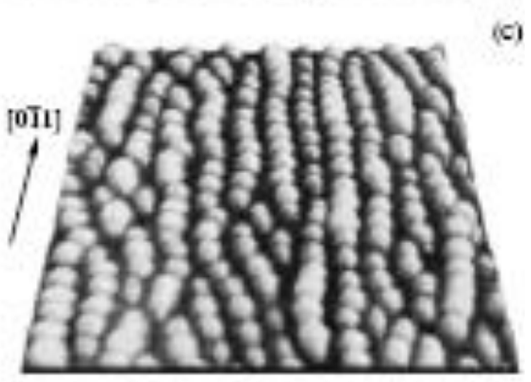
2 periods



7 periods



9 periods



V.V. Strel'chuk et al., Semiconductors 41 (2007).

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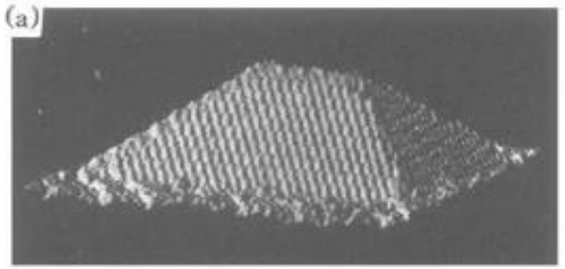
- Strain-induced ordering in the submonolayer growth regime
- Ordering of stacked quantum dots
- Inclusion of a step-edge barrier to model mound formation

Mound Formation due to an Additional Step-Edge barrier

Strain in heteroepitaxial systems leads to instabilities and the formation of quantum dots:

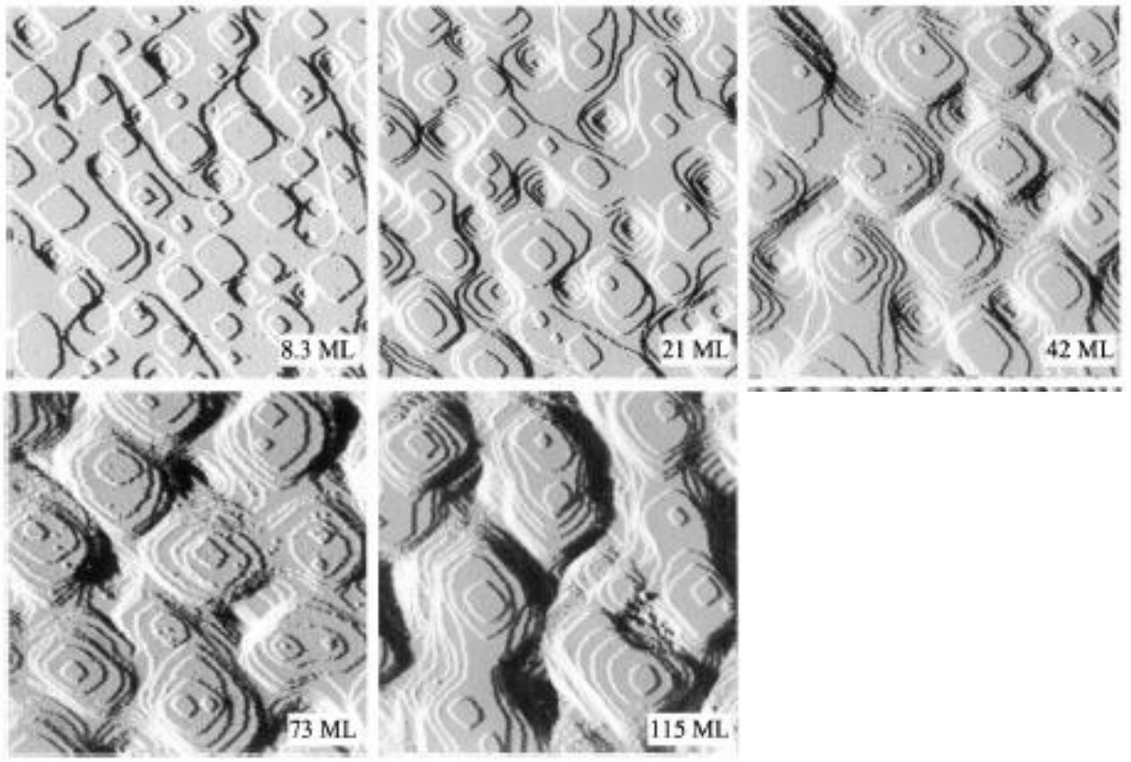
Self-Organized hut formation Ge/Si(001)



Y.-W. Mo et al., PRL **65** (1990)

But instabilities and mounds also form for homoepitaxial system (even though there is no strain as a driving force)

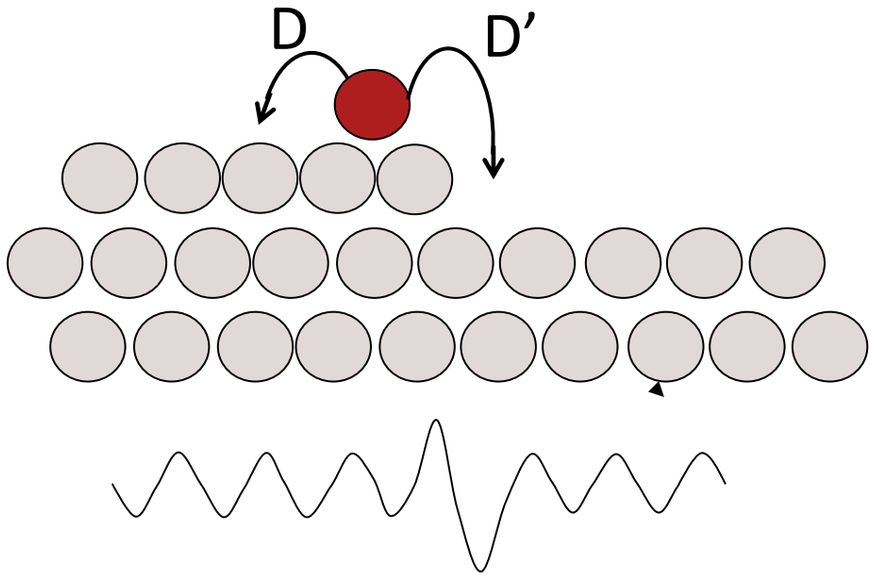
Example: Cu on Cu(100):



J.K. Zuo and J.F. Wendelken, PRL 78, 2791 (1997).

A Mixed Boundary Condition to Include a Step-Edge Barrier

Many systems have an additional barrier to diffuse down a step edge to a lower terrace:



This leads to a modified mixed (Robin) boundary condition:

$$\nabla\rho \cdot \mathbf{n} = \frac{D'}{D-D'}(\rho_{eq} - \rho)$$

$$-D\frac{\partial\rho}{\partial x}(x_n) = \quad D'\,\rho(x_{n-1}) \quad - \quad D'\rho_{eq}$$

Macroscopic flux on upper terrace = (microscopic) hop toward step - (microscopic) hop away from step

Approximate

$$\rho(x_{n-1}) = \rho(x_n) - \frac{\partial\rho}{\partial x}(x_n)$$

What are We Doing About ρ_{eq} ?

Extend Approach of Caflisch et al. (PRE, 1999): $\rho_{eq} = \frac{D_{det}}{D_T}\frac{16}{3}\left(\frac{16}{15}\frac{LF}{D_E}\right)^{2/3}$

To obtain

$$\rho_{eq} = \mathrm{C}\left[\frac{FL}{2}\frac{1}{c_{w1}D_{EK}+\mathrm{C}\left(c_{w2}D_{TK}{}^{+}+c_{w3}D_{TK}{}^{-}\right)}\right]^{2/3}\left[\frac{c_{h1}D_{EB}+\mathrm{C}\left(c_{h2}D_{TB}{}^{+}+c_{h3}D_{TB}{}^{-}\right)}{c_{g1}D_{EK}+\mathrm{C}\left(c_{g2}D_{TK}{}^{+}+c_{g3}D_{TK}{}^{-}\right)}\right]^{1/3}$$

with

$$\mathrm{C}=\frac{\frac{a}{L}\left(c_{f+}\frac{D_{ET}{}^{+}}{D_T}+c_{f-}\frac{D_{ET}{}^{-}}{D_T}\right)+c_{f+}c_{f-}\frac{A}{D_T{}^{2}}}{\frac{a}{L}\left(c_{f+}\frac{D_{TE}{}^{+}}{D_T}+c_{f-}\frac{D_{TE}{}^{-}}{D_T}\right)+c_{f+}c_{f-}\frac{D_{TE}{}^{+}D_{TE}{}^{-}}{D_T{}^{2}}}$$

J. Papac, D. Margetis, F. Gibou, and C. Ratsch, (almost) submitted.

Numerical Implementation of Robin Boundary Condition

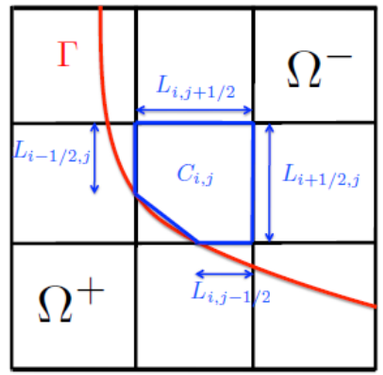
- The boundary condition is a PDE: $\nabla \rho \cdot \mathbf{n} = f - \alpha \rho$
- Ghost fluid approach is not easily applicable.
- Trick: Use the diffusion equation in integral form:

$$\int_{\mathcal{C}_{i,j} \cap \Omega^-} \left(\rho_{i,j}^{n+1} - \rho_{i,j}^n - \frac{k\Delta t}{2} \widetilde{\Delta} \rho^{n+1} - \frac{k\Delta t}{2} \widetilde{\Delta} \rho^n - \frac{\Delta t}{2} g_{i,j}^{n+1} - \frac{\Delta t}{2} g_{i,j}^n \right) dA = 0$$

Difficult terms

- Apply divergence theorem to cell in order to prescribe the boundary condition:

$$\int_{\mathcal{C} \cap \Omega^-} \nabla \cdot \nabla \rho \, dA = \int_{\partial(\mathcal{C} \cap \Omega^-)} \nabla \rho \cdot n \, dl.$$



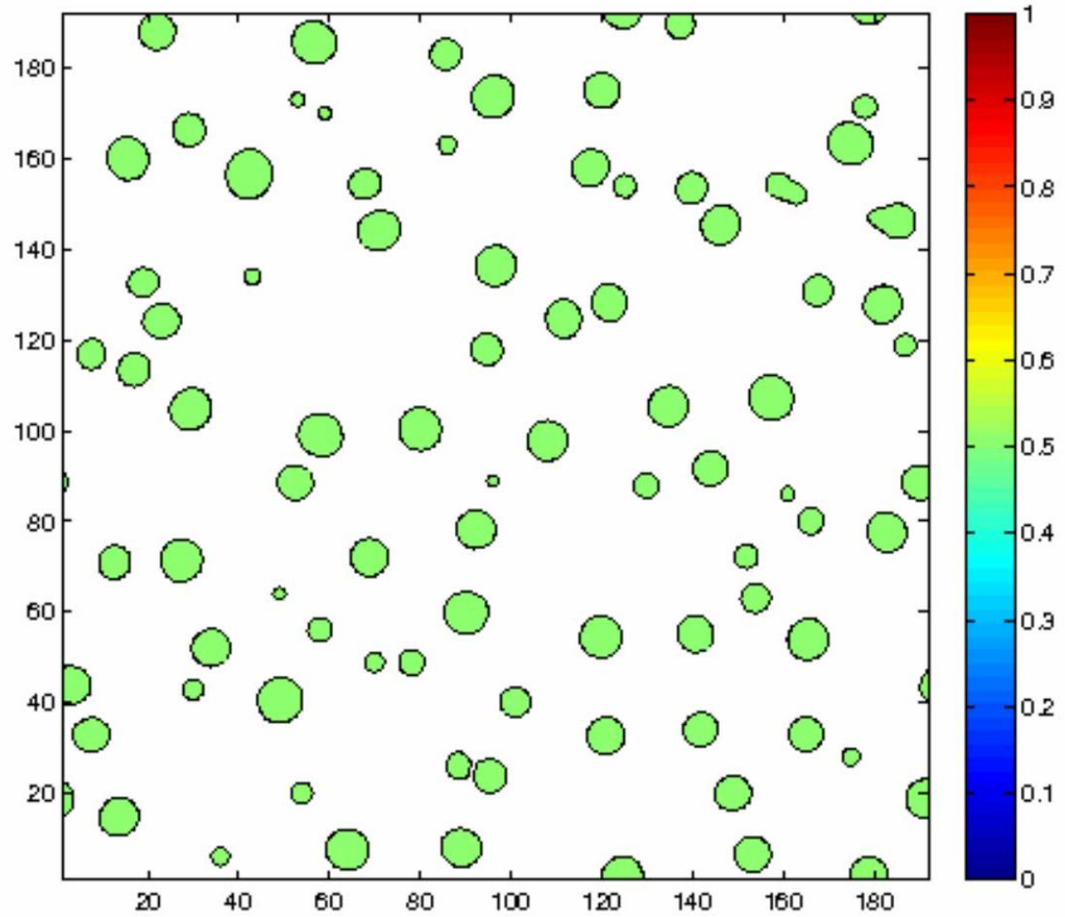
- Then use boundary condition: $\int_{\mathcal{C}_{i,j} \cap \Gamma} \nabla \rho \cdot \mathbf{n} \, dl = \int_{\mathcal{C}_{i,j} \cap \Gamma} (f - \alpha \rho) \, dl$

- This leads to a stable, second order accurate method.

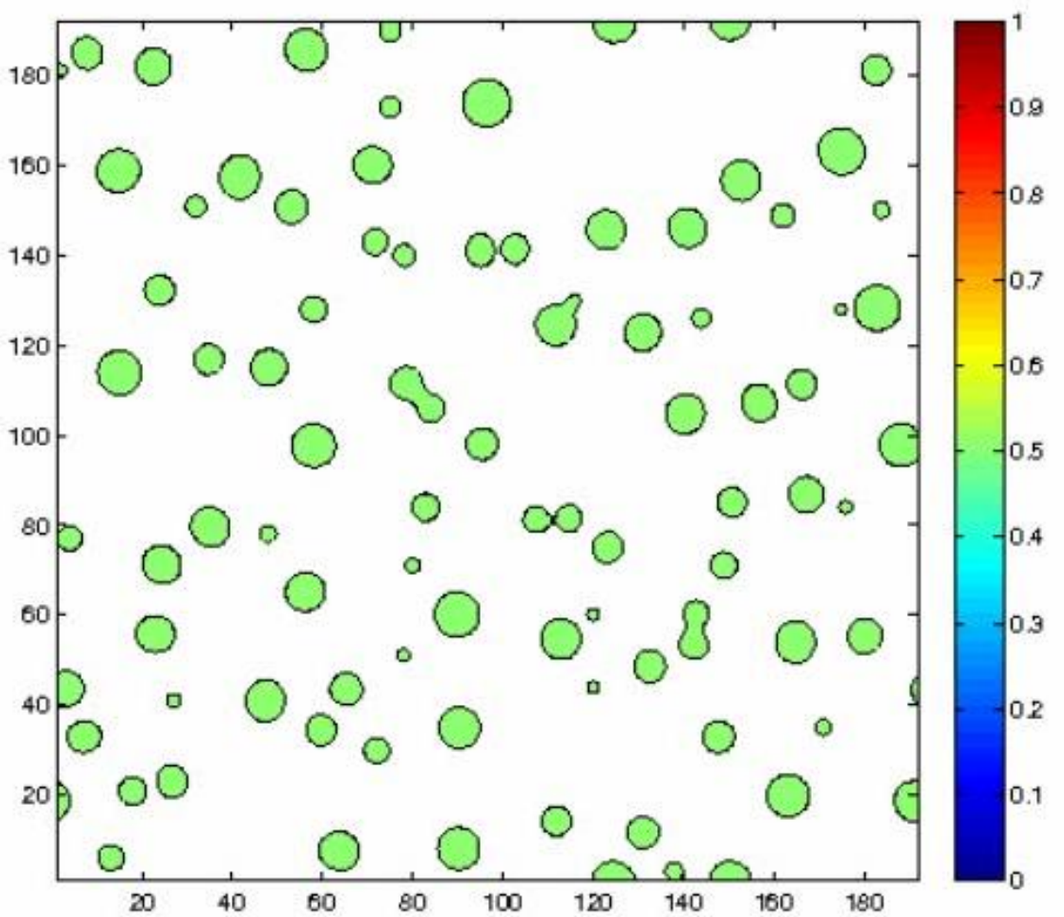
J. Papac, F. Gibou, and C. Ratsch, JCP, 229, 875 (2010).

Effect of a Step-Edge Barrier

No Step-edge barrier



Substantial Step-edge barrier



Conclusions

- We have developed a numerically stable and efficient island dynamics model that includes the effect of strain on all microscopic parameters that are relevant during epitaxial growth.
- We find that compressive strain leads to sharpening of the scaled island size distribution.
- We have modeled the formation and self-alignment of stacked quantum dots.
- We have included the effect of a step-edge barrier, via a Robin Boundary condition.

Thank You for Your Attention !

$$$** I would like to acknowledge support from the National Science Foundation **$$$

Happy Birthday, Russ!