Strain Dependence of Microscopic Parameters and its Effects on Ordering during Epitaxial Growth

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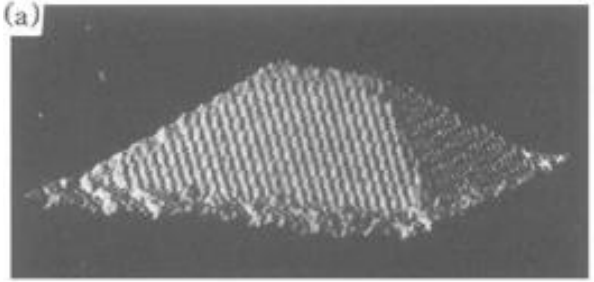
Collaborators: Xiaobin Niu, Peter Smereka, Jason DeVita.

Motivation: strain induces ordering.

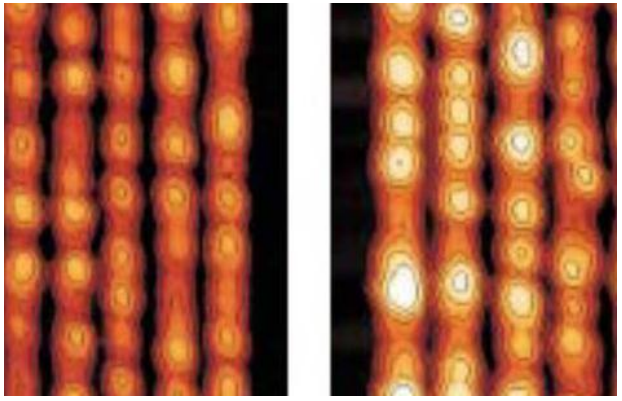
Self-Organized hut formation Ge/Si(001)



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

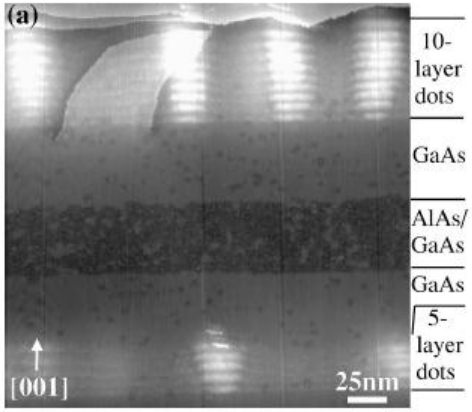
Horizontal ordering of quantum dots:

InAs on AlGaAs



E. Uccelli et al., Nanotech. **19** (20008)

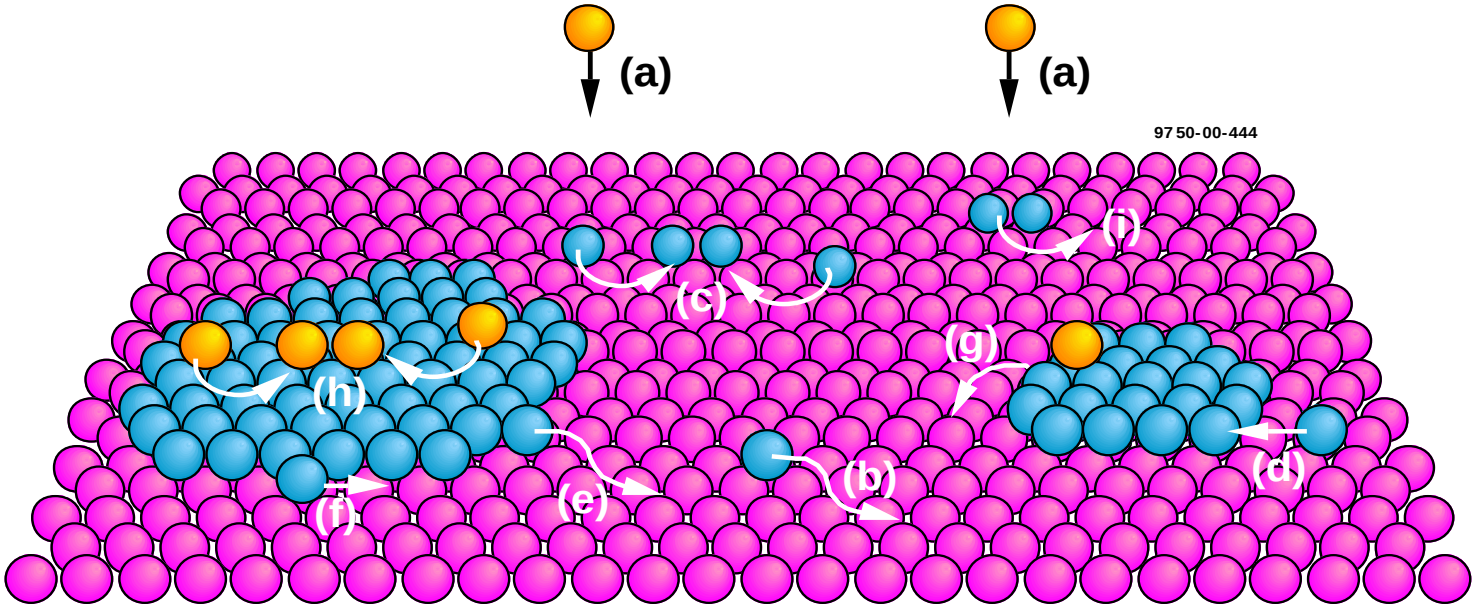
Vertical ordering of quantum dots: $Al_xGa_{1-x}As$ system



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

Goal: Develop a kinetic model that correctly includes strain!

How do we include strain in a growth model?



- Build a model that includes the above processes.
  - Straightforward in an atomistic model (KMC)
  - Tricky but possible in continuum type models, or island dynamics (level sets)
- Every process can have a different rate at different location (because of local strain)
- Our approach
  - Calculate the strain field at every time step of the simulation
  - Adjust rate for every process (at every location) based on local strain
  - Rates are based on density-functional theory calculations (pre-computed)

Outline

- Use density-functional theory (DFT) to calculate strain dependence of microscopic parameters.
- The level-set formalism to model epitaxial growth.
- Ordering in the submonolayer growth regime.
- Ordering of stacked quantum dots.

Computational Details of DFT Calculations

- We use the FHI-AIMS package.

  V. Blum et al., Comp. Phys. Comm. 180, 2175 (2009).

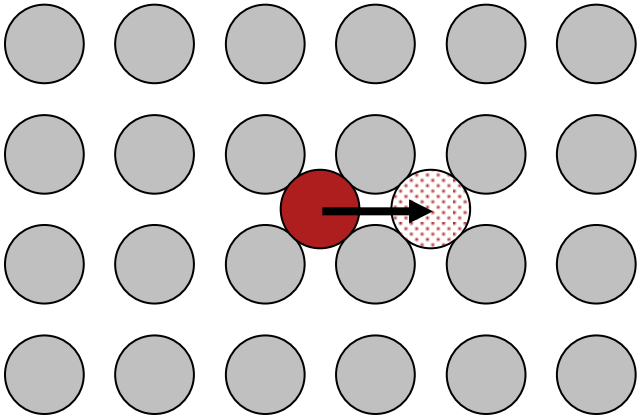
- This code uses numeric atom-centered basis sets.

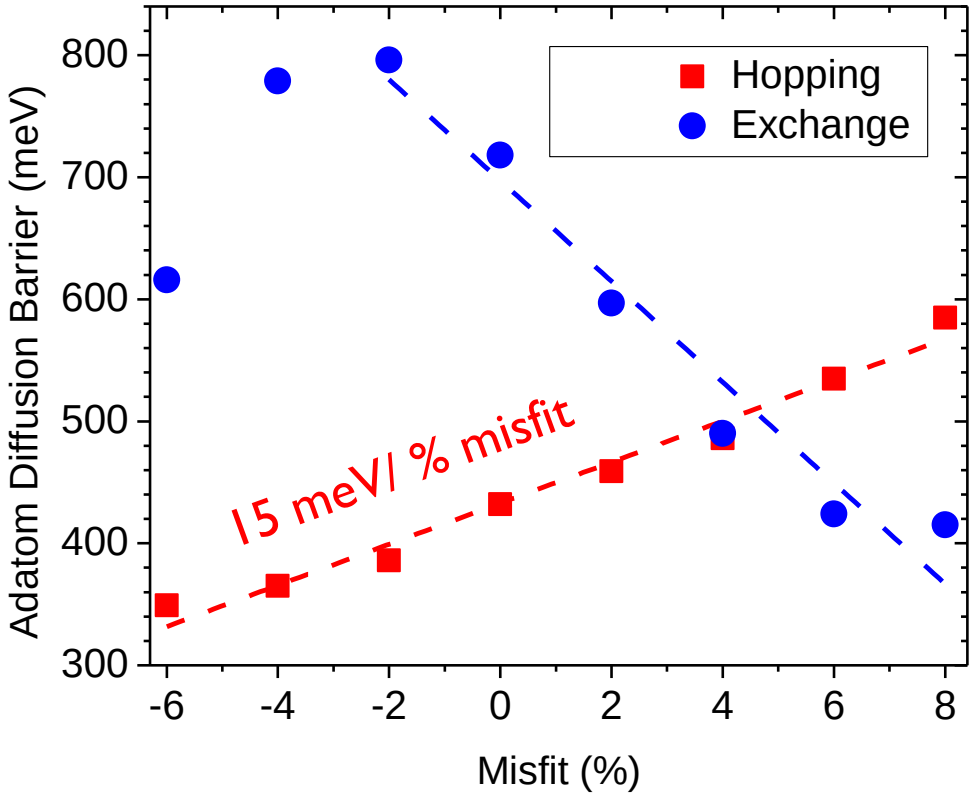
- GGA is used for exchange correlation (PBE parameterization).

- Periodic supercells with 6 layers that are fully relaxed.

- Model system: Ag/Ag(100)

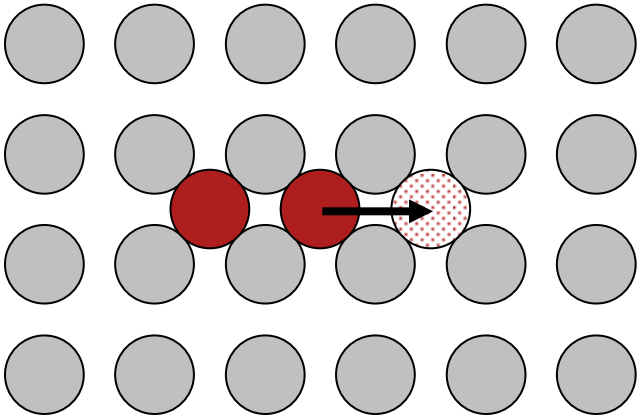
Strain Dependence for Adatom Diffusion for Ag on Ag(100)

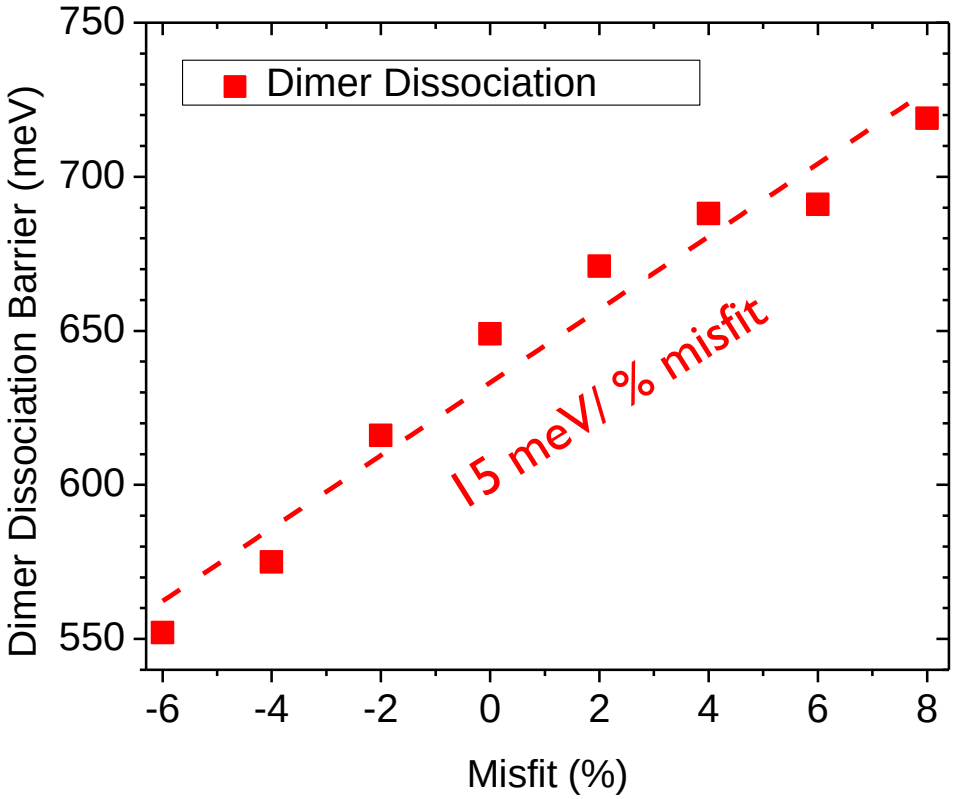




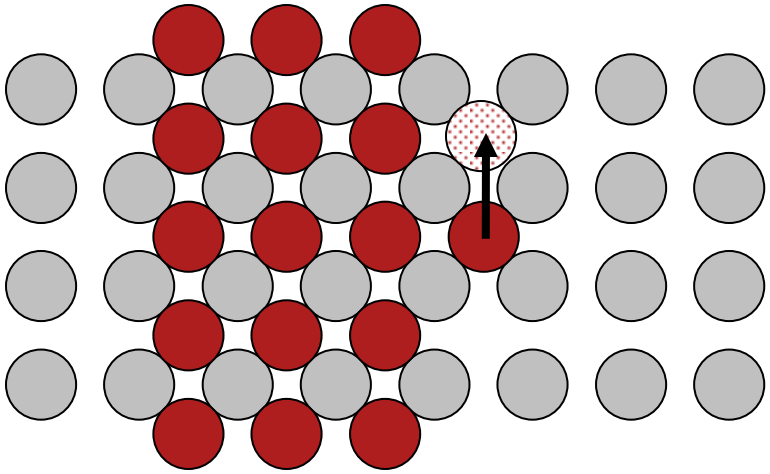
In agreement with Yu and Scheffler, PRB 56, R15569 (1997)

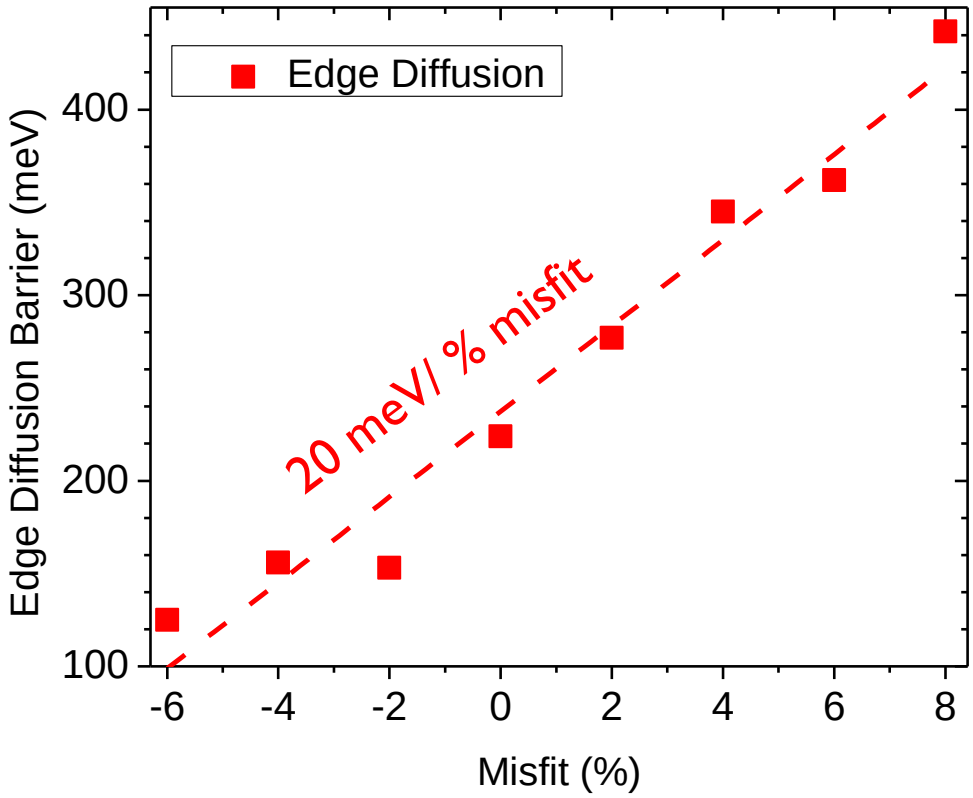
Strain Dependence for Dimer Dissociation for Ag on Ag(100)



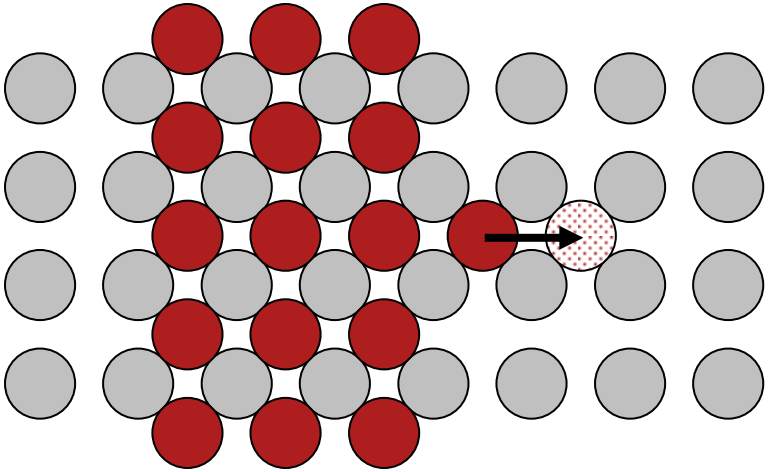


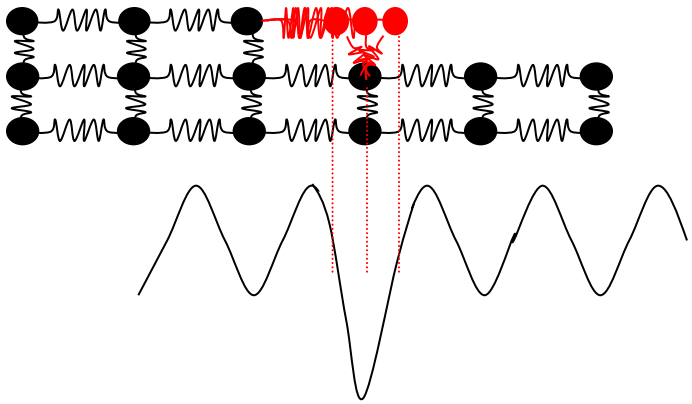
Strain Dependence for Edge Diffusion for Ag/Ag(100)

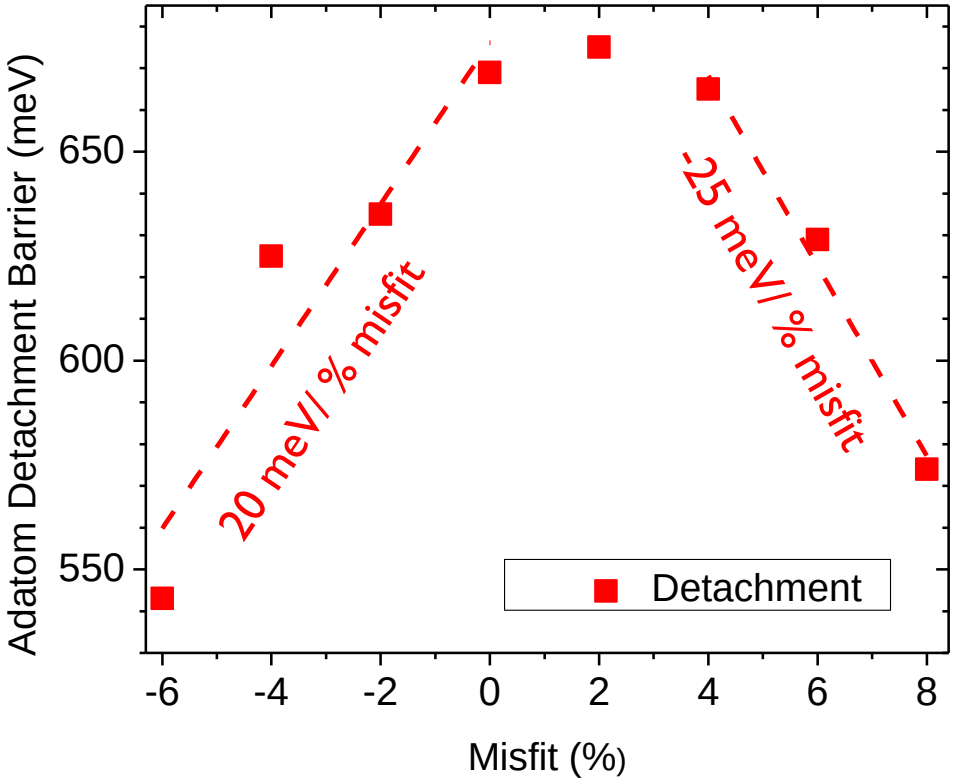




Strain Dependence for Adatom Detachment for Ag/Ag(100)







Transition site matters!

Outline

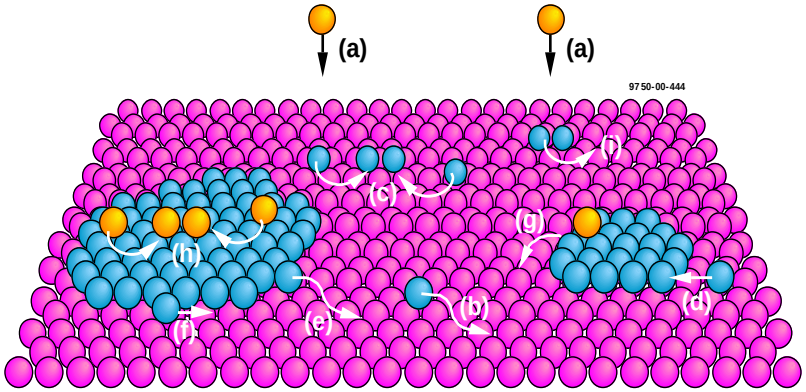
- Use density-functional theory (DFT) to calculate strain dependence of microscopic parameters.
- The level-set formalism to model epitaxial growth.
- Ordering in the submonolayer growth regime.
- Ordering of stacked quantum dots.

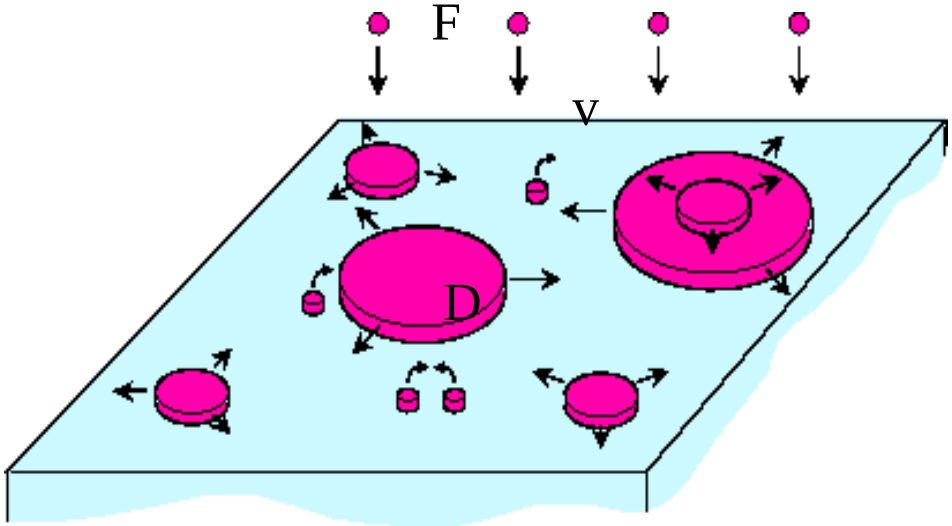
How Do We Incorporate These Strain Dependent Parameters into a Growth Model?

Atomistic picture
(i.e., kinetic Monte Carlo)
Time step: $O(10^{-6}s)$

replace →

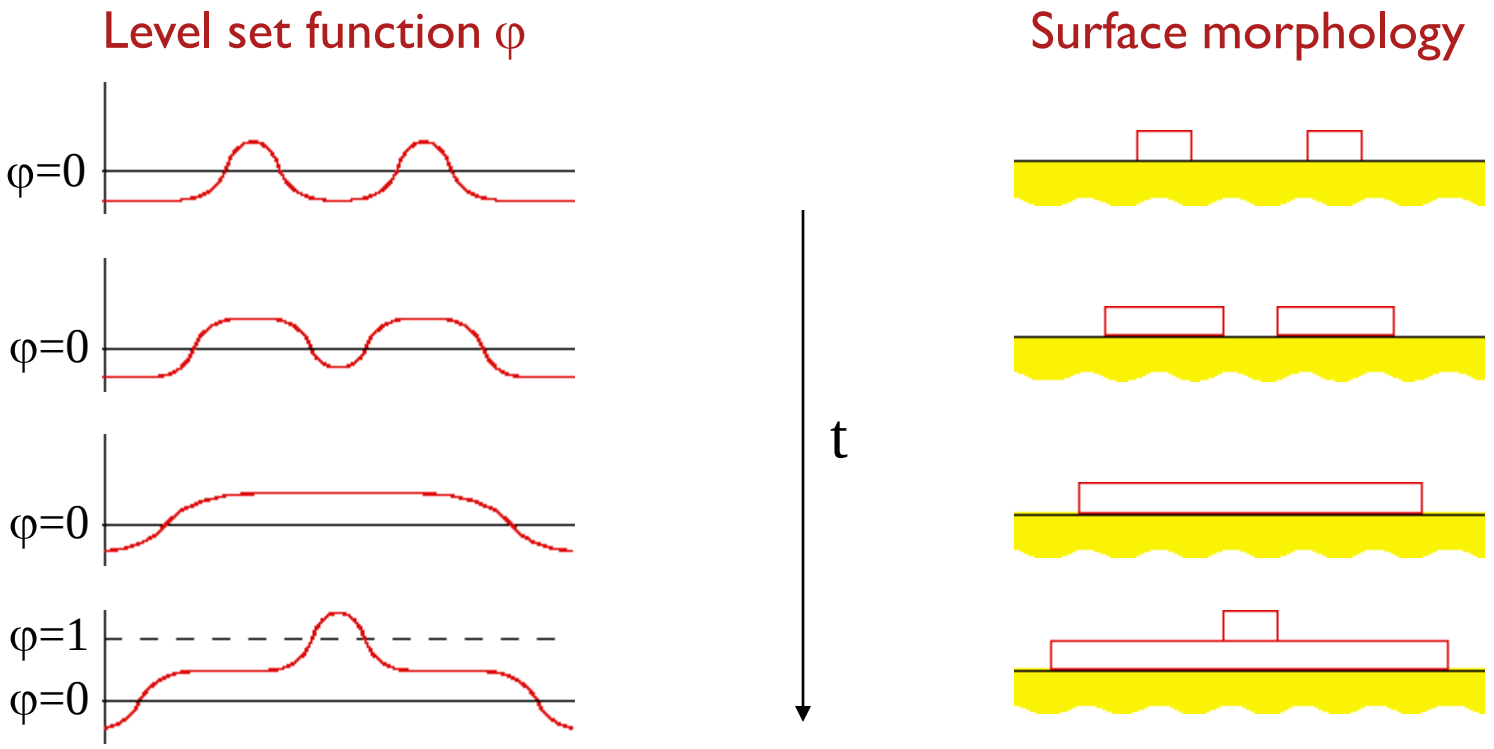
Island dynamics
Use level set method to track island boundaries
Time step: $O(10^{-2} – 10^{-3}s)$





- Assume that material that is deposited has a bigger lattice constant.
- Solve elastic equations at every time step (expensive!).
- This gives us strain (i.e., effective lattice constant) at every lattice site at every time step.
- Parameterize strain dependence of relevant parameters in growth model with DFT results.

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

The Level Set Method (Red Terms are Strain Dependent)

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

ρ : Adatom concentration

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

Diffusion in x-direction

Diffusion in y-direction

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

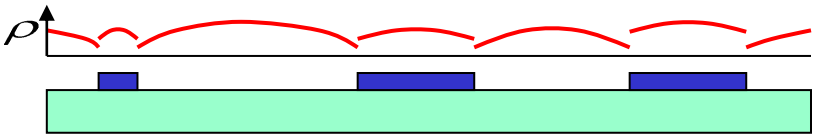
- Diffusion equation: $\frac{\partial\rho}{\partial t} = F + \nabla\cdot\mathbf{D}(\nabla\rho) - 2\frac{dN}{dt} + \text{drift}$

drift ~ $D_{xx}\nabla_x E_{ad} + D_{yy}\nabla_y E_{ad}$

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

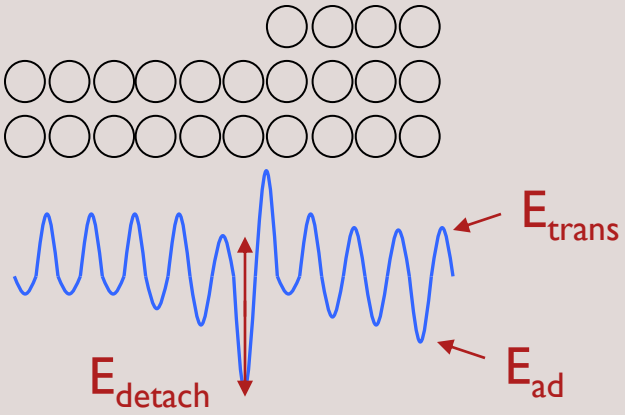
- Boundary condition: $\rho = \rho_{eq}$ $\mathbf{D}_{det}$ $,\mathbf{x})$

detachment rate

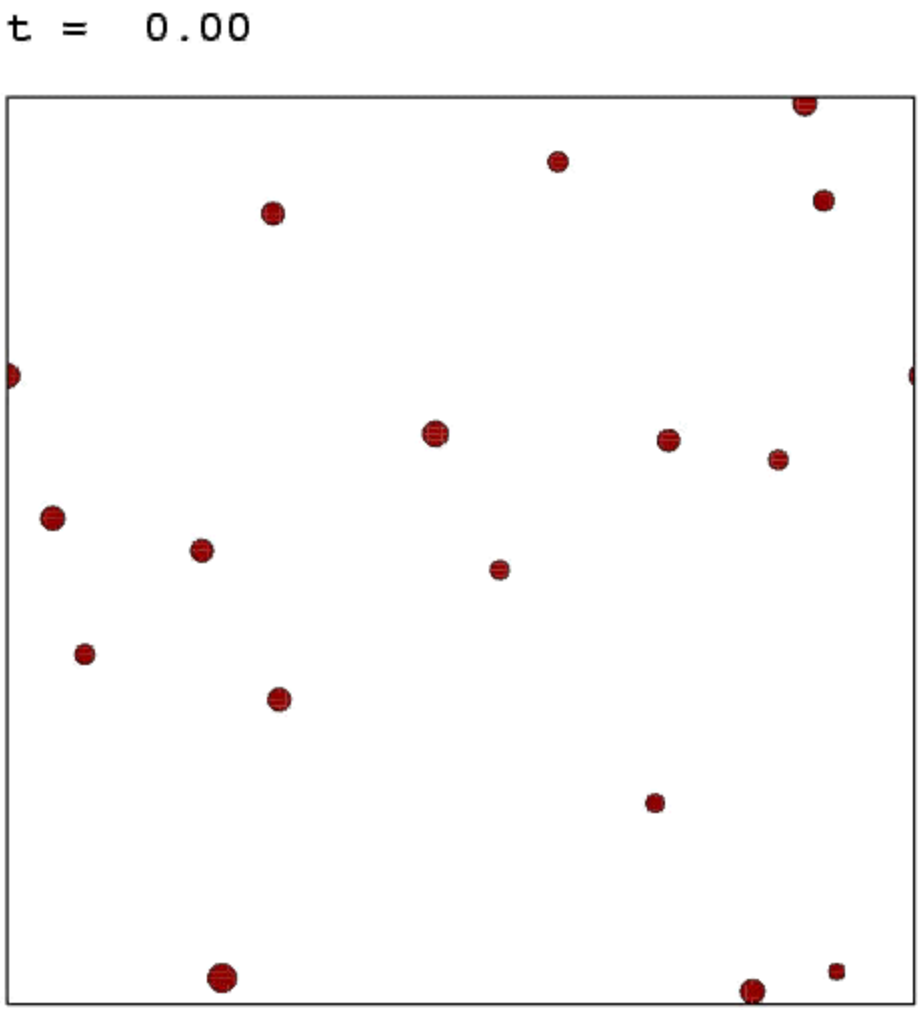


- Stochastic element needed for nucleation
- Stochastic rate for dissociation of islands

A typical potential energy surface

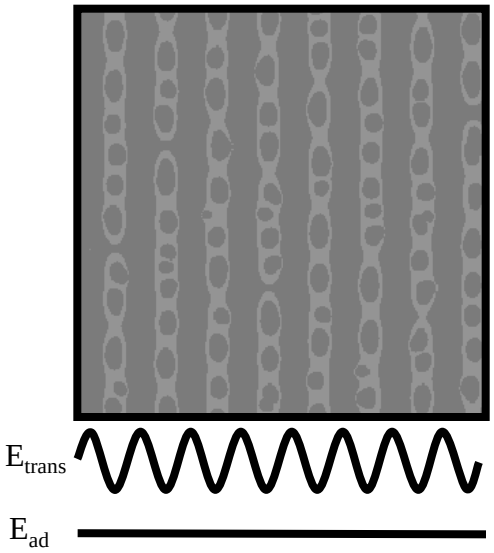


A typical level set simulation



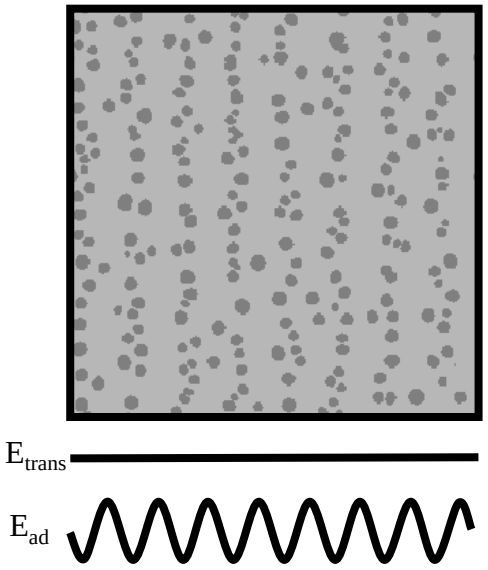
Variation of adsorption and transition energy (no strain yet)

Kinetic limit



Nucleation in region of fast diffusion

Thermodynamic limit



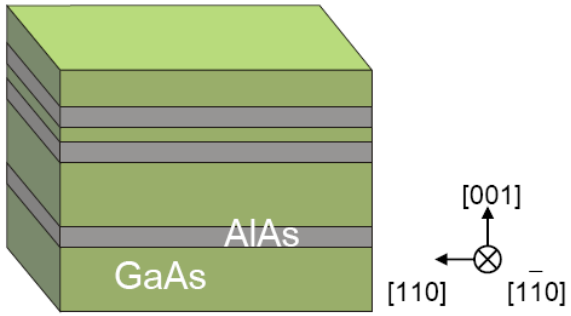
Nucleation in region of slow diffusion (but high adatom concentration), dominated by drift

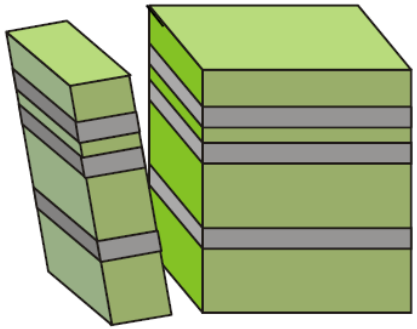
Nucleation rate ~ $D\rho(\mathbf{x},t)^2$

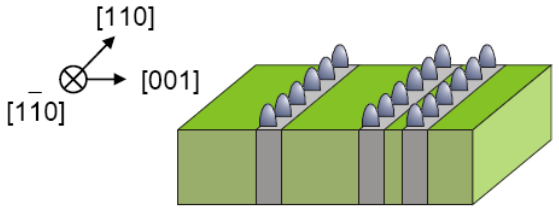
Ordering by Cleaved Edge Overgrowth

Work of E. Uccelli, G. Abstreiter, et al.

Grow AlAs/GaAs superlattice → Cleave and rotate → Grow InAs quantum dots





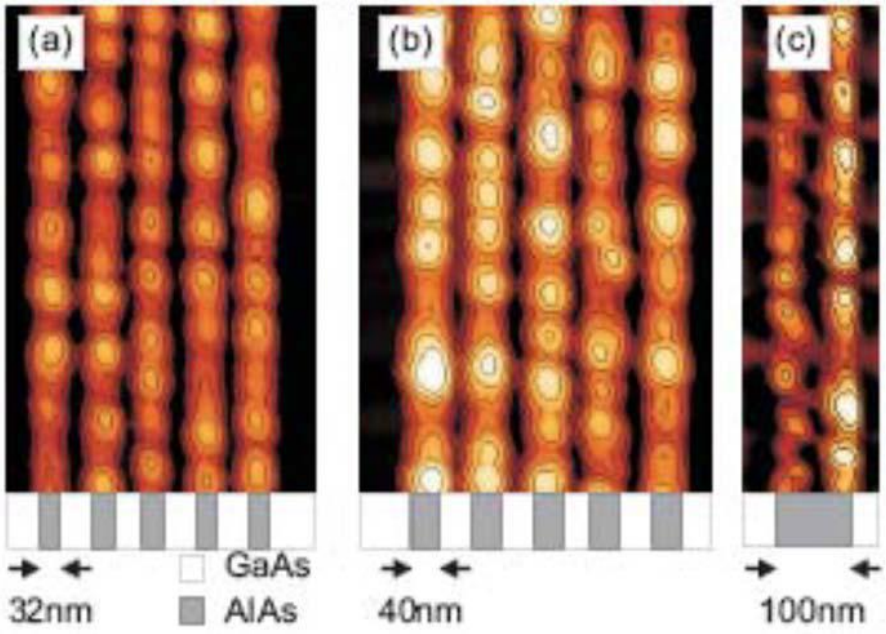


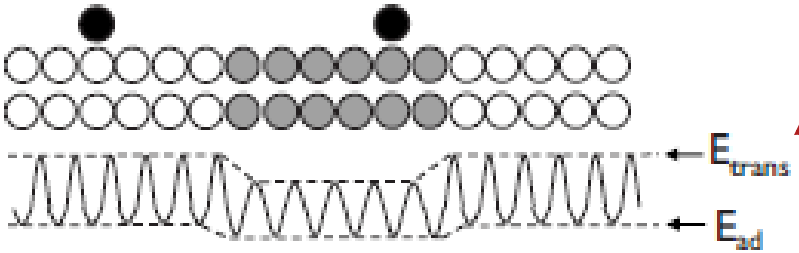
Quantum dots grow on top of the AlAs stripes

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

Ordering by Cleaved Edge Overgrowth

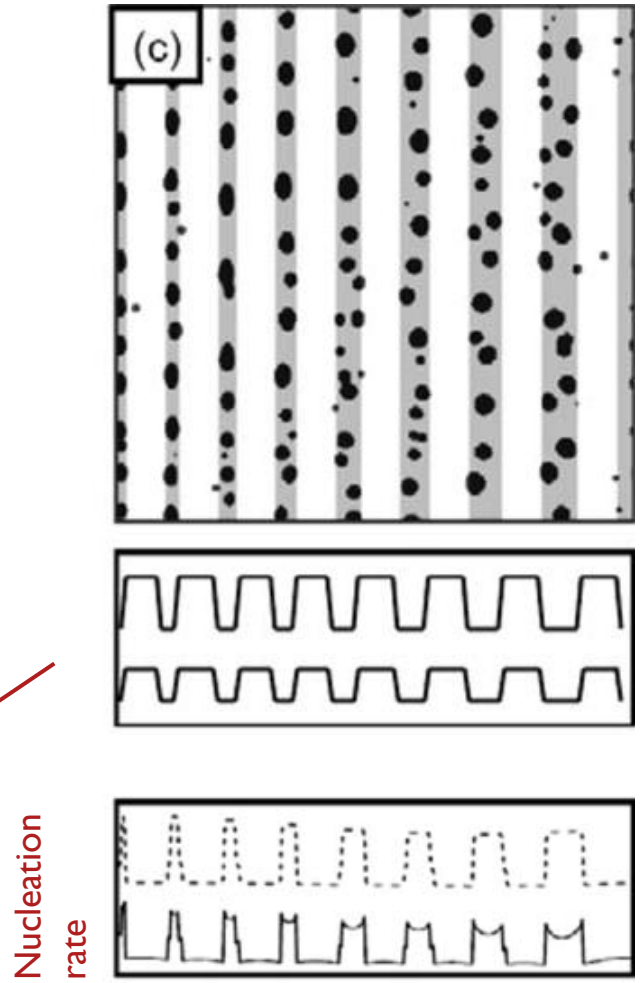
Experimental Result





PES

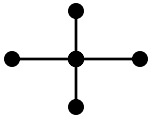
Simulations



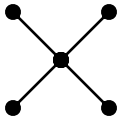
X. B. Niu, E. Uccelli, A. Fontcuberta i Morral, and C. Ratsch, APL 95 (2009)

Include Strain: Calculate Elastic Field at Every Timestep

- Our Model: Write down an atomistic energy density, 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) continuum energy density

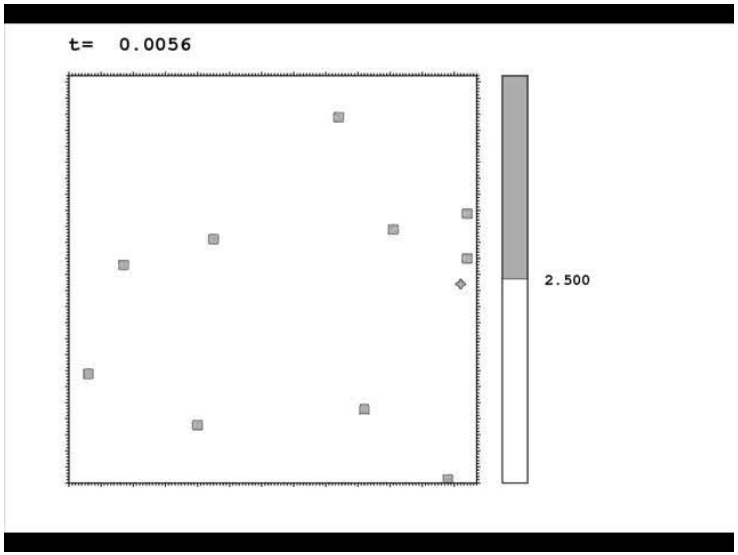
$$E = \alpha(S_{xx}^2 + S_{yy}^2) + \beta S_{xy}^2 + \gamma S_{xx} S_{yy}$$

- Minimize energy with respect to all displacements: ∂_u E [u] = 0

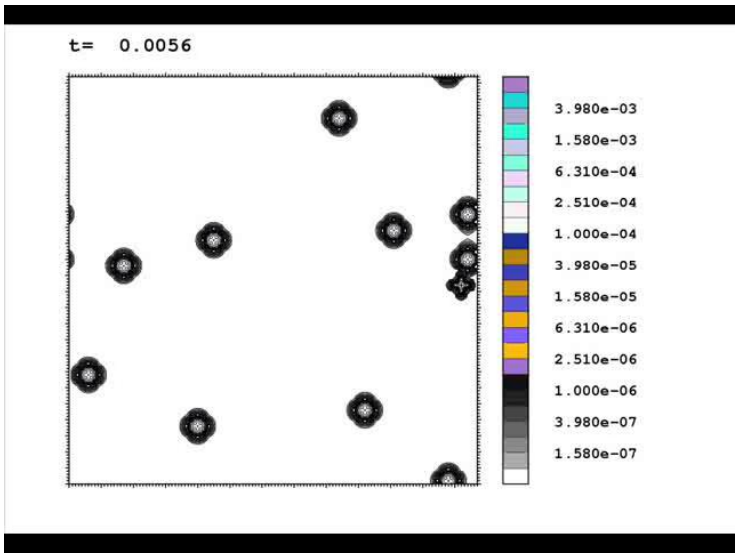
- The relevant microscopic parameters at every grid point are then varied as a function of the local strain, according to the strain dependence calculated by DFT

Effect of Strain in the Simulation

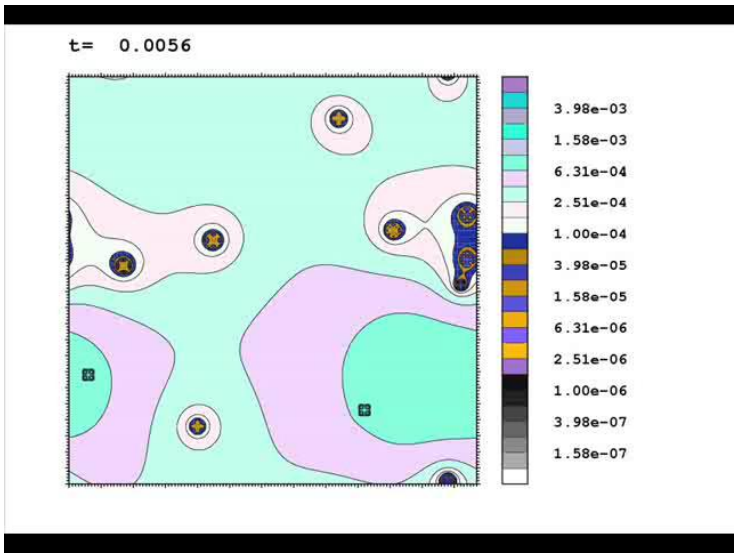
Morphologies



Elastic energies

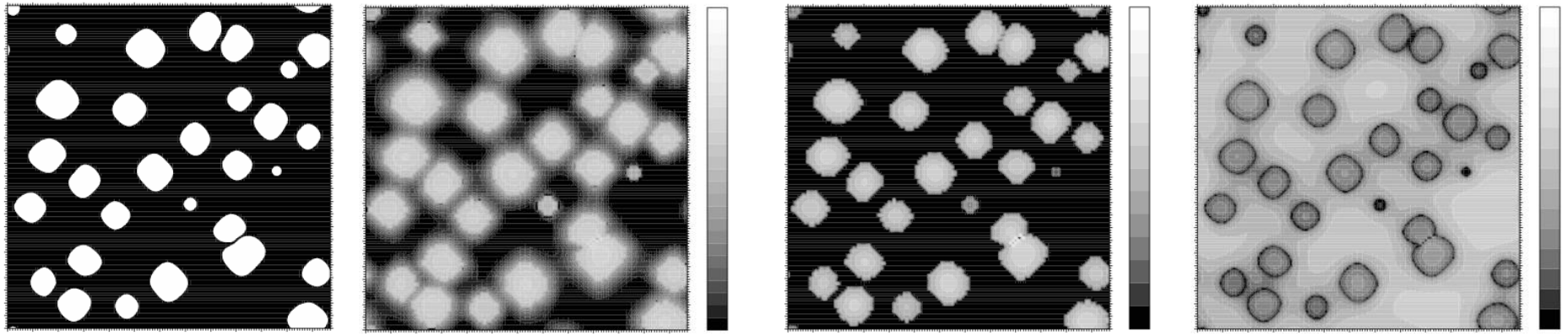


Adatom concentration



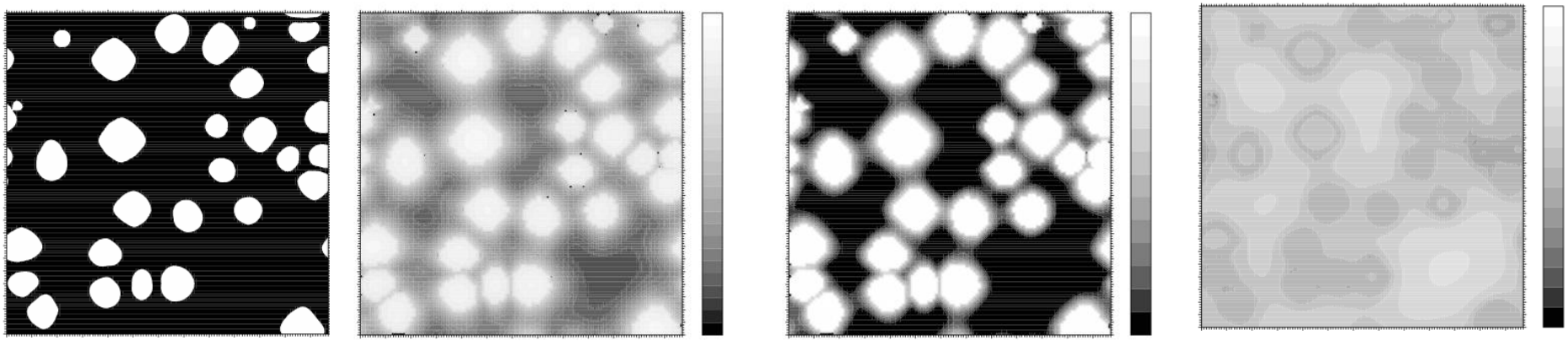
Effect of Strain in the Simulation

Morphologies | Elastic energies | Detachment rate | Adatom concentration



Top row: Compressive strain=1%

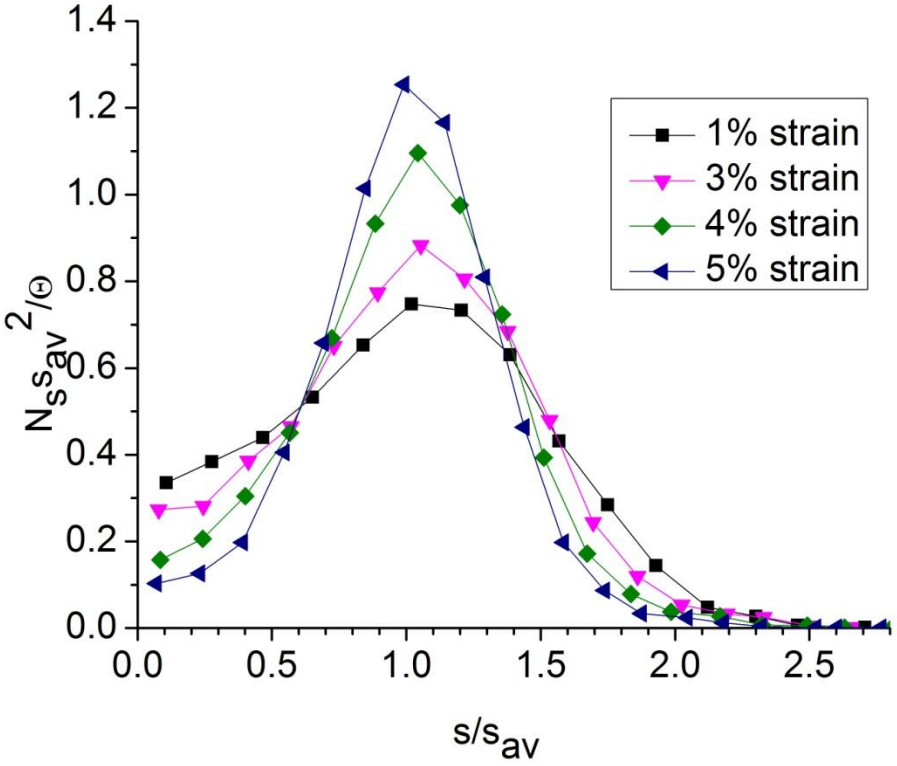
Bottom row: Compressive strain=5%



•With increasing compressive strain, islands become more regular, because i) small islands are more likely to break up, and ii) growth of large islands slows down.

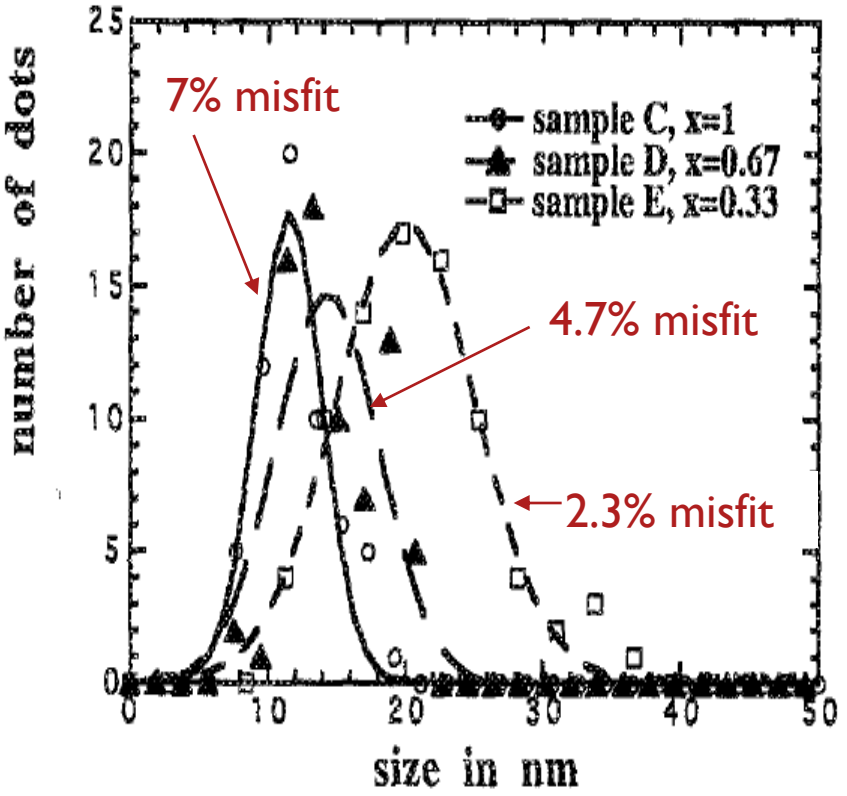
Sharpening of the Scaled Island Size Distribution during Submonolayer Epitaxy upon Compressive Strain

Level-set Simulation



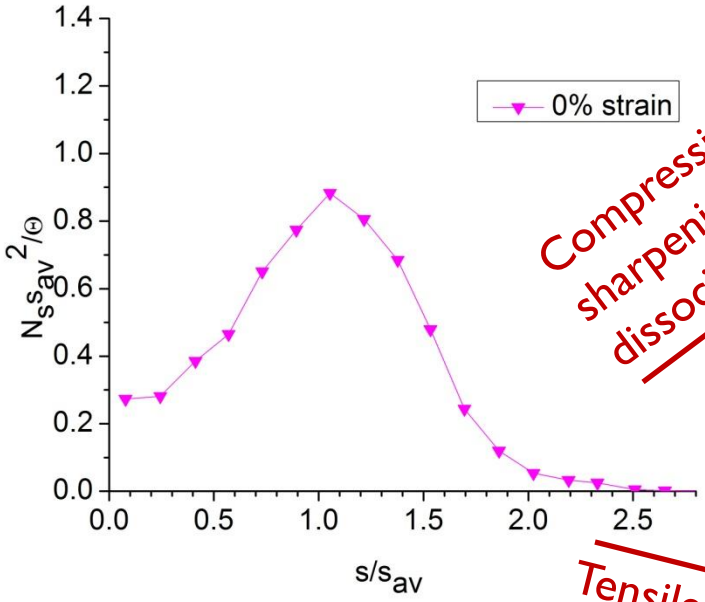
C. Ratsch, J. DeVita, and P. Smereka,
Phys. Rev. B **80**, 155309 (2009).

Experiment: $In_xGa_{1-x}As/GaAs(100)$

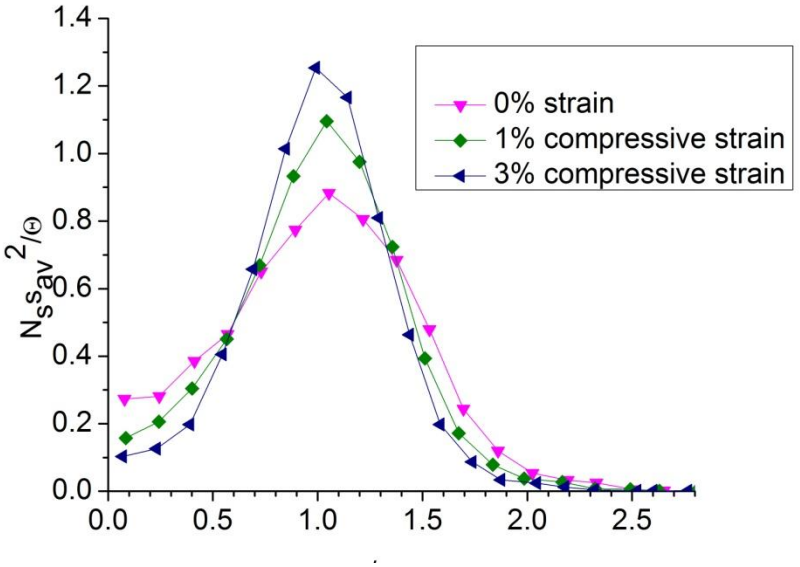


D. Leonard, M. Krishnamurthy, S. Fafard, J.L. Merz, and P.M. Petroff, J. Vac. Sci. Tech B 12, 1063 (1994)

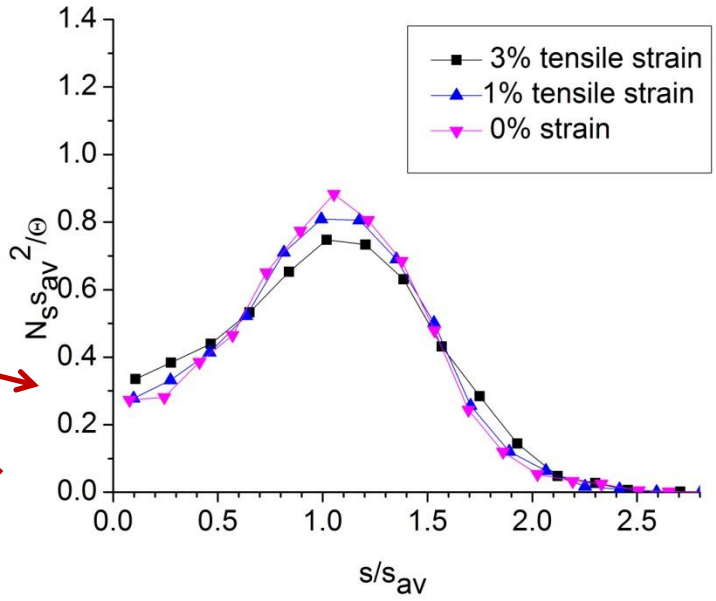
Scaled Island Size Distribution: Compressive versus Tensile Strain



Compressive strain leads to sharpening of ISD (dimer dissociation is enhanced)



Tensile strain leads to broadening of ISD (dimer dissociation is supressed)

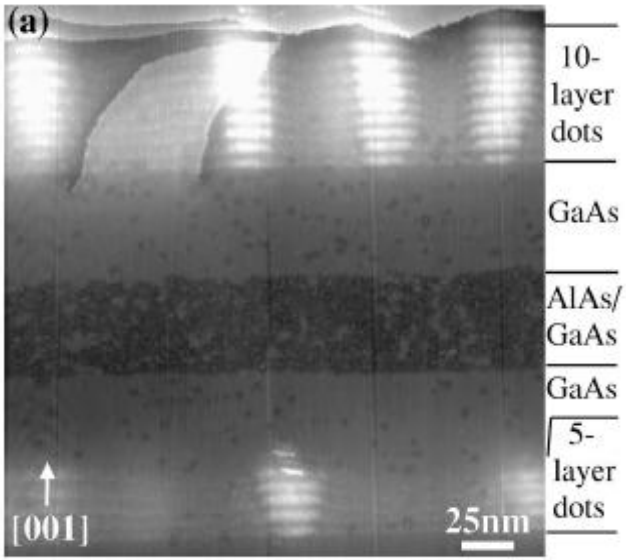


Outline

- Use density-functional theory (DFT) to calculate strain dependence of microscopic parameters.
- The level-set formalism to model epitaxial growth.
- Ordering in the submonolayer growth regime.
- Ordering of stacked quantum dots.

Simulation of Stacked Quantum Dots

$Al_xGa_{1-x}As$ system

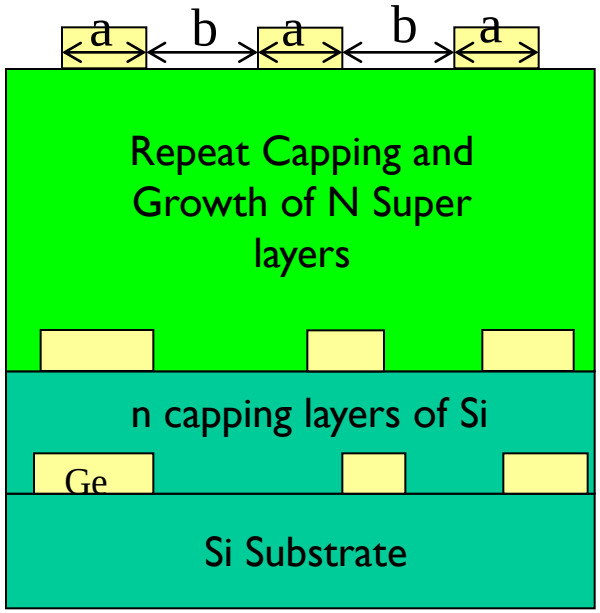


B. Lita et al., APL **74**, (1999)

Experimental observation: Stacked quantum dots align under certain conditions

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

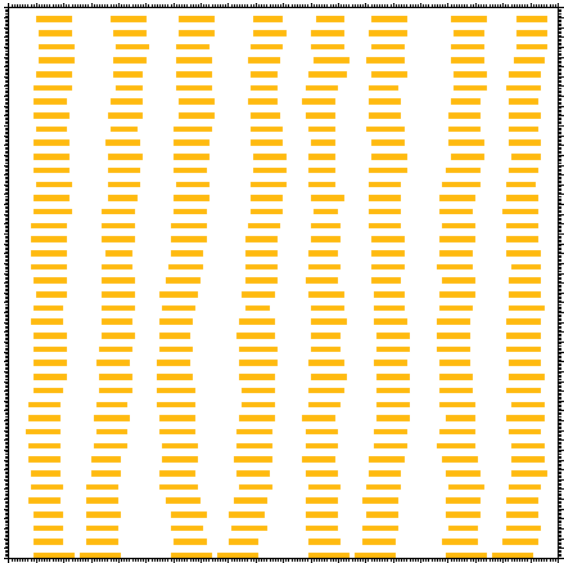
Simulation of Stacked Quantum Dots



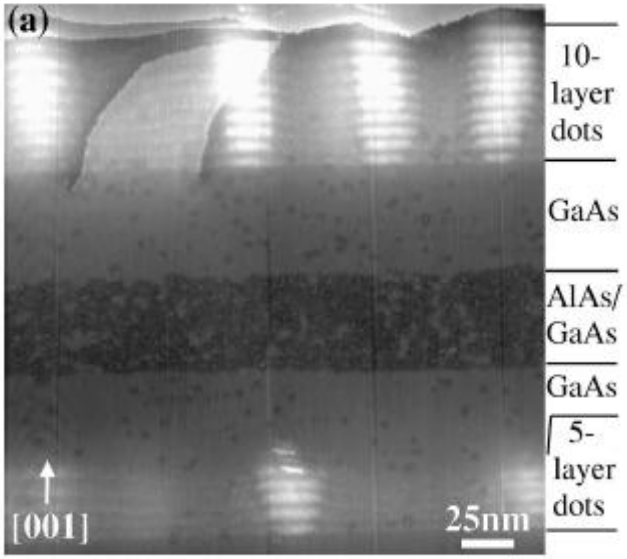
- Growth of islands on substrate without strain (constant diffusion and detachment)
- Fill in capping layer "by hand"
- Calculate strain on top of smooth capping layer
- Modify microscopic parameters for diffusion and detachment) according to strain
- Run growth model

Repeat procedure

Ordering of Stacked Quantum Dots



$Al_xGa_{1-x}As$ system

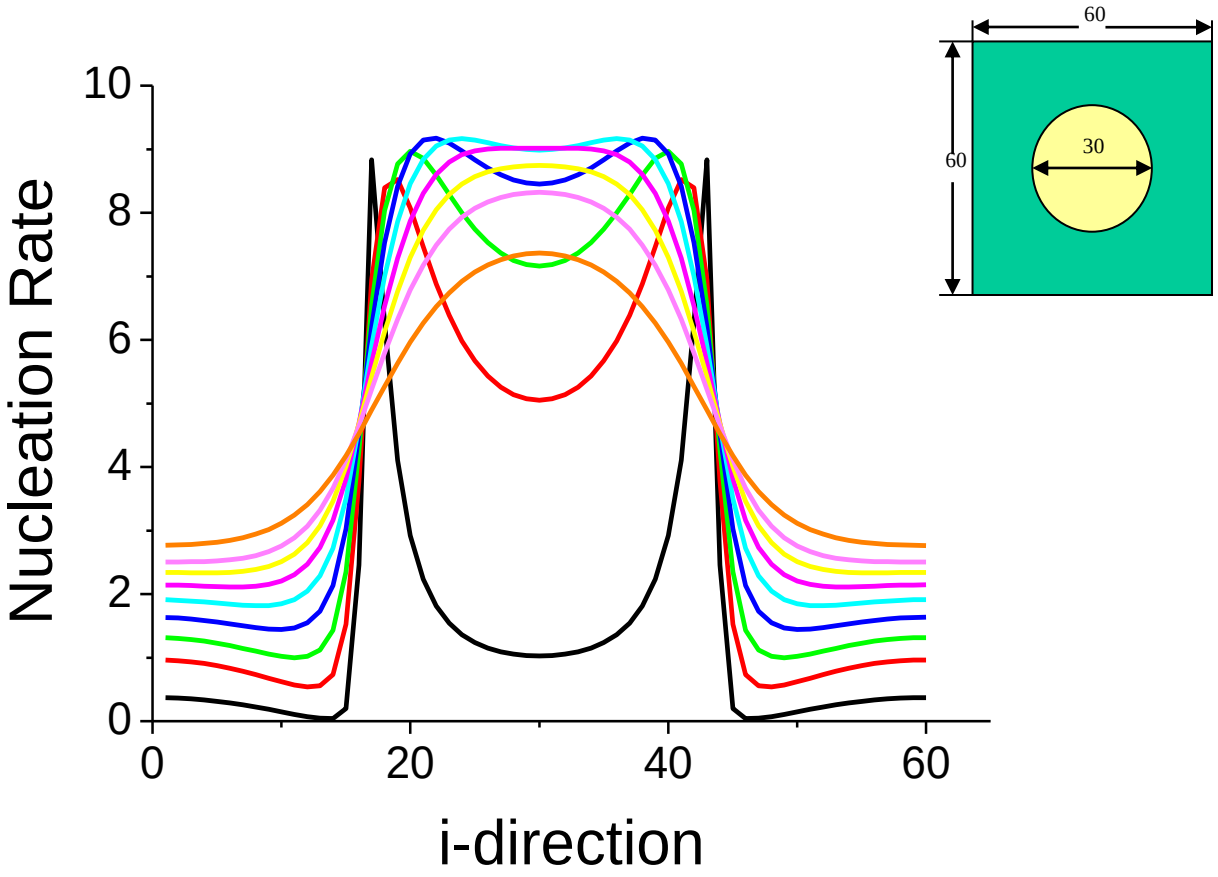


B. Lita et al., APL **74**, (1999)

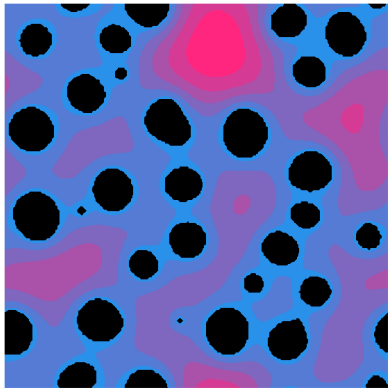
- Spacing and size of stacked dots becomes more regular

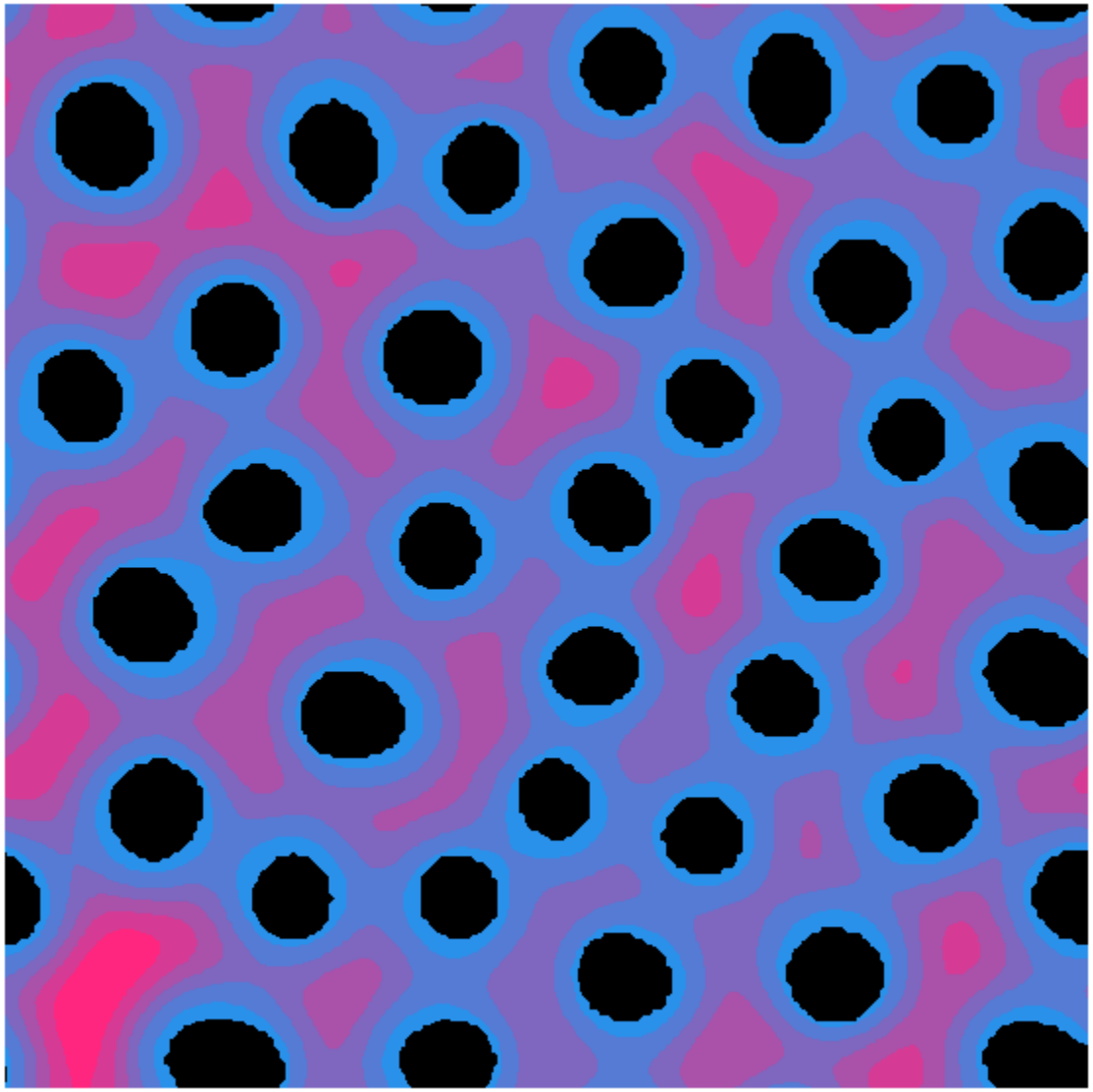
X. Niu, Y.-J. Lee, R.E. Caflisch, and C. Ratsch, Phys. Rev. Lett. 101, 086103 (2008).

Nucleation Rate as a Function of Capping Layer Thickness

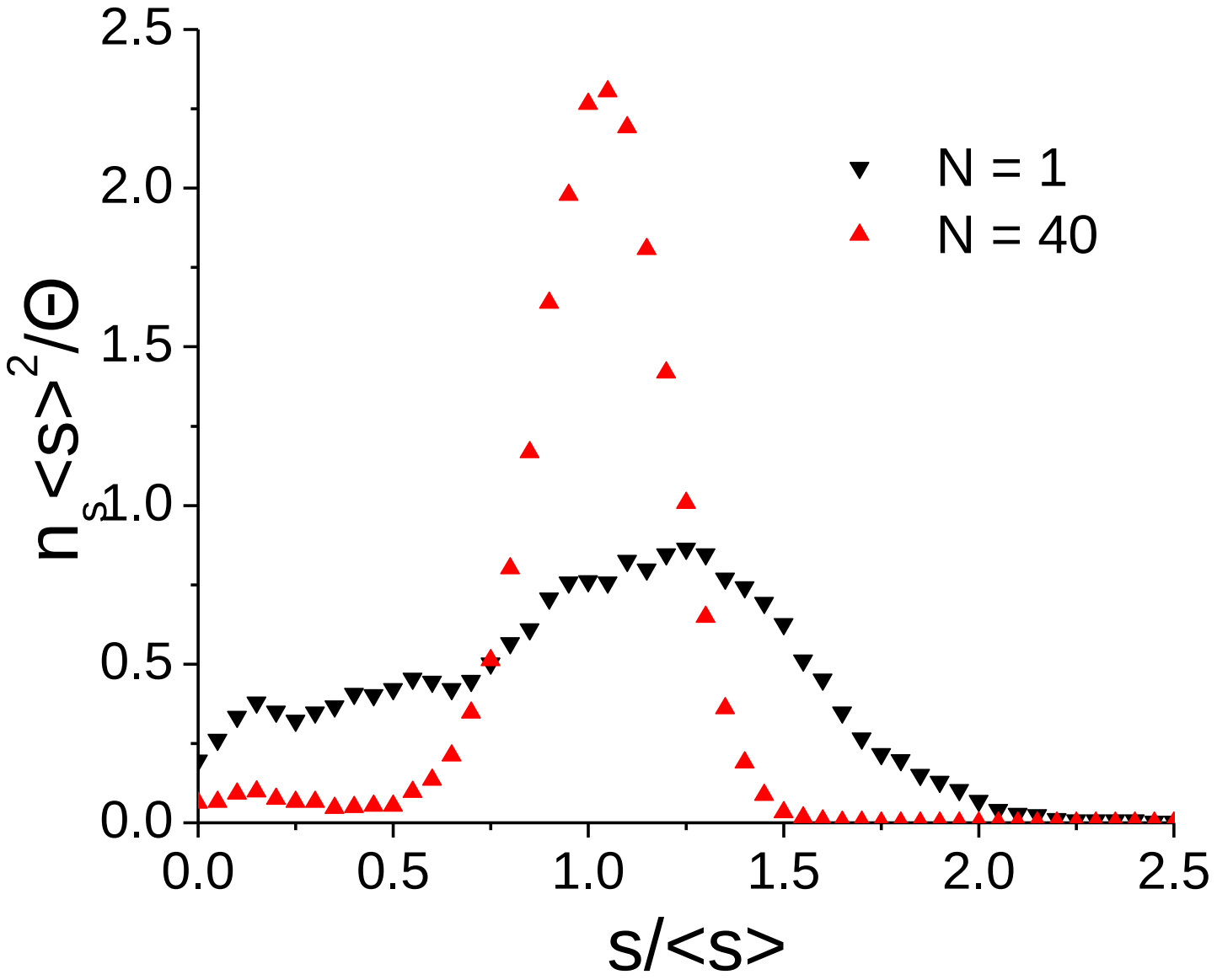


Simulation of Growth of 20 Superlayers





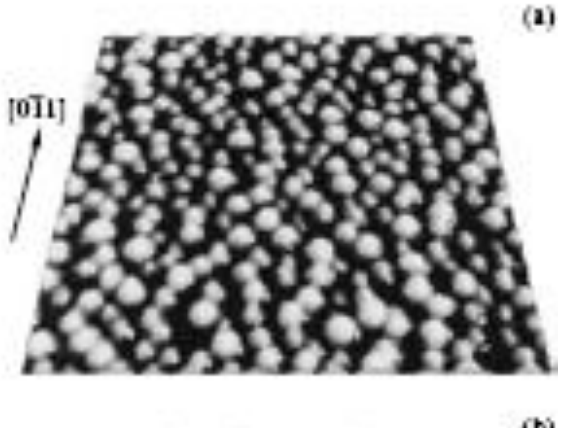
Regularization of Dot Size



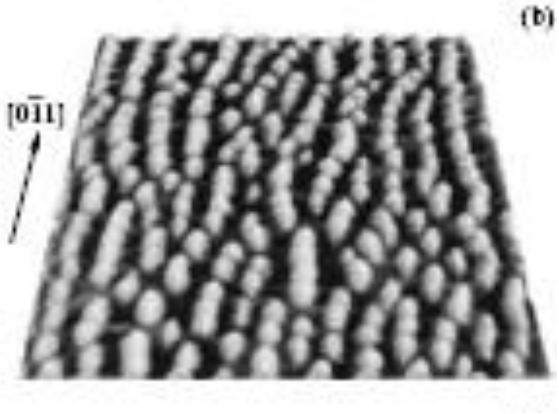
Ordering of Stacked Quantum Dots (Top View)

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

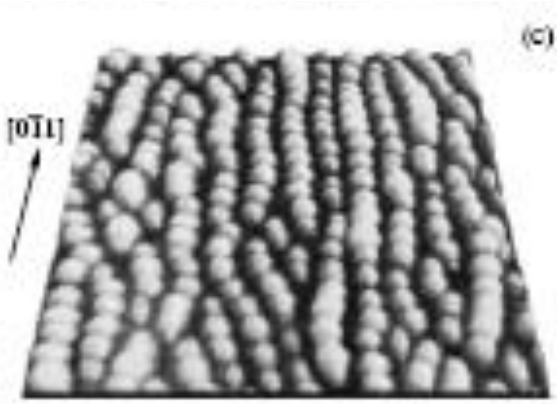
2 periods



7 periods



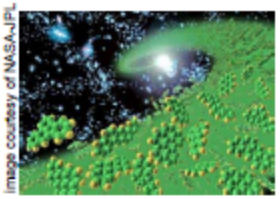
9 periods



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

Conclusions

- We have used DFT to calculate the strain dependence of adatom diffusion, dimer dissociation, edge diffusion, and detachment from island boundaries for Ag on Ag(100).
- 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, while tensile strain leads to broadening.
- We have modeled the aligned of stacked quantum dots.
- Ongoing work: Include step-edge barrier, via a Robin Boundary condition, and study effect of strain on the uphill current



INSTITUTE FOR PURE AND APPLIED MATHEMATICS
Los Angeles, California

Navigating Chemical Compound Space for Materials and Bio Design

March 14 – June 17, 2011

ORGANIZING COMMITTEE: Jean-Loup Faulon (Université d'Évry-Val d'Essonne), **William Hart** (Sandia National Labs), **Kendall Houk** (UCLA), **Peter Jones** (Yale), **Steven Lustig** (DuPont), **Tamar Seideman** (Northwestern), **Mark Tuckerman** (NYU), **Anatole von Lilienfeld** (Sandia National Labs)

Scientific Overview

Chemical compound space (CCS) is the combinatorial set which encompasses all chemical compounds. It can be viewed as the high dimensional space spanned by all the possible stoichiometries and configurations of electrons and atomic nuclei which form molecular or condensed matter. Due to the combinatorial nature of CCS, systematic screening for interesting properties or even simple enumeration is beyond any computational capacity. But CCS provides a natural framework in which to construct rigorous mathematical tools for the development of direct and inverse quantitative structure-property relationships, which can be applied to challenges in Materials and Bio design. Diverse scientific areas are involved, which benefit from historically grown experimental insights as well as advances made in theoretical and computational sciences. They include statistical mechanics, liquid and solid state physics, quantum chemistry, graph theory, molecular physics, condensed matter physics, optimization algorithms, data mining, statistical analysis, and others.

Workshop Schedule

- Chemical Compound Space Tutorials, March 15-18, 2011
- Workshop 1: Design of Drugs and Chemicals that Influence Biology, April 4-8, 2011
- Workshop 2: Optimization, Search and Graph-Theoretical Algorithms for Chemical Compound Space, April 11-15, 2011
- Workshop 3: Materials Design in Chemical Compound Space, May 2-6, 2011
- Workshop 4: Physical Frameworks for Sampling Chemical Compound Space, May 16-20, 2011
- Culminating Workshop at Lake Arrowhead Conference Center, June 12-17, 2011

Participation

This long program will bring together senior as well as junior researchers of diverse scientific communities, which are involved in addressing the question of how to best navigate CCS, such that they can discuss current bottlenecks with each other and, in particular, with the applied mathematics community. It is expected lead to fruitful collaborations where all participants benefit largely from mathematical insights on their specific optimization and design problems.

Full and partial support for long-term participants is available. We are especially interested in applicants who intend to participate in the entire program, but will consider applications for shorter periods. Funding is available to participants at all academic levels, though recent PhDs, graduate students, and researchers in the early stages of their careers are especially encouraged to apply. Encouraging the careers of women and minority mathematicians and scientists is an important component of IPAM's mission and we welcome their applications. More information and a link to an application are available online.

www.ipam.ucla.edu/programs/ccs2011

UCLA

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