

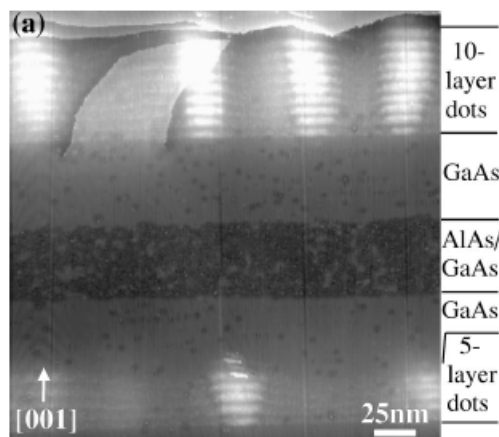
A Level-Set Method for Self-Organized Pattern Formation during Heteroepitaxial Growth

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Institute for Pure and Applied Mathematics, and Department of Mathematics

Collaborators: Xiaobin Niu, Russ Caflisch, Young-Ju Lee, Jason DeVita, Peter Smereka

Motivation: strain induced ordering in experiments

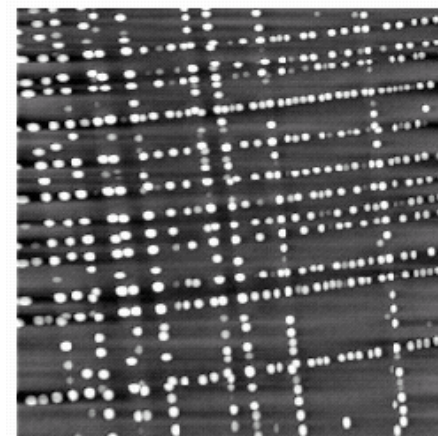


$\text{Al}_x\text{Ga}_{1-x}\text{As}$
system

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

GeSi
system

H. J. Kim, et al.,
PRB 68, (2003).

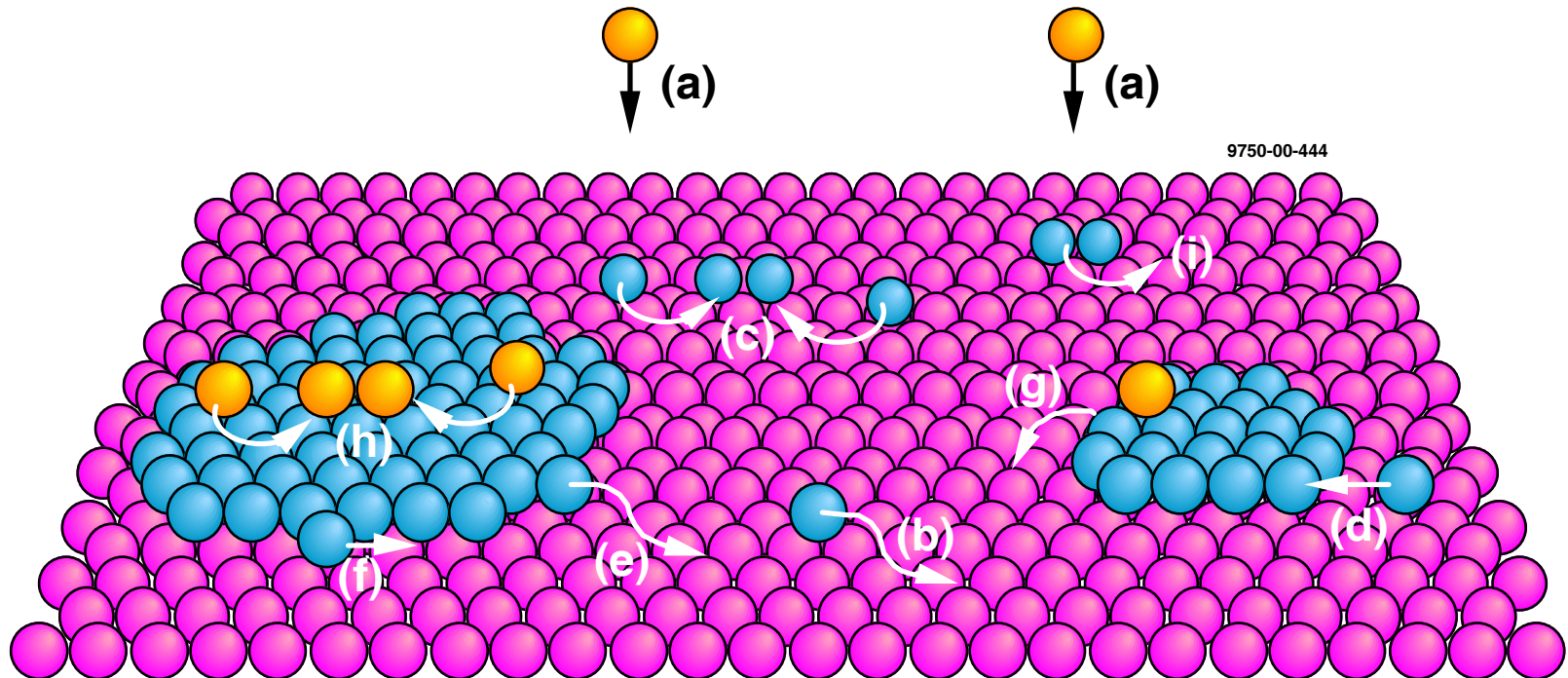


Goal: Develop a kinetic model that includes strain!

Outline of this Talk

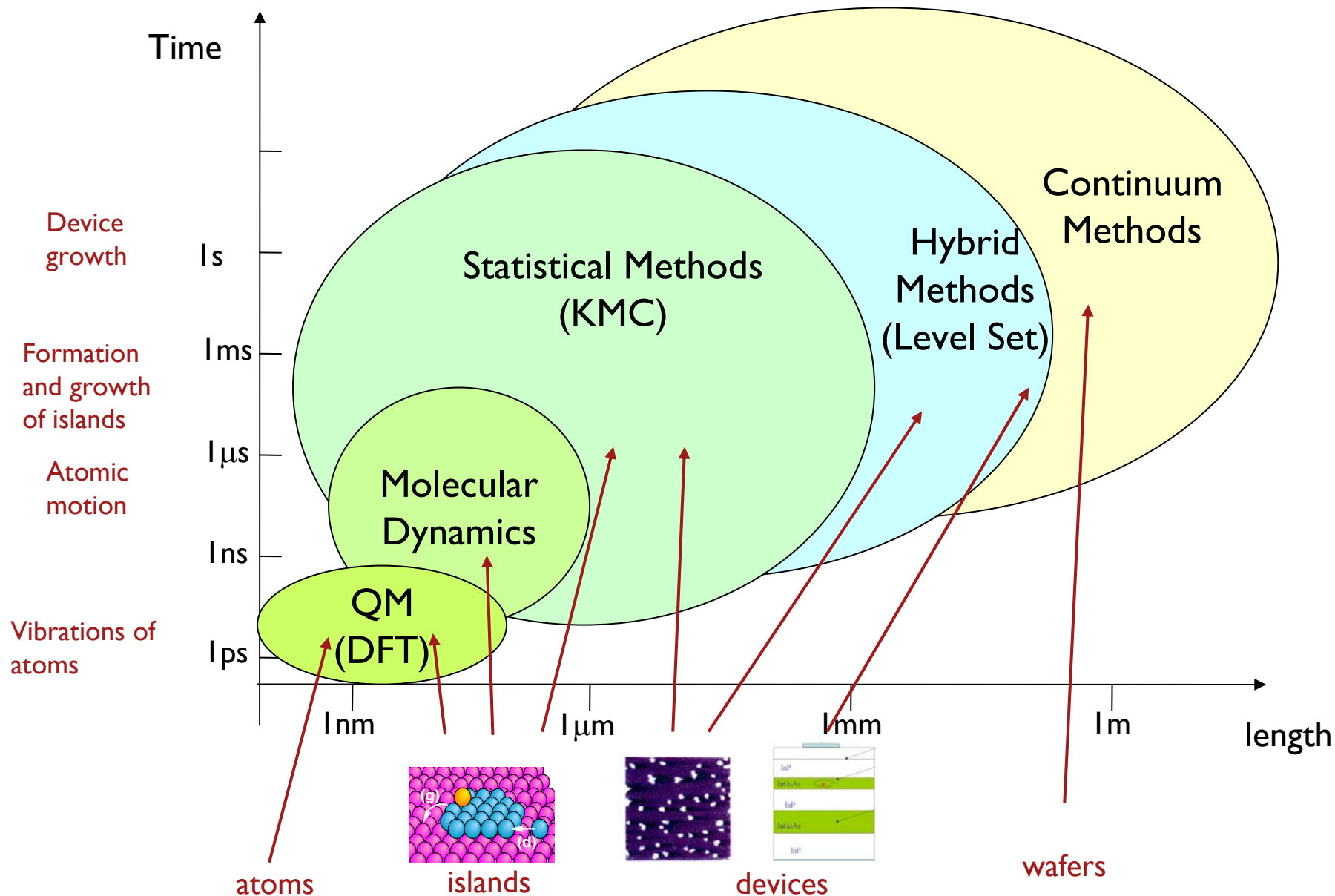
- **Introduction**
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- Spatially varying potential energy surface
(due to surface defects, reconstructions, ...)
- Our elastic model
- Ordering in the submonolayer growth regime
- Ordering of stacked quantum dots

Physical Processes During Epitaxial Growth

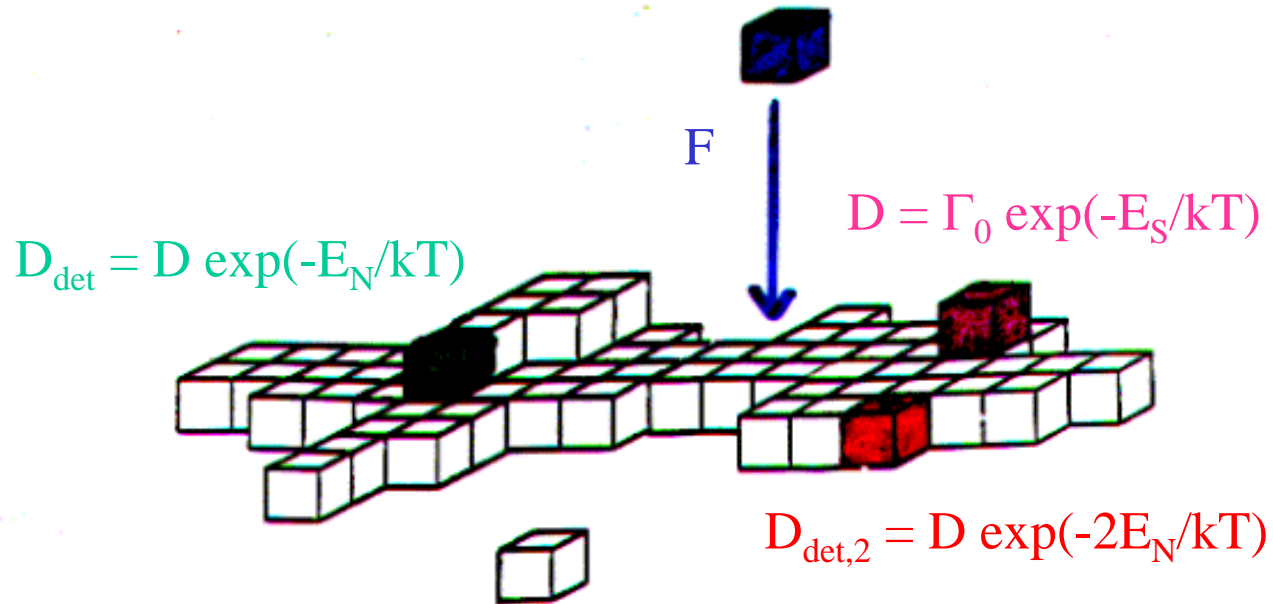


Atomic Motion	Time Scale $\sim 10^{-13}$ seconds	Length Scale: Ångstrom
Island Growth	Time Scale \sim seconds	Length Scale: Microns

Hierarchy of Theoretical Approaches



KMC Simulation of a Cubic, Solid-on-Solid Model



E_S : Surface bond energy

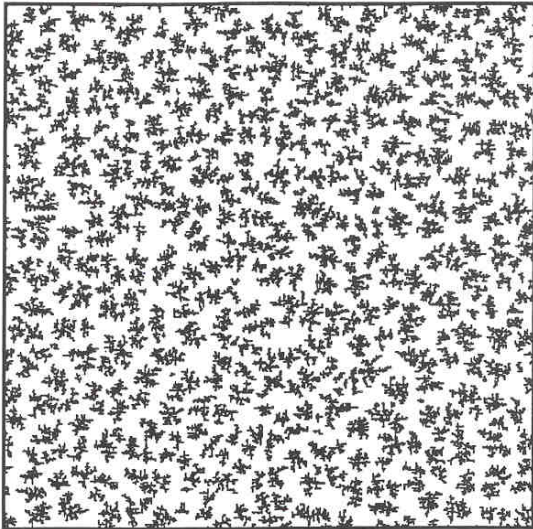
E_N : Nearest neighbor bond energy

G_0 : Prefactor [$O(10^{13}\text{s}^{-1})$]

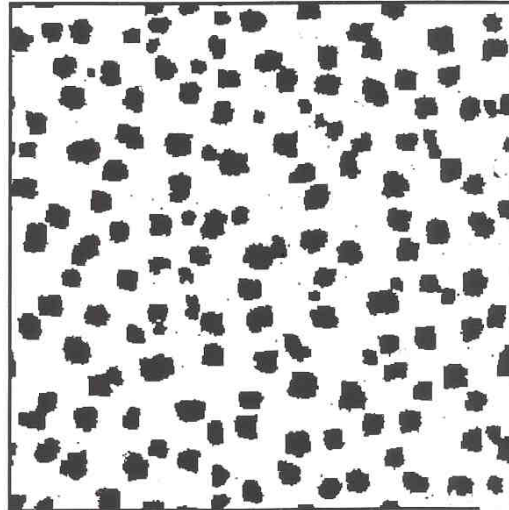
- Parameters that can be calculated from first principles (e.g., DFT)
- Completely stochastic approach

KMC Simulations: Effect of Nearest Neighbor Bond EN

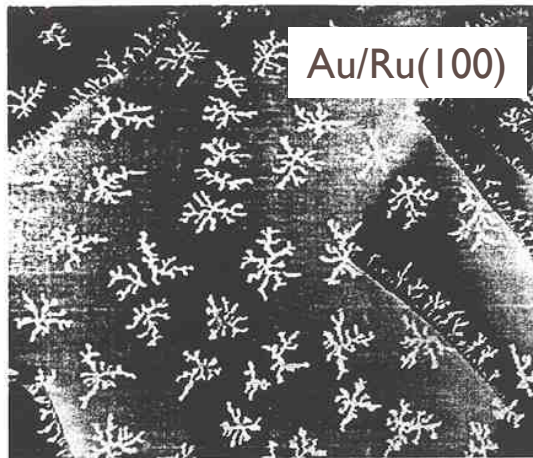
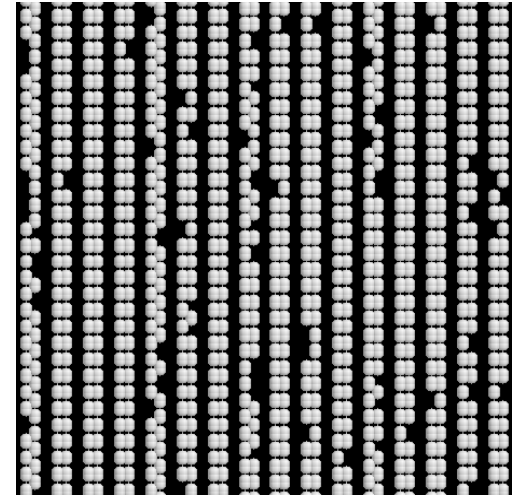
Large E_N :



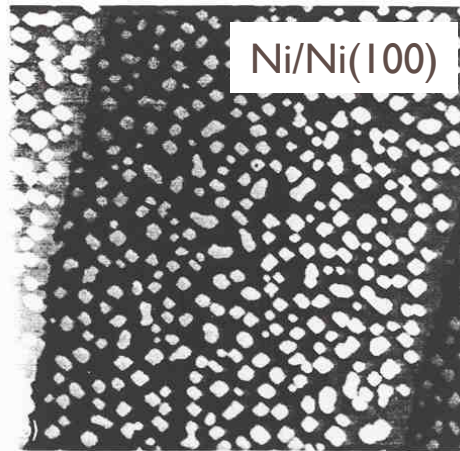
Small E_N :



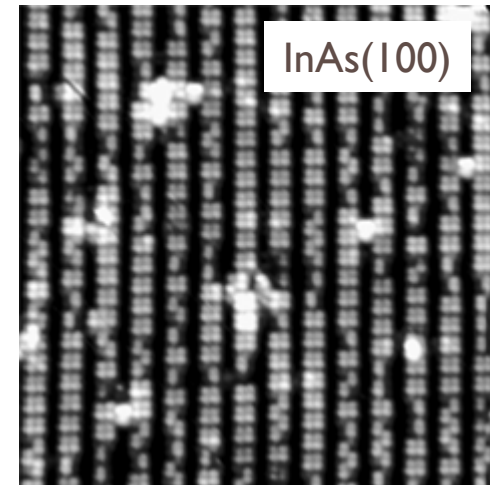
More Detailed KMC model



Au/Ru(100)



Ni/Ni(100)



InAs(100)

Hwang et al., PRL 67 (1991)

Kopatzki et al., Surf.Sci. 284 (1993)

F. Grosse et al. PRL 89 (2002)

Scaling of Island Size Distribution Function from KMC

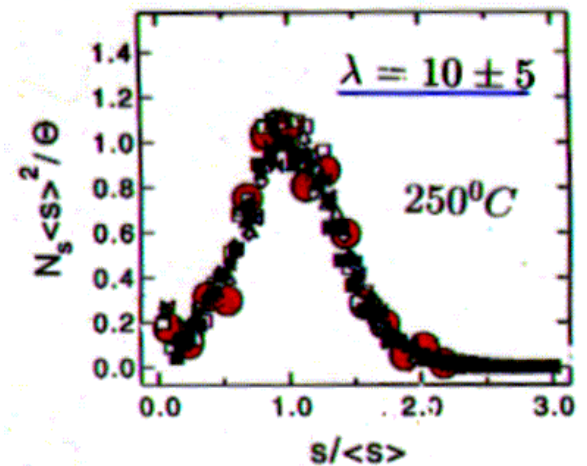
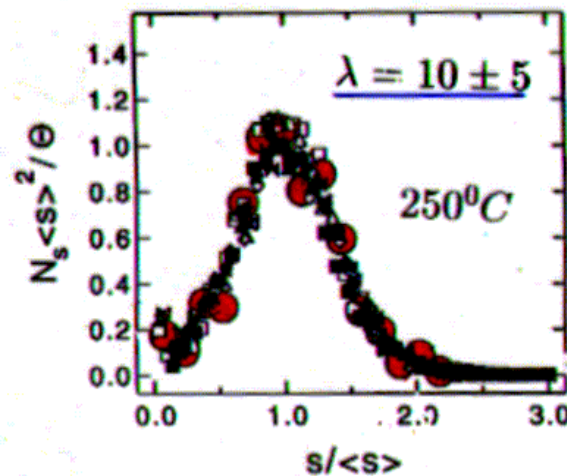
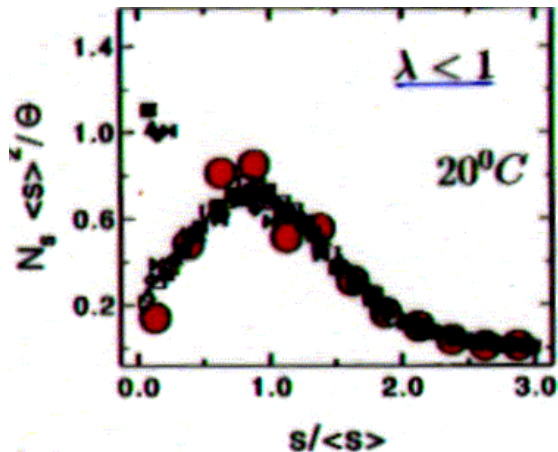
Scaling form for island densities N_s :

$$N_s = \frac{\Theta}{s_{av}^2} g\left(\frac{s}{s_{av}}\right)$$

Θ : Coverage

s_{av} : Average island size

KMC Simulation vs. Experiment [(Fe/Fe(100); Stroscio et al., 1993)]



- KMC reproduces scaling with D/F and coverage Q in agreement with experiment
- Scaling function depends only on degree of reversibility

Ratsch, Smilauer, Zangwill, Vvedensky, Phys. Rev. Lett., 1994; Surf. Sci. Lett., 1995

The challenge for including strain in a growth model

- 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

- solve it only locally, maybe not even every timestep
- do only 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 of this Talk

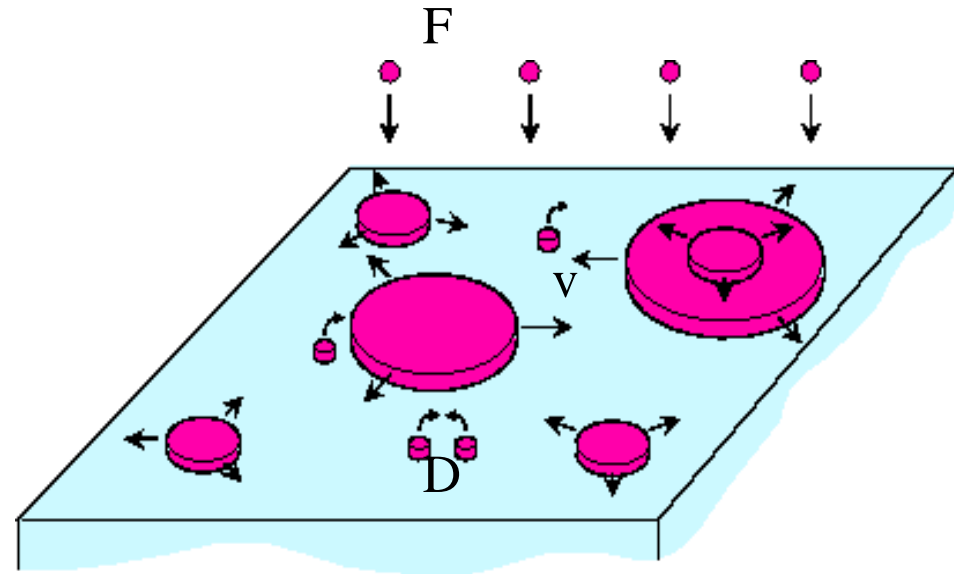
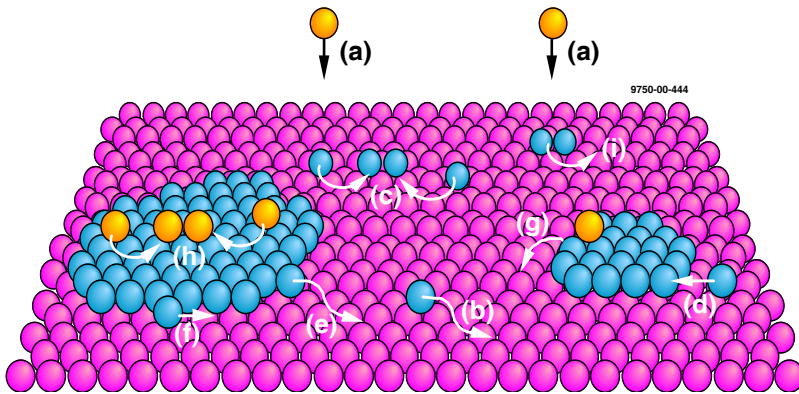
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The Island Dynamics Model for Epitaxial Growth

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

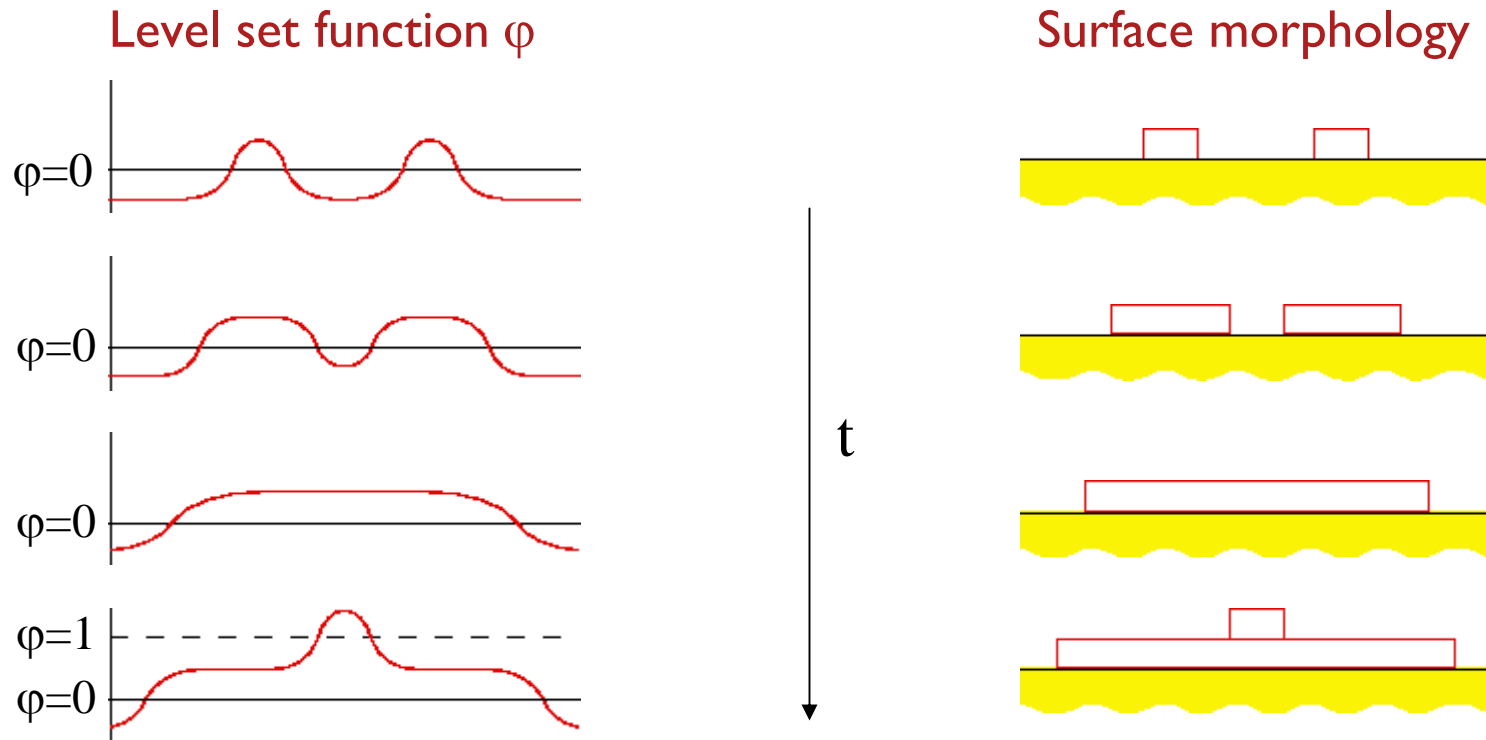


Island dynamics



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

The Level Set Method

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

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

ρ : Adatom concentration

Diffusion in x-direction

Diffusion in y-direction

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

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

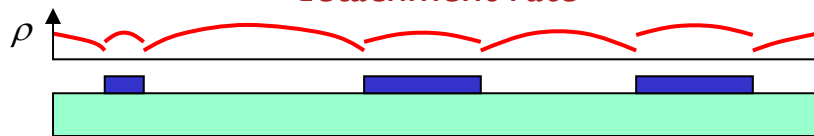
Nucleation rate $\sim D\rho(\mathbf{x}, t)^2$

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

- Boundary condition:

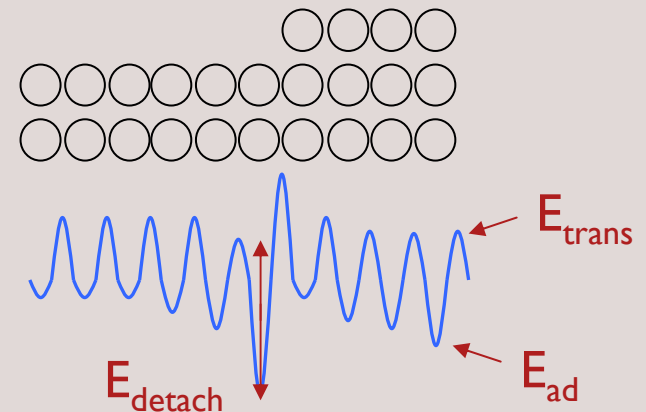
$$\rho = \rho_{eq}(D_{\text{det}}, \mathbf{x})$$

detachment rate



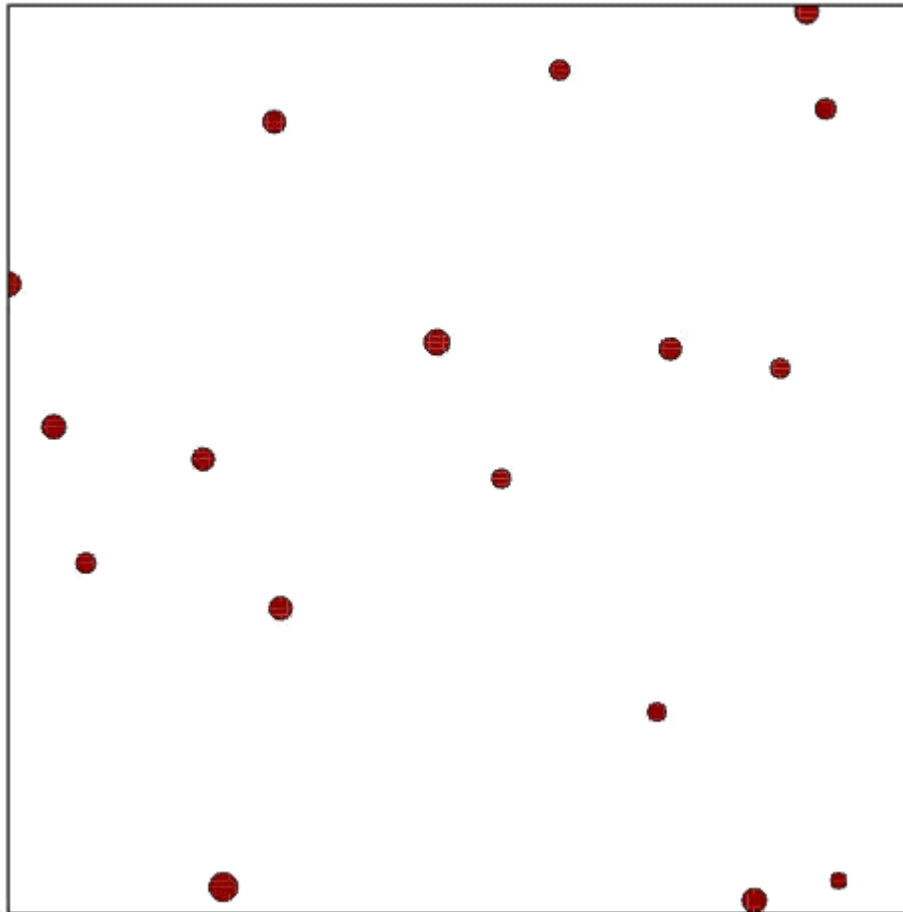
- Stochastic element needed for nucleation

A typical potential energy surface

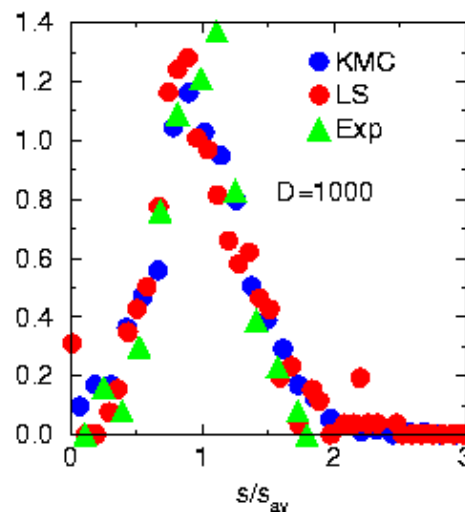
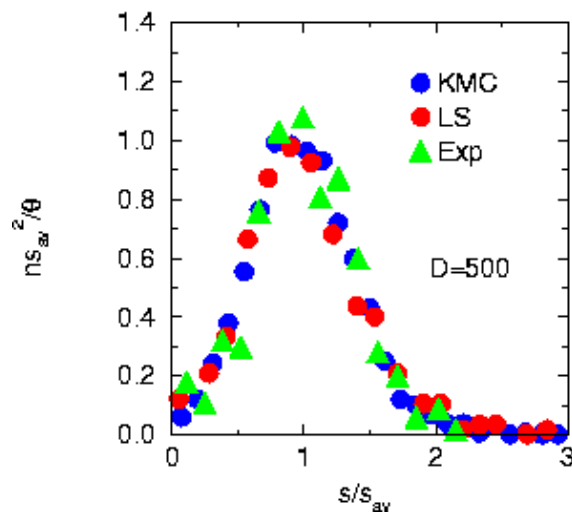
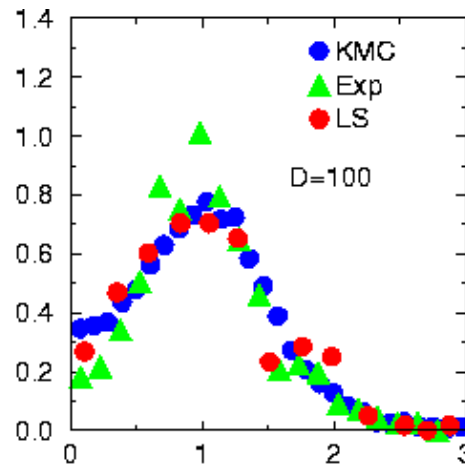
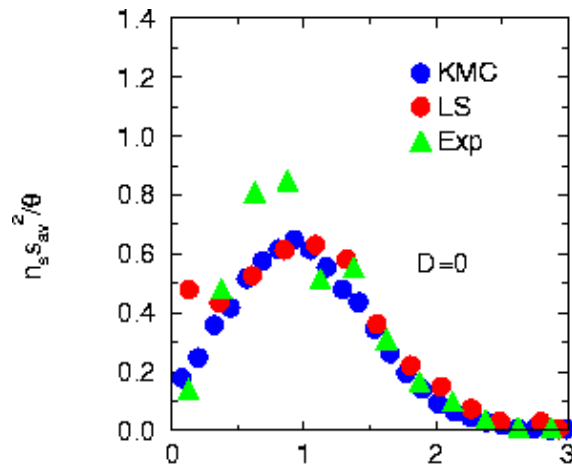


A typical level set simulation

$t = 0.00$



Validation by comparison of scaled island size distribution



Experimental Data
for Fe/Fe(001),

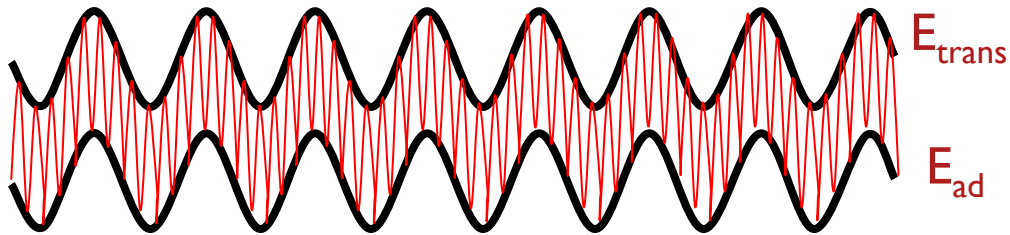
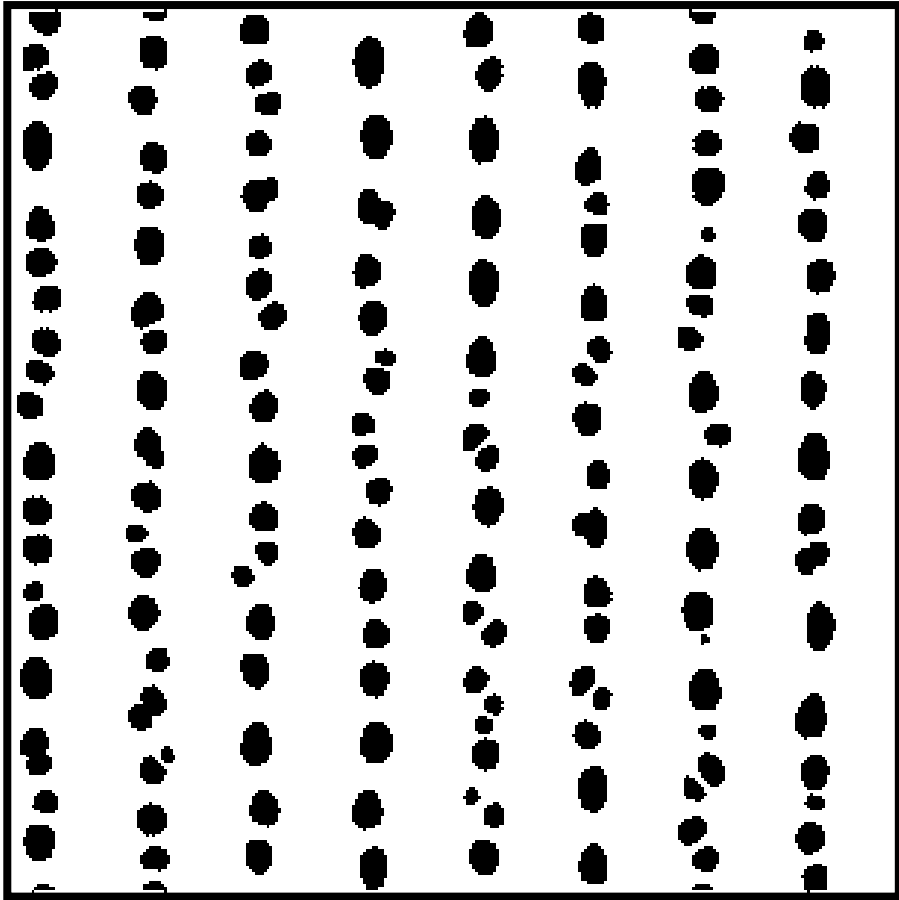
Stroscio and Pierce, Phys.
Rev. B 49 (1994)

Petersen, Ratsch, Caflisch, Zangwill, Phys. Rev. E 64, 061602 (2001).

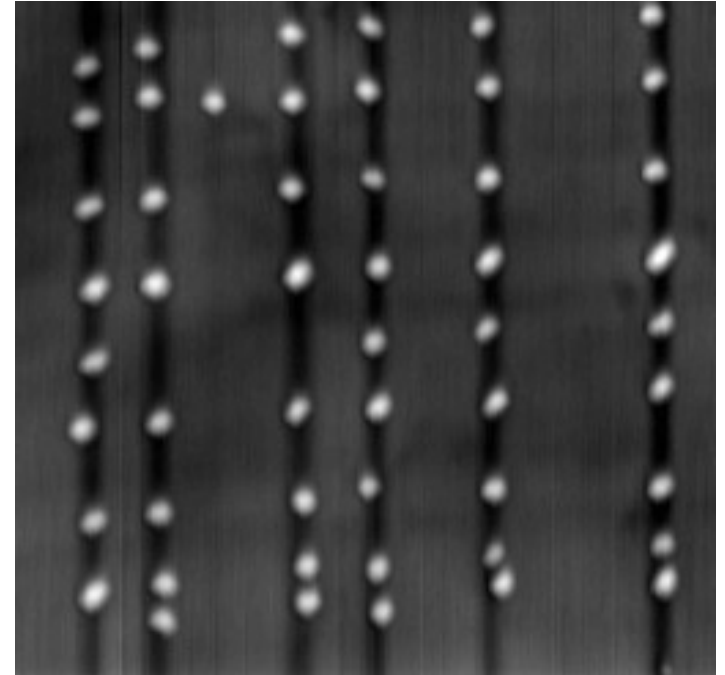
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- **Spatially varying potential energy surface**
(due to surface defects, reconstructions, ...)
- Our elastic model
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Assume spatially varying potential energy surface (no strain)



Experiment by Xie et al., UCLA

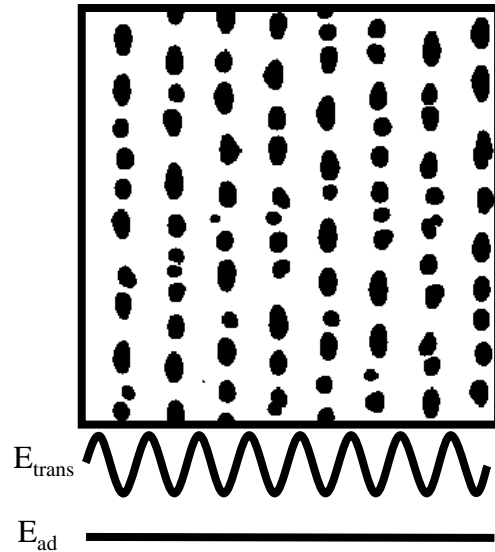


Interpretation:

Variation of potential energy surface is due to strain, that results from buried defect lines

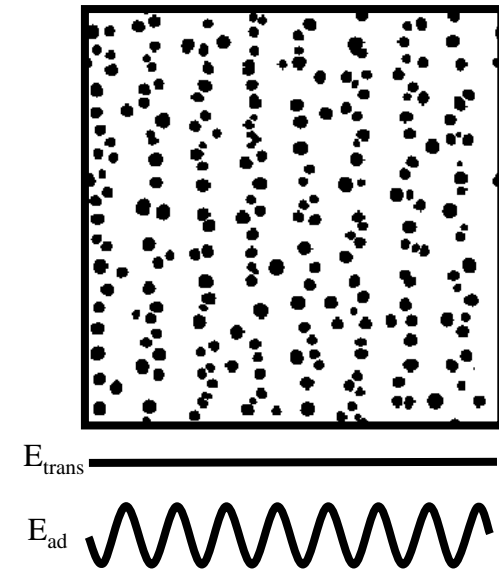
Variation of adsorption and transition energy

Kinetic limit



Nucleation in region of fast diffusion

Thermodynamic limit

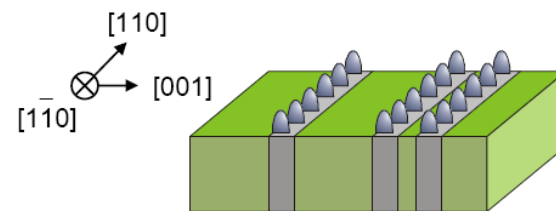
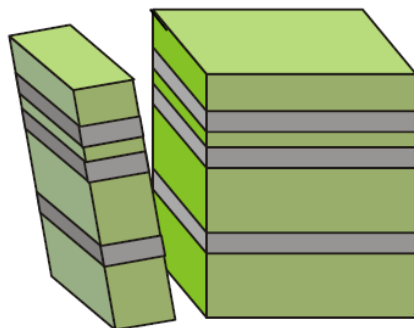
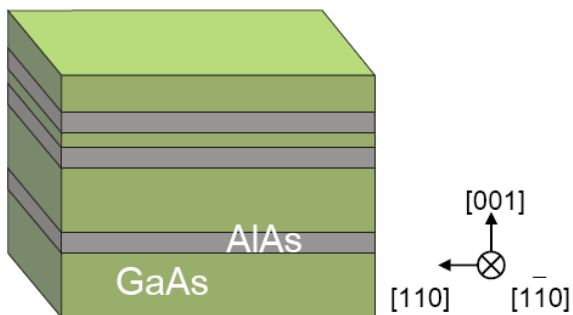


Nucleation in region of slow diffusion (but high adatom concentration), dominated by drift

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

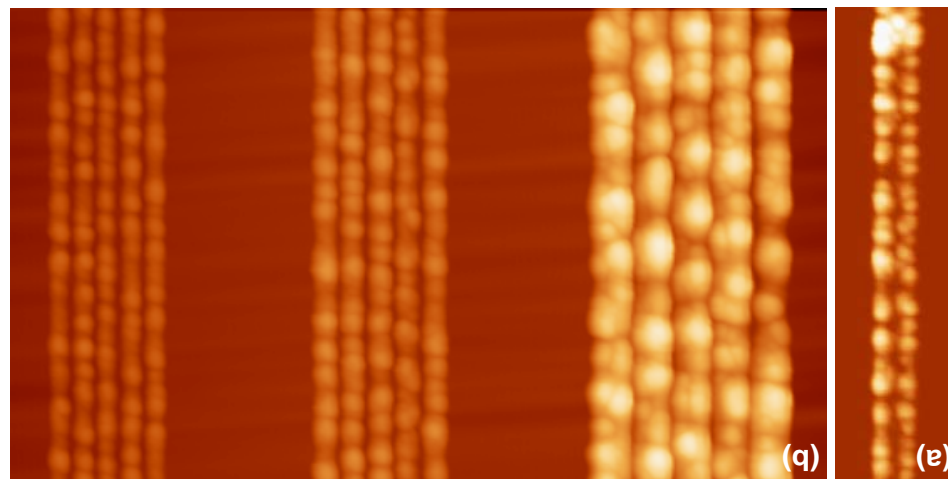
Ordering by Cleaved Edge Overgrowth

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



Quantum dots grow on top of the AlAs stripes

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



Width of AlAs stripe:

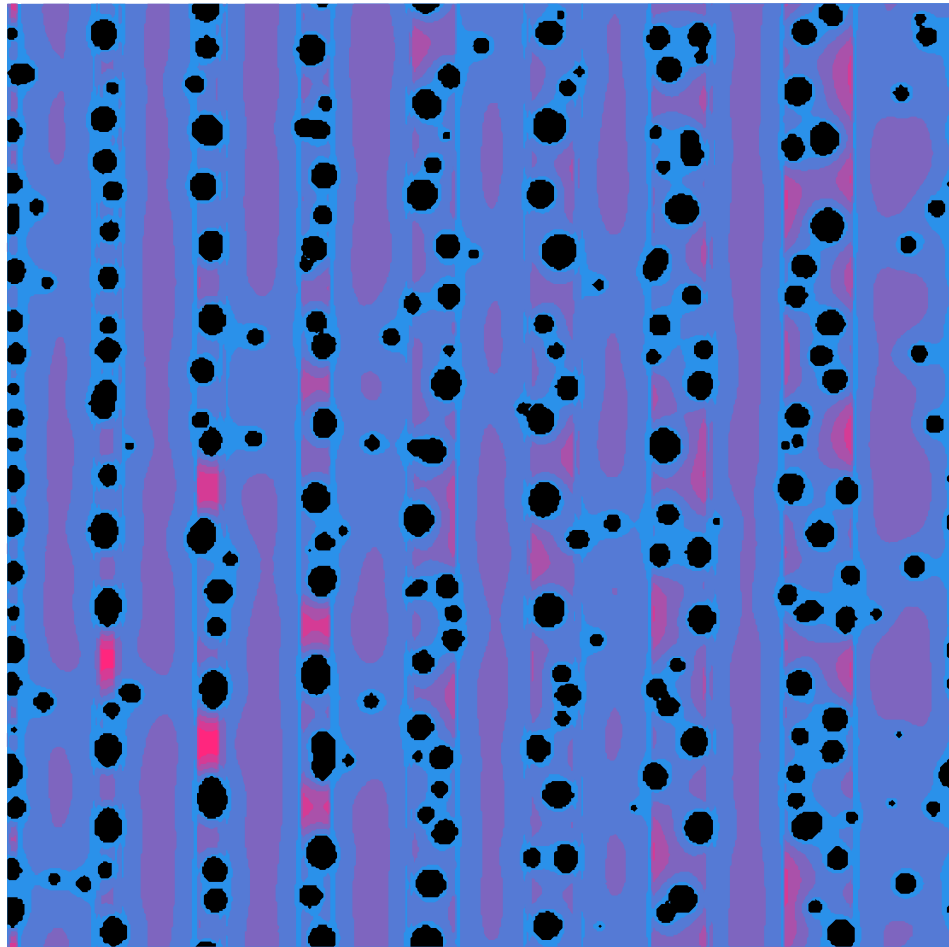
30 nm

42 nm

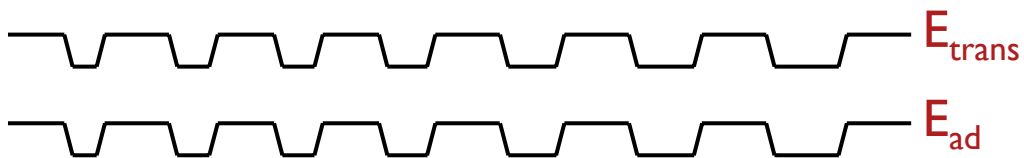
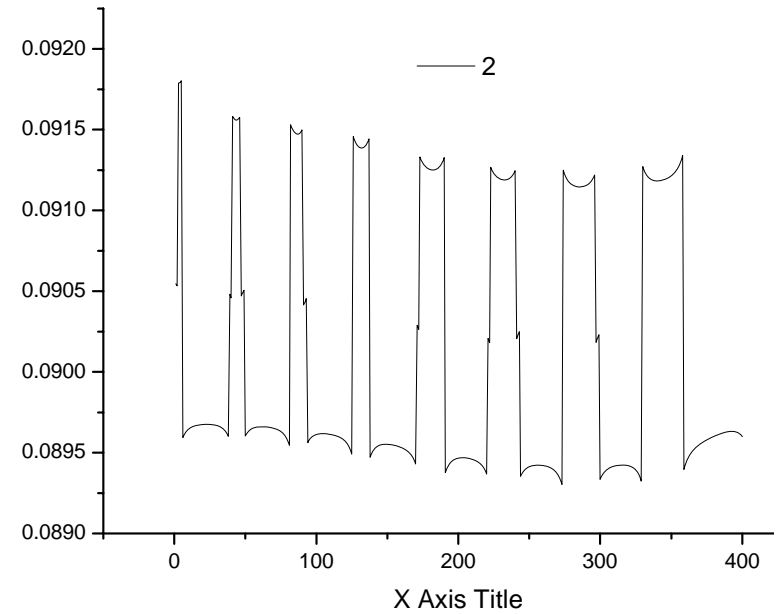
80 nm

100 nm

Level set simulations



Nucleation rate as function of position
for increasing width of AIAs stripe

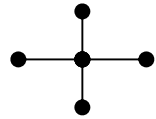


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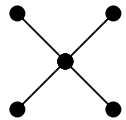
Include Strain: Calculate Elastic Field at Every Timestep

- 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

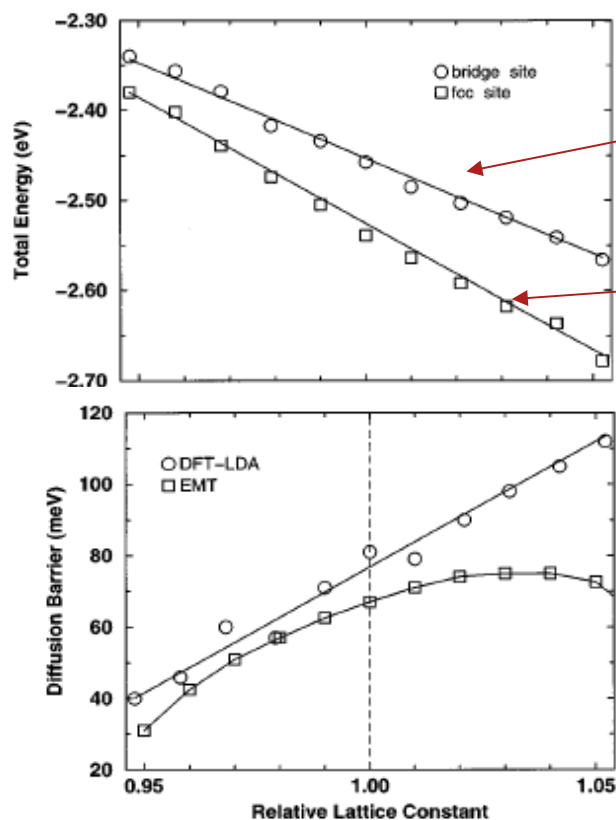
$$E = \alpha(S_{xx}^2 + S_{yy}^2) + \beta S_{xy}^2 + \gamma S_{xx} S_{yy}$$

- Minimize energy with respect to all displacements: $\partial_u E[u] = 0$
- The relevant microscopic parameters at every grid point can then be varied as a function of the local strain.

How does Strain affect the Parameters in our Model?

Density-functional theory (DFT) has been used to study strain dependence of surface diffusion D

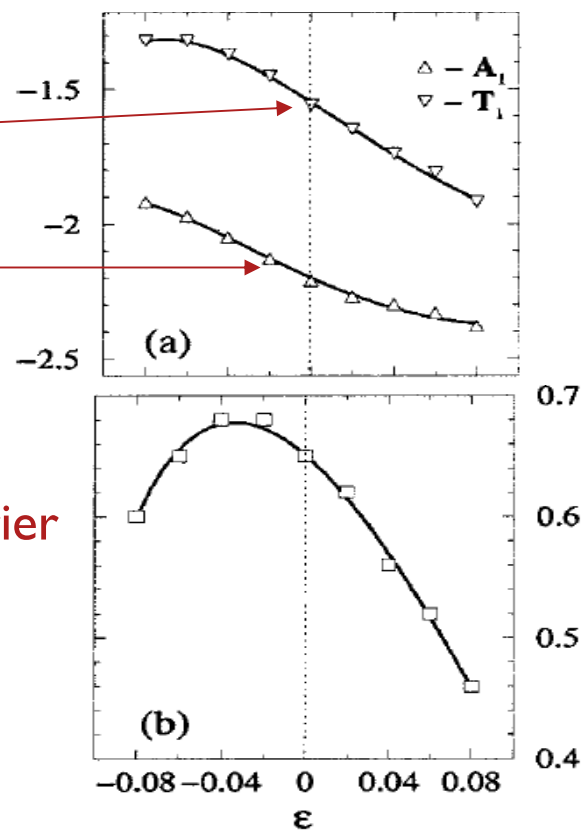
Ag/Ag(111) (a metal)



GaAs(100) (a semiconductor)

E_{trans}

E_{ad}



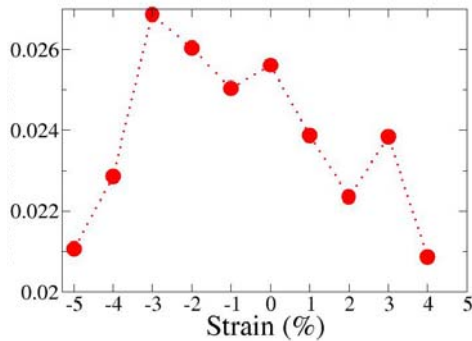
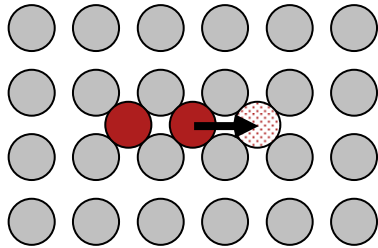
Energy barrier
for surface
diffusion

Ratsch et al. Phys. Rev. B **55**, 6750-6753 (1997).

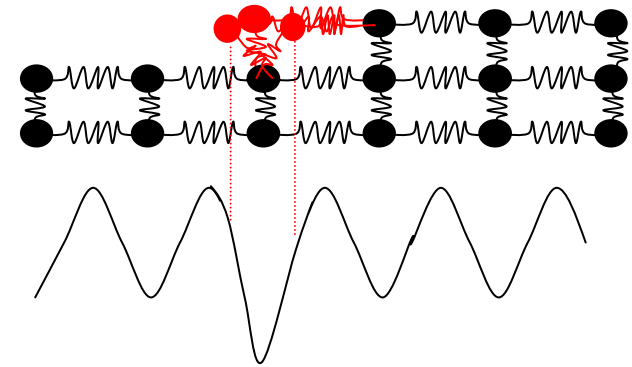
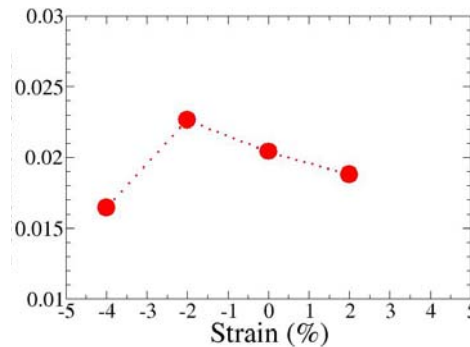
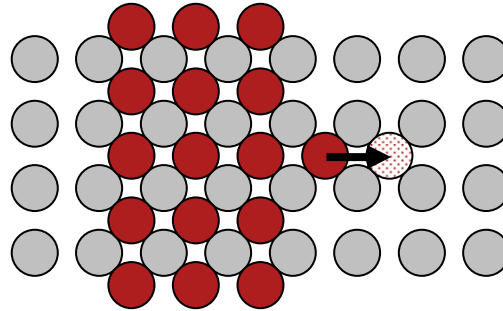
E. Penev et al., Phys. Rev. B **64**, 085401 (2001).

Dimer Dissociation and Detachment for Ag/Ag(100)

Dimer dissociation



Adatom detachment



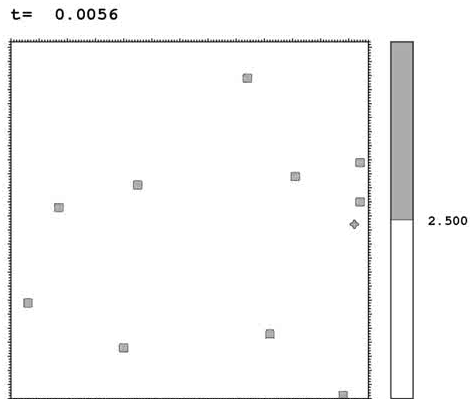
Preliminary DFT Results suggest decrease of energy barrier for dimer dissociation and adatom detachment upon tensile and compressive strain:

$$D_{\text{det}} = D_{\text{det},0} \exp\left(\frac{\Delta E_{\text{Strain}}}{k_B T}\right)$$

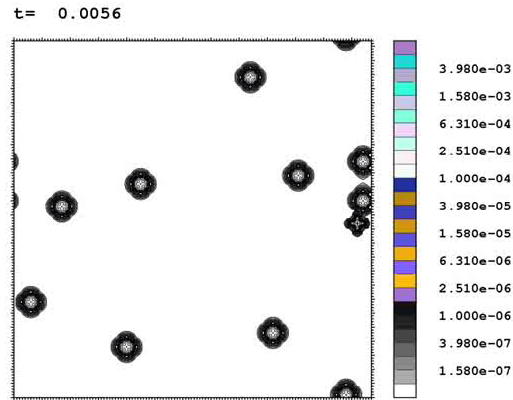
Tests show that the dependence of D_{det} is more important for ordering of island sizes, while dependence of D is more important for ordering of location.

Effect of Strain in the Simulation

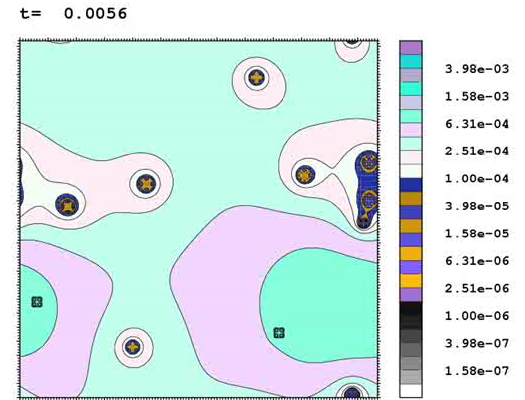
morphologies



Elastic energies

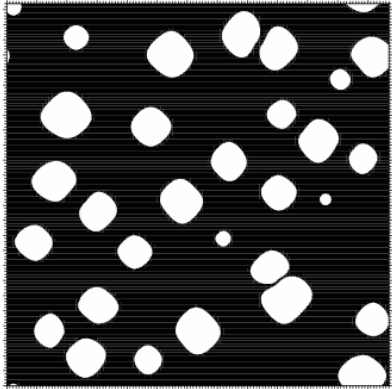


Adatom concentration

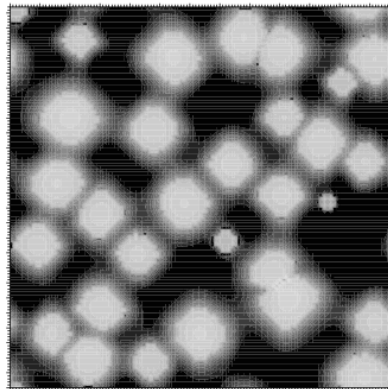


Effect of Strain in the Simulation

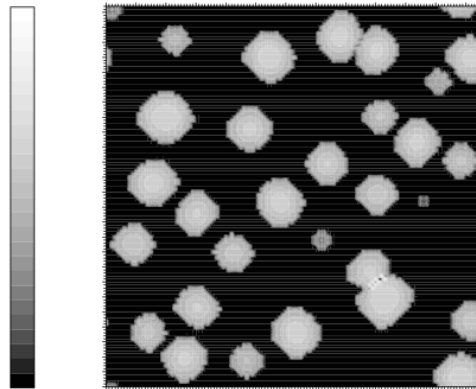
Morphologies



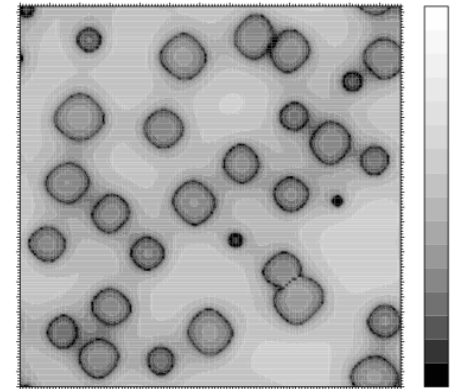
Elastic energies



Detachment rate

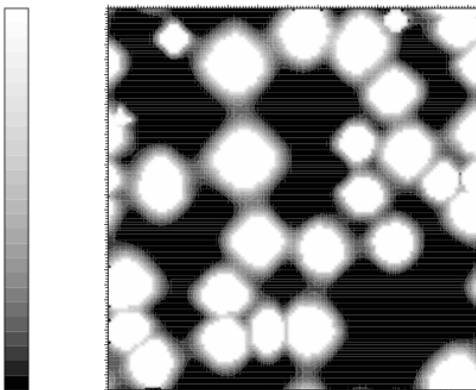
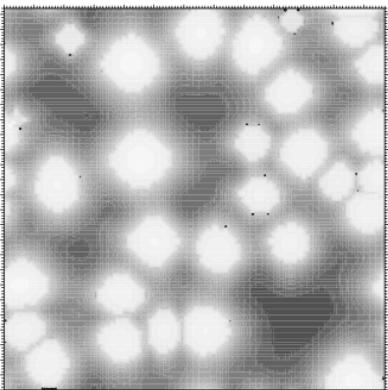
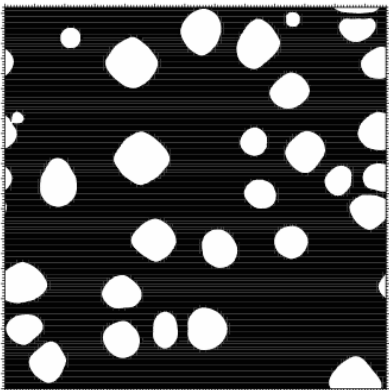


Adatom concentration



Top row: Strain=1%

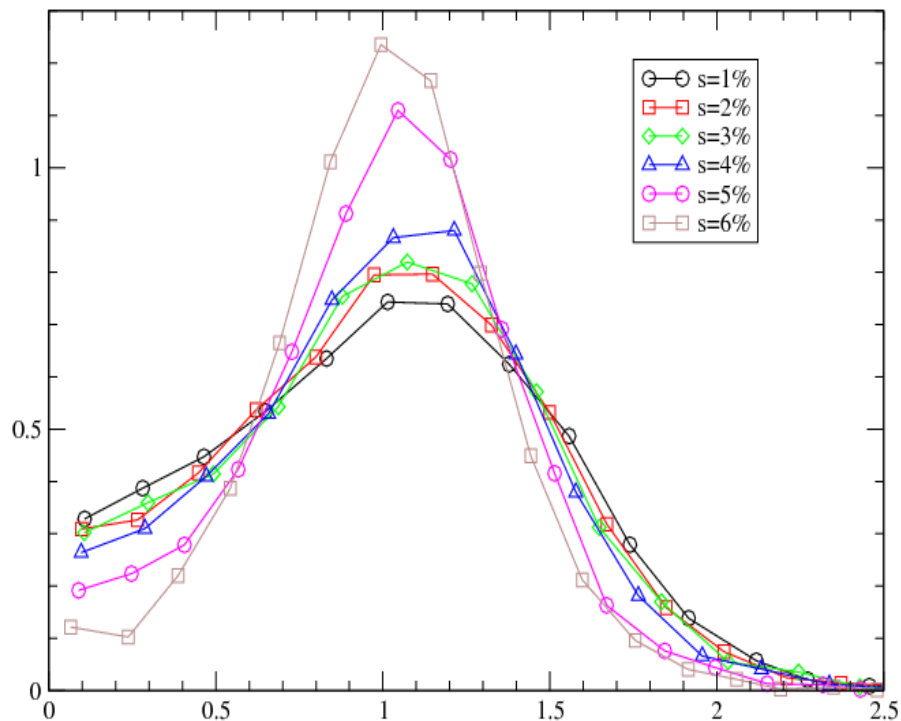
Bottom row: Strain=5%



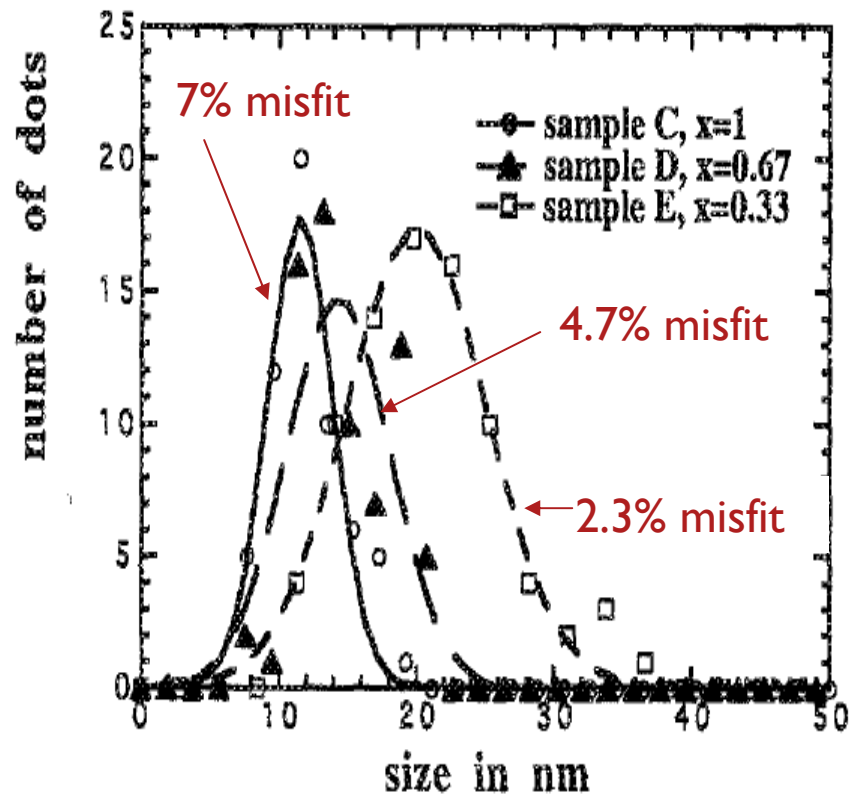
- With increasing strain, islands become more regular, because small islands are more likely to break up, and growth of large islands slows down.

Sharpening of the Scaled Island Size Distribution

Level-set simulation



Experiment: $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}(100)$

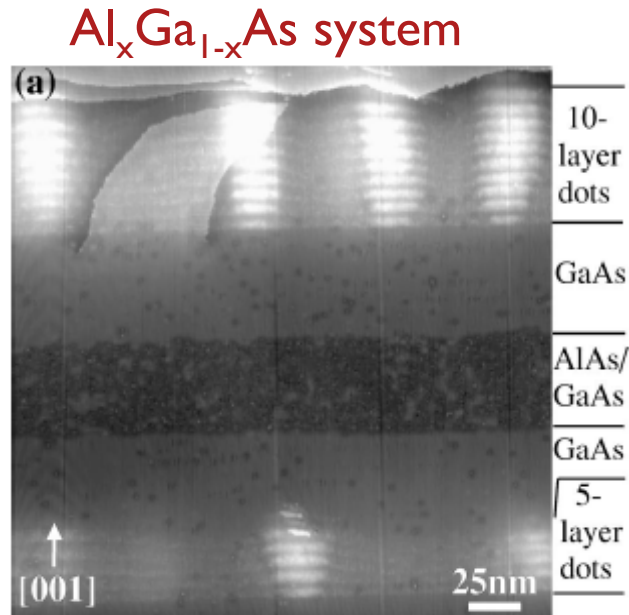


D. Leonard, M. Krishnamurthy, S. Fafard, J.L. Merz, and P.M. Petroff, *J. Vac. Sci. Tech B* 12, 1063 (1994)

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Simulation of Stacked Quantum Dots

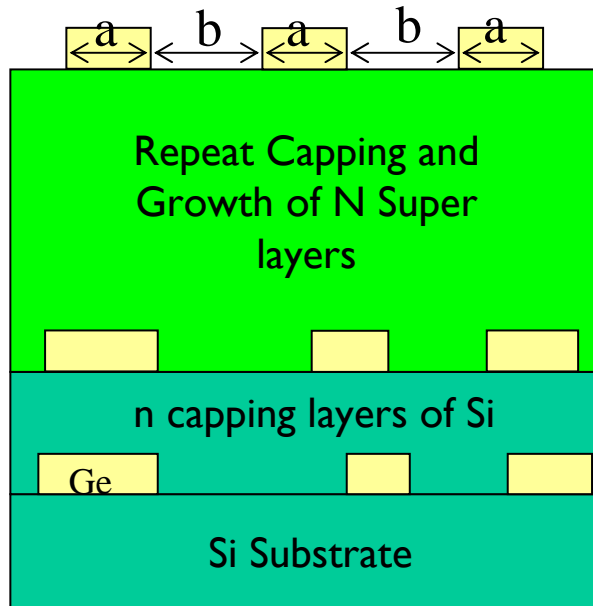


B. Lita et al., APL 74, (1999)

Experimental observation: Stacked quantum dots align under certain conditions

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

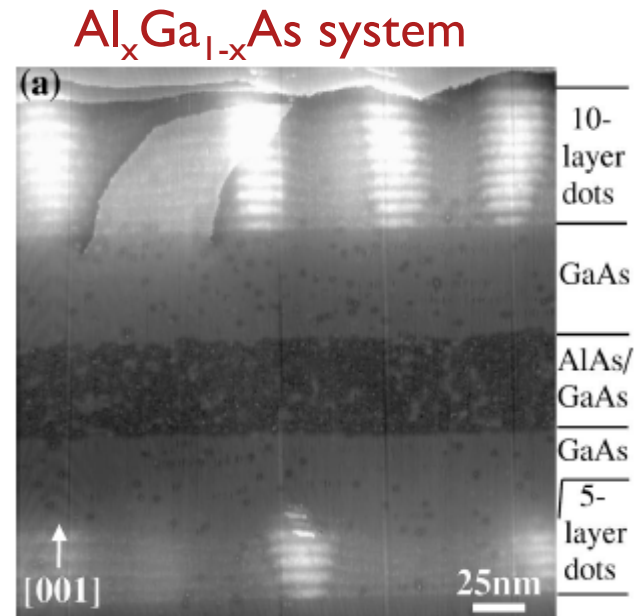
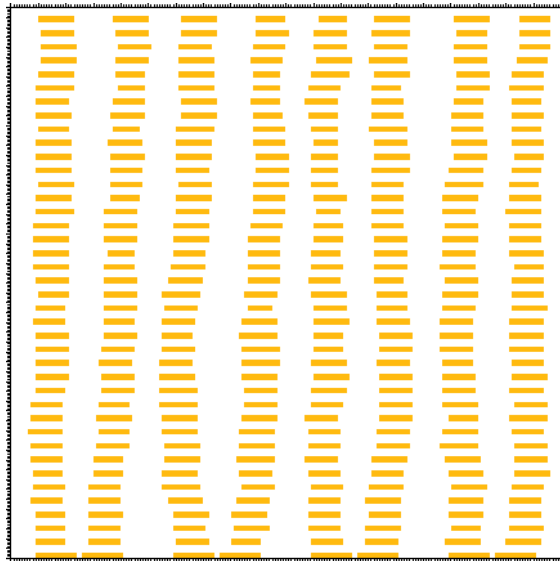
Simulation of Stacked Quantum Dots



- 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

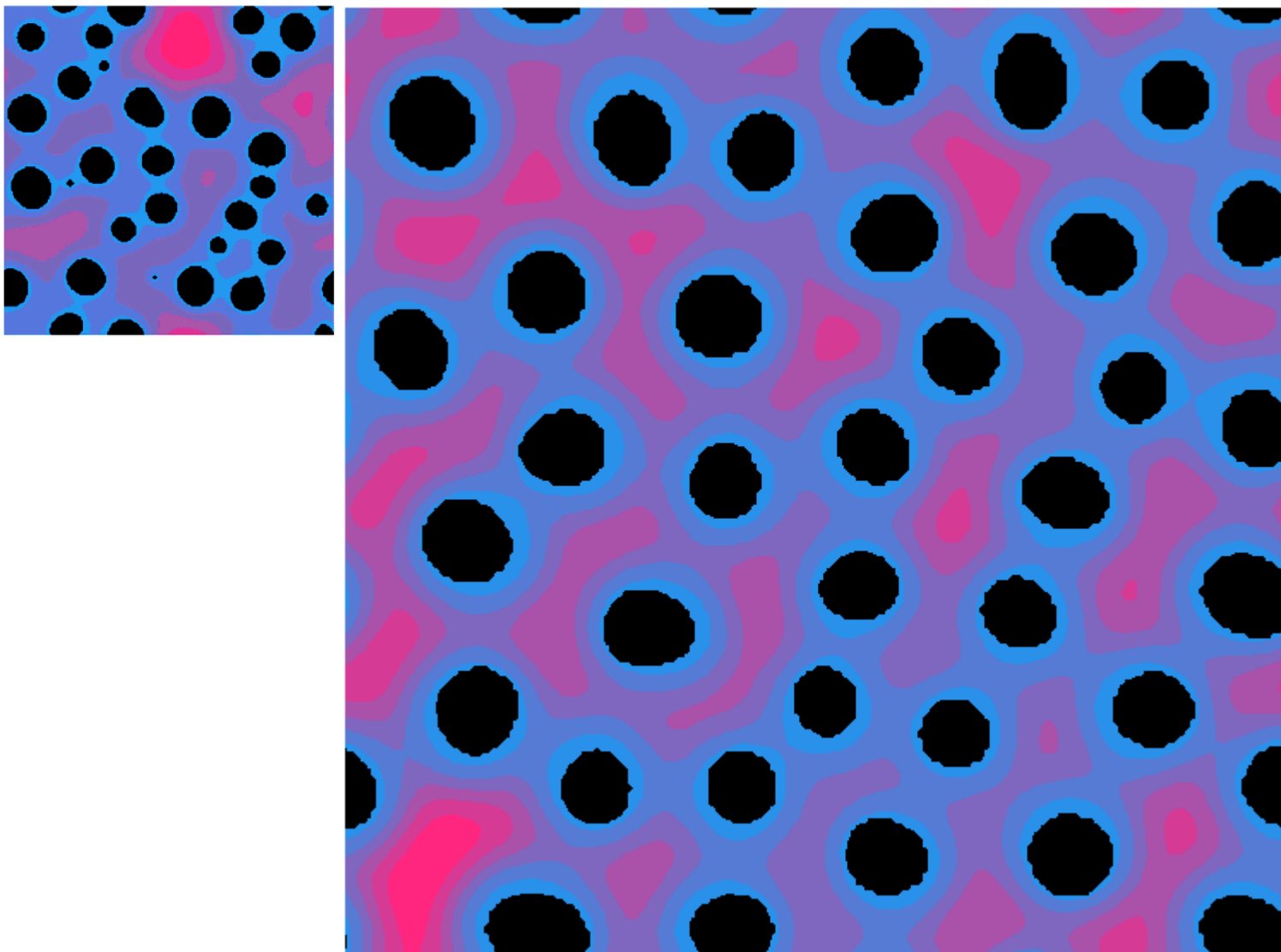


B. Lita et al., APL 74, (1999)

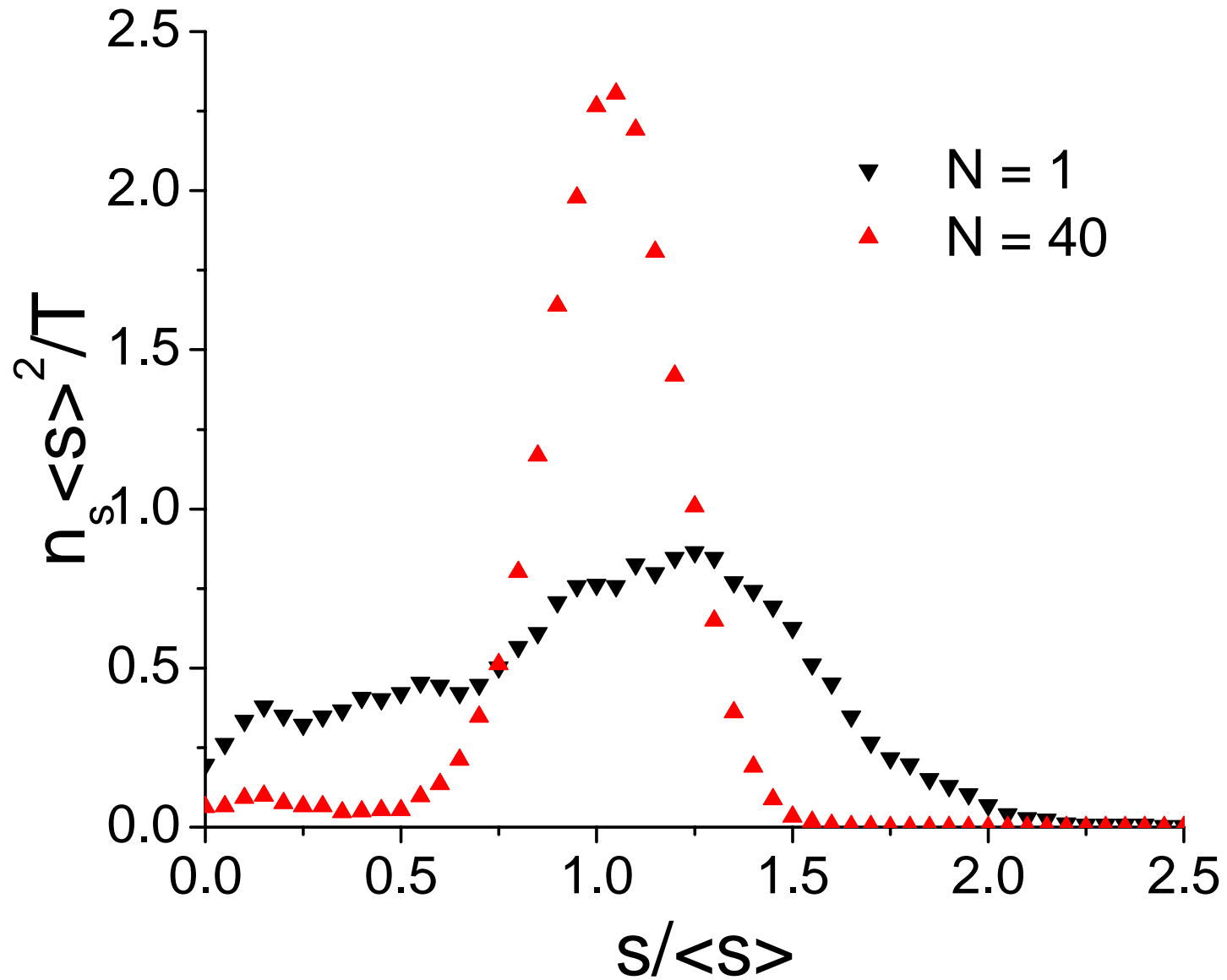
- Spacing and size of stacked dots becomes more regular

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

Simulation of growth of 20 superlayers



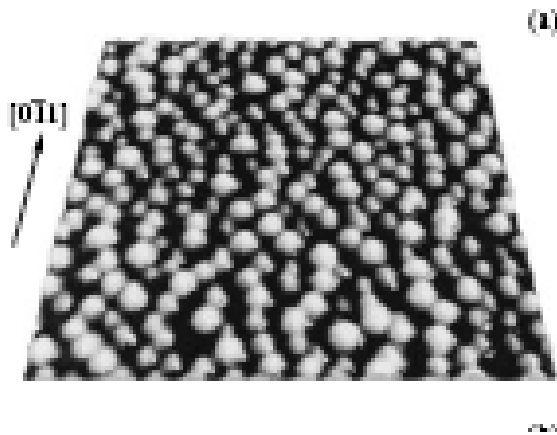
Regularization of dot size



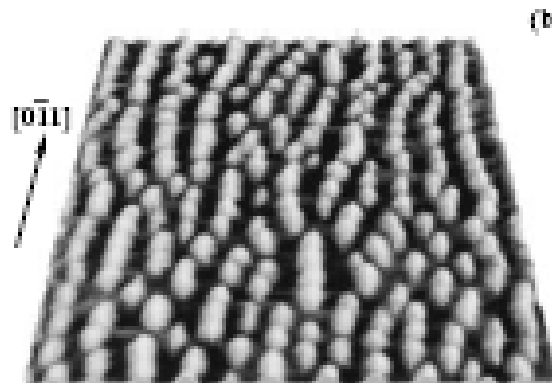
Ordering of stacked quantum dots (top view)

Growth of stacked quantum dots of $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}/\text{GaAs}(100)$

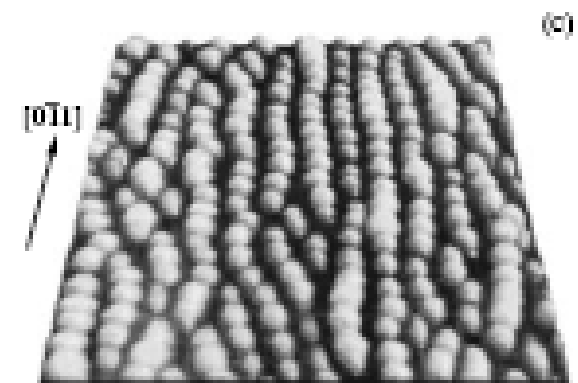
2 periods



7 periods



9 periods

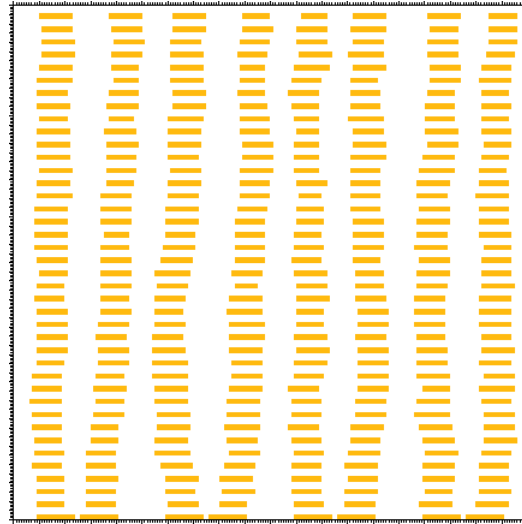


V.V. Strel'chuk et al., *Semiconductors* 41 (2007)

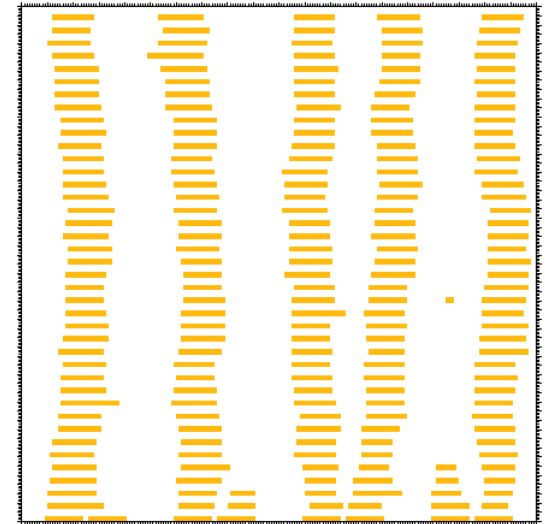
Thickness dependence of vertical ordering



0 capping layer



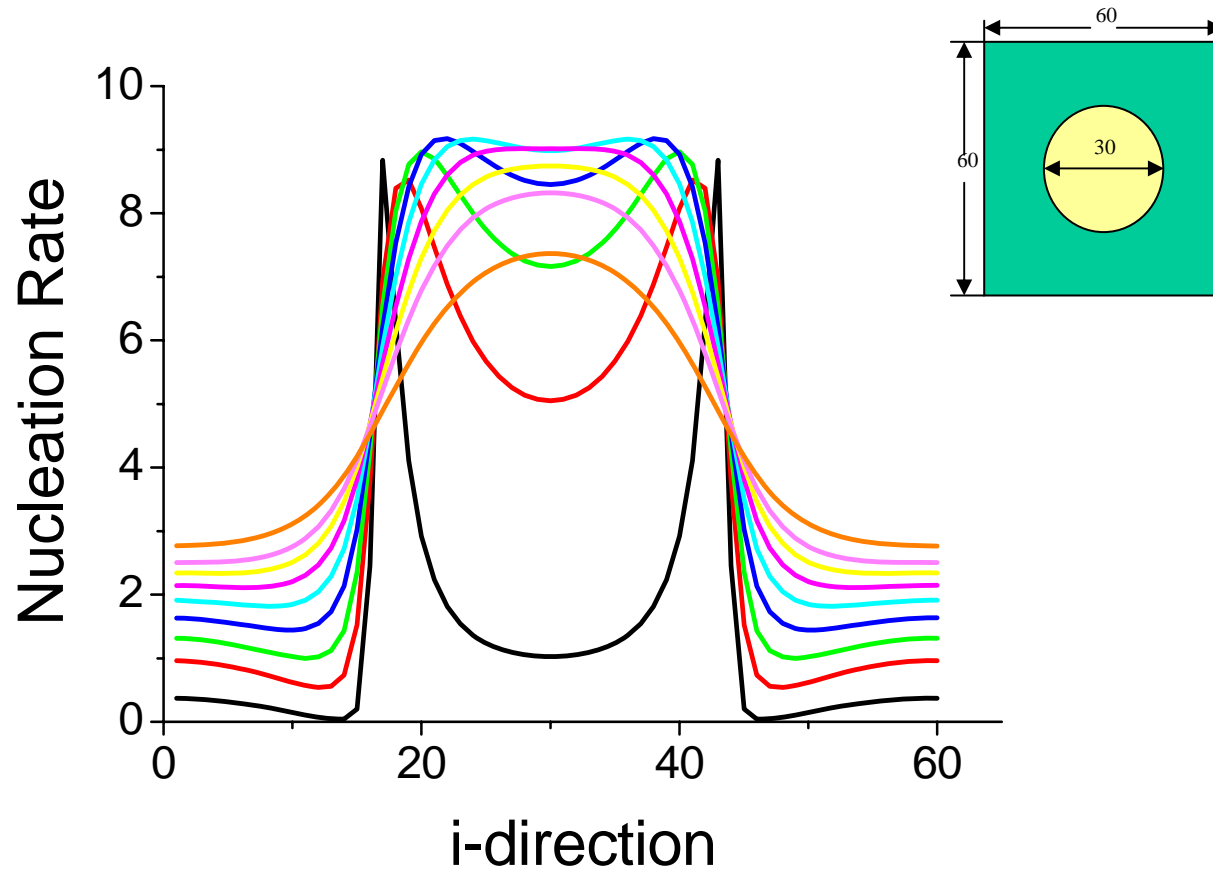
2 capping layers



4 capping layers

- We find an optimal thickness of capping layer for ordering

Nucleation rate as a function of capping layer thickness



Conclusions

- We have developed a numerically stable level-set method to model epitaxial growth
- A spatially varying potential energy surface can be exploited to obtain ordered structures.
- It is very efficient to include strain in the model, and solve the elastic equations at every numerical timestep.
- Strain leads to ordering in the submonolayer growth regime
- We model the formation and self organization of stacked quantum dots, and suggest that an optimal thickness exists.