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Lecture 3
Pattern formation on thin films

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Sapporo 2010

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Outline

- Directed self-assembly
  - A possible route to improved microelectronics
- Thin film growth with strain
  - Coupling the level set method & atomistic strain solver
  - Dependence of kinetic coefficients on strain
- Pattern formation over buried dislocation lines
- Alignment of stacked quantum dots

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Maintaining Moore's Law for Device Speed

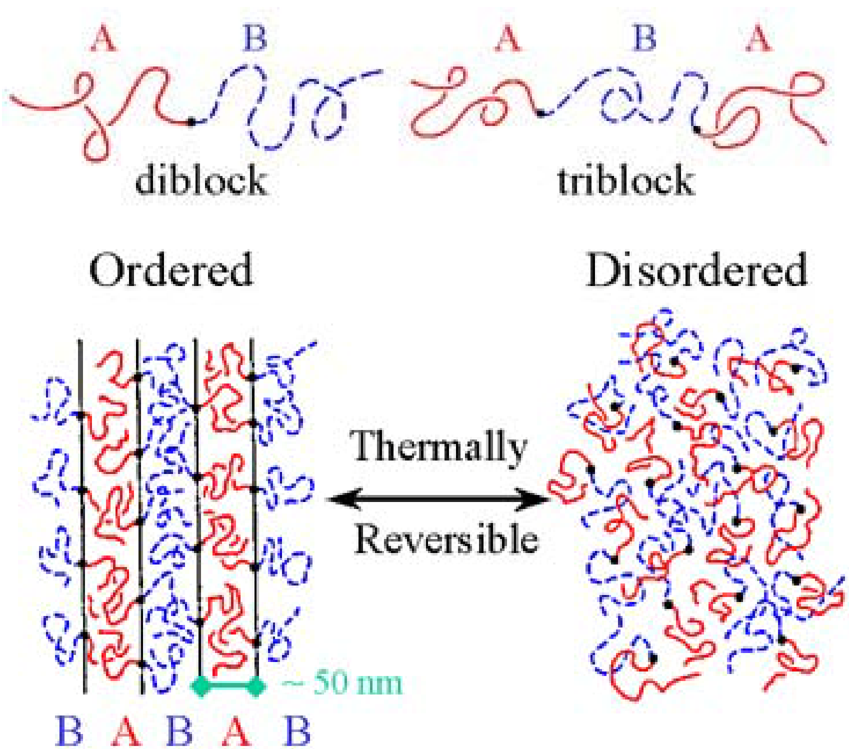
- Radically different devices will be required
- Feature sizes approaching the atomic scale
  - 50nm by 2010
  - Wavelength (visible light) = 400nm
- New device physics
  - photonics, spintronics, quantum computing
- New device structures
  - Massively parallel nanoscale structures
  - Constructed through self-assembly (bottom-up) or directed self-assembly
  - Too small for conventional lithography (top-down)
    - New approaches to lithography are emerging, e.g., using plasmons (edge waves)

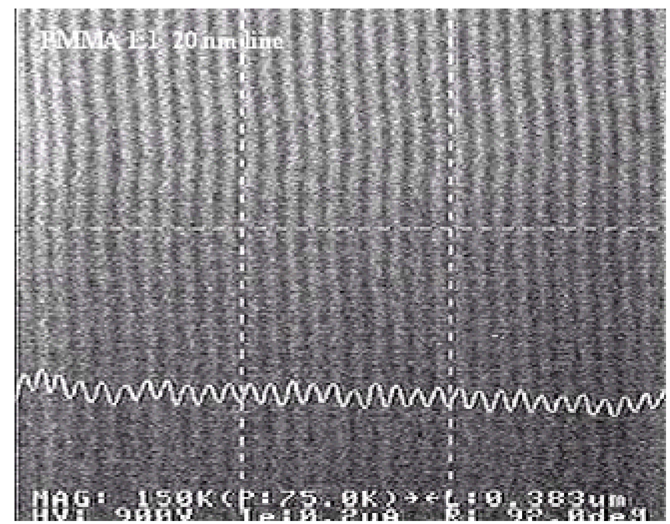
Approaches to Self-Assembly or Directed Self-Assembly

- Solid-state structures on thin films
  - Quantum wells, wires and dots
- Molecular systems
  - Self-assembled monolayers (SAMs)
- Bio/organic systems
  - E.g., DNA structures
- Block Copolymer systems

Block Copolymer Systems

- Composites of different polymeric strands
- Attraction/repulsion between strands leads to segregation and patterns
- Currently used to improve precision of lithographic patterns





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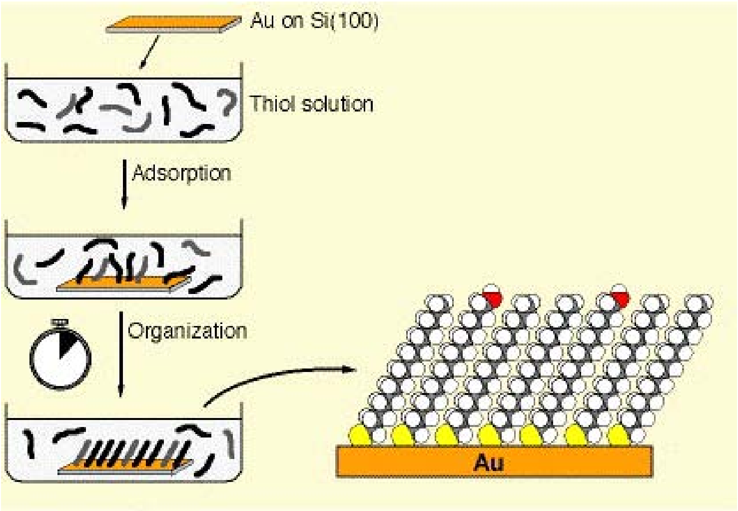
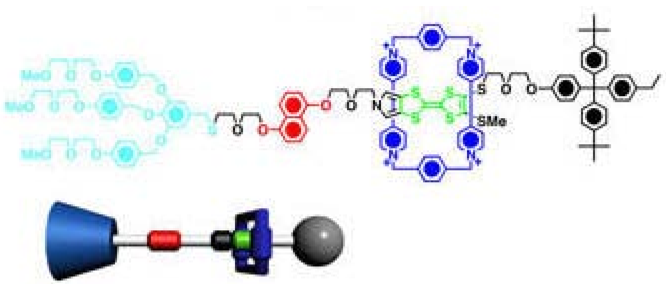
Self-Assembled Monolayers

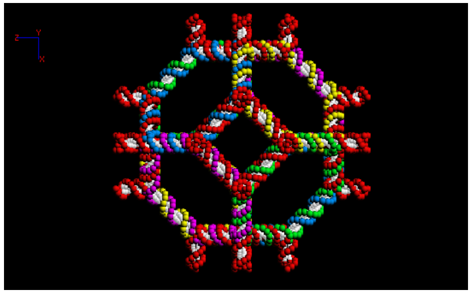
- Chemically-assembled molecular systems
- If each molecule has switching properties, the resulting system could be a massively parallel device

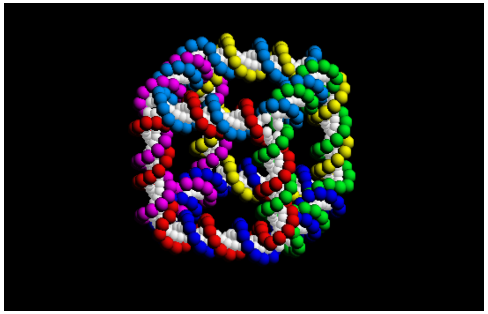


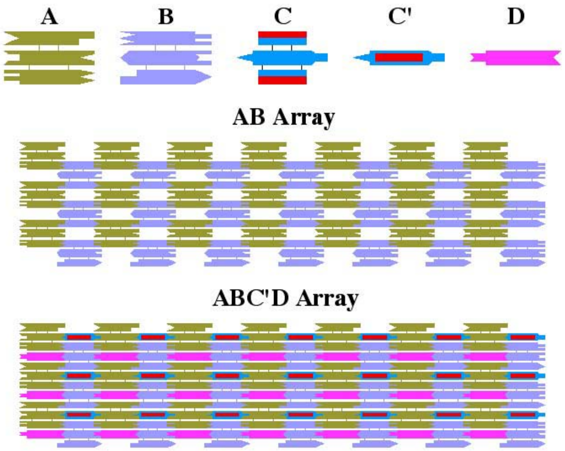


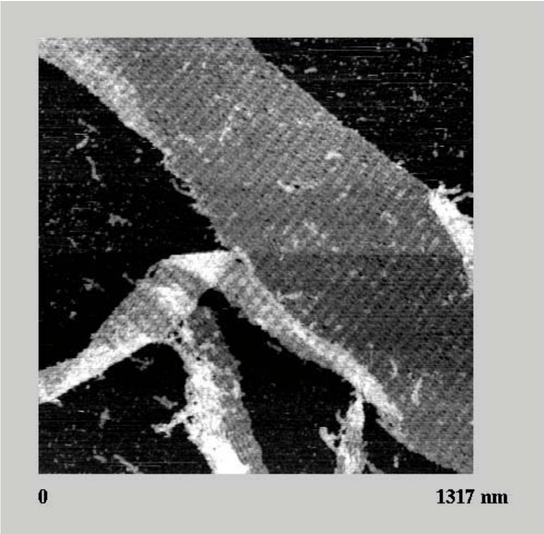
DNA Structures and Patterns

- Complex interactions of DNA strands can be used to create non-trivial structures
- The structures can be pieced together to make patterns









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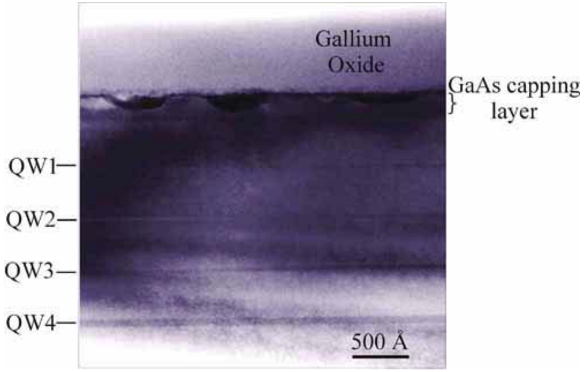
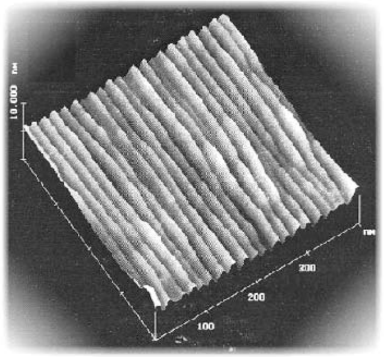
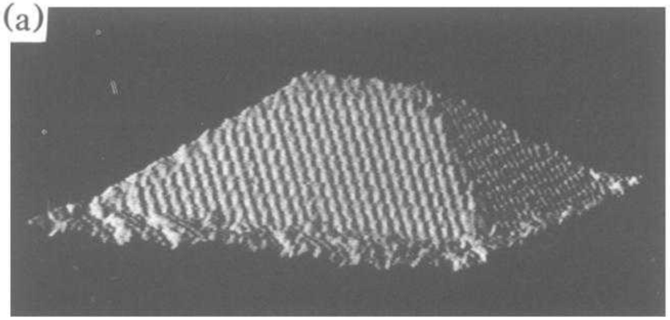
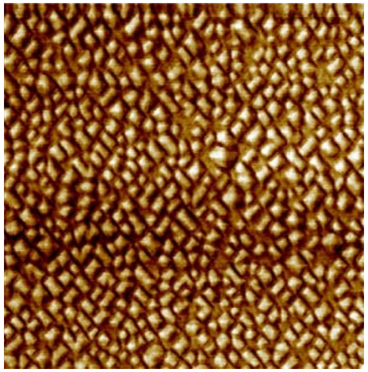
Solid-State Quantum Structures

- Quantum wells (2D)
  - "perfect" control of thickness in growth direction
  - Lasers, fast switches, semiconductor lighting
- Quantum wires (1D)
  - Various strategies for assembly
- Quantum dots (0D)
  - Self-assembled to relieve strain in systems with crystal lattice mismatch (e.g., Ge on Si)
  - Difficult to control geometry (size, spacing)





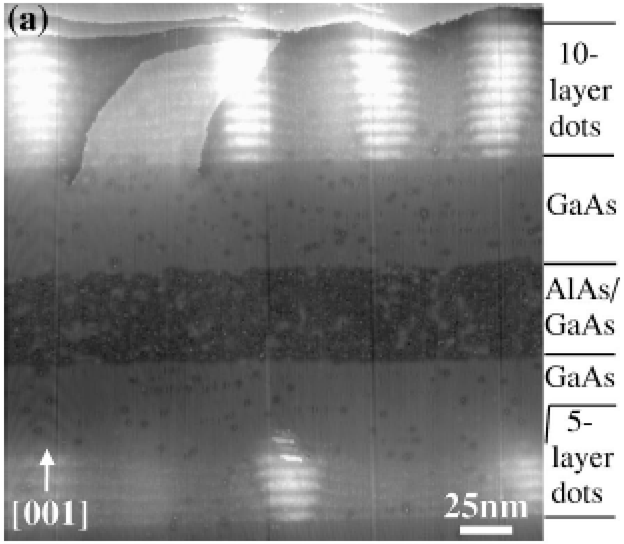




Directed Self-Assembly of Quantum Dots

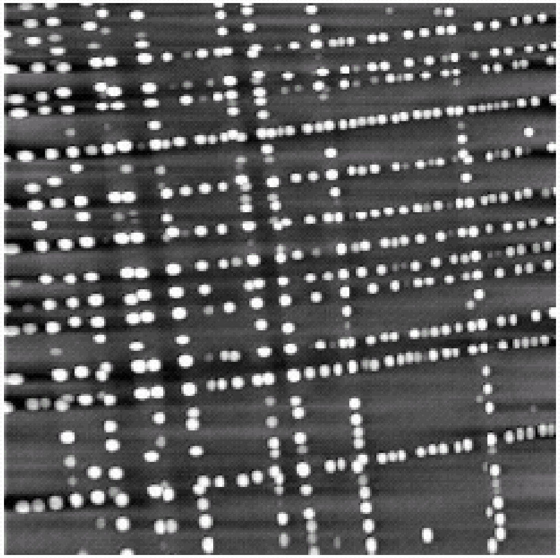
- Vertical allignment of q dots in epitaxial overgrowth (left)
- Control of q dot growth over mesh of buried dislocation lines (right)

$Al_xGa_{1-x}As$ system



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

GeSi system



H. J. Kim, Z. M. Zhao, Y. H. Xie, PRB **68**, (2003).

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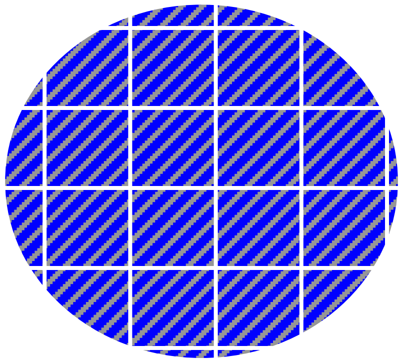
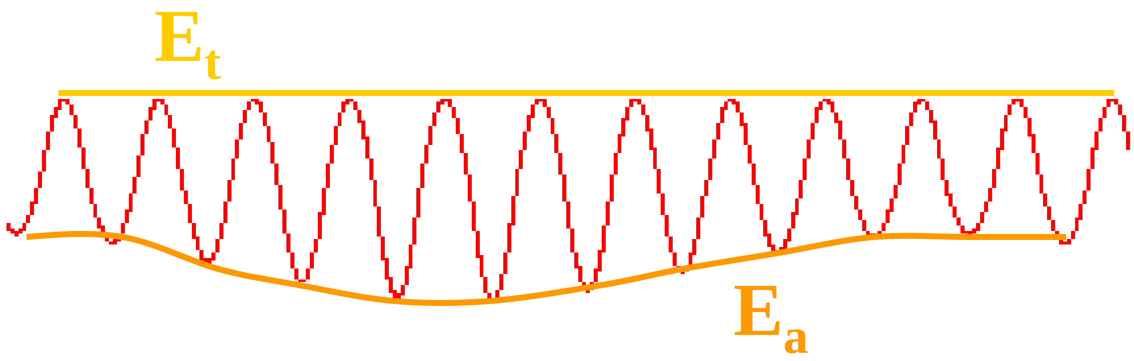
How do we combine Levelset code and strain solver?



Energetic Description of Prepatterning

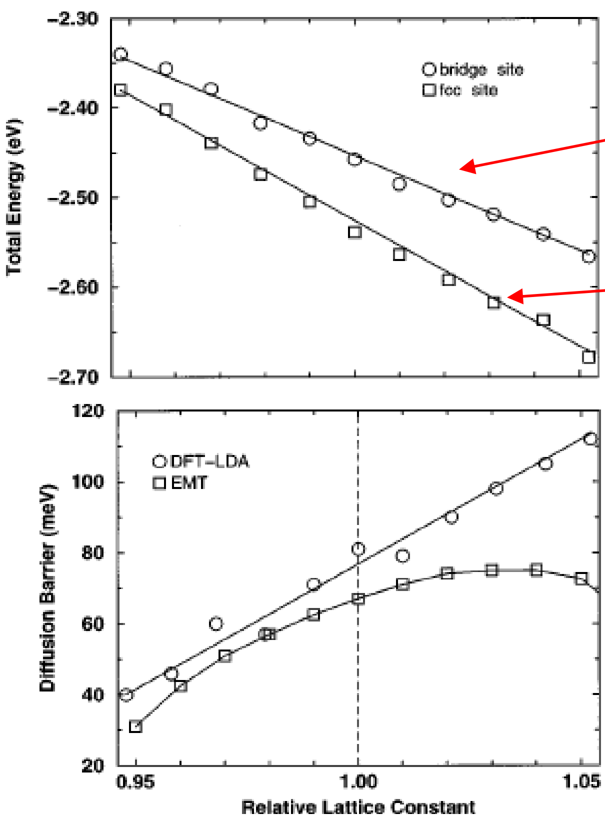
- Strain affects the energy landscape for a crystal
  - E_a = attachment energy
    = energy min above crystal atoms
  - E_t = transition energy
    = energy of barriers between energy min
- Kinetic parameters
  - Diffusion coefficient D depends on E_t - E_a
  - Variation in E_a → "thermodynamic drift velocity" v_t towards lower energy
- We propose these as the connection between strain and patterns
  - Theory of pattern formation and self-assembly is needed!



How does strain affect the parameters in our model?

D

Ag/Ag(111) (a metal)

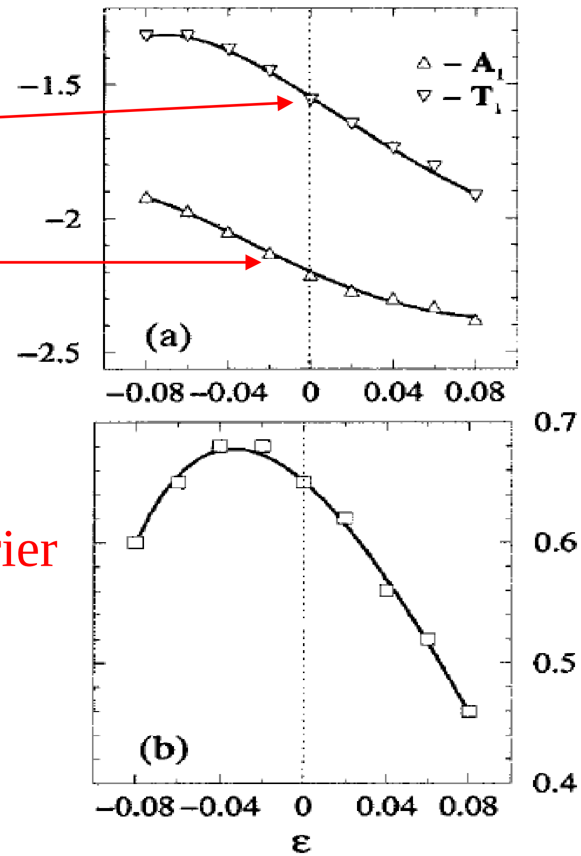


E_{trans}

E_{ad}

Energy barrier for surface diffusion

GaAs(100) (a semiconductor)



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

E. Penev, P. Kratzer, and M. Scheffler, Phys. Rev. B **64**, 085401 (2001).

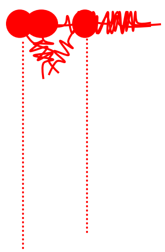
How does strain affect the parameters in our model, cont.?

D_{det}

D_{det}

- Assume that energy barrier for detachment is reduced by a strain energy:

$$E_{barrier} = E_{barrier,0} - \Delta E_{Strain} = E_{barrier,0} - K \cdot S_{tot}$$



D_{det}

$$D_{\det} = D_{\det,0} \exp\left(\frac{K \cdot S_{tot}}{k_B T}\right)$$

D_{det}

D

Diffusion Coefficient D and Thermodynamic Drift Velocity v_t for Variable E_a and E_t

- Diffusion coefficient D
  - comes from the energy barrier E_t - E_a

$$D = \exp(-(E_t - E_a)/kT)$$

- Equilibrium adatom density
  - depends on the attachment energy E_a

$$\rho_{eq} = \bar{\rho}\exp(-E_a/kT)$$

- Drift velocity v_t

$\partial_t \rho_{eq} = 0$ and $\partial_t \rho = \nabla\cdot(D\nabla\rho) - \nabla\cdot(v_t\rho)$

$$\begin{aligned} v_t &= \rho^{-1} D \nabla \rho \\ &= \bar{\rho}^{-1} e^{E_a/kT} D \nabla (\bar{\rho} e^{-E_a/kT}) \\ &= (D/kT)(-\nabla E_a) \end{aligned}$$

Modifications to the Level Set Formalism for non-constant Diffusion

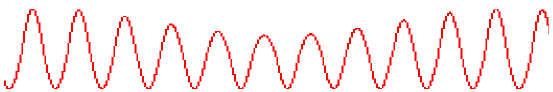
$$\mathbf{D} = \mathbf{D}(\mathbf{x}) = \begin{pmatrix} D_{xx}(\mathbf{x}) & 0 \\ 0 & D_{yy}(\mathbf{x}) \end{pmatrix}$$

Diffusion in x-direction

Diffusion in y-direction

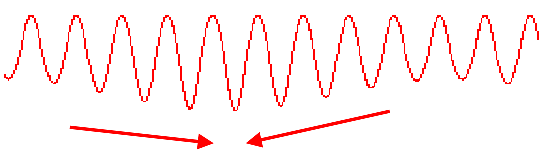
$$\frac{\partial \rho}{\partial t} = F + \nabla \cdot \mathbf{D}(\nabla \rho) - 2\frac{dN}{dt} + \nabla \cdot (\mathrm{v}_\mathrm{t}\,\rho)$$

$$v_t \sim D_{xx} \nabla_x E_{\mathrm{ad}} + D_{yy} \nabla_y E_{\mathrm{ad}}$$



no drift

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



drift

$$\frac{dN}{dt} = \left\langle \frac{D_{xx}(\mathbf{x}) + D_{yy}(\mathbf{x})}{2} \rho(\mathbf{x}, t)^2 \right\rangle$$

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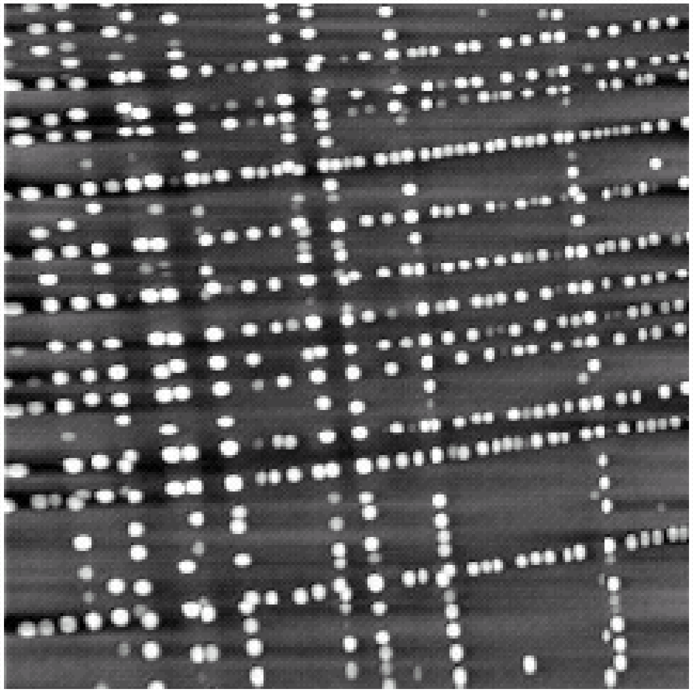
Directed Self-Assembly of Quantum Dots

Motivation: Results of Xie et al.



H. J. Kim, Z. M. Zhao, Y. H. Xie, PRB **68**, (2003).

Nucleation occurs in regions of fast diffusion

Level Set formalism is ideally suited to incorporate anisotropic, spatially varying diffusion without extra computational cost

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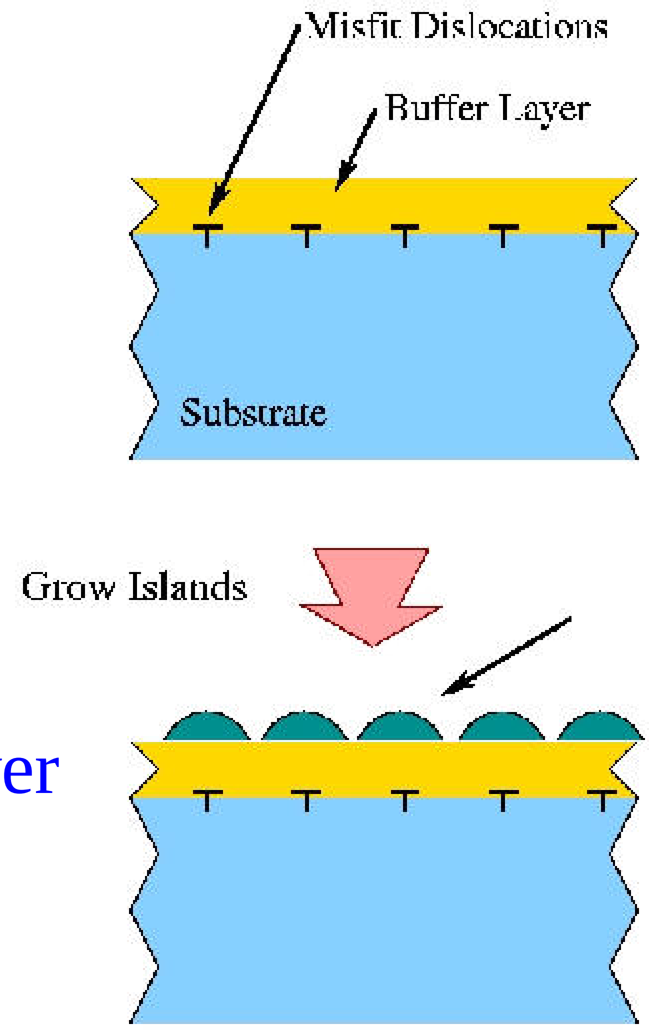
Creation of Dislocation Network

- Layered system
  - Substrate Si (001)
  - 800Å $Si_{.85}Ge_{.15}$ buffer layer
  - 100Å Si capping layer
  - Anneal to relax buffer layer
- Dislocation network
  - substrate/buffer interface
  - Mixed edge/screw type
- Q dots grow on top of 900Å layer
  - Ge or SiGe
  - Along slip plane from buried dislocations



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Q Dots and Dislocation Network

- TEM
  - Q dots on surface
  - Buried dislocation lines
  - --- is location of slip plane at surface
  - → are Burgers vectors

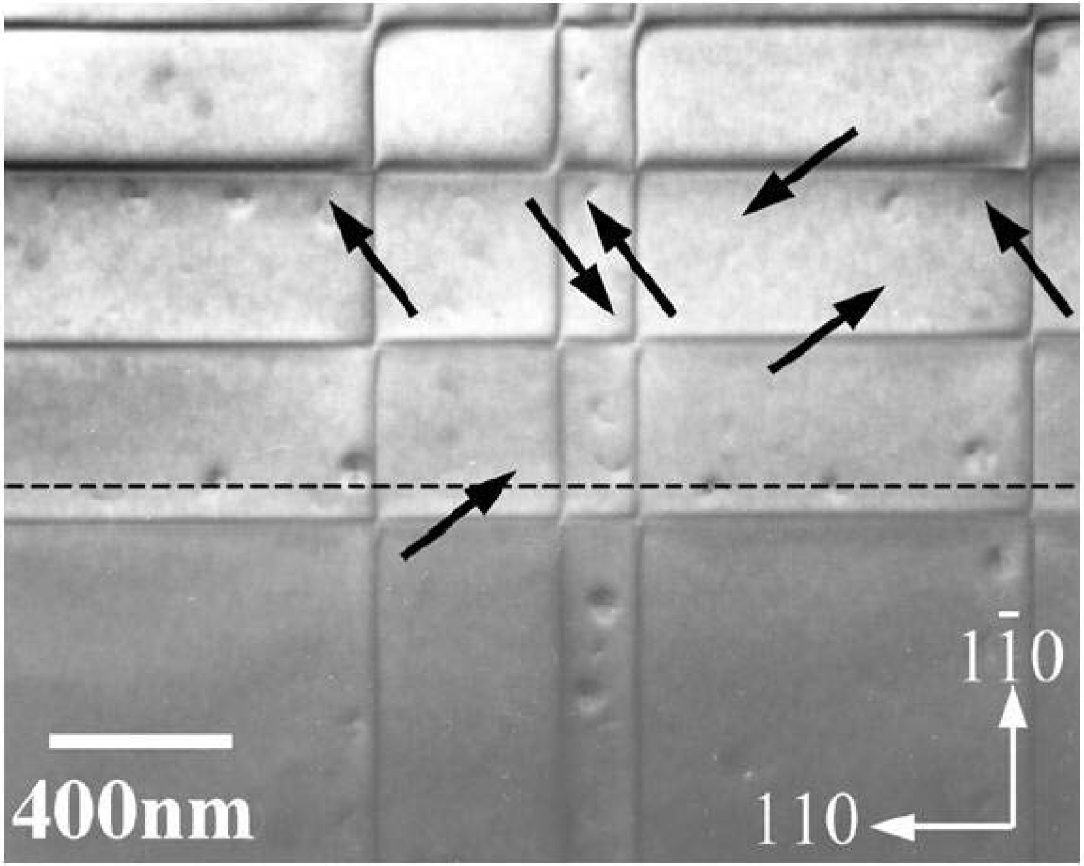

Fig. 2. Plan-view TEM micrograph of the sample with Ge SAQDs grown on the relaxed $Si_{0.85}Ge_{0.15}$ buffer layer.

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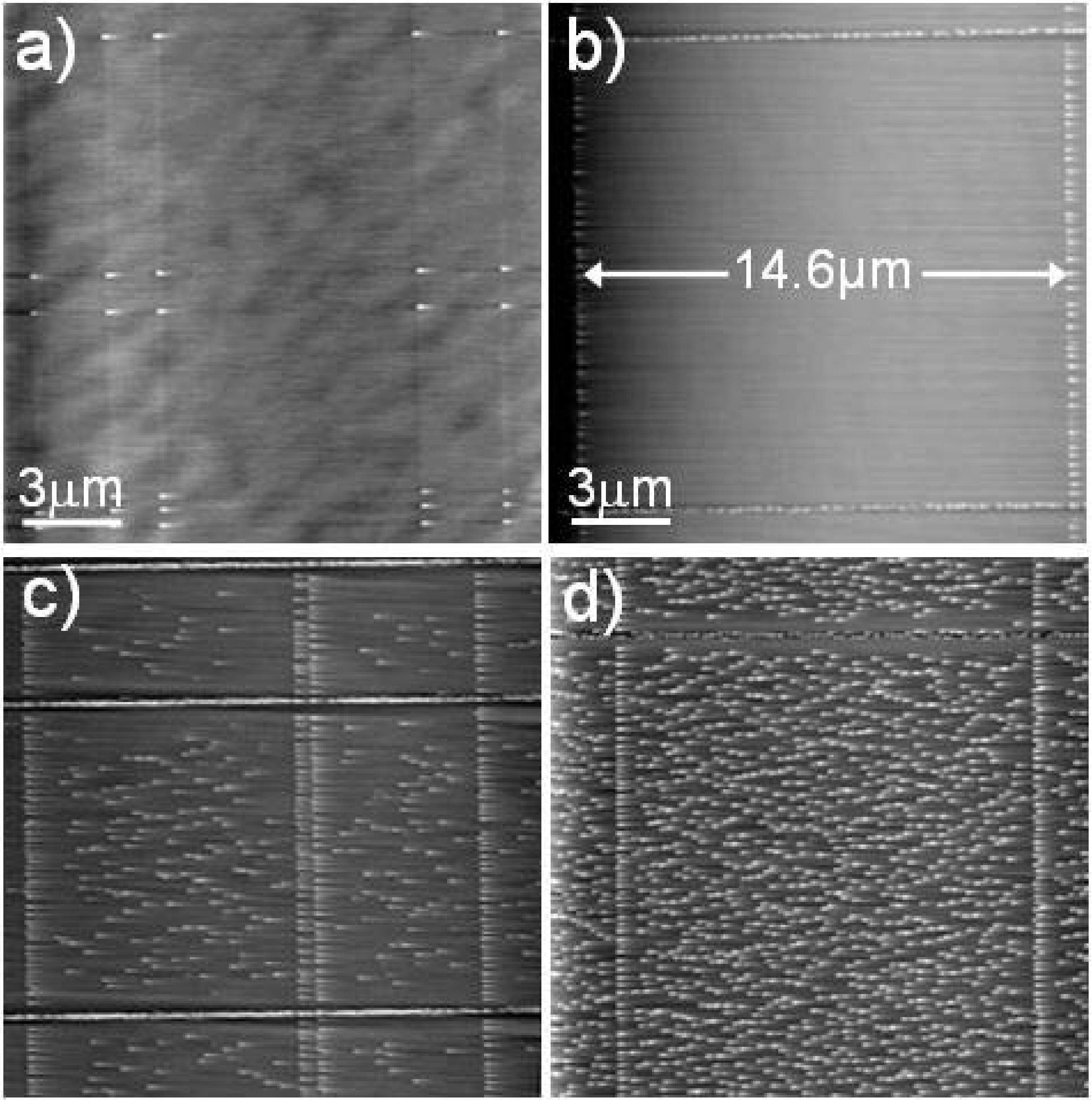
Growth over Buried Dislocation Lines

Ge coverage

- 4.0 Å
- 4.5 Å
- 5.0 Å

(d) 6.0 Å



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Model for Growth

- Prescribe variation in E_a, E_t
  - Variable D and v_t
- Perform growth using LS method
  - Nucleation occurs for larger values $D\rho^2$
- Pattern formation in islands positions
  - Seeds positions for quantum dots
  - Niu, Vardavas, REC & Ratsch PRB (2006)

$$\frac{\partial \rho}{\partial t} = F + \nabla \cdot \mathbf{D}(\nabla \rho) - 2\frac{dN}{dt} + \nabla \cdot (\mathrm{v}_{\mathrm{t}} \rho)$$

$$v_t \sim D_{xx} \nabla_x E_{\mathrm{ad}} + D_{yy} \nabla_y E_{\mathrm{ad}}$$

$$\frac{dN}{dt} = \left\langle \frac{D_{xx}(\mathbf{x}) + D_{yy}(\mathbf{x})}{2} \rho(\mathbf{x}, t)^2 \right\rangle$$

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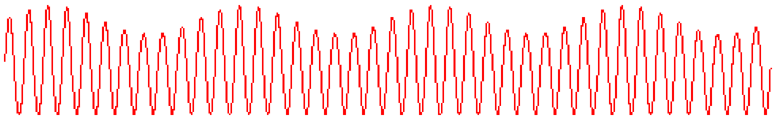
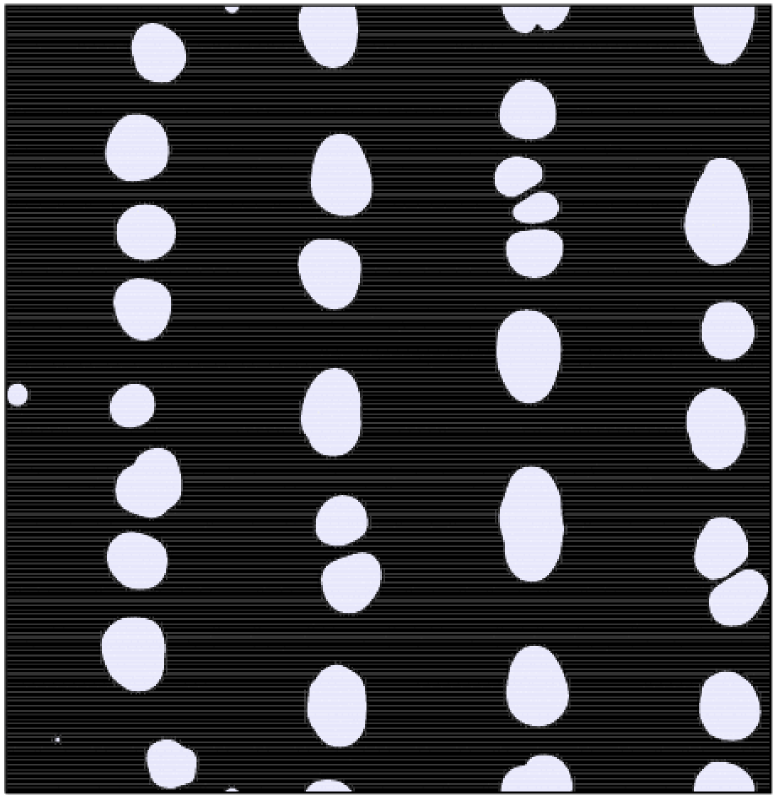
First part: assume isotropic, spatially varying diffusion



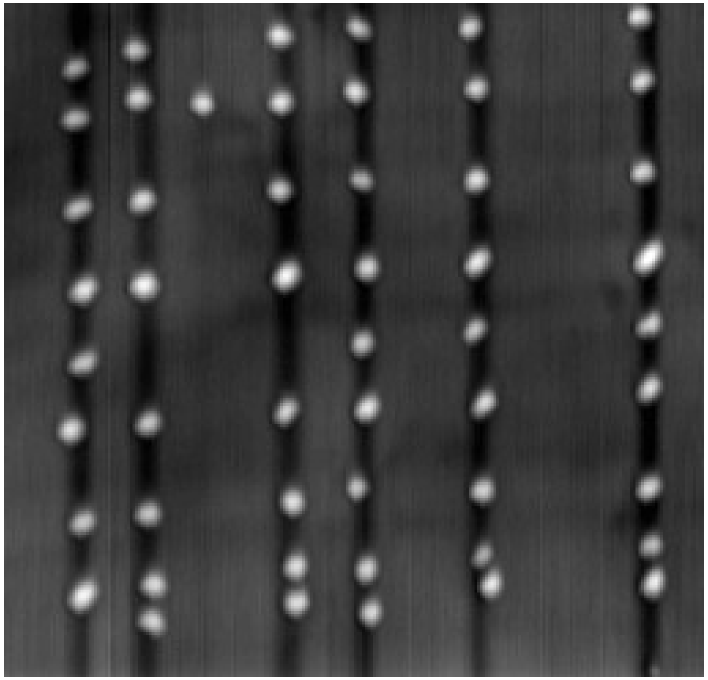
$$D_{xx} = D_{yy} \sim \sin(ax)$$



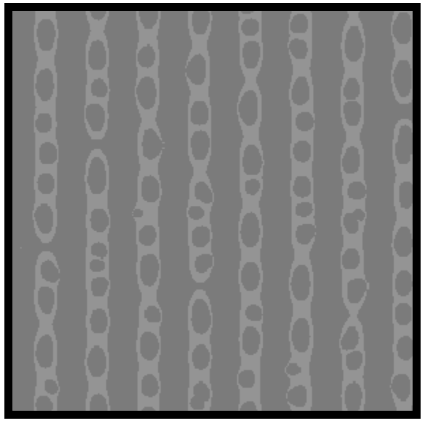
fast diffusion

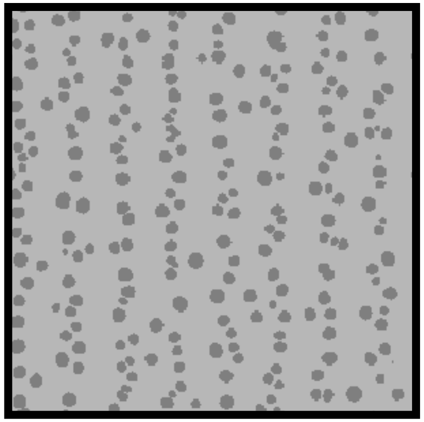
slow diffusion

Experiment by Xie et al., UCLA



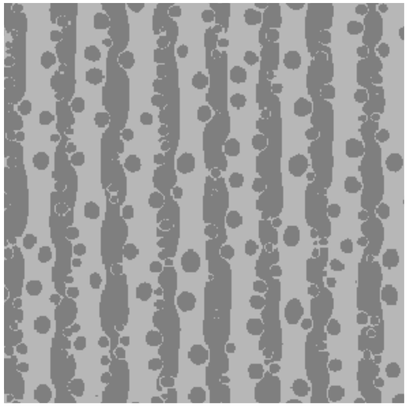
Variation of adsorption or transition energy

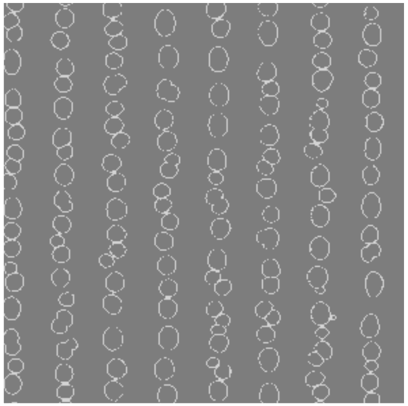




Nucleation rate ~ $D\rho(\mathbf{x},t)^2$

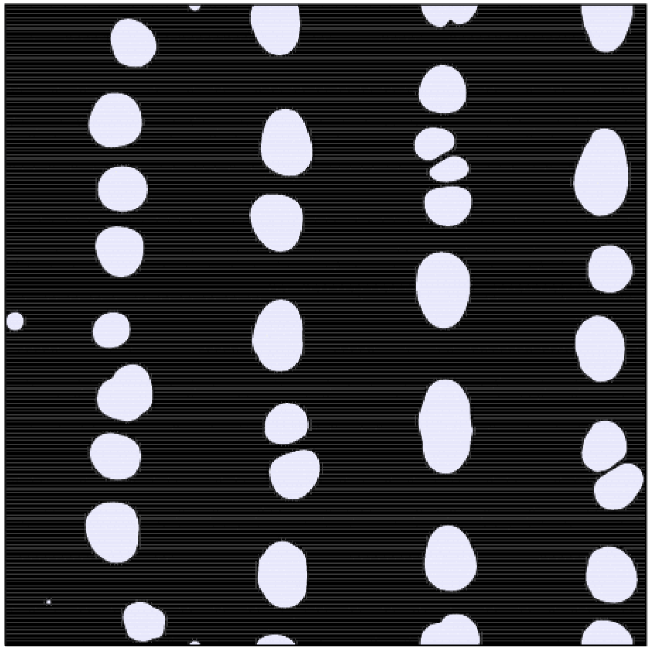
Variation of both, adsorption and transition energy



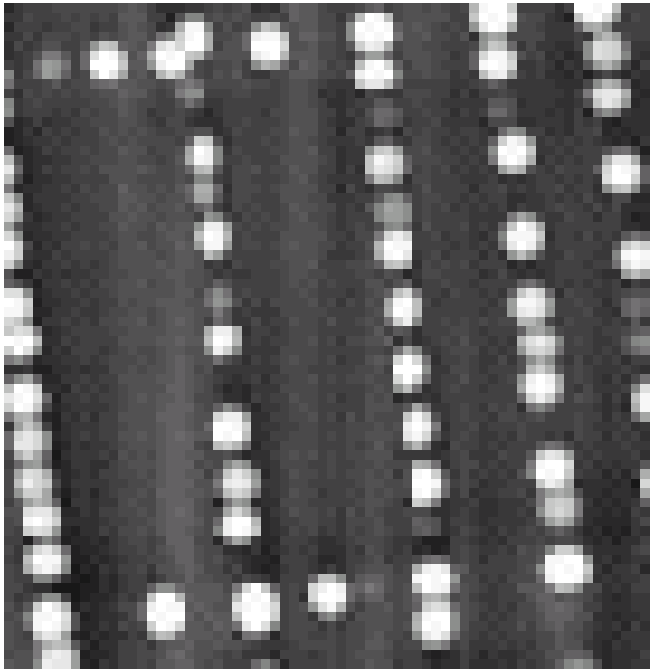


Comparison with Experimental Results

Simulations



Results of Xie et al.



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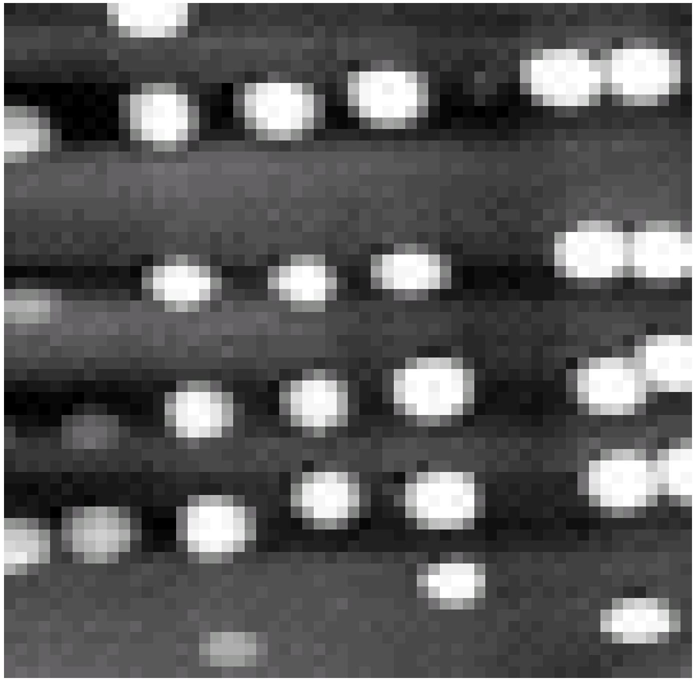
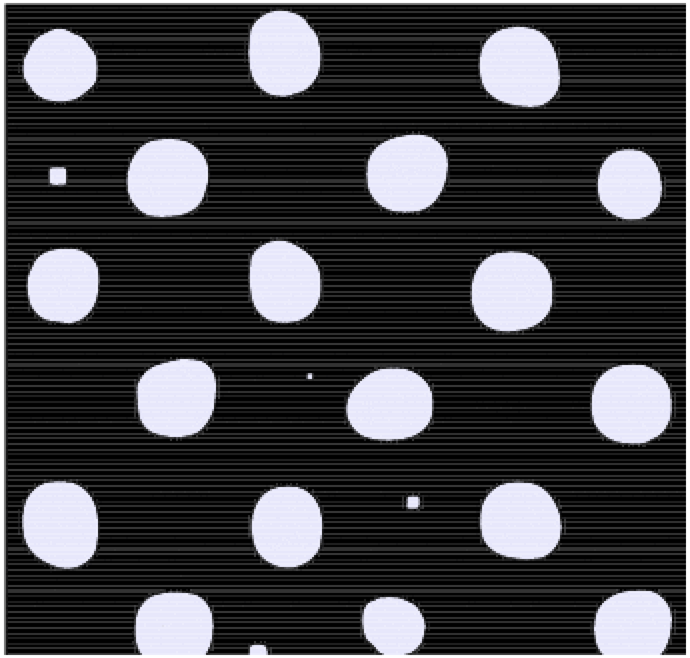


Simulations



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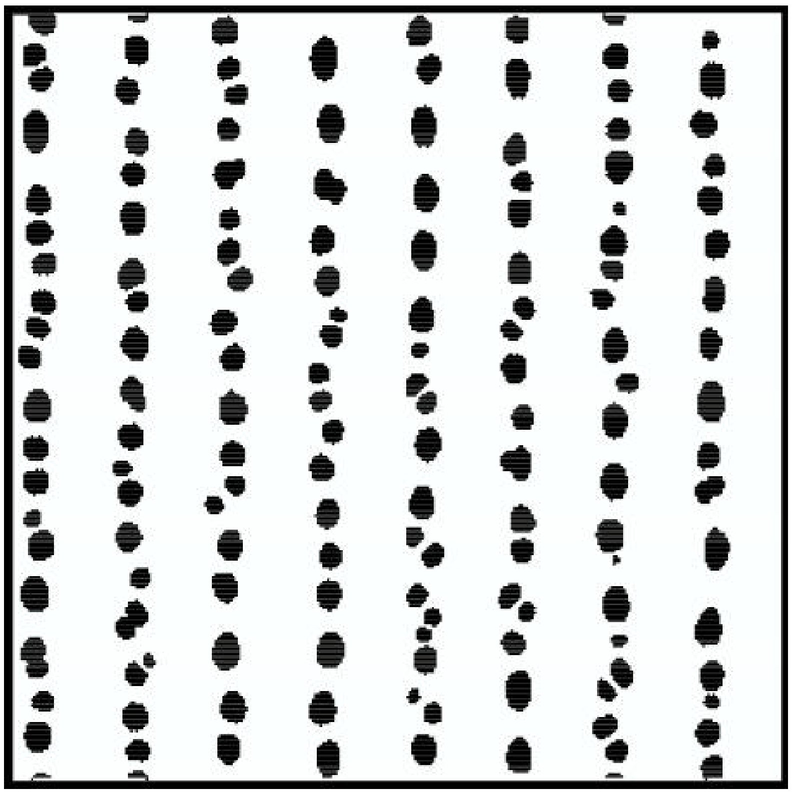
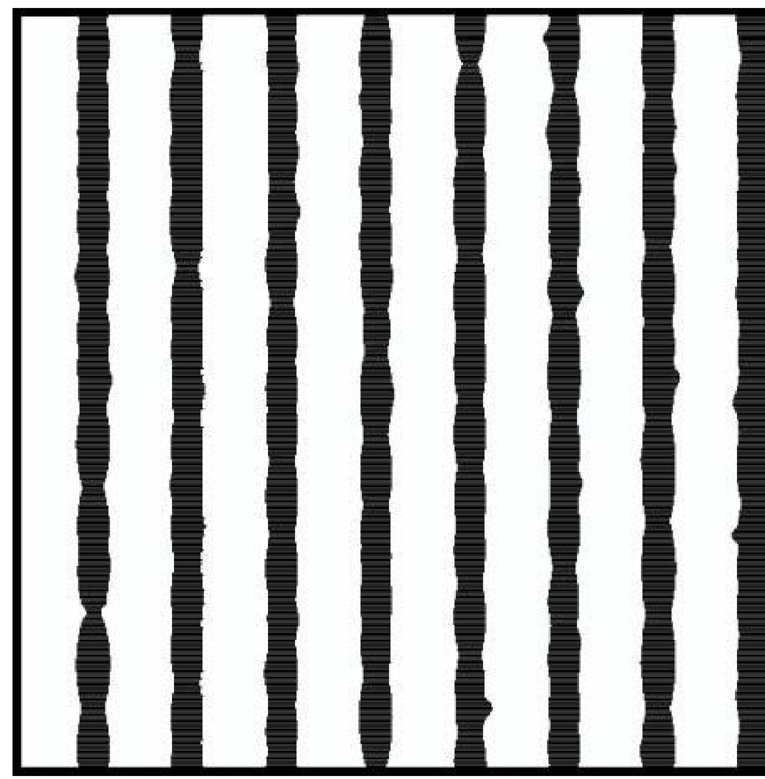
From islands to wires

For islands that are well aligned, due to prepatterning, further growth can lead to monolayer wires





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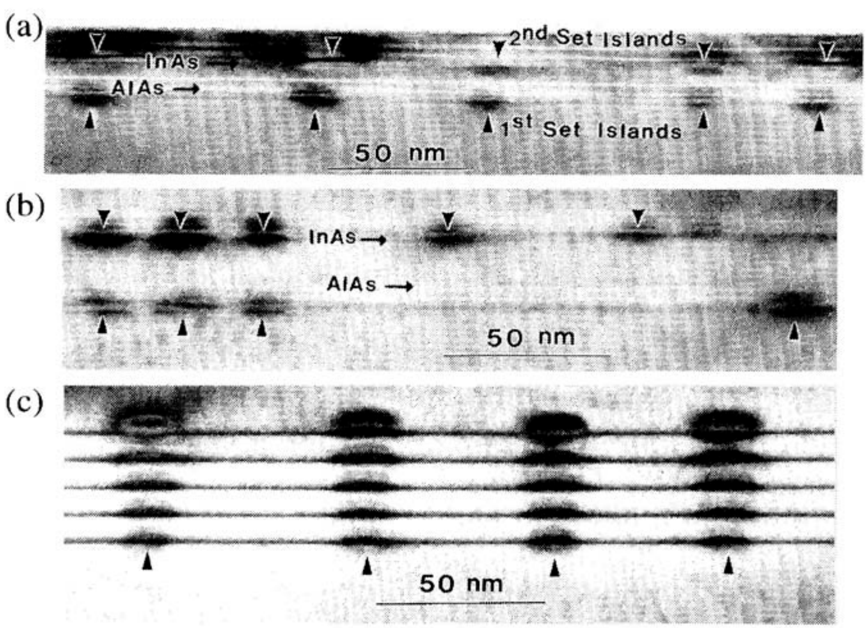
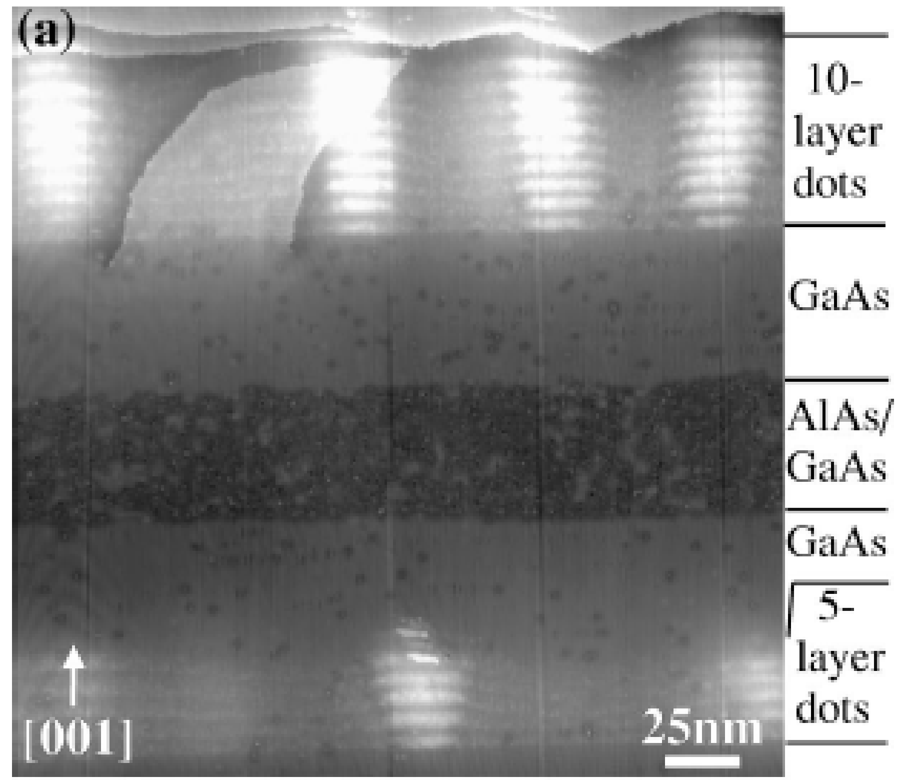
Vertically Aligned Quantum Dots



Q. Xie, et al. (Madhukar group), PRL **75**, (1995)



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

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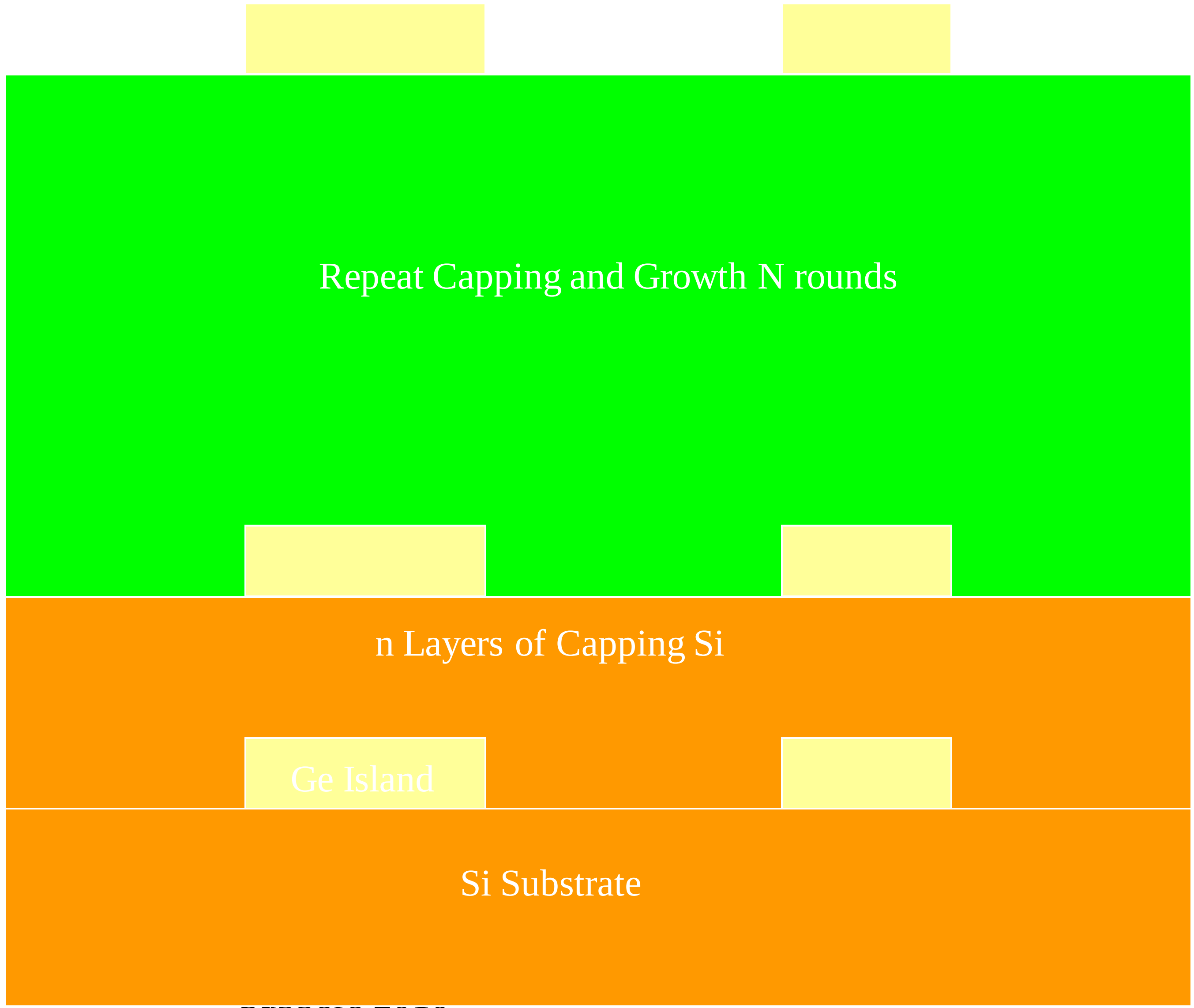
Simulation of stacked quantum dots


Repeat Capping and Growth N rounds
n Layers of Capping Si
Ge Island
Si Substrate

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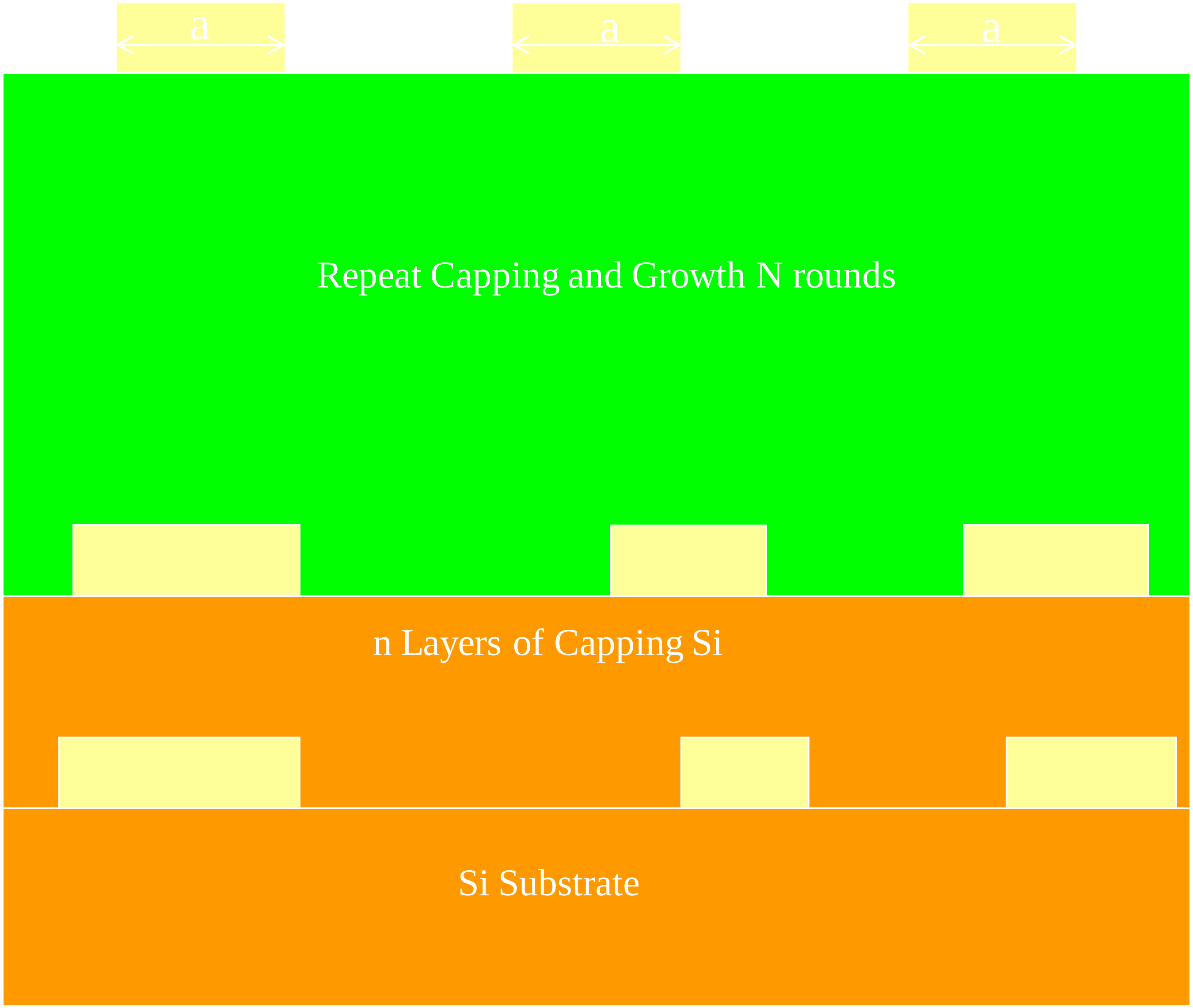
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Repeat Capping and Growth N rounds
n Layers of Capping Si
Si Substrate

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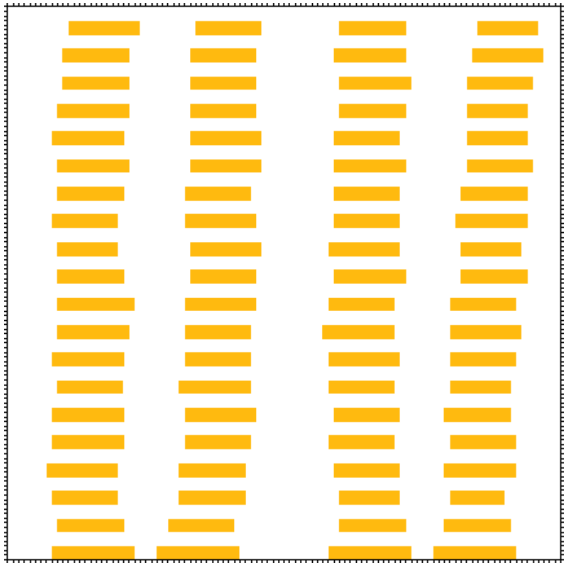
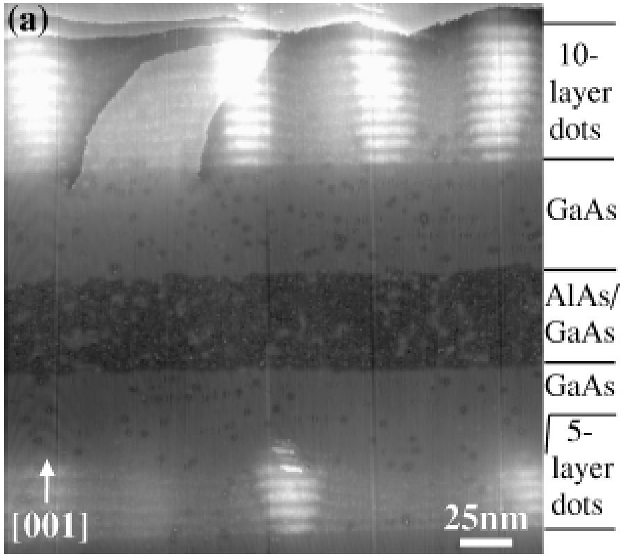
Ordering of stacked quantum dots

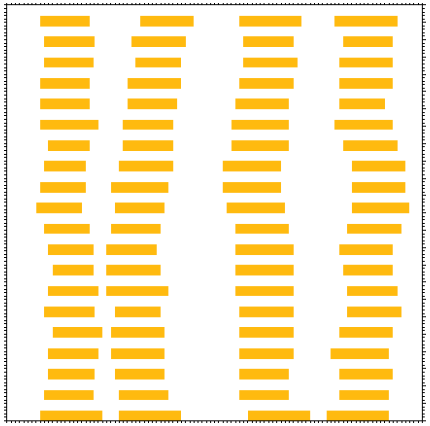


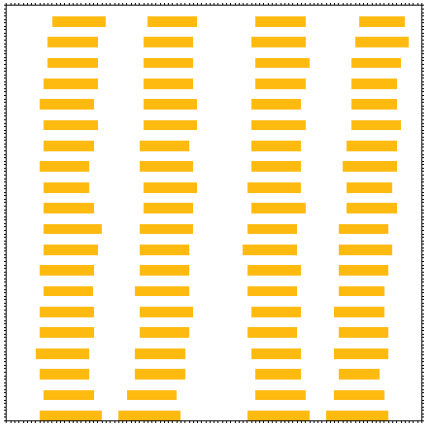
$Al_xGa_{1-x}As$ system

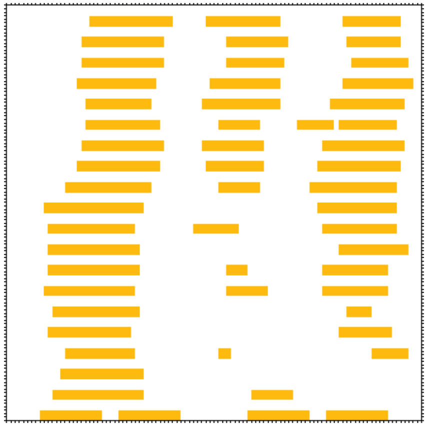


B. Lita et al., APL **74**, (1999)

Thickness dependence of vertical ordering







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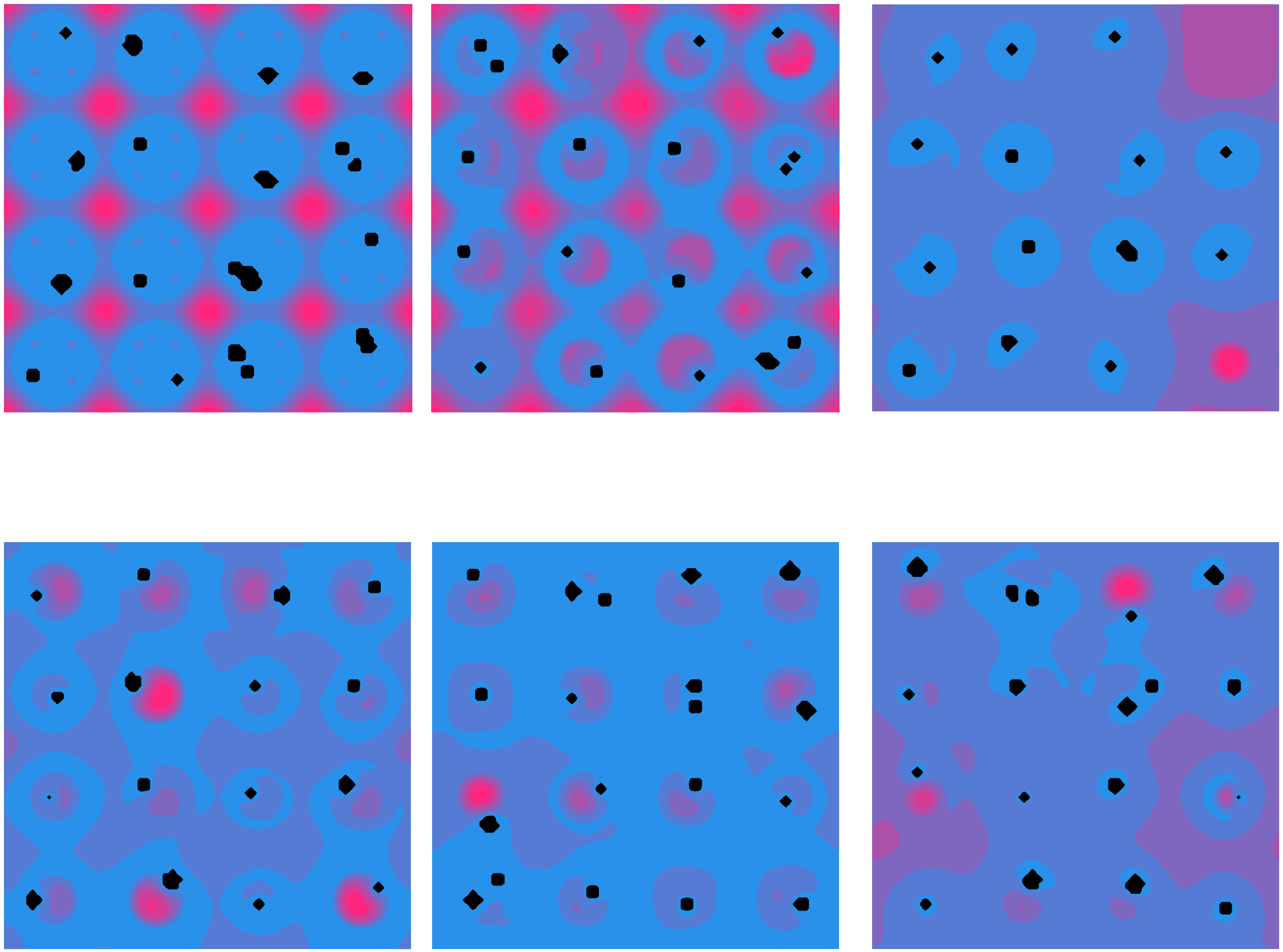
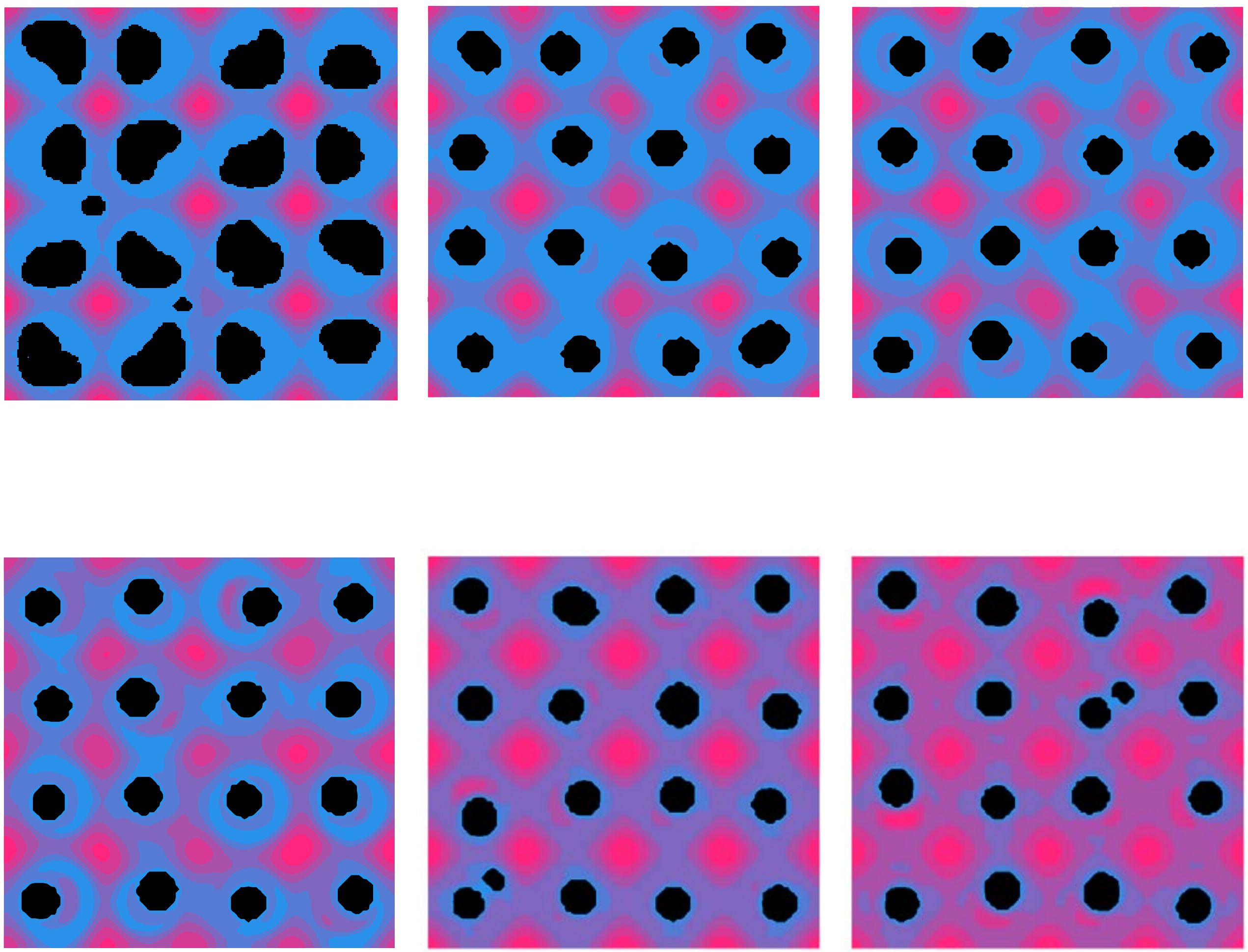


- Thin
  - nucleation at bdry
- Moderate
  - nucleation at center
- Thick
  - random nucleation



- Thin
    - misshaped islands
- Moderate
    - circular islands
    - regularly placed
- Thick
    - displaced islands

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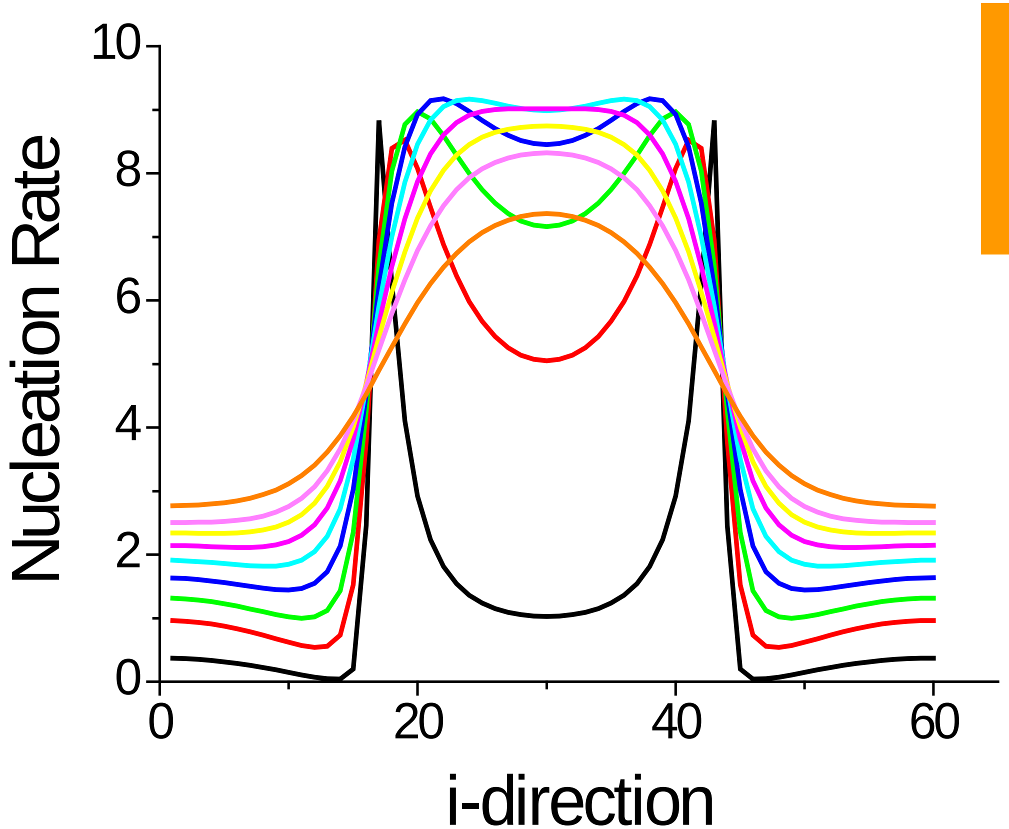
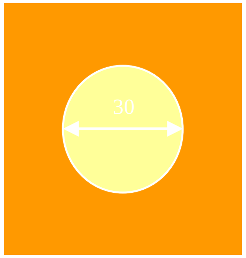
Nucleation rate as a function of capping layer thickness





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Conclusions

- Island dynamics/level set method
  - Combined to simulate strained growth
  - Kinetic parameters assumed to have strain dependence
- Directed Self-Assembly
  - Growth over a network of dislocation lines
  - Alignment of stacked quantum dots
- Unsolved problems
  - Growth mode selection (e.g., formation of wetting layer)
  - Pattern design and control (e.g., quantum dot arrays)
  - Optimizing material (and device) properties