EFFICIENT COMPUTATIONAL METHODS FOR HETEROEPITAXIAL GROWTH

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Joint work with
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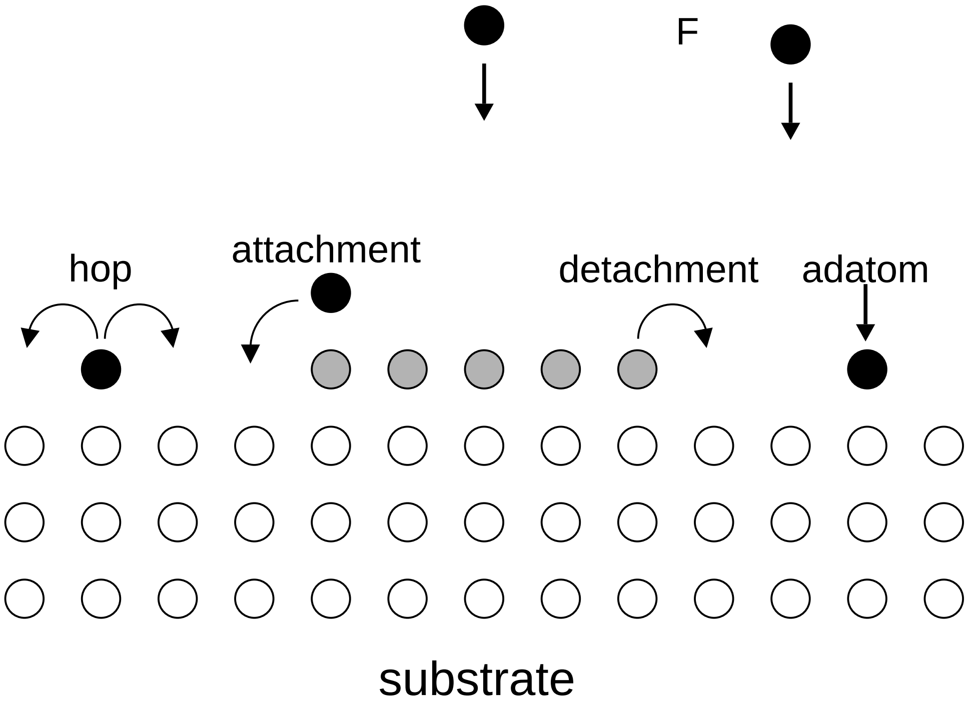
WHAT IS EPITAXIAL GROWTH?

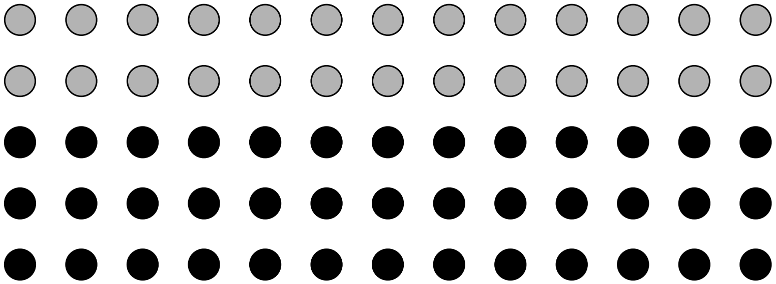


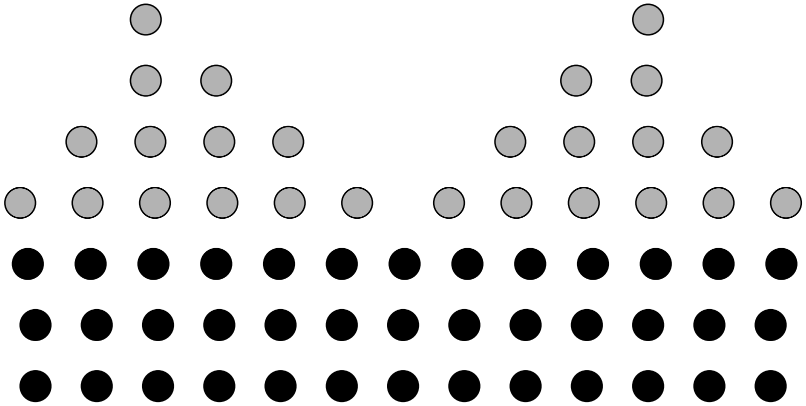
A schematic of the basic processes occurring in epitaxial growth as viewed in cross-section. The gray atoms are atoms that are part of an island and the black atoms are adatoms.

HETEROEPITAXY

- Heteroepitaxial growth occurs when one atomic species grows on a substrate of another material
- One example is when Germanium is grown on a substrate of Silicon
- One notable feature of such a system is that the lattice spacing of the deposited material is different from the substrate. This difference is called the misfit.
- The mismatch can be tuned by using alloys. For example by depositing a Si-Ge alloy the mismatch can be varied from 0 to 4 %.
- This gives rise to elastic effects which can destabilize a flat interface resulting in the formation of mounds.

ENERGETICS



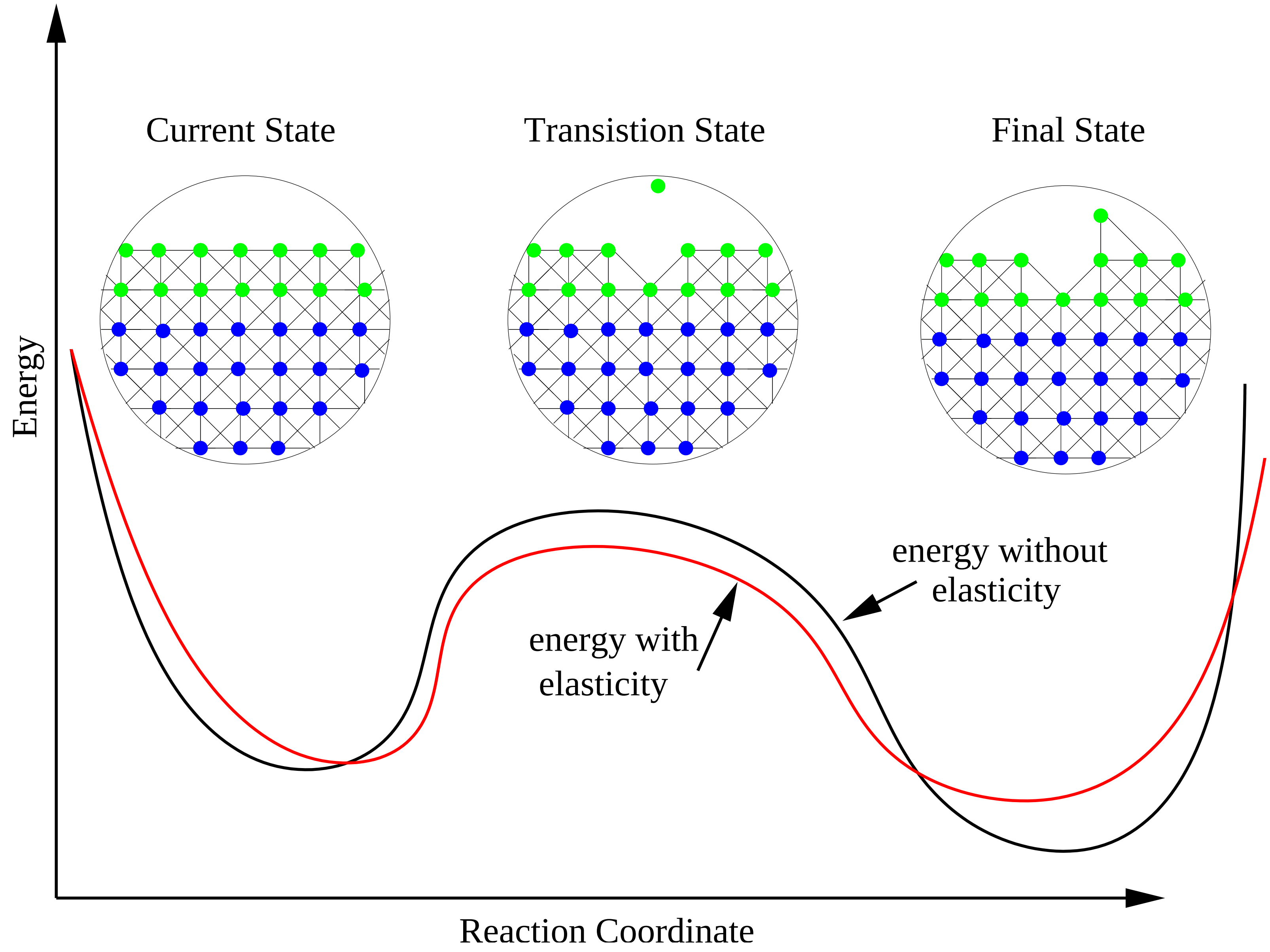


Germanium on Silicon– Due to elastic interaction
the bottom configuration can have less energy than the top one.

KINETIC MONTE CARLO (KMC)

- KMC is based on knowing or assuming the important events and estimating their rates
- Rates are based on transition state theory which gives $R = \omega \exp(-\Delta E/kT)$
- $\Delta E = E(\text{transition state}) - E(\text{current state})$
- ω is the attempt frequency, kT is the thermal energy

ENERGY DIAGRAM



TRANSITION RATE APPROXIMATION

- Transition Rate
  $= \omega \exp\left[(\text{Current Energy} - \text{Transition Energy})/kT\right]$
- Transition Rate
  $\approx \omega \exp\left[(\text{Current Energy} - \text{Energy with Atom Removed})/kT\right]$
- ω is the attempt frequency, kT is the thermal energy

HETEROEPITAXIAL MODEL– Lam, Lee, and Sander (2002)

- We use a solid-on-solid type model with a cubic lattice.
- We consider nearest and next nearest neighbor bonds.
- Elastic effects are modeled using a linear ball and spring model with springs connecting nearest and next nearest neighbor atoms.
- $W \equiv$ total elastic energy
- Suppose we wish compute hopping rate of atom p then we will need to compute:
  $\Delta W = W(\text{with atom p}) - W(\text{without atom p})$
- In most cases $\Delta W > 0$, removing an atom lowers the elastic energy

MODEL– continuued

- Hopping Rate is $R = \omega \exp\left[(-n\gamma + \Delta W + E_0)/kT\right]$
- where $n =$ number of bonds, $\gamma =$ bond energy, $\omega = 6D_0/((I+1)(2I+1))$, D_0 and E_0 are chosen to match experimental adatom diffusion
- Hop $\equiv$ movement to site $\pm i$ atoms away i chosen at random from $\{1, 2, 3, ..., I\}$ (uniformly)
- Effective Deposition rate $f = F(I+1)(2I+1)/6$ where F is actual deposition rate (atoms/sec)
- Evolution is relatively insensitive to I but larger values I improve simulation speed.

CLOSELY RELATED PREVIOUS WORK

- Orr, Kessler, Snyder, and Sander (1992) – KMC 1+1 dimensions
- Ratsch and Zangwill (1993) – KMC 1+1 dimensions
- Ratsch, Smilauer, Vvedensky, and Zangwill (1995) – KMC 2+1 dimensions
- Much,Ahr,Biehl and Kinzel (2002) – off lattice KMC 1+1 dimensions

OTHER APPROACHES

- Full Continuum (Phase Field, Sharp Interface)
- Semi-Discrete (BCF, Level Set Methods)

REJECTION-FREE KINETIC MONTE CARLO

- Make a list of rates, R_k, for all atoms
- $W = \sum_{k=1}^{N} R_k$ is the total transition rate
- $P_k = R_k/W$ is the hopping probablity
- Use $\{P_k\}_{k=1}^{N}$ as probability distribution from which pick an atom which is then allowed to hop
- Update $\{P_k\}_{k=1}^{N}$
- Repeat

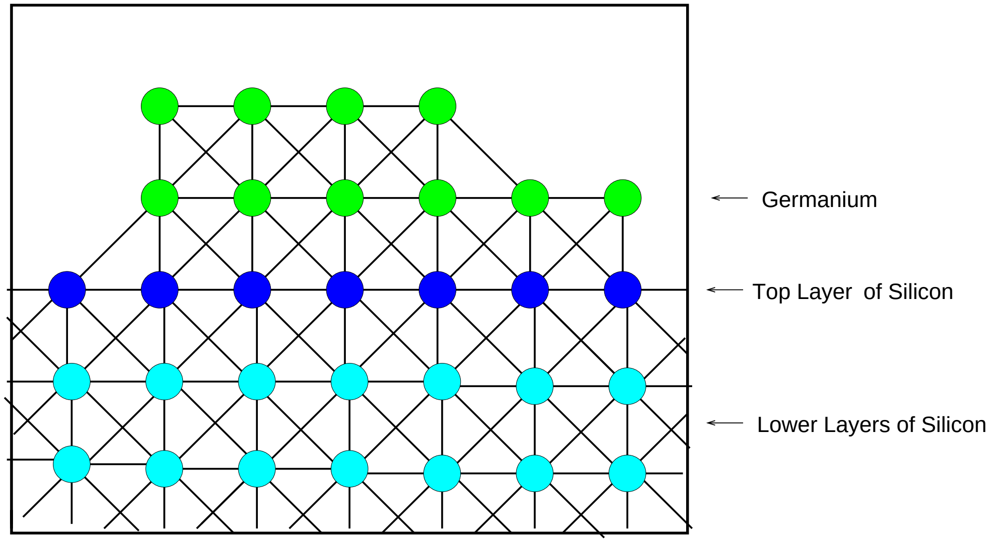
COMPUTATIONAL BOTTLE NECK

- In principle we need to compute R_k for all atoms
- This entails removing each and every atom, computing the resulting elastic field, and resulting change in energy
- Elastic interactions can be long ranged
- Consquently computations of heteroepitaxy are very challenging

WIDENING THE BOTTLE NECK

- Efficient algorithm for the elastic field based on a Multigrid-Fourier Method
- Ability to provide local updates to the elastic field when possible: Principle of energy localization & the expanding box method
- A reasonably sharp upper bound on R_k without needing an update on the elastic field allows us to formulate a rejection-reduced KMC method

DISCRETE ELASTICITY FORMULATION



Straight lines indicate springs

Since the interaction matrix for the substrate atoms is time independent the displacements of the light blue atoms can be specified in terms of the dark blue atoms in an efficient manner

Consequently the unknowns are the Germanium atoms and the top row of Silicon (the green and dark blue atoms)

DISPLACEMENT FIELD

u_s - displacement of the top row of Silicon atoms from their equilibrium position.

u_g - displacement of Germanium from "compressed" configuration.

b - the force that arises from the compressed configuration.

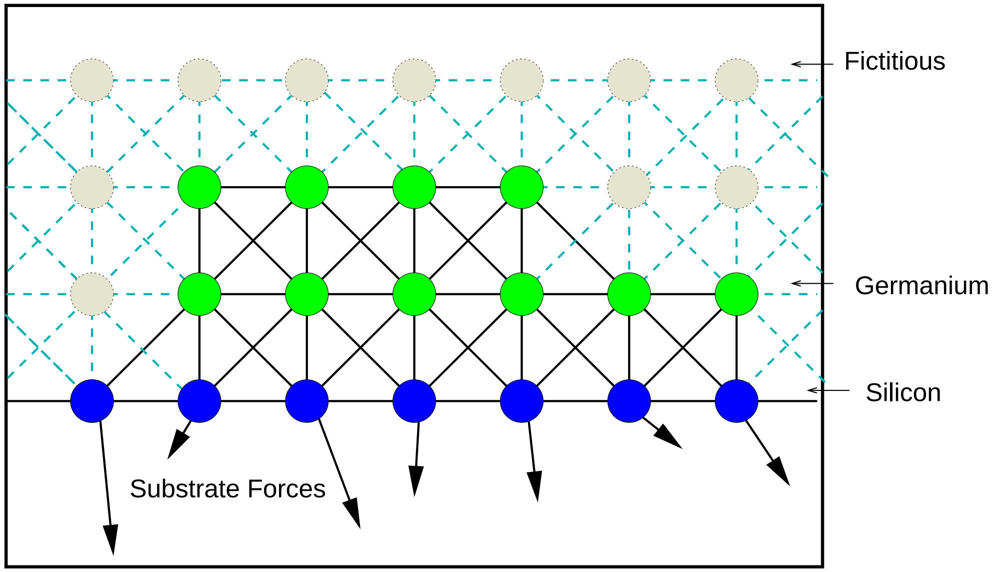
The Silicon substrate is periodic in the horizontal directions and semi-infinite in the vertical direction.

The displacements satisfy:

$$\begin{pmatrix} A & C^T \\ C & B \end{pmatrix} \begin{pmatrix} u_g \\ u_s \end{pmatrix} = \mathbf{b}$$

A and C are sparse matrices whereas B is dense matrix Bu_s is similar to a Dirichlet to Neumann map and it can be efficiently evaluated using FFTs.

MULTGRID-FOURIER METHOD



All atoms are located on a rectangular grid

Lines are real springs & dotted lines are vanishingly weak springs

All but the top row of substrate are removed
and replaced by equivalent forces

These forces are computed using the Fourier Algorithm

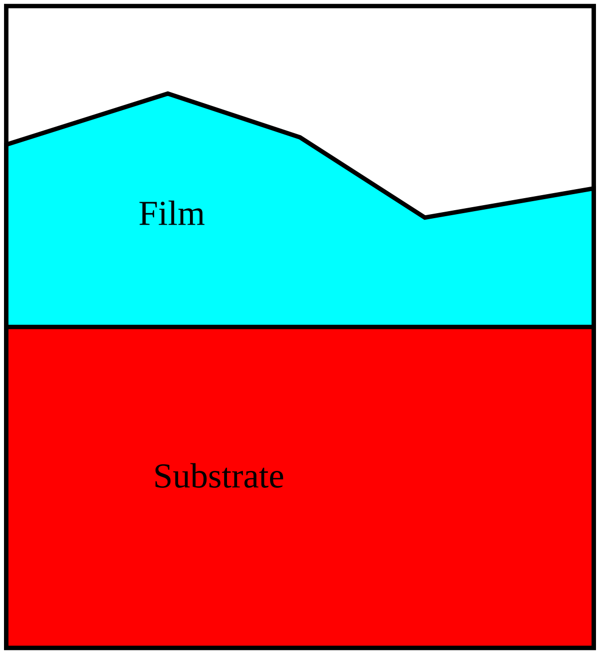
MULTGRID-FOURIER METHOD

- We use standard V-cycles
- Stopping criteria is: L^2 norm of relative residual $< 10^{-2}$
- 1 V-cycle is usually enough
- Elastic energy changes are accurate to 1%
- CPU time $\approx .1$ sec in 1+1 (1024×50) and ≈ 1 sec in 2+1 ($128 \times 128 \times 5$)
- Other work: Algebraic Multigrid + ABC (Caflisch and coworkers)

LOCAL ELASTIC COMPUTATIONS

- It is desirable to update the displacement field locally in the region near where an atom was moved
- Local calculations are difficult due to the long range nature of elastic interactions
- As we shall see elastic interactions are much longer ranging in the film due to the misfit strain
- We shall descibe a method that can provide very accurate estimates of energy changes using local calculations

Continuum Theory



$$W = \int_\Omega |e_{ij} - \chi_F e^0_{ij}|^2 dx$$

e_{ij} =strain tensor, e^0_{ij} misfit strain

$\delta W = 0:$

$$\nabla^2 \mathbf{u} + 2\nabla(\nabla \cdot \mathbf{u}) = 0 \quad x \in \Omega$$

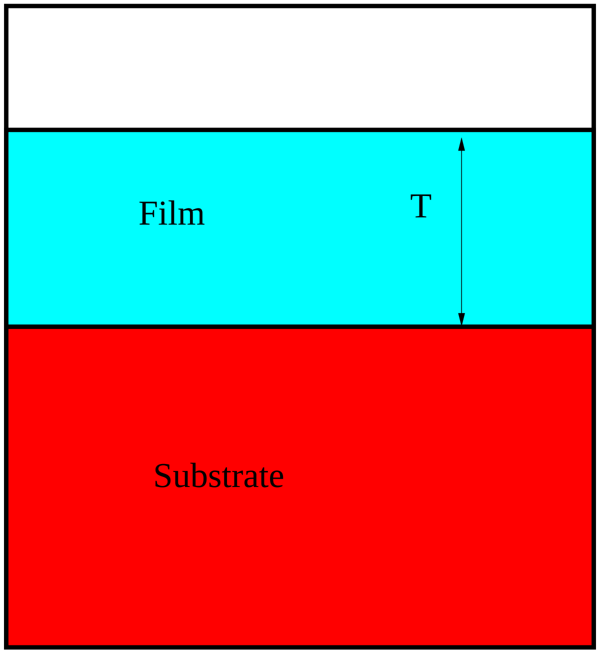
$$\tau_{ij} n_j = \tau^0_{ij} n_j \quad x \in \partial\Omega$$

Ω = Film + Substrate

$\partial\Omega$ = Film/Vacuum Interface

χ_F = characteristic function of the film

Approximate Continuum Theory



Ω = Film + Substrate

$y = T$: Film/Vacuum Interface

ϵ = misfit

Choose reference configuration

so that $e^0_{ij} = \begin{pmatrix} 0 & 0 \\ 0 & \epsilon \end{pmatrix}$

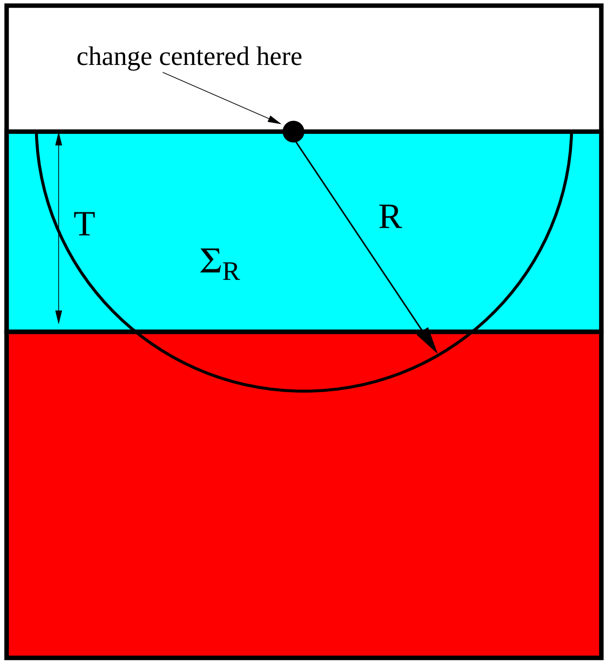
$$\nabla^2 \mathbf{u} + 2\nabla(\nabla \cdot \mathbf{u}) = 0 \quad \mathbf{x} \in \Omega$$

Approximation:

$h(x) \approx T, \quad h'(x) \ll 1$

$\tau_{yy} = 0, \quad \tau_{xy} = \epsilon k h'(x) \quad y = T$

Nonlocal Nature of Elastic Energy



Original state

$$\nabla^2\mathbf{u} + 2\nabla(\nabla\cdot\mathbf{u}) = 0 \quad \mathbf{x}\in\Omega$$

$$\tau_{yy} = 0, \tau_{xy} = \epsilon\partial_x h \quad y = T$$

Change film profile by δh (localized)

$$\nabla^2\mathbf{w} + 2\nabla(\nabla\cdot\mathbf{w}) = 0 \quad x\in\Omega$$

$$\tau_{yy} = 0, \tau_{xy} = \epsilon\partial_x(h - \delta h) \quad y = T$$

Compute Change in Elastic Energy

$$\Delta W = W(\mathbf{u}) - W(\mathbf{w})$$

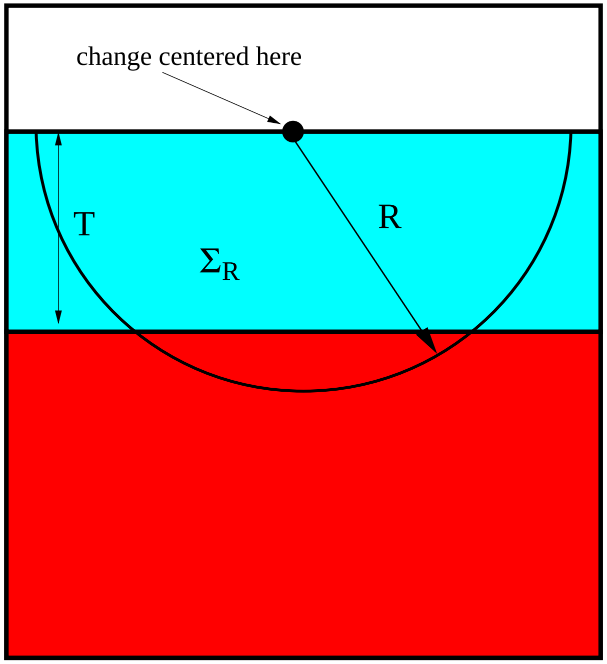
Nonlocality

$$\Delta W_R \equiv \int_{\Sigma_R} \Delta w dx$$

Theorem

$\Delta W = \Delta W_R + O(\epsilon T/R)$ (Large Error)

Elastic Energy Localization



Original state

$$\nabla^2\mathbf{u} + 2\nabla(\nabla\cdot\mathbf{u}) = 0 \quad \mathbf{x}\in\Omega$$

$$\tau_{yy} = 0, \tau_{xy} = \epsilon\partial_x h \quad y = T$$

Change film profile by δh (localized)

$$\nabla^2\mathbf{v} + 2\nabla(\nabla\cdot\mathbf{v}) = 0 \quad x\in\Sigma_R$$

$$\tau_{yy} = 0, \tau_{xy} = \epsilon\partial_x(h - \delta h) \quad y = T$$

$$\mathbf{v} = \mathbf{u} \quad \mathbf{x}\in\partial\Sigma_R$$

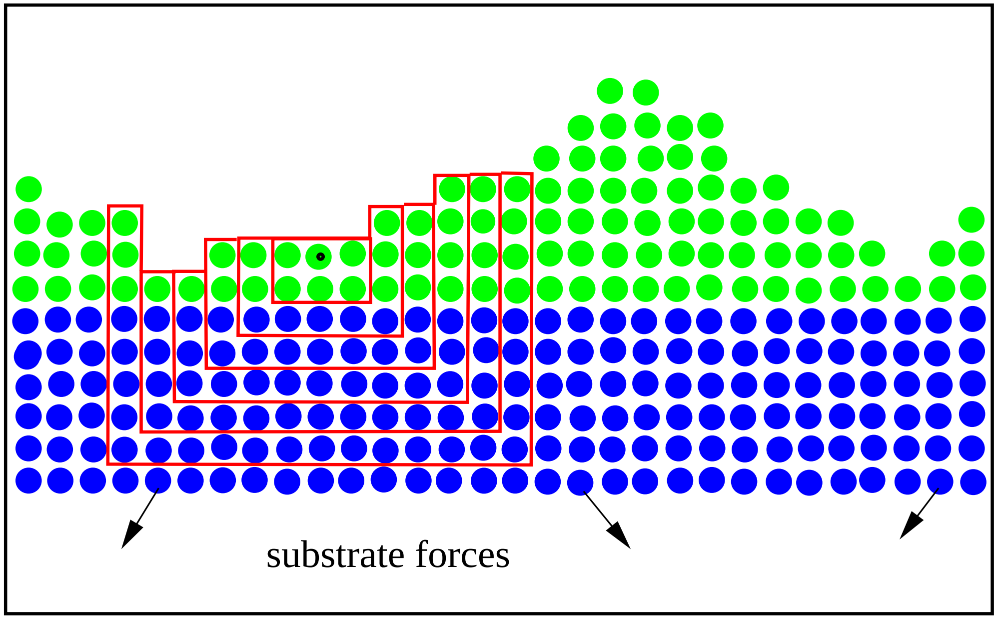
Compute Δw in Σ_R

$$\Delta W_R \equiv \int_{\Sigma_R} \Delta w dx$$

Theorem - Principle of Energy Localization

$\Delta W = \Delta W_R + O(1/R^2)$ (Small Error)

Expanding Box Method



Solve elastic equations with SOR in an expanding region

Use the exterior displacement field for Dirichlet conditions

The max norm of the residual is monitored

Success when $||\text{residual}||_\infty <$ tolerance and box is not too big

If local calculation fails we use a full elastic calculation

Calculation of Energy Changes

- The displacement field is not updated after an atom hops or after a deposition event
- The adatom hopping rate is assumed not to depend greatly on changes in the elastic energy. Therefore no updating the displacement field for adatom motion is performed
- Therefore when attempting to move an atom the displacement field must be updated

Calculation of Energy Changes

- One first attempts to update the displacement field with the expanding box method in a neighborhood of the selected atom. Say box size is N_1
- The atom is then removed and the displacement field is updated using the expanding box method which forced to expand to size N_1
- If successful, the change in elastic energy in the region of size N_1 is computed
- If N_1 is too small, one continues to expand until success. Size of new region is N_2
- One continues the previous calculation expanding to size N_2
- The change in elastic energy in the region of size N_2 is then computed

Calculation of Energy Changes

- If *either* of the expanding box calculations fail then both calculations must be replaced by updating the displacement field in the entire computational domain for both cases. (i.e. with and without the atom)
- The elastic energy is also must be computed on the entire domain with and without the atom

Calculation of Energy Changes

- In the case when both expanding box calculations are successful we will have updated the displacement field inside the same domain with the same Dirchlet boundary condtions at the boundary of the box.
- This the precise set up for the princple of energy localizations and explains why such an approach could be succesful.
- Careful numerical experiments demonstrate the method works extremely well.

A Remark

Suppose we do the following:

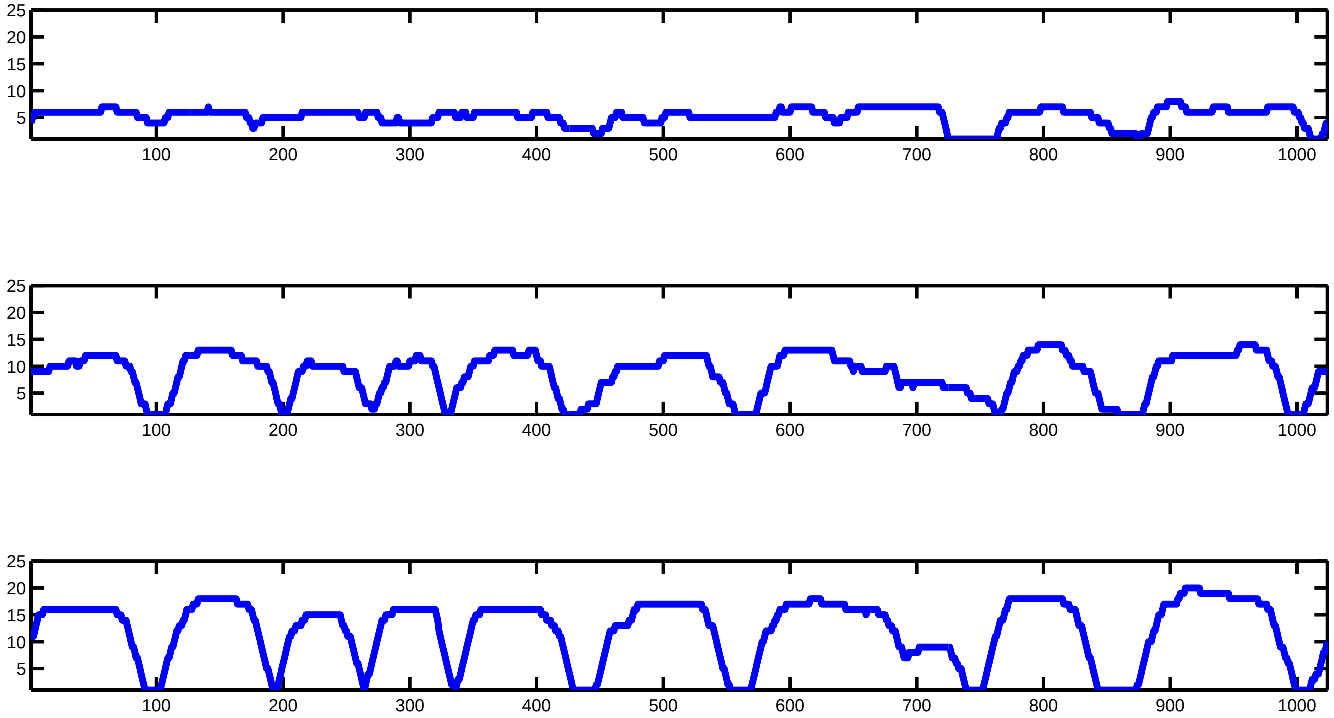
- A sucessful expanding box step with size N_1
- Remove atom
- A sucessful expanding box step with size N_2
- Compute the total elastic energy for both problems in a box of size N_3

Then unless $N_1 = N_2 = N_3$ the value for the change in elastic energy will often have large errors.

REJECTION-REDUCED KMC

- $\Delta E_s < C(n)e(p)$
- Change in elastic energy when removing atom p is bounded by the local energy density of the current configutation at p times a constant depending on the number of bonds
- This allows one to replace R_k by R_k^{upper}, an upper bound on the transition rate using $C(n)e(p)$ for ΔE_s
- When an atom is selected the actual transistion rate is computed. If $r < R_k^{actual}/R_k^{upper}$ move is accepted $(r = U[0, 1])$

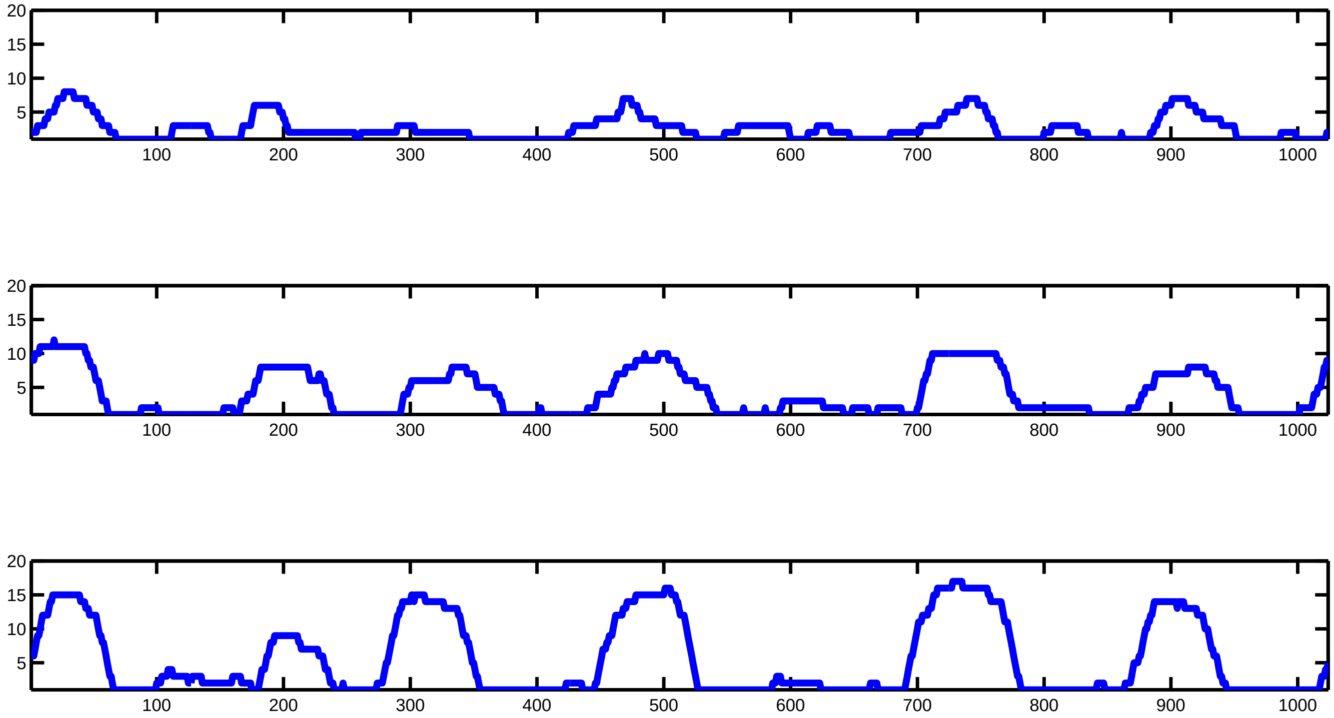
RESULTS



Germanium deposited on Silicon

$T = 600^\circ$ K and $D/F = 1.5 \times 10^7$, misfit = .04

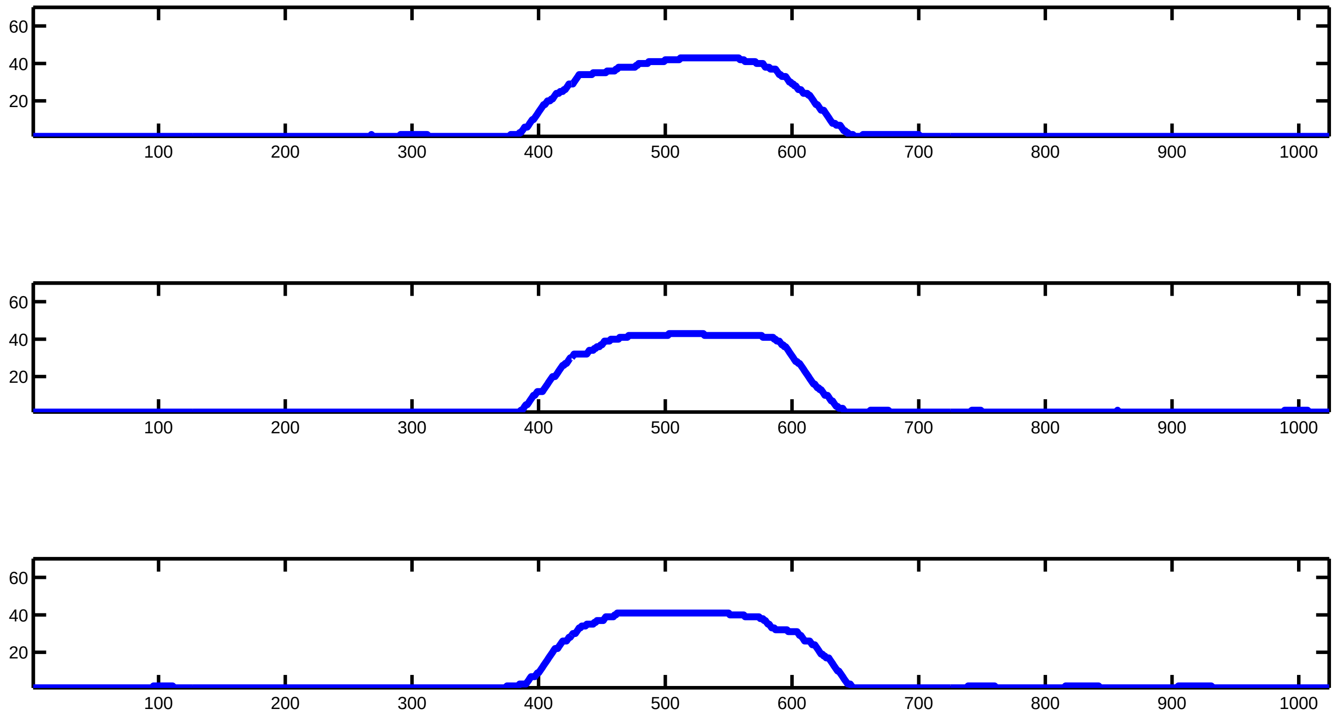
RESULTS



Germanium deposited on Silicon

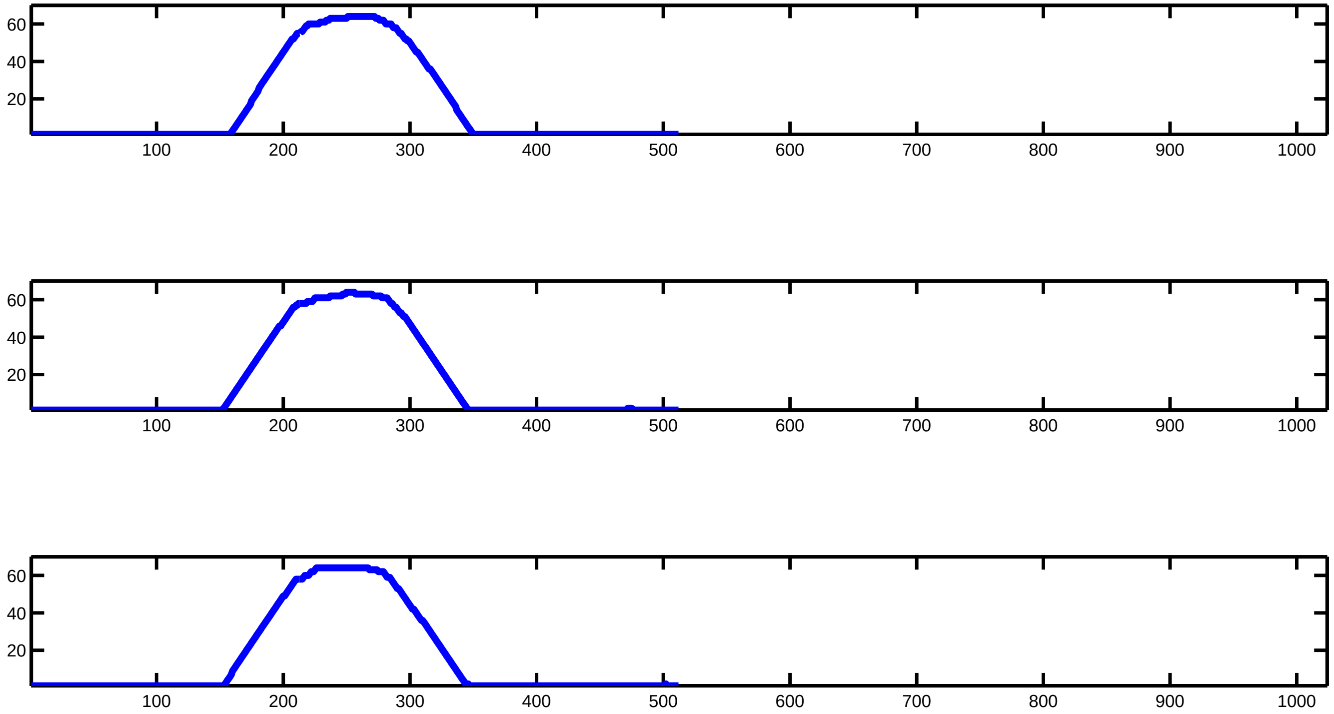
$T = 600^\circ$ K and $D/F = 1.5 \times 10^8$, misfit $= .04$

RESULTS



Annealling, $\sim 10^8$ hops, $T = 500^\circ$ K and misfit $= .02$

RESULTS



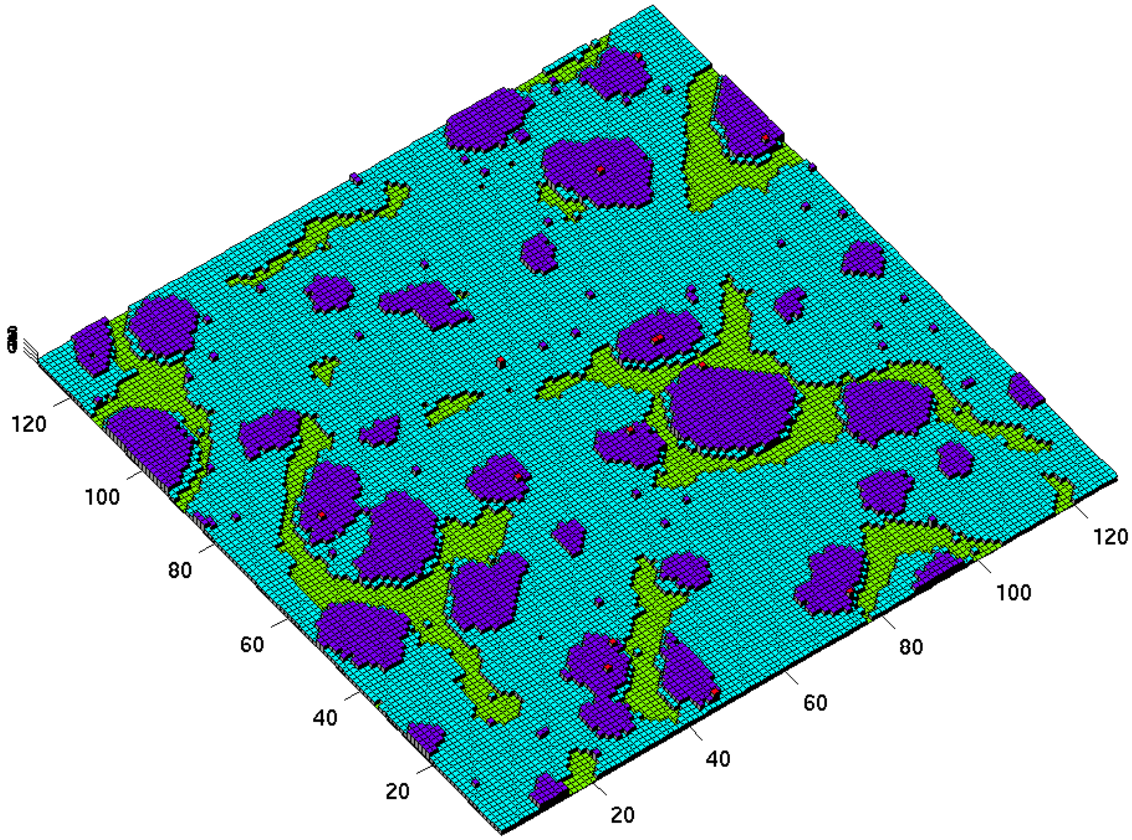
Annealling, $\sim 10^8$ hops, $T = 500^\circ$ K and misfit $=.04$

OBSERVATIONS

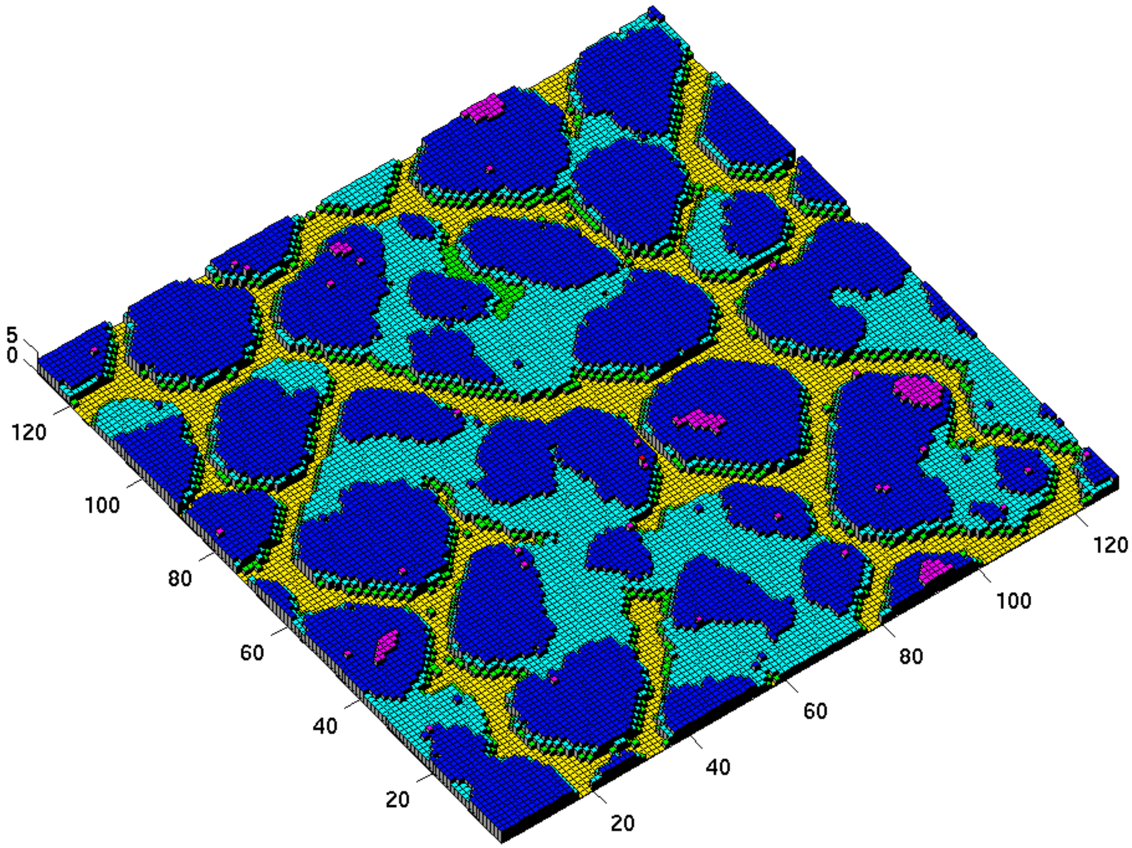
- As the misfit is increased we observe a direct transition from layer-by-layer growth to 3D islands with no wetting layer (Volmer-Weber growth).
- Experimentally this is not observed. Instead it is reported that if the misfit is not too large many layers of layer-by-layer growth will occur before 3D islands form (Stranski-Kratsanov growth).
- Clearly the model is incomplete. One possible missing ingredient is a more accurate model of the alloy. Perhaps phase segregation is important (Tu and Tersoff).

FUTURE PLANS

- Extend method to 2+1 dimensions
- Include intermxing



D/F = 25,000, Spring Constant = 500, misfit=.045



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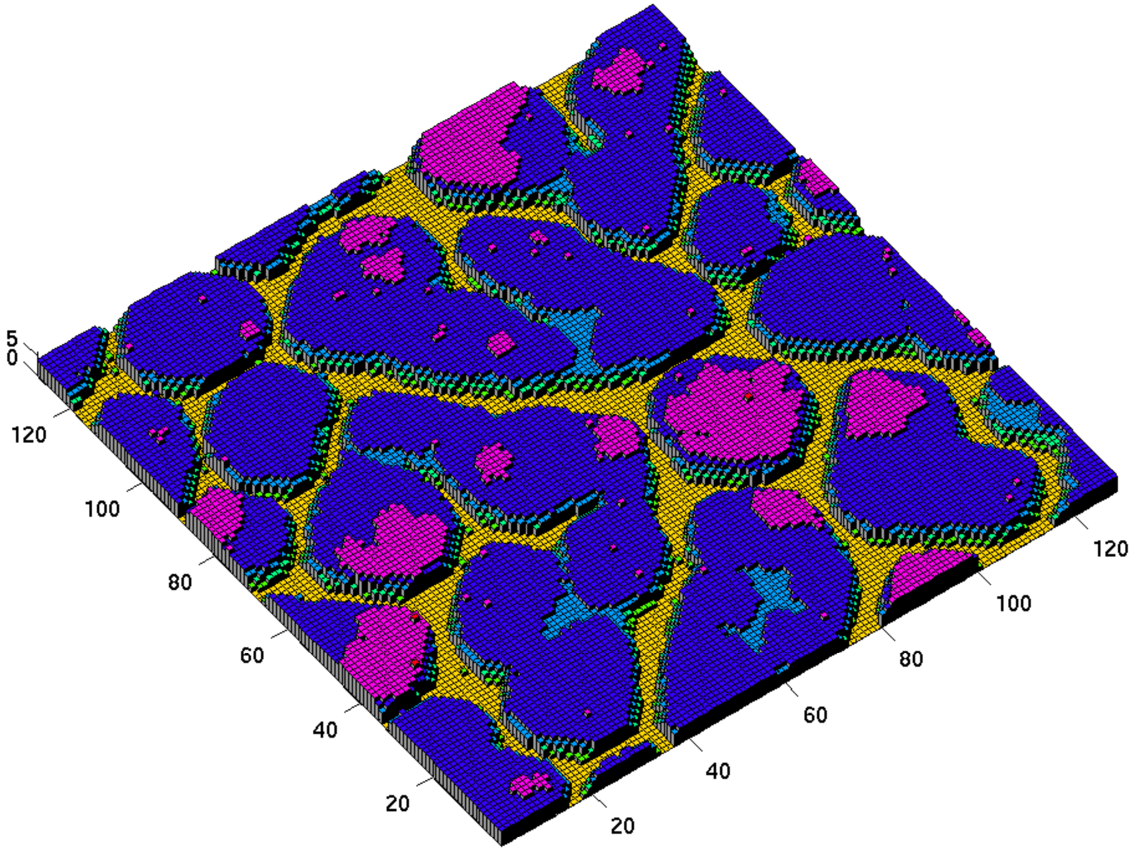

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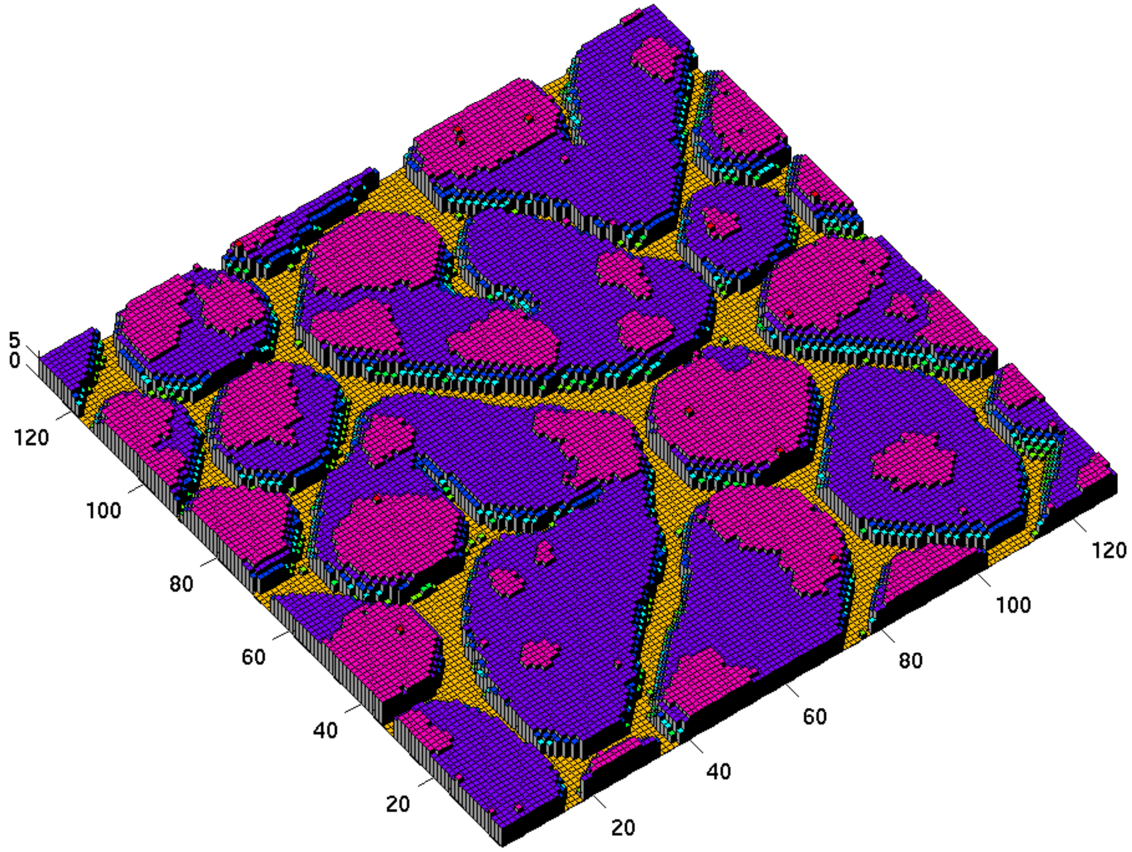
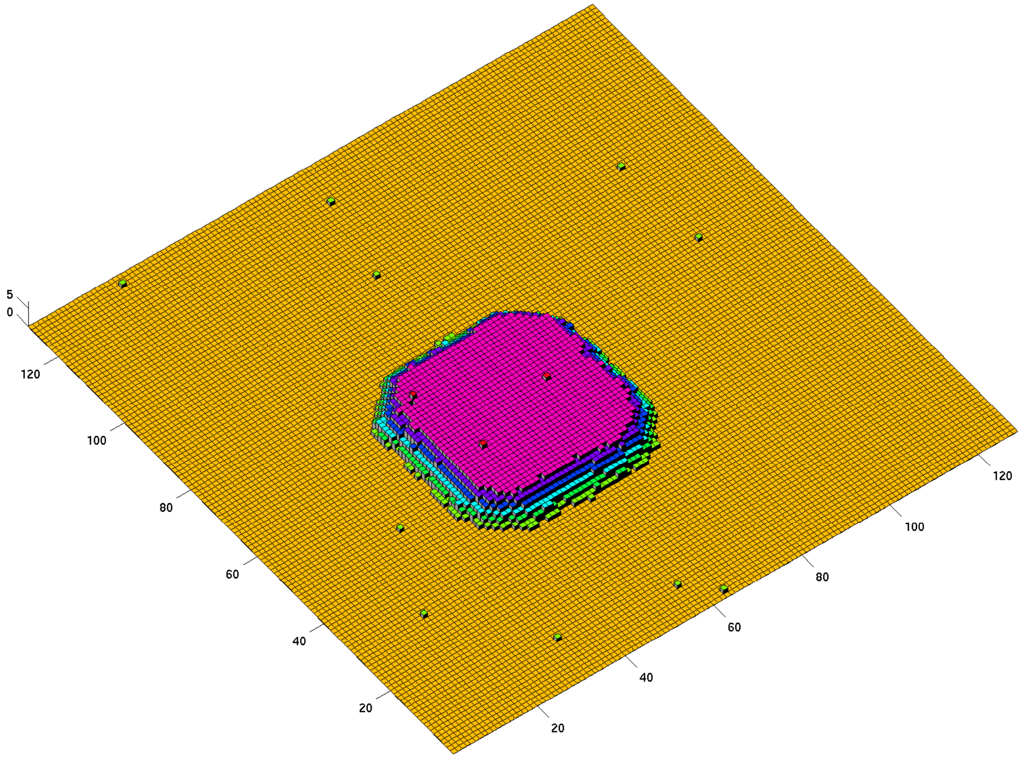


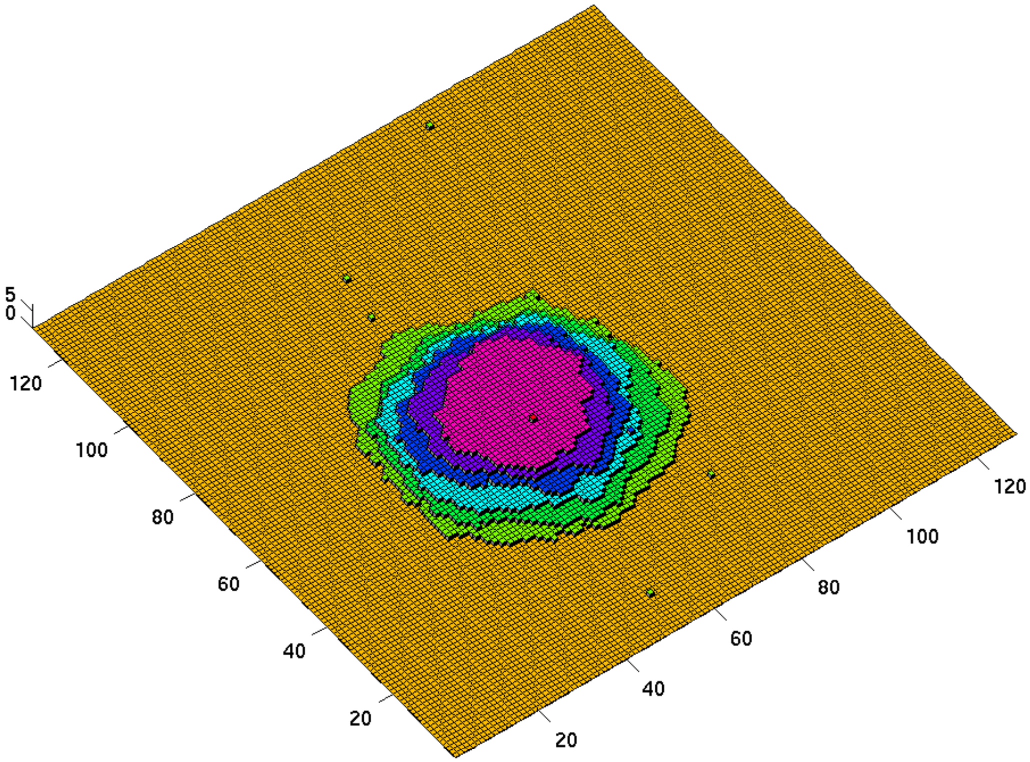
D/F = 25,000, Spring Constant = 500, misfit=.045

“EQUILIBRIUM” SHAPE



D/F = ∞, Spring Const. = 500, misfit=.03. For this misfit a flat surface is also stable. For D/F = 10^5 layer-by-layer growth occurs

RELAXATION



D/F = ∞, Spring Const. = 500, misfit=0. Initial condition: Equilibrium Shape for misfit=.03 (shown on previous slide).

INTERMIXING

Joint with Arvind Baskaran and Jason Devita

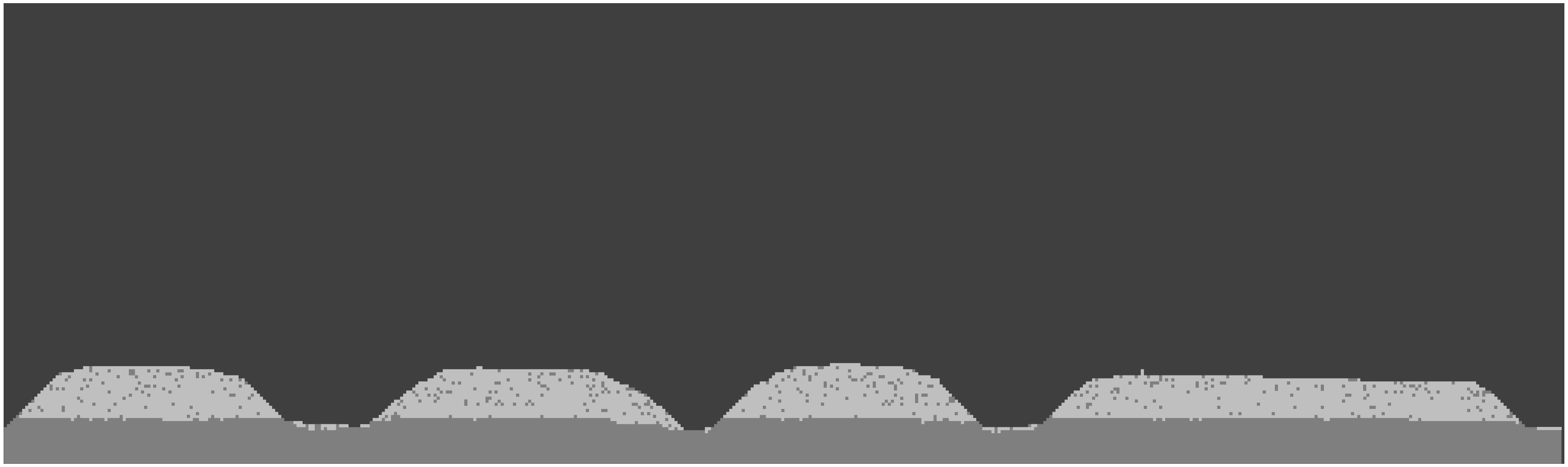


Figure 1: Germanium on Silicon - 10 monolayers. 600°K, 1 monolayer per second

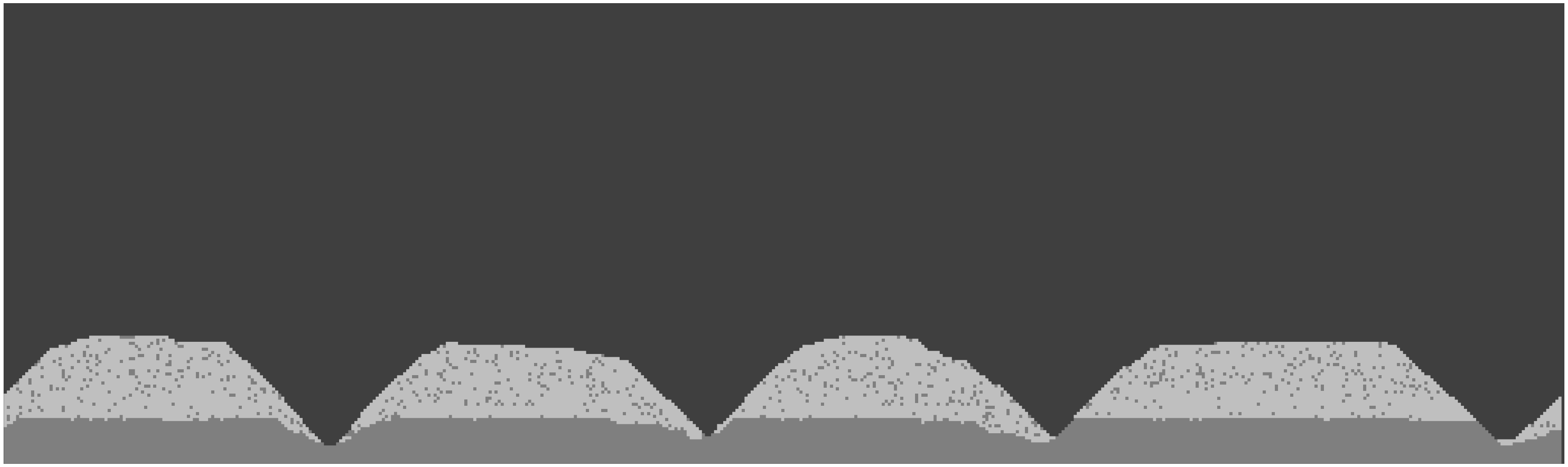


Figure 2: Germanium on Silicon - 16 monolayers. $600^\circ K$, 1 monolayer per second

Figure 3: Germanium on Silicon - 16 monolayers. $600^\circ K$, 1 monolayer per second. (Si-Ge bond energies 30% greater than Si-Si or Ge-Ge bonds)

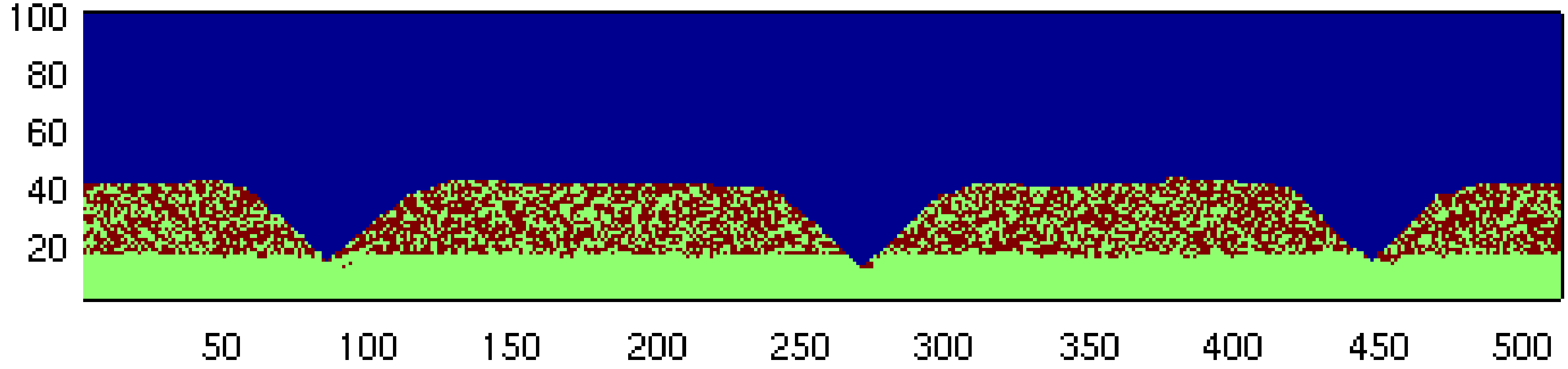


Figure 4: 60 % Germanium - 40% Silicon on Silicon- 15 monolayers. 1000° K, 1 monolayer per second.