Pattern Formation During Directional Epitaxy

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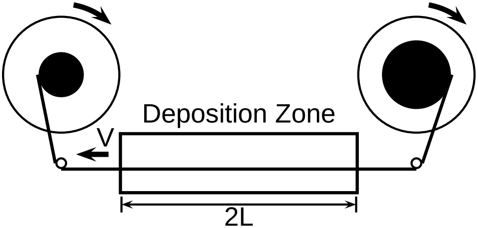
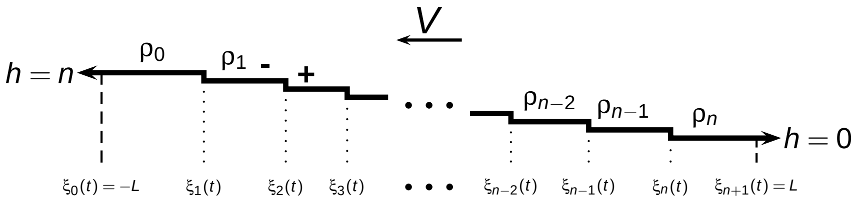
Epitaxial Growth and Continuous Processing

- Epitaxy describes ordered crystalline growth on a mono-crystalline substrate following the crystal structure of the substrate.
- Continuous processing has been used to create large quantities of thin film coated tapes/wire using a reel-to-reel system [Cui et al.,IEEE Trans. Appl. Supercond.,1999].
- "Directional Epitaxy" [Schulze, J. Crystal Growth, 2006] describes epitaxy on a continuously supplied substrate.
- For small deposition zones or near the ends of a larger system, finite size effects and boundary conditions are important.



Simple Reel-to-Reel Processing System (side view)

1-D Continuum Model



Geometry: n steps, $n+1$ terraces (side view)

1-D Burton-Cabrera-Frank (BCF) continuum model [Burton et al.,Phil. Trans. Roy. Soc.,1951]:

$$\partial_t \rho_j - V\partial_x \rho_j = D\partial_x^2 \rho_j + F, \quad \xi_j < x < \xi_{j+1}, \, j = 0,\dots,n$$
$$\pm D\partial_x \rho\big|_\pm \pm (V + \partial_t \xi_j)\rho\big|_\pm = k_\pm (\rho - \rho_e)\big|_\pm, \quad x = \xi_j(t), \, j = 1,\dots,n$$
$$\rho_a(V + \partial_t \xi_j) = D\,[\partial_x \rho]_-^+ + (V + \partial_t \xi_j)\,[\rho]_-^+, \quad x = \xi_j(t), \, j = 1,\dots,n$$
$$\rho_n = \rho_0 = \rho_e, \quad x = \pm L, \text{ respectively}$$

BCF Approximation

- 1-D BCF Model

$$
\begin{aligned}
\partial_t \rho_j - V \partial_x \rho_j &= D \partial_x^2 \rho_j + F, \quad \xi_j < x < \xi_{j+1}, \, j = 0, \ldots, n \\
\pm D \partial_x \rho \big|_{\pm} \pm (V + \partial_t \xi_j) \rho \big|_{\pm} &= k_{\pm} (\rho - \rho_e) \big|_{\pm}, \quad x = \xi_j(t), \, j = 1, \ldots, n \\
\rho_a (V + \partial_t \xi_j) &= D \left[\partial_x \rho \right]_{-}^{+} + (V + \partial_t \xi_j) \left[\rho \right]_{-}^{+}, \quad x = \xi_j(t), \, j = 1, \ldots, n \\
\rho_n = \rho_0 &= \rho_e, \quad x = \pm L, \text{ respectively}
\end{aligned}
$$

BCF Approximation

- Quasistatic approximation assumes the adatom density equilibrates fast compared to the motion of the steps.

$$\partial_t \rho_j - V\partial_x \rho_j = D\partial_x^2 \rho_j + F, \quad \xi_j < x < \xi_{j+1}, j = 0, \ldots, n$$

$$\pm D\partial_x \rho\big|_\pm \pm (V + \partial_t \xi_j)\rho\big|_\pm = k_\pm (\rho - \rho_e)\big|_\pm, \quad x = \xi_j(t), j = 1, \ldots, n$$

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$$\rho_n = \rho_0 = \rho_e, \quad x = \pm L, \text{ respectively}$$

BCF Approximation

- Quasistatic approximation assumes the adatom density equilibrates fast compared to the motion of the steps.
- $\rho = \rho - \rho_e$, scale by the diffusive time scale a^2/D

$$
\begin{aligned}
0 &= D\partial_x^2\rho_j + F, \quad \xi_j < x < \xi_{j+1}, j = 0, \ldots, n \\
\pm D\partial_x \rho\big|_\pm &= k_\pm(\rho - \rho_e)\big|_\pm, \quad x = \xi_j(t), j = 1, \ldots, n \\
\rho_a(V + \partial_t \xi_j) &= D\,[\partial_x \rho]_-^+, \quad x = \xi_j(t), j = 1, \ldots, n \\
\rho_n = \rho_0 &= \rho_e, \quad x = \pm L, \text{ respectively}
\end{aligned}
$$

BCF Approximation

- Quasistatic approximation assumes the adatom density equilibrates fast compared to the motion of the steps.
- $\rho = \rho - \rho_e$, scale by the diffusive time scale a^2/D
- 1-D BCF Quasistatic Approximation

$$
\begin{aligned}
0 = \partial_x^2 \rho_j + F, \quad \xi_j < x < \xi_{j+1}, j = 0, \ldots, n \\
\partial_x \rho_j = \rho_j, \quad x = \xi_j, j = 1, \ldots, n \\
-\partial_x \rho_{j-1} = \rho_{j-1}, \quad x = \xi_j, j = 1, \ldots, n \\
V + \partial_t \xi_j = \partial_x \left(\rho_j - \rho_{j-1} \right), \quad x = \xi_j, j = 1, \ldots, n \\
\rho_n = \rho_0 = 0, \quad x = \pm L, \text{ respectively.}
\end{aligned}
$$

1-D Dynamical System

The adatom density takes the form:

$$\rho_j(x;t) = A_j(\vec{\xi};t) + B_j(\vec{\xi};t)x - \frac{Fx^2}{2}$$

We then obtain the ODE system:

$$\partial_t \xi_j = B_j(\vec{\xi},t) - B_{j-1}(\vec{\xi},t) - V, \quad j = 1,\ldots,n.$$

where for fixed set of $\vec{\xi}$, one can easily solve for $\{A_j, B_j\}_{j=0}^{n}$ the following block diagonal linear system:

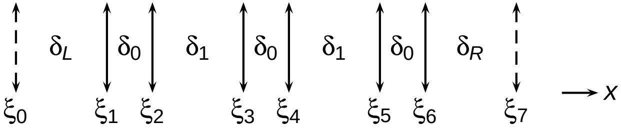
$$\mathbf{M}(\vec{\xi}) \begin{pmatrix} A_0 \\ B_0 \\ A_1 \\ B_1 \\ \vdots \\ A_n \\ B_n \end{pmatrix} = \vec{b}(\vec{\xi})$$

Pairwise Step Patterns (PSP)

- A Pairwise Step Pattern (PSP) is an alternating interior terrace width pattern denoted by δ_0 and δ_1.
- Step locations $\xi_j, j = 1, \ldots, n$ can be expressed as:

$$\xi_j = \begin{cases} -L + \delta_L + \frac{j}{2}\delta_0 + \left(\frac{j}{2} - 1\right)\delta_1, & j \text{ even} \\ -L + \delta_L + \frac{j-1}{2}\delta_0 + \frac{j-1}{2}\delta_1, & j \text{ odd} \end{cases} \qquad j = 1, \ldots, n.$$

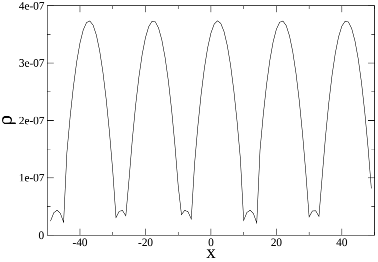
- BCF approximation indicates PSP's exist and have fundamentally different character when the number of steps n is odd or even.
- When seeking steady states, for a range of $V(n)$, there exists fixed points.



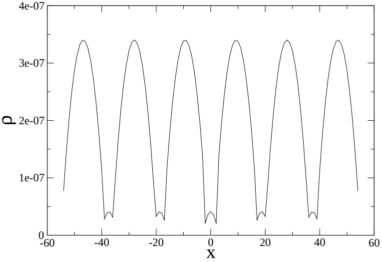
Pairwise Step Pattern for $n = 6$ (side view)

Steady State Adatom Density Profiles $\rho(x)$ (side view)

- Note the existence of PSP for both *n* odd and *n* even.
- Note the asymmetry for *n* odd. The two fixed points have the end profiles interchanged.



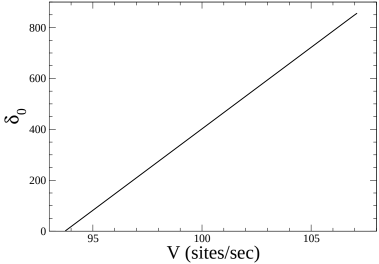
(a) $n = 9$ (odd)



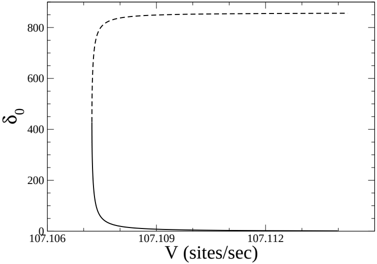
(b) $n = 10$ (even)

$\Delta - V$ Relation

- For n even, a wide range of velocities produces a wide range of δ_0's and the Jacobian evaluated at the valid fixed points has purely imaginary eigenvalues.
- For n odd, a narrow range of velocities produces two values of δ_0 for each V and the Jacobian evaluated at the two fixed points has eigenvalues, one with all negative real parts and one with all positive real parts.



(a) $\delta_0(V)$, $n = 14$ (even)



(b) $\delta_0(V)$, $n = 13$ (odd)

Kinetic Monte Carlo (KMC)

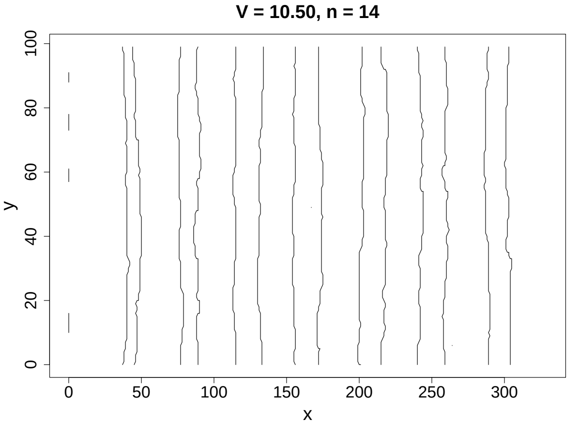
- Atomistic growth model in 2+1 dimensions with nearest neighbor interactions.
- State of surface described by integer height $h(x,y)$ on a square grid lattice of dimensions $M \times N$.
- Equilibrium boundary conditions at grid boundaries in the x direction; periodic boundary conditions in the y direction.
- We utilize an in-plane lateral nearest neighbor bond counting technique to determine rates (probabilities) of an adatom moving [Clarke and Vvedensky,J. Appl. Phys., 1988].
- Surface atoms hop to neighboring sites with rates given by R:

$$\Delta E = E_s + nE_n, \quad R = K(T)\exp(-\Delta E/(k_B T))$$

- These hopping rates vary based on the current surface morphology.

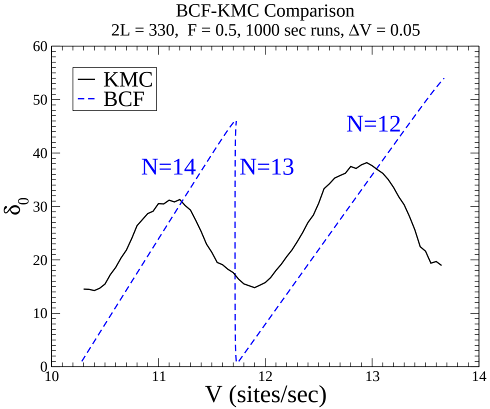
KMC Step Edge Contours (plan view)

- Evidence of PSP in 2+1 KMC clearly visible.
- Requires relatviely short terrace widths to reduce island nucleation influence.



KMC $\bar{\delta}_0$, BCF δ_0

- KMC appears to match slope correctly for *n* even.
- Possible reasons for not matching include nucleation, integer resolution of KMC, other stochastic and 2-D effects.

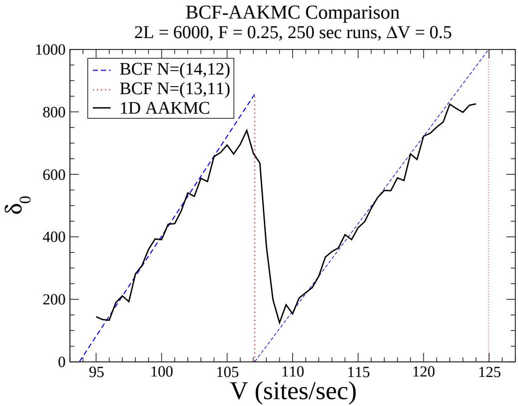


Adatom Kinetic Monte Carlo (AAKMC)

- 1-D version of KMC, no nucleation, developed in order to be more like our BCF approximation.
- Allows us to then go to larger terrace widths and thus minimize integer resolution issues and stochastic noise.
- Rather than describing the state of the surface, one tracks the positions of step edges and adatom locations.
- Adatom movement is treated as a random walk on flat surface covering whole domain.
- Adatoms do not interact with each other.
- When adatom lands in front of step edge, that step edge moves forward.

AAKMC $\bar{\delta}_0$, BCF δ_0

- Note very close matching throught velocity range.



Summary

- Nonperiodic BC are relevant for small systems or near the boundaries. Periodic BC are applicable to large systems or far from the boundaries.
- "Steady States" exist for continuously processed systems, i.e., the height profile is stationary and mean step positions don't change.
- These states take the form of a Pariwise Step Pattern (PSP) where the interior terrace widths alternate between two widths, δ_0 and δ_1.
- AAKMC confirms this with very good agreement with 1-D continuum approximation.