Pattern Formation During Directional Epitaxy

Michael A. Saum    Tim P. Schulze

Department of Mathematics
The University of Tennessee, Knoxville

Sixth International Congress on Industrial and Applied Mathematics, Zürich, July 16–20, 2007
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].


- 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)](image)
1-D Continuum Model

\[ h = n \]

\[ \xi_0(t) = -L \]
\[ \xi_1(t) \]
\[ \xi_2(t) \]
\[ \xi_3(t) \]
\[ \cdots \]
\[ \xi_{n-2}(t) \]
\[ \xi_{n-1}(t) \]
\[ \xi_n(t) \]
\[ \xi_{n+1}(t) = L \]

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

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

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

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

\[
\rho_a (V + \partial_t \xi_j) = D [\partial_x \rho]^+_\pm + (V + \partial_t \xi_j) [\rho]^+_\pm, \quad x = \xi_j(t), \ j = 1, \ldots, n
\]

\[
\rho_n = \rho_0 = \rho_e, \quad x = \pm L, \text{ respectively}
\]
BCF Approximation

1-D BCF Model

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

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

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

\[
\pm D \partial_x \rho \bigg|_{\pm} \pm (V + \partial_t \xi_j) \rho \bigg|_{\pm} = k_{\pm} (\rho - \rho_e) \bigg|_{\pm}, \quad x = \xi_j(t), \ j = 1, \ldots, n
\]

\[
\rho_a (V + \partial_t \xi_j) = D [\partial_x \rho]^+_\pm + (V + \partial_t \xi_j) [\rho]^+_\pm, \quad x = \xi_j(t), \ j = 1, \ldots, n
\]

\[
\rho_n = \rho_0 = \rho_e, \quad x = \pm L, \text{ respectively}
\]
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 $a^2/D$

\[
0 = D \frac{\partial^2 \rho_j}{\partial x^2} + F, \quad \xi_j < x < \xi_{j+1}, \ j = 0, \ldots, n
\]

\[
\pm D \frac{\partial \rho}{\partial x} \bigg|_\pm = k_\pm (\rho - \rho_e) \bigg|_\pm, \quad x = \xi_j(t), \ j = 1, \ldots, n
\]

\[
\rho_a(V + \partial_t \xi_j) = D [\partial_x \rho]_+, \quad x = \xi_j(t), \ j = 1, \ldots, n
\]

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

\[
\begin{align*}
0 &= \partial_x^2 \rho_j + F, \quad \xi_j < x < \xi_{j+1}, \ j = 0, \ldots, n \\
\partial_x \rho_j &= \rho_j, \quad x = \xi_j, \ j = 1, \ldots, n \\
-\partial_x \rho_{j-1} &= \rho_{j-1}, \quad x = \xi_j, \ j = 1, \ldots, n \\
V + \partial_t \xi_j &= \partial_x (\rho_j - \rho_{j-1}), \quad x = \xi_j, \ j = 1, \ldots, n \\
\rho_n &= \rho_0 = 0, \quad x = \pm L, \text{ respectively.}
\end{align*}
\]
1-D Dynamical System

The adatom density takes the form:

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

We then obtain the ODE system:

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

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

\[
M(\xi) \begin{pmatrix} A_0 \\ B_0 \\ A_1 \\ B_1 \\ \vdots \\ A_n \\ B_n \end{pmatrix} = \bar{b}(\xi)
\]
Pairwise Step Patterns (PSP)

- A Pairwise Step Pattern (PSP) is an alternating interior terrace width pattern denoted by $\delta_0$ and $\delta_1$.
- Step locations $\xi_j, j = 1, \ldots, n$ can be expressed as:
  \[
  \xi_j = \begin{cases} 
  -L + \delta_L + \frac{j}{2} \delta_0 + \left(\frac{j}{2} - 1\right) \delta_1, & j \text{ even} \\
  -L + \delta_L + \frac{j-1}{2} \delta_0 + \frac{j-1}{2} \delta_1, & j \text{ odd}
  \end{cases} 
  \]
- 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.
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 \( \delta_0 \)'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 \( \delta_0 \) 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.

\[ \delta_0 \]

\( V \) (sites/sec)

(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 relatively short terrace widths to reduce island nucleation influence.

\[ V = 10.50, \ n = 14 \]
KMC $\bar{\delta}_0$, BCF $\delta_0$

- 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 $\tilde{\delta}_0$, BCF $\delta_0$

- Note very close matching throughout 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, $\delta_0$ and $\delta_1$.
- AAKMC confirms this with very good agreement with 1-D continuum approximation.