Step Bunching in Epitaxial Growth with Elasticity Effects

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05 Jan 2017

Outline

1. Introduction

Step bunching phenomenon

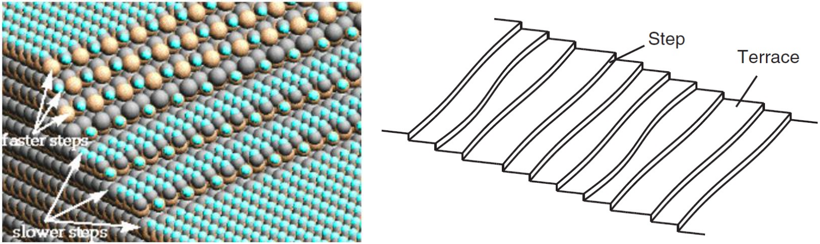


Figure: Stepped epitaxial surfaces.

Left: from the Internet; Right: [Xu and Xiang, SIAM J. Appl. Math., '09].

Step bunching phenomenon (cont.)

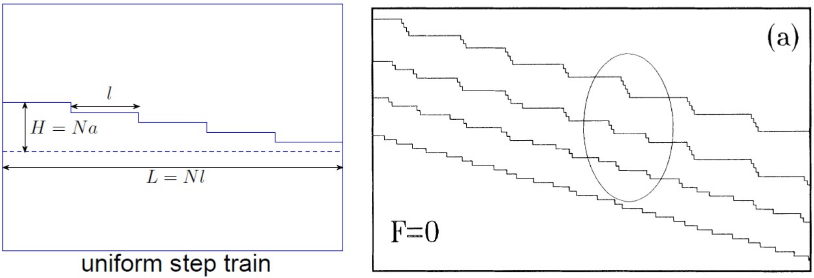
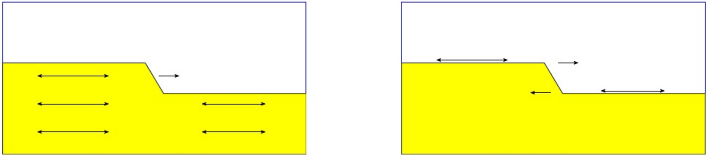


Figure: Left: uniform step train; right: step-bunching phenomenon.

Left: [L., Xiang, and Yip, Multiscale Model. Simul., '16];
Right: [Tersoff et al, Phys. Rev. Lett., '95].

Elasticity effects

- force monopole, by misfit stress in the bulk
  (attractive, destabilizing the uniform step train)
- force dipole, by steps
  (repulsive, stabilizing the uniform step train)



Monopole at step   Dipole at step

Figure: Elasticity effects in epitaxial growth. [L., Xiang, and Yip, Multiscale Model. Simul., '16]

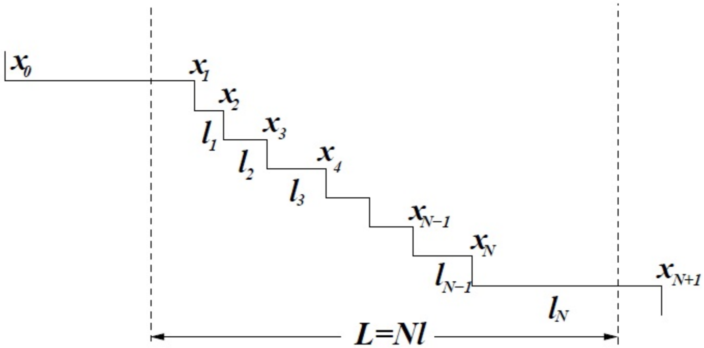


Figure: An example of step configuration. [L., Xiang and Yip, Multiscale Model. Simul., '16]

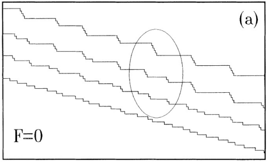
Tersoff's Discrete model [Tersoff et al, Phys. Rev. Lett., '95]

$$\frac{1}{a^2}\frac{\mathrm{d}x_n}{\mathrm{d}t} = F\frac{l_n + l_{n+1}}{2} + \frac{\rho_0 D}{k_B T}\left(\frac{f_{n+1} - f_n}{l_n} - \frac{f_n - f_{n-1}}{l_{n-1}}\right) \tag{1}$$

where

$$f_n = -\sum_{m\neq n}\left(\frac{\alpha_1}{x_m - x_n} - \frac{\alpha_2}{(x_m - x_n)^3}\right) \tag{2}$$

- a: lattice constant, F: adatom flux, $l_n = x_{n+1} - x_n$: step length.
- physical constants: ρ_0 equilibrium adatom density on a step in the absence of elastic interactions, D diffusion constant on the terrace, k_B Boltzmann constant, T temperature.
- $-\sum_{m\neq n}\frac{\alpha_1}{x_m - x_n}$ due to the misfit stress (attractive)
- $\sum_{m\neq n}\frac{\alpha_2}{(x_m - x_n)^3}$ due to the interaction between the steps (repulsive)



Xiang's continuum model [SIAM J. Appl. Math., '02]

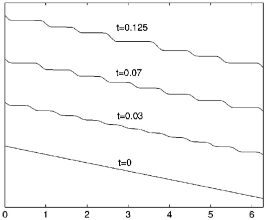
Assume $h \in C^4(\mathbb{R})$ and h is monotonically increasing. Let $x_n = x(h_n, t)$, $h_{n+1} - h_n = a$. Taking continuum limit $a \to 0$, they obatin the continuum equation

$$h_t = a^3 F + \frac{a^5 F}{12} \frac{\partial^2}{\partial x^2} \left(\frac{1}{h_x^2} \right) + \frac{a^2 \pi \alpha_1 \rho_0 D}{k_B T} \frac{\partial^2}{\partial x^2} \left[-H(h_x) - \eta \left(\frac{1}{h_x} + \gamma h_x \right) h_{xx} \right]. \qquad (3)$$

- $\eta = \frac{a}{2\pi}$, a is the lattice constant, $\eta \ll 1$. Other constants are $O(1)$.
- Hilbert transform $H(f)(x) = \frac{1}{\pi} p.v. \int_{-\infty}^{+\infty} \frac{f(y)}{x-y} \mathrm{d}y$.

Remarks:

1. [Xiang, SIAM J. Appl. Math., '02] and [Xiang & E Phys. Rev. B, '04] also derived a continuum model from the discrete model by [Duport et al., Phys. Rev. Lett., '95], including Schwoebel effect and interaction between adatoms and steps.
2. [Dal Maso et al, Arch. Rat. Mech. Anal., '14] and [Fonseca et al, Commun. Partial Differ. Equ., '15] established the well-posedness for Xiang's model.



Linear stability analysis (at the uniform step train)

- discrete: by [Tersoff et al., Phys. Rev. Lett., '95]
- continuum: by [Xiang & E, Phys. Rev. B, '04]

Numerical simulation

- both recover the step bunching phenomenon
- consistent to each other

The instability analysis only works for the profile very close to a uniform step train. However, the bunching happens after the initial stage. (Compare this with the Cahn–Hilliard theory: initially spinodal decomposition and then coarsening).

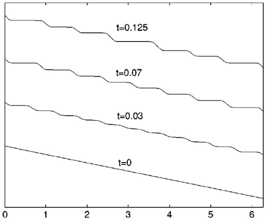


Figure: Continuum model for step-bunching. [Xiang and E, Phys. Rev. B, '04]

2. Energy scaling and asymptotic properties

Question: can we rigorously prove that the step bunching will eventually take place?

Q & A

- Q: Is there a solution for the minimization and dynamical problems?
  A: Existence of minimizer & gradient structure of the dynamics.
- Q: What is the energy of the minimizer?
  A: Energy scaling law.
- Q: Does the minimizer have only one step bunch?
  A: All steps concentrate in a narrow band.
- Q: What is the structure of this bunch?
  A: Size of the bunch & slope of the bunch profile.

For a finite system, consider the energy minimization problem: to find $X_N = (x_1, \cdots, x_N)^T$ such that

$$E[X_N] = \inf_{x'_1 < x'_2 < \cdots < x'_N} E[X'_N], \tag{4}$$

where

$$E[X_N] = \sum_{1 \le i < j \le N} e(x_j - x_i), \tag{5}$$

$$e(x) = \alpha_1 \log |x| + \frac{\alpha_2}{2} x^{-2}. \tag{6}$$

Remark: our results hold for an infinite system in the periodic setting.

Theorem (existence)

(1)There exists a (global) minimizer of E for each N.
(2)For any initial data, the ODE system has a solution on $[0, +\infty)$, i.e., no finite time blow up.

Indeed, there is a metric g, s.t. $\frac{\mathrm{d}E}{\mathrm{d}t} = -\langle \frac{\mathrm{d}x}{\mathrm{d}t}, \frac{\mathrm{d}x}{\mathrm{d}t} \rangle_g$.

Theorem (energy scaling law)

For any $\delta > 0$, *there exist positive constants* c_δ *and* C *such that*

$$\left(\frac{\alpha_1}{4} - \delta\right) N^2 \log N - c_\delta N^2 \le E_N \le \frac{\alpha_1}{4} N^2 \log N + C N^2 \quad \textit{for all} \quad N, \tag{7}$$

where

$$E_N := E[X_N] = \inf_{x_1' < x_2' < \cdots < x_N'} E[X_N'] \tag{8}$$

and $X_N = (x_1, \cdots, x_N)^T$ *is an energy minimizer.*

Theorem (terrace lengths)

For any $\delta > 0$, there exist positive constants c_δ, C_δ and C such that, for all N and for any energy minimizer X_N, we have

$$\text{(minimal terrace length)} \qquad c_\delta N^{-\frac{1}{2}-\delta} \leq \lambda_N \leq C_\delta N^{-\frac{1}{2}+\delta}, \tag{9}$$

$$\text{(maximal terrace length)} \qquad c_\delta N^{-\frac{1}{6}-\delta} \leq \lambda_N' \leq C, \tag{10}$$

where

$$\lambda_N := \min_{1\leq i\leq N-1} \{x_{i+1} - x_i\}, \tag{11}$$

$$\lambda_N' := \max_{1\leq i\leq N-1} \{x_{i+1} - x_i\}. \tag{12}$$

Theorem (bunch size)

For any $\delta > 0$ *and any* $0 < s < 1$, *there exist positive constants* C *and* $C_{\delta,s}$ *such that, for all* N *and for any energy minimizer* X_N, *we have*

$$\text{(lower bound)} \qquad w_N := x_N - x_1 \geq CN^{\frac{1}{2}} (\log N)^{-\frac{1}{2}}, \tag{13}$$

$$\text{(upper bound)} \qquad \min_i \left\{ x_{i+[sN]} - x_i \right\} \leq C_{\delta,s} N^{\frac{1}{2}+\delta}. \tag{14}$$

Since δ can be arbitrarily small in these theorems, we essentially obtain the relations as $N \to \infty$:

$$\text{(minimum energy)} \qquad E_N \sim N^2 \log N, \tag{15}$$

$$\text{(minimal terrace length)} \qquad \lambda_N \sim N^{-\frac{1}{2}}, \tag{16}$$

$$\text{(bunch size)} \qquad w_N \sim N^{\frac{1}{2}}, \tag{17}$$

Recall that the size of the reference state (uniform step train) is

$$L = Nl \sim N. \tag{18}$$

Lennard–Jones (m, n) potential with interaction range index γ

$$E[X_N] = \sum_{1\le i<j\le N,\ j-i\le N^{\gamma}} e(x_j - x_i) \tag{19}$$

$$e(x) = \begin{cases} -\frac{\alpha_1}{m}|x|^{-m} + \frac{\alpha_2}{n}|x|^{-n} & , -1 < m < n, m \neq 0, n \neq 0, \\ \alpha_1 \log|x| + \frac{\alpha_2}{n}|x|^{-n} & , 0 = m < n, \\ -\frac{\alpha_1}{m}|x|^{-m} - \alpha_2 \log|x| & , -1 < m < n = 0. \end{cases} \tag{20}$$

Notice that:
In epitaxial growth model, LJ $(0, 2)$, 'step-bunching' appears.
However, in the classical LJ $(6, 12)$, 'bunching' phenomenon is never mentioned.

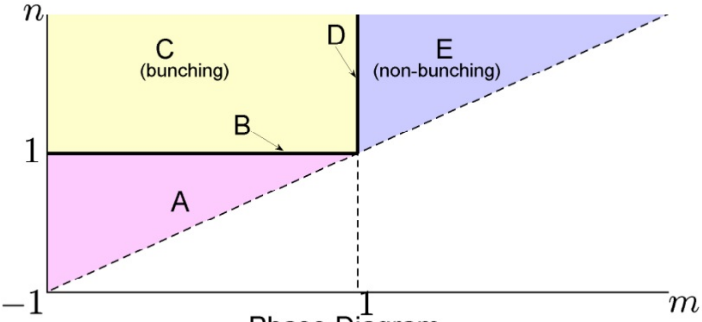


Figure: Phase Diagram of the Scaling Law. This diagram characterizes the scaling behaviors of Lennard-Jones (m, n) potential with $\gamma = 1$.

Case C: $-1 < m < 1 < n$, 'bunching' regime. $(\limsup_{N\to+\infty} \frac{w_N}{N} = 0)$
Case E: $1 < m < n$, 'non-bunching' regime. $(\liminf_{N\to+\infty} \frac{w_N}{N} > 0)$

Theorem (bunching regime)

Suppose that $-1 < m < 1$, $1 < n$, *and* $0 < \gamma \leq 1$. *There exist positive constants* C, C', θ, *and* N_0 *such that, for any* $N > N_0$ *and any energy minimizer* X_N, *we have*
(A) energy scaling law

$$CN^{1+\frac{(1-m)n\gamma}{n-m}} \leq E_N \leq C'N^{1+\frac{(1-m)n\gamma}{n-m}}, \qquad -1 < m < 0, \tag{21}$$

$$-CN^{1+\frac{(1-m)n\gamma}{n-m}} \leq E_N \leq -C'N^{1+\frac{(1-m)n\gamma}{n-m}}, \qquad 0 < m < 1, \tag{22}$$

$$\frac{n-1}{2n}N^2\log N - CN^2\log\log N \leq E_N \leq \frac{n-1}{2n}N^2\log N + C'N^2, \qquad m = 0, \gamma = 1, \tag{23}$$

$$\frac{(n-1)\gamma}{n}N^{1+\gamma}\log N - CN^{1+\gamma}\log\log N \leq E_N \leq \frac{(n-1)\gamma}{n}N^{1+\gamma}\log N + C'N^{1+\gamma}, \qquad m = 0, 0 < \gamma < 1; \tag{24}$$

Theorem (bunching regime)

(B) minimal terrace length

$$CN^{-\frac{(1-m)\gamma}{n-m}} \le \lambda_N \le C' N^{-\frac{(1-m)\gamma}{n-m}}, \qquad -1 < m < 1, m \neq 0, \tag{25}$$

$$CN^{-\frac{\gamma}{n}} (\log N)^{-\frac{1}{n}} \le \lambda_N \le C' N^{-\frac{\gamma}{n}}, \qquad m = 0; \tag{26}$$

(C) system size

$$CN^{1-\frac{(1-m)\gamma}{n-m}} \le w_N \le C' N^{1-\theta}, \qquad -1 < m < 1, m \neq 0, \tag{27}$$

$$CN^{1-\frac{\gamma}{n}} (\log N)^{-\frac{1}{n}} \le w_N \le C' N^{1-\theta}, \qquad m = 0. \tag{28}$$

In particular, we have $\lambda_N \ll 1$, and the system is in the bunching regime

$$\limsup_{N\to+\infty} \frac{w_N}{N} = 0.$$

Remark that C and C' may be different in part (A), (B), and (C).

Theorem (non-bunching regime)

Suppose that either (i) $1 < m < n$, $0 \leq \gamma \leq 1$ *or (ii)* $-1 < m < 1 < n$, $\gamma = 0$. *There exist positive constants* C, C', *and* N_0 *such that, for any* $N > N_0$ *and any energy minimizer* X_N, *we have*
(A) energy scaling law

$$-CN \leq E_N \leq -C'N; \tag{29}$$

(B) minimal terrace length

$$C \leq \lambda_N \leq C'; \tag{30}$$

(C) system size

$$CN \leq w_N \leq C'N. \tag{31}$$

In particular, we have $\lambda_N = O(1)$, *and the system is in the non-bunching regime,*

$$\liminf_{N\to+\infty} \frac{w_N}{N} > 0.$$

To sum up, we have shown the asymptotic behaviors for two different regimes:

$$\begin{aligned}
\text{bunching regime} \quad & -1 < m < 1 < n, 0 < \gamma \leq 1, && (32)\\
& \lambda_N \ll 1, \ \limsup_{N\to+\infty} \frac{w_N}{N} = 0; && (33)\\
\text{non-bunching regime} \quad & \gamma = 0 \text{ or } 1 < m < n, 0 < \gamma \leq 1, && (34)\\
& \lambda_N = O(1), \ \liminf_{N\to+\infty} \frac{w_N}{N} > 0. && (35)
\end{aligned}$$

Continuum model

$$h_t = \frac{\partial^2}{\partial x^2}\left[-H(h_x) - \eta\left(\frac{1}{h_x} + \gamma h_x\right) h_{xx}\right] \tag{36}$$

where Hilbert transform $H(f)(x) = \frac{1}{\pi} p.v. \int_{-\infty}^{+\infty} \frac{f(y)}{x-y} \mathrm{d}y$.
In the periodic case, the energy

$$E^c[h] = \int_{-L/2}^{L/2} \left(-\frac{\pi\alpha_1}{2}\tilde{h}H(\tilde{h}_x) + \pi\alpha_1\eta h_x \log h_x + \pi\alpha_1\frac{\eta\gamma}{6}h_x^3\right) \mathrm{d}x, \tag{37}$$

where $\tilde{h} = h - Ax$. A is the average slope.
Fix L and let $\varepsilon = \eta \to 0$.

- Existence. WLOG $h(0) = 0$.
- Slope h_x is symmetric and unimodal:

$$h_x(\xi) \le h_x(\xi'),\ -L/2 < \xi < \xi' < 0; \tag{38}$$
$$h_x(\xi) \ge h_x(\xi'),\ 0 < \xi < \xi' < L/2. \tag{39}$$

- Large slope parts concentrate in a narrow band

$$H - \varepsilon^{\beta} < h(\varepsilon^{\alpha}) - h(-\varepsilon^{\alpha}) < H. \tag{40}$$

- Step bunch size determined by matched asymptotics: size $\sim \eta^{1/2}$.

These results are consistent with the discrete ones.

4. Conclusion

For Tersoff's discrete model, we have

- Existence
- Energy scaling law
- One bunch structure
- Optimal bunch size and slope

For Xiang's continuum model, we have

- Existence
- One bunch structure
- Optimal bunch size and slope

For one-dimensional system with LJ (m, n) interaction, we have

- A phase diagram
- 'Bunching' depends on interaction range N^{γ}
- Energy and length scaling laws in bunching/non-bunching regime

Future work

- Dynamics (e.g. coarsening rate, different time stages)
- 2+1 dimensional model

Thank you for your attention!