Three Tales of Three Scales in Epitaxial Growth:
Lecture III:
On the atomistic origin of the BCF model

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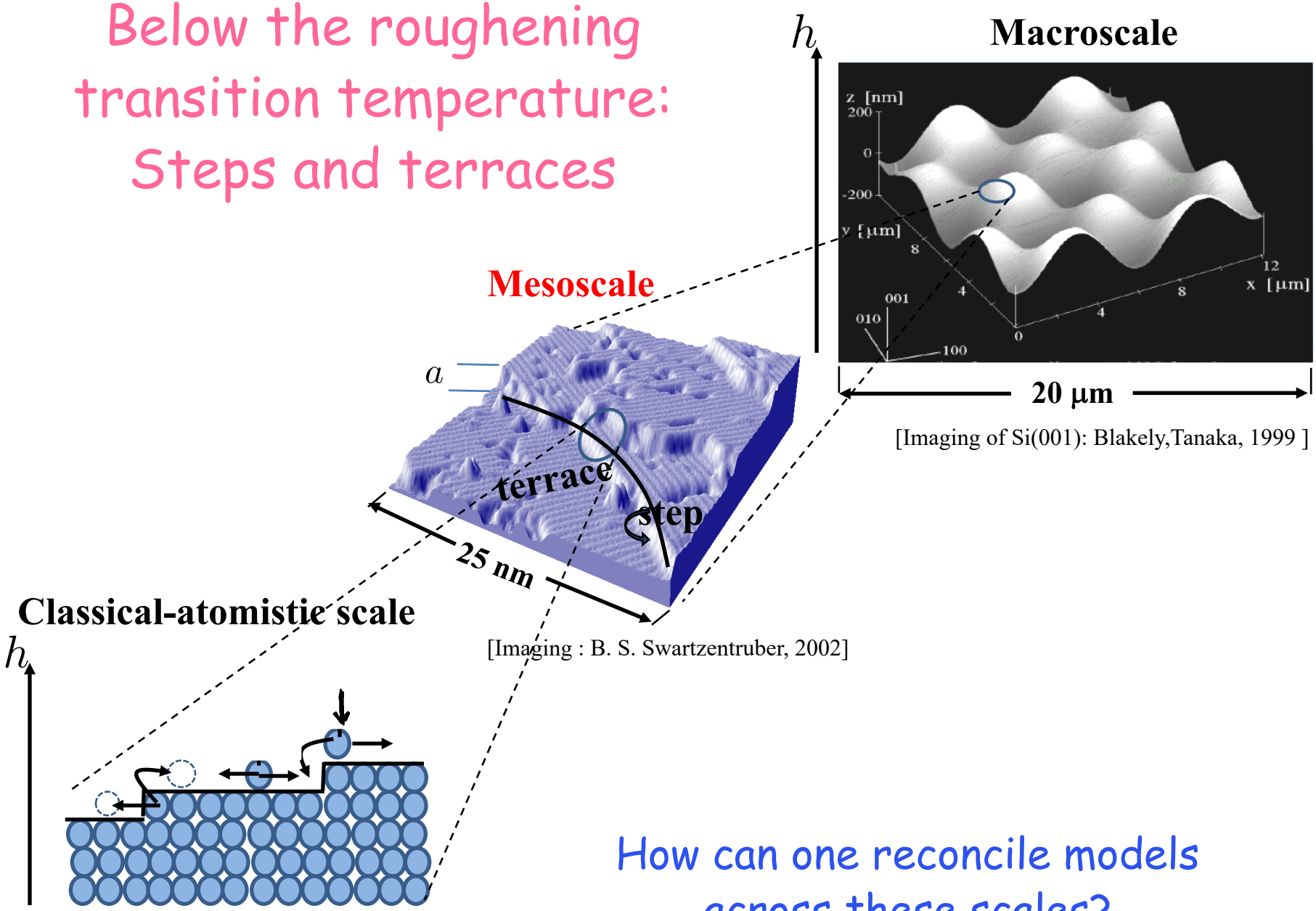
[*]Research supported by NSF DMS-1412769

Symposium on: Mathematical Aspects of Surface and Interface Dynamics
University of Tokyo, Japan
Friday, 19 October 2018

How can the motion of atoms give rise to organized (surface) structures and devices?

Below the roughening transition temperature: Steps and terraces



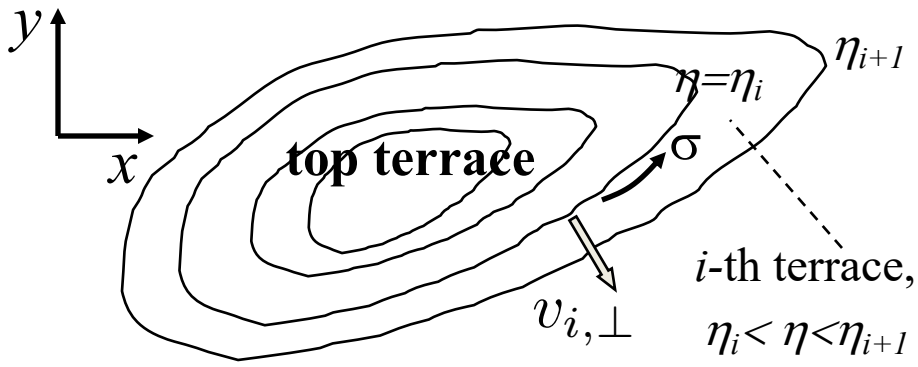
[Imaging of Si(001): Blakely,Tanaka, 1999]

[Imaging : B. S. Swartzentruber, 2002]

How can one reconcile models across these scales?

Mesoscale: Step flow: BCF model [Burton, Cabrera, Frank, 1951]

Local coordinates (η, σ);
descending steps of height a;
i-th step at $\eta = \eta_i$



- Step normal **velocity** :

$$v_{i,\perp} = a^2(J_{i-1,\perp} - J_{i,\perp})$$

- Adatom **diffusion** on i-th terrace:

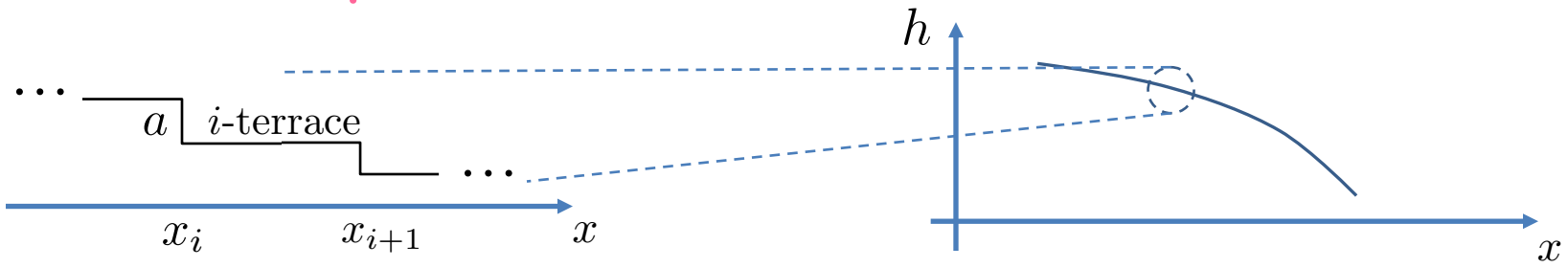
$$\mathbf{J}_i = -D_s \nabla \rho_i, \;\; D_s \Delta \rho_i + F = \frac{\partial \rho_i}{\partial t} \approx 0 \quad \eta_i < \eta < \eta_{i+1}$$

- Robin-type boundary conditions at bounding step edges :

$$-J^+_{i,\perp} = q_+[\rho_i^+ - \rho_i^{\rm eq}(\sigma,t)], \;\; \eta = \eta_i; \;\; J^-_{i,\perp} = q_-[\rho_i^- - \rho_{i+1}^{\rm eq}(\sigma,t)], \;\; \eta = \eta_{i+1}$$

$$\rho_i^{\rm eq} = \rho_s e^{\mu_i/T}$$ Gibbs-Thomson relation

Step motion and continuum limit (Review)



Step velocity

Mass flux on *i*-th terrace

$$\dot{x}_i = a(J_{i-1} - J_i) \qquad \text{at } x = x_i$$

$a \to 0$

$$\partial_{\tilde{t}} h = -\partial_x J$$

Mass conservation

Diffusion; Attachment/detachment

$$\begin{cases} -J_i = q(\rho_i - \rho_i^{\rm eq}) & \text{at } x = x_i^+ \\ J_i = q(\rho_i - \rho_{i+1}^{\rm eq}) & \text{at } x = x_{i+1}^- \end{cases} \quad (i\text{-th terrace})$$

Adatom density

$$D_s \partial_{xx} \rho_i = \partial_t \rho_i \approx 0 \ , \ J_i = -D_s \partial_x \rho_i \quad x_i < x < x_{i+1}$$

$$J = -\frac{D_s}{1 + \cancel{\frac{D_s}{qa}|\nabla h|}} \partial_x \rho^{\rm eq}$$

Diffusion-limited kinetics

"Fick's law" for surface diffusion

Step chemical potential, near equilibrium

$$\rho_i^{\rm eq} = \rho_s e^{\mu_i / T}$$

step chem. potential

Total step energy (*N* steps)

$$\mu_i = a \frac{\delta E_N^{\rm st}}{\delta x_i}$$

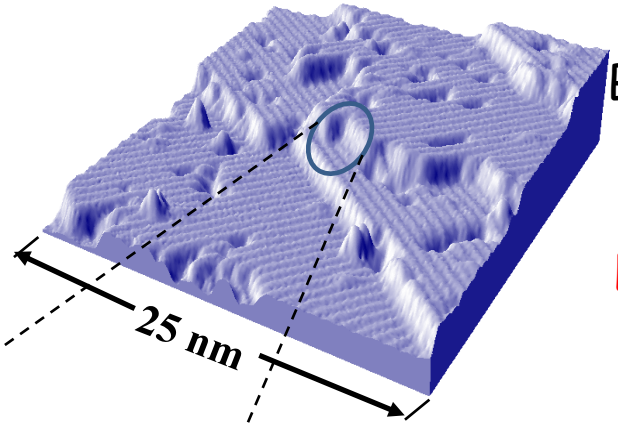
$$\rho^{\rm eq} = \rho_s e^{\mu/T}$$

$$\mu = a \frac{\delta E[h]}{\delta h}$$

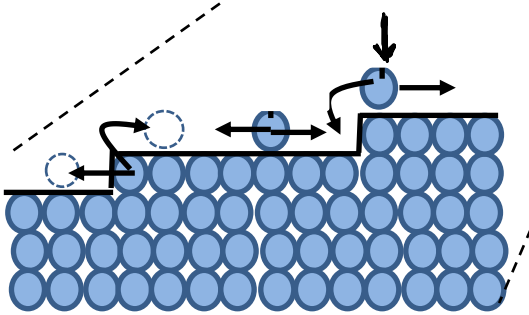
Near-equilibrium condition

From atoms to step motion

Mesoscale



BCF (phenomenological) model:
Adatom diffusion/Fick's law +
Step velocity law +
Linear kinetic relation at step



Kinetic restricted solid-on-solid model:
Atoms hop
in **1+1 dimensions**

Motivation

- The BCF model is primarily phenomenological.

We need to understand its plausible **atomistic** origin.

- If crystal surfaces have high supersaturations, the pileup of adsorbed atoms (adatoms) obstructs step motion.

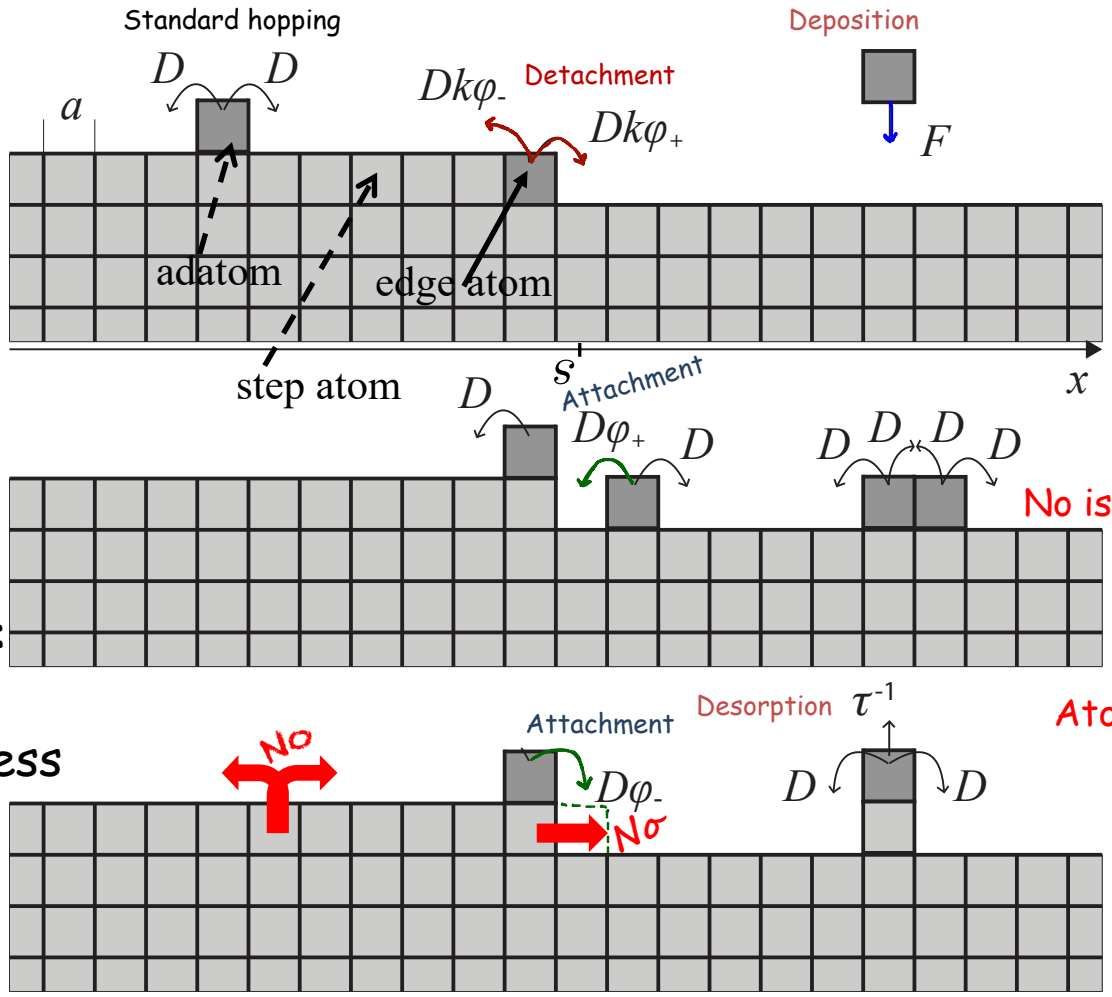
We need to understand how this may affect BCF-type step laws.

Heuristic study for 1+1 dims.
Pending issues, especially in 2+1 dimensions…

[Patrone, DM, 2014; Patrone, Einstein, DM, 2014; Lu, Liu, DM, 2015; Schneider, DM, 2017; Schneider, Patrone, DM, 2018]

Atomistic scale: Basic transitions: Toy model

N lattice sites; $Na = \mathcal{O}(1)$ as $a \to 0$



$k = e^{-E_b/T}$

Key parameters:

k and F/D

controlling diluteness of adatoms

Atoms do not interact elastically

Position of edge atom can only change by -1, 0, 1 on lattice

Program

Main assumptions:

Dimensionality: **1D**. Nucleation is **neglected**. Step is imposed.

Stages:

- Formulate a master equation for adatom configurations.
- Formally express statistical averages related to step motion.
- Examine limit of vanishing lattice spacing via scaling of atomistic parameters.

Atomistic configurations and step motion

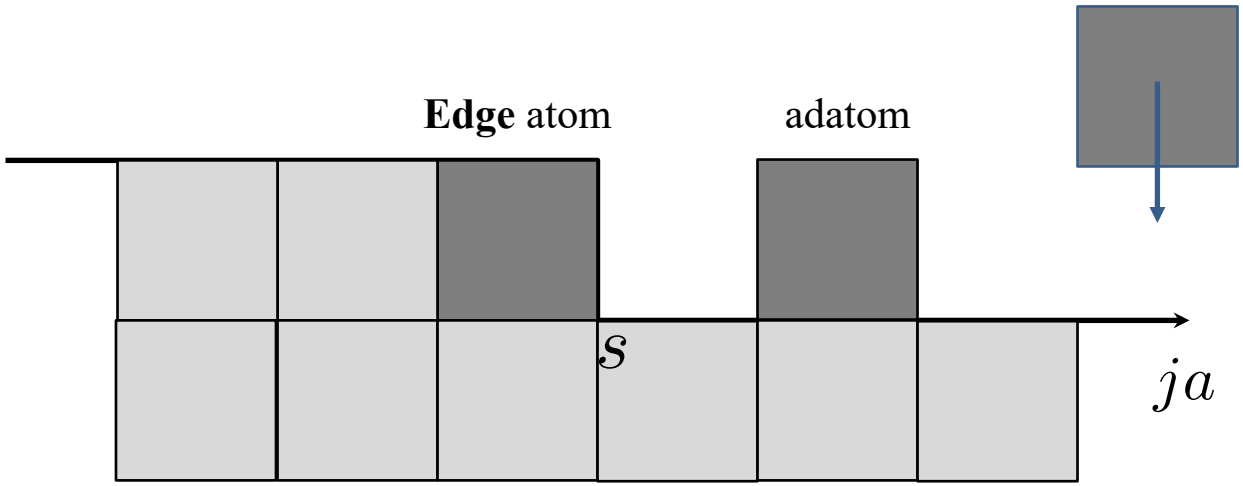
multiset: positions of adatoms

$(\boldsymbol{\alpha}, m)$: state of system

Mass of system (changed by deposition from above)

s_0: initial site for edge atom

m_0: initial mass



s: Microscale step position

$$s = \{s_0 - \underbrace{(|\boldsymbol{\alpha}| - |\boldsymbol{\alpha}_0|)}_{\text{adatom number increase}} + \underbrace{(m - m_0)}_{\text{mass increase (\# of deposited atoms)}}\}a$$

Master equation

$$\dot{p}_{\boldsymbol{\alpha},m}(t) = \sum_{\boldsymbol{\alpha}',m'} T_{(\boldsymbol{\alpha},m),(\boldsymbol{\alpha}',m')} p_{\boldsymbol{\alpha}',m'}(t) \qquad \left(\sum_{\boldsymbol{\alpha},m} T_{(\boldsymbol{\alpha},m),(\boldsymbol{\alpha}',m')} = 0 \right)$$

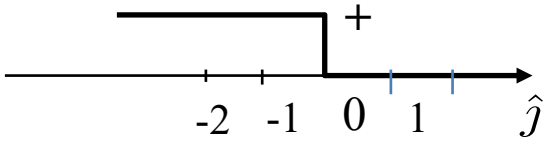
probability density for $(\boldsymbol{\alpha}', m')$

Average of Q: $\langle Q \rangle = \sum_{\boldsymbol{\alpha},m} Q(\boldsymbol{\alpha}, m) p_{\boldsymbol{\alpha},m}(t)$

Goal:
To obtain relations among
averages for step position and adatom density

[Schneider, DM, 2017; Schneider, Patrone, DM, 2018]

Master equation: More details



$$\dot{p}_{\boldsymbol{\alpha},m}(t) = \sum_{\boldsymbol{\alpha}',m'} T_{(\boldsymbol{\alpha},m),(\boldsymbol{\alpha}',m')} p_{\boldsymbol{\alpha}',m'}(t) \qquad \left(\sum_{\boldsymbol{\alpha},m} T_{(\boldsymbol{\alpha},m),(\boldsymbol{\alpha}',m')} = 0 \right)$$

probability density for $(\boldsymbol{\alpha}', m')$

$T_{(\boldsymbol{\alpha},m),(\boldsymbol{\alpha}',m')} = D,$ if $m = m'$ and $|\boldsymbol{\alpha}| = |\boldsymbol{\alpha}'|$ and $|\boldsymbol{\alpha} \setminus \boldsymbol{\alpha}'| = 1$

Hopping by 1 lattice site, away from step

and $\Big| ||\boldsymbol{\alpha} \setminus \boldsymbol{\alpha}'|| - ||\boldsymbol{\alpha}' \setminus \boldsymbol{\alpha}|| \Big| = 1;$

$T_{(\boldsymbol{\alpha},m),(\boldsymbol{\alpha}',m')} = D\phi_{\pm},$ if $m = m'$ and $|\boldsymbol{\alpha}| = |\boldsymbol{\alpha}'| - 1$

attachment

and $\boldsymbol{\alpha}' \setminus \tilde{\boldsymbol{\alpha}} = \{\pm 1\};$ $\tilde{\boldsymbol{\alpha}} = \{\hat{\imath} + 1 |$ for all $\hat{\imath} \in \boldsymbol{\alpha}\}$

relative to edge

$T_{(\boldsymbol{\alpha},m),(\boldsymbol{\alpha}',m')} = Dk\phi_{\pm},$ if $m = m'$ and $|\boldsymbol{\alpha}| = |\boldsymbol{\alpha}'| + 1$

detachment

and $\boldsymbol{\alpha} \setminus \tilde{\boldsymbol{\alpha}}' = \{\pm 1\};$

$T_{(\boldsymbol{\alpha},m),(\boldsymbol{\alpha}',m')} = \dfrac{F}{N-1},$ if $m = m' + 1$ and $|\boldsymbol{\alpha}| = |\boldsymbol{\alpha}'| + 1$

deposition

and $|\boldsymbol{\alpha} \setminus \boldsymbol{\alpha}'| = 1;$

$T_{(\boldsymbol{\alpha},m),(\alpha',m')} = \tau^{-1}$ if $m = m' - 1$, $|\boldsymbol{\alpha}| = |\boldsymbol{\alpha}'| - 1$, $|\boldsymbol{\alpha}' \setminus \boldsymbol{\alpha}| = 1$

Average: $\langle Q \rangle = \sum_{\boldsymbol{\alpha},m} Q(\boldsymbol{\alpha}, m) p_{\boldsymbol{\alpha},m}(t)$

Marginal density

Definition. The *marginal probability density* is

$$p_{\boldsymbol{\alpha}}(t) = \sum_m p_{\boldsymbol{\alpha},m}(t)$$

The marginalized master equation is

$$\begin{aligned}\dot{p}_{\boldsymbol{\alpha}} &= \sum_{\boldsymbol{\alpha}'} \mathcal{T}_{\boldsymbol{\alpha},\boldsymbol{\alpha}'} p_{\boldsymbol{\alpha}'}(t) \\ &= D \sum_{\boldsymbol{\alpha}'} [A_{\boldsymbol{\alpha},\boldsymbol{\alpha}'} + \epsilon B_{\boldsymbol{\alpha},\boldsymbol{\alpha}'}] p_{\boldsymbol{\alpha}'}(t); \quad \epsilon = F/D \ .\end{aligned}$$

Diffusion, attachment/detach.

Deposition

Property of marginalized master equation

PROPOSITION 1. *If a non-trivial steady-state solution, $p^{ss}_{\boldsymbol{\alpha}}$, of the marginalized master equation exists, then any solution $p_{\boldsymbol{\alpha}}(t)$ satisfies*

$$\max_{\boldsymbol{\alpha}} \frac{p_{\boldsymbol{\alpha}}(t)}{p^{ss}_{\boldsymbol{\alpha}}} \leq \max_{\boldsymbol{\alpha}} \frac{p_{\boldsymbol{\alpha}}(0)}{p^{ss}_{\boldsymbol{\alpha}}} \ , \quad t > 0 \ .$$

Proof. We have

$$\begin{aligned} \dot{p}_{\boldsymbol{\alpha}}(t) &= \mathcal{T}_{\boldsymbol{\alpha},\boldsymbol{\alpha}} p_{\boldsymbol{\alpha}}(t) + \sum_{\boldsymbol{\alpha}' \neq \boldsymbol{\alpha}} \mathcal{T}_{\boldsymbol{\alpha},\boldsymbol{\alpha}'} p_{\boldsymbol{\alpha}'}(t) \\ &= \mathcal{T}_{\boldsymbol{\alpha},\boldsymbol{\alpha}} p^{ss}_{\boldsymbol{\alpha}} \frac{p_{\boldsymbol{\alpha}}(t)}{p^{ss}_{\boldsymbol{\alpha}}} + \sum_{\boldsymbol{\alpha}' \neq \boldsymbol{\alpha}} \mathcal{T}_{\boldsymbol{\alpha},\boldsymbol{\alpha}'} p^{ss}_{\boldsymbol{\alpha}'} \frac{p_{\boldsymbol{\alpha}'}(t)}{p^{ss}_{\boldsymbol{\alpha}'}} \\ \left(\sum_{\boldsymbol{\alpha}'} \mathcal{T}_{\boldsymbol{\alpha},\boldsymbol{\alpha}'} p^{\mathrm{ss}}_{\boldsymbol{\alpha}'} = 0 \right) \quad &= \sum_{\boldsymbol{\alpha}' \neq \boldsymbol{\alpha}} \mathcal{T}_{\boldsymbol{\alpha},\boldsymbol{\alpha}'} p^{ss}_{\boldsymbol{\alpha}'} \left\{ \frac{p_{\boldsymbol{\alpha}'}(t)}{p^{ss}_{\boldsymbol{\alpha}'}} - \frac{p_{\boldsymbol{\alpha}}(t)}{p^{ss}_{\boldsymbol{\alpha}}} \right\} \ . \end{aligned}$$

Note that $\mathcal{T}_{\boldsymbol{\alpha},\boldsymbol{\alpha}'} p^{ss}_{\boldsymbol{\alpha}'} \geq 0$ for all $\boldsymbol{\alpha}' \neq \boldsymbol{\alpha}$. If $\boldsymbol{\alpha}$ maximizes (minimizes) $p_{\boldsymbol{\alpha}'}(t)/p^{ss}_{\boldsymbol{\alpha}'}$ over all $\boldsymbol{\alpha}'$, then $\dot{p}_{\boldsymbol{\alpha}}(t) \leq 0$ ($\dot{p}_{\boldsymbol{\alpha}}(t) \geq 0$). □

Near equilibrium evolution: $\max_{\boldsymbol{\alpha}} \{p_{\boldsymbol{\alpha}}(0)/p^{ss}_{\boldsymbol{\alpha}}\} \leq C$.

Equilibrium prob. density in closed form

- $\tau \to \infty$, $F\tau \to 0$: (Mass conserving)

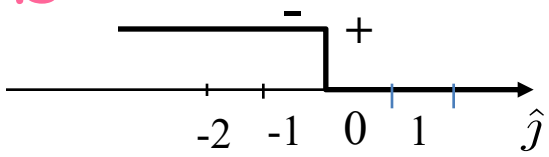
$$p^{ss}_{\boldsymbol{\alpha},m} = p^{eq}_{\boldsymbol{\alpha}} = (1-k)^{N-1} k^{|\boldsymbol{\alpha}|}$$

- $1 < F\tau < (N-1)k$:

$$p^{eq}_{\boldsymbol{\alpha},m} = (1-k)^{N-1} k^{|\boldsymbol{\alpha}|} (1-\mathcal{R}) \mathcal{R}^{m-m_0}$$

$$\mathcal{R} = \frac{F\tau}{(N-1)k}$$

Discrete averages: Definitions



Definition 1. The average step position is

$$\varsigma(t) = a \sum_{\boldsymbol{\alpha},m} \{s_0 - (|\boldsymbol{\alpha}| - |\boldsymbol{\alpha}_0|) + (m - m_0)\} p_{\boldsymbol{\alpha},m}(t) \ .$$

Definition 2. (i) The Eulerian adatom density is

$$\rho_j(t) = \sum_{\boldsymbol{\alpha},m} \nu_{j-s(\boldsymbol{\alpha},m)}(\boldsymbol{\alpha}) p_{\boldsymbol{\alpha},m}(t)/a \ .$$

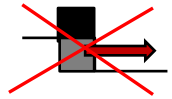
number of adatoms per site

(ii) The Lagrangian-type adatom density is defined by

$$c_{\hat{\jmath}} = \sum_{\boldsymbol{\alpha},m} \nu_{\hat{\jmath}}(\boldsymbol{\alpha}) p_{\boldsymbol{\alpha},m}(t)/a = \sum_{\boldsymbol{\alpha}} \nu_{\hat{\jmath}}(\boldsymbol{\alpha})\, p_{\boldsymbol{\alpha}}(t)/a \ .$$

Definition 3. Discrete adatom fluxes at step edge:

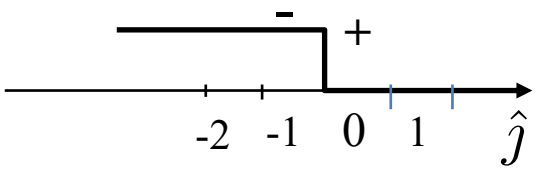
$$J_{\pm}(t) = \pm \sum_{\boldsymbol{\alpha},m} \mathbb{I}(\nu_{-1}(\boldsymbol{\alpha},m) = 0)$$



$$\times \left[T_{(\boldsymbol{\alpha}_{\pm},m),(\boldsymbol{\alpha},m)} p_{\boldsymbol{\alpha},m}(t) - T_{(\boldsymbol{\alpha},m),(\boldsymbol{\alpha}_{\pm},m)} p_{\boldsymbol{\alpha}_{\pm},m}(t) \right]$$

where $\boldsymbol{\alpha}_{\pm} = \widetilde{\boldsymbol{\alpha}} \cup \{\pm 1\}$ results by atom detachment $\left(\widetilde{\boldsymbol{\alpha}} = \{\hat{\imath}+1 \mid \forall \hat{\imath} \in \boldsymbol{\alpha}\}\right)$

Discrete averages: Adatom density $(\tau \to \infty)$



Evolution of discrete adatom density (by **Def. 2** and master eq.):

$$\dot{\rho}_j(t) = D[\rho_{j-1}(t) - 2\rho_j(t) + \rho_{j+1}(t)] + \frac{F}{(N-1)a}$$ discrete diffusion

$$- D[R_{j-1}(t) - 2R_j(t) + R_{j+1}(t)]$$ high-occupation correction

$+$ boundary (step-edge) terms

$$R_j(t) = \sum_{\boldsymbol{\alpha},m} \left[\nu_{j-s(\boldsymbol{\alpha},m)}(\boldsymbol{\alpha}) - \mathbb{1}(\nu_{j-s(\boldsymbol{\alpha},m)}(\boldsymbol{\alpha}) > 0)\right] p_{\boldsymbol{\alpha},m}(t)/a$$

by 2-particle or highly occupied states

By vanishing bdry terms

$$J_\pm(t) = \mp Da\,[c_{\pm 2}(t) - c_{\pm 1}(t)] \mp Da\left[\hat{R}_{\pm 2}(t) - \hat{R}_{\pm 1}(t)\right]$$

discrete Fick's law — Mass accumulation near step edge

From average step position: **(Def. 1)**

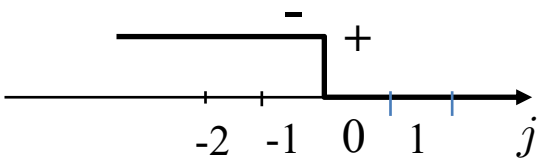
$$\dot{\varsigma}(t) = a\,[J_-(t) - J_+(t)]$$

Discrete averages: Kinetic relation at step edge

Mass Flux (Def.3)

Linear kinetic term correction

$$J_{\pm}(t) = \mp D\phi_{\pm} a\,[c_{\pm 1}(t) - c^{eq}] \mp D\phi_{\pm} a f_{\pm}(t)$$



From F=0: equil. soln. of master eqn.

$$c^{eq} = \frac{\langle n \rangle}{(N-1)a} = \frac{k/a}{1-k}$$

$$\begin{aligned}
f_+(t) &= k\left[c^{eq} + \sum_{\boldsymbol{\alpha}} \mathbb{1}(\nu_{-1}(\boldsymbol{\alpha}) > 0) p_{\boldsymbol{\alpha}}(t)/a\right] \\
&\quad - \sum_{\boldsymbol{\alpha}} \mathbb{1}(\nu_1(\boldsymbol{\alpha}) > 1)\nu_1(\boldsymbol{\alpha}) p_{\boldsymbol{\alpha}}(t)/a \\
f_-(t) &= k\left[c^{eq} + \sum_{\boldsymbol{\alpha}} \mathbb{1}(\nu_{-1}(\boldsymbol{\alpha}) > 0) p_{\boldsymbol{\alpha}}(t)/a\right] \\
&\quad - \sum_{\boldsymbol{\alpha}} \mathbb{1}(\nu_1(\boldsymbol{\alpha}) > 0)\nu_{-1}(\boldsymbol{\alpha}) p_{\boldsymbol{\alpha}}(t)/a \\
&\quad - \sum_{\boldsymbol{\alpha}} \mathbb{1}(\nu_1(\boldsymbol{\alpha}) = 0)\mathbb{1}(\nu_{-1}(\boldsymbol{\alpha}) > 1) \\
&\quad \times [\nu_{-1}(\boldsymbol{\alpha}) - 1]\, p_{\boldsymbol{\alpha}}(t)/a\ .
\end{aligned}$$

Accumulation of adatoms

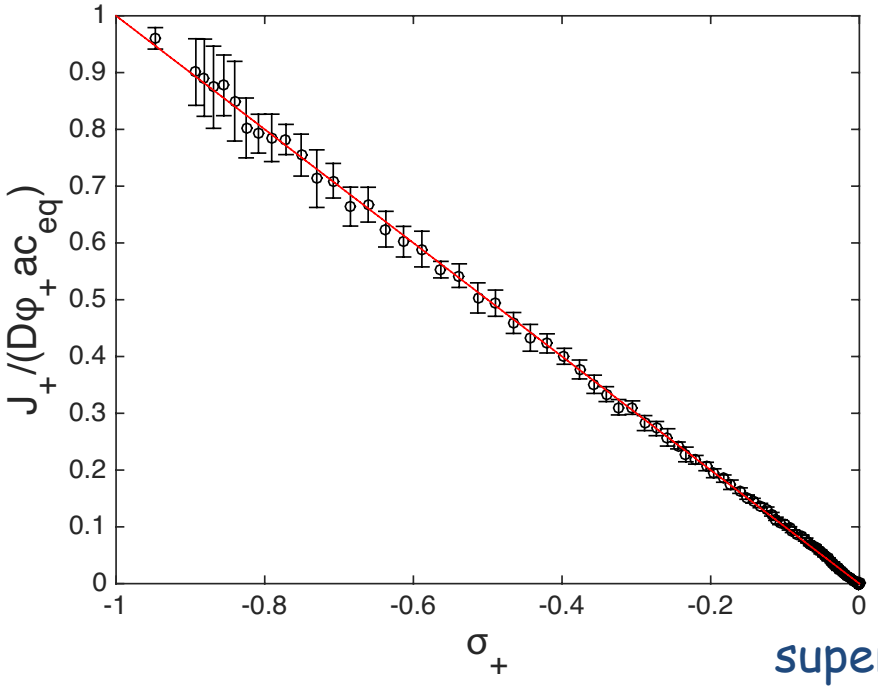
Kinetic Monte Carlo (KMC) simulations: Flux

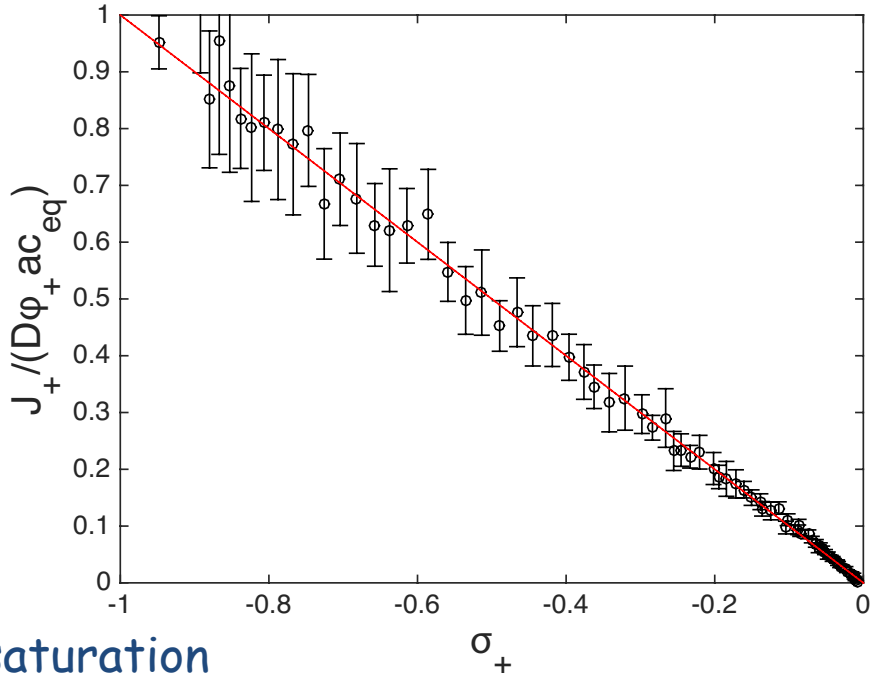
Dilute regime

$F = 0,\ k = 2.5 \times 10^{-3}$

$\epsilon = F/D = 10^{-3}$

$k = 2.5 \times 10^{-3}$





supersaturation

$$\sigma_+ := (c_1 - c^{eq})/c^{eq}$$

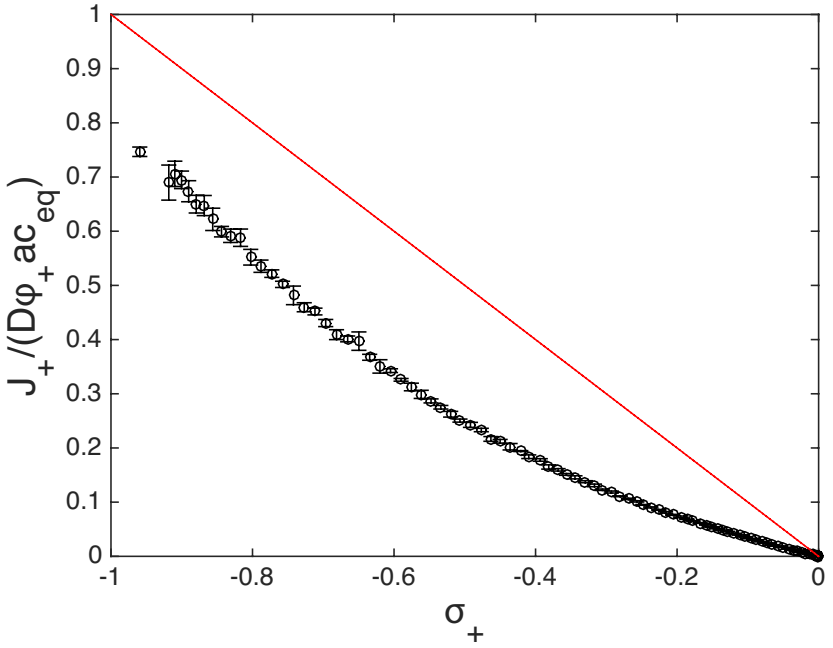
KMC simulations: Flux (cont.)

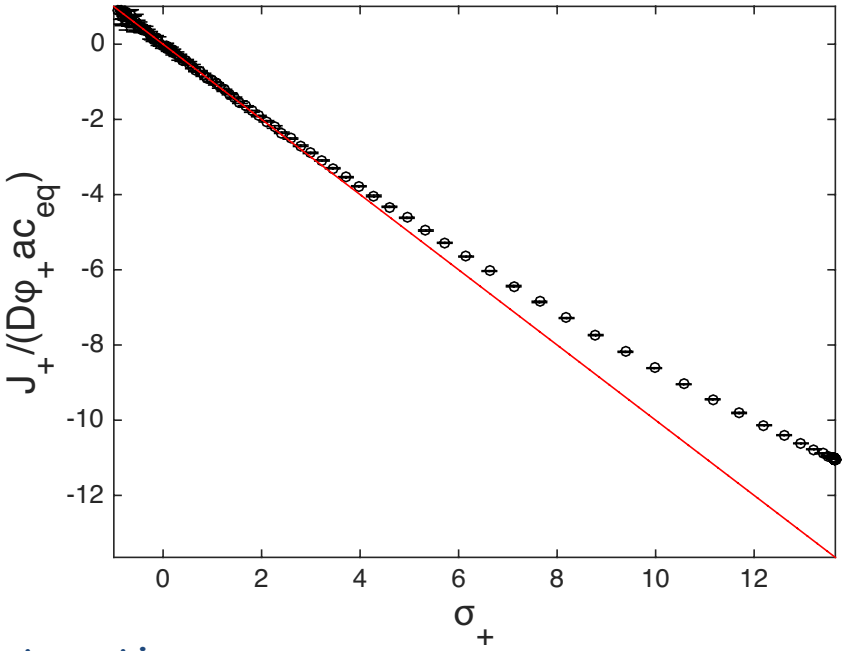
Non-dilute regime

$$\epsilon = F/D = 4 \times 10^{-2}$$

$$F = 0,\ k = 0.2$$

$$k = 2.5 \times 10^{-3}$$





supersaturation

$$\sigma_+ := (c_1 - c^{eq})/c^{eq}$$

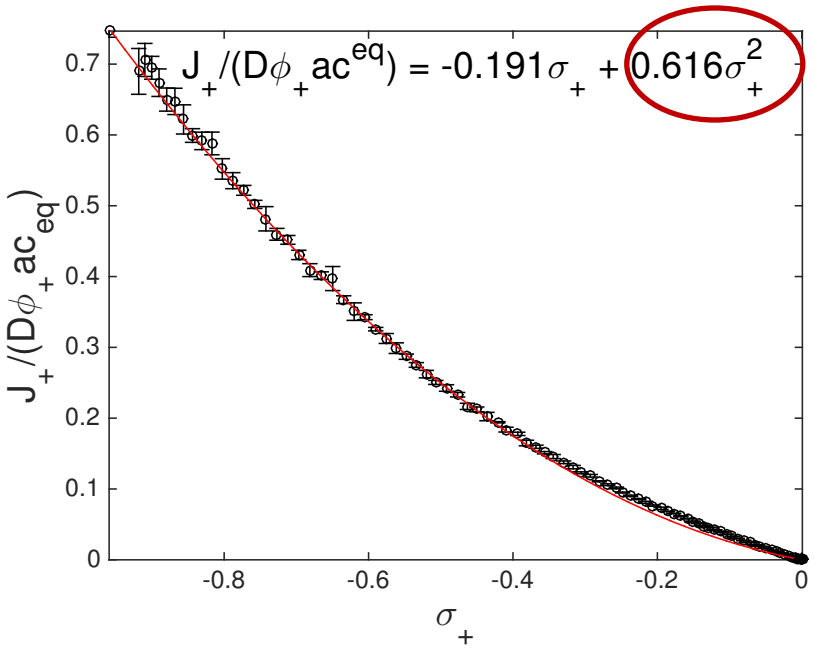
Corrections to BCF model: Fitting to KMC data

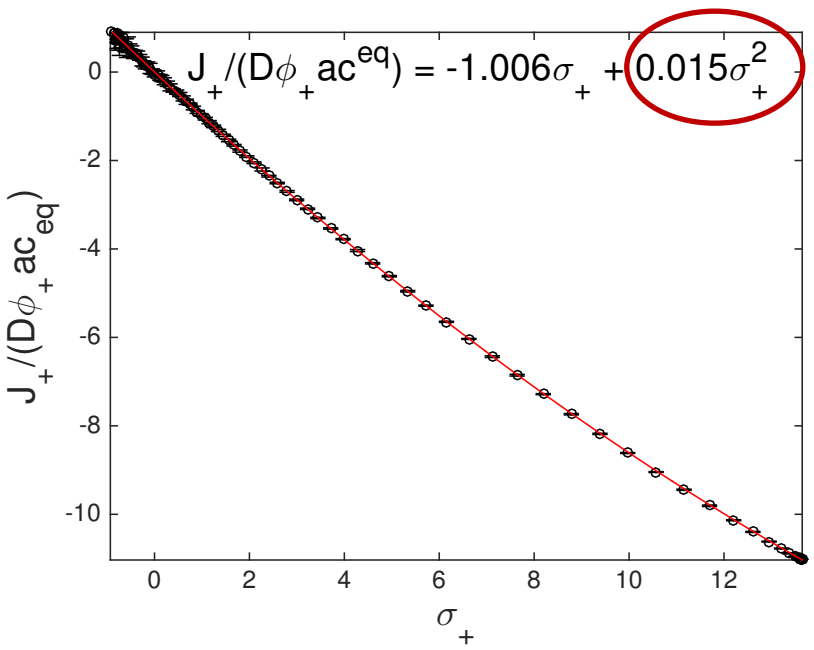
Non-dilute regime

$$\epsilon = F/D = 4 \times 10^{-2}$$

$$F = 0,\ k = 0.2$$

$$k = 2.5 \times 10^{-3}$$





Can one derive explicit formulas for the coefficients?

Extensions to higher dimension (curved steps) ?

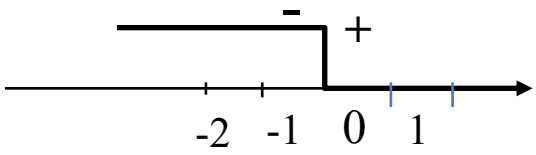
BCF model, corrections, and estimates

[Schneider, Patrone, DM, 2018]

Linear kinetic relation $D\phi_\pm a = O(1)$

$$J_\pm(t) = \mp D\phi_\pm a\,[c_{\pm 1}(t) - c^{eq}] \mp D\phi_\pm a f_\pm(t)$$

correction



correction

Also: $$J_\pm(t) = \mp Da\,[c_{\pm 2}(t) - c_{\pm 1}(t)] \mp Da\left[\hat{R}_{\pm 2}(t) - \hat{R}_{\pm 1}(t)\right]$$

PROPOSITION 2. *For near-equilibrium evolution,* $f_\pm(t)$ *obey*

$$f_\pm(t) \le \mathcal{C}_1 \frac{k}{1-k}\frac{k}{a} + \mathcal{C}_2 \frac{\epsilon N}{(1+\phi_\pm)a} \ .$$

In the same vein, the corrections $\hat{R}_j(t)$ *satisfy*

$$\hat{R}_j(t) \le \mathcal{C}_1 \frac{k}{1-k}\frac{k}{a} + \mathcal{C}_2 \frac{\epsilon N}{a} \qquad (\epsilon = F/D) \ .$$

Dilute regime: $k \le \mathcal{O}(a)$ and $\epsilon N \ll \mathcal{O}(a)$; corrections are negligible

Heuristics of proof (sketch)

- Assume $\max_{\boldsymbol{\alpha}}\{p_{\boldsymbol{\alpha}}(0)/p_{\boldsymbol{\alpha}}^{ss,\epsilon}\} \leq C$.
- Apply "Max. principle" (**Proposition 1**): $p_{\boldsymbol{\alpha}}(t) \lesssim p_{\boldsymbol{\alpha}}^{ss,\epsilon}$.
- Expand $p_{\boldsymbol{\alpha}}^{ss,\epsilon} \sim p_{\boldsymbol{\alpha}}^{ss,(0)} + \epsilon\, p_{\boldsymbol{\alpha}}^{ss,(1)}$, assuming small enough ϵ.
- Consequently, e.g.,

$$|f_+(t)| \lesssim \tilde{f}_+^{ss,\epsilon} \sim kc^{eq} + \frac{1}{a}\left\{\left[kS_1^{(0)} + S_2^{(0)}\right] + \epsilon\left[kS_1^{(1)} + S_2^{(1)}\right]\right\}$$

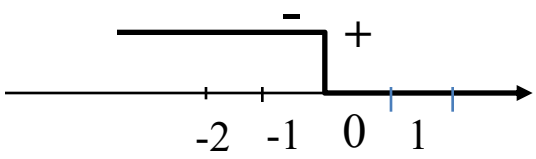
$$S_1^{(l)} := \sum_{\boldsymbol{\alpha}} \mathbb{1}(\nu_{-1}(\boldsymbol{\alpha}) > 0) p_{\boldsymbol{\alpha}}^{ss,(l)} \ , \ S_2^{(l)} := \sum_{\boldsymbol{\alpha}} \mathbb{1}(\nu_1(\boldsymbol{\alpha}) > 1)\nu_1(\boldsymbol{\alpha}) p_{\boldsymbol{\alpha}}^{ss,(l)} \ ; \ l = 0,\, 1 \ .$$

- Compute $S_j^{(0)}$ exactly in closed form; and approximate each $S_j^{(1)}$ by a sum.

$$S_1^{(0)} = \frac{1}{Z}\sum_{n=1}^{\infty}\left(\begin{array}{c} n+N-3 \\ n-1 \end{array}\right) k^n = k \ , \quad S_2^{(0)} = \frac{1}{Z}\sum_{\ell=2}^{\infty} \ell k^l \sum_{n=l}^{\infty}\left(\begin{array}{c} n-\ell+N-3 \\ n-\ell \end{array}\right) k^{n-\ell} = \frac{2k^2-k^3}{1-k} \ .$$

$$|S_j^{(1)}| \lesssim \frac{N}{1+\phi} \ ; \ j = 1,\, 2$$

Scaling limit for averages ($a \to 0$)



Assume $\mathcal{D} := Da^2 = \mathcal{O}(1)$

1. Flux towards edge (Robin boundary condition):

$$\frac{1}{1-k}\,\frac{k}{a} \Rightarrow \boxed{k = \mathcal{O}(a)}$$

$$J_\pm(t) = \mp D\phi_\pm a\,[c_{\pm 1}(t) - c^{eq}] \mp \underbrace{D\phi_\pm a f_\pm(t)}_{\mathcal{O}(a)}$$

Correction (see Proposition 2)

Suppose $\boxed{D\phi_+ a = \mathcal{O}(1)}$ $[\phi_\pm = \mathcal{O}(a)]$ *and* $\boxed{\epsilon = \mathcal{O}(a^3)}$ ⇒ $\mathcal{O}(a)$

BCF model emerges

2. Diffusion on terrace, $\rho_j(t) \to \rho(x,t)$

$$J_\pm(t) = \underbrace{\mp Da\,[c_{\pm 2}(t) - c_{\pm 1}(t)]}_{\to\, -\mathcal{D}\,\partial_x \rho} \mp \underbrace{Da\left[\hat{R}_{\pm 2}(t) - \hat{R}_{\pm 1}(t)\right]}_{\mathcal{O}(a)}$$

$$\underbrace{\dot{\rho}_j(t)}_{\mathcal{O}(a) \text{ if } t = \mathcal{O}(1/a)} = \underbrace{D\,[\rho_{j-1}(t) - 2\rho_j(t) + \rho_{j+1}(t)]}_{\to\, \mathcal{D}\,\partial_{xx}\rho} + \frac{F}{(N-1)a} - \underbrace{D\,[R_{j-1}(t) - 2R_j(t) + R_{j+1}(t)]}_{\mathcal{O}(a)}$$

Epilogue

- By a model of master eq. in 1+1 dims, BCF description arises as low-supersaturation limit adatom gas.

- For high enough supersaturation, linear kinetic relation for adatom flux acquires (nonlinear) corrections.

- These corrections have been indicated by KMC simulations.
  Incomplete analytical understanding.

- Model used so far implies regime near diluteness. Improvements?

- Stochastic fluctuations in step motion? [Lu, Liu, DM, 2015]

- Need to study 2+1 dims, allowing for island formation.