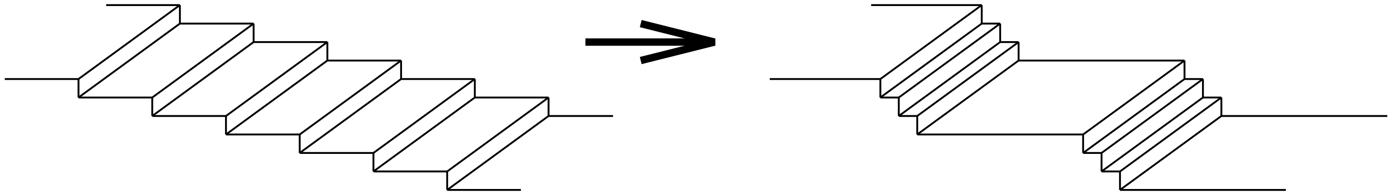
Universality Classes of Step Bunching?



- Mechanisms of step bunching
- Step-dynamical equations and continuum limit
- Scaling properties of a single bunch
- Towards a global scaling picture

Joint work with V. Tonchev, S. Stoyanov and A. Pimpinelli

Nonequilibrium mechanisms for step bunching

- Ehrlich-Schwoebel-barriers in sublimation — R.L. Schwoebel (1969)
- Surface electromigration — S. Stoyanov (1991)

in growth:

- Pinning of steps by impurities — N. Cabrera, D.A. Vermilyea (1958)
- Step edge diffusion — P. Politi, J.K. (2000)
- Chemical precursors [e.g. GaAs] — A. Pimpinelli, A. Videcoq (2000)
- Dimer mobility — M. Vladimirova, A. De Vita, A. Pimpinelli (2001)
- Impurity-induced mobility gradients — J.K. (2002)
- Anisotropic diffusion [e.g. Si(001)] — J. Mysliveček et al. (2002)

Issues:

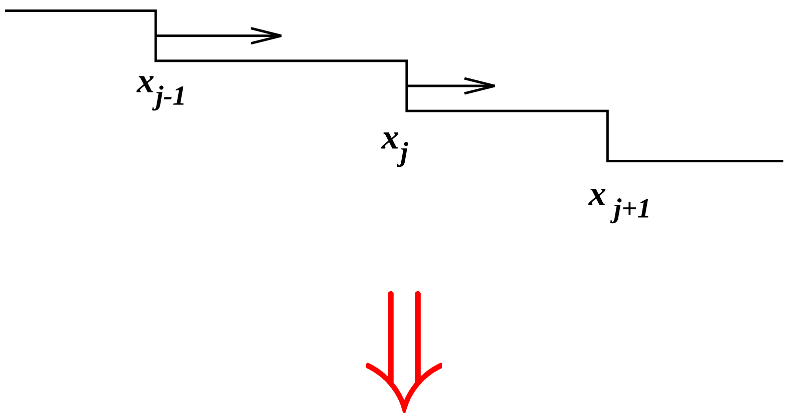
- shape and scaling of individual bunches
- global evolution of the morphology (coarsening)

Levels of description:

- step evolution equations:

$$\frac{dx_j}{dt} = f_+(x_{j+1} - x_j) + f_-(x_j - x_{j-1})$$

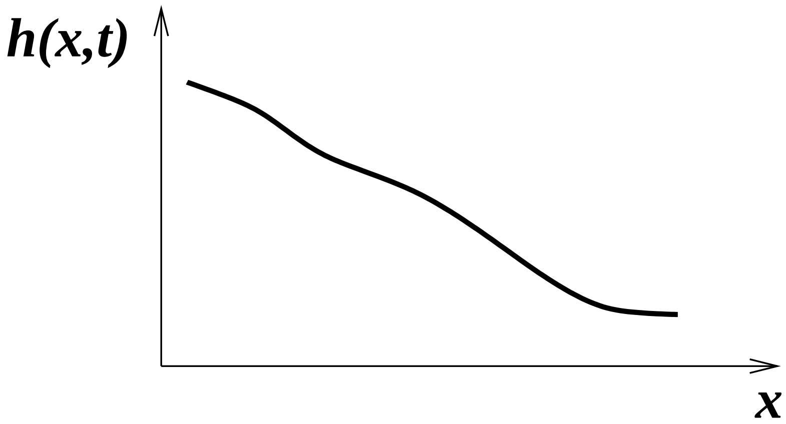
[BCF 1951; Schwoebel & Shipsey 1966]



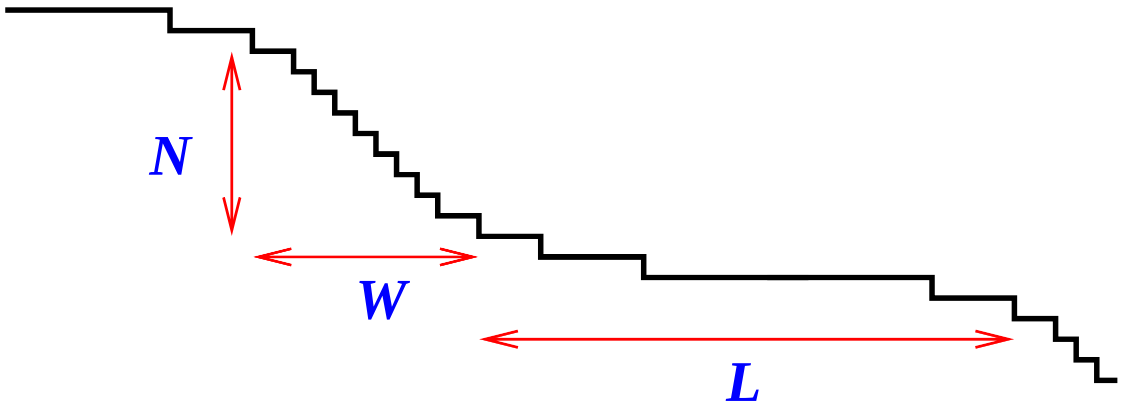
- continuum height equation:

$$\frac{\partial h}{\partial t} = \mathscr{F}(\nabla h, \nabla^2 h, \ldots)$$

[Mullins 1959; Villain 1991]



Scaling properties of step bunches



- bunch height
  $N \sim W^{\alpha}, \; \alpha > 1$
  $N \sim L \sim t^{\beta}$

- minimal terrace size:
  $l_{\min} \sim N^{-\gamma} \sim W/N$
  $\Rightarrow \gamma = 1 - 1/\alpha$

Experiments for electromigration-induced step bunching on Si(111):

- $\gamma \approx 2/3$ [Fujita et al., Phys. Rev. B 60, 16006 (1999)]

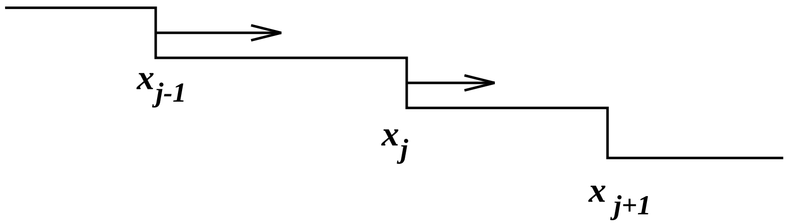
- $\beta \approx 1/2$ [Yang et al., Surf. Sci. 356, 101 (1996)]

Goal: Consistent derivation of power laws **and prefactors**

Stability of step trains

- step evolution:

$$\frac{dx_j}{dt} = f_+(x_{j+1} - x_j) + f_-(x_j - x_{j-1})$$



$f_\pm$: flux from lower/upper terrace

- homogeneous step train: $x_j^{(0)} = [f_+(l) + f_-(l)]t + jl$ $\quad l$: step spacing

  linear stability analysis: $x_j(t) = x_j^{(0)} + \varepsilon_j(t) \quad \Rightarrow \quad \varepsilon_j(t) \sim e^{i\phi j + \omega t}$ with

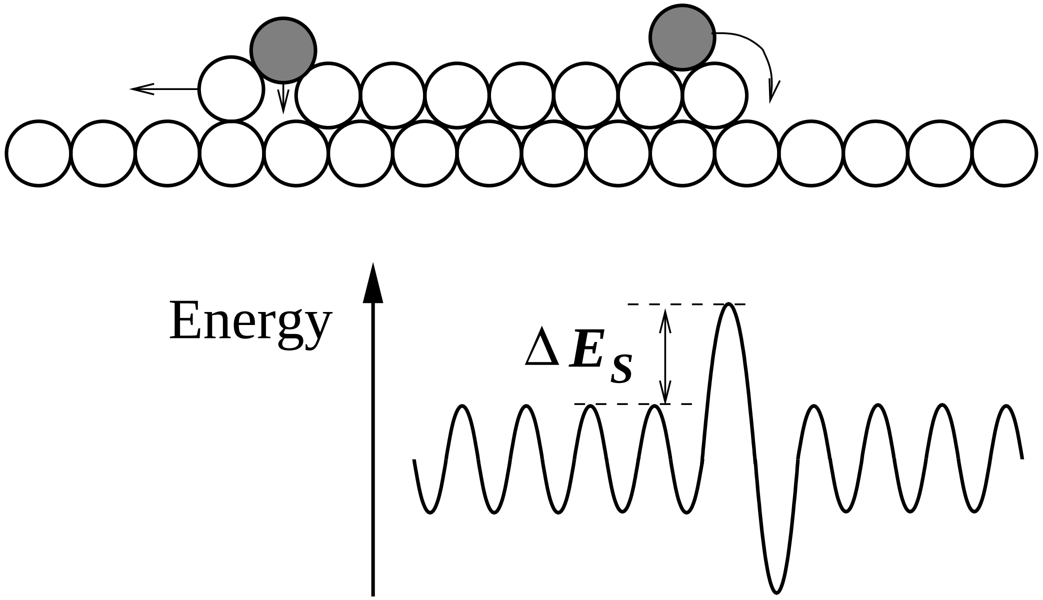
$$\mathscr{R}(\omega(\phi)) = -(1 - \cos\phi)[f'_+(l) - f'_-(l)]$$

  $\Rightarrow$ step train is stable iff $f'_+(l) - f'_-(l) > 0$

- step bunching during growth requires preferential attachment from the **upper** terrace

The Ehrlich-Schwoebel effect

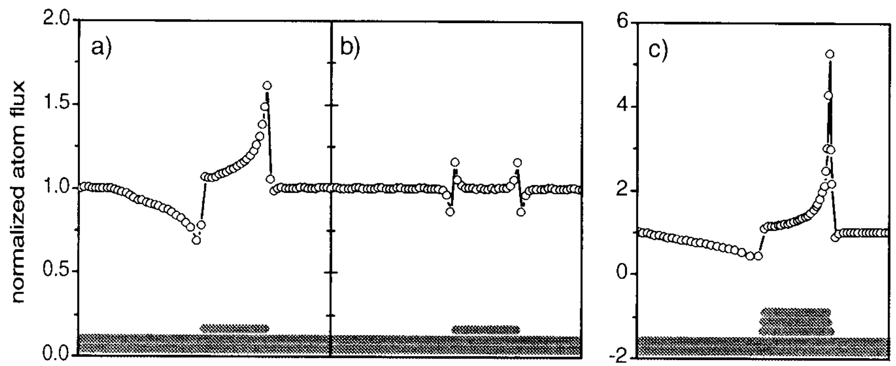
[G. Ehrlich, F. Hudda (1966); R.L. Schwoebel, E.J. Shipsey (1966)]



- Additional energy barrier suppresses adatom descent across step edges
- Preferential attachment to step edges from the **lower** terrace
  $\Rightarrow$ **stabilization** during growth, **destabilization** during sublimation

Step bunching by steering

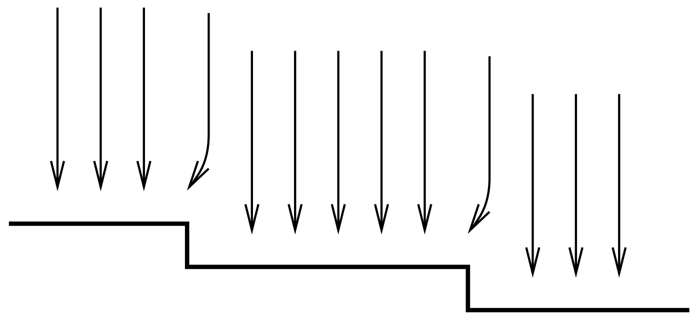
- **Steering:** Attraction of incident atoms to the substrate implies inhomogeneous deposition flux



Lennard-Jones trajectories for Cu(100)

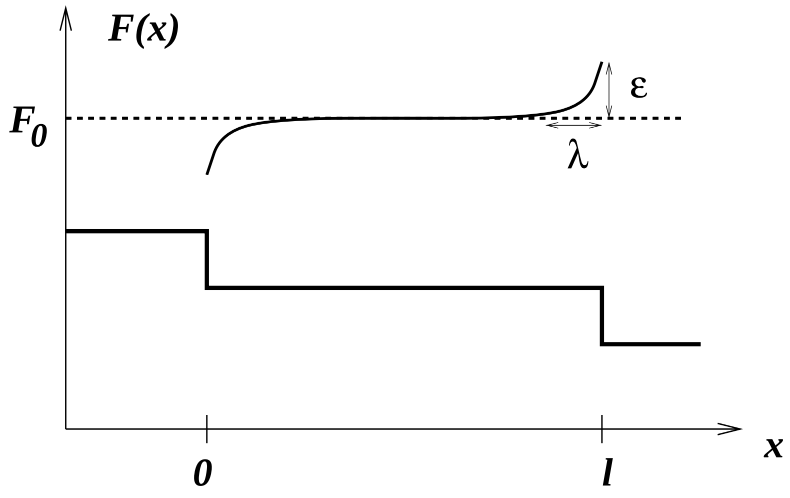
[van Dijken et al., PRB 61, 14047 (2000)]

At **vicinal surfaces** steering leads to enhanced deposition near descending steps



- Flux model:

$$F(x) = F_0[1 - \varepsilon e^{-x/\lambda} + \varepsilon e^{-(l-x)/\lambda}]$$



- Burton-Cabrera-Frank (BCF) equation for stationary adatom density $n(x)$:

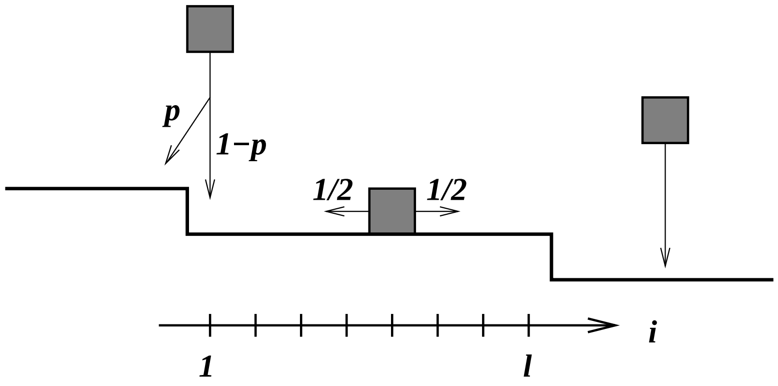
$$Dn'' + F(x) = 0 \quad \text{with boundary conditions} \quad n(0) = n(l) = 0$$

$$\Rightarrow \quad f_+ - f_- = -2\varepsilon[\lambda(1 + e^{-l/\lambda}) - 2(\lambda^2/l)(1 - e^{-l/\lambda})] \approx -2\varepsilon[\lambda - 2(\lambda^2/l)]$$

$$\Rightarrow \quad f'_+(l) - f'_-(l) < 0,$$ instability for all l

- Time scale for bunching: $\theta_c \approx (8\varepsilon)^{-1}(l/\lambda)^2$ ML

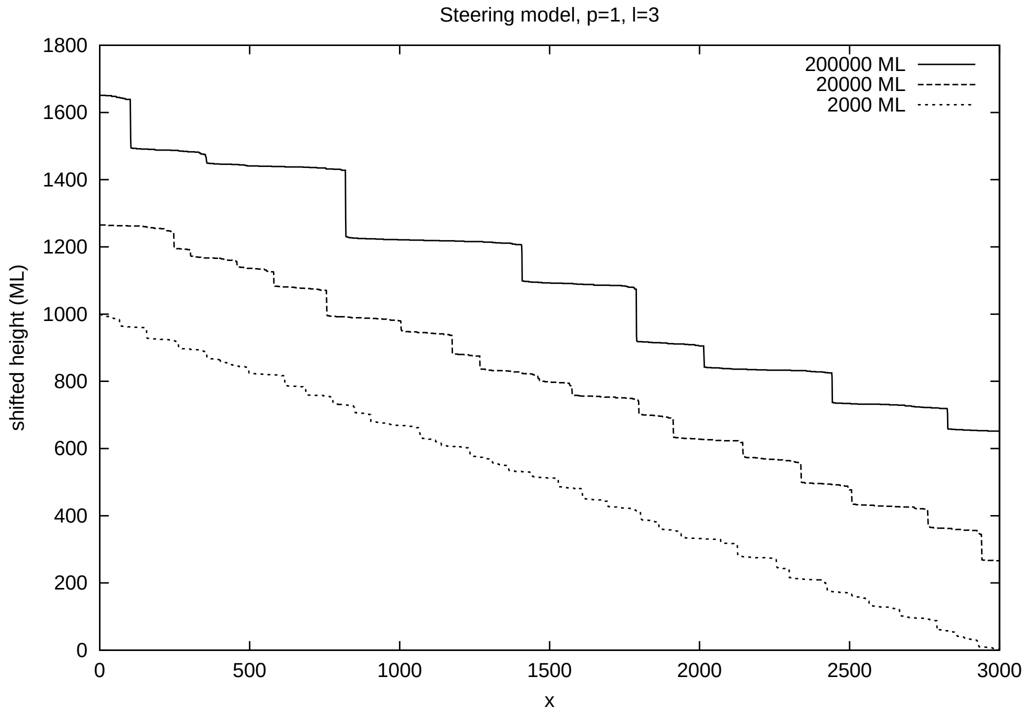
One-dimensional stochastic model



- Atom deposited at site i reaches upper step with probability $P_i = 1 - (i-1)/l$

- Atoms deposited at $i = 1$ are "deflected" with probability p

- Simulation with $l = 3$ and $p = 1$:



Step bunching during sublimation

- BCF equation for the stationary adatom density $n(x)$:

$$D\frac{d^2n}{dx^2} - \frac{n}{\tau} = 0 \qquad \text{b.c.}: \quad D\frac{dn}{dx}(x_i) = \pm k_\pm [n(x_i) - n_{\text{eq}}(x_i)]$$

τ: adatom lifetime $\qquad k_\pm$: attachment rates to ascending/descending step

- step-step interactions: $n_{\text{eq}}(x_i) = n^0_{\text{eq}} \exp[\beta\Delta\mu(x_i)]$ with $\beta = 1/k_\text{B}T$ and

$$\beta\Delta\mu(x_i) = -\left(\frac{l_0}{x_{i+1} - x_i}\right)^3 + \left(\frac{l_0}{x_i - x_{i-1}}\right)^3$$

$l_0 = (2\Omega\beta g)^{1/3}$: interaction length $\qquad g$: step repulsion coefficient

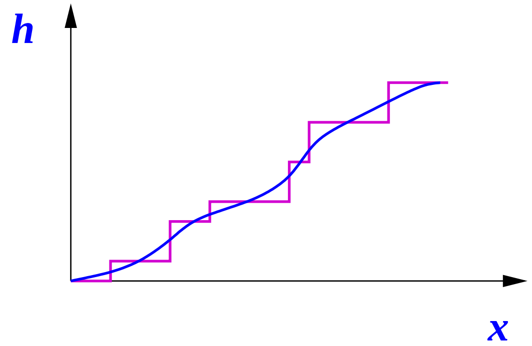
- additional lengths: $\lambda_D = \sqrt{D\tau}$ diffusion length $\qquad d = D/k_-$ kinetic length
- Ehrlich-Schwoebel parameter: $S = k_-/k_+ = \exp[-\beta\Delta E_\text{S}] < 1$

- For $x_i - x_{i-1} \ll d$ and $x_i - x_{i-1} \ll \lambda_D$ step dynamics become **linear**:

$$\frac{dx_i}{dt} = \frac{D\Omega n_{\rm eq}^0}{\lambda_D^2}\left[\frac{S}{1+S}(x_{i+1}-x_i) + \frac{1}{1+S}(x_i - x_{i-1})\right] \quad + \text{interaction terms}$$

$\Rightarrow$ exact continuum limit for slowly varying profiles: [J.K. (1997)]

$$\frac{\partial h}{\partial t} + \frac{\partial J}{\partial x} = -Rh_0$$



$$J = -\frac{Rh_0^2(1-S)}{2(1+S)}\frac{1}{m} \qquad - \qquad \frac{Rh_0^3}{6m^3}\frac{\partial m}{\partial x} \qquad + \qquad \frac{3\Omega l_0^3 D n_{\rm eq}^0 S}{2d(1+S)m}\frac{\partial^2 m^2}{\partial x^2}$$

destabilizing **symmetry-breaking** **stabilizing**

$m = \partial h / \partial x > 0$ slope $\quad R = \Omega n_{\rm eq}^0/\tau$ desorption rate $\quad h_0$ monolayer height

Step bunching by surface electromigration

- BCF equation with an electromigration force f [Stoyanov, 1990]:

$$D\frac{d^2n}{dx^2} - \beta D f \frac{dn}{dx} - \frac{n}{\tau} = 0 \qquad \text{b.c.}: \quad D\frac{dn}{dx} - \beta D f n\Big|_{x=x_i} = \pm k[n - n_{\text{eq}}]\Big|_{x=x_i}$$

$\Rightarrow$ linear step equations in the attachment-limited regime:
[Liu & Weeks, 1998]

$$\frac{dx_i}{dt} = \frac{D\Omega n^0_{\text{eq}} f}{2dk_{\text{B}}T}\left(x_{i+1} + x_{i-1} - 2x_i\right) + R(x_{i+1} - x_{i-1}) \quad + \quad \text{interaction terms}$$

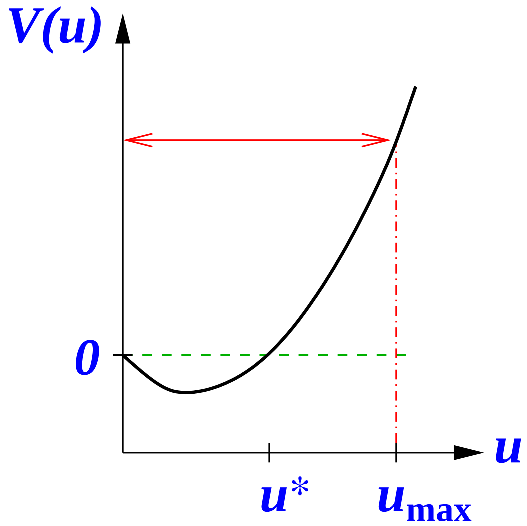
$\Rightarrow$ continuum equation:

$$\frac{\partial h}{\partial t} + \frac{\partial}{\partial x}\left[\frac{Dn^0_{\text{eq}} h_0 f}{2dk_{\text{B}}T}\frac{1}{m} - \frac{Rh_0^3}{6m^3} + \frac{3\Omega l_0^3 D n^0_{\text{eq}}}{4dm}\frac{\partial^2 m^2}{\partial x^2}\right] = -Rh_0$$

destabilizing for $f < 0$

- Neglecting the symmetry-breaking term $\sim m^{-3}\partial m/\partial x$, the stationarity condition $J(x) \equiv J_0$ takes the form of Newton's equation for $u = m^2$:

$$K\,\frac{d^2u}{dx^2} = \sqrt{u}(J_0 + B/\sqrt{u}) = -V'(u), \quad V(u) = -\frac{2}{3}J_0 u^{3/2} - Bu$$



- Bunch shape corresponds to trajectory $0 \le u(x) \le u_{\max}$
- Two types of trajectories: $K(du/dx)^2/2 + V(u) \le 0$ or > 0
- How to fix J_0?

- Liu & Weeks [PRB 57, 14891 (1998)]: $J_0 = -\frac{1-S}{2(1+S)}Rh_0 l < 0$

$\Rightarrow$ current remains at its initial value throughout the bunching process

$\Rightarrow$ bunch is described by trajectories with $u_{\max} \gg u^*$

- Scaling law for minimal terrace size l_{min} in a bunch of N steps:

$$\frac{l_{\text{min}}}{l} = 2^{4/3}\left(\frac{S}{1-S}\right)^{1/3}\left(\frac{l}{d}\right)^{1/3}\left(\frac{\lambda_D}{l}\right)^{2/3}\left(\frac{l_0}{l}\right) N^{-2/3}$$

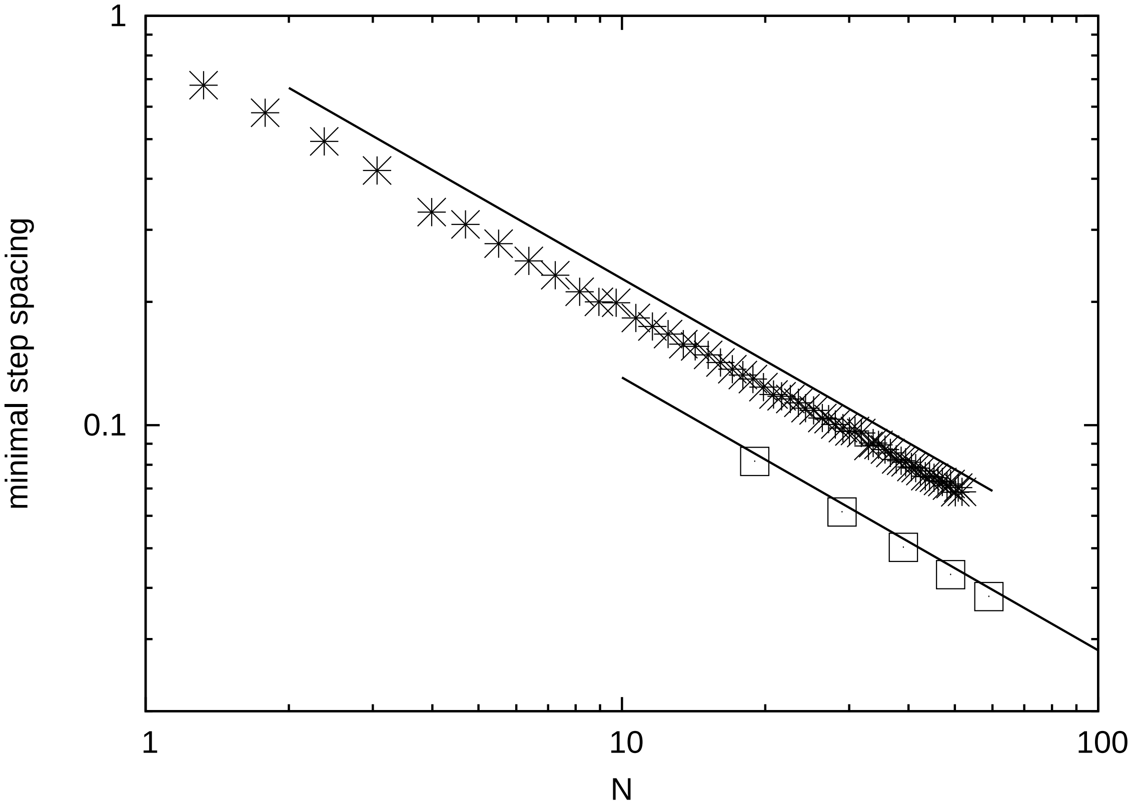
Numerical simulations:

(V. Tonchev)

$\lambda_D/l = d/l = 100$

$*$ $S = 0.3$, $l_0/l = 0.12$
500 steps

$\square$ $S = 0.01$, $l_0/l = 0.24$
single bunches



Universality classes of step bunching?

- Generic continuum equation for step bunching:
  [Pimpinelli et al., PRL 88, 206103 (2002)]

$$\frac{\partial h}{\partial t} + \frac{\partial J}{\partial x} = \text{const.}, \qquad J = B\, m^{\rho} + K\, m^{-k} \frac{\partial^2}{\partial x^2} m^n$$

with $B\rho > 0$, $K > 0$ **destabilizing** **stabilizing**

$k = 0/1$: diffusion limited/attachment limited kinetics

n: exponent of step-step interaction $[V_{\text{step}}(l) \sim l^{-n}]$

- Postulate invariance of $h(x,t)$ under scale transformation

$$h(x,t) \;\rightarrow\; b^{-\alpha}\, h(bx, b^z t)$$

$\Rightarrow \alpha = 1 + 2/(n-k-\rho)$, $z = 2(1+n-k-2\rho)/(n-k-\rho)$, $\beta = \alpha/z$

- For sublimation and electromigration $\rho = -1$, $k = 1$

$$\Rightarrow \quad \alpha = \frac{n+2}{n}, \quad \beta = \frac{1}{2}, \quad \gamma = \frac{2}{2+n}$$

- For $n = 2$ this implies $\gamma = 1/2$, $l_{\mathrm{min}} \sim N^{-1/2}$???

- The scaling argument for l_{min} fails because in the stationarity condition $J(x) \equiv J_0$ the destabilizing current is **irrelevant** compared to the mean current J_0 when $\rho < 0$ $\quad \Rightarrow \quad$ effectively $\rho = 0$ $\quad \Rightarrow \quad \gamma = 2/(n+1)$

- On the other hand, the numerics is consistent with the predictions of the scaling theory for α and β with $\rho = -1$

  $\Rightarrow$ violation of the "obvious" scaling relation $\gamma = 1 - 1/\alpha$!!!!

- Local and global properties described by **different** continuum equations ?

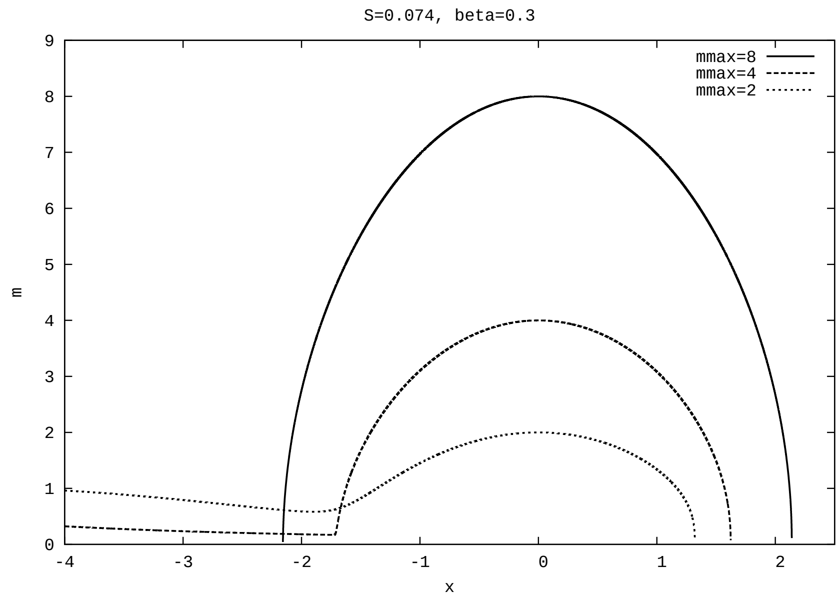
Bunch asymmetry and bunch motion

- Bunches are distinctly asymmetric:
  $l_{\text{first}} \sim N^{-1/3}$ as predicted by continuum theory, but $l_{\text{last}} \sim N^0$

- Asymmetry due to symmetry-breaking term is much too weak and of the wrong sign:

Numerically integrated bunch shape for different values of the maximal slope

$S = 0.3, \lambda_D/l = d/l = 100, l_0/l = 0.12$



- Bunch asymmetry reflects the accelerating trajectories of steps escaping from one bunch and attaching to the next