Crystal Growth: Physics, Technology and Modeling

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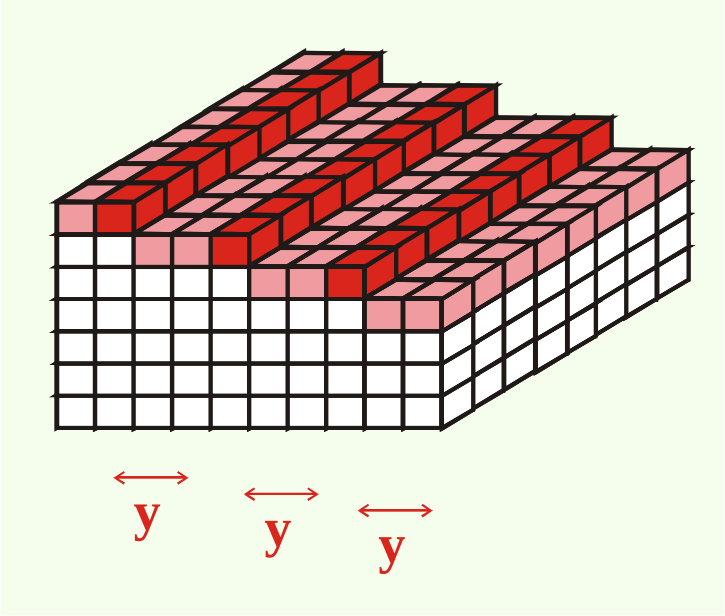
Lecture 7. Step motion

http://w3.unipress.waw.pl/~stach/cg-2022-23/

Scope

- **Vicinal surface - step trains**
- **Step meandering**
- **Step bunching**
- **Double steps**
- **Macro steps**
- **Mounds & Islands**

Vicinal surface – step trains

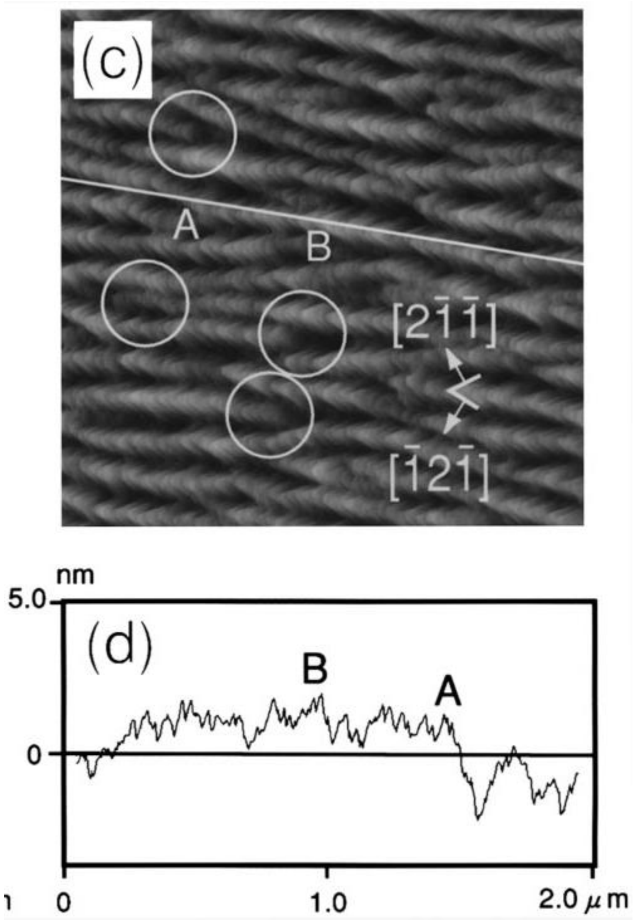


Misorientation

$$\mathbf{a} = y\, tg(\alpha) \quad \Rightarrow \quad \alpha = atan\left(\frac{a}{y}\right)$$

Step instabilities - meandering

- **Si(111) surface deposition**



H. Omi, T. Ogino, Thin Solid Films 380 (2000) 15

- **GaN(0001) - MOVPE**

AFM – G. Nowak

S. Krukowski et al. Cryst. Res. Technol. 42 (2007) 1281

Step fluctuations - equilibrium

- **Drumhead – elastic model**

$$F = \int dx \left[\frac{G}{2}\left(\frac{\partial z}{\partial x}\right)^2 + Uz^2\right] = \sum_k \left[\frac{Gk^2}{2} + U\right] z_k^2$$

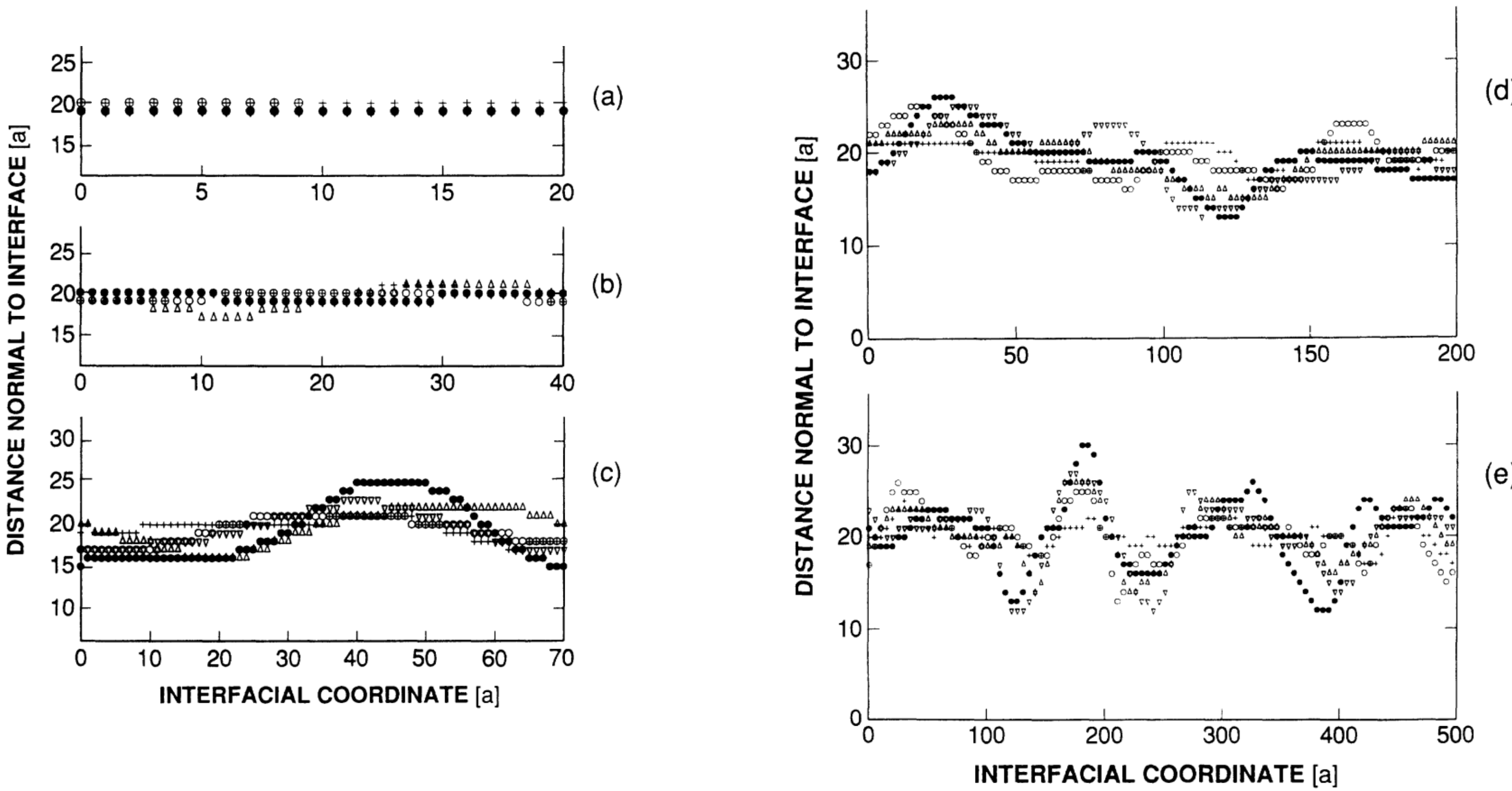
z – deviation from the straight line (z = 0)

- **Average width**

$$\langle z^2 \rangle = \frac{\int dz\, z^2\ exp(-F/kT)}{\int dz\ \ exp(-F/kT)} \cong \int \frac{2\pi k}{Gk^2 + 2U} dk$$

$$\langle z^2 \rangle \cong \pi\, ln\left(\frac{G}{2}\left(\frac{\pi}{2L}\right)^2 + U\right) = 2 \lim_{L\to\infty, U\to 0} ln(L) = \infty$$

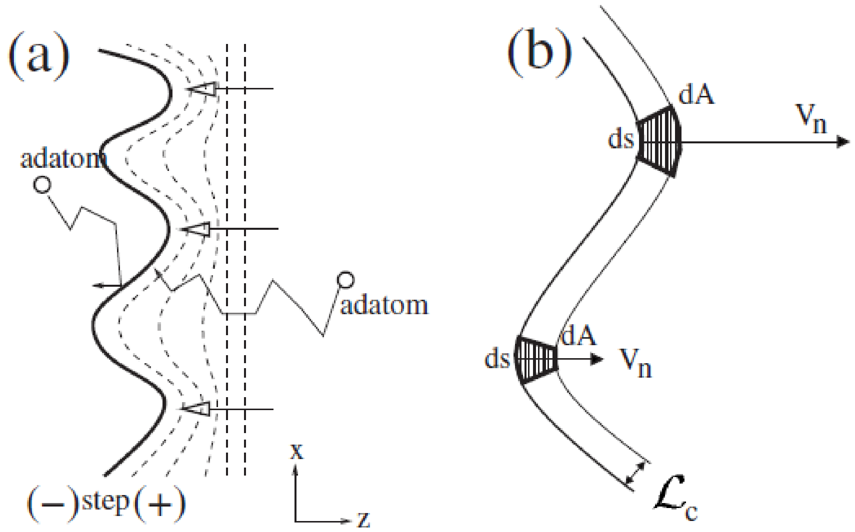
Step fluctuations - Monte Carlo equilibrium simulations



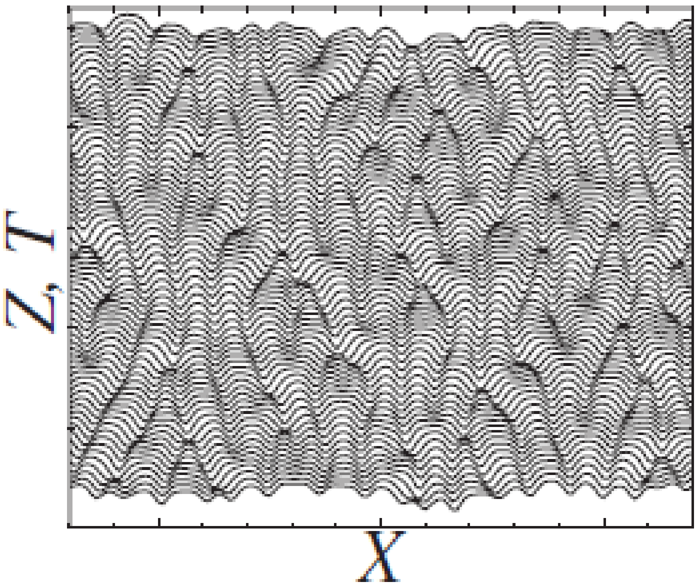
S. Krukowski, F. Rosenberger,
Phys. Rev. B 49 (1994) 12 464

Step meandering mechanism

- **Ehrlich-Schwoebel effect**



- **K-S instability**

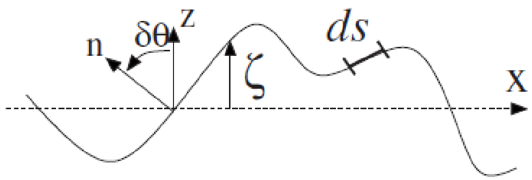


C. Misbah, O. Pierre-Luis, Y. Saito Rev. Mod. Phys. 82(2010) 981

Step fluctuations in nonequilibrium - Kuramoto-Sivashinski equation

- **Deviation from straight step - $\zeta = \zeta(x, t)$ in BCF solution**

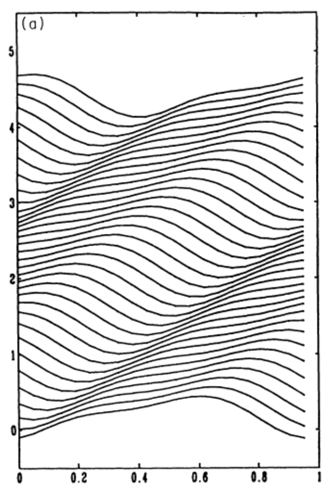
$$\frac{\partial \zeta}{\partial t} = -a\,\frac{\partial^2 \zeta}{\partial x^2} - b\,\frac{\partial^4 \zeta}{\partial x^4} + v\left(\frac{\partial \zeta}{\partial x}\right)^2$$

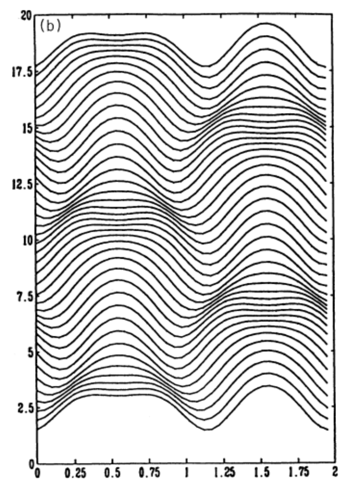


Lyapunov stability

$$\zeta = \zeta_o exp(\lambda t - ikx) \qquad \lambda = ak^2 - bk^4 + f(k) = \begin{cases} > 0 & unstable \\ < 0 & stable \end{cases}$$

Broken parity travelling mode





Vacillating-breathing mode

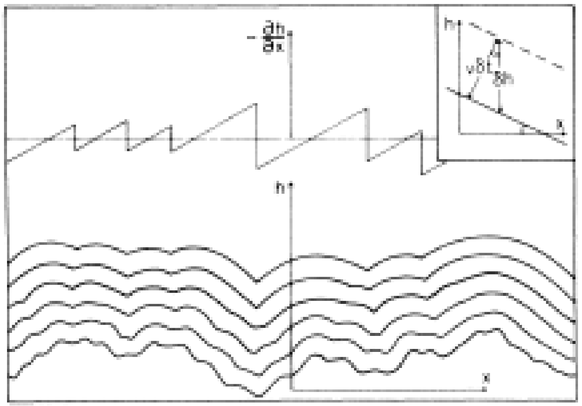
I. Bena, C. Misbah, A. Valence, Phys. Rev. B 47 (1993) 7408

Step fluctuations in nonequilibrium – Kardar-Parisi-Zhang (KPZ) equation

- **Deviation from straight step - $\boldsymbol{h = h(x,t)}$**

$$\frac{\partial h}{\partial t} = \nu \frac{\partial^2 h}{\partial x^2} + \frac{\lambda}{2}\left(\frac{\partial h}{\partial x}\right)^2 + \eta(x,t)$$

$\boldsymbol{\eta(x,t)}$ - random force

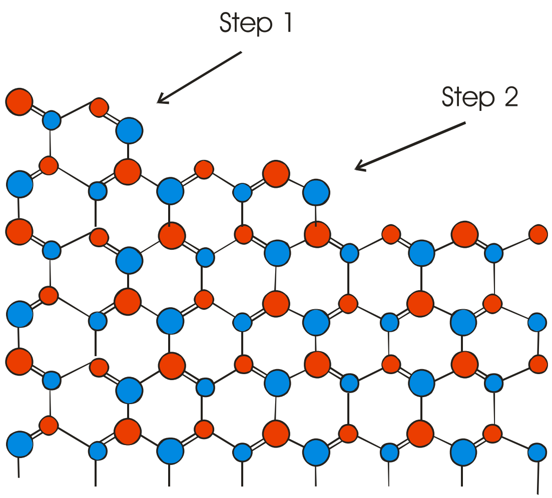


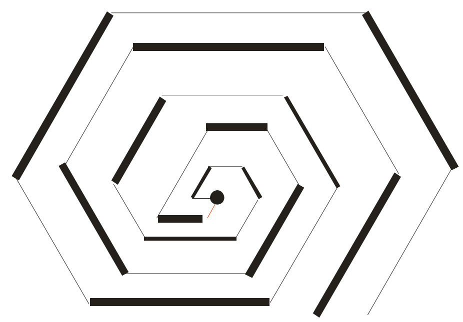
Successive profiles obtained from KPZ equation

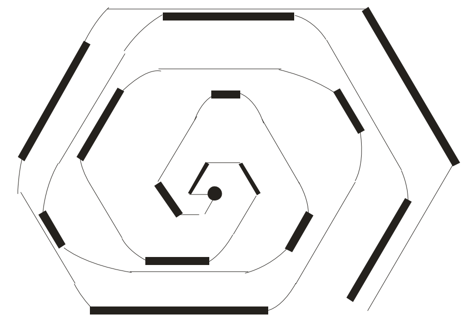
Inset – perpendicular growth mechanism

M. Kardar, G. Parisi, Y-C. Zhang
Phys. Rev. Lett. 56 (1986) 889

Step difference related meandering mechanism



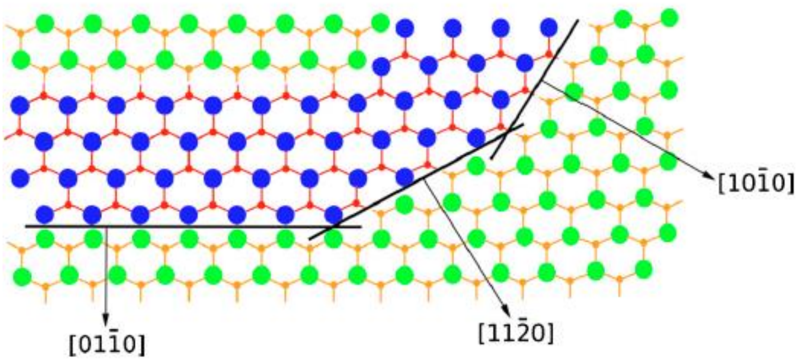


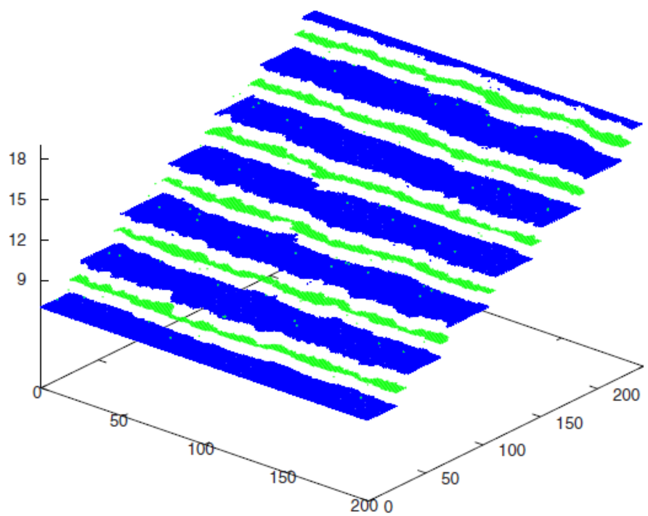


S. Krukowski, et al.
Cryst. Res. Technol. 42 (2007) 1281

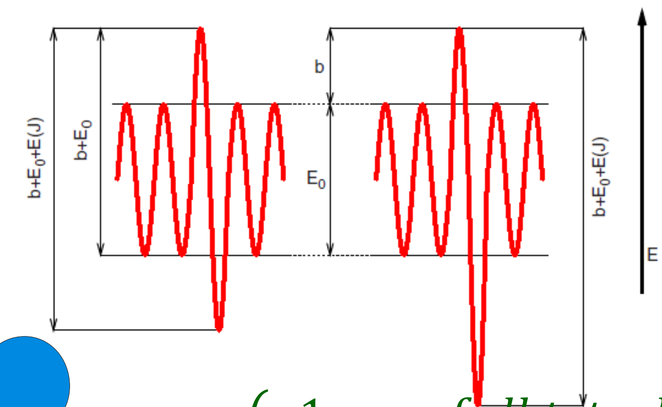
Monte Carlo simulation of surface diffusion – GaN(0001) surface

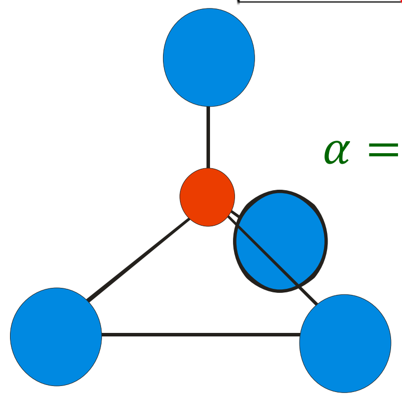
- **Step structures**





- **Ehrlich-Schwoebel effect - E_{bar}**





$$\alpha = \begin{cases} 1 & full\ tetrahedron \\ \frac{1}{3} rn & not\ full\ tetrahedron \end{cases}$$

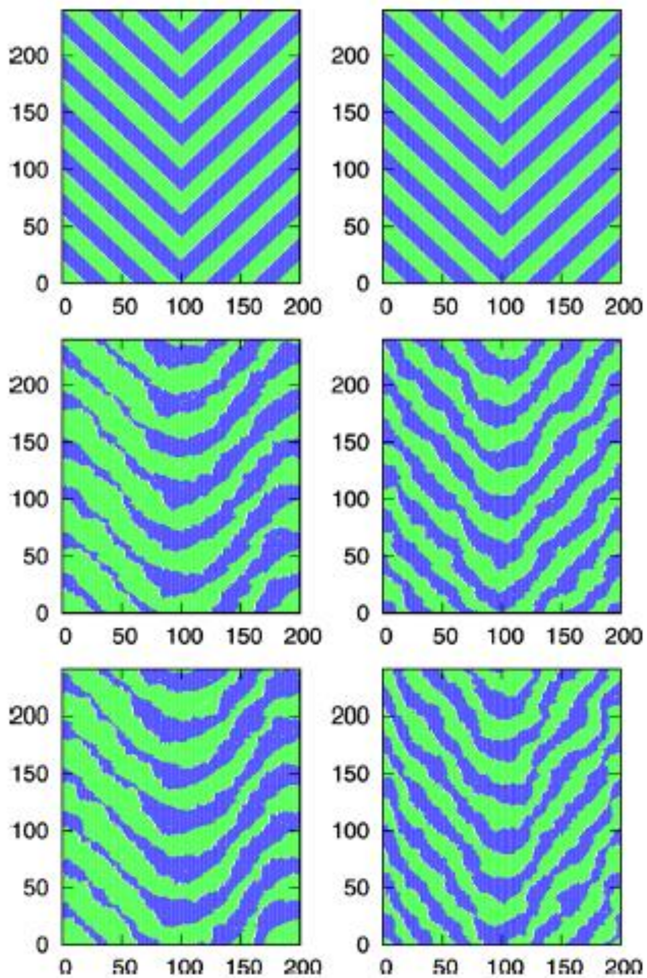
$$E = \phi \sum_{i=1}^{4} \alpha_i$$

$$P = \nu\ exp\left(-\frac{\Delta E + E_{bar}}{kT}\right)$$

M. Załuska-Kotur et al.
J. Appl. Phys. 109 (2011) 023515

MC simulation – GaN(0001) surface – difference in dynamics

$r = 0.4$ $r = 1$



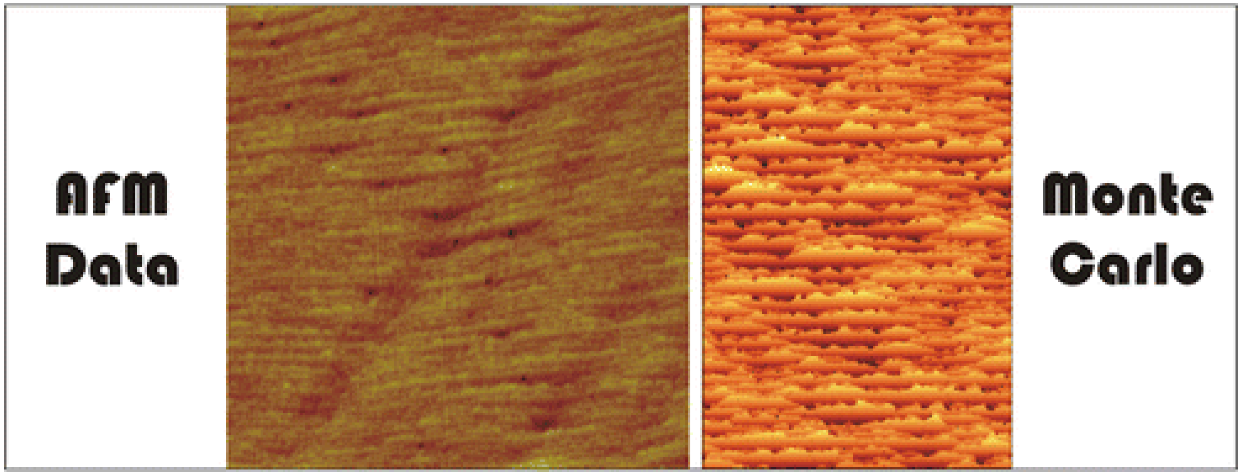
$t = 0\ MC\ steps$

$t = 5 \times 10^6\ MC\ steps$

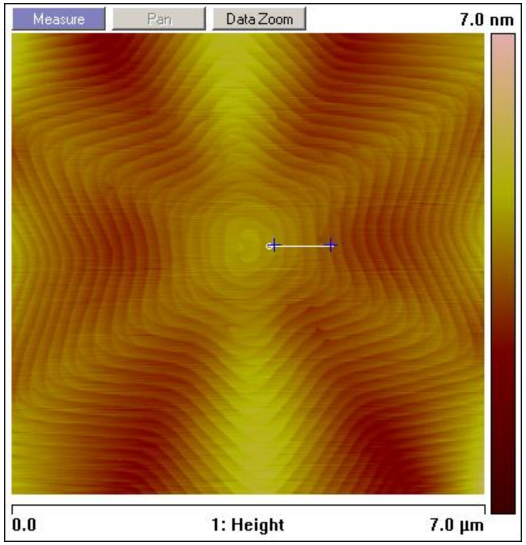
$t = 1 \times 10^7\ MC\ steps$

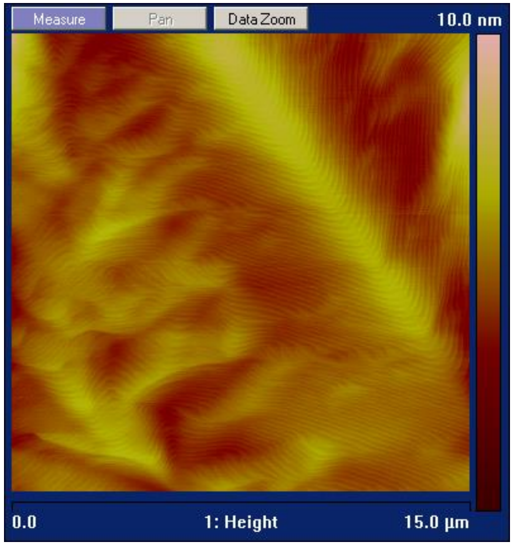
M. Załuska-Kotur et al.
J. Appl. Phys. 109 (2011) 023515

MOVPE growth and MC simulations of GaN(0001) surface



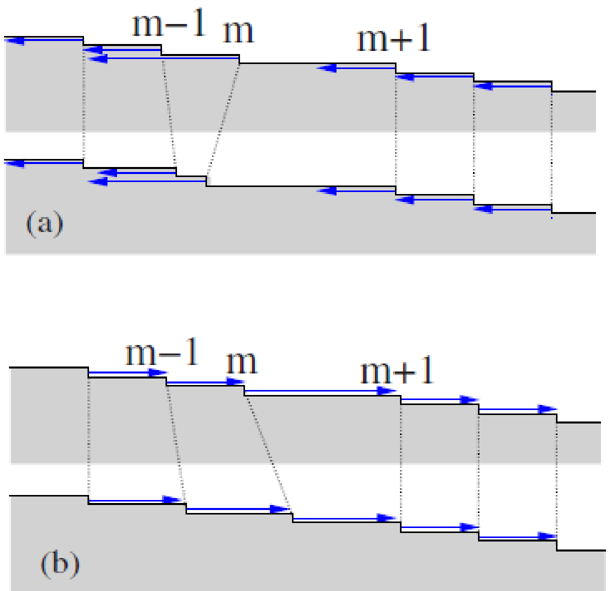
M. Załuska-Kotur et al.
Cryst. Growth Des.
13 (2013) 1006

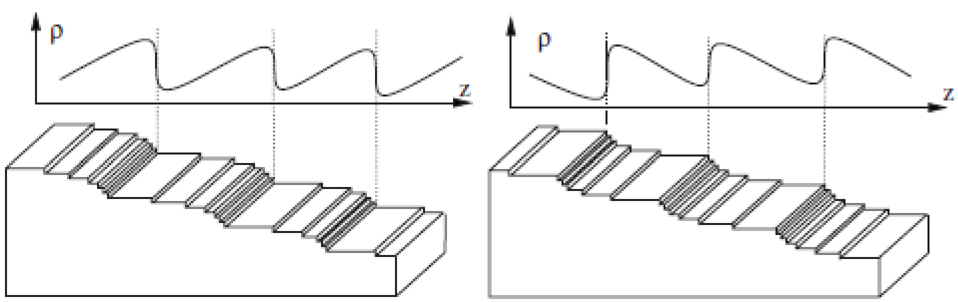




R. Czernecki, AFM data

Step bunching

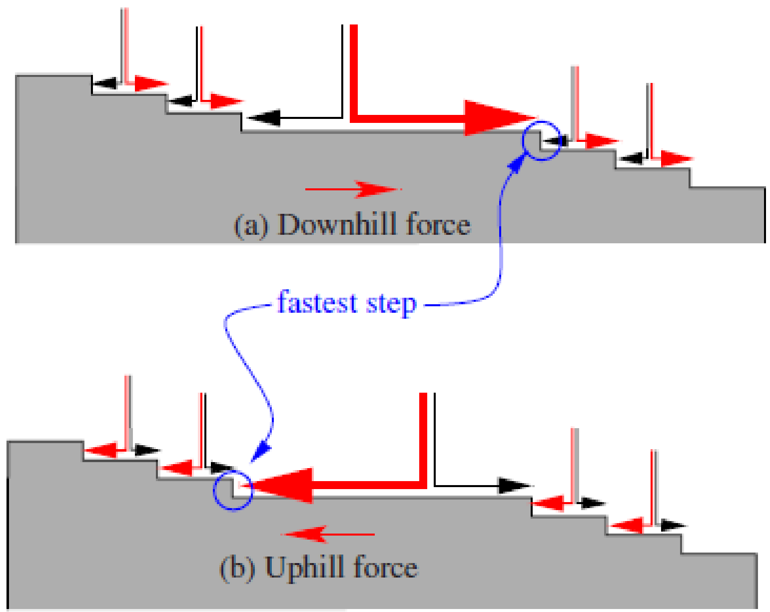




- **Ehrlich-Schwoebel effect**

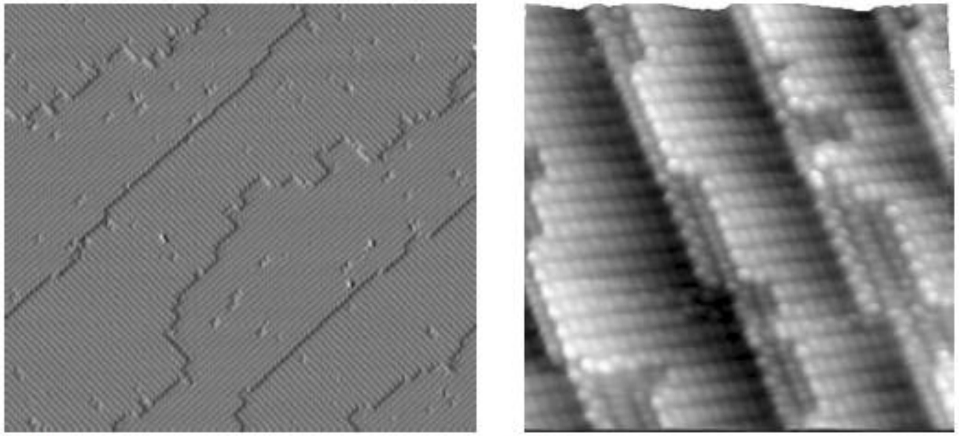
C. Misbah, O. Pierre-Luis, Y. Saito, Rev. Mod. Phys. 82(2010) 981

Step bunching – Si growth – instability mechanism



C. Misbah, O. Pierre-Luis, Y. Saito, Rev. Mod. Phys. 82(2010) 981

Si growth – double steps



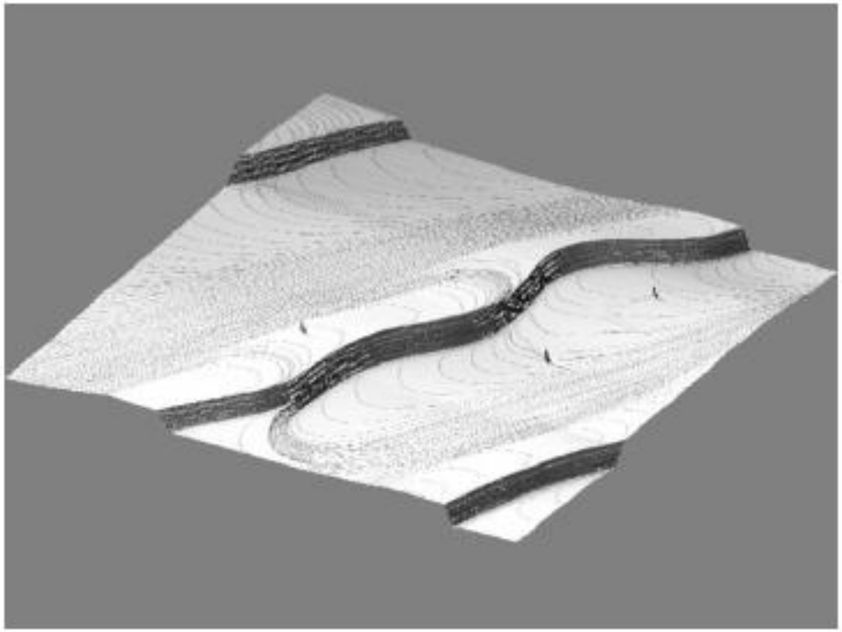
(a) (b)

- **AFM – M. Lagally**

C. Misbah, O. Pierre-Luis, Y. Saito,
Rev. Mod. Phys. 82(2010) 981

Step bunching – Si growth

- **Macrosteps**
- **Single steps on terraces**



C. Misbah, O. Pierre-Luis, Y. Saito, Rev. Mod. Phys. 82(2010) 981

Kinetics of anisotropic steps

$$R_{\pm} = k_{\pm}\nu\left(c_{sur} - c_{sur-eq}\right) = \frac{k_{\pm}c_{sur-eq}\sigma}{\tau_o}$$

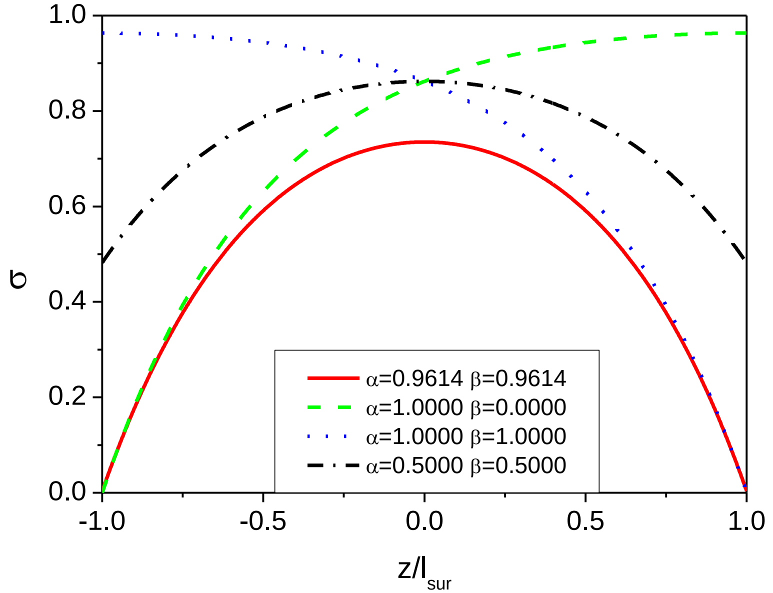
$$\sigma = \sigma_v\left[1 - \frac{\alpha \cosh\left(\frac{2z-y}{l_{sur}}\right)}{\cosh\left(\frac{2y}{l_{sur}}\right)} - \frac{\beta \cosh\left(\frac{2z+y}{l_{sur}}\right)}{\cosh\left(\frac{2y}{l_{sur}}\right)}\right] \quad -\frac{y}{2} < z < \frac{y}{2}$$

- **Supersaturation at the terrace**

$$\alpha = \frac{g_-}{g_- + \tanh\left(\frac{y}{l_{sur}}\right)} = \frac{2k_-\tau_{sur}a}{2k_-\tau_{sur}a + l_{sur}\tau_o \tanh\left(\frac{y}{l_{sur}}\right)}$$

$$\beta = \frac{g_+}{g_+ + \tanh\left(\frac{y}{l_{sur}}\right)} = \frac{2k_+\tau_{sur}a}{2k_+\tau_{sur}a + l_{sur}\tau_o \tanh\left(\frac{y}{l_{sur}}\right)}$$

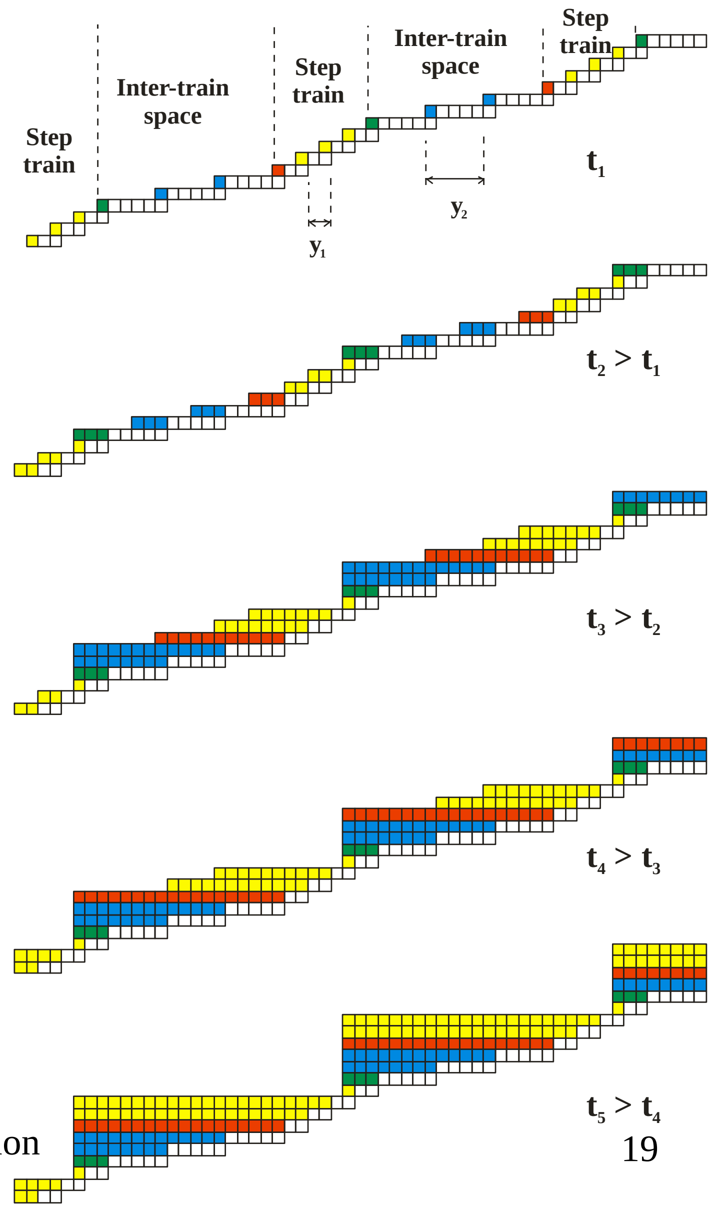
$$g_- = \frac{\tau_{sur}k_-a}{l_{sur}} \qquad g_+ = \frac{\tau_{sur}k_+a}{l_{sur}}$$



S. Krukowski et al., Prog. Cryst. Growth Charact. 68 (2022) 100581

Evolution of the step train – creation of double steps

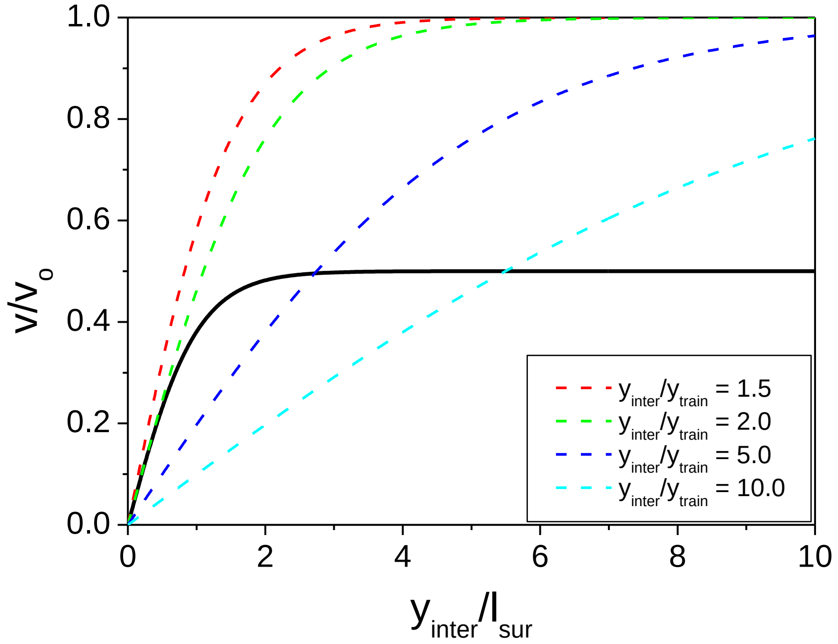
- **Nonuniform step distribution**
- **Motion of: the last step in the train & first step in the interstep**
- **Criterion: motion of single side alimented step is faster than the two-side alimented step**
- **Creation of double step**
- **Coalescence of the following steps**

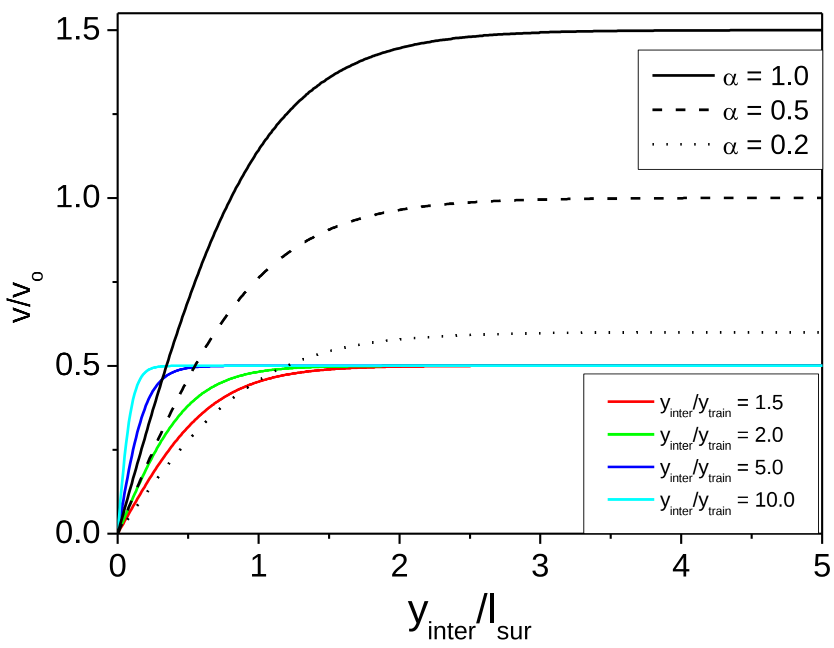


S. Krukowski et al., Prog. Cryst. Growth Charact. 68 (2022) 100581

Creation of double step

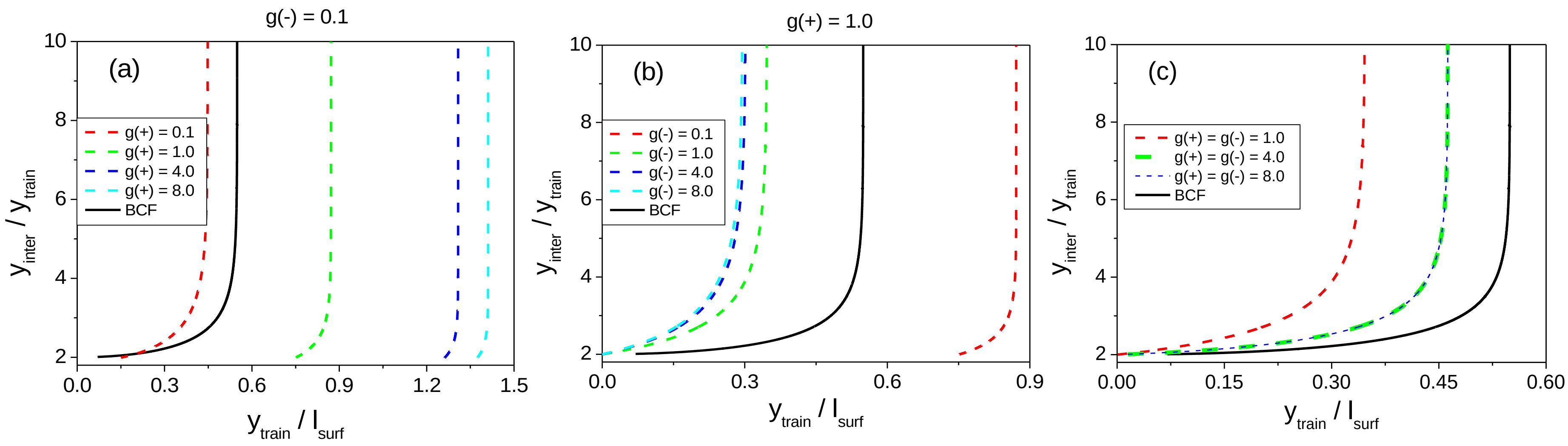
- **BCF kinetics**
- **Symmetric kinetics**





S. Krukowski et al., Prog. Cryst. Growth Charact. 68 (2022) 100581

Step bunching – coalescence regions



S. Krukowski et al., Prog. Cryst. Growth Charact. 68 (2022) 100581

Macrostep evolution – emission of single steps

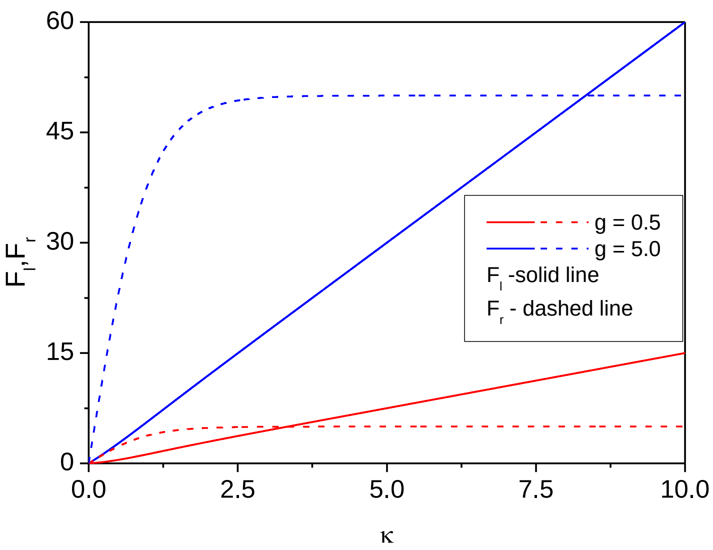
- **Macrosteps – slow motion**
- **Single steps on superterraces – fast motion**

$$z(t) = v_3\, t\, tanh\left(\frac{t}{t_1}\right)$$

$$\kappa[g + tanh(\kappa)] = \sigma_v c_{sur-eq}\left(\frac{t_1}{\tau_{sur}}\right) g\, tanh(\kappa)$$

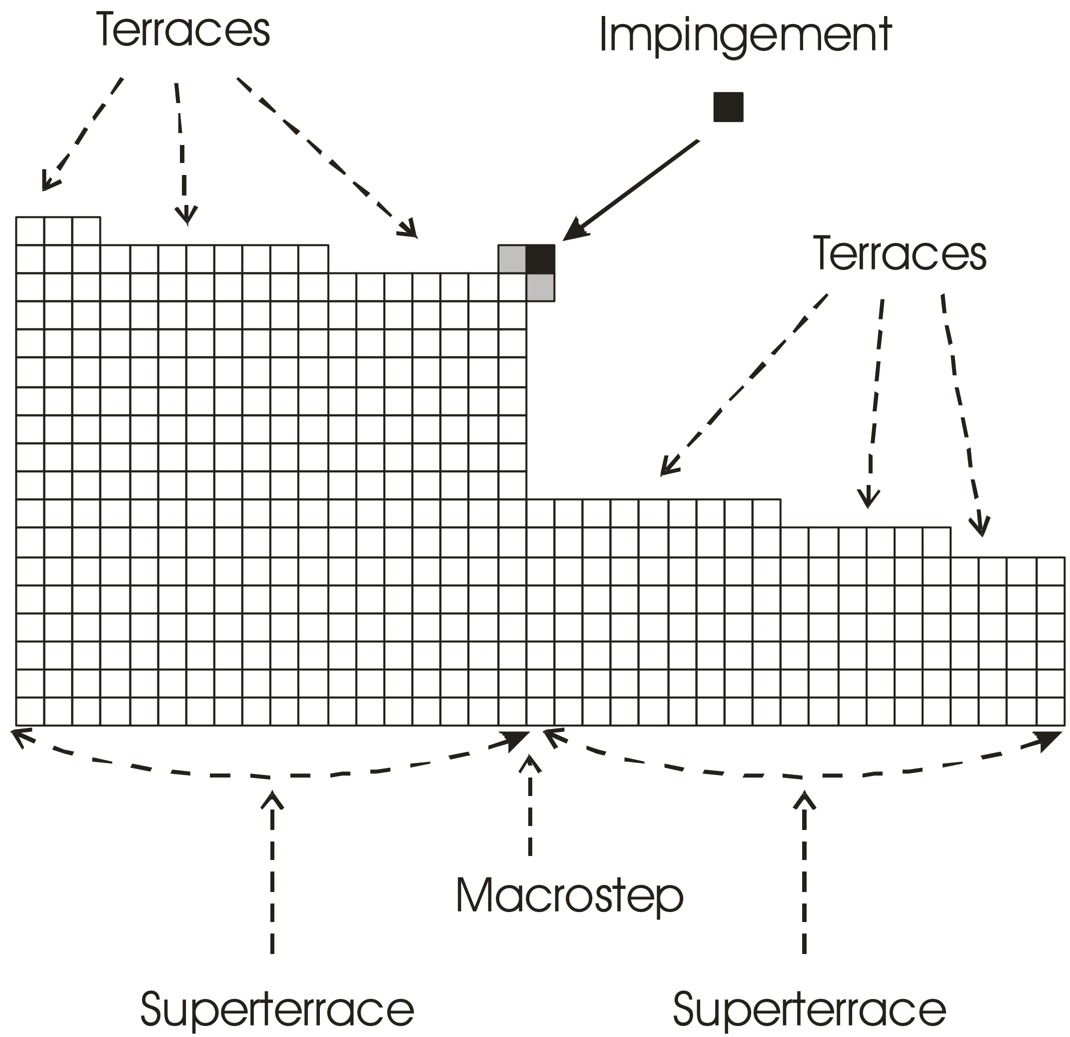
$$\kappa \equiv \frac{\Delta z_3}{l_{sur}} = \frac{v_3\, t_1}{l_{sur}}$$

$$g = \frac{\tau_{sur} k a}{l_{sur}}$$



S. Krukowski et al., Prog. Cryst. Growth Charact. 68 (2022) 100581

Surface structure – macrosteps & superterraces



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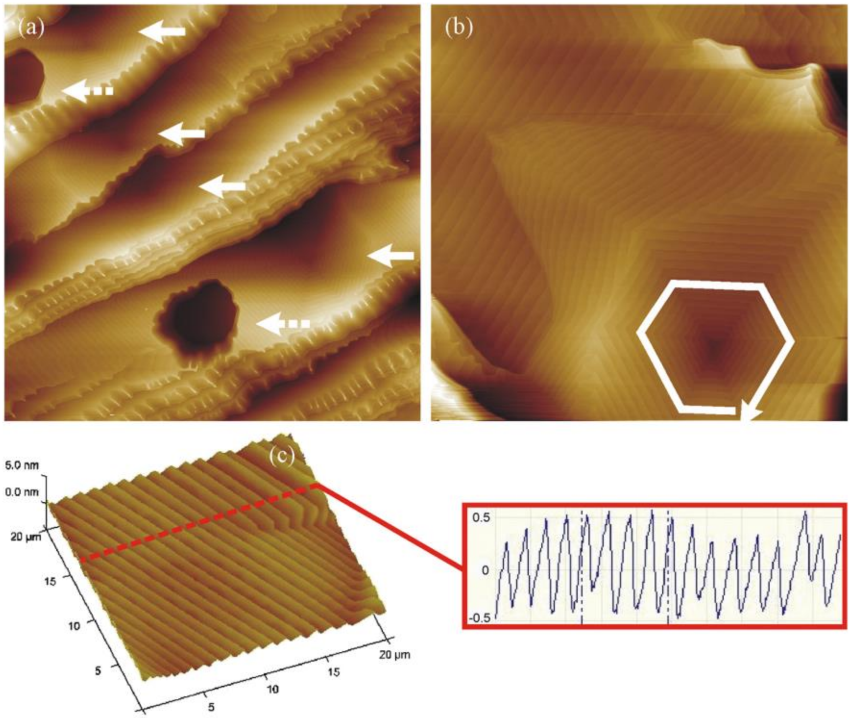
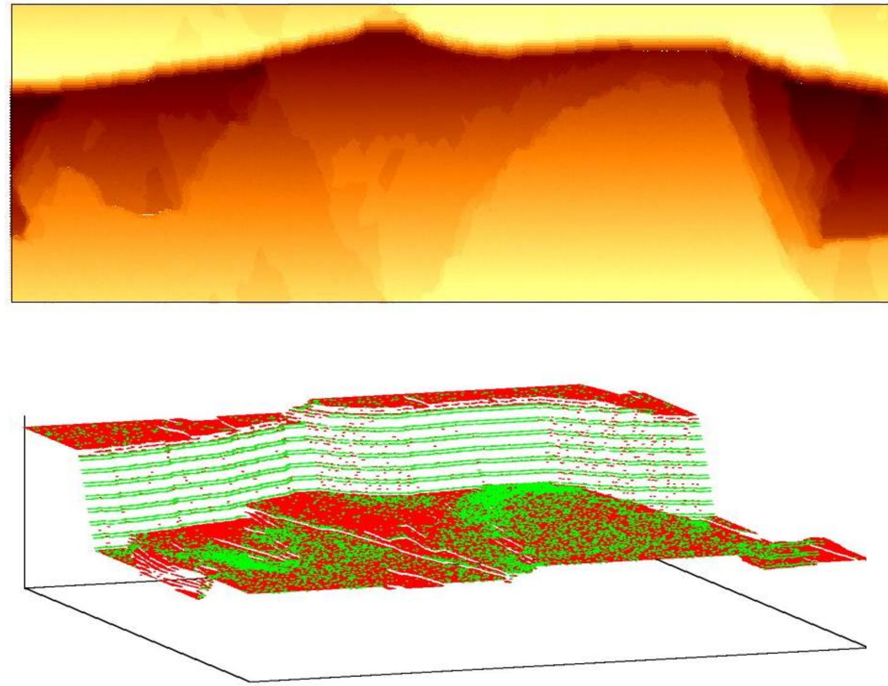
Si(111) surface - annealing





J. Hassan et al., J. Cryst. Growth 310 (2008) 4430

2-d nucleation on terraces – Pt on Pt(111)

0.3 ML **3 ML** **12 ML** **90 ML**

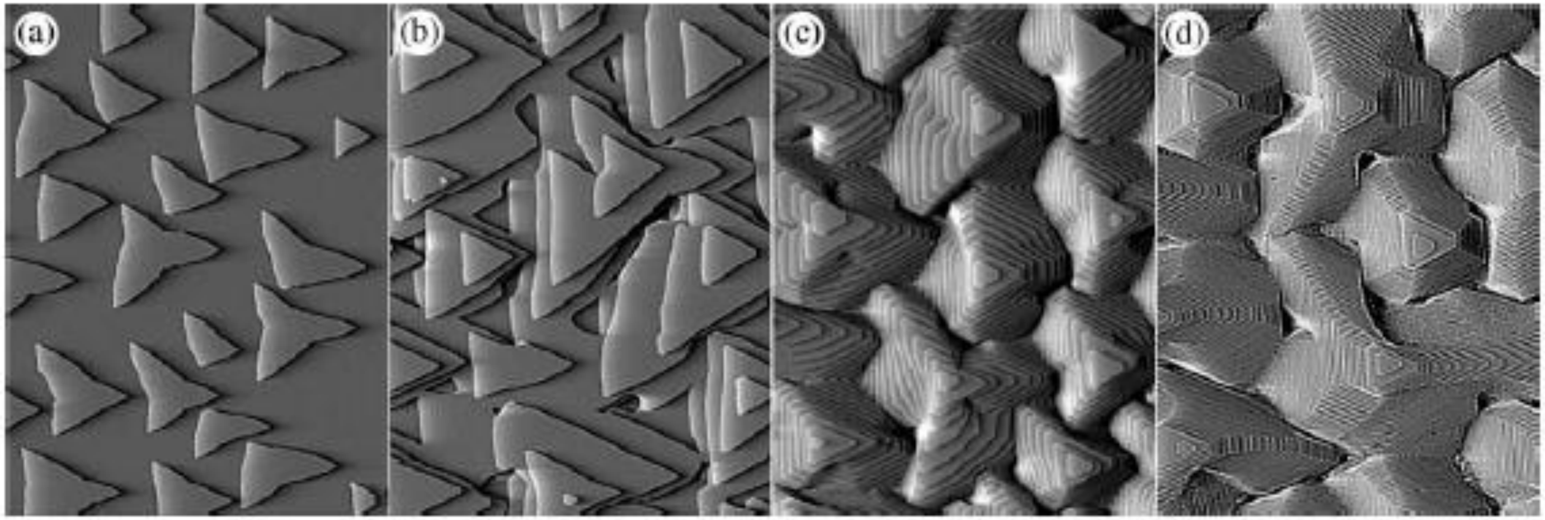
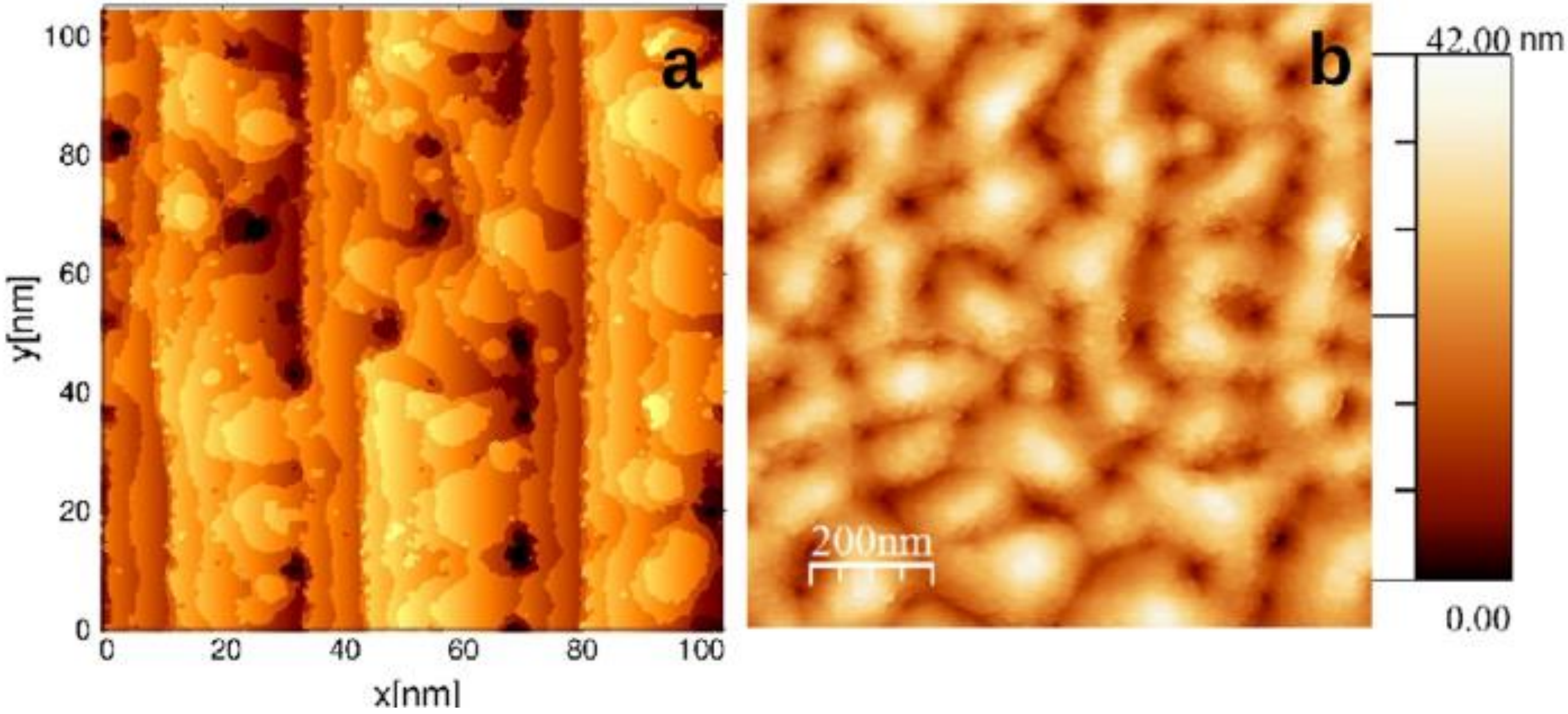


Fig. 2. Growth of Pt on Pt(111) at $T = 440$ K [5]. The total coverage is (a) 0.3 monolayers (ML), (b) 3 ML, (c) 12 ML and (d) 90 ML. The image size is 2600 Å × 3450 Å. Courtesy of T. Michely.

J. Krug, Physica A 313 (2002) 47

2-d nucleation on terraces – GaN on GaN (0001)

MC simulation **MBE growth**

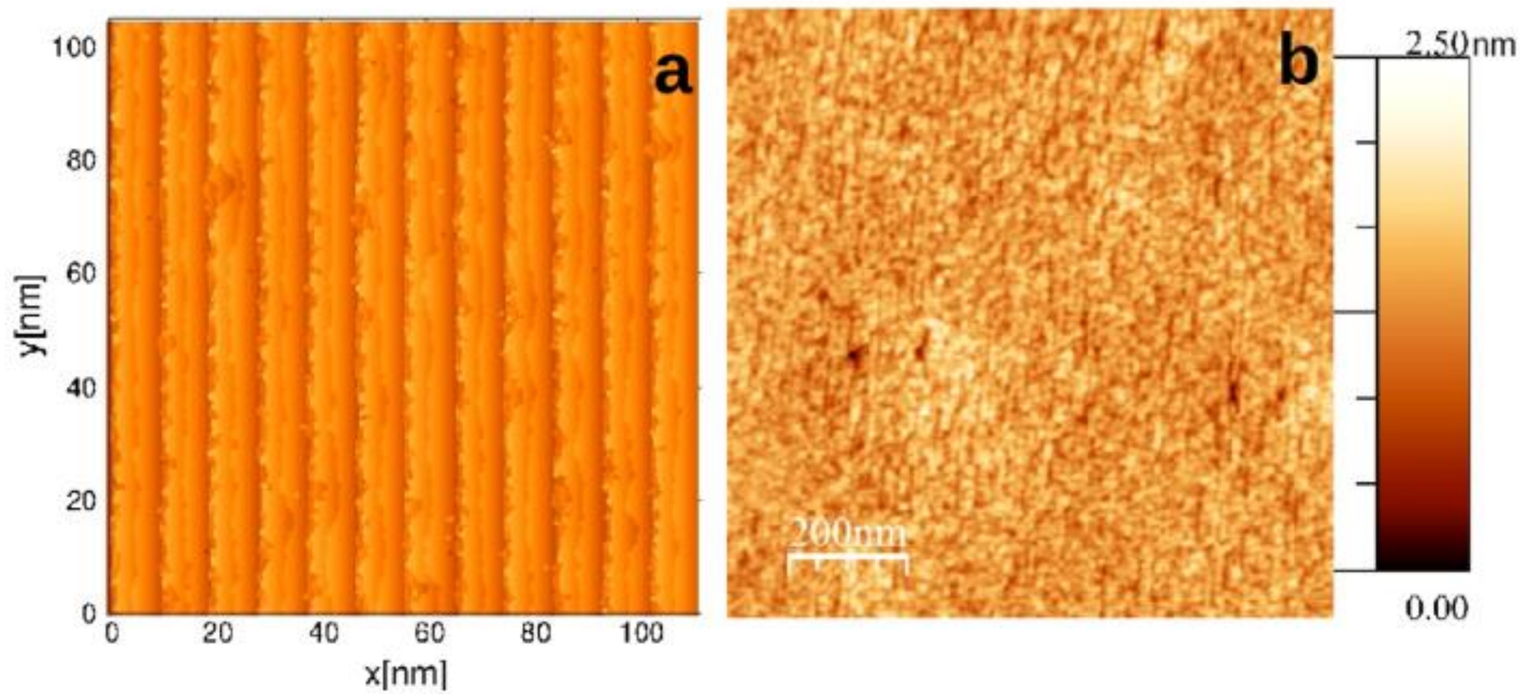


$\boldsymbol{\alpha\ = 2\ arc\ deg}$ $\boldsymbol{T = 750\ °C}$

F. Krzyzewski et al., J. Cryst. Growth 457 (2017) 38

2-d nucleation on terraces – GaN on GaN (0001)

MC simulation **MBE growth**

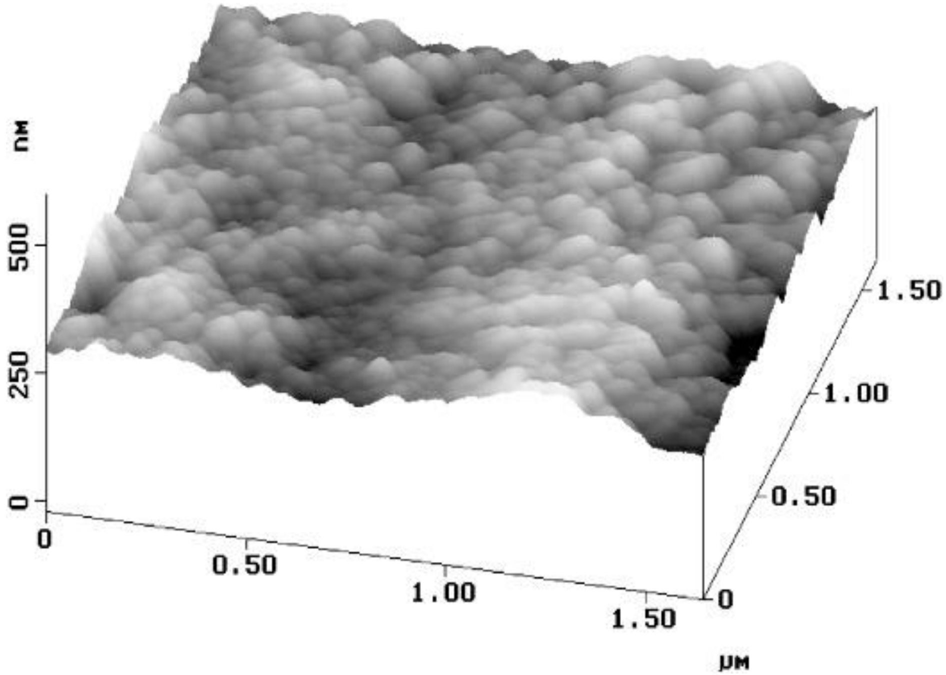


$\alpha = 4\ arc\ deg$ $T = 750\ °C$

F. Krzyzewski et al., J. Cryst. Growth 457 (2017) 38

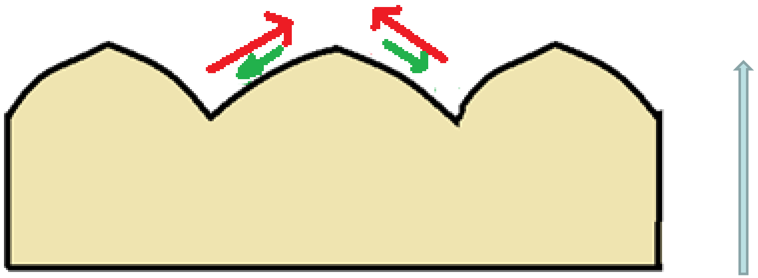
Subsequent layers - mounds

AFM image of a nonideal synthetic dolomite from the Mg:Ca = 1.0 experiment showing mound nanotopography.

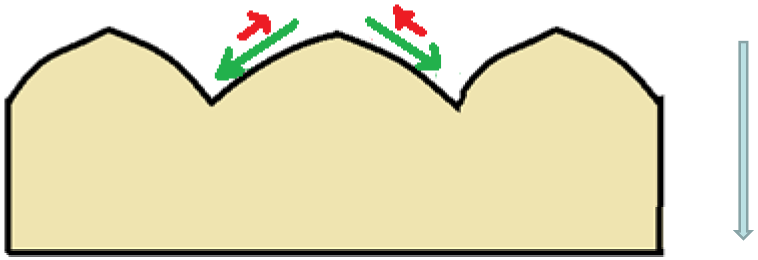


S.E. Kaczmarek & D.F. Sibley,
Journal of Sedimentary Research, 77 (2007) 424

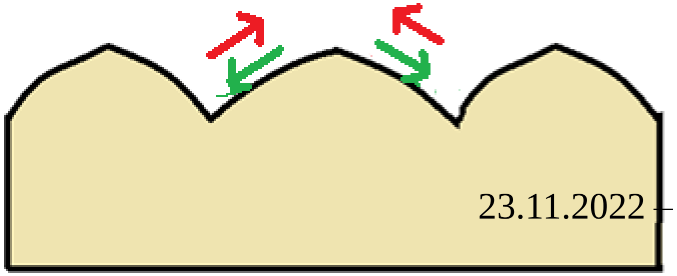
Surface – flux connection



Unstable – growth of the mounds



Stable flat surface – mounds disappering

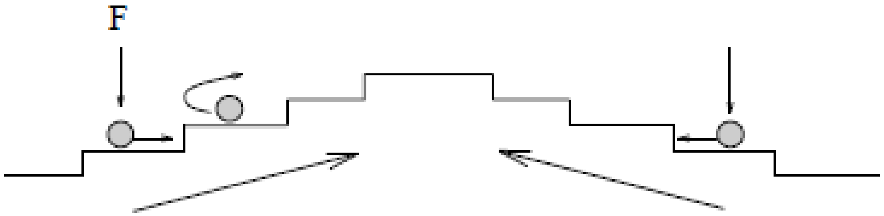


Stable mounds

Surface dynamics – mounds growth

- **Flat surface growth :**

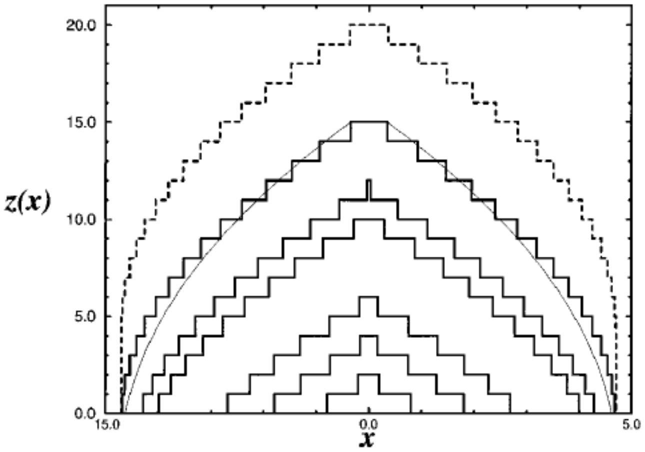
Mounds growth – monostep motion



$$\frac{d\theta_n}{dt} = F\,(\theta_{n-1} - \theta_n)$$

$$\theta_n = 1 - e^{-\Theta} \sum_{j=0}^{n-1} \frac{\Theta^n}{j!}$$

$$\Theta = \sum_{j=0}^{\infty} \theta_j = Ft$$



P. Politi & J. Villain, Phys. Rev. B 54 (1996) 5114

Literature

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