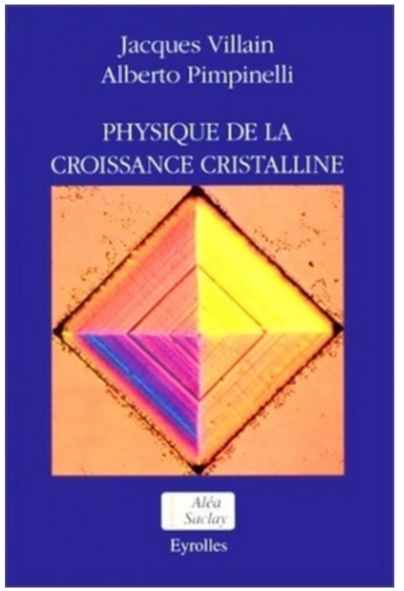
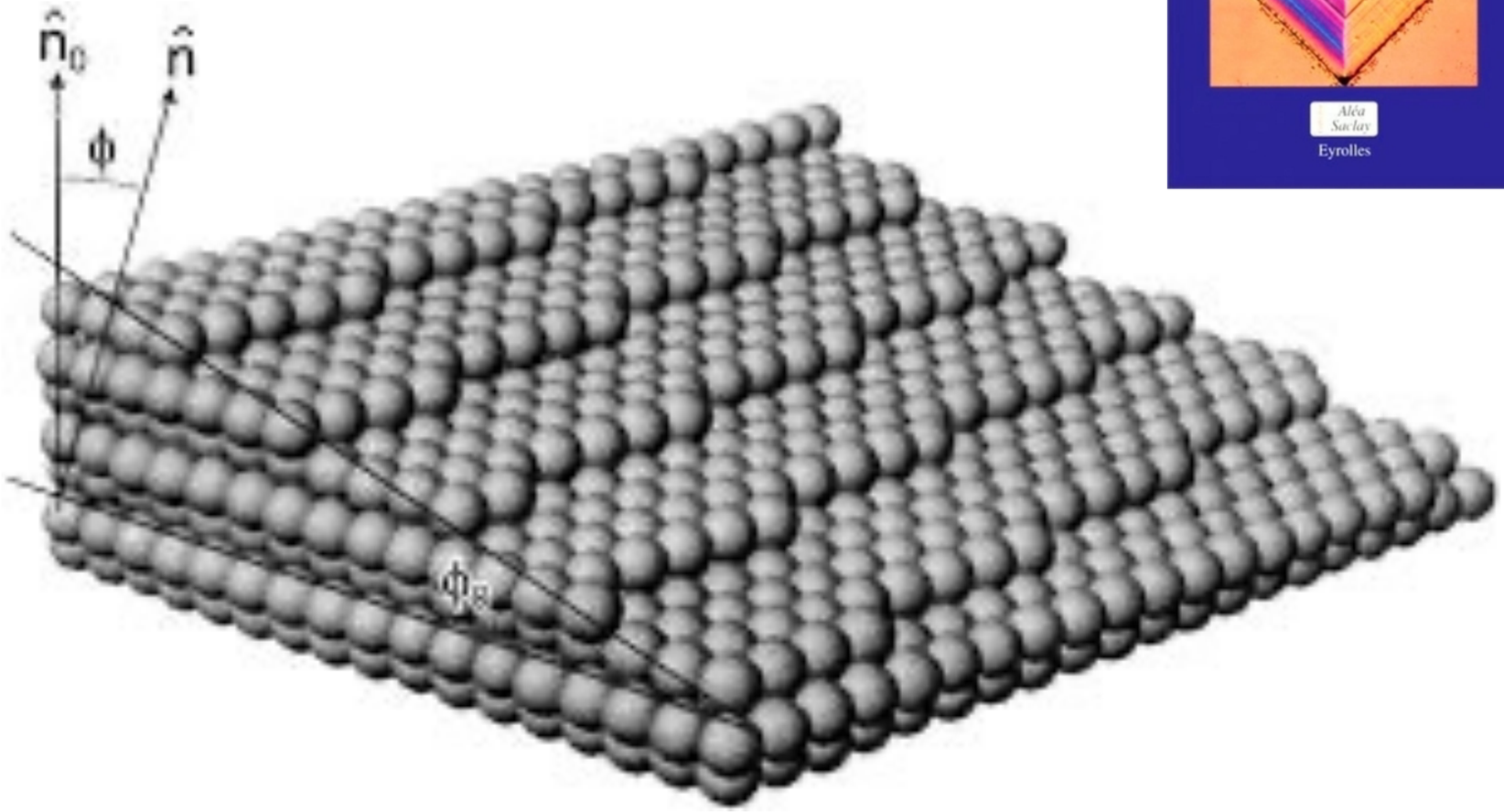
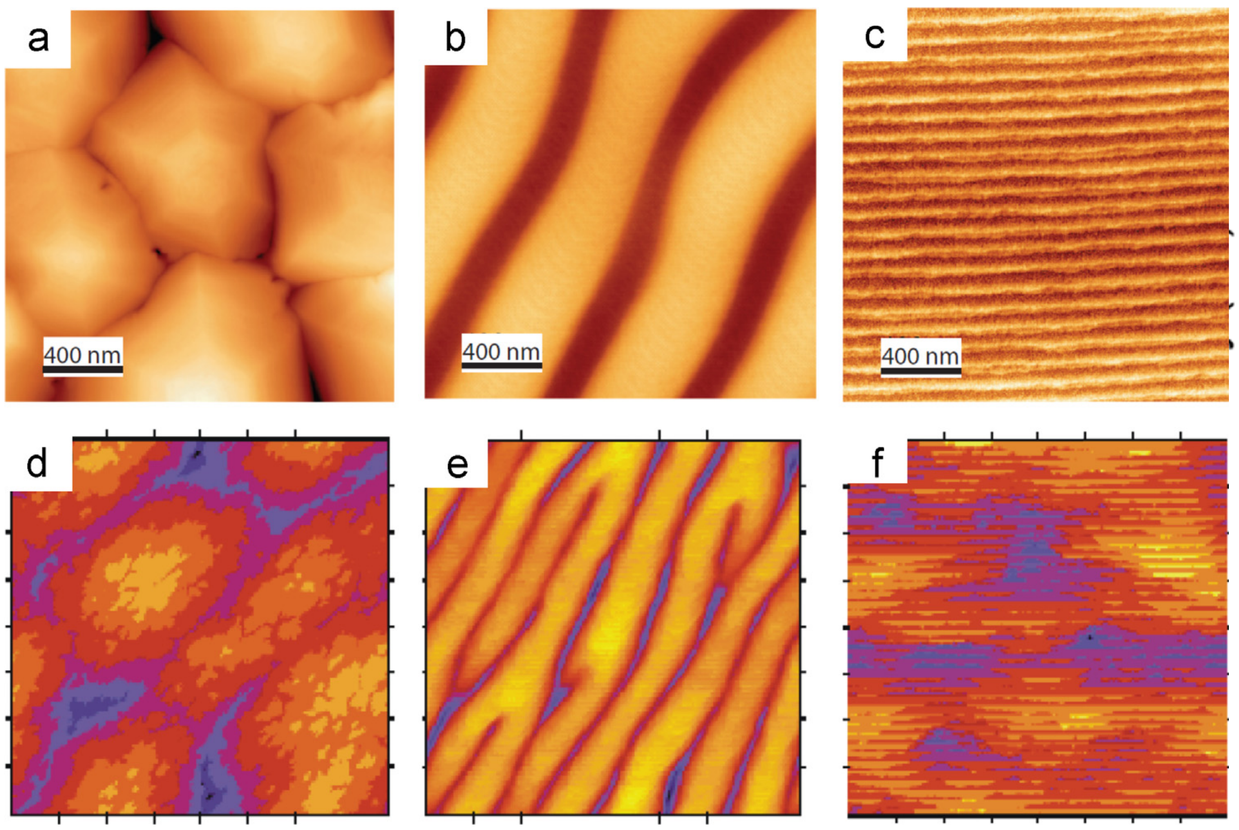
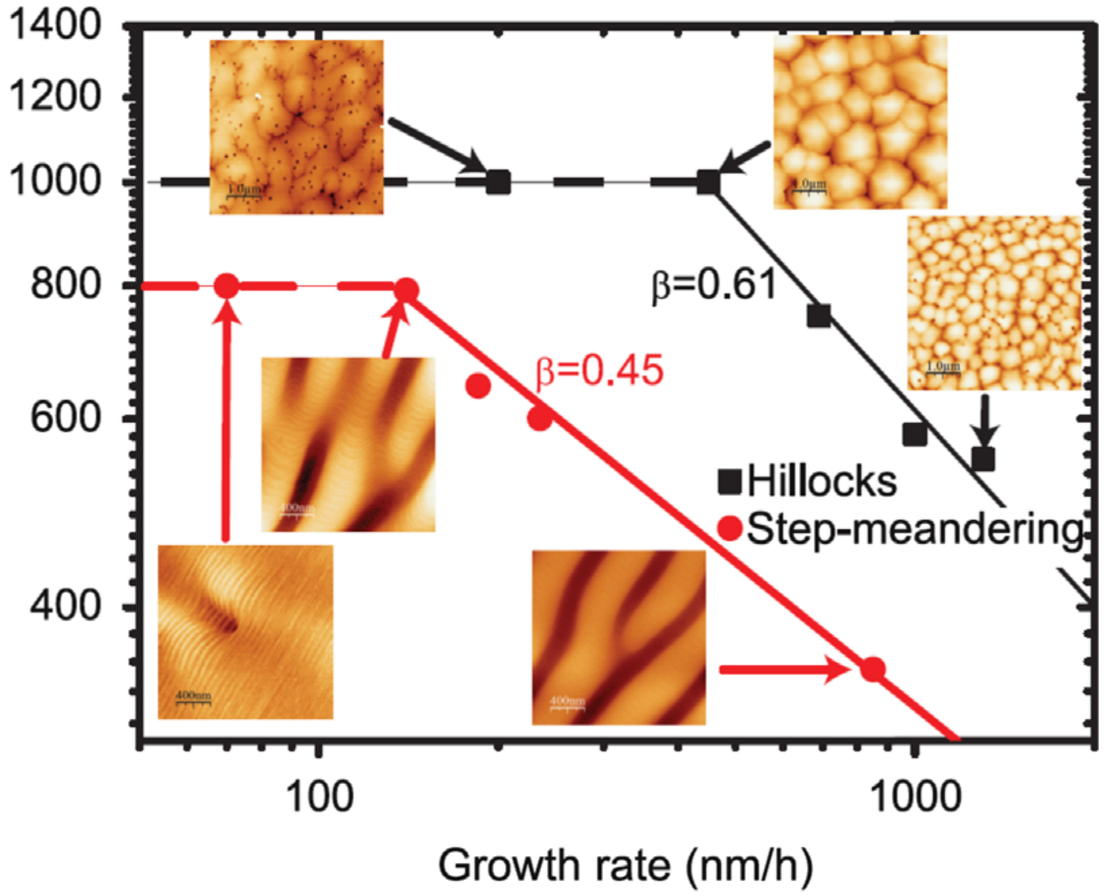
Steps, terraces, and step flow growth





Epitaxial Growth — Morphological Instabilities





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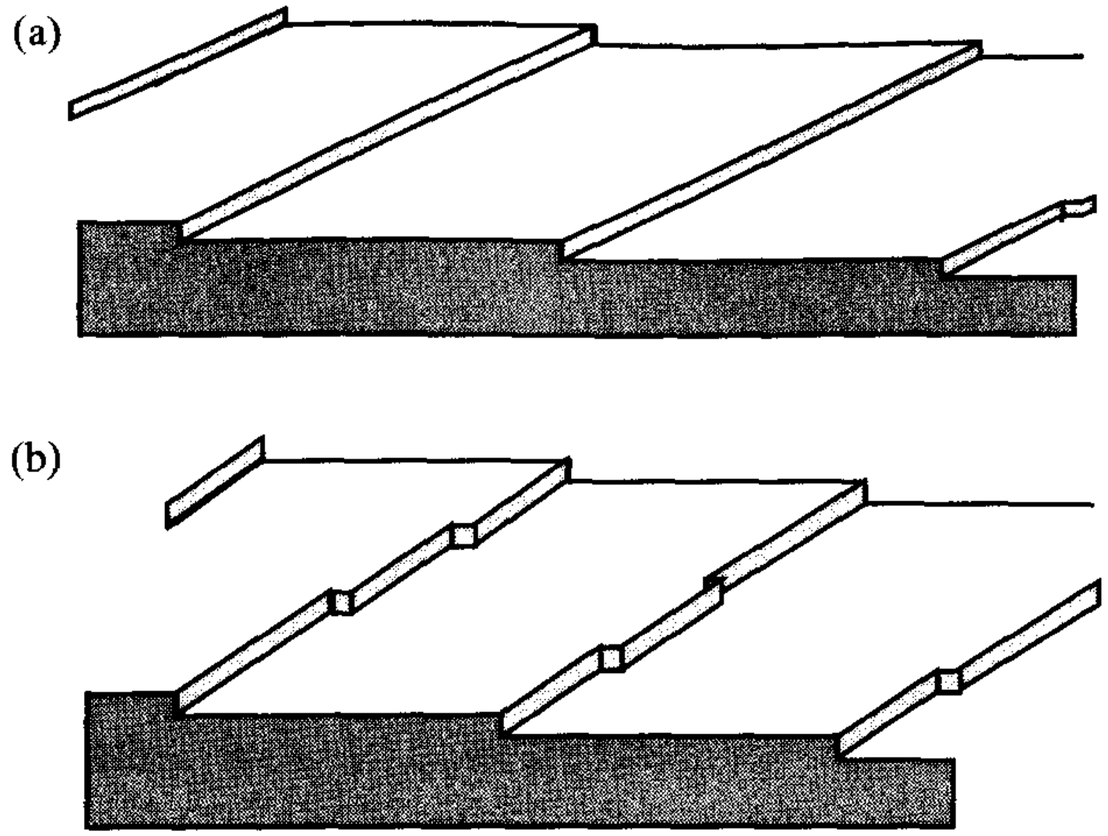
Steps, terraces, and the Burton, Cabrera and Frank (BCF) model of step flow growth



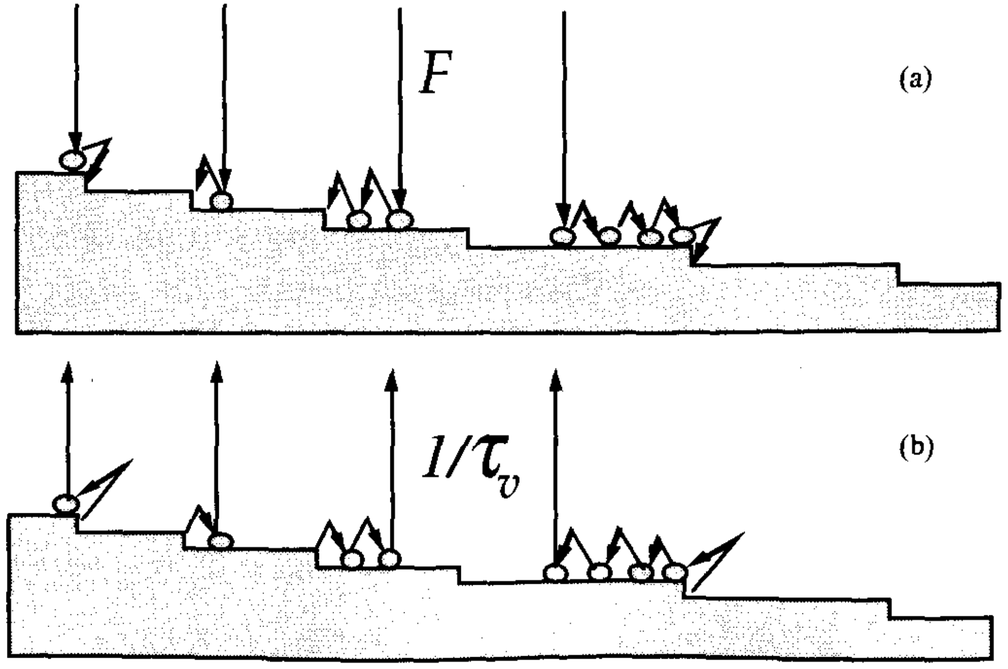


Fig. 6.1. One-dimensional picture of the growth (a) and evaporation (b) of a stepped surface.

$$\frac{\partial n}{\partial t} = D\nabla^2 n + F - \frac{n}{\tau_v}$$

$$\vec{J} = -D\nabla n$$

Boundary conditions at step edge :

$$n = n|_{step};\ \nabla n = \nabla n|_{step}$$

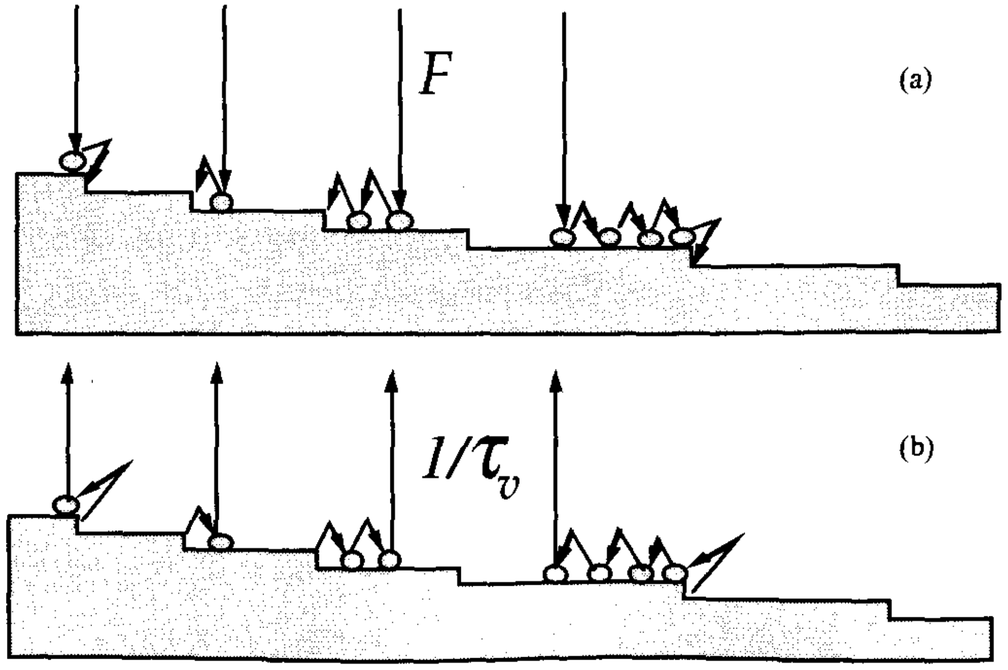


Fig. 6.1. One-dimensional picture of the growth (a) and evaporation (b) of a stepped surface.

Quasi-static approximation

$$\vec{J} = -D$$

ep edge :

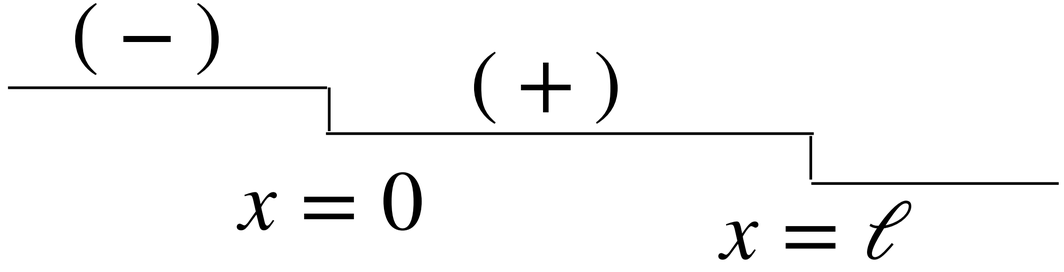
$$n|_{step}$$

Adatom density computed for stationary steps

Example – no evaporation

$$\frac{\partial n}{\partial t} = D\nabla^2 n + F$$

Boundary conditions at step edge :



$$n(0) = n(\ell) = 0$$

$$n(x) = (F/D)\frac{x(\ell - x)}{2}$$

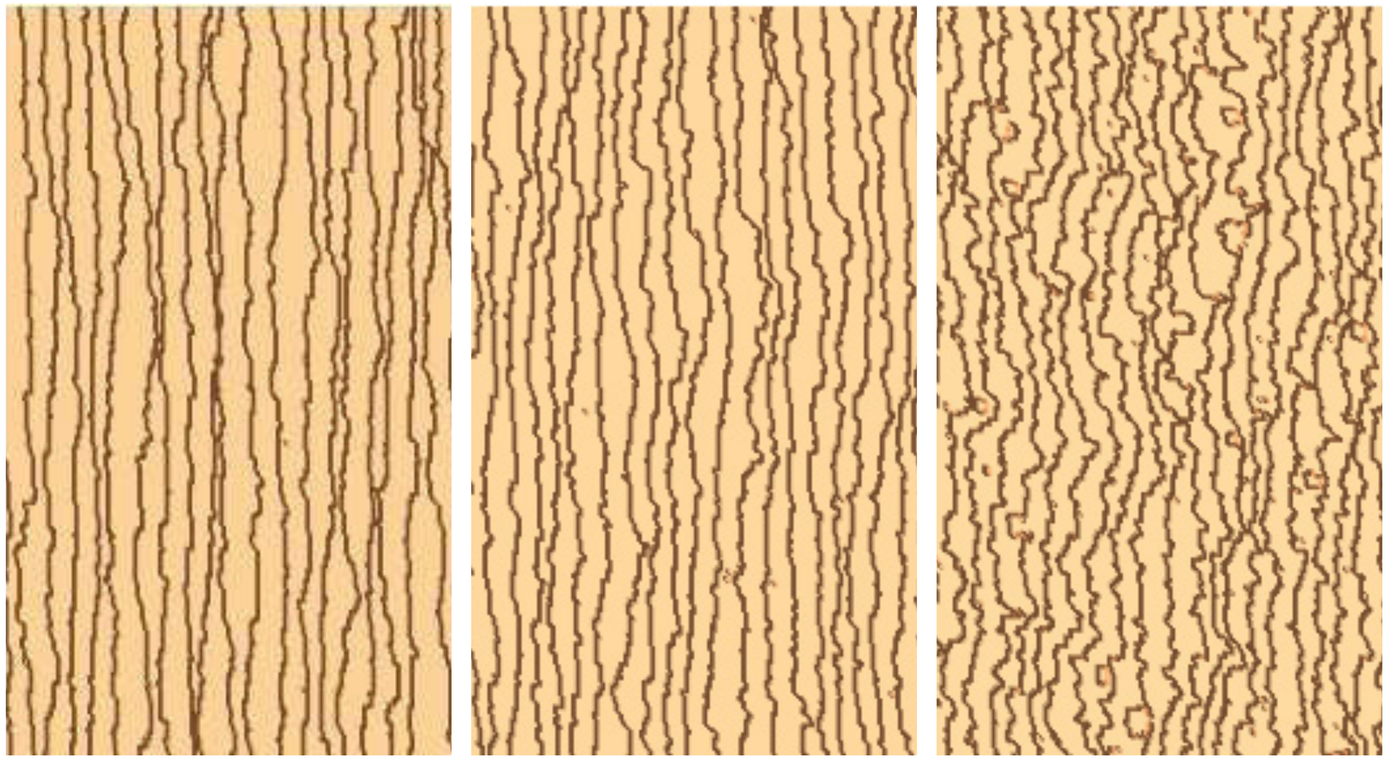
$$v_{\text{step}} = -D\frac{dn^-}{dx}\Big|_{x=0} + D\frac{dn^+}{dx}\Big|_{x=0}$$

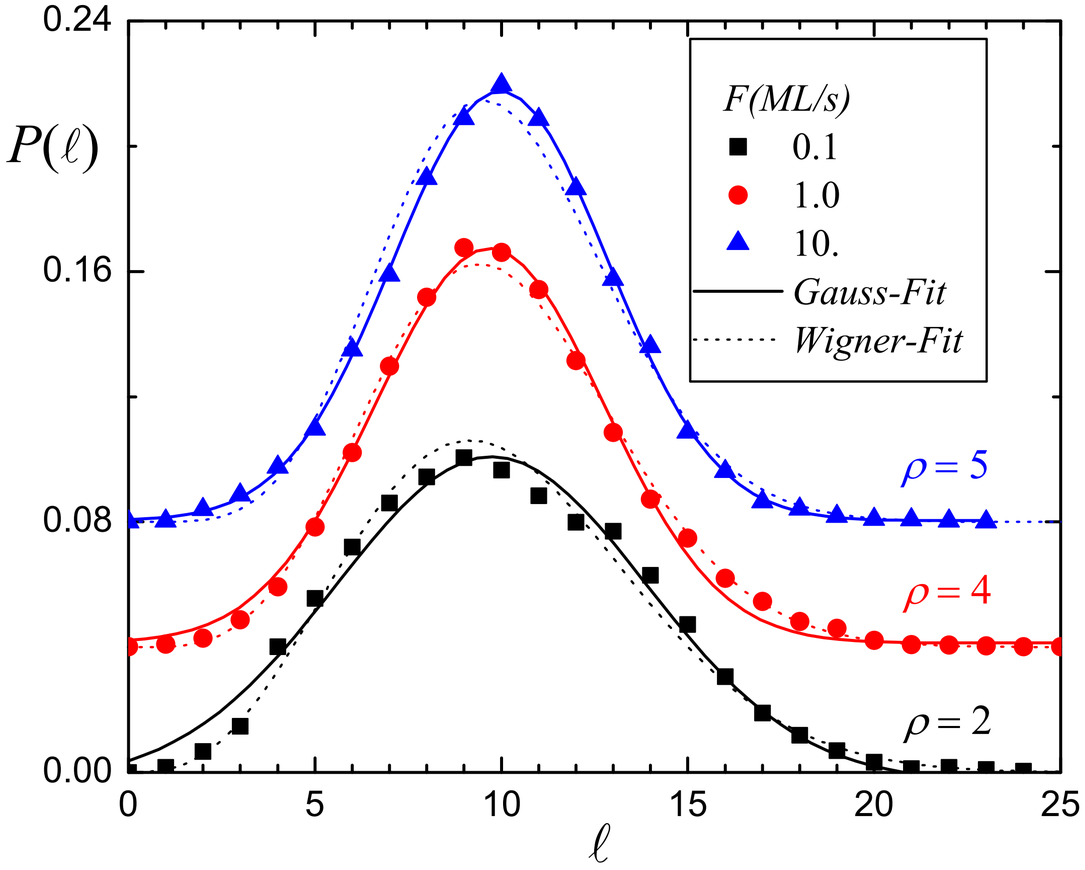
$$v_{\text{step}} = F\ell/2 + F\ell/2 = F\ell$$

Growth rate $= v_{\text{step}} \times$ step density

$$G = F\ell \cdot 1/\ell = F$$

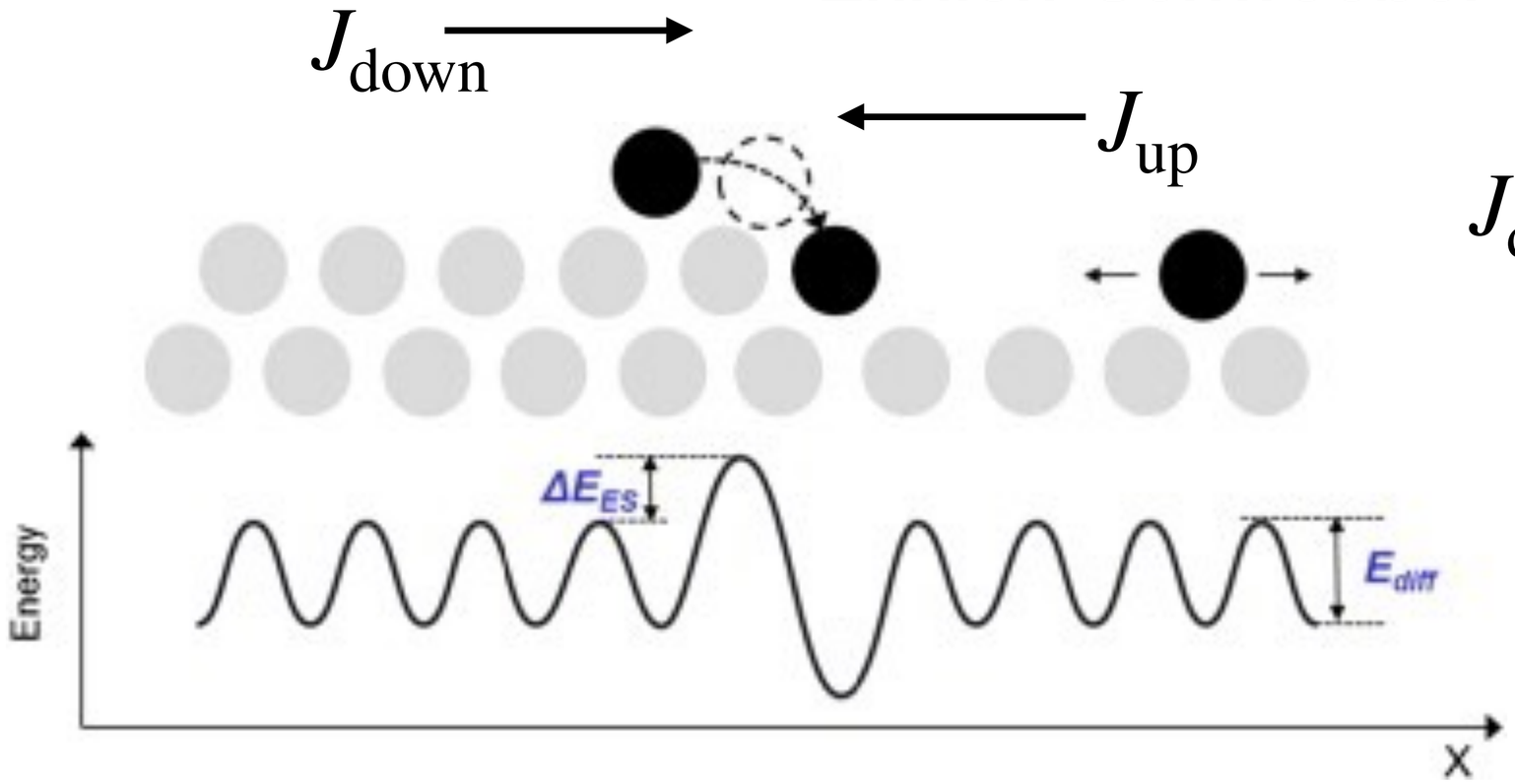
kMC — Moving Steps





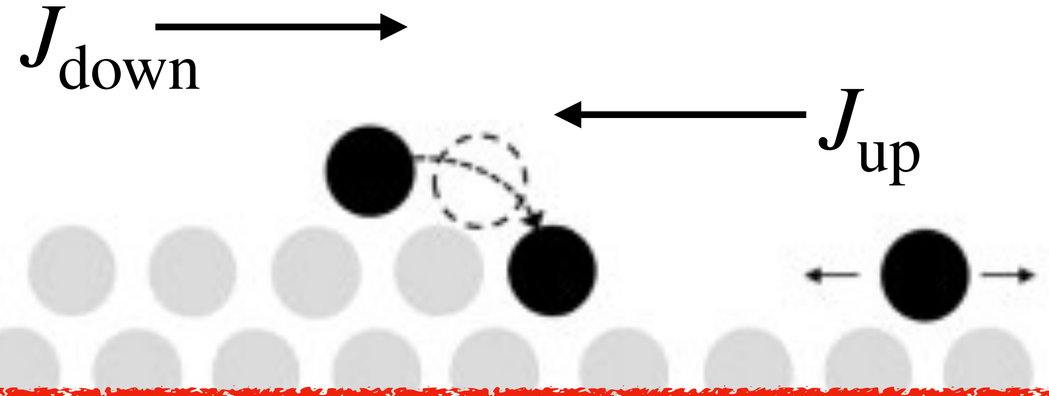
EPL, **88** (2009) 26005
doi: 10.1209/0295-5075/88/26005

Ehrlich-Schwoebel effect



$$J_{\text{down}} < J_{\text{up}}$$

Ehrlich-Schwoebel effect



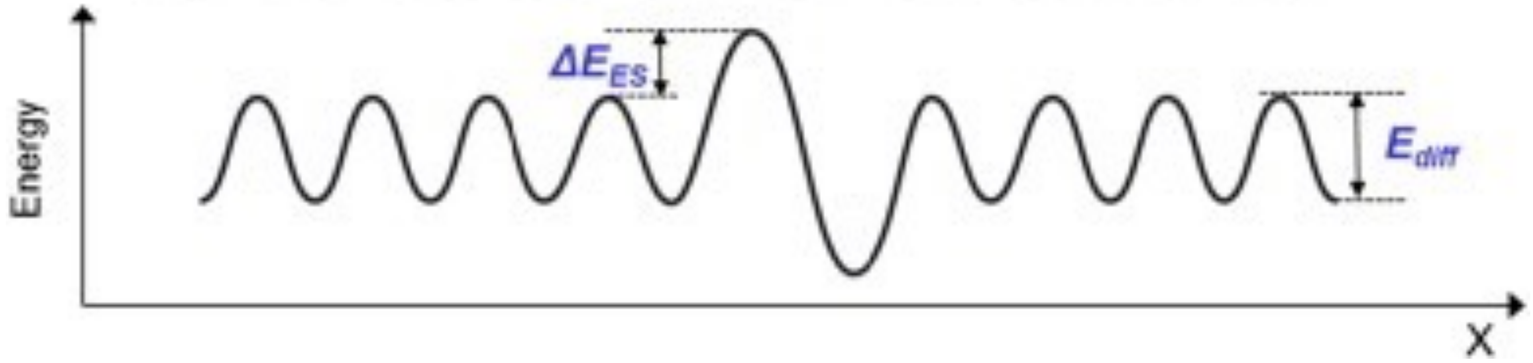
$J_{\text{down}} < J_{\text{up}}$

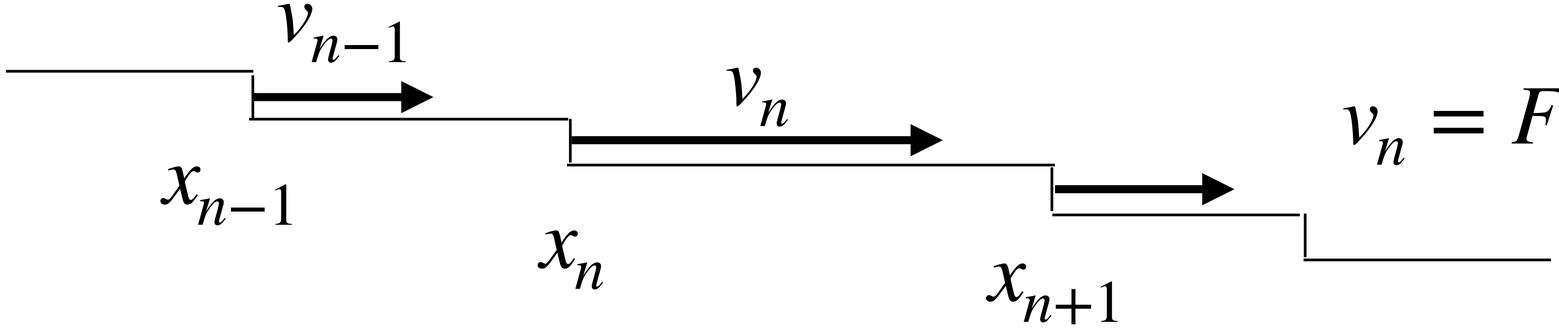
Extreme case: infinite ES effect

Ehrlich-Schwoebel effect

J_{down} J_{up}

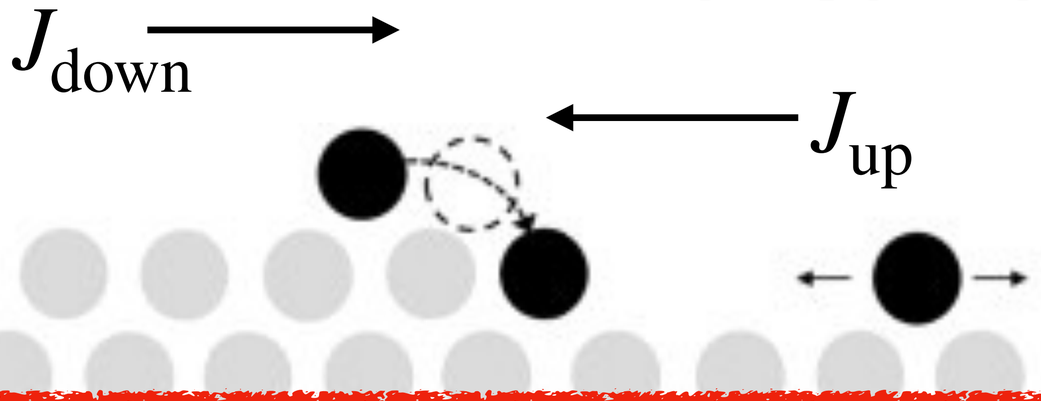
$J_{\text{down}} < J_{\text{up}}$





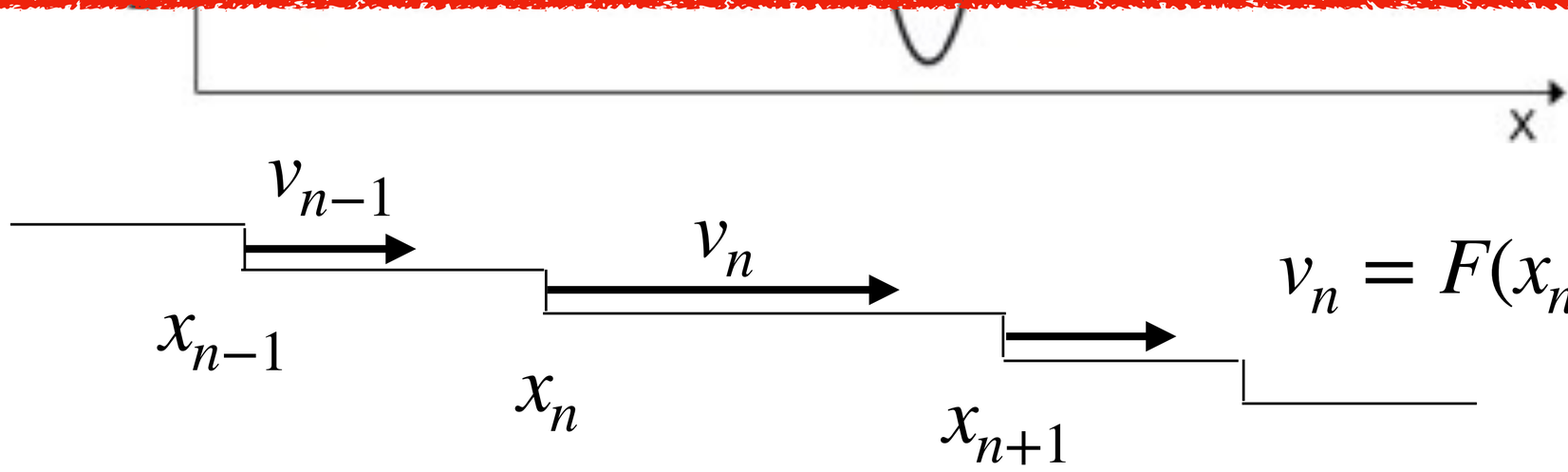
$v_n = F(x_{n+1} - x_n)$

Ehrlich-Schwoebel effect



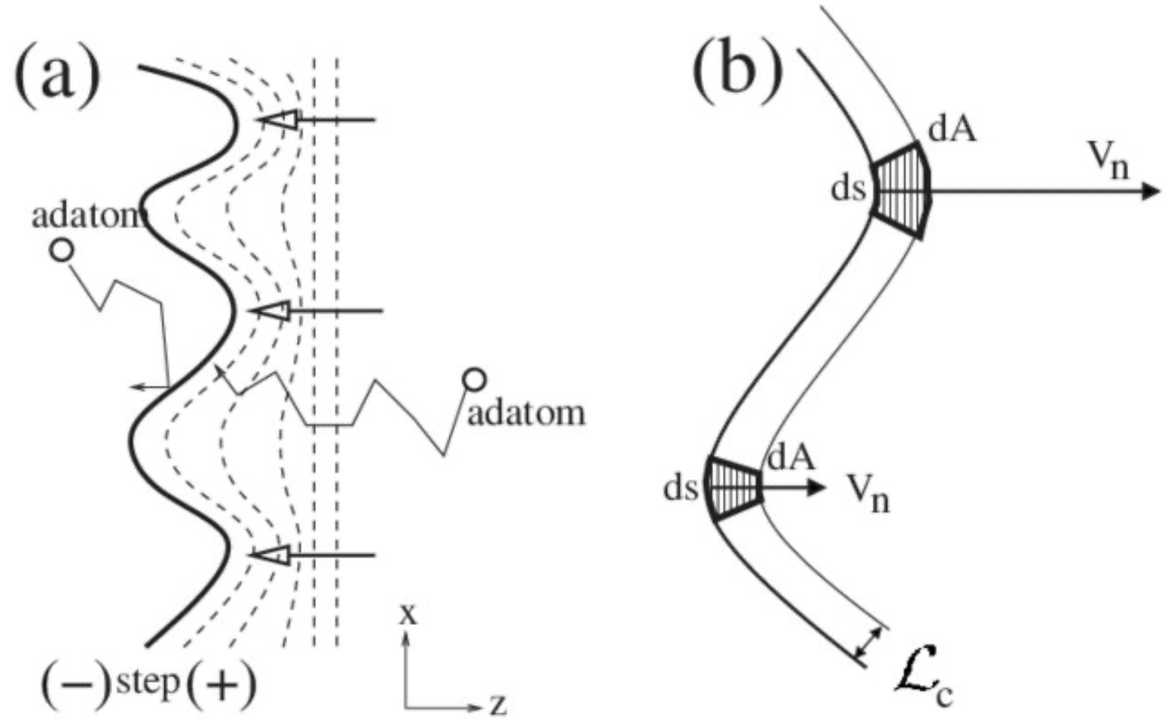
$$J_{\text{down}} < J_{\text{up}}$$

Kinetic repulsive interaction

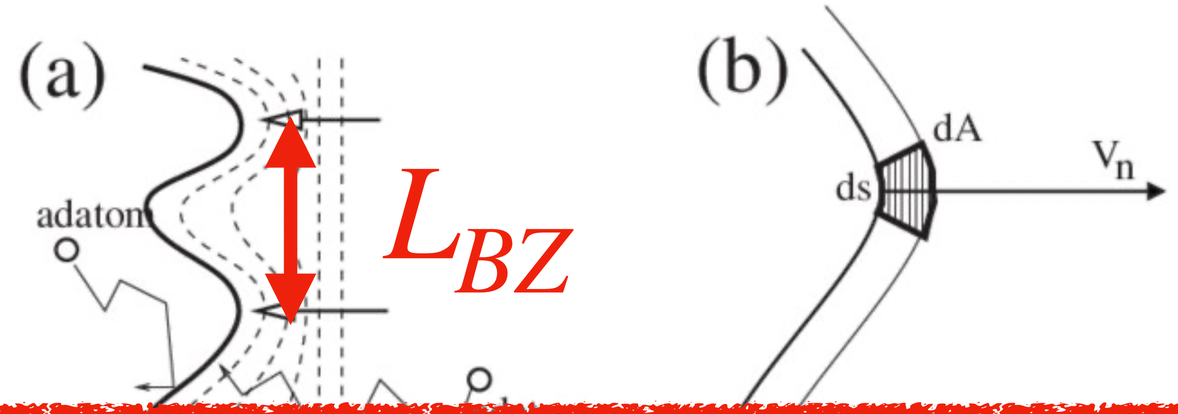


$$v_n = F(x_{n+1} - x_n)$$

Diffusional instability
(“Schwoebel gives meanders”)



Diffusional instability ("Schwoebel gives meanders")

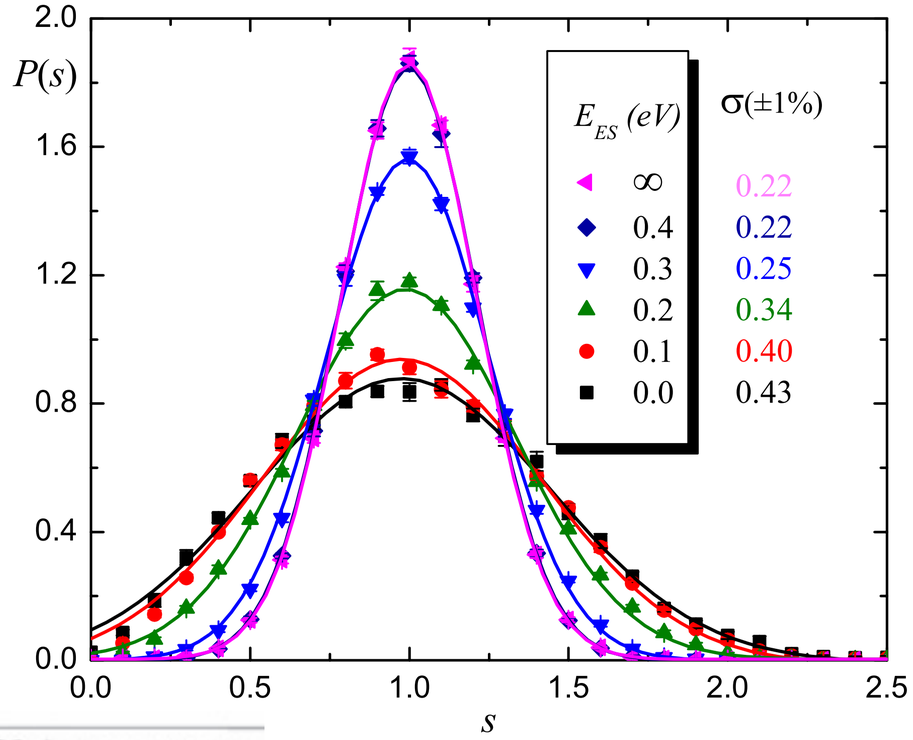


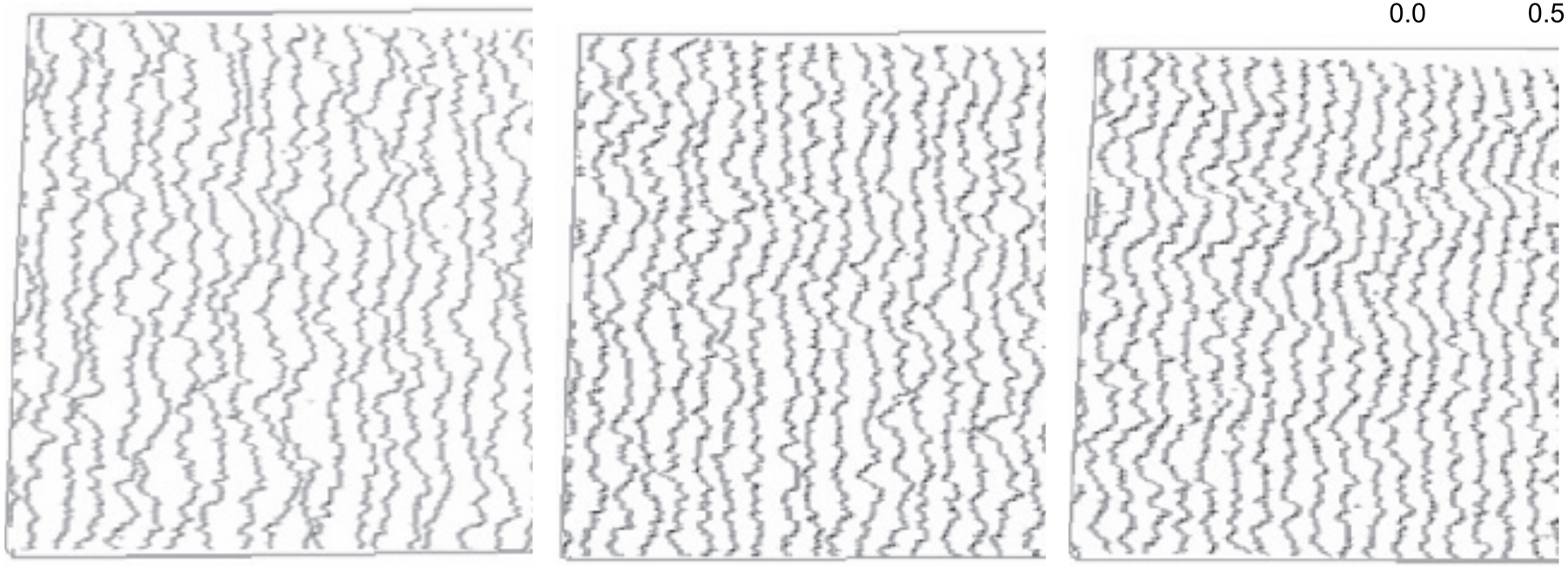
$$L_{BZ} \approx (D/F)^{1/2}$$

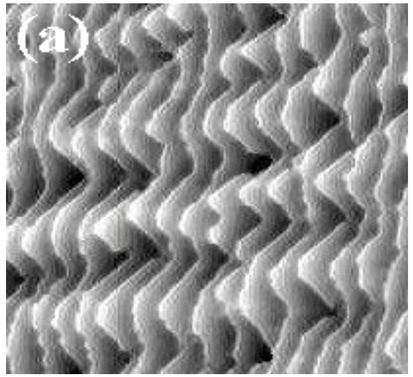
Bales & Zangwill, 1990

Morphological instability of a terrace edge during step-flow growth

Diffusional instability ("Schwoebel gives meanders")

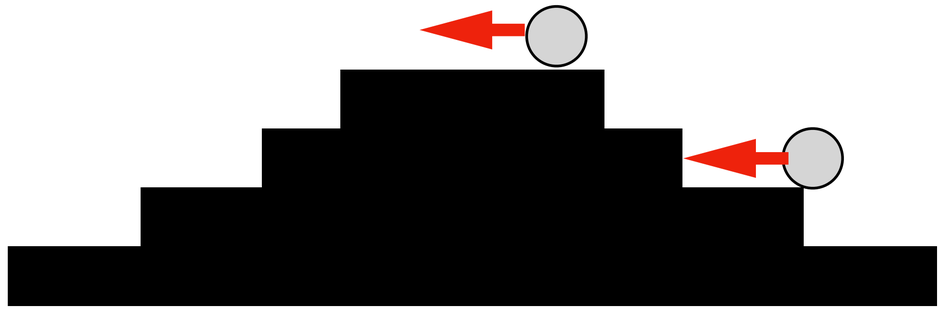






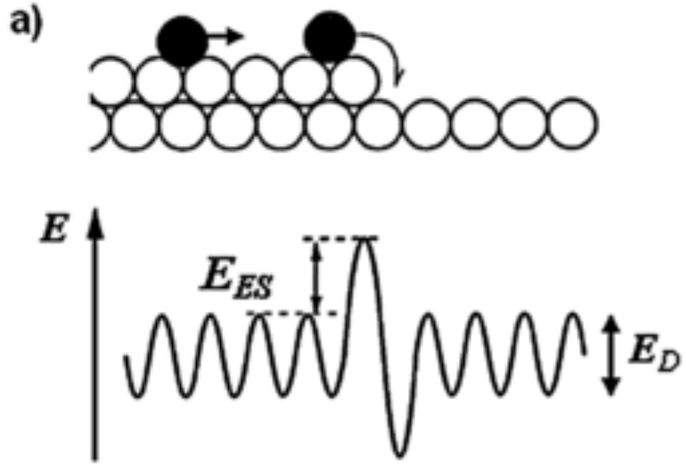
Cu (0,2,24)

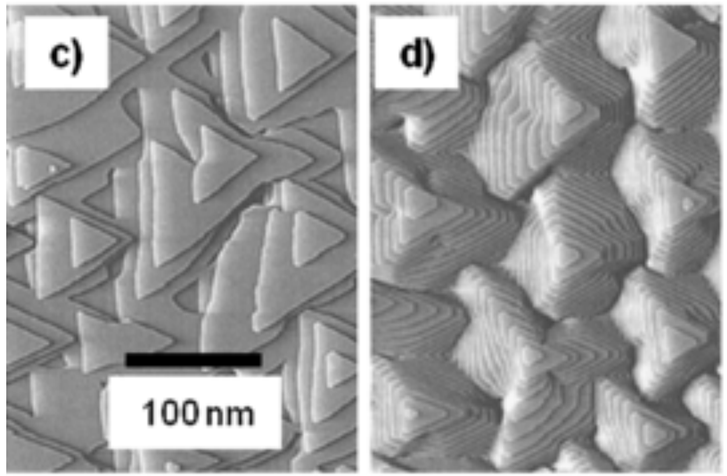
2D to 3D: interlayer transport barriers ("Schwoebel gives mounds")

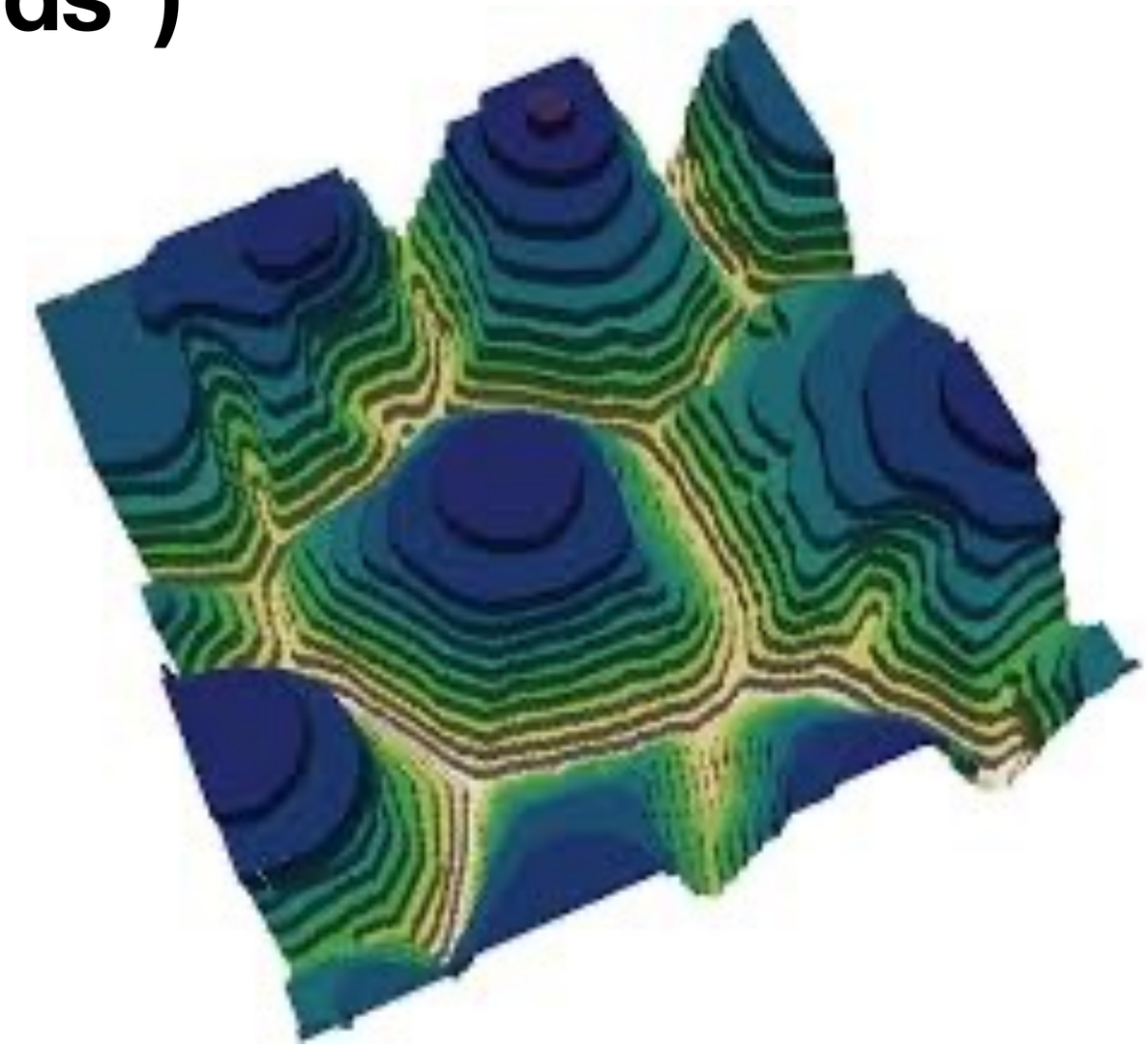


$$J_{\text{down}} < J_{\text{up}}$$

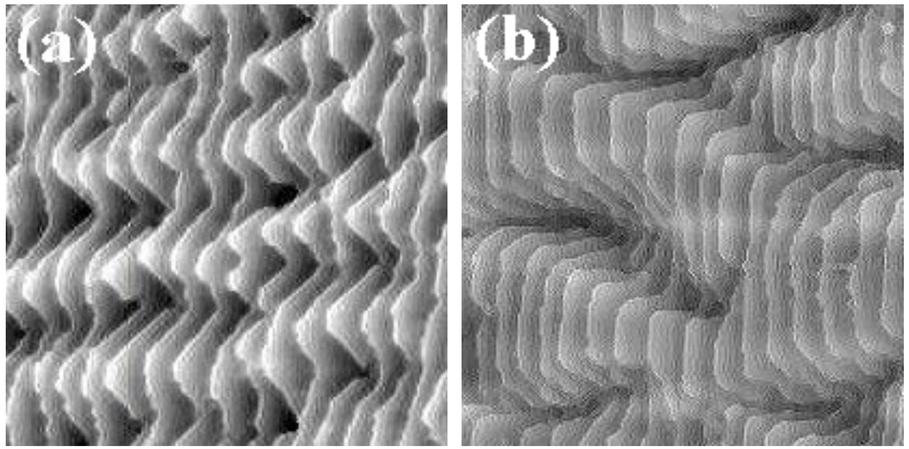
2D to 3D: interlayer transport barriers ("Schwoebel gives mounds")



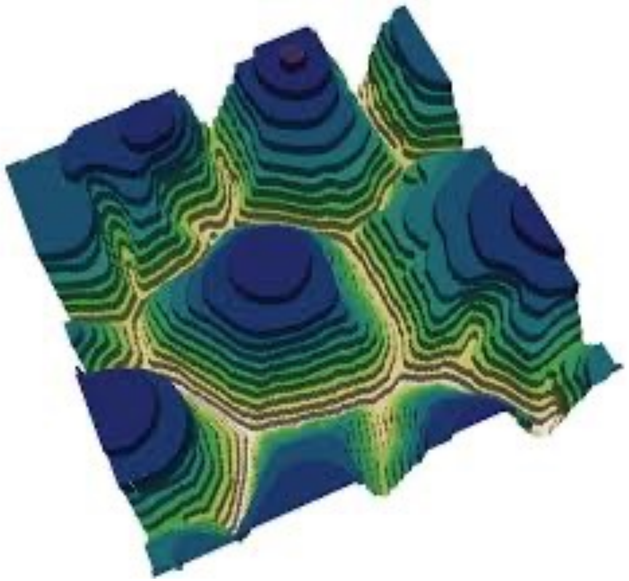




2D to 3D: interlayer transport barriers ("Schwoebel gives mounds")



$$J_{\text{down}} < J_{\text{up}}$$



Net current "up": leads to meandering steps and/or mounds

Unstable morphologies

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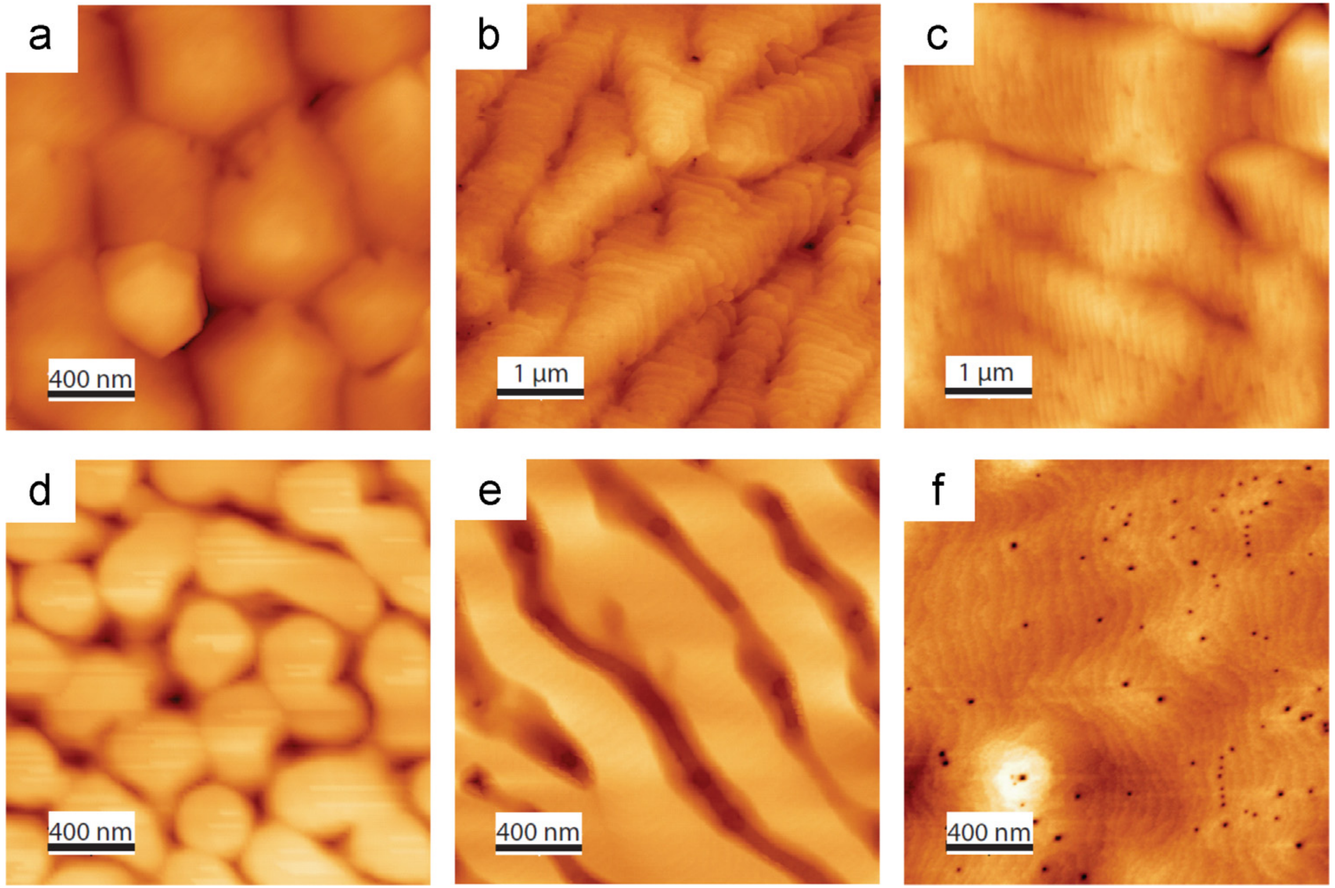


AFM images of NH3-MBE-grown GaN layers in hillock, step-meandering, and step-flow regime. For (a), (b) and (c) the growth temperatures were 800ºC, 865ºC and 920ºC, respectively.
The sample displayed in (d) and (e) was grown by nitrogen-rich PAMBE at 790ºC on FS GaN (unintentionally offcut variations of the GaN FS substrate are responsible for the different morphologies). (f) Step- ow growth surface obtained by PAMBE at 7401C under metal-rich conditions.

GaN vicinals — meandering steps or mounds

Unstable morphologies

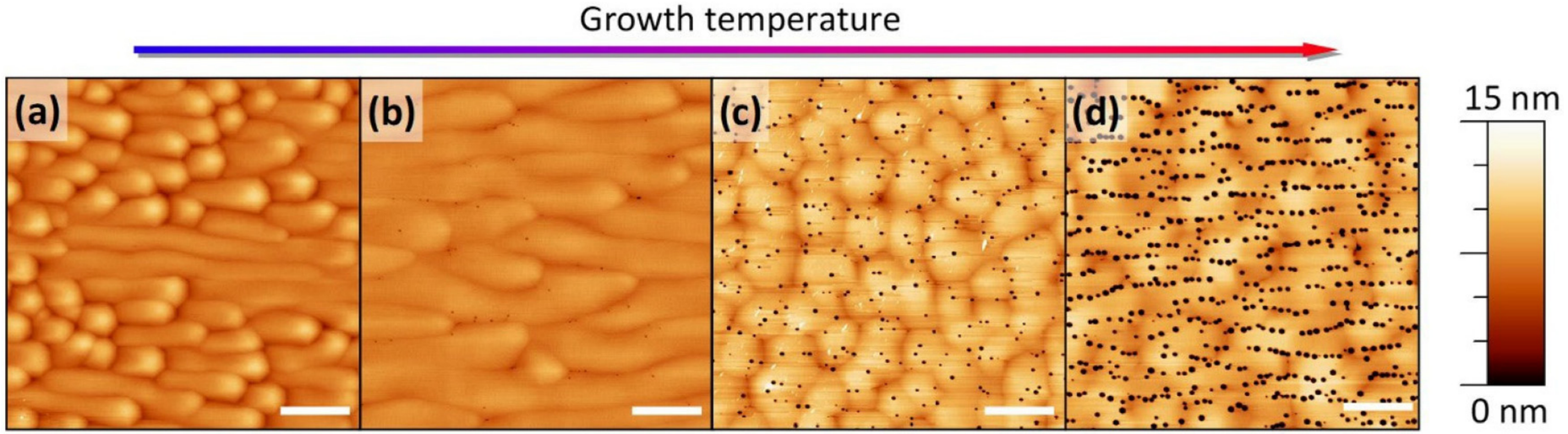


FIG. 1. $10 \times 10\,\mu m^2$ atomic force microscopy images of the AlGaN surface of samples A–D [(a)–(d)]. The growth temperature increases from 743 °C (left) to 818 °C (right). The scale bar corresponds to $2\,\mu m$.

GaN vicinals — meandering steps + small mounds or mounds

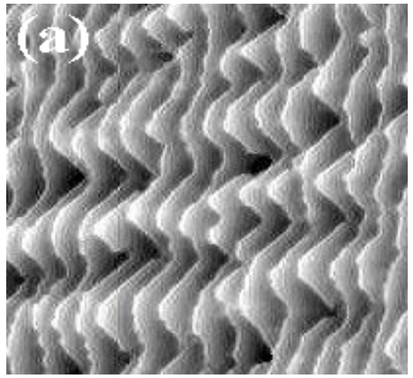
Morphological instabilities in epitaxial growth: a case study

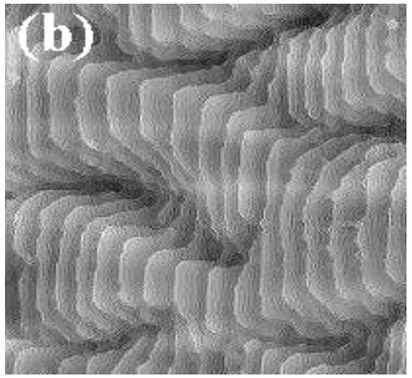
PHYSICAL REVIEW B, VOLUME 64, 165401

Morphological instability of Cu vicinal surfaces during step-flow growth

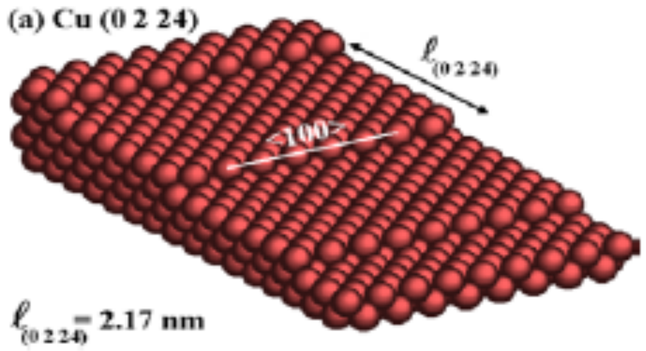
T. Maroutian, L. Douillard, and H.-J. Ernst
CEA Saclay, DSM/DRECAM/SPCSI, F-91191 Gif sur Yvette, France
(Received 13 February 2001; published 28 September 2001)

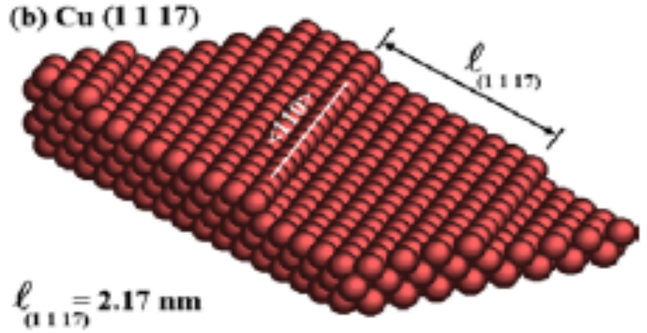
Cu (0,2,24)



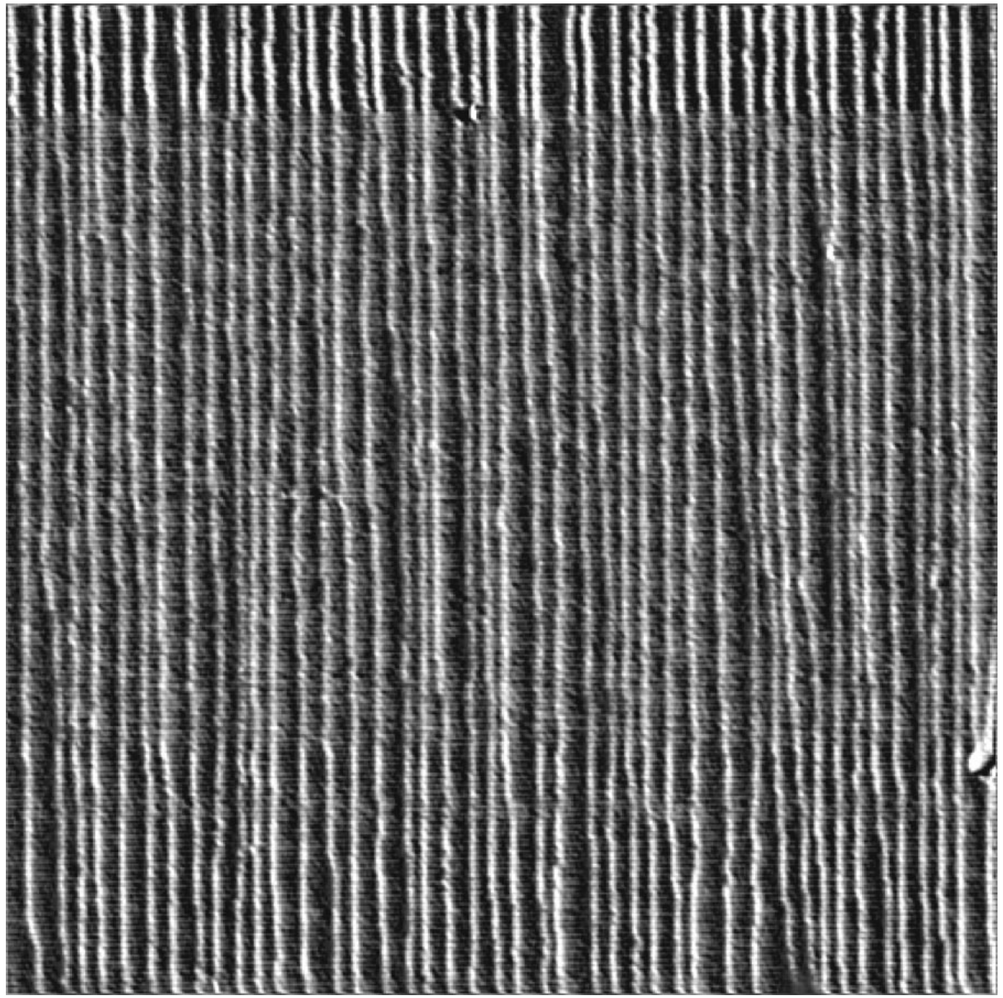


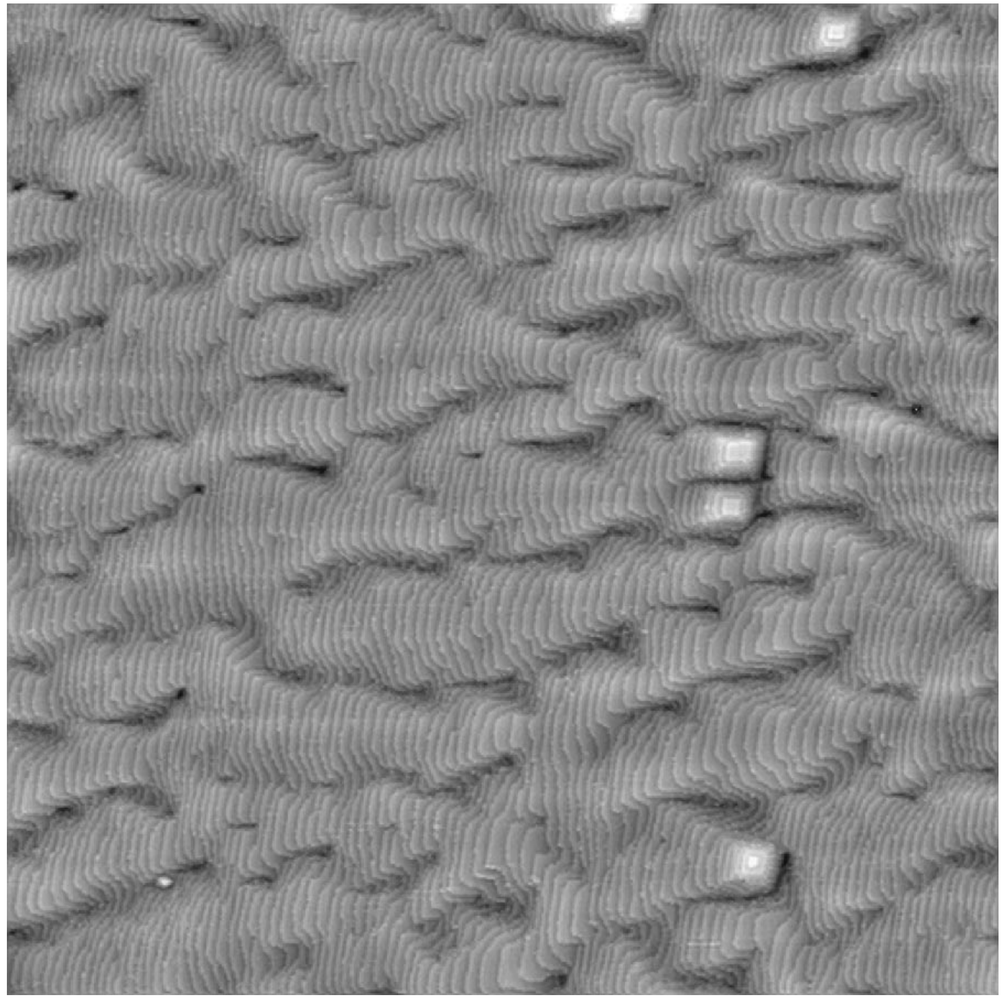
Cu (1,1,17)

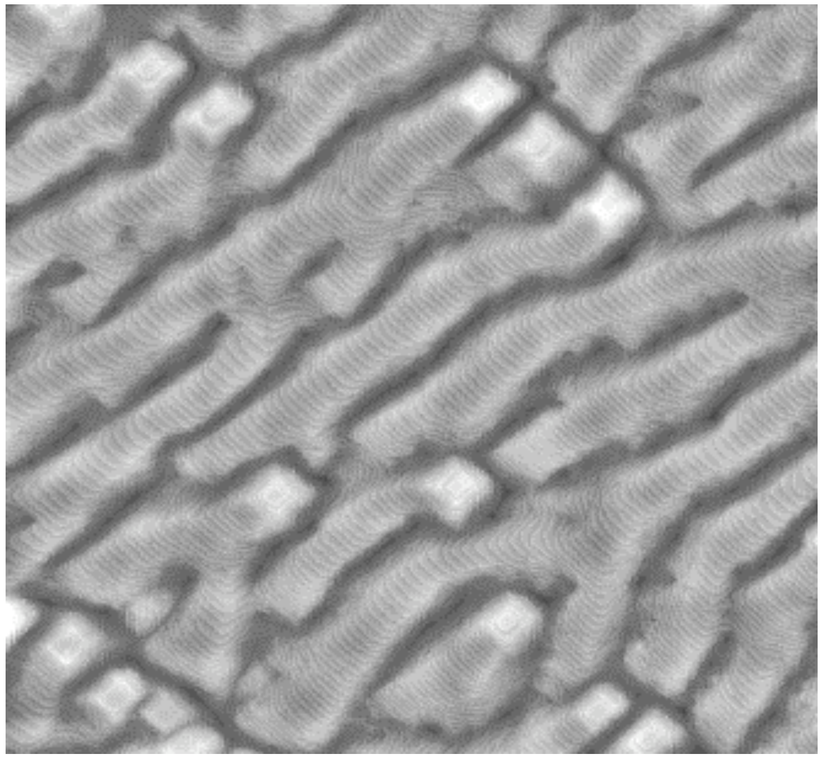




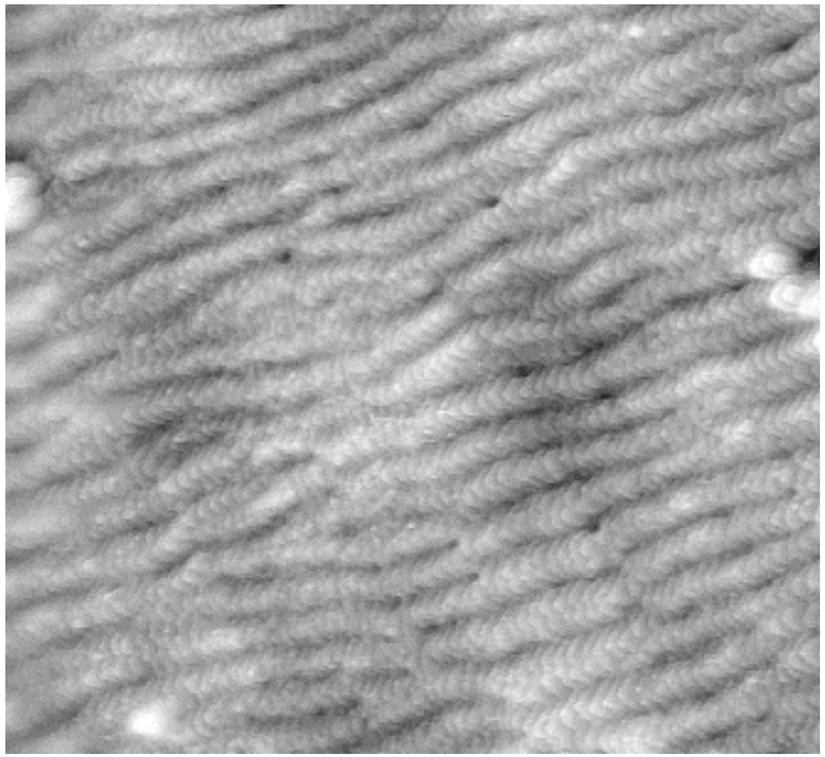
Cu (1,1,17)



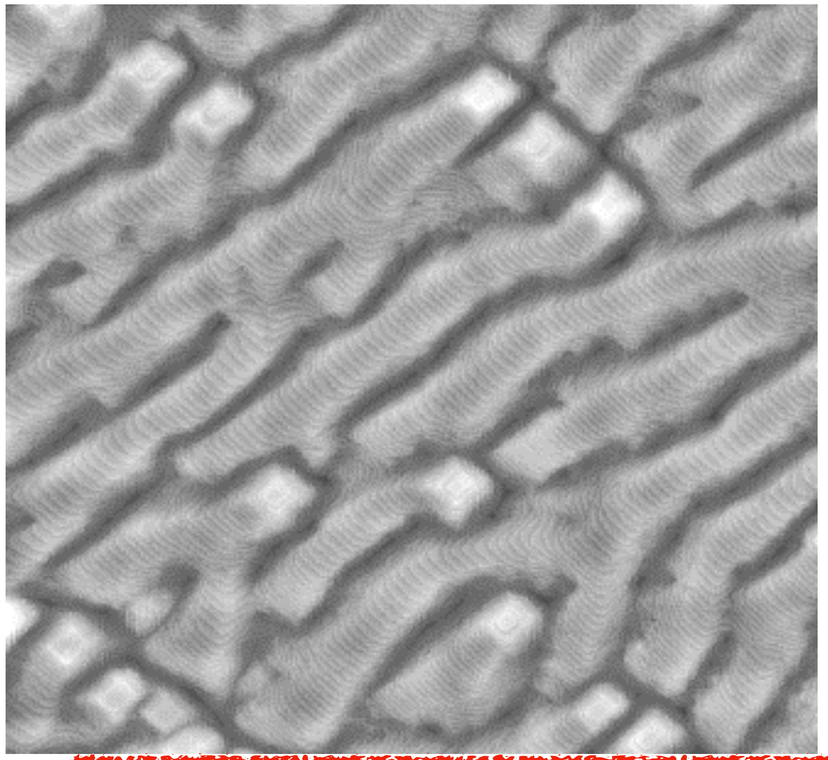


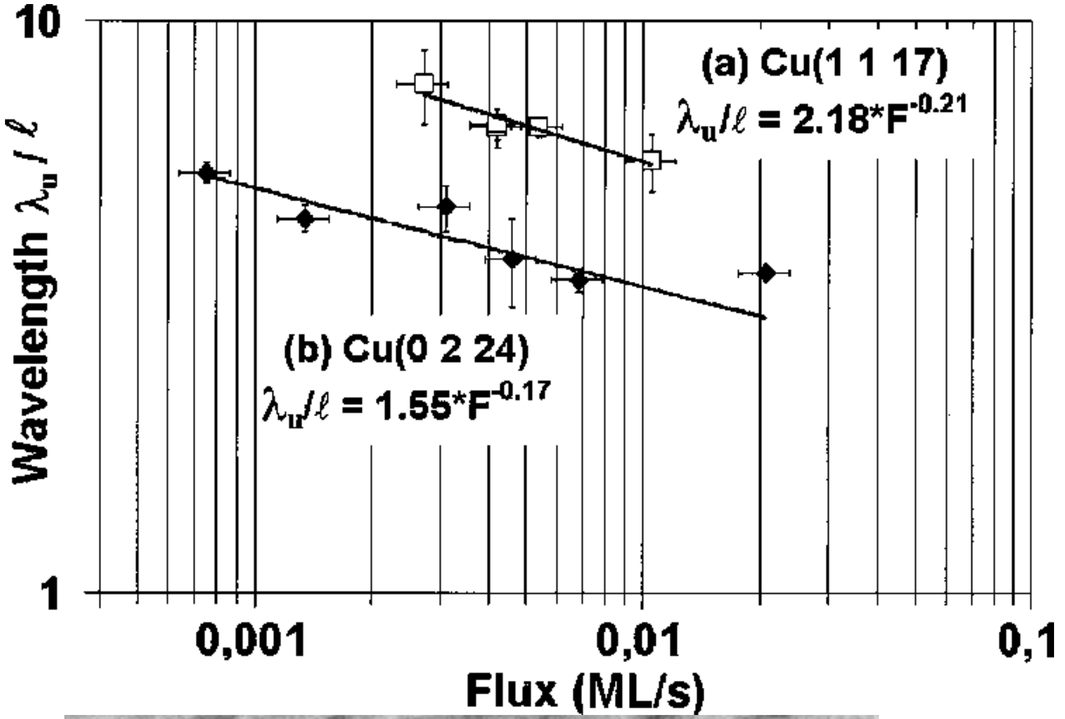


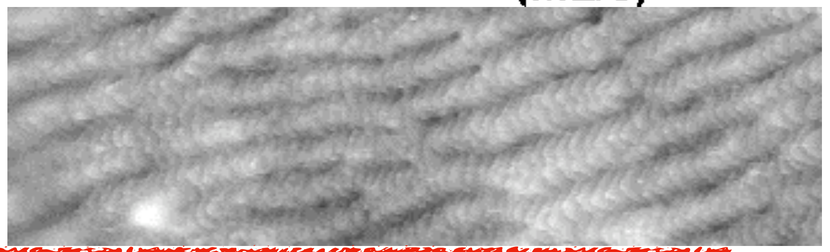
Cu (1,1,17)



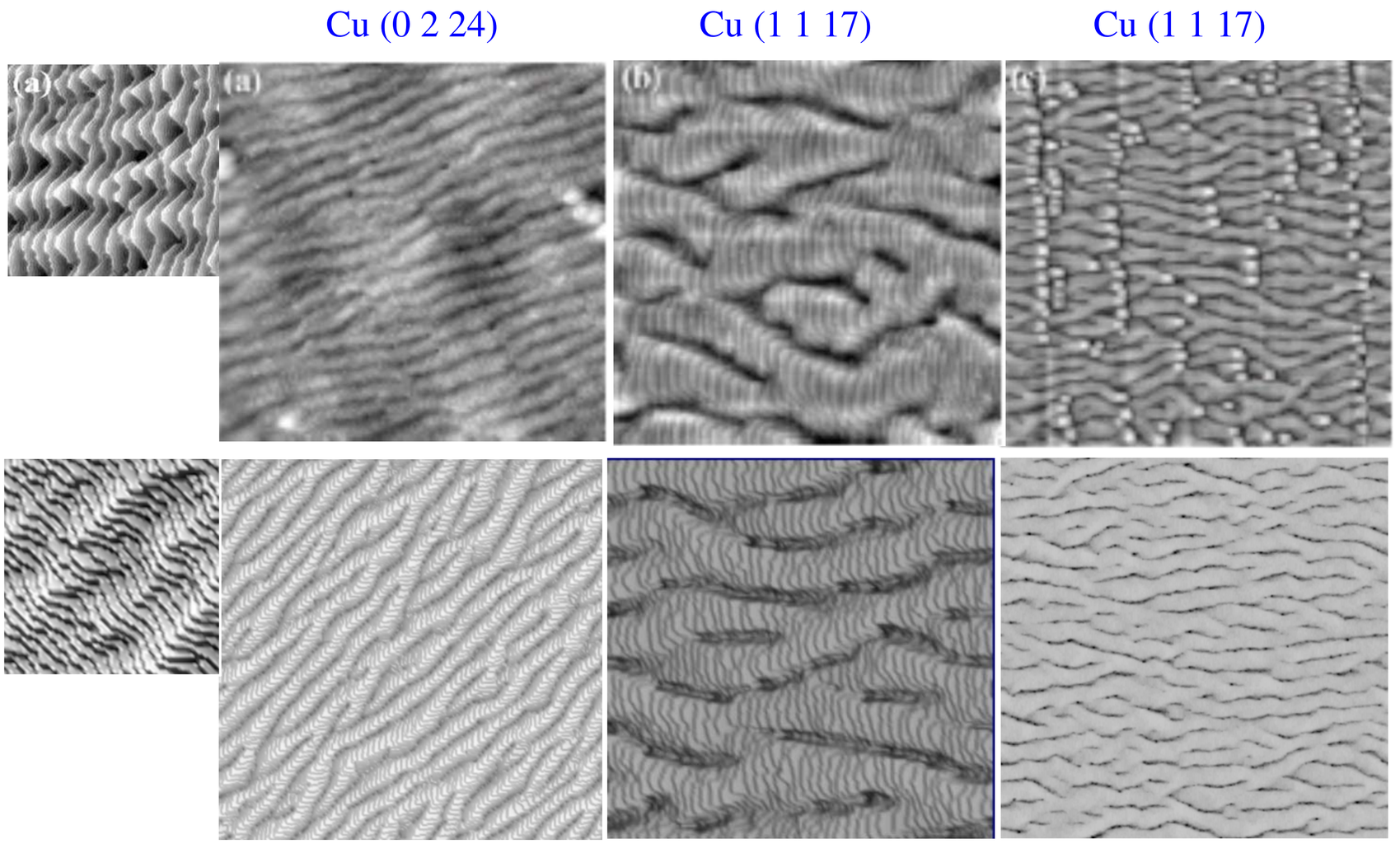
Cu (0,2,24)







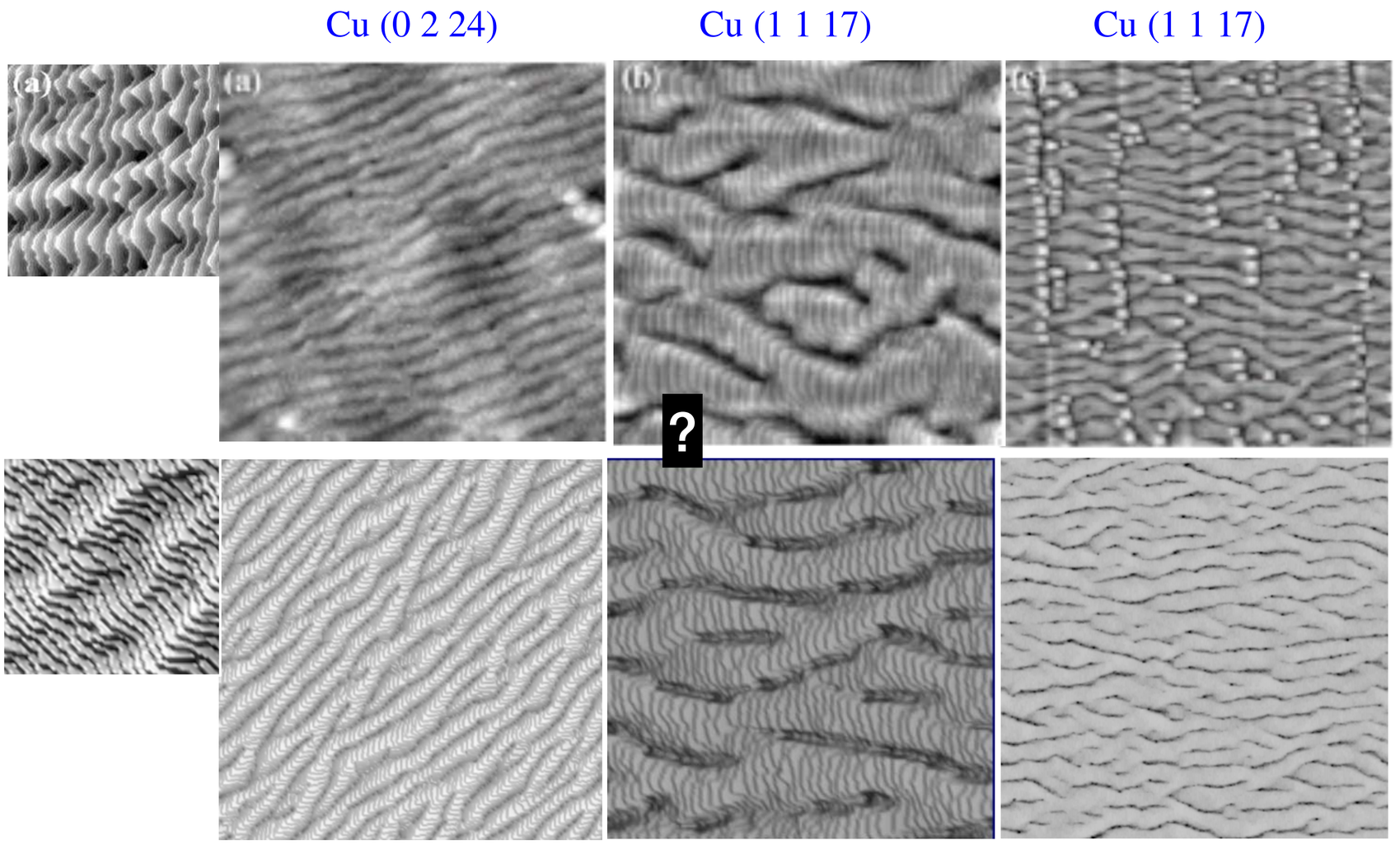
Cu vicinals — unstable growth:
meandering steps (?) + pyramids (?)



800x800, L=15, T=250K, F=5e-3ML/s
E_b=0.1eV, E_d=0.4eV, E_a=0.15eV,
20 ML

360x360, L=5, T=280K, F=5e-3ML/s
E_b=0..07eV, E_d=0.4eV, E_a=0.12eV,
20 ML

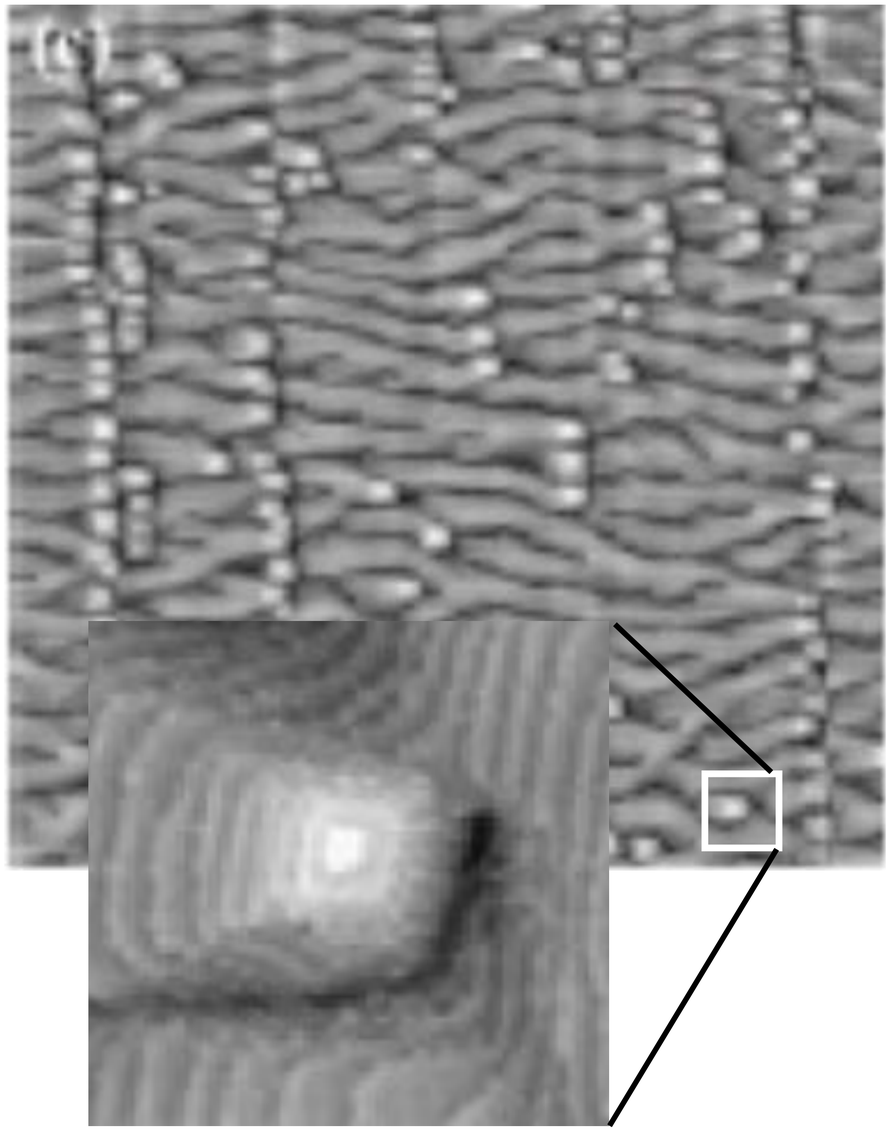
800x800, L=5, T=285K, F=5e-2ML/s,
E_b=0..07eV, E_d=0.4eV, E_a=0.12eV,
40 ML

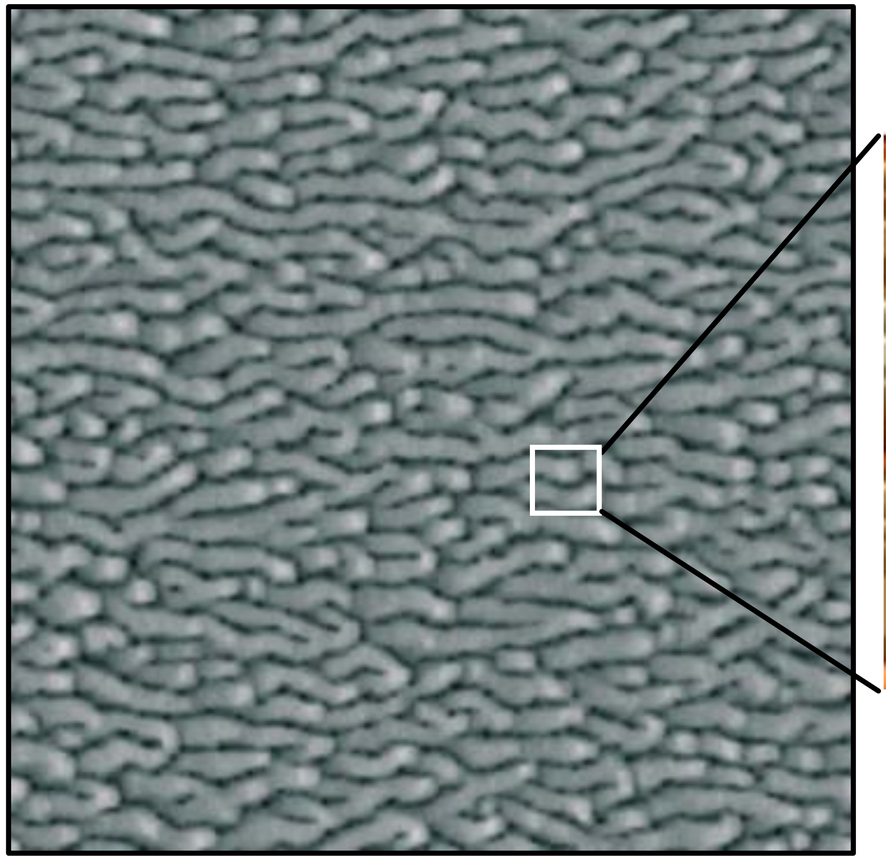


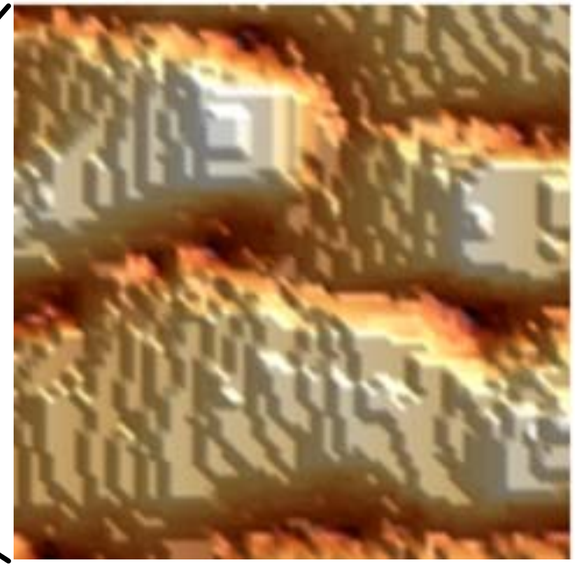
800x800, L=15, T=250K, F=5e-3ML/s
E_b=0.1eV, E_d=0.4eV, E_a=0.15eV,
20 ML

360x360, L=5, T=280K, F=5e-3ML/s
E_b=0..07eV, E_d=0.4eV, E_a=0.12eV,
20 ML

800x800, L=5, T=285K, F=5e-2ML/s,
E_b=0..07eV, E_d=0.4eV, E_a=0.12eV,
40 ML







Add an impurity

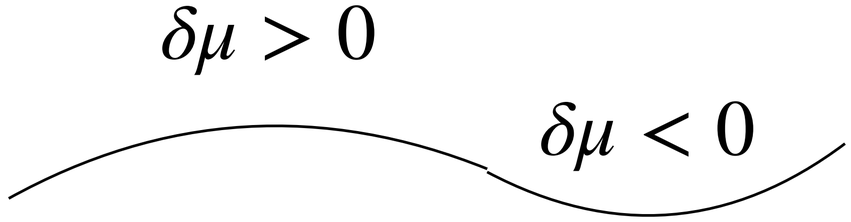
DFT points to W

Imp.	Cu	C (Set 1)	Al (Set 2)	Ni (Set 3)	W (Set 4)
γ	0.45 ± 0.05	0.35 ± 0.06	0.50 ± 0.06	0.41 ± 0.05	0.17 ± 0.02
Zoom View					

FIG. 2. (Color online) Rough estimate of the exponent γ ($\lambda_m \sim F^{-\gamma}$) and of the possibility of pyramid formation for pure Cu and for Cu codeposited with a member of each of the four sets of impurities, the impurity being 2% of the flux. Simulations are done for five values of F: 0.005, 0.01, 0.05, 0.1, 0.5 ML/s.[41] For pure Cu (Cu-Cu) γ is consistent with the Bales-Zangwill[7] value ($\gamma = 0.5$) while for Cu-W ($\gamma = 0.17$) it is in the range of the experiment.[3] The zoomed views (color online) are 7% of images as in Fig. 1: after 40 ML are deposited with $F = 0.05$ ML/s at $T = 425$ K.

Growth Instabilities at Long Length Scales

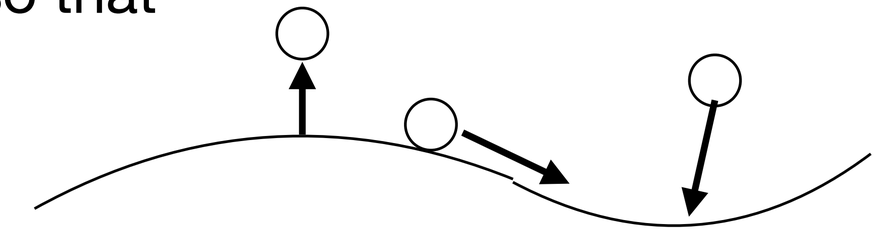
- At scales $\ell \gg a$, where a is the lattice spacing, a crystal surface can be treated as a continuous object $z = z(x, y)$
- Assuming as reference state a flat surface, $z(x, y) = z_0$, an undulating surface is characterized by an excess chemical potential, proportional to the local curvature $\nabla^2 z$: $\delta\mu = -\tilde{\sigma}\left(\frac{\partial^2 z}{\partial x^2} + \frac{\partial^2 z}{\partial y^2}\right)$



Growth Instabilities at Long Length Scales

- Linear thermodynamics yields two types of dynamics, corresponding to two different transport modes:
  - Non conserved: $\dot{z} = -K\delta\mu = K\tilde{\sigma}\nabla^2 z$
  - Conserved: $\dot{z} = -\mathrm{div}\vec{J};\ \vec{J} = -D_s\vec{\nabla}\delta\mu$, so that

$$\dot{z} = -\tilde{\sigma}D_s\nabla^4 z$$

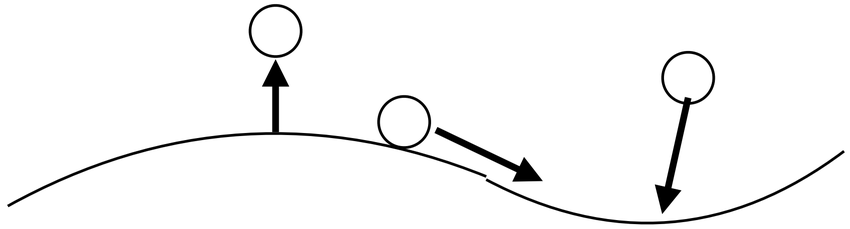


- These equations can describe the growth, dissolution or equilibration kinetics of a rough surface at long length scales

Growth Instabilities at Long Length Scales

- In particular, if $\tau(L)$ is the characteristic time scale of the surface evolution at length scale L:
  - Non conserved: $\dot{z} = K\tilde{\sigma}\nabla^2 z$ implies $\tau(L) \sim L^2$
  - Conserved: $\dot{z} = -\tilde{\sigma}D_s\nabla^n z$ implies $\tau(L) \sim L^n$
  - Ex. Kardar-Parisi-Zhang Eq.

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



Growth Instabilities at Long Length Scales

The surface morphology can be quantified by measuring its height-height correlation function

$$G(r,t) = \langle \overline{[h(x+r,t) - h(x,t)]^2} \rangle$$

Such function is observed to possess the scaling form

$$G(r,t) = t^{2\beta} g(r/\xi)$$

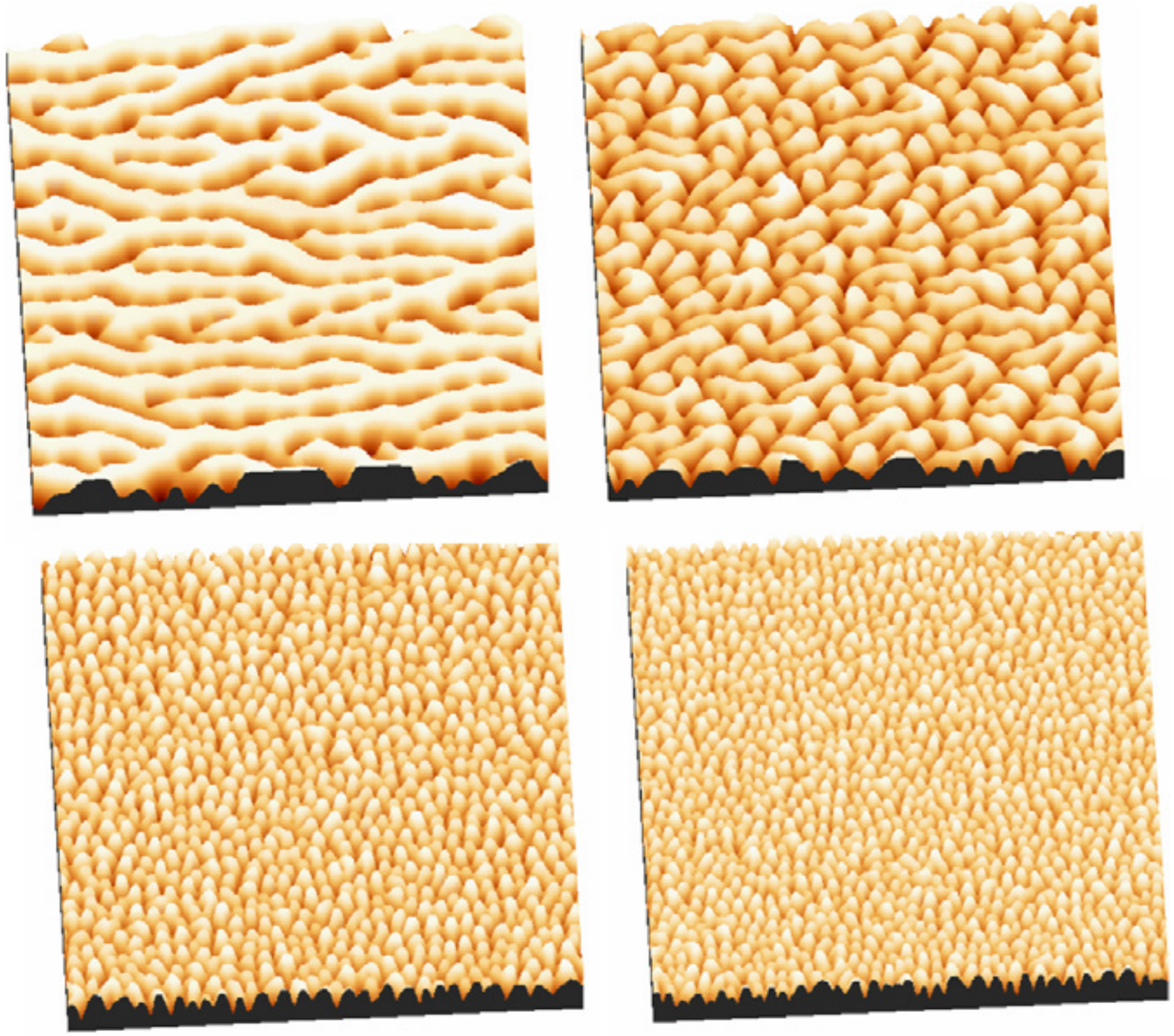
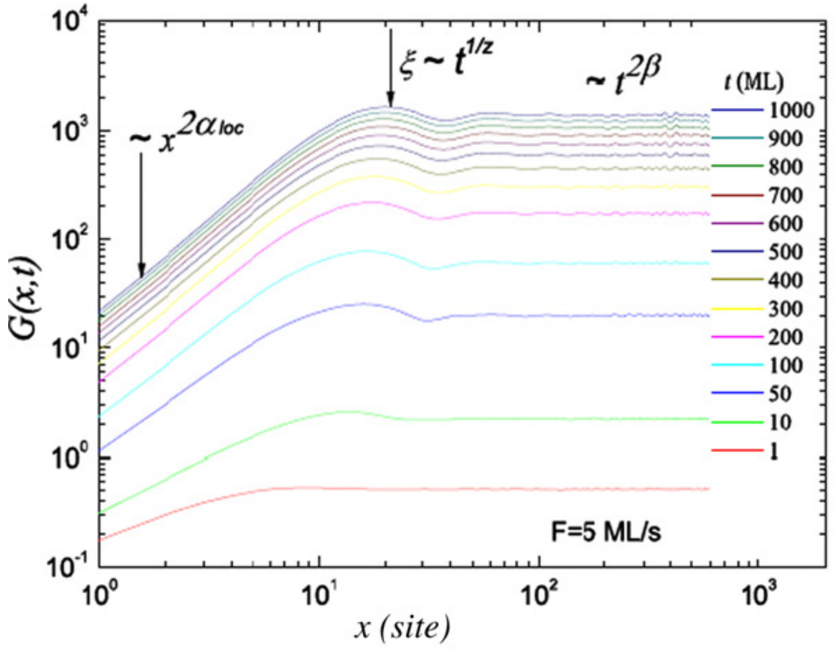
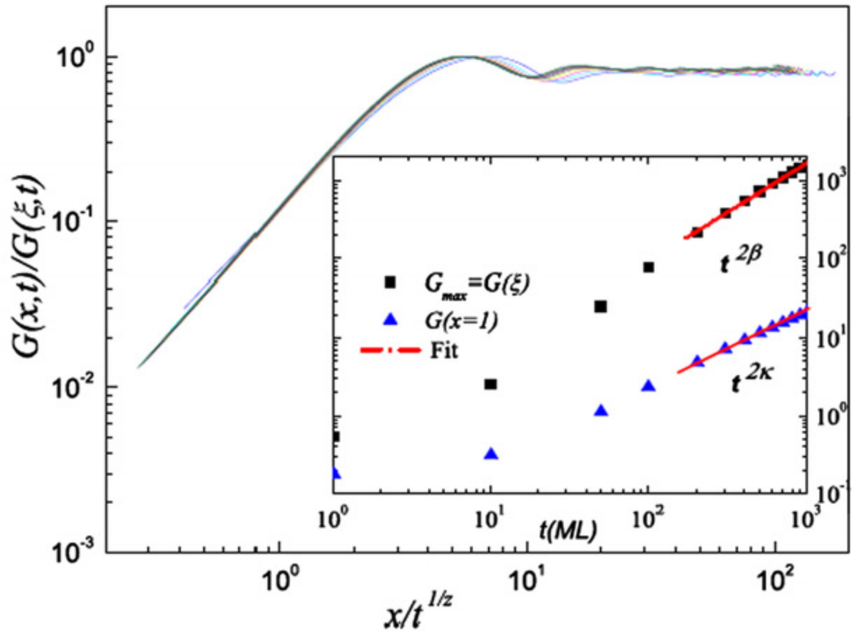


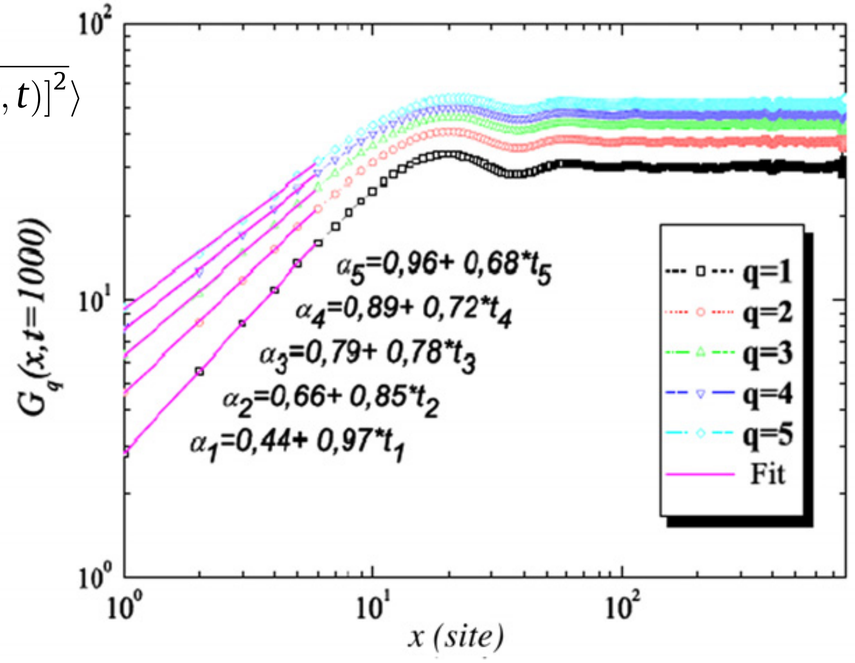
Fig. 5. Monte Carlo simulation images of the surface morphology evolution with increasing deposition flux: $F = 1$, 5, 20, 40 ML/s. The initial surface is a vicinal (800×800) sites (in lattice unit) with terrace width $L = 5$ sites.





$$G(r,t) = t^{2\beta} g(r/\xi)$$

$$G(r,t) = \langle \overline{[h(x+r,t) - h(x,t)]^2} \rangle$$



$$\frac{\partial}{\partial t} h(x,y,t) = v_x \frac{\partial^6}{\partial x^6} h(x,y,t) + v_y \frac{\partial^6}{\partial y^6} h(x,y,t) + \eta(x,y,t),$$

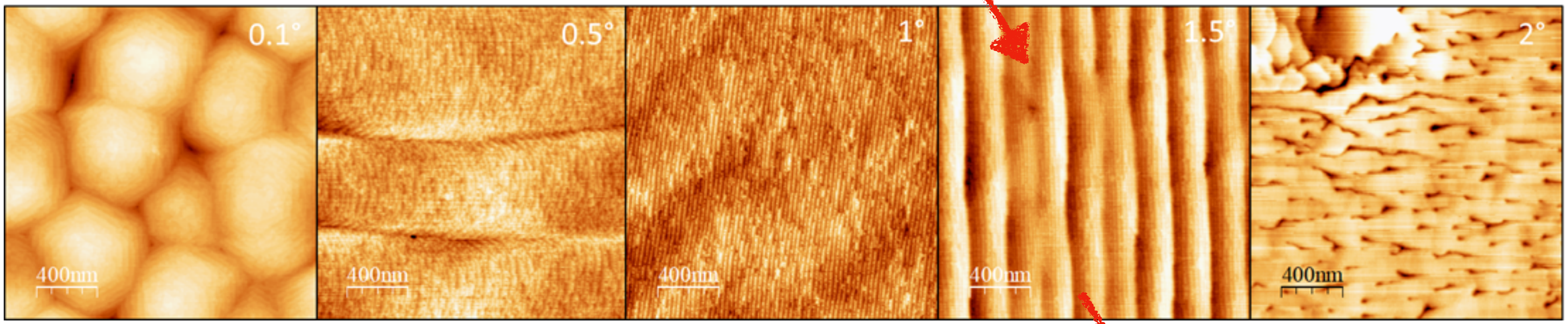
with $< \eta(x,y,t) >= 0,$

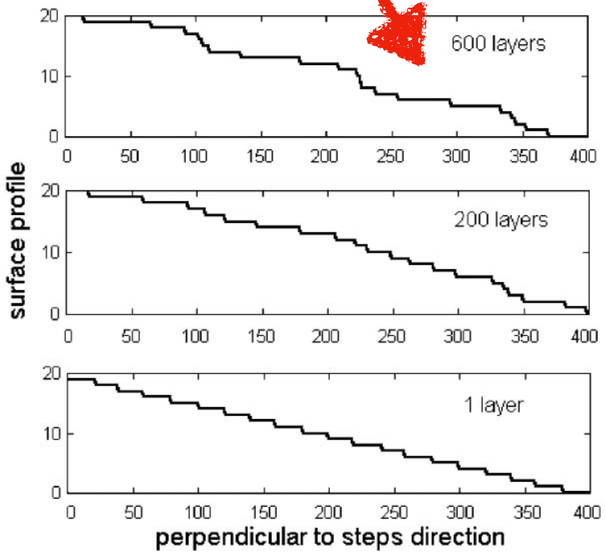
and

$$\langle \eta(x,y,t)\eta(x',y',t') \rangle = D((x-x')^2 + (y-y')^2)^{(2\rho-1)/2} |t-t'|^{2\theta-1}.$$

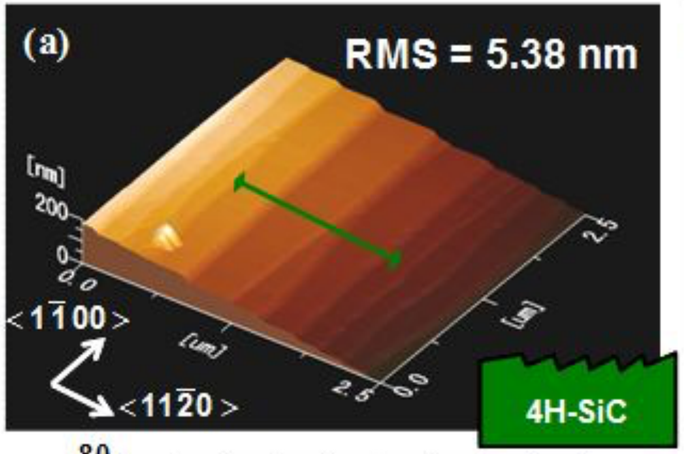
Multiscaling!

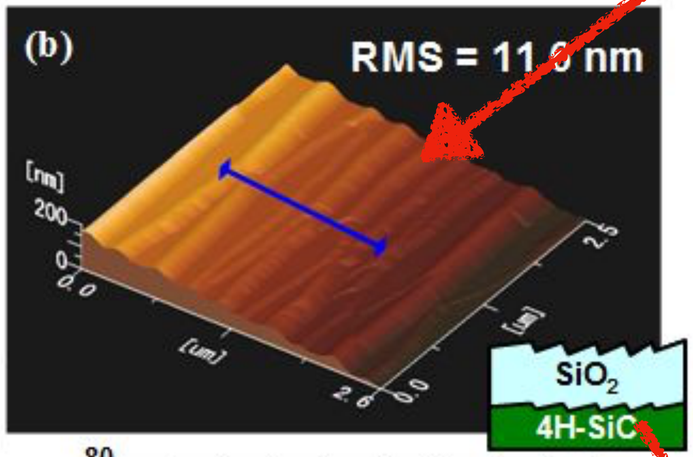
Other Instabilities: Step Bunching

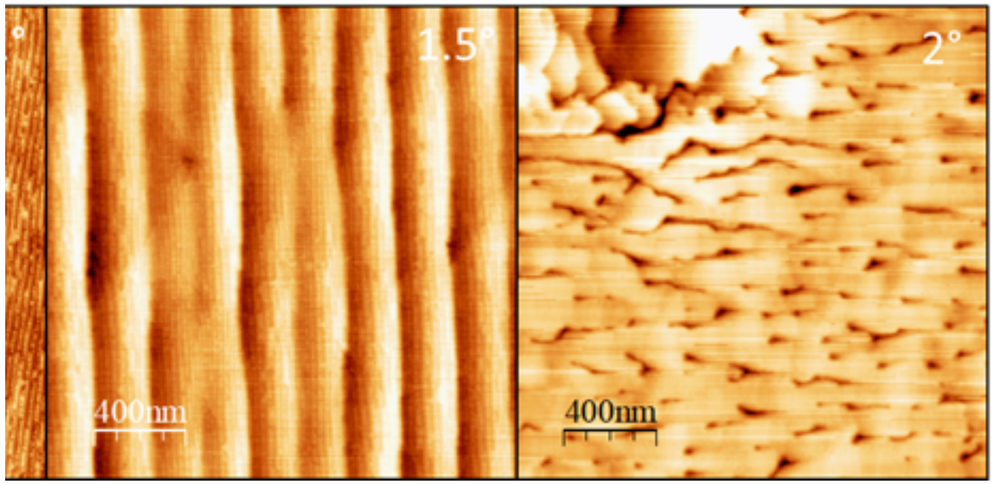


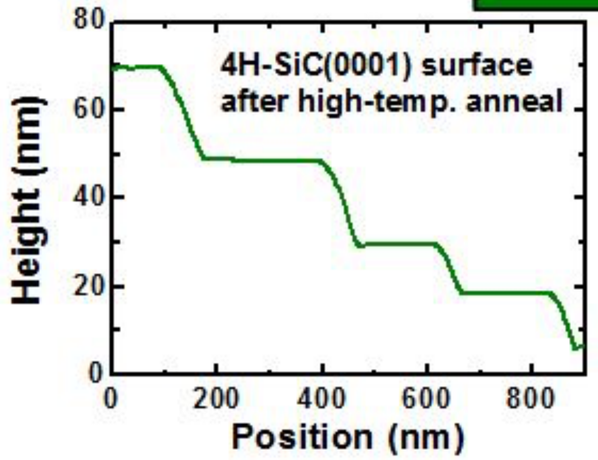


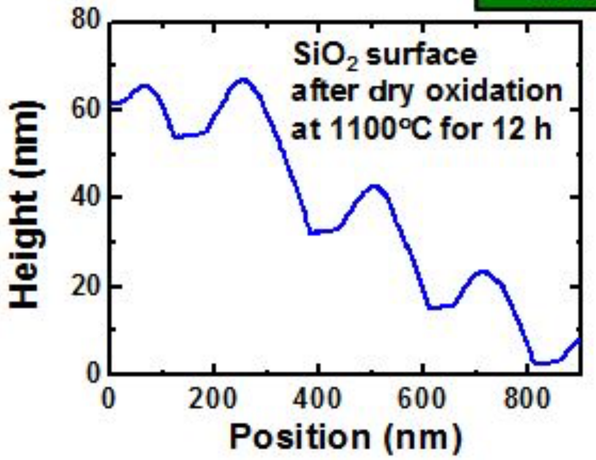
Other Instabilities: Step Bunching

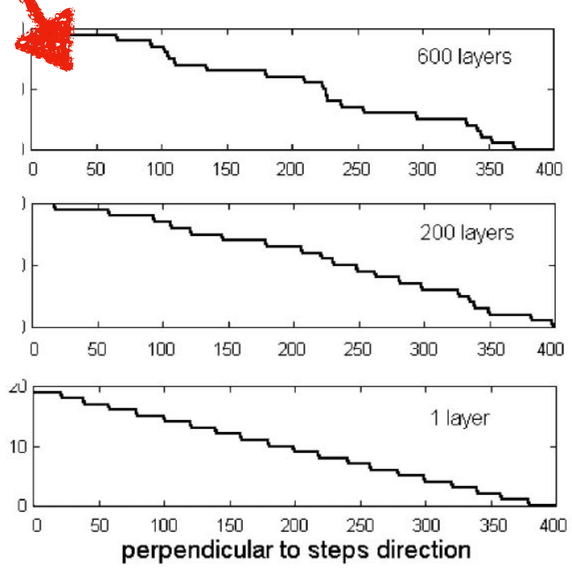












Inverse Schwoebel effect (iES)

R.L. Schwoebel, J. Appl. Phys. 40 (1968) 614.

Bunching: Net current “down”

$$J_{\text{down}} > J_{\text{up}}$$

Inverse Schwoebel effect (iES)

$$v_n = F(x_n - x_{n-1})$$

Extreme case: infinite iES effect

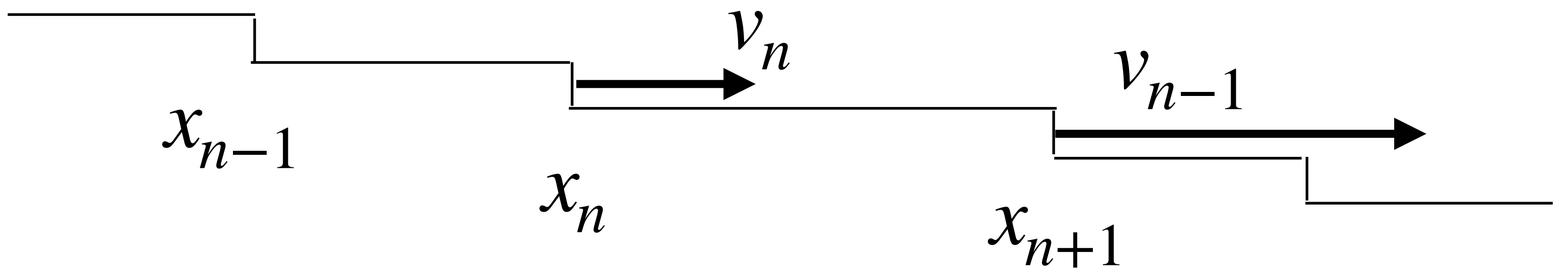
Net current "down"

$$J_{\text{down}} > J_{\text{up}}$$

R.L. Schwoebel, J. Appl. Phys. 40 (1968) 614.

Inverse Schwoebel effect (iES)

$$v_n = F(x_n - x_{n-1})$$



Atoms can only “step down”

$$J_{\text{up}} = 0$$

R.L. Schwoebel, J. Appl. Phys. 40 (1968) 614.

Inverse Schwoebel effect (iES)

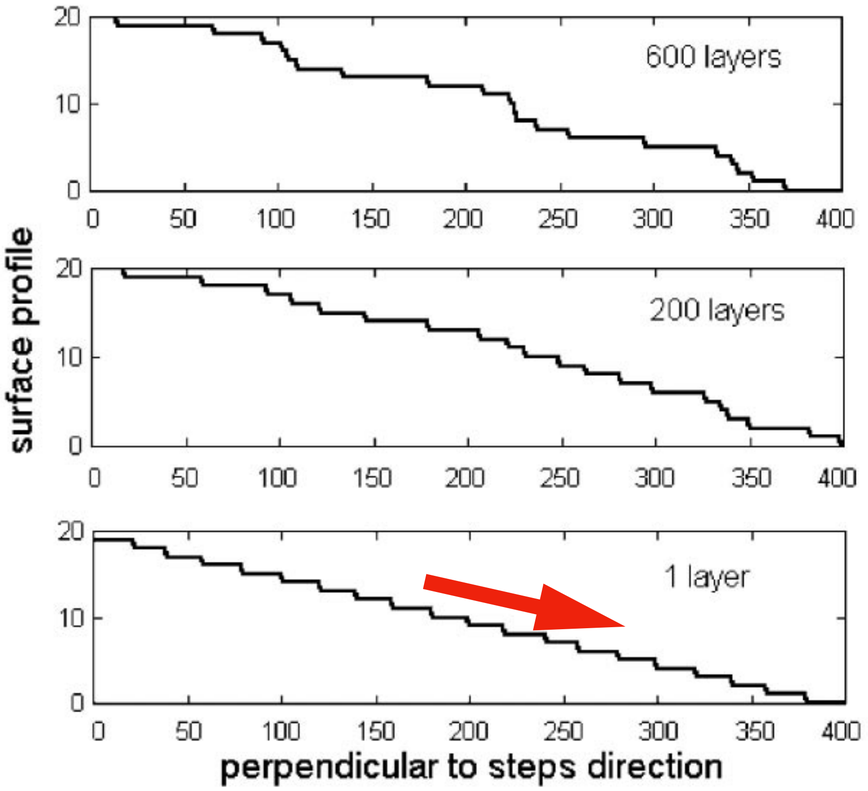
$$v_n = F(x_n - x_{n-1})$$

Net current "down"

$$J_{\text{down}} > J_{\text{up}}$$

R.L. Schwoebel, J. Appl. Phys. 40 (1968) 614.

Inverse Schwoebel effect



$$J_{\text{down}} > J_{\text{up}}$$

Net current “down”

Steps “attract”

Leads to step bunching

R.L. Schwoebel, J. Appl. Phys. 40 (1968) 614.

Common wisdom is:

- Surface current “up”: step meandering;
- Surface current “down”: step bunching

Bunching and meandering cannot coexist

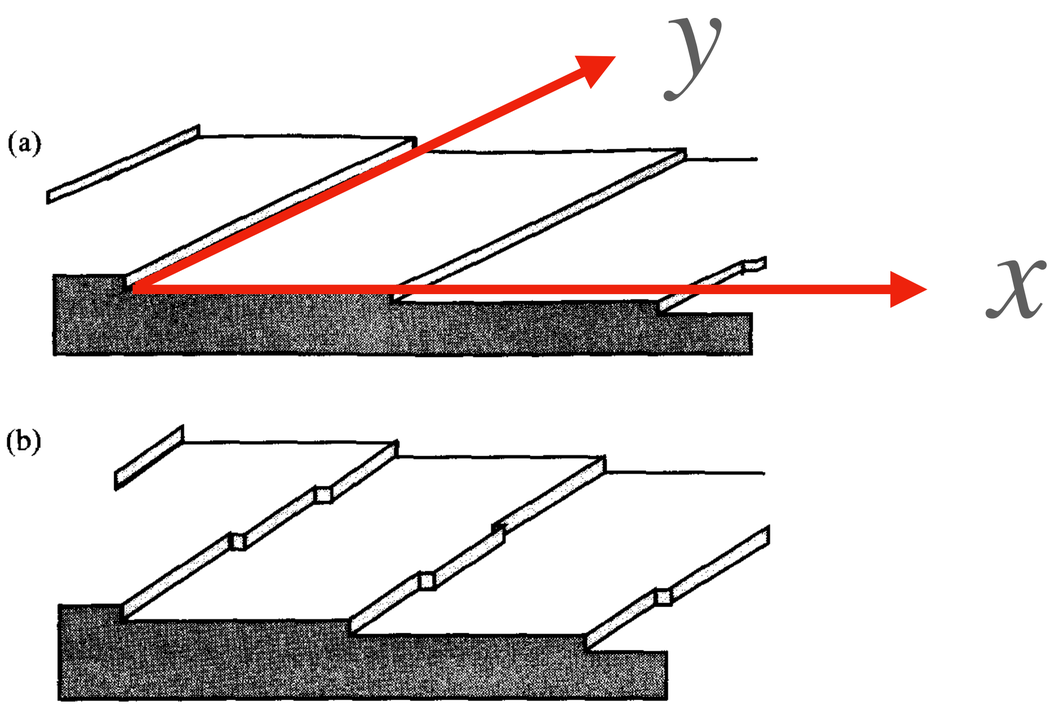
Common wisdom is:

- Surface current “up”: step meandering;
- Surface current “down”: step bunching

Linear stability analysis

Common wisdom is:

- Surface current “up”: step meandering;
- Surface current “down”: step bunching



Common wisdom is:

- Surface current “up”: step meandering;
- Surface current “down”: step bunching

$$\frac{\partial h}{\partial t} = \nu_{\parallel} \frac{\partial^2 h}{\partial y^2} + \nu_{\perp} \frac{\partial^2 h}{\partial x^2}$$

Common wisdom is:

- Surface current “up”: step meandering;
- Surface current “down”: step bunching

Common wisdom is wrong

$$\nu_{\parallel} = \ell J(\ell)$$

step meandering if $\nu_{\parallel} < 0$

$$\nu_{\perp} = -\ell^2 \frac{dJ(\ell)}{d\ell}$$

step bunching if $\nu_{\perp} < 0$

Common wisdom is:

- Surface current “up”: step meandering;
- Surface current “down”: step bunching

Common wisdom is wrong

$$\nu_{\parallel} = \ell J(\ell)$$

step meandering if $\nu_{\parallel} < 0$

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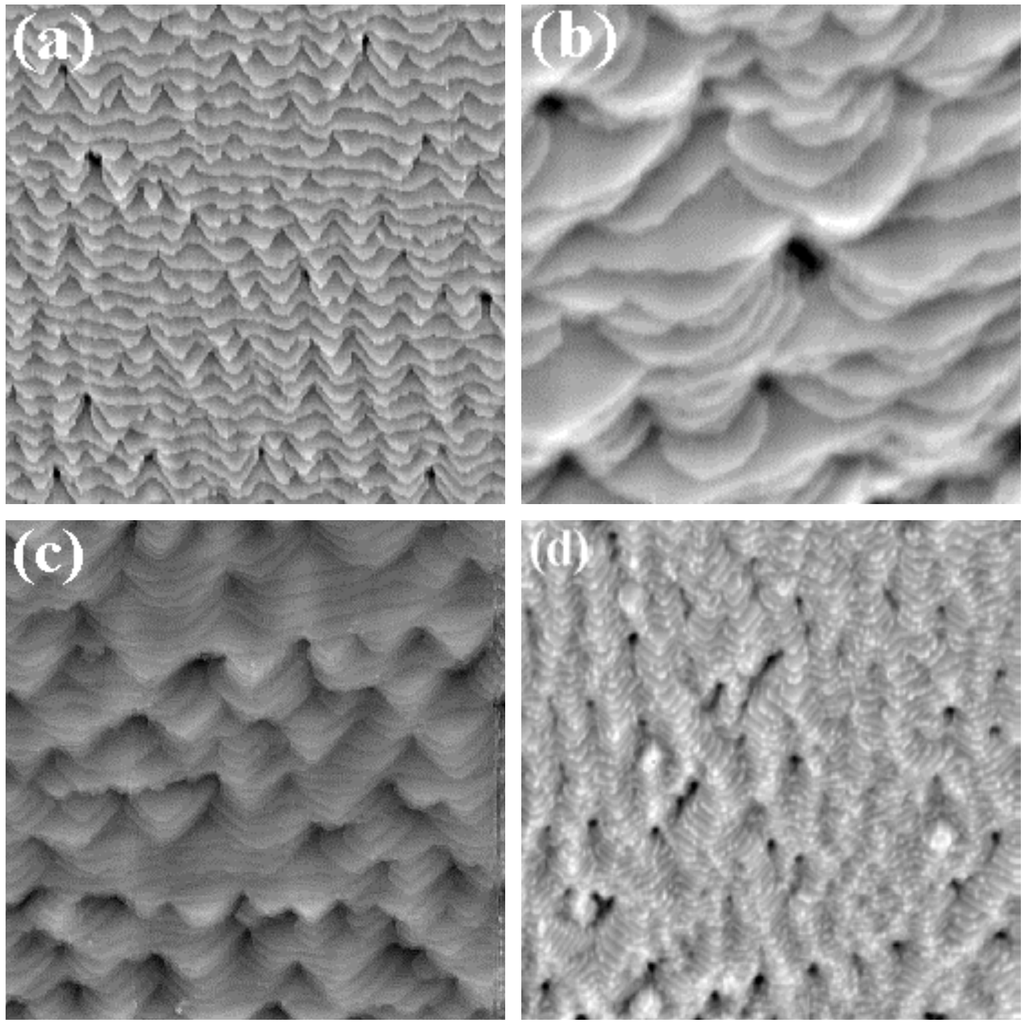
step bunching if $\nu_{\perp} < 0$

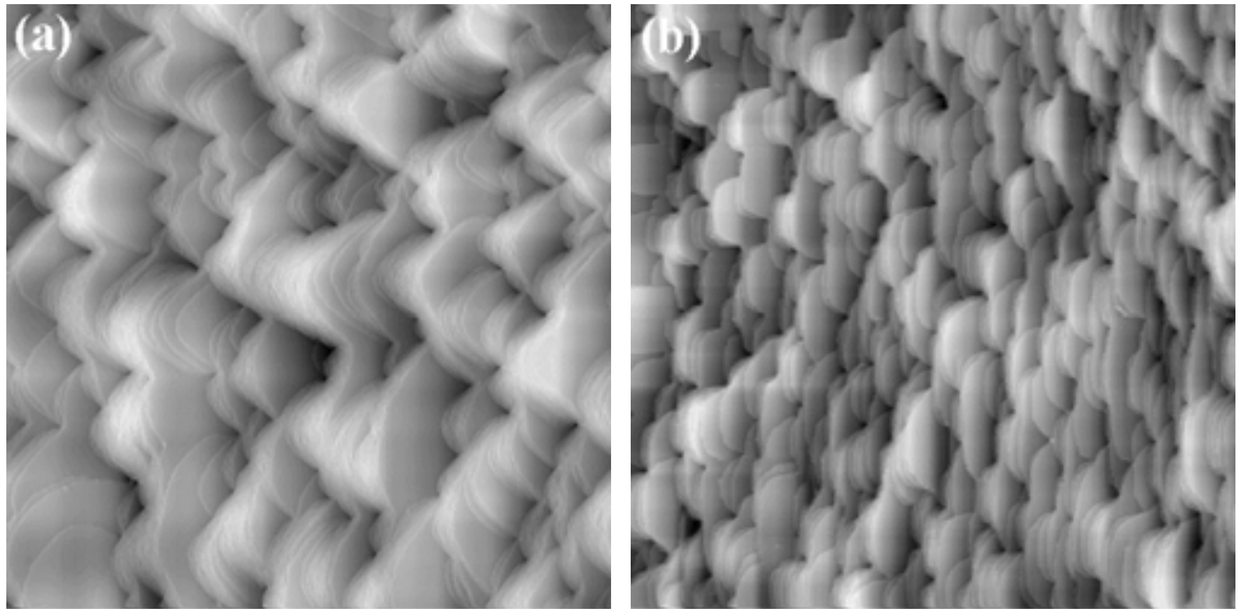
Bunching and meandering can coexist

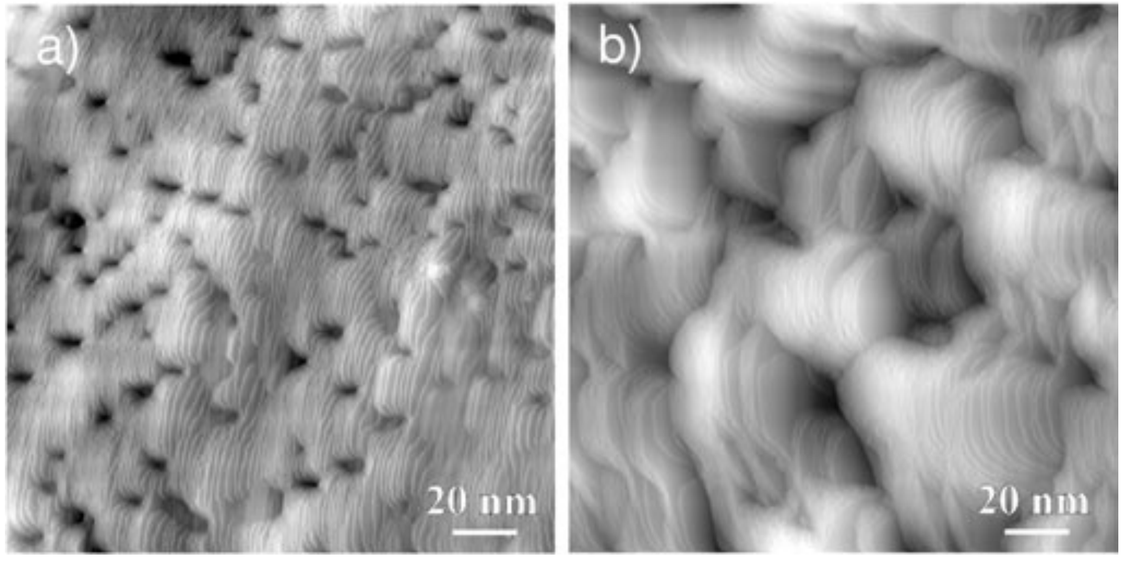


Spontaneous structural pattern formation at the nanometre scale in kinetically restricted homoepitaxy on vicinal surfaces

N Néel, T Maroutian, L Douillard and H-J Ernst

CEA Saclay, DSM/Drecam/Spcsi, 91191 Gif Sur Yvette, France





Complex growth: multiple components

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JOURNAL OF CRYSTAL GROWTH

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A new model of morphological instabilities during epitaxial growth: from step bunching to mounds formation

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[b]LASMEA, UMR 6602 CNRS/Université Blaise Pascal, Clermont 2, F-63177 Aubière Cedex, France

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Two-particle surface diffusion-reaction models of vapour-phase epitaxial growth on vicinal surfaces

Alberto Pimpinelli*, Robert Cadoret, Evelyne Gil-Lafon, Jérôme Napierala, Agnès Trassoudaine

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Direct condensation modelling for a two-particle growth system: application to GaAs grown by hydride vapour phase epitaxy

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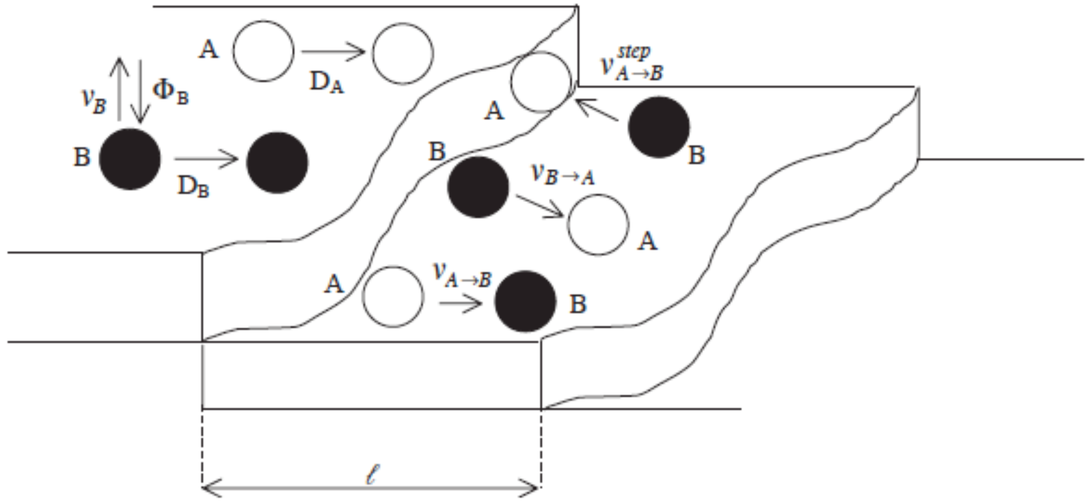
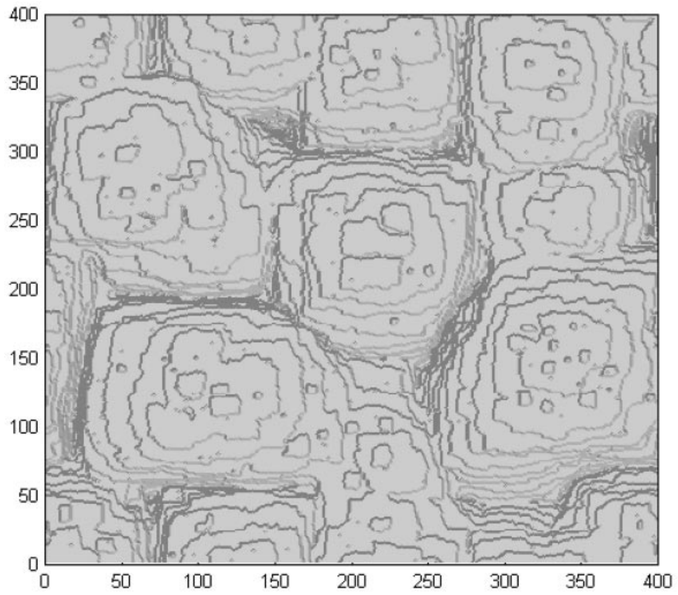
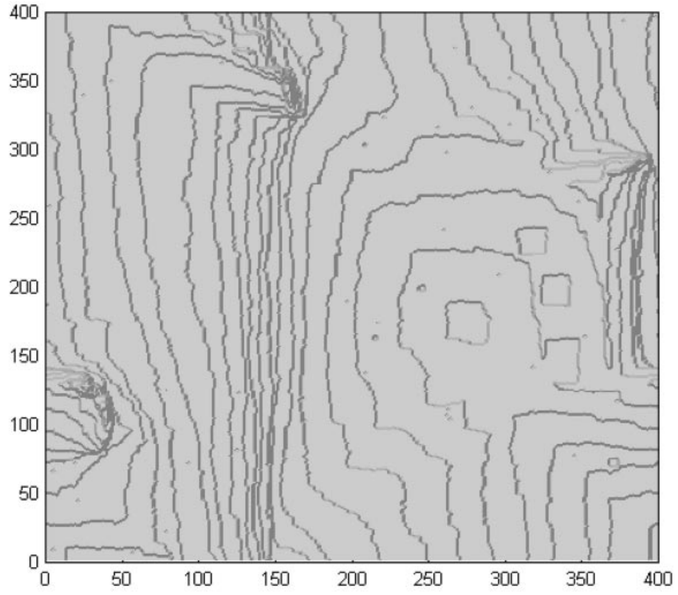


Mounds

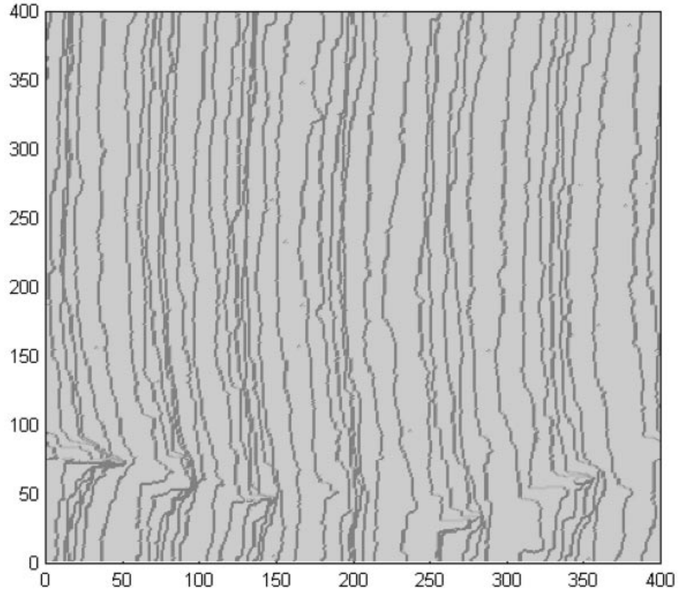


(a)

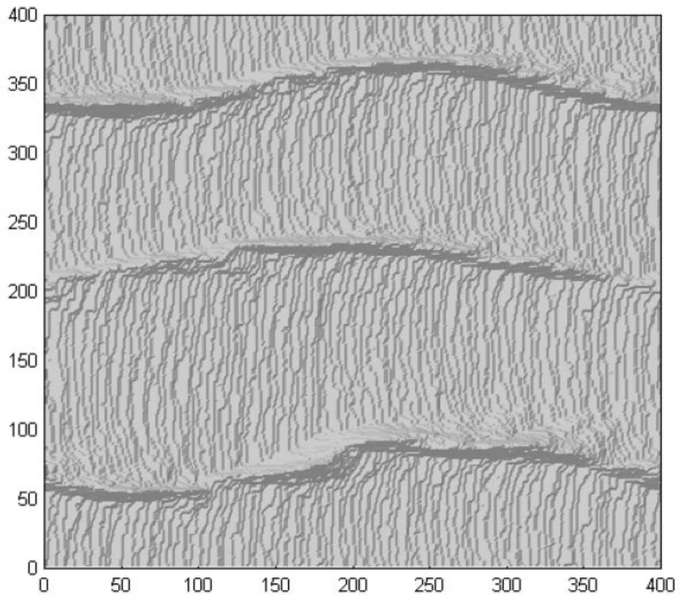


(b)

Bunches



(c)



(d)

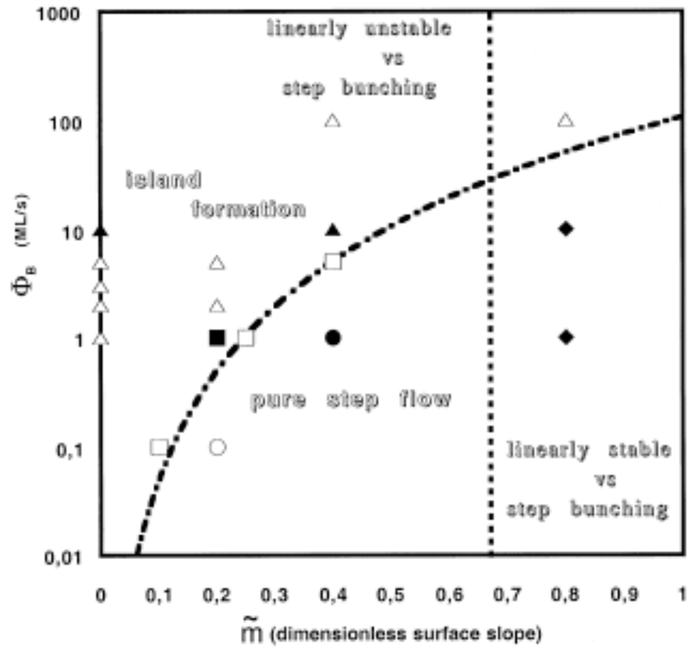
Meanders

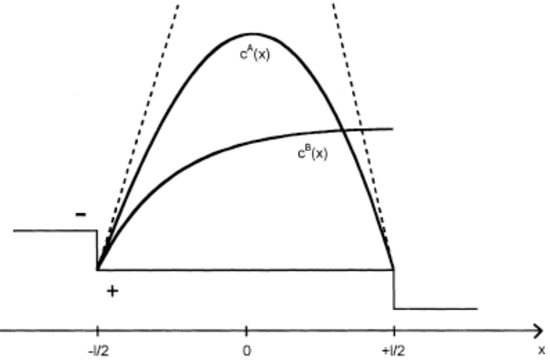
Université Blaise Pascal

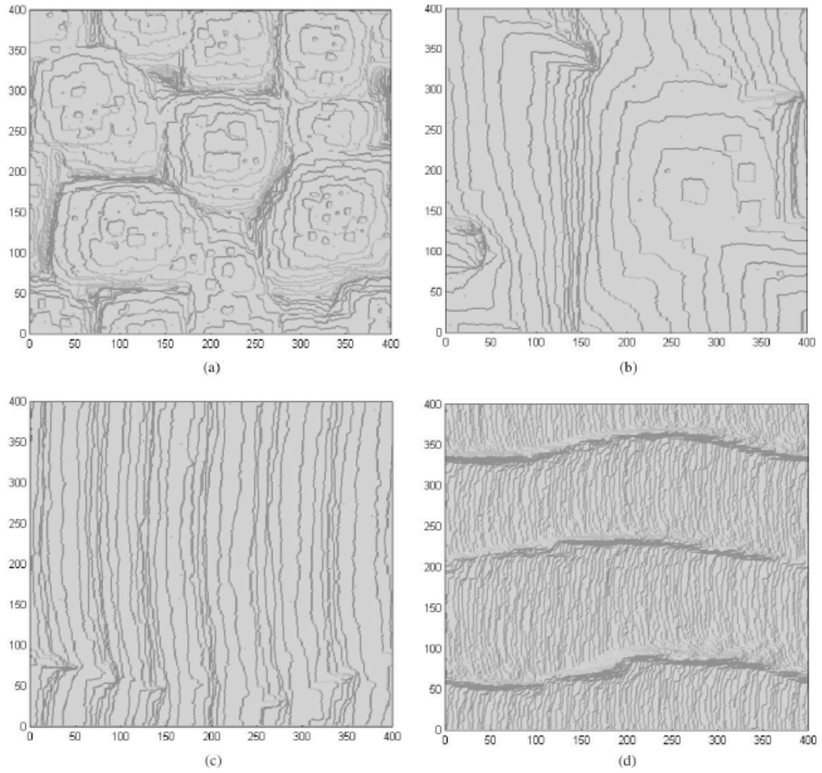
ES barrier at step edges implies coexistence of mounding, bunching *and* meandering...

$$D_B \nabla^2 c^B - v_B c^B + \Phi_B = 0,$$

$$D_A \nabla^2 c^A + v_B c^B = 0,$$



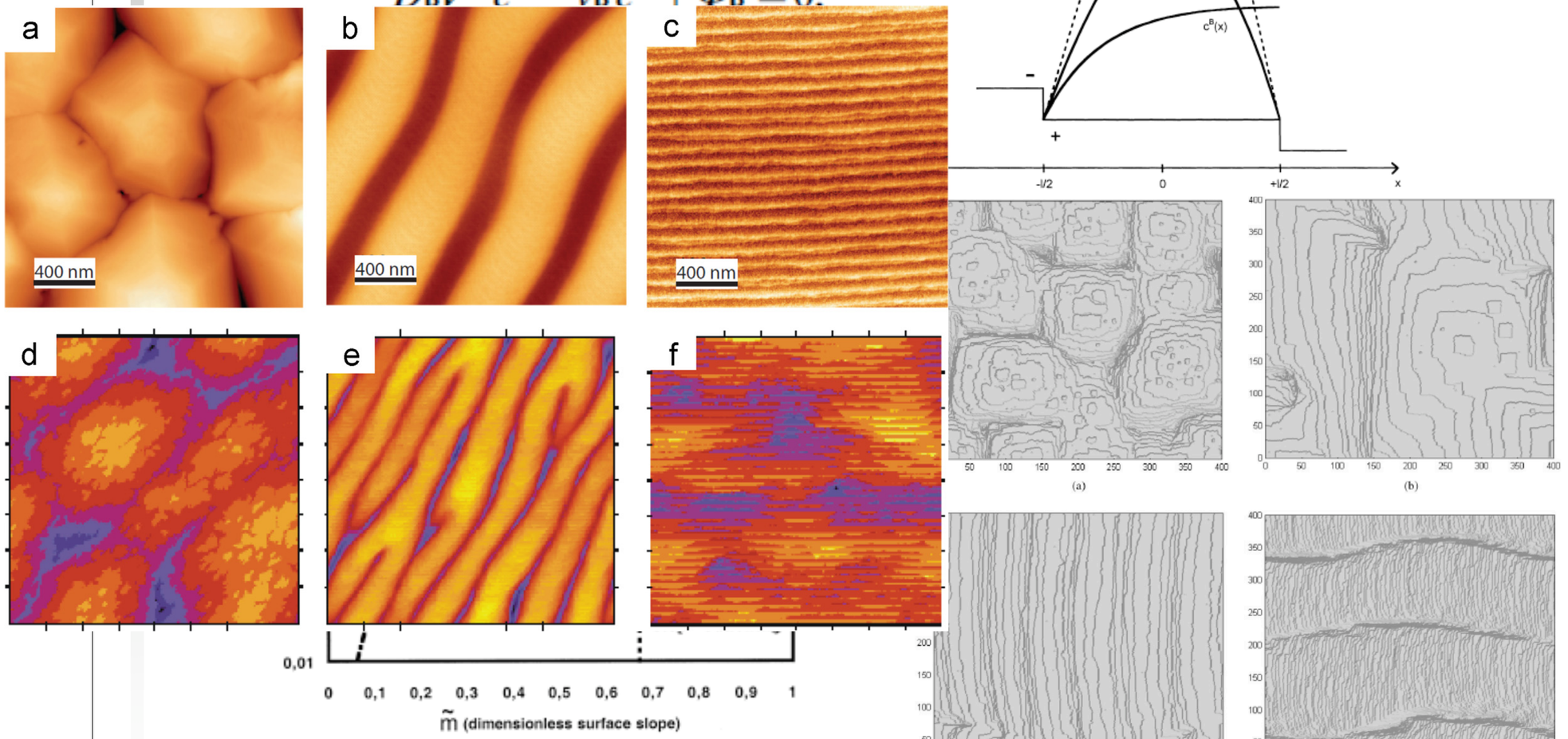




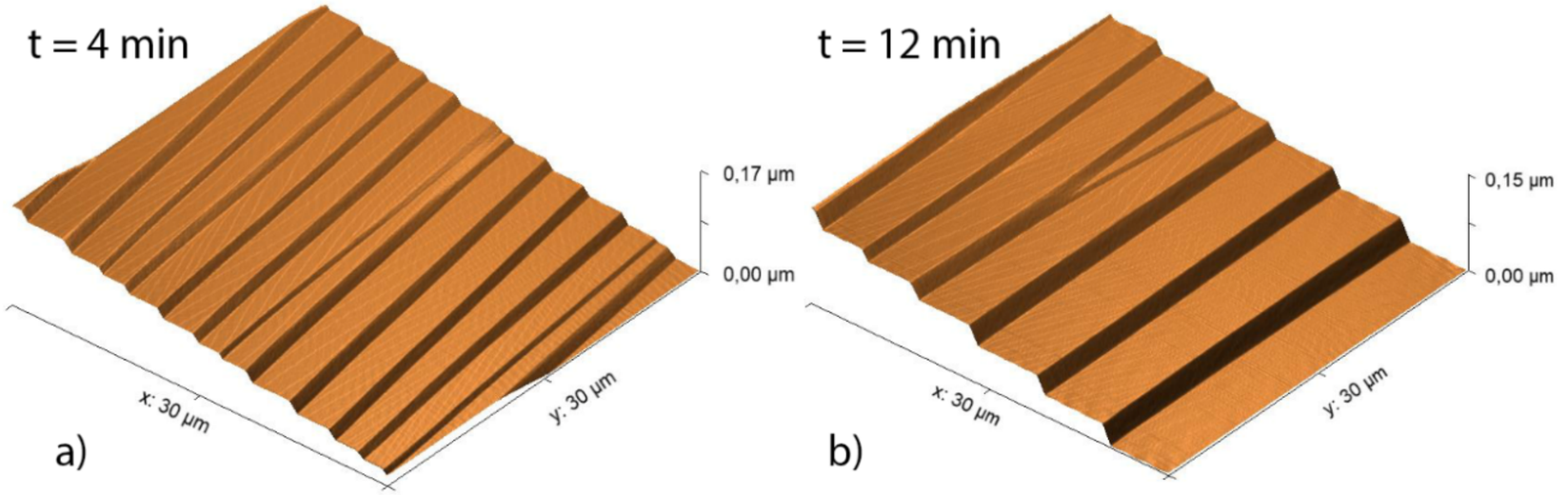
Université Blaise Pascal

ES barrier at step edges implies coexistence of mounding, bunching *and* meandering...

$$D_B \nabla^2 c^B - \nu_B c^B + \Phi_B = 0.$$

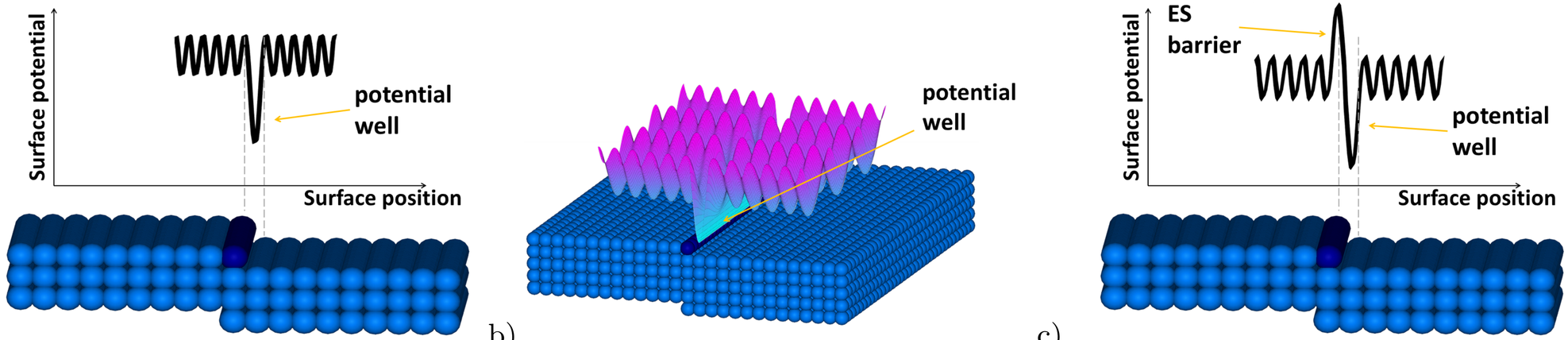


More Exotic Instabilities: Electromigration



AFM images of sublimating Si(111) vicinal surface,
miscut in the [11-2] direction by 0.3°,
heated by step-down direct current for 4 min (a) and 12 min (b).

More Exotic Models: Cellular Automata



Step meandering: The balance between the potential well and the Ehrlich – Schwoebel barrier

Marta A. Chabowska[1],* Hristina Popova[2], and Magdalena A. Załuska-Kotur[1]

More Exotic Models: Cellular Automata

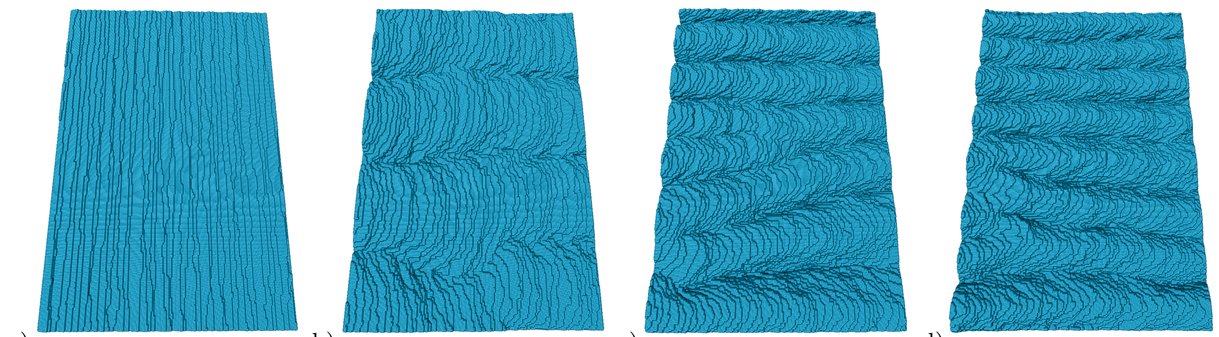


FIG. 2. Meanders obtained for $c_0 = 0.003$, $l_0 = 5$, $n_{DS} = 5$ and a) $\beta E_V = 0.0$, b) $\beta E_V = 2.0$, c) $\beta E_V = 3.5$, d) $\beta E_V = 6.($ simulation time $t = 2 \cdot 10^6$. System size 200 x 300.

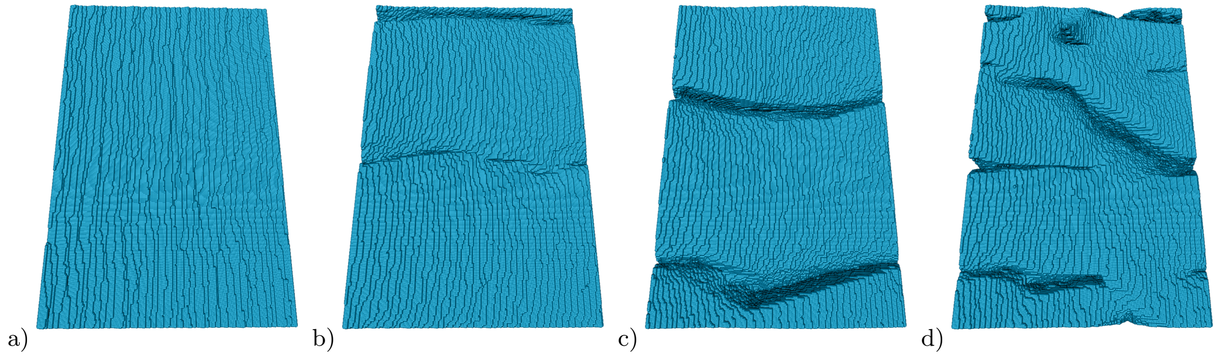


FIG. 3. Meanders obtained for $c_0 = 0.005$, $l_0 = 5$, $n_{DS} = 5$, $\beta E_V = 0.0$ and a) $\beta E_{ES} = 2.0$, b) $\beta E_{ES} = 4.0$, c) $\beta E_{ES} = 6.0$, d) $\beta E_{ES} = 8.0$, simulation time $t = 2 \cdot 10^6$. System size 200 x 300.

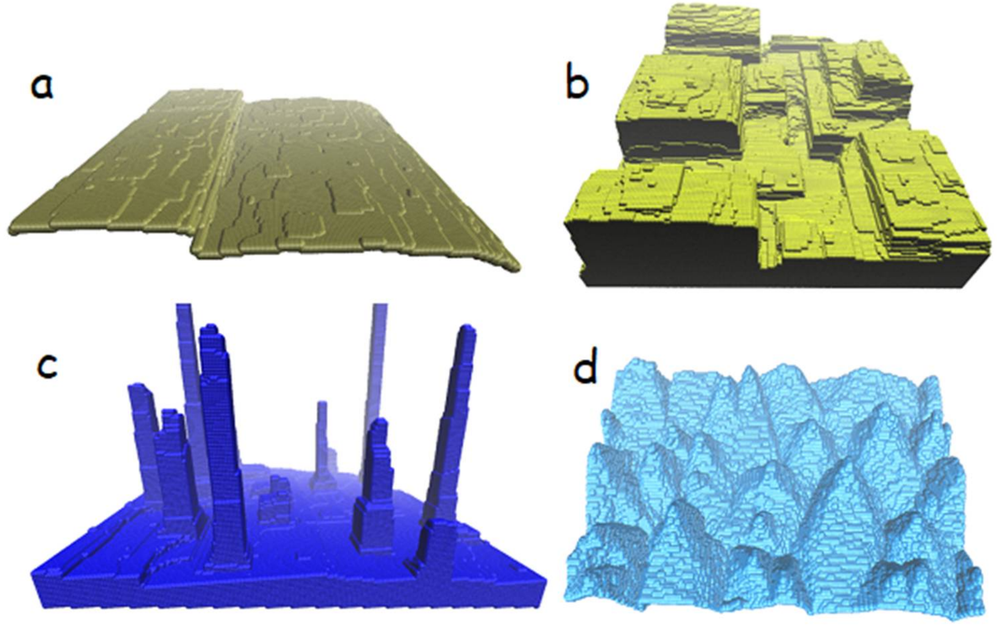
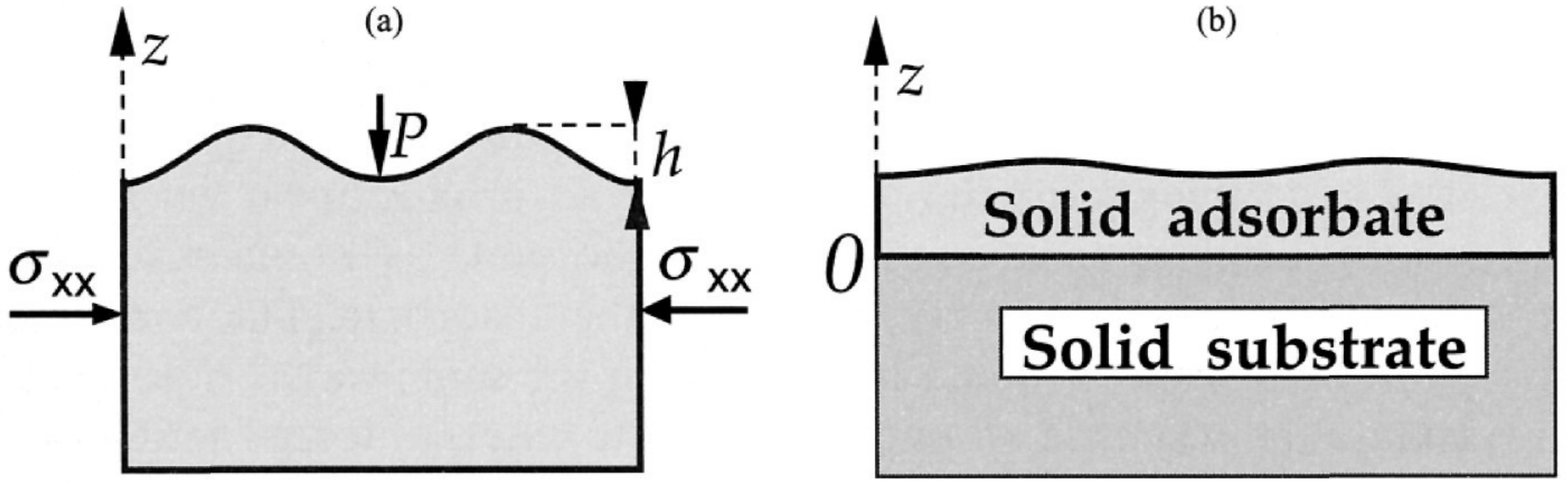


Figure 5. (**a**) Antibunches, c_0 = 0.02, P_{iES} = 0.2, P_{dES} = 0.6, p_w = 1.66, l_0 = 10, n_{DS} = 10, time steps 3×10^6. (**b**) Nanopillars, c_0 = 0.02, P_{iES} = 0.2, P_{dES} = 0.4, p_w = 2.5, l_0 = 10, n_{DS} = 10, time steps 2×10^5. (**c**) Nanowires, c_0 = 0.02, P_{iES} = 0.1, P_{dES} = 0.2, p_w = 5, l_0 = 10, n_{DS} = 10, time steps 3×10^5. (**d**) Pyramids, c_0 = 0.02, P_{iES} = 1.0, P_{dES} = 0.0, p_w = 1.0, l_0 = 5, n_{DS} = 10, time steps 4×10^5.

Elastic instabilities: Grinfeld



Misfit-induced strain: $\epsilon_{xx} = \epsilon_{yy} = \epsilon_0 = \delta a/a$

$$p_0 = -\frac{E}{1-\zeta}\epsilon_0 = \frac{E}{1-\zeta}\frac{\delta a}{a}$$

Contributions to the free energy of the undulating film $\delta Z(x,y) = Z(x,y) - \bar{Z}$

$$\delta Z(x) = h\cos(qx)$$

1) $\mathrm{d}\mathscr{F}_{\mathrm{cap}}/\mathrm{d}\mathscr{A} = \sigma h^2 q^2/2$

Capillarity (surface free energy)

$$\text{curvature} \sim \partial^2 z/\partial x^2 \rightarrow z_q q^2 \sim h q^2$$

$$dV = h d\mathscr{A}$$

2) $\mathrm{d}\mathscr{F}_{\mathrm{relax}}/\mathrm{d}\mathscr{A} \approx -h p_0 \epsilon$

Relaxation, proportional to elastic energy and to volume change

$$dV = h d\mathscr{A}$$

3) $\mathrm{d}\mathscr{F}_{\mathrm{el}}/\mathrm{d}\mathscr{A} \approx C\epsilon^2/(2q)$

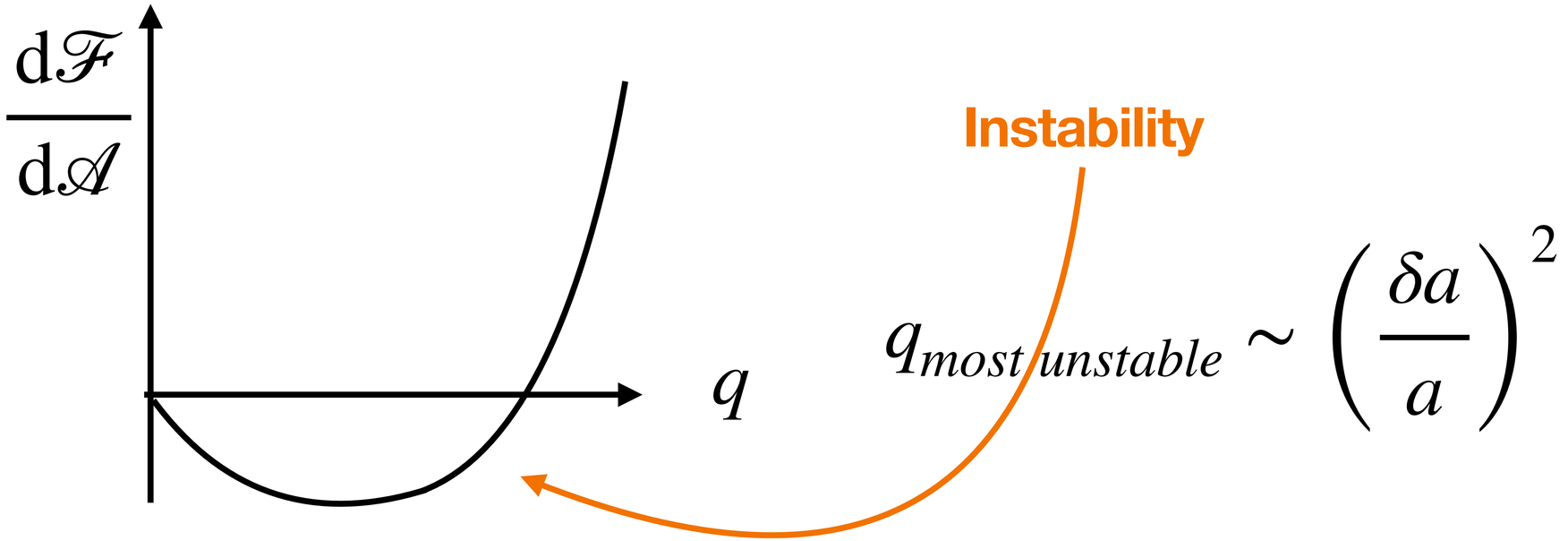
Elastic energy cost proportional to penetration length of the strain $\lambda \approx 1/q$

Minimizing 2 and 3 with respect to ϵ yields:

$$\epsilon \approx hqp_0/C$$

The total free energy in the undulating film is the sum of 1), 2) and 3):

$$\mathrm{d}\mathscr{F}/\mathrm{d}\mathscr{A} = \sigma h^2 q^2/2 - h^2 p_0^2\, q/(2C)$$



Questions?