Continuum theories of ion-induced pattern formation: A critical introduction

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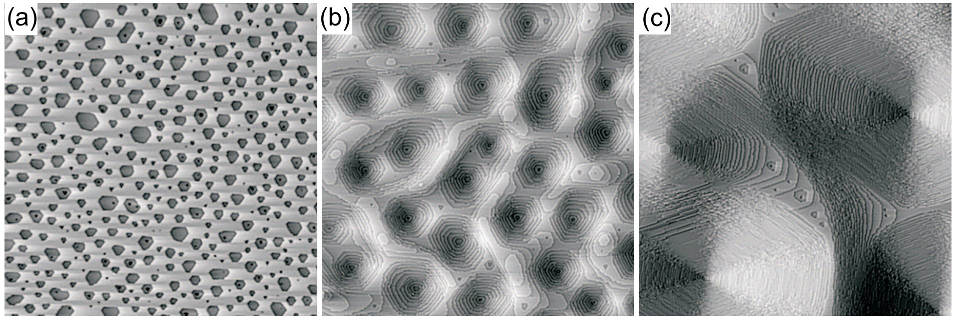


Courtesy of Th. Michely

- Instability mechanisms: Bradley-Harper, Villain and all that
- Wavelength selection in the linear regime
- Scenarios of nonlinear evolution: Coarsening, chaos and order
- Recent developments and open issues

Review: In *Collective Dynamics of Nonlinear and Disordered Systems*, ed. by G. Radons, W. Just & P. Häussler (Springer, Berlin 2005), pp. 5–37.

The continuum approach to surface morphology evolution

- Description of the surface by a height function $z = h(\vec{r}, t), \quad \vec{r} = (x, y)$.

- Surface evolution equation:

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

  with the required symmetries [$h \to h + c$, rotation in the plane, $h \to -h$,....]

- **Pattern formation:** Instability of the flat solution

  $h_0(\vec{r}, t) = \vec{m} \cdot \vec{r} + Vt$ $\quad$ $\vec{m}$: mean inclination $\quad$ V : growth/erosion speed

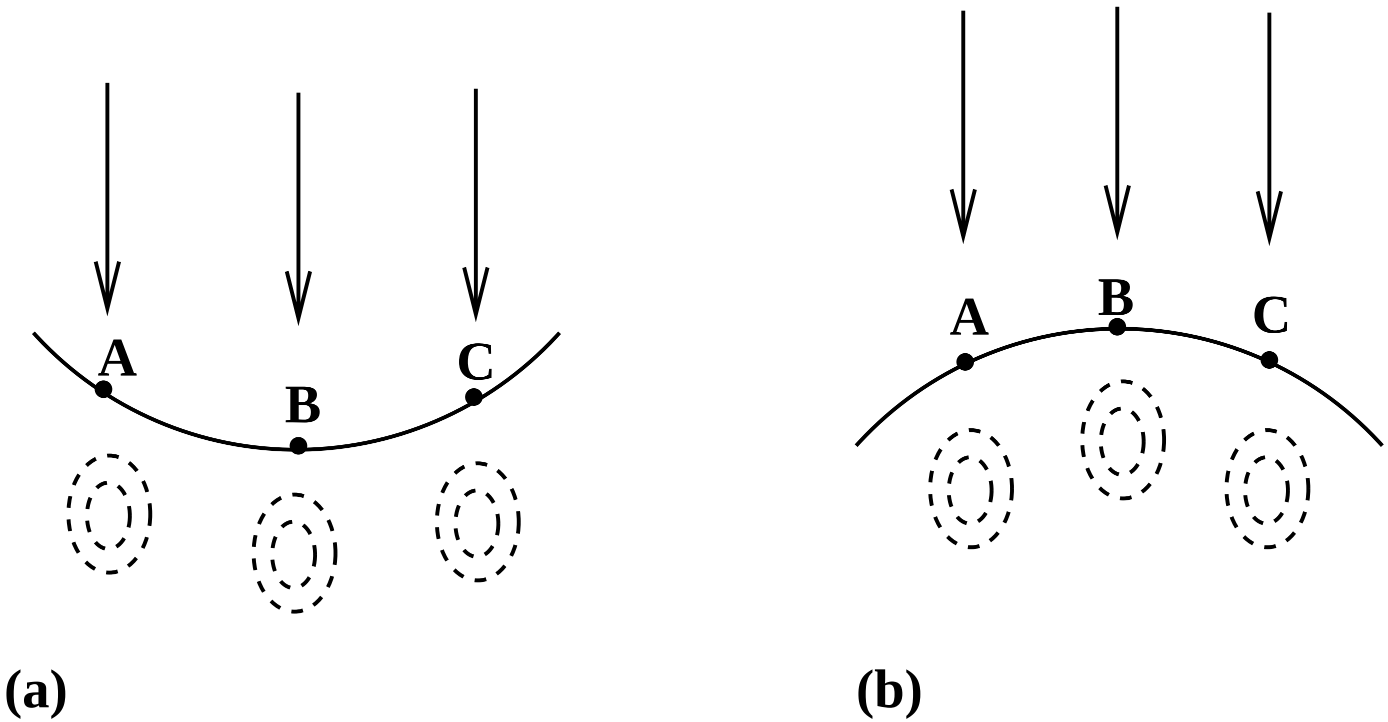
- **Advantages:** Compact analytic or numerical treatment of global aspects of the morphology; comparison between different problems (growth vs. erosion, solid vs. sand surfaces....)

- **Challenges:** **(i)** Relation to microscopic processes
  (ii) Analysis of nonlinear evolution.

The Bradley-Harper instability

R.M. Bradley, J.M.E. Harper, J. Vac. Sci. Technol. A 6, 2390 (1988)

Larger energy contribution from ions incident near minima implies **curvature dependence** of the sputtering yield Y:



minimum

maximum

- Ions incident along the x-axis at angle θ from surface normal

- Evolution equation to leading order:

$$\frac{\partial h}{\partial t} = -Y_0(\theta) - \frac{dY_0}{d\theta}\frac{\partial h}{\partial x} + v_{\parallel}(\theta)\frac{\partial^2 h}{\partial x^2} + v_{\perp}(\theta)\frac{\partial^2 h}{\partial y^2}$$

$Y_0(\theta)$: sputtering yield from flat surface

- Stability requires $v_{\parallel}, v_{\perp} > 0$

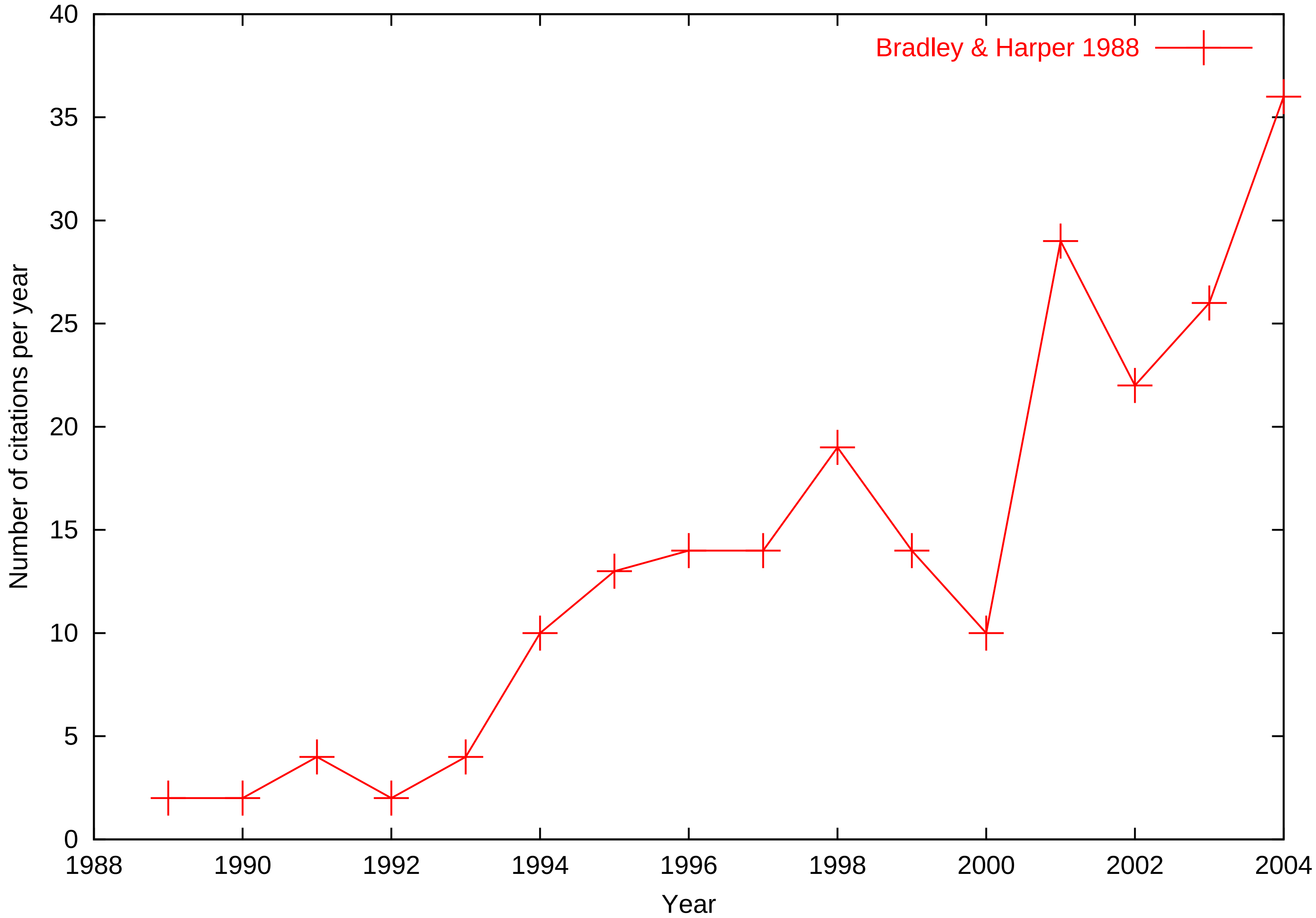
- Normal incidence: $v_{\parallel} = v_{\perp} < 0 \quad \Rightarrow$ **pits**

- Near grazing incidence: $v_{\parallel} > 0$, $v_{\perp} < 0 \quad \Rightarrow$ ripples **parallel** to the beam

- $0 < \theta < \theta_c$: $v_{\parallel} < v_{\perp} < 0 \quad \Rightarrow$ ripples **perpendicular** to the beam

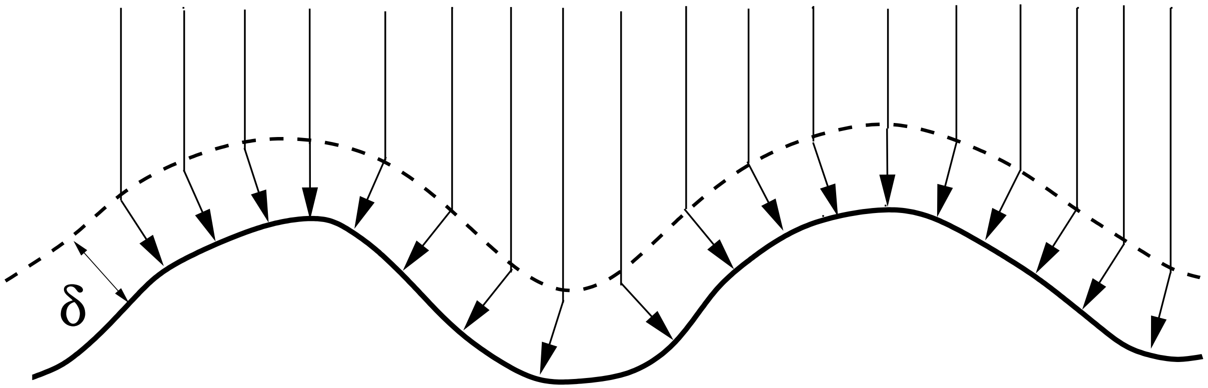
- Ripple rotation well established in experiments on amorphous surfaces and molecular dynamics simulations

Citation statistics



An analogy in growth: Steering

Deflection of incident atoms towards the surface normal due to attractive substrate forces implies enhanced growth flux at maxima:



$$\Rightarrow \quad \frac{\partial h}{\partial t} = F(1+\delta\kappa) \approx F - F\delta\nabla^2 h$$

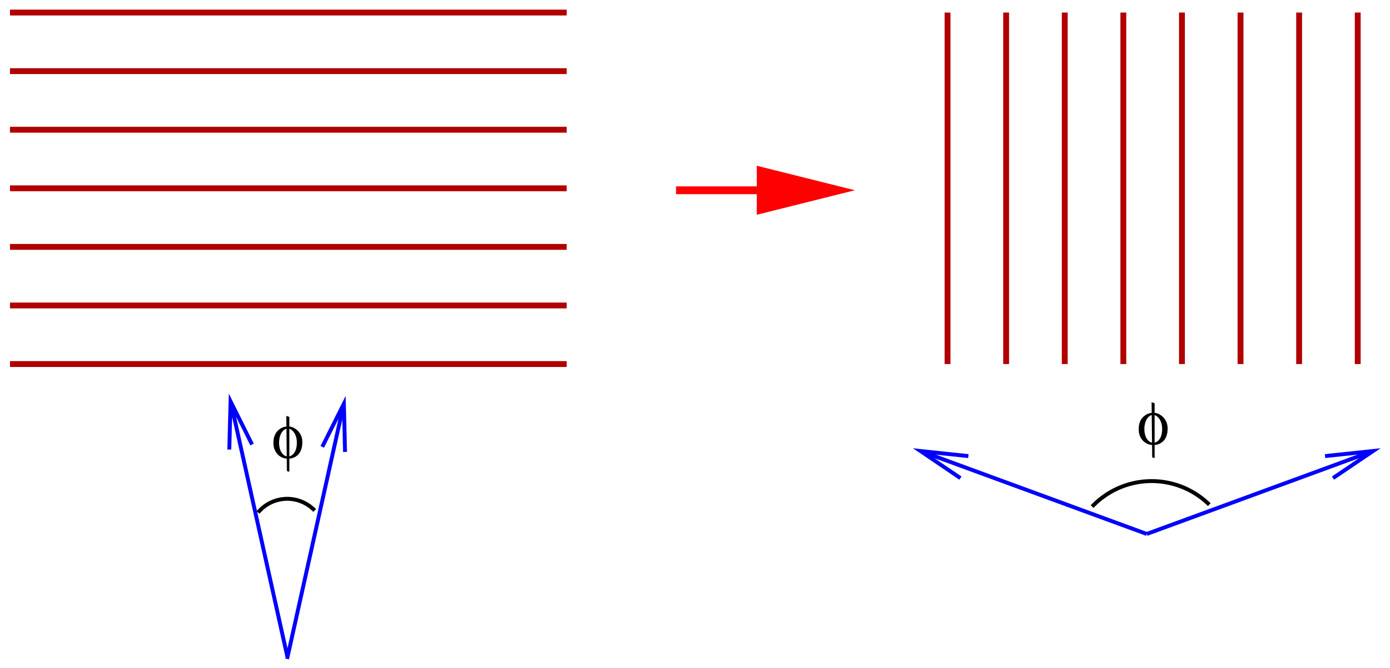
F: deposition flux κ: surface curvature δ: deflection length

H. Park, A. Provata, S. Redner, J. Phys. A 24, L1391 (1991)
A. Mazor, D.J. Srolovitz, P.S. Hagan, B.G. Bukiet, Phys. Rev. Lett. 60, 424 (1988)

Rotation of aeolian sand ripples

D.M. Rubin, R.E. Hunter, Science 237, 276 (1987)

- Sand ripples formed under bidirectional wind switch from tranverse to longitudinal as a function of the divergence angle ϕ

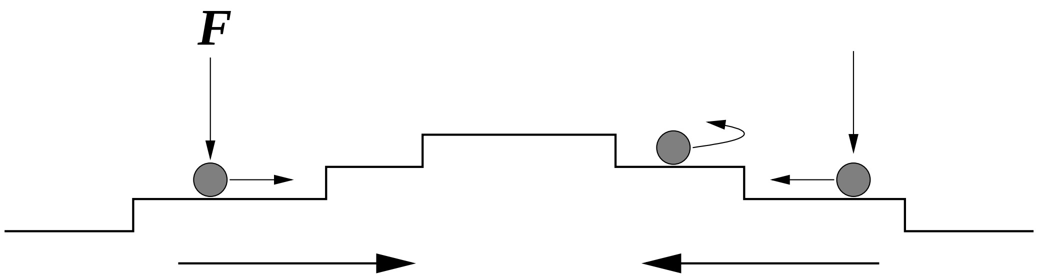


- Critical angle $\phi_c = \pi/2$ is determined by maximization of sand transport.

The Villain instability in epitaxial growth

J. Villain, J. Phys. I France 1, 19 (1991)

- Preferential attachment of atoms to ascending steps [Ehrlich-Schwoebel effect] induces an uphill mass current $\vec{j}(\nabla h)$:



- Continuum evolution equation:

$$\frac{\partial h}{\partial t} + \nabla \cdot \vec{j} = F, \quad \vec{j} \approx f \nabla h \ \Rightarrow \ \frac{\partial h}{\partial t} = -f \nabla^2 h + F$$

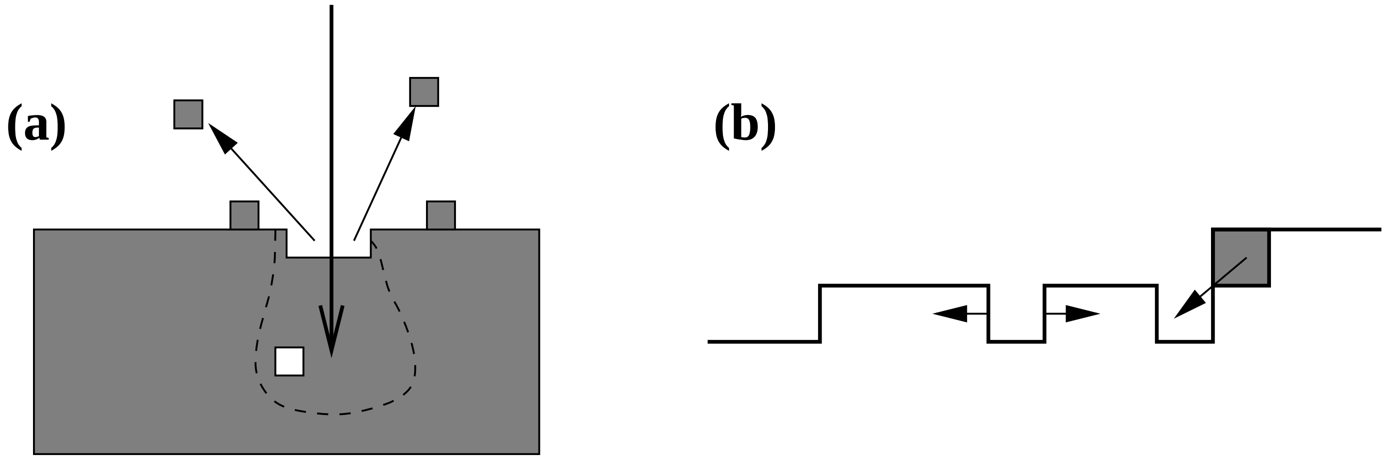
instability for $f > 0$

- Ripple instability on vicinal surfaces [JK, M. Schimschak, J. Phys. I 5, 1065 (1995)]

The Villain instability in erosion: Negative growth

Costantini et al., PRL 86, 838 (2001); Kalff et al., Surf. Sci. 486, 103 (2001)

- Net outcome of ion impact: Creation of adatoms and advacancies in correlated positions



- Recombination of vacancy at step edge requires detachment of step atom $\Rightarrow$ generic strong Ehrlich-Schwoebel effect for vacancies

- Complication compared to growth: **Two** mobile species on the surface

- Coexistence with Bradley-Harper type effects at larger scales?

Wavelength selection in the linear regime

- Linear evolution equation is mathematically **ill-posed**:

$$\frac{\partial h}{\partial t} = \nu \nabla^2 h + F \;\Rightarrow\; h = Ft + h_0 e^{i\vec{q}\cdot\vec{r} + \omega(\vec{q})t} \text{ with } \omega(\vec{q}) = -\nu|\vec{q}|^2$$

  Unbounded growth of short wavelength modes $(|\vec{q}| \to \infty)$ when $\nu < 0$.

- Regularisation by surface diffusion: W.W. Mullins, J. Appl. Phys. 30, 77 (1959)

$$\frac{\partial h}{\partial t} = \nu \nabla^2 h - K(\nabla^2)^2 h + F \;\Rightarrow\; \omega(\vec{q}) = -\nu|\vec{q}|^2 - K|\vec{q}|^4$$

  with **maximally amplified wavelength** $\Lambda^* = 2\pi\sqrt{2K/|\nu|}$

- Smoothening by surface diffusion requires **thermal creation of adatoms**

- Mullins term is inappropriate for surfaces with thermally stable steps

Bradley-Harper:

- Importance of ion-induced mobility at low temperature

- Derivation of smoothening term $-K(\nabla^2)^2 h$ as higher order correction to Bradley-Harper theory Makeev et al., Nucl. Instr. Meth. B 197, 185 (2002)

$\Rightarrow$ initial wavelength determined by the ion penetration depth

Villain:

- Initial wavelength determined by Λ^* only if it is larger than the submonolayer island spacing **(weak Ehrlich-Schwoebel effect)**

- For strong ES effect wavelength is set by submonolayer processes

- Effective smoothening term $-K(\nabla^2)^2 h$ generated by island nucleation ? P. Politi, J. Villain, Phys. Rev. B 54, 5114 (1996)

Nonlinear evolution: Conserved and nonconserved equations

- **Villain:** Nonlinear contributions to the slope-dependent current

$$\frac{\partial h}{\partial t} = -\nabla \cdot [\vec{j} + K\nabla(\nabla^2 h)] + F = -\nabla \cdot f(|\nabla h|^2)\nabla h - K(\nabla^2)^2 h + F$$

slope selection: $f(m^2) = f(0)[1-(m/m_0)^2]$ Siegert & Plischke, PRL 73, 1517 (1994)
no slope selection: $f(m^2) = f(0)/[1+(m/m_0)^2]$ Johnson et al., PRL 72, 116 (1994)
+ crystal anisotropy, up-down symmetry breaking terms, ...

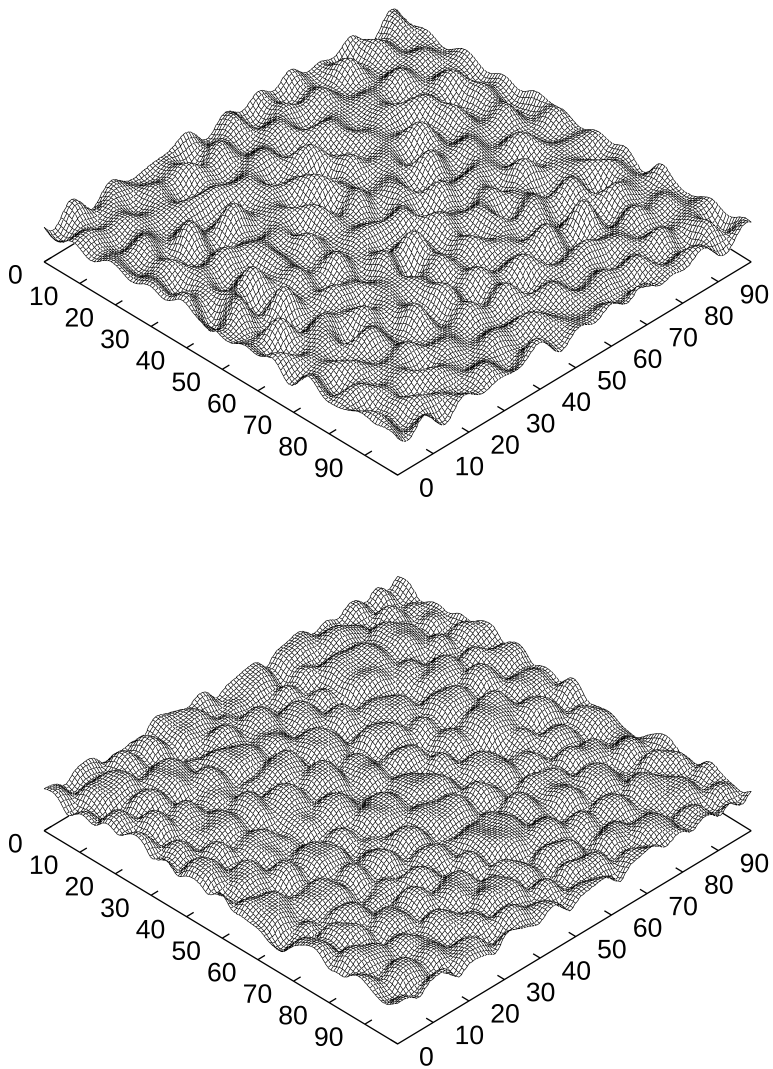
- **Bradley-Harper:** Nonlinear contributions to the sputtering yield
R. Cuerno, A.-L. Barabási, PRL 74, 4746 (1995)

$$\frac{\partial h}{\partial t} = \nu_{\parallel}\frac{\partial^2 h}{\partial x^2} + \nu_{\perp}\frac{\partial^2 h}{\partial y^2} + \frac{\lambda_{\parallel}}{2}\left(\frac{\partial h}{\partial x}\right)^2 + \frac{\lambda_{\perp}}{2}\left(\frac{\partial h}{\partial y}\right)^2 - K(\nabla^2)^2 h$$

anisotropic Kuramoto-Sivashinsky equation

- Nonlinearities suppress exponential instability of the linear theory, except in the presence of cancellation modes [M. Rost, JK, PRL 75, 389, 1995]

Nonlinear evolution of the isotropic KS-equation: Chaotic bubbling



Courtesy of M. Rost

From chaos to noise

- Description of KS dynamics by an effective **stochastic** interface equation on scales large compared to Λ^* V. Yakhot, PRA 24, 642 (1981)

- Chaotic fluctuations generate **(i)** effective noise $\eta(\vec{r},t)$
  (ii) positive surface tension $\bar{\nu} > 0$

$$\Rightarrow \frac{\partial h}{\partial t} = \bar{\nu}\nabla^2 h + \frac{\lambda}{2}(\nabla h)^2 + \eta$$

Kardar-Parisi-Zhang equation (KPZ)

- Asymptotic morphology is a self-affine rough surface with scaling relations

$$\langle |h(\vec{r},t) - h(\vec{r}',t)|^2 \rangle \sim |\vec{r} - \vec{r}'|^{2\alpha}, \quad \langle |h(\vec{r},t) - h(\vec{r},t')|^2 \rangle \sim |t - t'|^{2\beta}$$

  with $\alpha \approx 0.38$, $\beta \approx 0.24$.

- Large value of $\bar{\nu}$ implies long intermediate scaling regime
  Sneppen et al., PRA 46, R7351 (1992); Drotar et al., PRE 59, 177 (1999)

Coarsening behavior of conserved evolution equations

- Coarsening laws: Pattern wavelength $\Lambda \sim t^{1/z}$
  Surface width $W = \sqrt{\langle (h - \bar{h})^2 \rangle} \sim t^{\beta}$

- Exact equation for the surface width: M. Rost, JK, PRE 55, 3952 (1997)

$$\frac{1}{2}\frac{dW^2}{dt} = \langle \vec{j} \cdot \nabla h \rangle - K \langle (\nabla^2 h)^2 \rangle$$

$\Rightarrow$ determination of scaling exponents by estimating the two terms

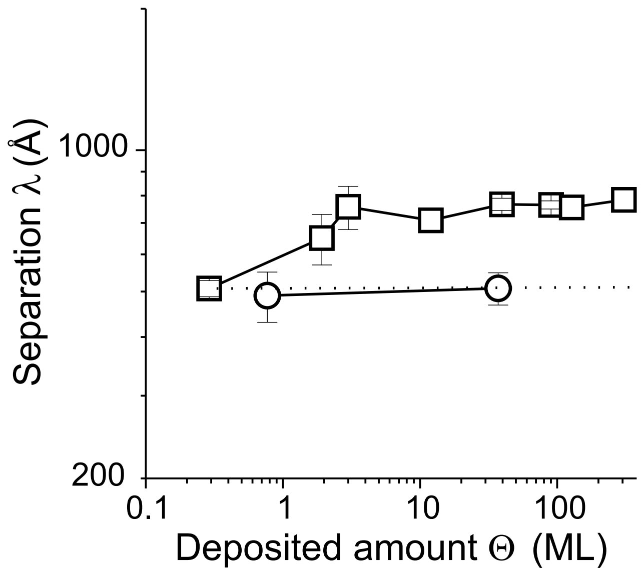
- In-plane isotropy without slope selection: $\beta = 1/2, 1/z = 1/4$
  L. Golubovic, PRL 78, 90 (1997)

- In-plane isotropy with slope selection: $\beta = 1/z = 1/3$
  D. Moldovan, L. Golubovic, PRE 61, 6190 (2000); R.V. Kohn, X. Yan, Comm. Pure Appl. Math. 56, 1549 (2003)

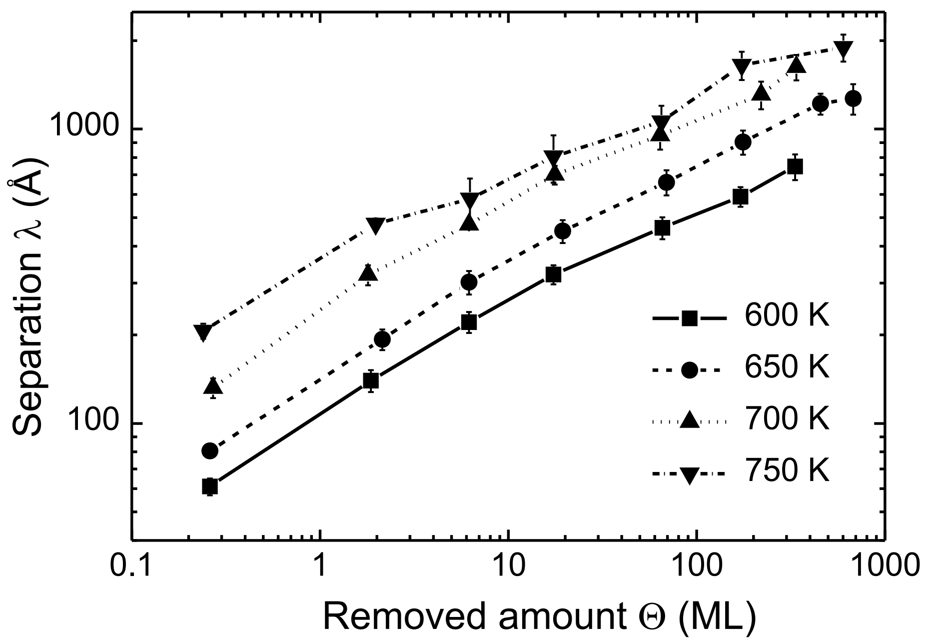
- In-plane anisotropy of low symmetry (fourfold) destroys simple scaling
  M. Siegert, PRL 81, 5481 (1998)

Coarsening on Pt(111)

Growth at 440 K



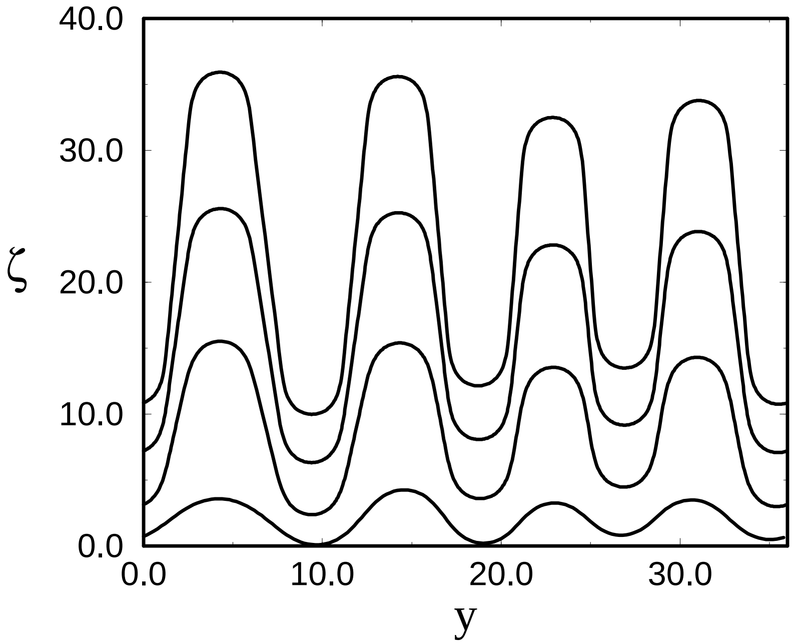
Erosion



Courtesy of Th. Michely

Steepening without coarsening

- One-dimensional evolution equations for surface steps often show unbounded amplitude growth without coarsening $(1/z = 0, \ \beta > 0)$



J. Kallunki, JK, PRE 62, 6229 (2000)

- Steepening with/without coarsening is the **only available scenario** for conserved, one-dimensional evolution equations

  P. Politi, C. Misbah, PRL 92, 090601 (2004)

Breaking the symmetry between hills and valleys

- The conserved KPZ (CKPZ) term [J. Villain, 1991]

$$\frac{\partial h}{\partial t} = -\nabla \cdot [\vec{j} + \frac{\lambda_c}{2}(\nabla h)^2 - K\nabla(\nabla^2 h)] + F$$

reflects slope dependence of the concentration of mobile species

- Influence on coarsening behavior appears to be minor (?)

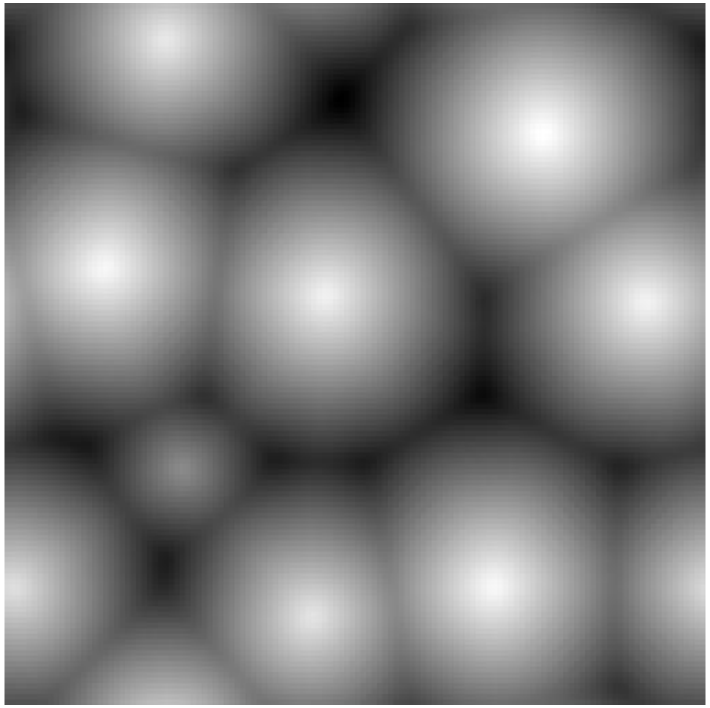
  Stroscio et al., PRL 75, 4246 (1995); P. Politi, PRE 58, 281 (1998)

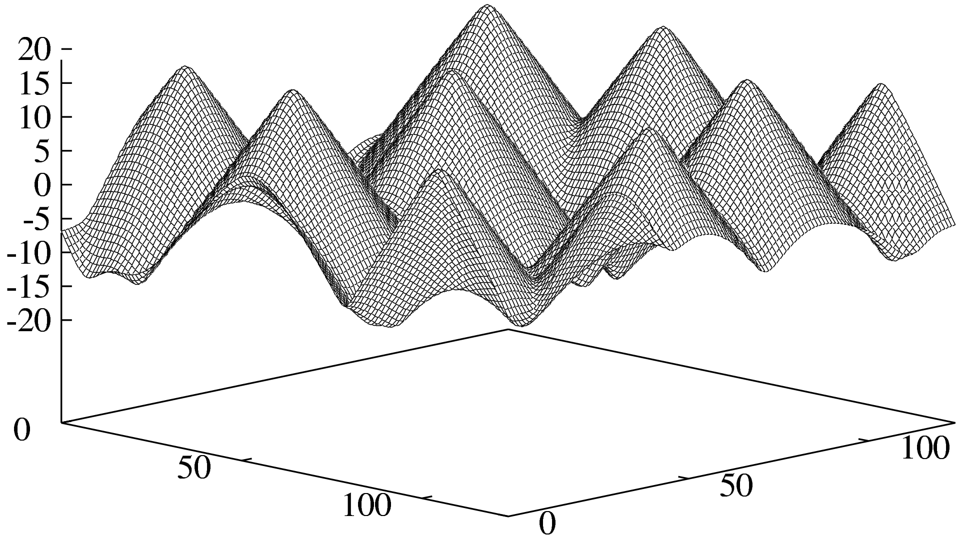
- Nonconserved terms arisising from slope-dependent flux or desorption rate

$$\frac{\partial h}{\partial t} = -\nabla \cdot [\vec{j} - K\nabla(\nabla^2 h)] + F(\nabla h)$$

speed up coarsening: $t^{1/3} \rightarrow t^{1/2}$ in the presence of slope selection

- Growth with desorption: $F(\nabla h) = F(0) - A/[1 + (\nabla h)^2]$





P. Šmilauer, M. Rost, JK, PRE 59, R6263 (1999)

From coarsening to chaos

- How does the transition occur from conserved (coarsening) to nonconserved (chaotic) dynamics ?

- Convective Cahn-Hilliard equation in one dimension:
  Golovin et al., PRL 86, 1550 (2001); Watson et al., Physica D 178, 127 (2003)

$$\frac{\partial h}{\partial t} = -\frac{\partial}{\partial x}\left[\frac{\partial h}{\partial x} - \left(\frac{\partial h}{\partial x}\right)^3\right] - \frac{\partial^4 h}{\partial x^4} - \rho\left(\frac{\partial h}{\partial x}\right)^2$$

  $\rho = 0$: slow coarsening $\Lambda \sim \ln t$
  $\rho \to \infty$: Kuramoto-Sivashinsky equation

- Fast coarsening $\Lambda \sim \sqrt{\rho t}$ for $\rho \ll 1$ on time scales $1/\rho \ll t \ll 1/\rho^3$

- Onset of spatiotemporal chaos at $\rho_c \approx 3.5$

- Stable or propagating fixed wavelength, fixed amplitude solutions for intermediate ρ

Recent developments

- Coupled equations for height and concentration of mobile atoms

  T. Aste, U. Valbusa, Physica A 332, 548 (2004)

- Mechanisms for stable periodic patterns:

  (i) Damped Kuramoto-Sivashinsky equation

$$\frac{\partial h}{\partial t} = -\nabla^2 h - (\nabla^2)^2 h + \frac{\lambda}{2}(\nabla h)^2 - \alpha h$$

  breaks $h \to h + c$ symmetry   Facsko et al., PRB 69, 153412 (2004)

  (ii) KS-equation with CKPZ term

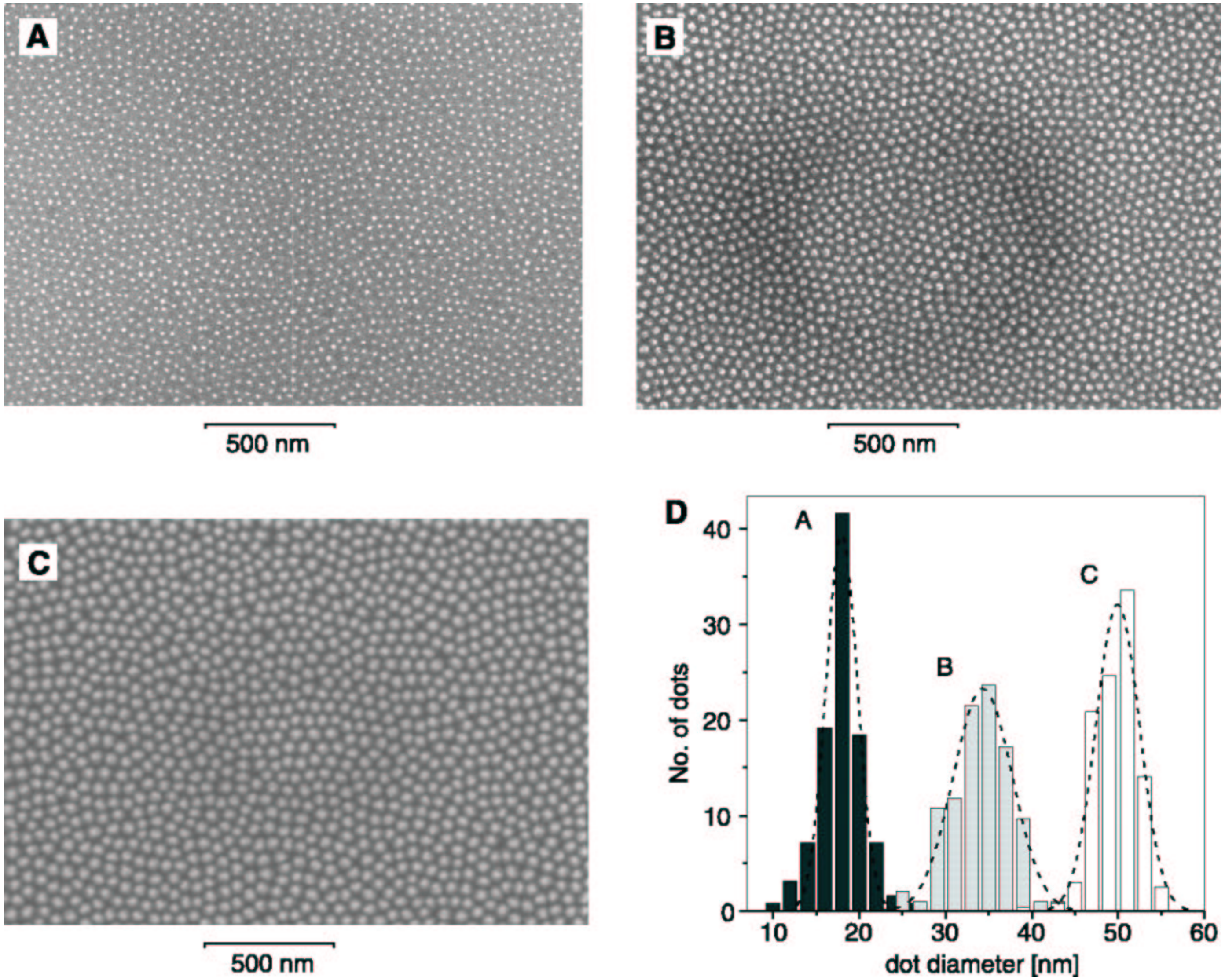
$$\frac{\partial h}{\partial t} = -\nabla^2 h - (\nabla^2)^2 h + \frac{\lambda}{2}(\nabla h)^2 - \frac{\lambda_c}{2}\nabla^2(\nabla h)^2$$

  shows transition from coarsening to chaos

  Castro et al., PRL 94, 016102 (2005)

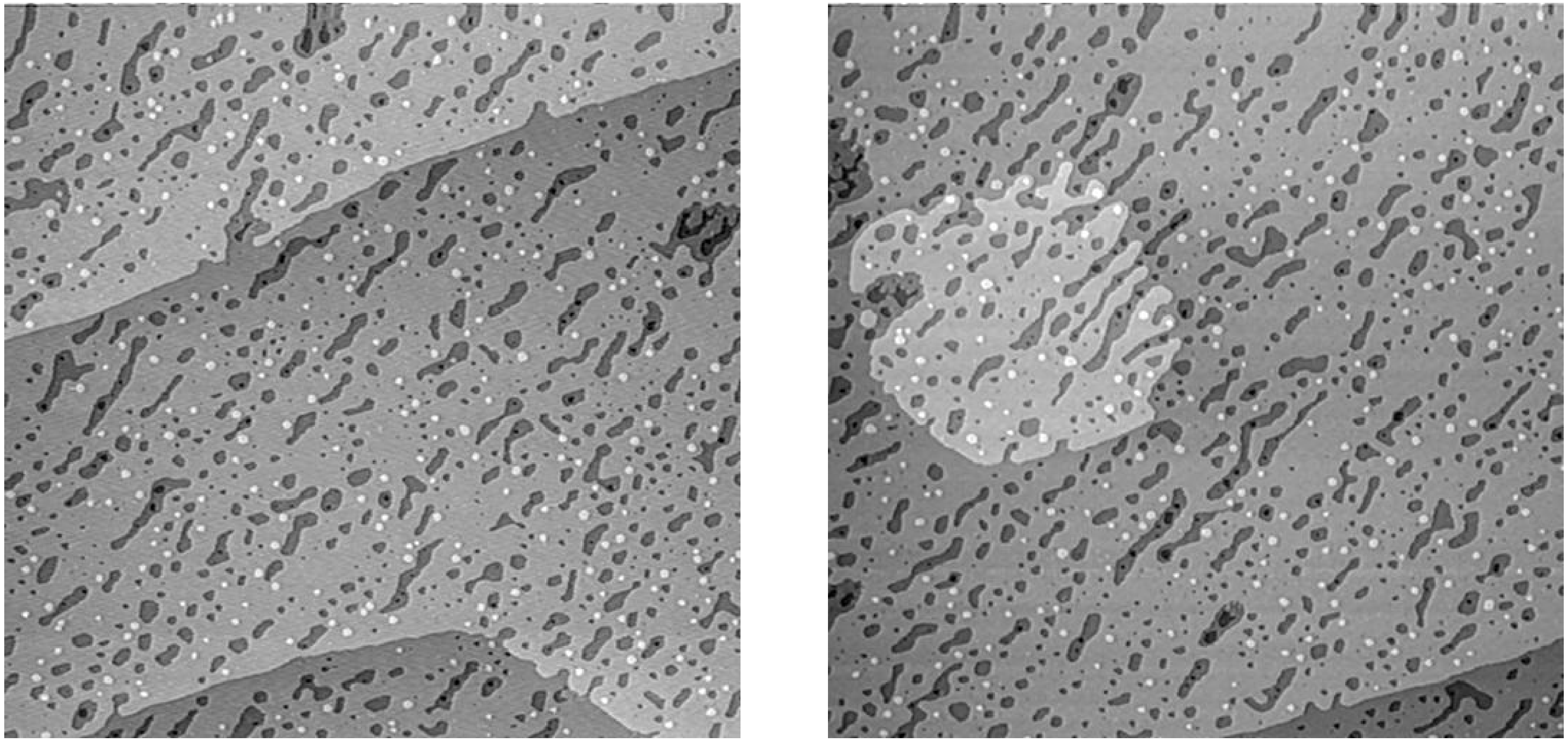
- Ion-induced step propagation   Hansen et al., PRL 92, 246106 (2004)

Ordered nanodots on GaSb(100)



S. Fascko et al., Science **285**, 1551 (1999)

Grazing incidence erosion of Pt(111)



H. Hansen et al., PRL **92**, 246106 (2004)

Outlook

- Continuum height models of surface evolution are fundamentally limited by their neglect of the **discrete atomic structure** in the h-direction.
- At least in situations without nucleation or coalescence, an attractive alternative is provided by **step dynamics**, which is continuous in the plane ($\vec{r}$) but discrete in height (h)
- Simplest assumption: Ion erosion shifts illuminated steps at constant speed
- Progress has been made in deriving continuum height equations from step dynamics in relaxation [D. Margetis, submitted] and step flow [JK et al., PRB 71, 045412 (2005)]
- Treatment of nucleation of adatom- or vacancy clusters in a continuum framework remains a challenge