Non-equilibrium interface dynamics in crystal growth

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Contents

Modeling Crystal surfaces at different scales

- Atomistic
  - Electrons
    Ab Initio, quantum effects
    Density Functional Theory
    (10^3 at., 10^2 CPU, 10^{-12}s/day)
  - Atoms
    Newton equations
    Molecular Dynamics
    (10^5 at., 10 CPU, 10^{-9}s/day)
  - Lattice
    Effective moves: translation, rotation, etc.
    Kinetic Monte Carlo
    (10^6 surface sites, 1 CPU, 10^2s/day)
- ...Intermediate
  - Lattice Boltzmann
    hydrodynamics
  - Phase field Crystal
    continuum but atomic positions
- Continuum
  Diffusion, hydrodynamics, elasticity, etc.
  - Diffuse interface
    Phase field
  - sharp interface
    Continuum macroscopic
  - Stochastic Differential Equations
    Langevin equations

Direct derivation Vs. Asymptotic analysis

Direct derivation of models
(phenomenological models)

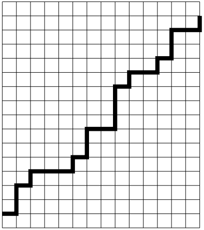
- Symmetries
- Conservation Laws
- Linear Irreversible Thermodynamics (Onsager)
- ... etc.

Asymptotic methods relating different scales

- Transition state theory
  Atomistic (DFT or MD)
  $\rightarrow$ energy landscape
  $\rightarrow$ rates for KMC on continuum
- Sharp interface limit
  Diffuse interface
  $\rightarrow$ Sharp interface
- Multiple scale expansions
  Instability
  $\rightarrow$ effective nonlinear (amplitude) equations
  $\rightarrow$ Morphology
- Renormalization group
  Integrate over small scales
  $\rightarrow$ large scale behavior
  (large distances, large times)
- Homogenization
  Heterogeneous medium
  $\rightarrow$ effective medium

Contents

The roughening transition



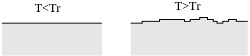
Broken bond energy J
Length N

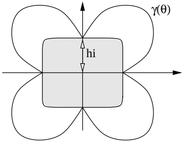
Free energy

$$F = E - TS = NJ - k_B T \ln 2^N = N(J - k_B T \ln 2)$$

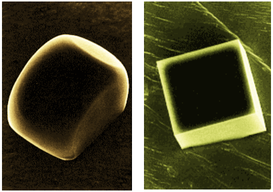
$F \to 0$ as $T \to T_R$
Roughening transition temperature

$$T_R = \frac{J}{k_B \ln 2}$$



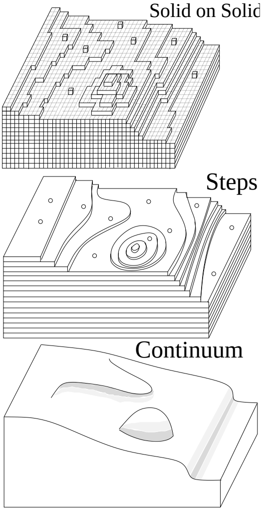


For usual crystals $T_r \sim T_M$



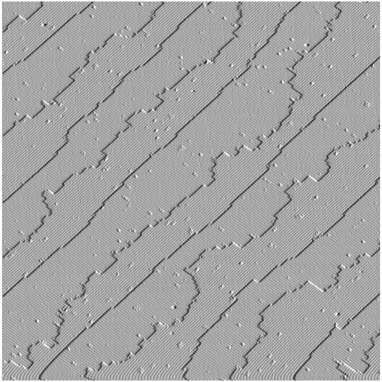
NaCl, Métois et al (620-710^{o}C)

Levels of description



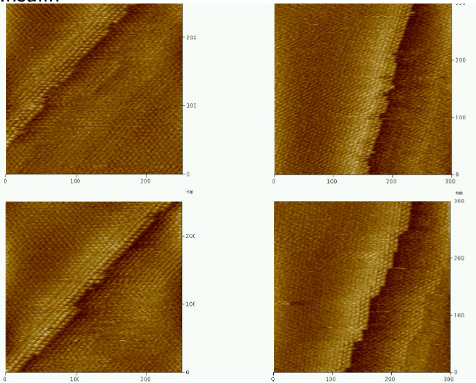
Atomic steps

Si(100)



M. Lagally, Univ. Visconsin

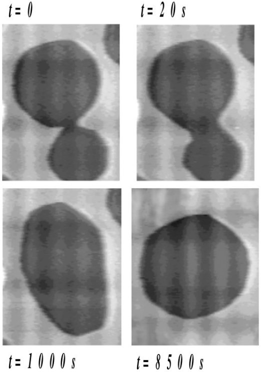
Insulin



P. Vekilov, Houston

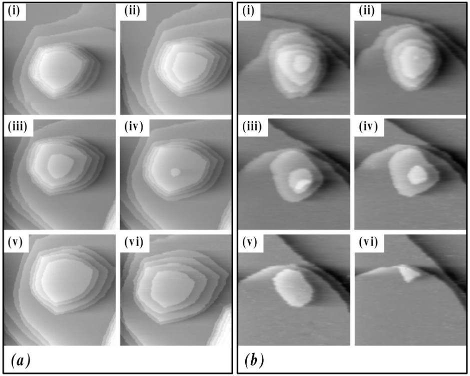
Nano-scale Relaxation

Ag(111), Sintering



M. Giesen, Jülich Germany

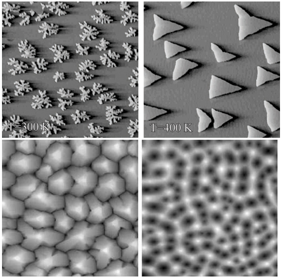
Decay Cu(1,1,1) 10mM HCl at (a) -580mV; and (b) 510 mV



Broekman et al, (1999) J. Electroanal. Chem.

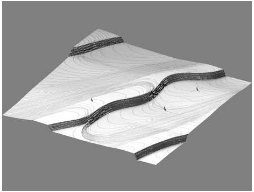
Nano-scale crystal Surface Instabilities out of equilibrium

Growth and Ion Sputtering, Pt(111)



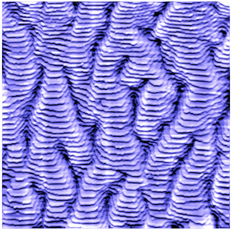
T. Michely, Aachen, Germany

Electromigration Si(111)



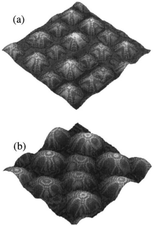
E.D. Williams, Maryland, USA

Growth Cu(1,1,17)



Maroutian, Ernst, Saclay, France

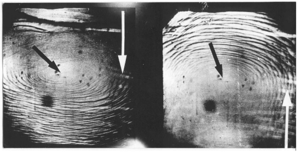
SiGe MBE growth



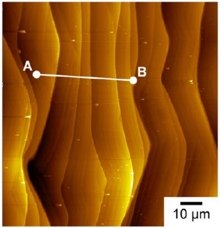
Floro et al 1999

Solution Growth $NH_4H_2PO_4$ (ADP) (5mm)

Chernov 2003



Solution Growth SiC

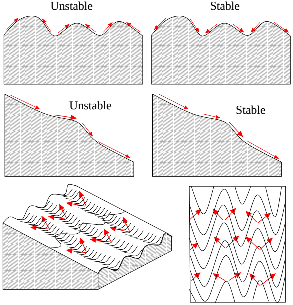


Zu et al Cryst. Growth & Des. 2013

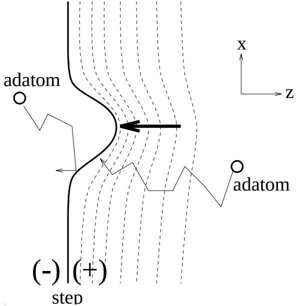
Nano-scale crystal Surface Instabilities out of equilibrium

Kinetic instabilities

Mass fluxes **along** the surface



Mass fluxes **to** the surface



Energy relaxation driven instabilities:

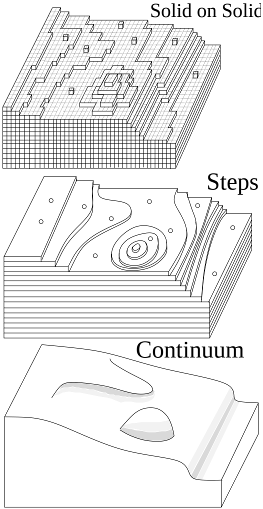
surface energy, elastic energy, electrostatic energy, electronic energy, etc.

Nano-scale crystal Surface Instabilities out of equilibrium

Movie...

Contents

Levels of description



Master equation

Discrete set of configurations, index $n = 1, .., N_{tot}$
Physics: Transition rates $R(n \to m)$
Markovian dynamics (no memory)
Master Equation

$$\partial_t P(n,t) = \sum_{m=1}^{N_{tot}} R(m \to n) P(m,t) - \sum_{m=1}^{N_{tot}} R(n \to m) P(n,t)$$

Ising lattice (0,1), $L \times L = L^2$ sites, $N_{tot} = 2^{L^2}$ configurations
$L = 10 \Rightarrow N_{tot} \sim 10^{30}$ ... too large for direct numerical solution!

Note: number of possible moves from n: $N_{poss}(n) \ll N_{tot}$
$\Rightarrow R(m \to n)$ sparse, i.e. $\Rightarrow R(m \to n) = 0$ for most values of m, n.

Equilibrium and Detailed Balance

Master Equation

$$\partial_t P(n,t) = \sum_{m=1}^{N_{tot}} R(m \to n) P(m,t) - \sum_{m=1}^{N_{tot}} R(n \to m) P(n,t)$$

Equilibrium, steady-state ($\partial_t P_{eq}(n,t) = 0$), Hamiltonian $\mathcal{H}(n)$

$$P_{eq}(n) = \frac{1}{Z} \exp\left[-\frac{\mathcal{H}(n)}{k_B T}\right]$$

$$Z = \sum_{n=1}^{N_{tot}} \exp\left[-\frac{\mathcal{H}(n)}{k_B T}\right]$$

leading to

$$\frac{P_{eq}(n)}{P_{eq}(m)} = \exp\left[-\frac{\mathcal{H}(n) - \mathcal{H}(m)}{k_B T}\right]$$

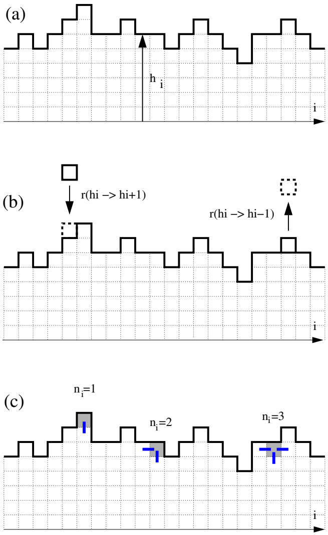
Stonger condition:Detailed Balance

$$R(n \to m) P_{eq}(n) = R(m \to n) P_{eq}(m)$$

or

$$R(n \to m) = R(m \to n) \exp\left[\frac{\mathcal{H}(n) - \mathcal{H}(m)}{k_B T}\right]$$

Example : Attachment-Detachment model

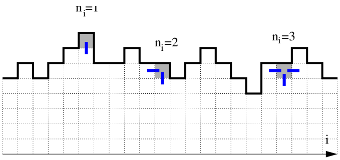


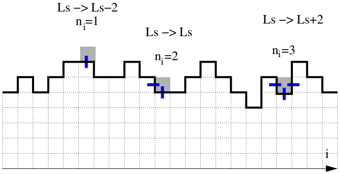
State $n = \{h_i; i = 1.., L\}$
Rates

$$
\begin{aligned}
R(h_i \to h_i + 1) &= F \\
R(h_i \to h_i - 1) &= r_0 \exp[-\frac{n_i J}{k_B T}]
\end{aligned}
$$

n_i number nearest neighbors at site i before detachment
(Breaking all bonds to detach / Transition state Theory)

Example: Attachment-Detachment model





Detailed Balance when $F = F_{eq}$

$$
\begin{aligned}
R(h_i - 1 \to h_i) P_{eq}(h_i - 1) &= R(h_i \to h_i - 1) P_{eq}(h_i) \\
F_{eq} P_{eq}(h_i - 1) &= r_0 \exp[-\frac{n_i J}{k_B T}] P_{eq}(h_i)
\end{aligned}
$$

Bond energy $J \Rightarrow$ Broken Bond energy $J/2$
Hamiltonian $\mathcal{H}$, surface length L_s

$$\mathcal{H} = L_s \frac{J}{2}$$

Energy change

$$
\begin{aligned}
L_s(h_i) - L_s(h_i - 1) = 2(2 - n_i) \\
\mathcal{H}(h_i) - \mathcal{H}(h_i - 1) = (2 - n_i) J
\end{aligned}
$$

Thus

$$\frac{P_{eq}(h_i - 1)}{P_{eq}(h_i)} = \exp\left[\frac{\mathcal{H}(h_i) - \mathcal{H}(h_i - 1)}{k_B T}\right] = \exp\left[\frac{(2 - n_i) J}{k_B T}\right]$$

and

$$F_{eq} = r_0 \exp\left[-\frac{2J}{k_B T}\right]$$

Example: Attachment-Detachment model

Link to the Ising Hamiltonian with field H, $S_i = \pm 1$

$$\mathcal{H}_{Ising} = -\frac{J}{4} \sum_{\langle i,j \rangle} S_i S_j - H \sum_i S_i$$

Define $n_i = (S_i + 1)/2 \Rightarrow J$ is the bond energy

$$\mathcal{H}_{Ising} = -J \sum_{\langle i,j \rangle} n_i n_j - (J - 2H) \sum_i n_i + \mathrm{const}$$

re-writing $\mathcal{H}$

$$\mathcal{H}_{Ising} = \frac{J}{2} \sum_{\langle i,j \rangle} [n_i(1 - n_j) + n_j(1 - n_i)] - 2H \sum_i n_i + \mathrm{const} = \frac{J}{2} L_s - \Delta\mu \sum_i n_i + \mathrm{const}$$

Chemical potential $\Delta\mu = 2H$
Number of broken bonds $L_s = \sum_{\langle i,j \rangle} [n_i(1 - n_j) + n_j(1 - n_i)]$

Example: Attachment-Detachment model

Chemical potential $\Delta\mu = 2H$

Detailed Balance imposed to the Ising system with field H

$$\begin{aligned} F &= R(h_i - 1 \to h_i) = R(h_i \to h_i - 1)\frac{P_{eq}^{Ising}(h_i)}{P_{eq}^{Ising}(h_i - 1)} \\ &= \exp\left[\frac{-n_i J + \mathcal{H}_{Ising}(h_i) - \mathcal{H}_{Ising}(h_i - 1)}{k_B T}\right] \\ &= \exp\left[-\frac{n_i J}{k_B T} + \frac{[L_s(h_i) - L_s(h_i - 1)]J}{2k_B T} + \frac{\Delta\mu}{k_B T}\right] \\ &= \exp\left[-\frac{n_i J}{k_B T} + \frac{(n_i - 2)J}{2k_B T} + \frac{\Delta\mu}{k_B T}\right] \\ &= F_{eq}\exp\left[\frac{\Delta\mu}{k_B T}\right] \end{aligned}$$

$$F_{eq} = r_0 \exp\left[-\frac{2J}{k_B T}\right]$$

Attachement-Detachment model equivalent to Ising in Magnetic field

Equilibrium Monte Carlo: Metropolis algorithm

Algorithm

1. Choose an event $n \to m$ at random
2. implement the event with probability

$$\mathcal{H}(n) \geq \mathcal{H}(m) \quad \Rightarrow \quad P(n \to m) = 1$$
$$\mathcal{H}(n) \leq \mathcal{H}(m) \quad \Rightarrow \quad P(n \to m) = \exp[-\frac{\mathcal{H}(m) - \mathcal{H}(n)}{k_B T}]$$

Obeys Detailed Balance

$$\frac{R(n \to m)}{R(m \to n)} = \exp\left[-\frac{\mathcal{H}(n) - \mathcal{H}(m)}{k_B T}\right]$$

Chain of events conv. to equil. $\Rightarrow$ sample configuration space $n \Rightarrow$ Thermodynamic averages
No time!

Contents

A simple Monte Carlo method with time: random attempts

Using physical rates $R(n \to m)$ from a given model

Algorithm

1. Choose an event $n \to m$ at random
2. Implement the event with probability

$$P(n \to m) = \frac{R(n \to m)}{R_{max}}$$

   where $R_{max} = \max_{n'}(R(n \to n'))$.
3. implement the time by $\Delta t \sim 1/(N_{poss}(n) R_{max})$
   $N_{poss}(n)$ number of possible moves from state n

Problem: when most $R(n \to m) \ll R_{max}$, then most $P(n \to m) \ll 1 \Rightarrow$ most attempts rejected!

Questions

- a rejection-free algorithm?
- time implementation?

Kinetic Monte Carlo algorithm 1: Rejection-free algorithm

implement the FIRST event that occurs

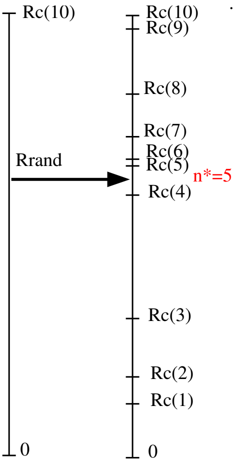
Probability first chosen event $n \to m$ is $\sim R(n \to m)$

Choose event with probability

$$P(n \to m) = \frac{R(n \to m)}{R_{tot}(n)}$$

Rate that one event occurs

$$R_{tot}(n) = \sum_{m=1}^{N_{tot}} R(n \to m)$$



Algorithm

1. Build cumulative rates $R_c(m) = \sum_{p=1}^{m} R(n \to p)$, with $R_c(N_{tot}) = R_{tot}(n)$.
2. Choose random number R_{rand}, uniformly distributed with $0 < R_{rand} \leq R_{tot}(n)$.
3. Choose event n_* such that $R_c(n_* - 1) < R_{rand} \leq R_c(n_*)$.

Kinetic Monte Carlo algorithm 3: comments

Algorithm

1. Build cumulative rates $R_c(m) = \sum_{p=1}^{m} R(n \to p)$, with $R_c(N_{tot}) = R_{tot}(n)$.
2. Choose random number R_{rand}, uniformly distributed with $0 < R_{rand} \leq R_{tot}(n)$.
3. Choose event n_* such that $R_c(n_* - 1) < R_{rand} \leq R_c(n_*)$.

Improved algorithm:

- Number of possible events from one state $N_{poss}(n) \ll$ number of states $N_{tot}(n)$
- Groups of events with same rates

Kinetic Monte Carlo algorithm 2: implementing time

Define τ time with $\tau = 0$ when arriving in state n
Proba $Q(\tau)$ that no event occurs up to time τ, with $Q(0) = 1$

$$Q(\tau + d\tau) = Q(\tau)(1 - R_{tot}(n)d\tau) \Rightarrow \frac{dQ(\tau)}{d\tau} = -R_{tot}(n)Q(\tau) \Rightarrow Q(\tau) = \exp[-R_{tot}(n)\tau]$$

probability density ρ, i.e. $\rho(\tau)d\tau$ first event occuring between τ and $\tau + d\tau$

$$\rho(\tau)d\tau = -dQ(\tau) = R_{tot}(n)\exp[-R_{tot}(n)\tau]d\tau$$

Easy to generate: uniform distribution $\rho(u) = 1$, and $0 < u \leq 1$
Variable change

$$u = \exp[-R_{tot}(n)\tau]$$

same probability density

$$\rho(\tau)d\tau = -\rho(u)du$$

Algorithm

1. Choose random number u, uniformly distributed with $0 < u \leq 1$.
2. Time increment $t \to t + \tau$ with

$$\tau = \frac{-\log(u)}{R_{tot}(n)}$$

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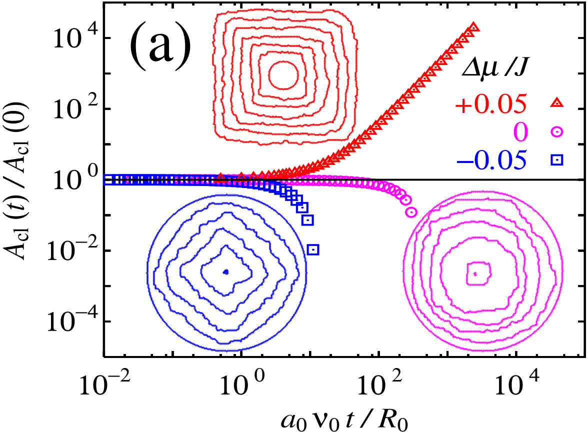
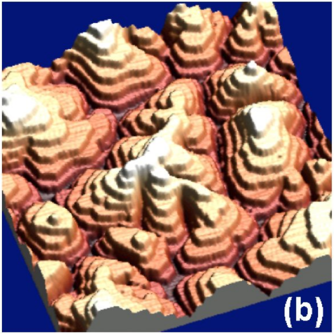
Attachment-detachment model



$F = F_{eq} \mathrm{e}^{-\Delta\mu/k_B T}$

Saito, Pierre-Louis, PRL 2012

KMC results



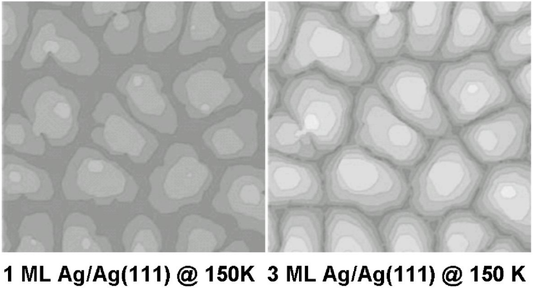
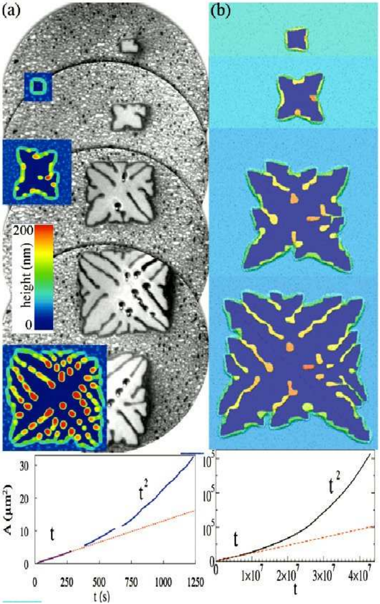


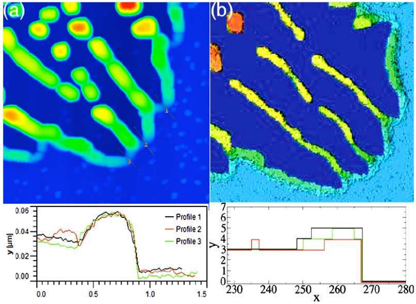
FIGURE 12. Simulation predictions for the morphologies of multilayer Ag/Ag(111) films grown at 150 K with F= 0.004 ML/s. Images sizes are ~160×160 nm^2.

Thiel and Evans 2007

KMC vs SOI solid-state dewetting



$h = 3,\ E_S = 1,\ T = 0.5$



E. Bussman, F. Leroy, F. Cheynis, P. Müller, O. Pierre-Louis NJP 2011

KMC conclusion

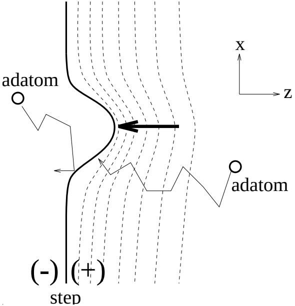
- KMC versatile method to look at crystal shape evolution
- Improved methods to obtain physical rates (DFT, MD)
- Build-in thermal fluctuations

Problem: difficult to parallelize

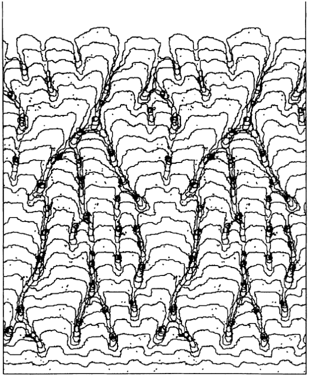
Step meandering Instabilities

Non-equilibrium meandering of a step
Schwoebel effect + terrace diffusion

Bales and Zangwill 1990



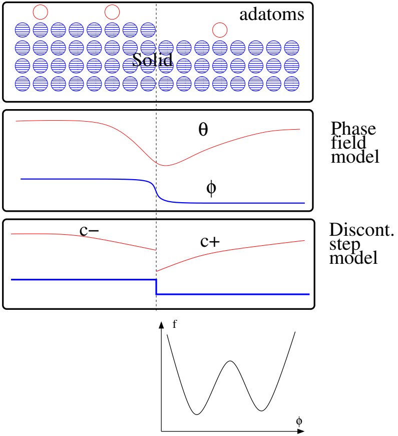
Y. Saito, M. Uwaha 1994



Solving step dynamics
→ morphology and coupling to a diffusion field

Contents

Phase field Model



Concentration c

$$\partial_t c = \nabla[M\nabla c] + F - \frac{c}{\tau} - \partial_t h/\Omega$$

Phase field ϕ

$$\tau_p \partial_t \phi = W^2 \nabla^2 \phi - f'(\phi) + \lambda(c - c_{eq}) g'(\phi)$$

$g_\phi = 0$ at min of f

Equilibrium at $F = F_{eq} = c_{eq}/\tau$
Free energy if $f = -g/[g]_-^+$

$$\mathcal{F} = \int d^2\mathbf{r} \left[\frac{W^2}{2} (\nabla\phi)^2 + f(\phi) + \frac{\lambda}{2} (c - c_{eq})^2 \right]$$

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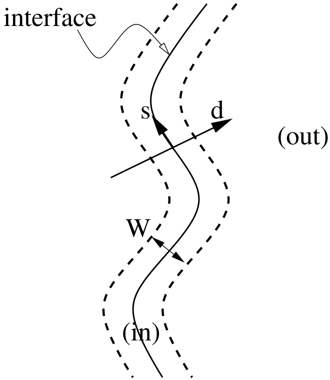
Asymptotic expansion



$\epsilon = W/\ell_c$; $\eta = d/\epsilon$; $\nabla d = \mathbf{n}$

Two expansions: "inner domain" and "outer domain"

$$c^{in}(\eta, s, t) = \sum_{n=0}^{\infty} \epsilon^n c_n^{in}(\eta, s, t)$$

$$c^{out}(x, y, t) = \sum_{n=0}^{\infty} \epsilon^n c_n^{out}(x, y, t)$$

Matching conditions $c^{in}(\eta) = c^{out}(\epsilon\eta)$
with $\eta \to \infty$, $\epsilon \to 0$, $\epsilon\eta \to 0$

$$\lim_{\eta\to\pm\infty} c_0^{in} = \lim_{d\to 0\pm} c_0^{out}$$

$$\lim_{\eta\to\pm\infty} \partial_\eta c_0^{in} = 0$$

$$\lim_{\eta\to\pm\infty} c_1^{in} = \lim_{d\to 0\pm} c_1^{out} + \eta \lim_{d\to 0\pm} \mathbf{n}.\nabla c_0^{out}$$

$$\lim_{\eta\to\pm\infty} \partial_\eta c_1^{in} = \lim_{d\to 0\pm} \mathbf{n}.\nabla c_0^{out}$$

$$\lim_{\eta\to\pm\infty} \partial_{\eta\eta} c_1^{in} = 0$$

Expansion inner domain

Change to inner variables with

$$\nabla d = \mathbf{n}; \qquad \nabla \cdot \mathbf{n} = \Delta d = \kappa$$

Laplacian u

$$\Delta u = \epsilon^{-2}(\partial_{\eta\eta} u + \epsilon\kappa\partial_\eta u + h.o.t.)$$

Co-moving frame at velocity V

$$\partial_t = -V\partial_d$$

Model equations inner region

$$\begin{aligned} o(\epsilon^2) &= \partial_\eta[M\partial_\eta c^{in}] + \epsilon(V + M\kappa)\partial_\eta c^{in} + \epsilon V\partial_\eta h/\Omega \\ o(\epsilon^2) &= \partial_{\eta\eta}\phi^{in} - \partial_\phi f + \lambda(c^{in} - c_{eq})\partial_\phi g + \epsilon(Va + \kappa)\partial_\eta\phi^{in} \end{aligned}$$

with

$$\frac{1}{a} = \frac{W^2}{\tau_\phi}$$

Expansion order by order

$\lambda(c - c_{eq}) \sim \epsilon$
No coupling to leading order
0th order

$$\partial_\eta [M^0 \partial_\eta c_0^{in}] = 0$$

$\rightarrow c_0^{in} = cst$ in step region

$$\partial_{\eta\eta}\phi_0 - f_\phi^0 = 0$$

$\rightarrow$ frozen step
Diffusion equation in the outer region

$$\partial_t c^{out} = D\nabla^2 c^{out} + F - \frac{c^{out}}{\tau}$$

1st order
Mass conservation

$$(\Omega^{-1} + c_- - c_+)V = -D\mathbf{n}.\nabla c_- + D\mathbf{n}.\nabla c_+$$

Expansion order by order

higher order

Sharp interface asymptotics Caginalp 1989

Weak coupling (permeable steps): $\lambda \sim \epsilon$

$$c_+ = c_- = \tilde{c}_{eq}$$

$$\tilde{c}_{eq} = c_{eq}(1 + \Gamma\kappa) + \tilde{\beta}\frac{V}{\Omega}$$

with

$$\begin{aligned} \beta &= \frac{\tau_\phi}{\lambda W}\int d\eta(\partial_\eta\phi_0)^2 \\ \Gamma c_{eq} &= \frac{W}{\lambda}\int d\eta(\partial_\eta\phi_0)^2 \end{aligned}$$

Expansion order by order

higher order

Thin interface asymptotics Karma Rappel 1996, OPL 2003

Fast kinetics: $(c - c_{eq}) \sim \epsilon$

$$\begin{aligned} D\mathbf{n}.\nabla c_+ &= \tilde{\nu}_+(c_+ - \tilde{c}_{eq}) \\ -D\mathbf{n}.\nabla c_- &= \tilde{\nu}_-(c_- - \tilde{c}_{eq}) \end{aligned}$$

with

$$\tilde{c}_{eq} = c_{eq}(1 + \Gamma\kappa) + \tilde{\beta}\frac{V}{\Omega}$$

$$\begin{aligned} \frac{1}{\tilde{\nu}_+} &= W\int d\eta \left(\frac{1}{M^0} - \frac{1}{D}\right)(h^0_- - h^0) \\ \frac{1}{\tilde{\nu}_-} &= W\int d\eta \left(\frac{1}{M^0} - \frac{1}{D}\right)(h^0 - h^0_+) \\ \tilde{\beta} &= \frac{\tau_\phi}{W\lambda}\int d\eta(\partial_\eta\phi_0)^2 - W\int d\eta \frac{(h^0_+ - h^0)(h^0 - h^0_-)}{M^0} \\ c_{eq}\Gamma &= \frac{W}{\lambda}\int d\eta(\partial_\eta\phi_0)^2 \end{aligned}$$

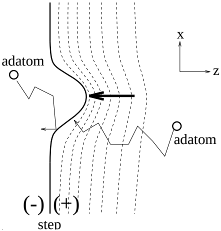
$\tilde{\nu}_\pm \sim \tilde{\beta} \sim \epsilon^{-1}$

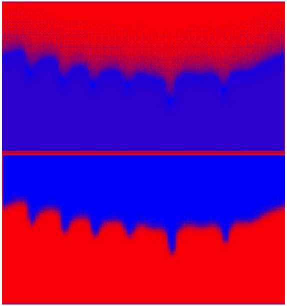
Contents

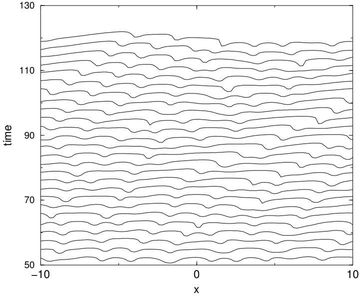
Simulations of step dynamics with phase field

Phase field models → no interface tracking
Phase field model with Schwoebel effect

OPL 2003



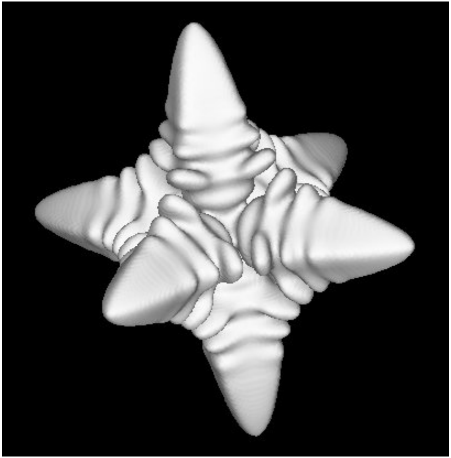




Other phase field simulations

Dendritic Growth

M. Plapp, E.P., France



Conclusion

- Interface tracking vs Interface Capturing $\rightarrow$ very effective numerical method
- Can add, elastic strain, hydrodynamics, temperature fields, etc...
- Can include fluctuations

Contents

Burton-Cabrera-Frank step model

At the steps

Thermodynamic Fluxes

$$J_\pm = \pm D\mathbf{n}.\nabla c_\pm$$

Fluxes prop. to forces + Onsager reciprocity

$$\begin{aligned} J_+ &= \nu_+(c_+ - c_{eq}) + \nu_0(c_+ - c_-) \\ J_- &= \nu_-(c_- - c_{eq}) - \nu_0(c_+ - c_-) \end{aligned}$$

Gibbs-Thomson $\mu = \Omega\tilde{\gamma}\kappa$, with $\tilde{\gamma} = \gamma + \gamma''$

$$c_{eq} = c_{eq}^0 \mathrm{e}^{\mu/k_B T}$$

Mass conservation

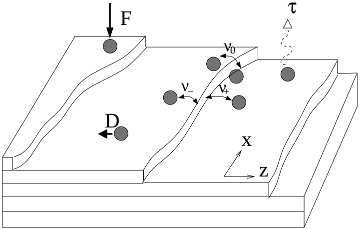
$$V\Delta c = J_+ + J_-$$

with $\Delta c = 1/\Omega + c_- - c_+$

On terraces:

diffusion + incoming flux + evaporation

$$\partial_t c = D\nabla^2 c + F - c/\tau$$



$\nu_+ > \nu_-$ Ehrlich-Schwoebel effect

$\nu_0 \neq 0$ Step transparency or permeability

re-Writing Step Boundary Conditions

Linear combination of BC

$$\begin{aligned} J_+ &= \tilde{\nu}_+(c_+ - \tilde{c}_{eq}) \\ J_- &= \tilde{\nu}_-(c_- - \tilde{c}_{eq}) \end{aligned}$$

Gibbs-Thomson $\mu = \Omega\tilde{\gamma}\kappa$, with $\tilde{\gamma} = \gamma + \gamma''$

$$\tilde{c}_{eq} = c_{eq}^0 \mathrm{e}^{\mu/k_B T} + \tilde{\beta}\frac{V}{\Omega}$$

with

$$\begin{aligned} \tilde{\nu}_+ &= \frac{\nu_+\nu_- + \nu_0(\nu_+ + \nu-)}{\nu_-} \\ \tilde{\nu}_- &= \frac{\nu_+\nu_- + \nu_0(\nu_+ + \nu-)}{\nu_-} \\ \tilde{\beta} &= \frac{\nu_0}{\nu_+\nu_- + \nu_0(\nu_+ + \nu-)} \end{aligned}$$

Identical to thin interface limit of phase field!

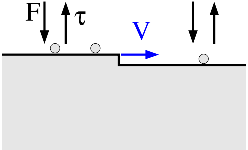
Stationary states

No permeability (transparency) $\nu_0 = 0$ and quasistatic approx $\partial_t c = 0$

1) Isolated straight step velocity with evaporation

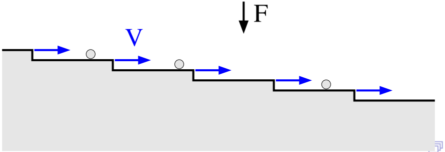
$$\bar{V} = \Omega(F - c_{eq}^0/\tau)x_s^2 \frac{2x_s + d_+ + d_-}{x_s^2 + x_s d_+ + x_s d_- + d_+ d_-}$$

diffusion length $x_s = (D\tau)^{1/2}$
Schwoebel lengths $d_\pm = D/\nu_\pm$



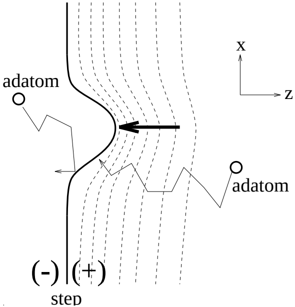
2) Vicinal surface: Step Flow velocity without evaporation

$$\bar{V} = \Omega F \ell$$



Contents

Schwoebel effect → Meandering instability



Bales and Zangwill 1990
(∼ Mullins and Sekerka)

isolated step; one-sided model
($\nu_0 = 0$, $\nu_- = 0$, $\nu_+ \to \infty$)
Model equations

$$\begin{aligned} \partial_t c &= \partial_{zz} c + \partial_{xx} c + F - \frac{c}{\tau} \\ c(z = h(x,t)) &= c^0_{eq} \exp[-\Gamma\kappa] \\ \frac{1}{\Omega D}\partial_t h(x,t) &= \partial_z c(z = h) - \epsilon \partial_x h \partial_x c(z = h) \end{aligned}$$

with $\Gamma = \Omega\tilde{\gamma}/k_B T$, and

$$\kappa = \frac{-\partial_{xx} h}{(1 + (\partial_x h)^2)^{3/2}}$$

Meandering/ Linear Analysis: Isolated step

isolated step; one-sided model ($\nu_0 = 0$, $\nu_- = 0$, $\nu_+ \to \infty$)

Stationary state at constant velocity

$$\bar{V} = \Omega(F - c_{eq}/\tau)(D/\tau)^{1/2}$$

Linear Stability
Small perturbations, Fourier Mode

$$\zeta(x,t) \sim Ae^{i\omega t + iqx}$$

Dispersion relation
Long wavelength $qx_s \ll 1$

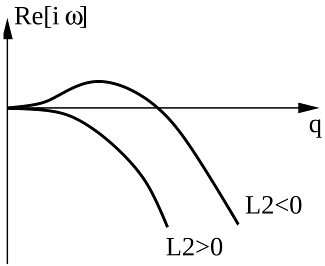
$$i\omega = \frac{x_s^2}{2}\Omega(F - F_c)q^2 - \frac{3}{4}x_s D\Omega c_{eq}^0 \Gamma q^4$$

$\to$ Morphological instability
for $F > F_c = F_{eq}(1 + \frac{2\Gamma}{x_s})$

Bales and Zangwill 1990

An instability occurs if: $\Re e[i\omega(q)] > 0$

$$\Re e[i\omega] \quad = \quad -L_2 q^2 + L_4 q^4$$



Weakly unstable $\epsilon \sim (F - F_c)$
$q \sim \epsilon^{1/2}$; $i\omega \sim \epsilon^2$
$x \sim \epsilon^{-1/2}$; $t \sim \epsilon^{-2}$

Meandering/ Nonlinear behavior: Isolated step, Epavoration

Weakly unstable $\epsilon \sim (F - F_c)$
$x \sim \epsilon^{-1/2}$; $t \sim \epsilon^{-2}$; $\zeta \sim \epsilon$

$$x = X\epsilon^{-1/2}; \qquad t = T\epsilon^{-2}$$
$$\zeta(x,t) = \epsilon H(X,T) = \epsilon[H_0(X,T) + \epsilon H_1(X,T) + \epsilon^2 H_2(X,T) + ...]$$
$$c(x,t) = c_0(X,T) + \epsilon c_1(X,T) + \epsilon c_2(X,T) + ...$$

Model equations

$$\epsilon^2 \partial_T c = \partial_{zz} c + \epsilon \partial_{XX} c + F - \frac{c}{\tau}$$
$$c(z = \epsilon H(X,T)) = c_{eq}^0 \exp[-\Gamma\kappa]$$
$$\frac{\epsilon^3}{\Omega D}\partial_T H = \partial_z c(z = \epsilon H) - \epsilon \partial_X H \partial_X c(z = \epsilon H)$$

with

$$\kappa = \epsilon^2 \frac{-\partial_{XX} H}{(1 + \epsilon^3(\partial_X H)^2)^{3/2}}$$

Meandering/ Nonlinear behavior: Isolated step, Epavoration

0th order in $\epsilon \to$ Straight step solution

...

3rd order in ϵ

$$\partial_t \zeta = -\frac{x_s^2}{2}(F - F_c)\Omega \partial_{xx}\zeta - \frac{3}{4} x_s D \Omega c_{eq}^0 \Gamma \partial_{xxxx}\zeta + \frac{\bar{V}}{2}(\partial_x \zeta)^2$$

Kuramoto-Sivashinsky Equation

Out of equilibrium $\to$ Non-variational

Rescaled

$$\partial_t \zeta = -\partial_{xx}\zeta - \partial_{xxxx}\zeta + (\partial_x \zeta)^2$$

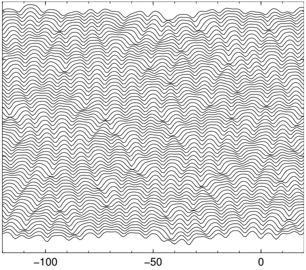
Geometric origin of nonlin $V_n = \bar{V}$

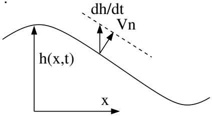
$$V_n = \frac{\partial_t h}{[1 + (\partial_x h)^2]^{1/2}}$$

$$\partial_t h = \bar{V}[1 + (\partial_x h)^2]^{1/2} \approx \bar{V}[1 + \frac{1}{2}(\partial_x h)^2]$$

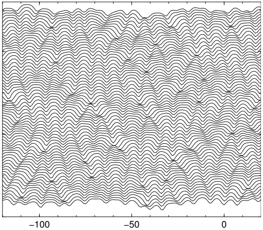
Chaotic dynamics

Bena-Misbah-Valance 1994

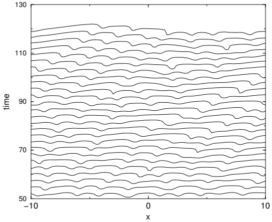




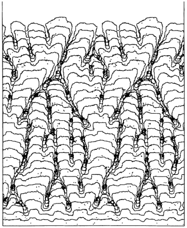
Meandering/ Nonlinear behavior: Isolated step, Epavoration



Kuramoto-Sivashinsky Bena-Misbah-Valance 1994



Phase field OPL 2003



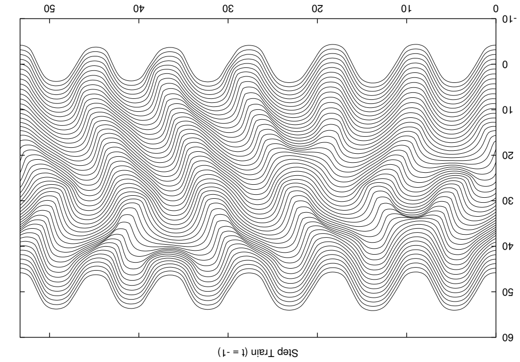
KMC Saito, Uwaha 1994

Meandering/ Nonlinear behavior: Vicinal surface, no evaporation

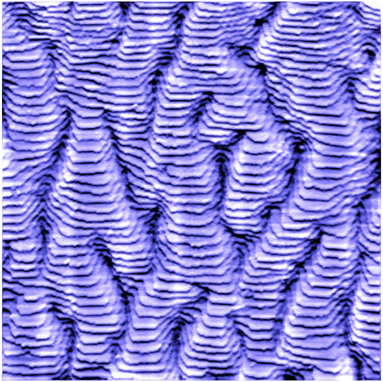
$\epsilon \sim F$

Highly nonlinear equation $x \sim \zeta \sim \epsilon^{-1/2}$

$$\partial_t \zeta = -\partial_x \left[\frac{\epsilon \partial_x \zeta}{1 + (\partial_x \zeta)^2} + \frac{1}{1 + (\partial_x \zeta)^2} \partial_x \left(\frac{\partial_{xx} \zeta}{(1 + (\partial_x \zeta)^2)^{3/2}} \right) \right]$$



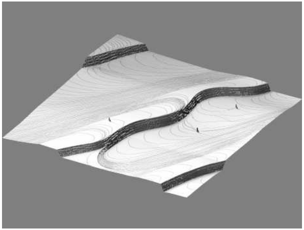
Danker, OPL, Misbah 2005



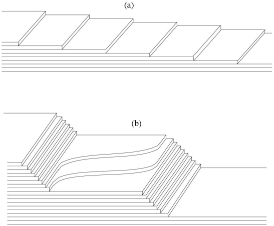
Maroutian et al 2000

Step bunching

Inverted ES effect, or electromigration
Electromigration Si(111)



E.D. Williams, Maryland, USA

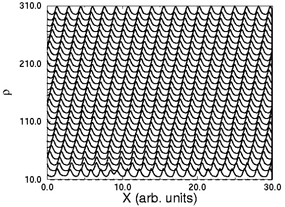


Step bunching

with evaporation
distance to instability threshold ϵ
Generic equation Benney:

$$\partial_t h = -\epsilon \partial_{xx} h + b \partial_{xxx} h - \partial_{xxxx} h + (\partial_x h)^2$$

Ordered/chaotic bunches $\rho = \partial_x h$



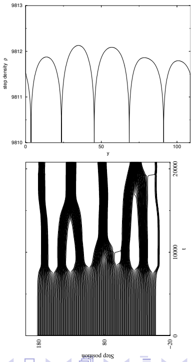
Electromigration on vicinal surfaces

M. Sato, M. Uwaha, 1995 O. Pierre-Louis, C. Misbah 1995

without evaporation: Highly nonlinear dynamics
$\rightarrow$ singular facets appear
$\rho = \partial_x h$
migration force $\sim \epsilon$

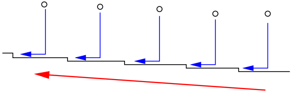
$$\frac{V}{\Omega} = a \partial_y \left[\frac{\epsilon c_{eq} + a \rho \partial_y c_{eq}}{1 + (d_+ + d_-)\rho} \right]$$

Separation of the bunches

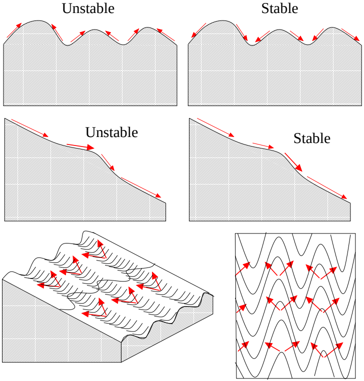


Non-equilibrium mass fluxes and morphological stability

Growth with Ehrlich-Schwoebel effect $\rightarrow$ mass flux



Mass fluxes **along** the surface



Ehrlich-Schwoebel effect: Mound formation

Growth with Ehrlich-Schwoebel effect
$\rightarrow$ mass flux

$$J = F\frac{\ell}{2} = \frac{F}{2|\nabla h|}$$

Continuum limit:

$$\partial_t h = F - \partial_x J$$

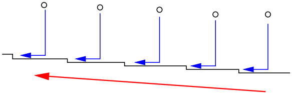
Separation of varliables: $h = Ft + A(t)g(x)$

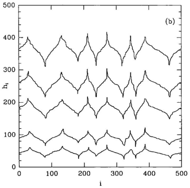
$$2A\dot{A} = \frac{g''}{g(g')^2}$$

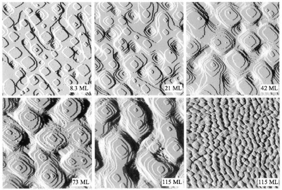
leading to

$$A \sim t^{1/2} \quad \text{and} \quad g \sim erf^{-1}(x)$$

Singular shape $\rightarrow$ mounds separated by sharp trenches
2D nucleation controls the shape of mound tops







Conclusion

- Steps dynamics $\rightarrow$ nanoscale morphology
- Growth, or evaporation/dissolution ...
  but also: electromigration, sputtering, dewetting dynamics, strain-induced instabilities, etc.
- Non-equilibrium steady-states and morphological instabilities
- Universality in pattern formation: Kuramoto Sivashinsky example

Contents

What happens when the surface is unstable? Nonlinear Dynamics?

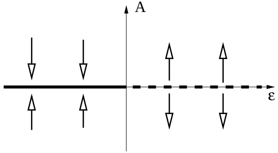
Preamble: simpler case with 1 degree of freedom $A(t)$

One dimensional dynamics with one parameter:

$$\partial_t A = F(A, \lambda)$$

Linear stability around threshold $\epsilon = \lambda - \lambda_c$

$$dA/dt = \epsilon A$$

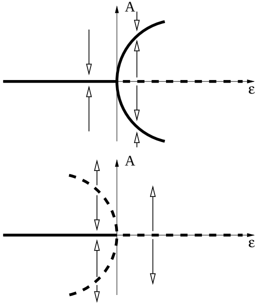


Steady state loosing its stability
Time scale: $t \sim \epsilon^{-1}$
critical slowing down
Unstable → large amplitude → nonlinear dynamics

Symmetry $A \to -A$

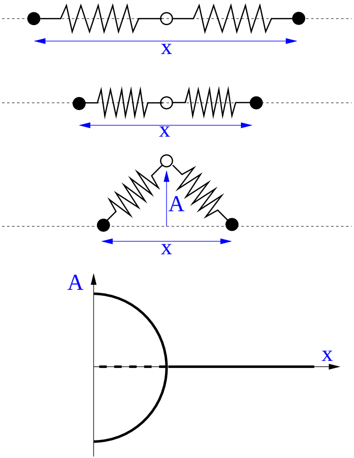
$$dA/dt = \epsilon A + c_3 A^3$$

Super-critical or Sub-critical bifurcation



Scales $t \sim \epsilon^{-1}$, $A \sim \epsilon^{1/2}$
next order $A^5 \sim \epsilon^{5/2}$ negligible $\ll$ other terms $\sim \epsilon^{3/2}$

Two springs in a plane

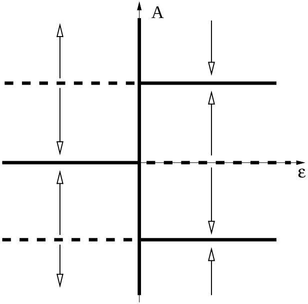


Constraint on ϵ

$\partial_t A = F(A, \epsilon)$

If $F(A, \epsilon) \to 0$ when $\epsilon \to 0$

$$dA/dt = \epsilon Q(A) + o(\epsilon^2)$$

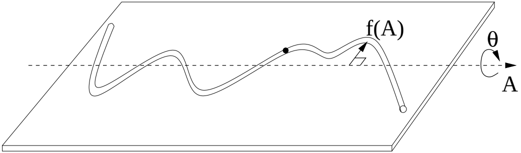


with $Q(A) = \partial_\epsilon F(A, \epsilon)|_{\epsilon=0}$

Scales $t \sim \epsilon^{-1}$, $A \sim 1$

Highly nonlinear dynamics

A tube on a plane



h local height
$f(A)$ in plane meander of the tube
m particle mass
g gravity
ν friction
Particle velocity

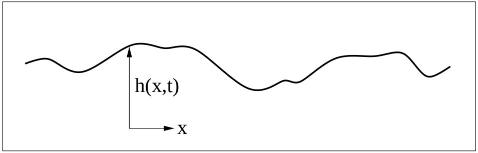
$$V = -\nu \partial_s [mgh]$$

$$\partial_t A = -\nu mg \sin(\theta) \frac{f'(A)}{1 + f'(A)^2}$$

$\epsilon = \sin(\theta)$
Dynamics are highly nonlinear around $\theta = 0$

Contents

Front dynamics



front $h(x, t)$
Translational invariance in x, and in h
($\rightarrow$ dynamics depends on derivs of h only)
local dynamics
Instability at long wavelength
Examples: crystal growth, flame fronts, sand ripple formation, etc....

O. Pierre-Louis EPL 2005

Systematic analysis

Linear analysis, Fourier modes:

$$h(x,t) = h_{\omega q}\exp[i\omega t + iqx]$$

Dispersion relation $D[i\omega, iq] = 0$

Local dynamics

Expansion of the dispersion relation at large scales:

$$\begin{aligned} i\omega &= L_0 + iL_1 q - L_2 q^2 - iL_3 q^3 + L_4 q^4 + \dots \\ \partial_t h &= L_0 h + L_1 \partial_x h + L_2 \partial_{xx} h + L_3 \partial_{xxx} h + L_4 \partial_{xxxx} h \end{aligned}$$

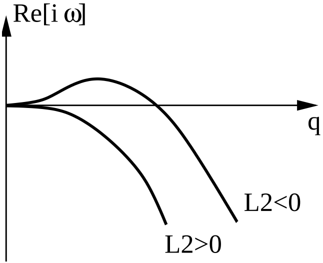
Translational invariance $L_0 = 0$

Gallilean transform $x \to x + L_1 t$ $(\partial_t \to \partial_t + L_1 \partial_x)$

$$\partial_t h = L_2 \partial_{xx} h + L_3 \partial_{xxx} h + L_4 \partial_{xxxx} h + \dots$$

An instability occurs if: $\Re e[i\omega(q)] > 0$

$$\Re e[i\omega] = -L_2 q^2 + L_4 q^4$$



Long wavelength instab. $L_4 < 0$, $-L_2 = \epsilon$
Instability scales $t \sim \epsilon^{-2}$, $x \sim \epsilon^{-1/2}$
Fast propagative time scale $t \sim \epsilon^{-3/2}$

Weakly nonlinear expansion

Generic formal expansion

$$\partial_t h = -\epsilon \partial_{xx} h + L_3 \partial_{xxx} h + L_4 \partial_{xxxx} h + \epsilon^\gamma [\partial_x]^n [\partial_t]^l [h]^m$$

Power counting

scaling of $h \sim \epsilon^\alpha$

Linear terms $\sim \epsilon^{2+\alpha}$

Nonlinear term $\sim \epsilon^{\gamma + n/2 + l + m\alpha}$

$$\alpha = \frac{2 - \gamma - n/2 - l}{m - 1}$$

Nonlinearity which saturates the amplitude the soonest wins!

$\rightarrow$ selection of the nonlinearity having the biggest α

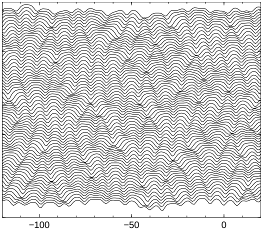
Condition for WNL

$\partial_t h, \partial_x h \ll 1 \rightarrow \alpha > -1/2$

Generic equation Benney:

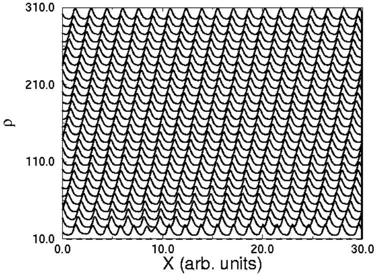
$$\partial_t h = -\epsilon\partial_{xx} h + b\partial_{xxx} h - \partial_{xxxx} h + (\partial_x h)^2$$

$b = 0$ in the presence of $x \to -x$ symmetry
$\to$ Kuramoto-Sivashinsky equation



Step growth with Schwoebel effect

I Bena, A. Valance, C. Misbah, 1993

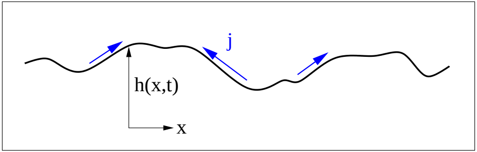


Electromigration on vicinal surfaces

M. Sato, M. Uwaha, 1995 O. Pierre-Louis, C. Misbah 1995

Conservation law

$$\partial_t h = -\partial_x j$$



Vicinity to thermodynamic equilibrium

(or variational steady-state)
driving force ϵ

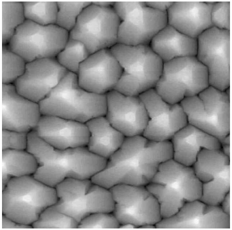
$$\partial_t h = \partial_x \left[M \partial_x \frac{\delta \mathcal{F}}{\delta h} - \epsilon J \right] + o(\epsilon^2)$$

Highly nonlinear dynamics

$$\partial_t h = \partial_x \left[\epsilon A + B \partial_{xx} C \right]$$

where A, B, C functions of $\partial_x h \sim 1$
$x \to -x$ symmetry; non-variational
$\to$ Examples in Molecular Beam Epitaxy

Mound formation in MBE



Experiments:
T. Michely, Aachen, Germany.
Deposition of 300 atomic layers of Pt at 440K; 660 nm x 660 nm.
Theory:
J. Villain, CEA Grenoble, France
Non-equilibrium mass flux due to ES effect
used Highly nonlinear equations

$$\partial_t h = \partial_x[\epsilon A + B\partial_{xx} C]$$

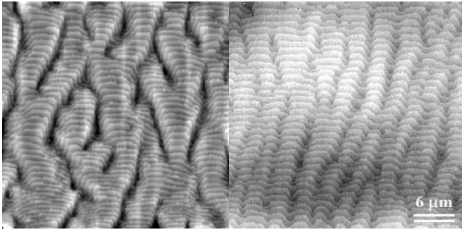
$\epsilon = F$

Step meandering in MBE

Experiments:

on Cu(100) Maroutian, Néel, Douillard, Ernst, CEA-Saclay, France.

Si(111) H. Hibino, NTT, Japan.



Theory: O. Pierre-Louis, C. Misbah, *LSP Grenoble.*

G. Danker, K. Kassner, *Univ. Otto von Guerricke Magdeburg, Germany.*

Multi-scale expansion from step model

$\rightarrow$ Highly nonlinear dynamics

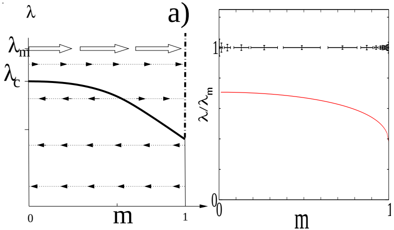
$$\partial_t h = \partial_x[\epsilon A + B\partial_{xx} C]$$

$\epsilon = F$

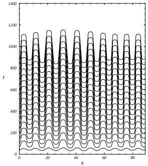
In phase meander

Free energy $\mathcal{F} = \int ds\gamma$
Chemical potential $\mu = \gamma\kappa$
κ step curvature
Relaxation via terrace diffusion

$$V_n = \partial_s \left[\frac{D\ell_\perp c_{eq}}{k_B T} \partial_s \mu \right]$$

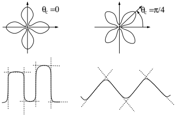


$$\partial_t \zeta = -\partial_x \left[\frac{\alpha \partial_x \zeta}{1 + (\partial_x \zeta)^2} + \frac{\beta}{1 + (\partial_x \zeta)^2} \partial_x \left(\frac{\partial_{xx} \zeta}{(1 + (\partial_x \zeta)^2)^{3/2}} \right) \right]$$

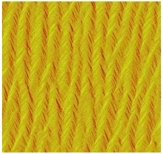


with $\alpha = F\ell^2/2$ and $\beta = Dc_{eq}\ell\gamma/k_B T$.
obtained from step model
$m = max[\partial_x \zeta/(1 + (\partial_x \zeta)^2)^{1/2}]$

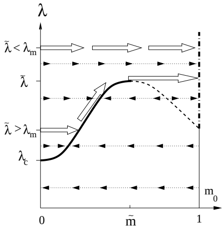
Can steps be pinned by anisotropy??
Energetic: $\gamma(\theta)$
or
Kinetic: Ehrlich-Schwoebel effect, Edge diffusion.
Same qualitative change:
1) $\theta_c = 0 \rightarrow$ no effect
2) $\theta_c = \pi/4 \rightarrow$ interrupted coarsening.

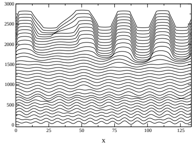


Si(111) growth



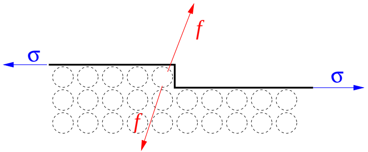
Hiroo Omi, Japan.



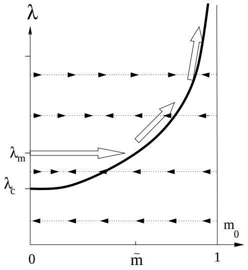


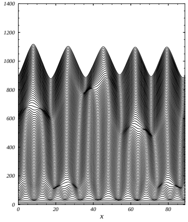
Anisotropy → Interrupted coarsening

Elastic interactions in homo-epitaxy:



Force dipoles at step:
$\rightarrow$ interaction energy $\sim 1/\ell^2$
between straight steps.





Elastic interactions $\rightarrow$ endless coarsening

From the study of step meandering

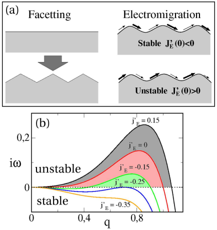
3 scenarios:

- frozen wavelength (no coarsening) growing amplitude OPL et al PRL 1998
- interrupted coarsening (nonlin wavelength selection) Danker et al PRL 2004
- endless coarsening Paulin et al PRL 2001

Can coarsening dynamics can generically be guessed from the branch of steady states only?

P. Politi - C. Misbah PRL 2004, Pierre-Louis 2014 $\longrightarrow$ phase stability

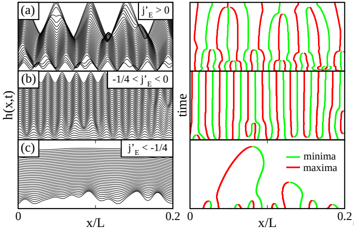
Facetting+Electromigration



$$\phi = \partial_x h \qquad \partial_t \phi = \partial_{xxxx}\left[\partial_{xx}\phi + \phi - \phi^3\right] - j'_E \partial_{xx}\phi \ .$$

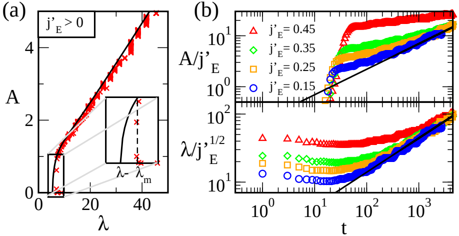
4th order equation for steady-states

3 regimes: $j'_E > 0$ reinforced instability; $0 > j'_E > -1/4$ weak instability; $-1/4 > j'_E$ stability

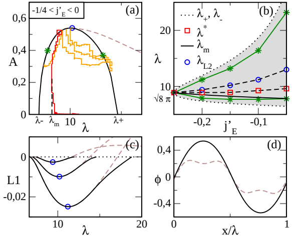


Facetting+Electromigration

$j'_E > 0$ reinforced instability $\rightarrow$ endless coarsening



$0 > j'_E > -1/4$ weak instability $\rightarrow$ frozen wavelength



F. Barakat, K. Martens, O. Pierre-Louis, Phys Rev Lett 2012

Conclusion

- weakly vs highly nonlinear dynamics
- Fronts close to thermodyn equil, or variational steady-state (Lyapunov functional)
- Higher order problems?

Other examples of Highly nonlinear dynamics

- Step bunching

  J. Chang, O. Pierre-Louis, C. Misbah, PRL 2006
- Oscillatory driving of crystal surfaces

  O Pierre-Louis, M.I. Haftel, PRL 2001
- Similar results without translational invariance

  O. Pierre-Louis EPL 2005

Contents

Example: Ballistic Deposition (from Barabasi and Stanley Fractal Concepts in Surf. Growth)

Irreversible attachment
Ballistic Deposition

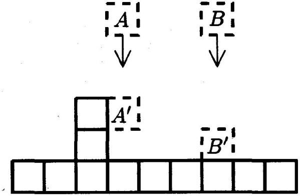
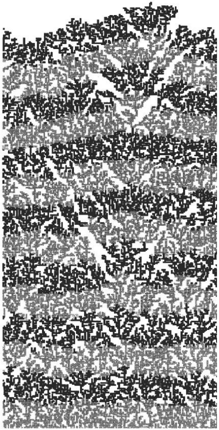


Figure 2.2 A BD cluster obtained by depositing 35 000 particles on a substrate of horizontal size $L = 200$. The shading reflects the arrival time of the particles: after the deposition of each set of 2500 particles, the shading changes. The eye can readily recognize that the roughness increases in time.

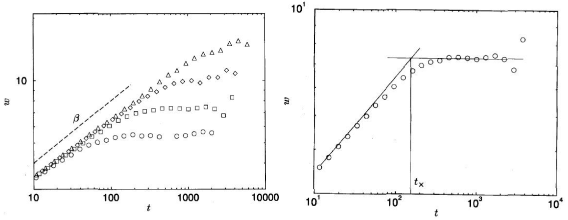


Zinc Electrodeposition

Sawada 1985

Roughness

Figure 2.4 The time evolution of the surface width for BD. Logarithmic scales are used so that power-law dependence can be seen as a straight line. The different curves correspond to simulations for different system sizes L: $L = 100$ (○), $L = 200$ (□), $L = 400$ (◇), and $L = 800$ (△). The system shown on Fig. 2.2 corresponds to the squares on this figure. The dashed line has slope β.



$$W(t, L) = \left[\frac{1}{L}\int_0^L dx\, \zeta^2(x,t)\right]^{1/2}$$

$$\zeta(x,t) = h(x,t) - \frac{1}{L}\int_0^L dx\, h(x,t)$$

Short times $t \ll t_\times$, roughening, *growth exponent* β

$$W(t, L) \sim t^\beta$$

Long times $t \gg t_\times$, Steady-state, *roughness exponent* α

$$W(t, L) \sim L^\alpha$$

Scaling

Growth of the correlation length , *dynamic exponent z*

$$\xi \sim t^{1/z}$$

Correlations reach the system size at $\xi \sim L$

$$t_\times \sim L^z$$

Crossover condition $W(t,L) \sim t_\times^\beta \sim L^\alpha$

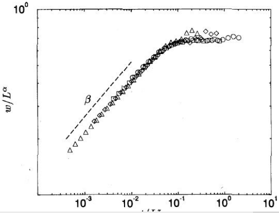
$$\beta = \frac{\alpha}{z}$$

Family-Viscek ansatz

$$W(t) \sim L^\alpha f\left(\frac{t}{L^z}\right).$$

$f(u) \sim u^\beta$ for $u \to 0$
$f(u) \sim cst$ for $u \to \infty$.

Figure 2.6 The BD simulations of Fig. 2.4 rescaled according to (2.8). The obtained curve is in fact the scaling function $f(u)$, with the properties (2.9) and (2.10). The different symbols correspond to runs with different system sizes $L = 100$ (○), $L = 200$ (□), $L = 400$ (◇), and $L = 800$ (△). The slope of the dashed line is β.



Contents

Continuum Models 1: Edwards-Wilkinson model

Linear Equation:

$$\partial_t h(x,t) = V + \nu \partial_{xx} h(x,t) + \eta(x,t)$$
$$\langle \eta(x,t)\eta(x',t')\rangle = 2D\delta(x-x')\delta(t-t')$$

η white Gaussian, zero average $\langle \eta(x,t)\rangle = 0$

For $d=1$, $\alpha = 1/2$, $\beta = 1/4$, $z=2$

$$\langle W(t)^2\rangle^{1/2} = \left(\frac{D}{\nu}\right)^{1/2} L^{1/2} f\left(\frac{\nu t}{L^2}\right)$$
$$f(u) = \left[2\sum_{n=1}^{\infty}\frac{1-\mathrm{e}^{-8\pi^2 n^2 u}}{4\pi^2 n^2}\right]^{1/2}$$

$f(u) \to 1/12^{1/2}$ for $u \to \infty$,
$f(u) \to (2u/\pi)^{1/4}$ for $u \to 0$.

For arbitrary d

$$\alpha = \frac{2-d}{2}, \quad \beta = \frac{2-d}{4}, \quad z = 2$$

Continuum Models 2.1: Kardar Parisi Zhang model

Nonlinear Equation:

$$\partial_t h(x,t) = V + \nu \partial_{xx} h(x,t) + \frac{\lambda}{2}(\partial_x h)^2 + \eta(x,t)$$

For $d = 1$, many exact results

$$\alpha = 1/2, \quad \beta = 1/3, \quad z = 3/2$$
$$h(t) = v_\infty t + (\Gamma t)^\beta \chi + ...$$

χ obeys the Tracy-Widom distribution Spohn et al 2000, 2010

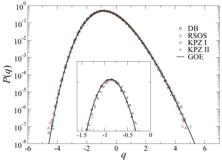


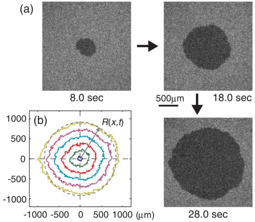
FIG. 2. (Color online) Height distributions scaled accordingly Eq. (2) for all investigated models. A simulation time of $t = 2 \times 10^4$ was used for BD and RSOS and $t = 800$ for the integrated KPZ equation. The solid line is the GOE distribution. The inset shows a zoom around the peak of the distributions.

T. J. Oliveira, S. C. Ferreira, and S. G. Alves et al 2012

For $d > 1$ no exact expression of the exponents
But for arbitrary d: $\alpha + z = 2$ (Galilean invariance)
Numerical results, e.g. for d=2: $\beta \approx 0.241..$, and $\alpha \approx 0.393..$

Continuum Models 2.2: Kardar Parisi Zhang model

Roughening is a universal phenomena also valid for other systems
Topological-defect turbulence in electroconvection of nematic liquid crystals Takeuchi, Sano PRL 2010



Paper burning M. Myllys et al Phys rev E 2001

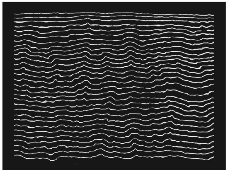
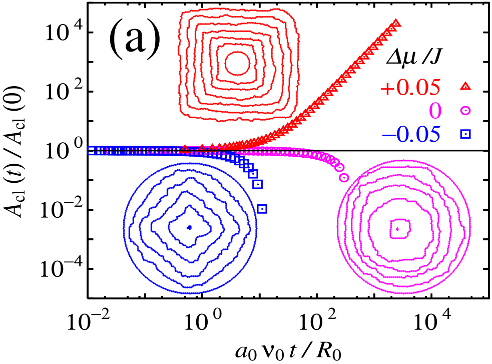


FIG. 2. Series of typical digitized fronts. The time step between successive fronts is 10 s, and the width of the digitized area is 310 mm.

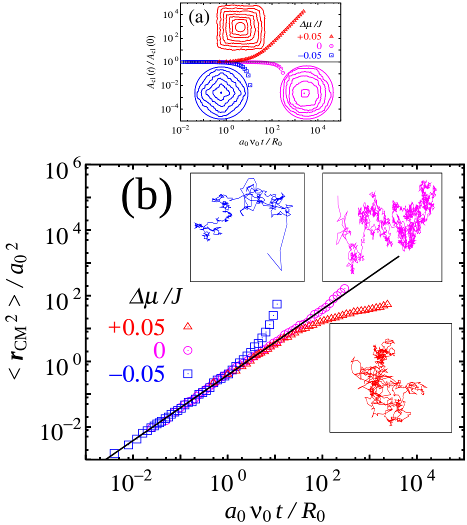
Kinetic Monte Carlo Model

Kinetic Monte Carlo simulations (Y. Saito)
attachment rate $\nu_0 e^{\Delta\mu/k_B T}$
detachment rate $\nu_0 e^{-(n-2)J/k_B T}$
temperature $T = 0.2J/k_B < T_c \approx 0.567J/k_B$



average over 100 runs

Center of mass diffusion



average over 100 runs

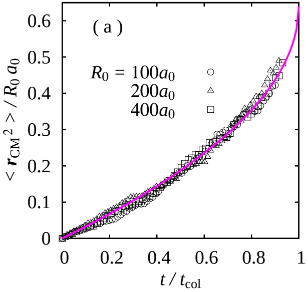
Comparison to KMC model

Curvature-driven evaporation

$\Delta\mu = 0$

$$\langle \mathbf{r}_{CM}^2(t) \rangle = \frac{4\Omega R_0}{\pi\Gamma}\left[1 - (1 - t/t_{col})^{1/2}\right],$$

$t_{col} = R_0^2/(2\Gamma K)$



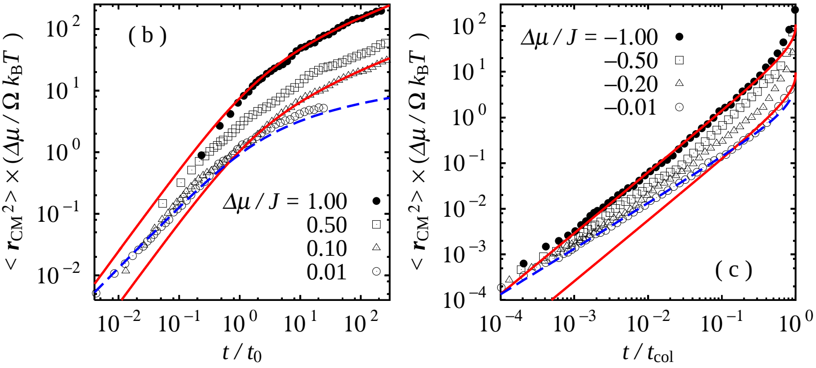
Comparison with KMC

KPZ $b = 1/3$

Short times $\langle \mathbf{r}^2_{CM}(t) \rangle \sim t^{4/3}$

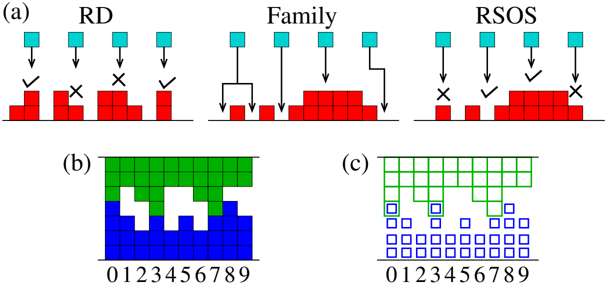
Long times $\langle \mathbf{r}^2_{CM}(t) \rangle \sim t^{1/3}$

Noise amplitude B_f obtained from KMC simulations



Collision of two interfaces

Simulation Models (irreversible)



Random Deposition (RD): $\beta = 1/2$
Family (EW): $\alpha = 1/2$, $\beta = 1/4$, $z = 2$
RSOS (KPZ): $\alpha = 1/2$, $\beta = 1/4$, $z = 2$
MOVIE ...
Extract from simulations:

- Collision times (duration of the collision) $t_c(x) = t_0 + \delta t_c(x)$
- locus of collision (roughness of the collision line) $h_c(x) = h_0 + \delta h_c(x)$

Collision of two interfaces

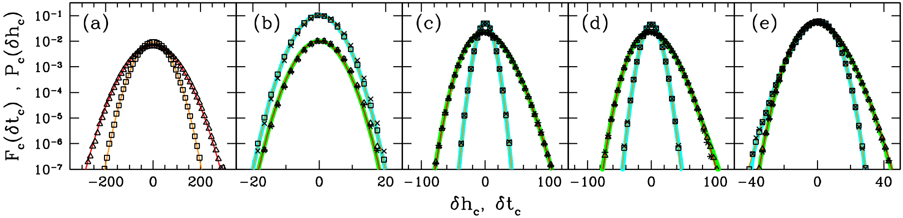
- Interactions during collision irrelevant
- Fluctuations during collision irrelevant

Interfaces without collision: distributions $P_\pm\left(\zeta_\pm;t\right)$
Distributions of collision times δt_c

$$F_c\left(\delta t_c\right) \quad = \quad \left(v_+ + v_-\right)\int d\zeta_+ P_+\left(\zeta_+;t_0\right)P_-\left(-\delta t_c(v_+ + v_-) - \zeta_+;t_0\right)$$

Distributions of collision locus (grain boundary roughness) δh_c

$$P_c\left(\delta h_c\right) \quad = \quad \frac{v_+ + v_-}{v_+}\int d\zeta_+ P_+\left(\zeta_+;t_0\right)P_-\left(\frac{-\delta h_c(v_+ + v_-) + \zeta_+ v_-}{v_+};t_0\right)$$



Conclusions on Kinetic Roughening

Many other models with other exponents, depending on

- Symmetries
- Conservation laws

→ Universality classes (EW, KPZ, ...)

More:

- correlation functions and power spectrum
- Persistence properties, Extremal Value distributions
- Fluctuations Macroscopic quantities
- Flat and curved geometries

Conclusion

Modelling Crystal Surface Dynamics

Tools

- KMC
- Phase field
- Sharp interface models (such as BCF Step models)

Phenomena

- Growth-dissolution rate
- Kinetic Roughening
- Morphology: relaxation or instabilities $\rightarrow$ patterns

Global remarks

- Surface Diffusion vs Bulk diffusion
- Faithful microscopic vs Effective dynamics
- Multi-scale modeling

Conclusion

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