Continuum Modeling of Surface Relaxation Below the Roughening Temperature

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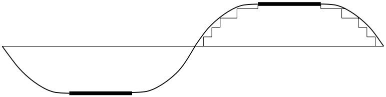
Mainly: PhD thesis of *Irakli Odisharia*
Also: current work with *Henrique Versieux* (numerical analysis)
and recent work with *Dionisios Margetis* (coarse-graining)

The subject

Physically: this talk concerns relaxation of a crystalline surface below the roughening temperature:



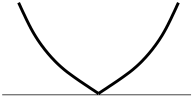
Surface consists of steps and terraces. Facets form at peaks and valleys, as surface relaxes to a single flat facet.

Mathematically: Main focus is the 4th-order steepest descent

$$h_t = -\left[\gamma'(h_x)_x\right]_{xx} \quad \text{assoc to} \quad E = \int \gamma(h_x)$$

where γ is convex but not smooth:

$$\gamma(h_x) = \beta|h_x| + \tfrac{1}{3}|h_x|^3$$



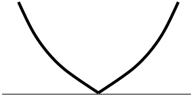
Outline

Mullins' viewpoint

Fourth-order PDE model is well-established for use above the roughening temperature (when surface energy is smooth):

$E = \int \gamma(\nabla h)$	surface energy
$\mu = \frac{\delta E}{\delta h} = -\text{div}\left(\frac{\partial \gamma}{\partial \nabla h}\right)$	chemical potential
$J = -M(\nabla h)\nabla \mu$	J = surface current, M = mobility,
$h_t + \text{div} J = 0$	conservation of mass
$h_t = -\left[\gamma'(h_x)_x\right]_{xx}$	in 1D, if $M = 1$ (diffusion-limited)

Equilibrium shape has facets, so energy density is singular at preferred slope $h_x = 0$.



General mobility

Mullins' argument gives, more generally:

$$h_t = -\left\{ M(h_x) \left[\gamma'(h_x)_x \right]_x \right\}_x .$$

Coarse-graining of step motion law gives

$$M(h_x) = \frac{1}{1 + \frac{D}{ak} |h_x|}$$

where

$$\begin{aligned} D &= \text{terrace diffusion constant} \\ k &= \text{sticking coefficient at step edge} \\ a &= \text{atomic lattice size.} \end{aligned}$$

Diffusion-limited setting corresponds to $\frac{D}{ak} |h_x| \ll 1$.

Steepest descent

Our 4th-order PDE describes H^{-1} steepest descent for E.

Use periodic bc. If $\int f = \int g = 0$ then

$$\begin{aligned} \langle f, g \rangle_{H^{-1}} &= \langle \nabla \Delta^{-1} f, \nabla \Delta^{-1} g \rangle_{L^2} \\ &= \langle f, -\Delta^{-1} g \rangle_{L^2} = \langle -\Delta^{-1} f, g \rangle_{L^2} \end{aligned}$$

When $E = \int \gamma(h_x)$ we have $\nabla_{H^{-1}} E = [\gamma'(h_x)_x]_{xx}$, since

$$\begin{aligned} \frac{d}{dt} E[h(x,t)] &= \int \gamma'(h_x) h_{xt} \\ &= \langle -\gamma'(h_x)_x, h_t \rangle_{L^2} = \langle \Delta \gamma'(h_x)_x, h_t \rangle_{H^{-1}} \end{aligned}$$

So

$$h_t = -\nabla_{H^{-1}} E \quad \Leftrightarrow \quad h_t = -\left[\gamma'(h_x)_x\right]_{xx}$$

A numerical scheme

Implicit Euler solves steepest descent $h_t = -\nabla E$ robustly:

$$\frac{h^{n+1} - h^n}{\Delta t} = -\nabla E(h^{n+1}) \quad \Longleftrightarrow \quad \min_{h^{n+1}} E(h^{n+1}) + \frac{\|h^{n+1} - h^n\|^2}{2\Delta t}$$

Time-step variational problem in our setting is

$$\min_h \int \left(\beta |h_x| + \frac{1}{3} |h_x|^3 \right) + \frac{1}{2\Delta t} \|h - h^n\|^2_{H^{-1}}$$

Reduces to quadratic programming problem

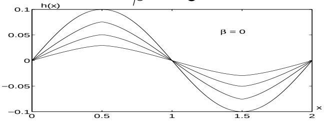
$$\min_{-\sigma(x) \le h_x \le \sigma(x)} \int \beta\sigma + \frac{1}{3} |h_x|^2 \sigma + \frac{1}{2\Delta t} \|h - h^n\|^2_{H^{-1}}$$

(approximate $|h_x|^2\sigma$ by 2nd-order Taylor expnsn around h^n).

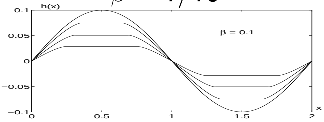
Use finite differences for spatial approximation, so facet is clearly defined as set where $h_x = 0$.

$$E = \int \beta |h_x| + \frac{1}{3}|h_x|^3$$

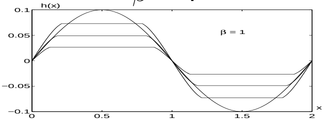
$\beta = 0$



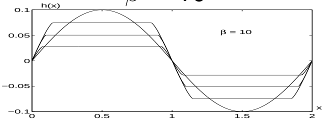
$\beta = 1/10$



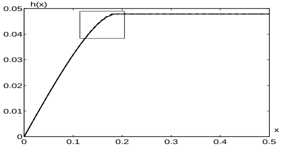
$\beta = 1$

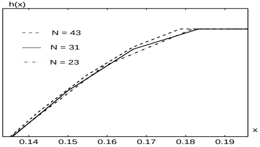


$\beta = 10$

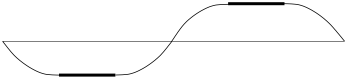


profiles

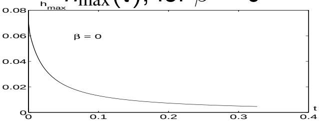




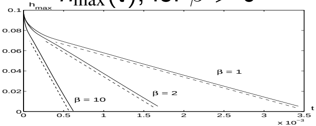
discretization effects



$h_{\max}(t)$, for $\beta = 0$

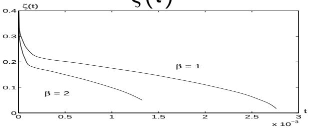


$h_{\max}(t)$, for $\beta > 0$

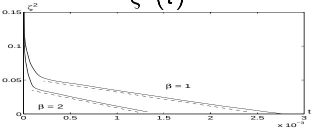


$$h_{\max} \sim \begin{cases} 1/t & \text{if } \beta = 0 \\ T - t & \text{if } \beta > 0 \end{cases}$$

$\zeta(t)$

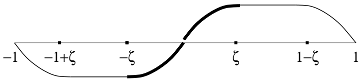


$\zeta^2(t)$



$$\zeta(t) = |\text{complement of facet}| \sim \sqrt{T - t}$$

Self-similarity

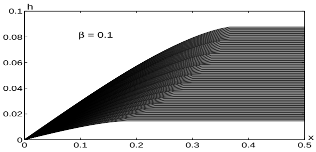


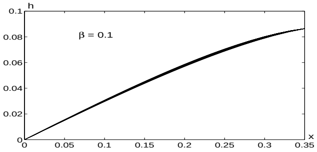
For “sinusoidal” initial data, profile away from facets is asymptotically self-similar:

$$h \sim h_{\max}(t)\phi_0(x/\zeta(t)) \quad \text{for } |x| < \zeta(t)$$

with $h_{\max}(t) \sim H_0(1 - t/T)$ and $\zeta(t) \sim \zeta_0(1 - t/T)^{1/2}$.

Graphical meaning: scaled profiles are independent of time





Coming soon: an explanation (including formulas for ϕ_0, H_0, ζ_0).

But first, a digression . . .

1. Getting started
2. Scaling and self-similarity
3. Alternative numerical schemes
4. Analysis of self-similarity
5. Is the PDE model correct?

The competition

Most widely-used method is a spectral Galerkin scheme (Shenoy, Freund, Ramasubramaniam):

- look for $h(x,t) = \sum_{|k|<N} a_k(t)e^{ikx}$
- do steepest-descent in this finite-dim'l space

Amounts to a nonlinear ODE for mode amplitudes $\{a_k(t)\}$:

$$da/dt = \frac{ik^3}{2\pi} \int \mathrm{sgn}(h_x)(\beta + h_x^2)e^{-ikx}\, dx.$$

Advantages: a) conceptually simple
b) equally easy in 2+1 dimensions

Disadvantages: a) slow convergence in N= # of modes
b) edge of facet isn't sharply defined

Source of slow convergence in N: h is singular at facet edge ($h_x = 0$ there, but $h_{xx} = \infty$).

Extension of implicit time-stepping to 2+1 dimensions

Problem: Need good numerical scheme for (discrete version of)

$$\min \left\{ \int \beta|\nabla h| + \tfrac{1}{3}|\nabla h|^3 \, dx + \frac{\|h - h^n\|^2}{2\Delta t} \right\}$$

Can't use lin progr since $|\nabla h| \leq \sigma$ is not equiv to list of ineq's.

Solution: Primal-dual (mixed) method. Explain idea by focusing on simpler problem (from image segmentation):

$$\min \int |\nabla h| + \tfrac{1}{2}|h - h^n|^2 \, dx.$$

Main steps:

- regularize: replace $|\nabla h|$ by $(|\nabla h|^2 + \delta)^{1/2}$
- write EL eqn as a system: $$\begin{aligned} -\text{div}\xi + (h - h^n) &= 0 \\ \xi(|\nabla h|^2 + \delta)^{1/2} - \nabla h &= 0 \end{aligned}$$
- solve for h and ξ using Newton's method

Key point: method is robust in limit $\delta \to 0$.

Back to self-similarity . . .

Precise formulation of PDE

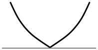
Analogue of $h_t = -\nabla_{H^{-1}} E$ for non-smooth E:

$$-h_t = \text{element of } \partial_{H^{-1}} E(h) \text{ of minimal } H^{-1} \text{ norm.}$$

Same as solution obtained by regularization, e.g. using

$$E_\delta = \int \beta(h_x^2 + \delta^2)^{1/2} + \tfrac{1}{3}|h_x|^3 \quad \text{as } \delta \to 0.$$

$$\begin{aligned} \partial\gamma &= \text{subdifferential of } \gamma \\ &= \{\text{slopes of supporting planes}\} \end{aligned}$$



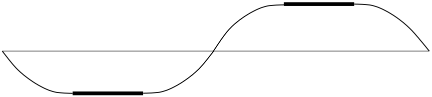
$$\gamma(z) = \beta|z| + \tfrac{1}{3}|z|^3 \Rightarrow \partial\gamma(z) = \begin{cases} \beta + z^2 & z > 0 \\ [-\beta, \beta] & z = 0 \\ -\beta - z^2 & z < 0 \end{cases}$$

$$\begin{aligned} \partial_{H^{-1}} E(h) &= \{w \,:\, E(g) \geq E(h) + \langle w, g - h \rangle_{H^{-1}}\} \\ &= \{v_{xxx} \,:\, v(x) \in \partial\gamma(h_x) a.e.\} \end{aligned}$$

The evolution as a free boundary problem

Finally: $h_t = -v_{xxx}$ where

- off facets: $v = \text{sgn}(h_x)(\beta + h_x^2)$
- on facets: $v =$ cubic polynomial, $|v| \leq \beta$
- at facet edge: v, v_x, v_{xx}, v_{xxx} are cont's

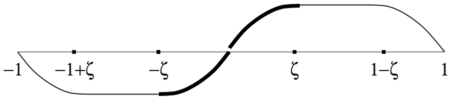


Notes:

- off facets we get the expected 4th order PDE
- on facets, h_t is constant
- conditions at facet edge assure continuity of h; they set both bc for PDE and velocity of facet edge
- h is somewhat singular: $h_x \to 0$ but $h_{xx} \to \infty$ at facet edge

Understanding self-similarity via formal asymptotics

Recall eqn: $h_t = -v_{xxx}$. Assume "sinusoidal symmetry." Then h is odd, v even for $|x| < \zeta$; and h is even, v odd abt facet center.



In central region: expect $h(x,t) = h_{\max}(t)\phi(x/\zeta(t), t)$ with

$$\begin{aligned} h_{\max}(t) &= H_0(1 - t/T) + \text{higher order terms} \\ \zeta(t) &= \zeta_0\sqrt{1 - t/T} + \cdots \\ \phi(\xi, t) &= \phi_0(\xi) + \cdots \end{aligned}$$

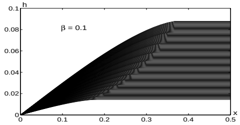
On upper facet: expect $v = a(t)(x - 1/2) + b(t)(x - 1/2)^3$,

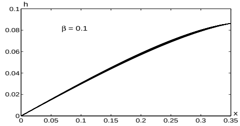
$$a(t) = a_0 + \cdots, \quad b(t) = b_0 + \cdots$$

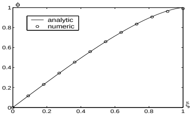
Solve to leading order . . .

Leading order profile is

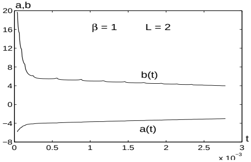
$$\phi_0(\xi) = \frac{4}{\pi}\int_0^{\xi}\sqrt{1-y^2}\,dy = \frac{2}{\pi}(\arcsin\xi + \xi\sqrt{1-\xi^2})$$







On the facet, $v = a(t)(x-1/2) + b(t)(x-1/2)^3$. At leading order:



$$a_0 = -3\beta, \quad b_0 = 4\beta$$

Understanding self-similarity via PDE

Dynamical systems approach to self-similarity: change to *similarity variables*

$$h(x,t) = H_0(1-t/T)\phi(y,s), \quad y = \frac{x}{\zeta_0\sqrt{1-t/T}}, \quad s = \frac{2}{\sqrt{1-t/T}}$$

Key properties:

- no loss of generality; $s \to \infty$ as $t \to T$;
- h becomes self-similar with profile ϕ_0 if $\phi \sim \phi_0$ as $s \to \infty$.

To see (linear) stability of self-similar solution, substitute

$$\phi(y,s) = \phi_0(y) + \frac{1}{s}\eta(y,s)$$

into evolution equation and linearize in η. This gives

$$\eta_s + g\left(\sqrt{1-y^2}\,\eta_y\right)_{yyy} + \frac{1}{s}(y\eta_y - 3\eta) = \frac{4}{\pi}\arcsin(y)$$

with g constant, and bc from linearizing condns at facet edge.

Stabilization

$$\eta_s + g\left(\sqrt{1-y^2}\,\eta_y\right)_{yyy} + \frac{1}{s}(y\eta_y - 3\eta) = \frac{4}{\pi}\arcsin(y)$$

As $s \to \infty$, term with $1/s$ becomes negligible, and η stabilizes to soln of assoc stationary problem.

Why? Problem is linear and operator has gradient structure. (Same reason $u_t - \Delta u = f$ stabilizes.) To see gradient structure: for homogeneous eqn $\eta_s + \left(\sqrt{1-y^2}\,\eta_y\right)_{yyy} = 0$,

$$\begin{aligned}
\frac{d}{ds}\int \frac{1}{2}\sqrt{1-y^2}\,\eta_y^2 &= -\int \left(\sqrt{1-y^2}\,\eta_y\right)_y \eta_s \\
&= \int \left(\sqrt{1-y^2}\,\eta_y\right)_y \left(\sqrt{1-y^2}\eta_y\right)_{yyy} \\
&= -\int \left(\sqrt{1-y^2}\,\eta_y\right)_{yy}^2
\end{aligned}$$

when the bc are homogeneous.

Enough mathematics. Now some physics.

1. Getting started
2. Scaling and self-similarity
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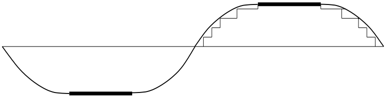
5. Is the PDE model correct?

Summary thus far:

- We've discussed a widely-used model for dynamics "below the roughening temperature."
- Good PDE theory via steepest-descent structure (even in 2D); more detail in 1D (numerics, self-similarity)



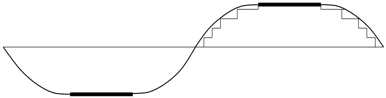
But: is this PDE model correct?

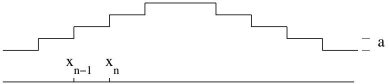
- Maybe (best argument: regularization)
- Maybe not (if the truth is a step-flow model)

Briefly: continuum limit of step dynamics justifies the PDE, but not the "free bdry condition" at the facet edge.

Step-flow model

Burton-Cabrera-Frank picture: surface has steps and terraces. Atoms detach from steps, diffuse along terraces, and attach at nearby steps.





Solve diffusion eqn on each terrace. When dust clears we get:

$E = c_1 \cdot \#steps + c_2 \sum \frac{1}{(x_{n+1}-x_n)^2}$	energy
$\mu_i = \frac{\partial E}{\partial x_i}$	chemical potential
$J_i = -M \frac{\mu_{i+1}-\mu_i}{x_{i+1}-x_i}$	surface current
$\dot{x}_i = -\Omega(J_{i+1} - J_i)$	mass conservation

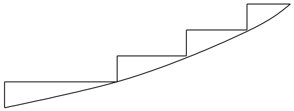
moreover M is constant if $|x_{n+1} - x_n| \gg$ diffusion length.

But tracking steps is laborious. We would rather solve a PDE.

For what PDE is the step motion law a numerical scheme?

Coarse-graining

Assume $h(x_j(t), t) = ja$



- self-energy = $\#steps = \sum_i 1 = \sum \frac{1}{a}\frac{\Delta h}{\Delta x}\Delta x_i \rightarrow \frac{1}{a}\int |h_x|$
- inter'n energy = $\sum \frac{1}{(x_{i+1}-x_i)^2} = \sum \frac{1}{a^3}\left(\frac{\Delta h}{\Delta x}\right)^3 \Delta x_i \rightarrow \frac{1}{a^3}\int |h_x|^3$
- μ_i = first varn of $E \rightarrow \mu = \pm \frac{3}{a^3}(h_x^2)_x$
- $J_i = -M\frac{\mu_{i+1}-\mu_i}{x_{i+1}-x_i} \rightarrow J = -M\mu_x$
- $\dot{x}_i = -\Omega(J_{i+1} - J_i) \rightarrow \frac{h_t}{h_x} = -\Omega a \frac{J_{i+1}-J_i}{h_{i+1}-h_i} = -\Omega a \frac{J_x}{h_x}$

In short: we get the continuum PDE model. (This is why $\gamma(\nabla h) = \beta|\nabla h| + \frac{1}{3}|\nabla h|^3$.)

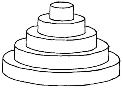
But the argument isn't applicable at the facet edge.

Is the PDE right?

No. The continuum limit of step motion is apparently not the solution of the continuum steepest-descent model.

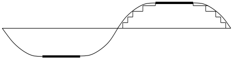
Reason 1: why should it be? Coarse-graining does not support idea that μ is a polynomial in x on the facet.

Reason 2: Recently Margetis, Fok, Aziz, Stone studied analogous question in radial setting. They found:



- PDE is correct away from the facet, but
- Limit of step flow does not equal steepest-descent solution

Should we be surprised? No. Singular perturbations induce bndry layers. Finite step-height is a singular perturbation.



Can we resolve the boundary layer at the facet edge?

Coarse-graining is more interesting in 2D

Focus has been on 1D setting. Was that a good idea?

In favor: Must crawl before learning to walk. Also: approx 1D ripples are easy to achieve experimentally.

Against: 1D picture may be non-physical. Possibly unstable due to 2D fluctuations of steps bounding facet. Also, if the avg slope isn't exactly 0, the ridges won't be flat.

Radial case is more physical. Coarse-graining issues are same as 1D setting. No self-similar asymptotics (yet).

Coarse-graining is richer in 2D (Kohn-Margetis, MMS, 2006)

- PDE is 4th order, but anisotropic (not an obvious extension of 1D or radial case)
- Source of anisotropy: directions $\parallel$ and $\perp$ to steps are very different.

Conclusions

Steepest descent PDE model:

- interesting, nonlinear PDE
- numerics via implicit time-stepping
- self-similar decay in 1D with “sinusoidal” data

Relaxation of crystal surfaces:

- continuum modeling still poorly understood
- coarse-graining of step flow model is a current challenge
- does finite step-size induce “boundary layer” at facet edge?

Some references

1D finite difference numerics, and self-similar asymptotics: I. Odisharia, PhD Thesis, NYU 2006; R. Kohn, I. Odisharia, in preparation

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