Home Page

Title Page

Page 1 of 100

Go Back

Full Screen

Close

Quit

Analytical and Computational Modeling of the Stability and Dynamics of a Dewetting Ultrathin Solid Film

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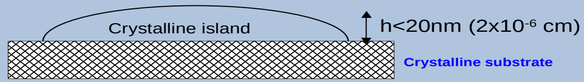
Outline:

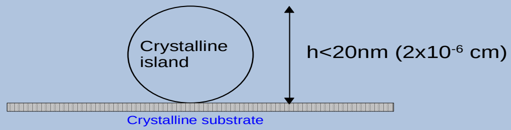
- Motivation
- Physical factors
- Mathematical model
- Stability analysis
- Numerical method
- Computational results
- Directions for future work

Graduate Student Seminar, February 5, 2010

What is “wetting” ?



``Wetting" island (solid 1 on substrate 1):
likes the substrate, tries to spread and maximize the contact area



``Non-wetting" island (solid 1 on substrate 2):
hates the substrate, tries to contract and minimize the contact area

Dewetting = dynamical transition from “wetting” to “non-wetting”

Motivations

Non-mathematical:

For technological applications, the structural integrity of thin solid films must be maintained in tough environments such as high temperatures and stress levels

Increased understanding of film dynamics is needed in order to develop reliable methods for controlling film *morphology* (= shape)

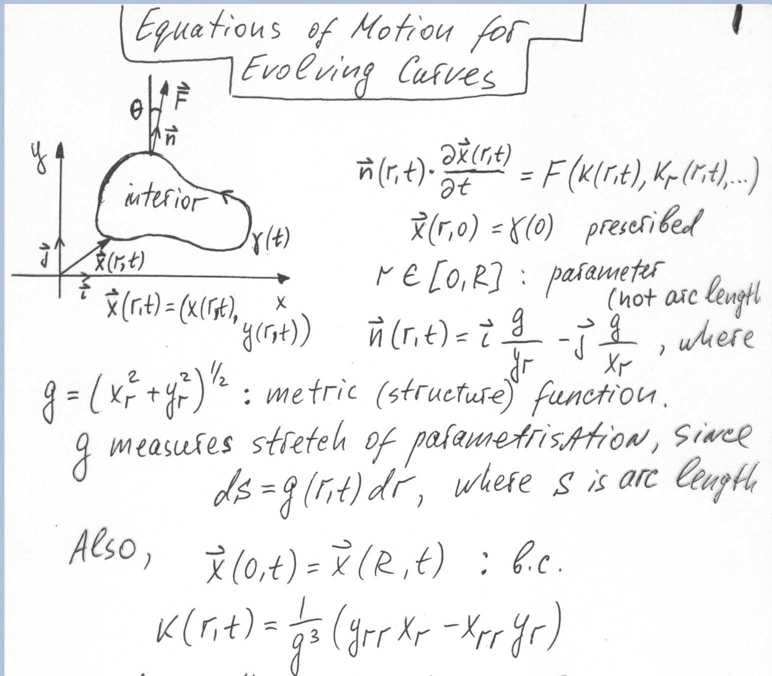
Mathematical:

Models of thin films are often formulated in terms of high-order, heavily nonlinear "geometric" evolution PDEs that have some unusual properties, such as unstable and non-differentiable solutions. This calls for nonstandard methods of analysis and solution that are interesting in their own right.

A word of caution:

The field of geometric evolution PDEs is comprised of two subfields that rarely intersect: the theory of evolving curves and surfaces and modeling ("applied"). This work is on the "applied" side.

Next talk (someday): Introduction to curve/surface evolution problem


Equations of Motion for Evolving Curves

$$\vec{n}(r,t)\cdot\frac{\partial\vec{x}(r,t)}{\partial t} = F(\kappa(r,t), \kappa_r(r,t), \dots)$$

$\vec{x}(r,0) = \gamma(0)$ prescribed

$r \in [0,R]$: parameter (not arc length)

$\vec{n}(r,t) = \vec{i}\,\frac{g}{y_r} - \vec{j}\,\frac{g}{x_r}$, where

$g = (x_r^2 + y_r^2)^{1/2}$: metric (structure) function.

g measures stretch of parametrisation, since $ds = g(r,t)\,dr$, where s is arc length

Also, $\vec{x}(0,t) = \vec{x}(R,t)$: b.c.

$$\kappa(r,t) = \frac{1}{g^3}(y_{rr}x_r - x_{rr}y_r)$$

Key physical factors

- In high-temperature environment (~ 800 C) the driving force for film morphology changes is the *surface diffusion* of adsorbed atoms (= surface atoms that are weakly bonded to the interior of the film)
- Because film is very thin, its surface "feels" the substrate through the *long-range intermolecular attraction or repulsion*
- *Surface energy anisotropy* gives rise to surface faceting

These three factors influence the dynamics and outcomes of surface evolution

Impacts on film stability and integrity must be addressed

Our model: Interaction with the substrate is *attractive*. This is motivated by a real-life situation (next slide)

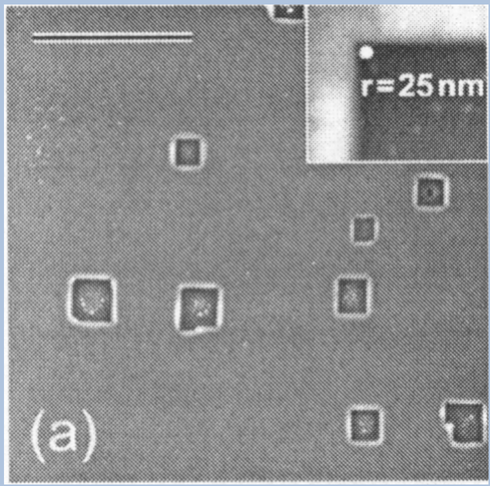
Dewetting experiment summary

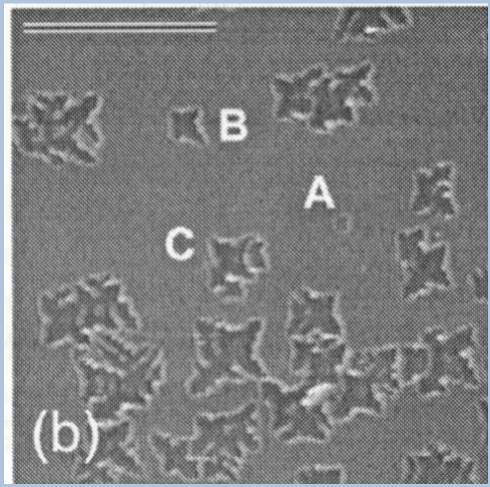
- After deposition, film surface features a finite number of randomly distributed pinhole defects ...
- Pinholes start to change shape and deepen (or contract) under the influence of the surface diffusion, attraction to the substrate, surface energy anisotropy, etc. ...
- Most often, the surface reaches the substrate and exposes it in the “dewetted windows” ...
- Windows expand until boundaries of neighboring windows meet and merge; ultimately, a large area of the film ceases to exist

The experiment

Sutter et al., Appl. Phys. Lett. 88, 141924 (2006)

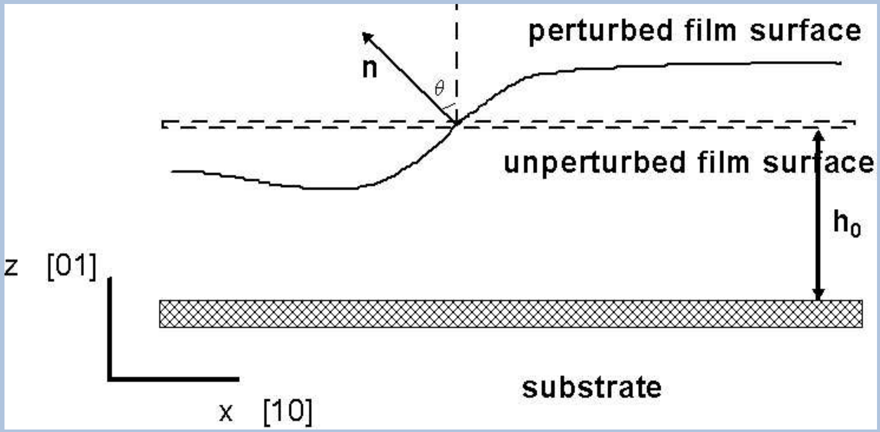
10 nm Si film on SiO_2 (quartz): deposited, heated to 800 C, held at this T for 2 min (left image), > 2 min (right image). Substrate is exposed in windows.





Our model is currently capable of describing the destabilization of the planar surface (= state of equilibrium) and the evolution of the surface morphology until the film height vanishes at the bottom of the pinhole, or the surface shape returns to planar state. i.e. the expansion and mergers of dewetted "windows" are not treated

The model is rooted in the classical theory of morphological evolution by surface diffusion (Mullins-Herring theory, developed in 1950s))



Mullins-Herring theory

Assume surface energy $\gamma =$ const. independent of the surface orintation θ: $\gamma =$ const.

$$\mathbf{z} = \mathbf{h}(\mathbf{x}, \mathbf{t}), \quad \mathbf{h_t} \cos\theta = \mathbf{h_t} \left(\mathbf{1} + \mathbf{h_x^2}\right)^{-1/2} = \mathbf{V} = -\boldsymbol{\Omega}\mathbf{j_s}, \quad \frac{\partial}{\partial \mathbf{s}} = \cos\theta \frac{\partial}{\partial \mathbf{x}}$$

$$\mathbf{j} = -\frac{\mathbf{D}\nu}{\mathbf{kT}}\mu_{\mathbf{s}}, \quad \mu \equiv \mu^{(\kappa)} = \boldsymbol{\Omega}\gamma\kappa, \quad \kappa = \frac{-\mathbf{h_{xx}}}{(\mathbf{1} + \mathbf{h_x^2})^{3/2}}$$

$$\text{Thus } \mathbf{h_t} = \mathbf{B}\frac{\partial}{\partial \mathbf{x}}\left((\mathbf{1} + \mathbf{h_x^2})^{-1/2}\left[\frac{-\mathbf{h_{xx}}}{(\mathbf{1} + \mathbf{h_x^2})^{3/2}}\right]_{\mathbf{x}}\right), \quad \mathbf{B} = \frac{\boldsymbol{\Omega}^2\nu\mathbf{D}\gamma}{\mathbf{kT}}$$

Next, make the PDE dimensionless →$h_0 = 1$

Linear Stability Analysis::

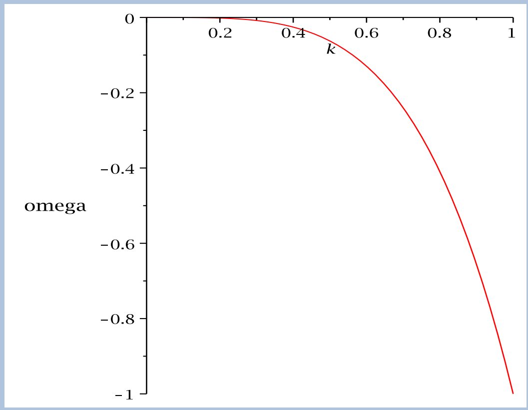
$$\mathbf{h = 1 + \xi(x,t) \quad \rightarrow \quad \xi_t = -B\xi_{xxxx}}$$

Let

$$\mathbf{\xi(x,t) = e^{ikx+\omega t}}$$

Solution is shape-preserving,

$$\mathbf{h(x,t) = 1 + \exp\left(-Bk^4 t\right)\cos kx, \quad \omega(k) = -Bk^4}$$



Dispersion curve $\omega(k)$

We add surface energy anisotropy, wetting interactions to M-H theory

Two-layer exponential wetting model:

$$\gamma(\mathbf{h},\theta) = \gamma^{(\mathbf{f})}(\theta) + \left(\gamma_{\mathbf{S}} - \gamma^{(\mathbf{f})}(\theta)\right)\exp\left(-\mathbf{h}/\ell\right)$$

$$\gamma(\mathbf{h},\theta) \to \gamma^{(\mathbf{f})}(\theta) \;\; \textbf{as} \quad \mathbf{h}/\ell \to \infty, \qquad \gamma(\mathbf{h},\theta) \to \gamma_{\mathbf{S}} \;\; \textbf{as} \quad \mathbf{h}/\ell \to \mathbf{0}$$

$$\gamma^{(\mathbf{f})}(\theta) = \gamma_{\mathbf{0}}(\mathbf{1} + \epsilon\cos 4\theta) + \frac{\delta}{\mathbf{2}}\kappa^{\mathbf{2}} \equiv \gamma_{\mathbf{p}}(\theta) + \frac{\delta}{\mathbf{2}}\kappa^{\mathbf{2}}$$

$(\delta/\mathbf{2})\kappa^{\mathbf{2}}$, $\delta \ll \mathbf{1}$: **regularization to preserve well-posedness of the evolution PDE for strong anisotropy,** $\epsilon > \mathbf{1}/\mathbf{15}$ **(*see works by, say, Angenent & Gurtin, Golovin, Spencer*)**

$$\mu = \mu^{(\kappa)} + \mu^{(\mathbf{w})}$$

$$\mu^{(\kappa)} = \left(\gamma_{\mathbf{p}}(\theta) + \frac{\partial^{\mathbf{2}}\gamma_{\mathbf{p}}}{\partial\theta^{\mathbf{2}}}\right)\left(\mathbf{1} - \exp\left(-\mathbf{h}/\mathbf{r}\right)\right)\kappa + \mathbf{\Gamma}\exp\left(-\mathbf{h}/\mathbf{r}\right)\kappa - \mathbf{\Delta}\left(\frac{\kappa^{\mathbf{3}}}{\mathbf{2}} + \kappa_{\mathbf{ss}}\right)$$

$$\mu^{(\mathbf{w})} = \mathbf{\Omega}\frac{\partial\gamma(\mathbf{h},\theta)}{\partial\mathbf{h}}\cos\theta = \left(\gamma_p(\theta) - \Gamma\right)\frac{\exp\left(-h/r\right)}{r}\cos\theta$$

Here $\mathbf{\Gamma} = \gamma_{\mathbf{S}}/\gamma_{\mathbf{0}}$, $\mathbf{r} = \ell/\mathbf{h_0}$ **and** $\mathbf{\Delta} = \delta/(\gamma_{\mathbf{0}}\mathbf{h}_{\mathbf{0}}^{\mathbf{2}})$

Typically, $\mathbf{\Delta} \ll \mathbf{1}$, $\mathbf{r} < \mathbf{1}$ **and** $\mathbf{\Gamma} < \mathbf{1}$ **(non-wetting)**

Now, the new evolution equation (6th-order) is:

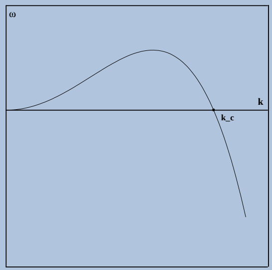
$$h_t = B(1+h_x^2)^{1/2}\frac{\partial^2}{\partial s^2}\left(\mu^{(\kappa)}+\mu^{(w)}\right), \qquad \frac{\partial}{\partial s} = (1+h_x^2)^{-1/2}\frac{\partial}{\partial x}$$

Linear Stability Analysis: $h = 1 + \xi(x,t)$:

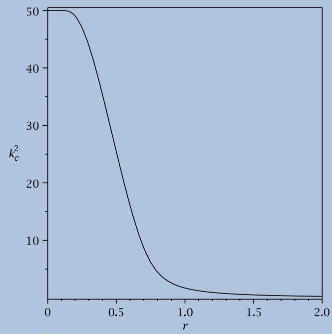
$$\xi_t = B\left(\Lambda\xi_{xxxx} + \Delta\xi_{xxxxxx} + \exp(-1/r)\left[r^{-2}g\xi_{xx} - (\Gamma-1)\xi_{xxxx}\right]\right)$$

Taking $\xi = e^{ikx+\omega t}$ gives

$$\omega(k) = B\left[\left(\Lambda - \exp(-1/r)(\Gamma+\Lambda)\right)k^4 - \Delta k^6 - \exp(-1/r)r^{-2}gk^2\right]$$



Dispersion curve $\omega(k)$ for $\Lambda = 15\epsilon - 1 > 0$, $g = \Gamma - 1 - \epsilon < 0$, $\Delta > 0$



k_c^2 vs. r

If $\Delta = 0$ when $\Lambda > 0$:
$\omega(k) \geq 0 \quad \forall k \quad \rightarrow$ absolute instability

From linear stability analysis of small deviations from planar equilibrium $h = 1$ to understanding the nonlinear dynamics of large deviations:

Computations of the pinhole dynamics using the nonlinear PDE

The initial condition:

$$\mathbf{h}(\mathbf{x}, \mathbf{0}) = \mathbf{1} - \mathbf{d} \exp\left[-\left(\frac{\mathbf{x}-\mathbf{5}}{\mathbf{w}}\right)^{\mathbf{2}}\right], \quad \mathbf{0} \leq \mathbf{x} \leq \mathbf{10}$$

$$\mathbf{d} = \mathbf{0.5} \ \text{(shallow)}$$

$$\mathbf{w} = \mathbf{0.15} \ \text{(narrow), or}$$

$$\mathbf{w} = \mathbf{1} \ \text{(intermediate), or}$$

$$\mathbf{w} = \mathbf{2} \ \text{(wide)}$$

Periodic b.c.'s at $\mathbf{x} = \mathbf{0}$ and $\mathbf{x} = \mathbf{10}$

$$\boldsymbol{\Gamma} = \mathbf{0.5}, \ \epsilon = \mathbf{1/12}, \ \boldsymbol{\Delta} = \mathbf{0.005}, \ \mathbf{B} = \mathbf{3.57} \times \mathbf{10^{-3}}, \ \mathbf{r} = \mathbf{0.1}, \mathbf{0.02}$$

Numerical method

Non-graph (overhanging) surfaces are possible for large deviations from equilibrium → evolution PDE is re-written in the parametric form for the coordinates $(\mathbf{x}, \mathbf{z})$ of a point on a surface (*a marker particle*):

$$\mathbf{x_t} = V \frac{z_u}{g} \quad (1)$$

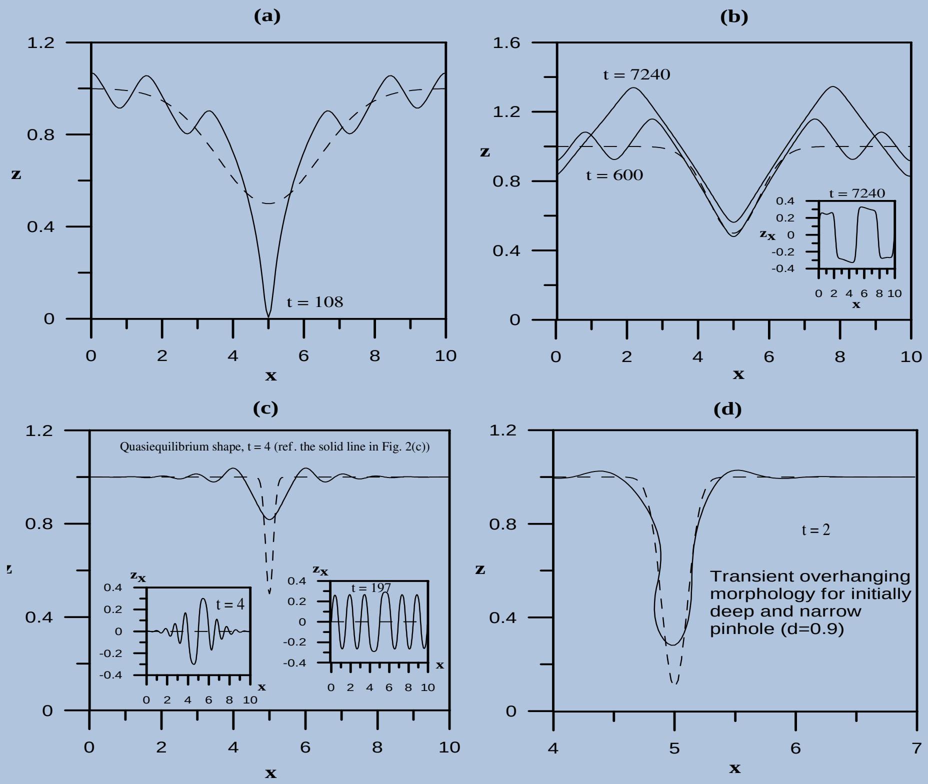
$$\mathbf{z_t} = -V \frac{x_u}{g}, \quad 0 \le u \le U : \textbf{the parameter} \quad (2)$$

$$\mathbf{V} = \mathbf{B}\left(\mu_{\mathbf{ss}}^{(\kappa)} + \mu_{\mathbf{ss}}^{(\mathbf{w})}\right), \quad \mathbf{g} = \sqrt{\mathbf{x_u^2} + \mathbf{z_u^2}}, \quad \frac{\partial}{\partial \mathbf{s}} = \frac{\mathbf{1}}{\mathbf{g}} \frac{\partial}{\partial \mathbf{u}}$$

Numerical method of lines:

- **PDE is discretized in u using the 2nd-order finite differences, time is left continuous → large system of coupled ODEs in time**
- **Off-the-shelf ODE solver (IRK-RADAU by Hairer & Wanner)**
- **To maintain uniform accuracy, need to remesh periodically to keep marker particles optimally spaced**

RESULTS: Morphologies I



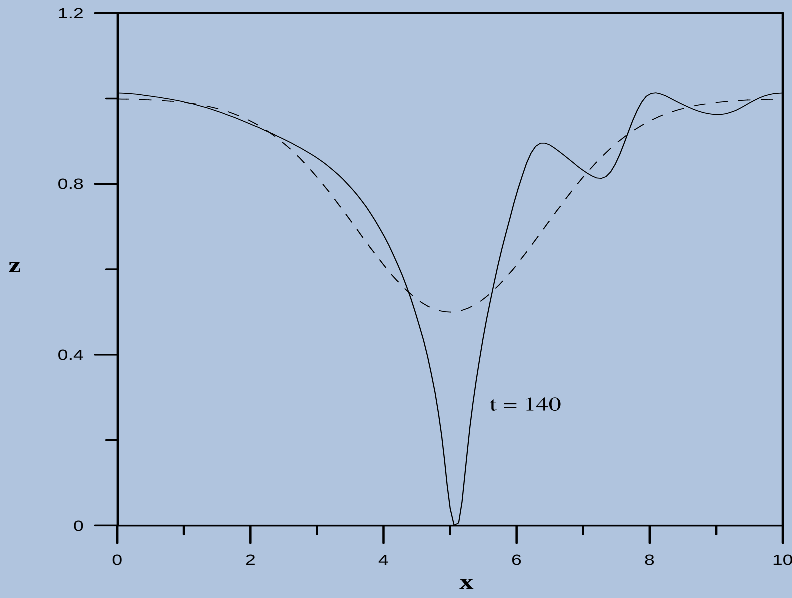
(a): w = 2, (b): w = 1, (c): w = 0.15

Quasiequilibrium: (b), (c). Tip is stationary, but surface structure coarsens in time.

Home Page
Title Page
Page 15 of 100
Go Back
Full Screen
Close
Quit

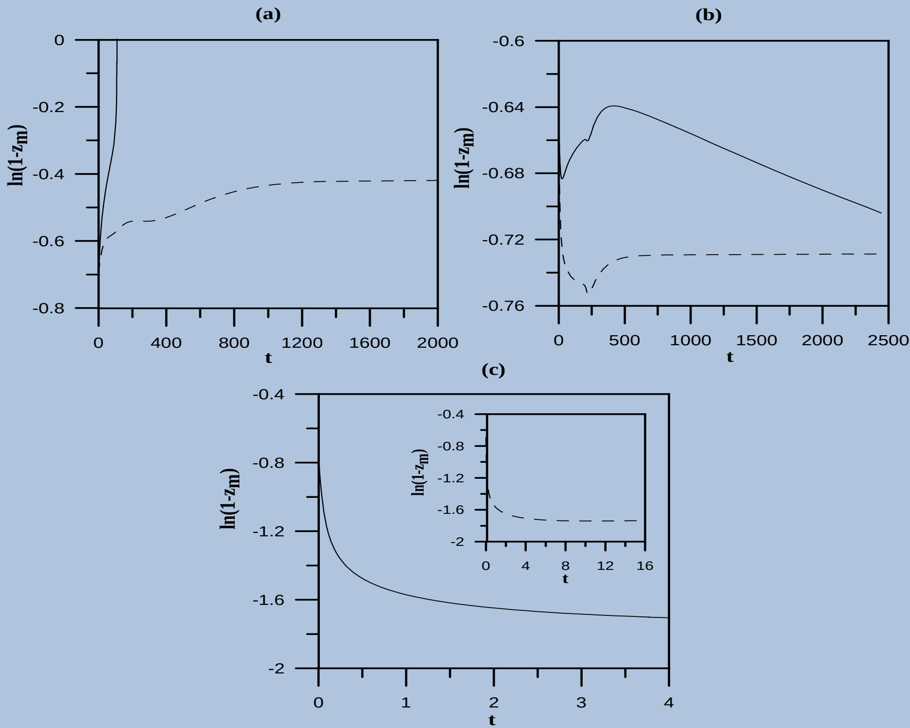
RESULTS: Morphologies II

Rotate sample, expose different crystallographic orientation: $\gamma^{(\mathrm{f})}(\theta) = \gamma_0(1 + \epsilon \cos 4(\theta + \beta))$



$\mathbf{w = 2,\ \beta = 10^\circ}$

RESULTS: Kinetics



(a): $w = 2$, (b): $w = 1$, (c): $w = 0.15$

Solid line: $r = 0.1$, dashed line: $r = 0.02$

Pinhole does not dewet in all cases except $w = 2$, $r = 0.1$. Instead, quasiequilibrium.

Directions for future work

- Weakly nonlinear analysis near instability threshold to study the bifurcation into a nonlinear pattern
- Direct analysis of steady-states (quasiequilibrium)
- Development of high-order adaptive methods (FD and/or spectral) for very accurate computation of film faceting, thinning and rupture
- 2D modeling (highly nontrivial in the presence of surface energy anisotropy)
- Inclusion of surface/epitaxial stress
- Inclusion of short-range repulsive interaction with the substrate
- Inclusion of the “external” stabilizing mechanisms, such as the electric fields
- Analysis of the influence of substrate patterning
- Modeling of multilayer films

SUMMARY

- Developed PDE-based model for high-T thin solid film dewetting
- Carried out linear stability analysis and computations in the strongly nonlinear regimes
- Determined parameter windows corresponding to film dewetting
- Demonstrated nontrivial surface morphologies

Publications:

M. Khenner, *Phys. Rev. B* 77, 245445 (2008)
M. Khenner, *Phys. Rev. B* 77, 165414 (2008)
M. Khenner, *Mathematical Modeling of Natural Phenomena* 3(5), 16-29 (2008)
M. Khenner, in preparation

THE END THANKS !

AFTERWORD

Other models of wetting interactions have been proposed, for instance:

Two-layer algebraic model

$$\gamma(\mathbf{h},\theta) = \frac{1}{2}\left(\gamma^{(\mathbf{f})}(\theta) + \gamma_{\mathbf{S}}\right) + \frac{1}{2}\left(\gamma^{(\mathbf{f})}(\theta) - \gamma_{\mathbf{S}}\right)\mathbf{f}(\mathbf{h}/\ell)$$

$$\lim_{\mathbf{h}\to\infty}\mathbf{f}(\mathbf{h}/\ell) = 1, \qquad \lim_{\mathbf{h}\to-\infty}\mathbf{f}(\mathbf{h}/\ell) = -1$$

Choose

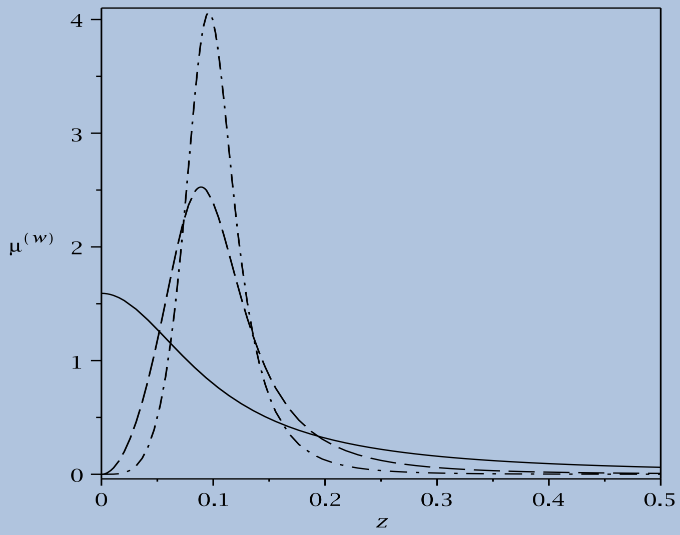
$$\mathbf{f}(\mathbf{h}/\ell) = \frac{2}{\pi}\mathbf{arctan}\left[\left(\frac{\mathbf{h}}{\ell}\right)^{\mathbf{m}}\right], \qquad \mathbf{m} = 1, 3, 5, \ldots$$

Note that

$$\mathbf{f}(\mathbf{h}/\ell) = 1 - \frac{2}{\pi}(\mathbf{h}/\ell)^{-\mathbf{m}} + \ldots \quad \text{as} \ \ \mathbf{h}\to\infty.$$

i.e., this tends to the limiting value +1 as an algebraic power.

Chemical potential:



$$\mu^{(\mathbf{w})} \sim \mathbf{1/h^{m+1}} \textbf{ for } \mathbf{h \gg 1}$$

- **Increased stability of the equilibrium h = 1**
- **Different kinetics and dewetting outcomes for different values of m (only wide and deep pinholes dewet for the same set of parameters, and only for m = 1)**