Capillary Instabilities on Thin Solid Films

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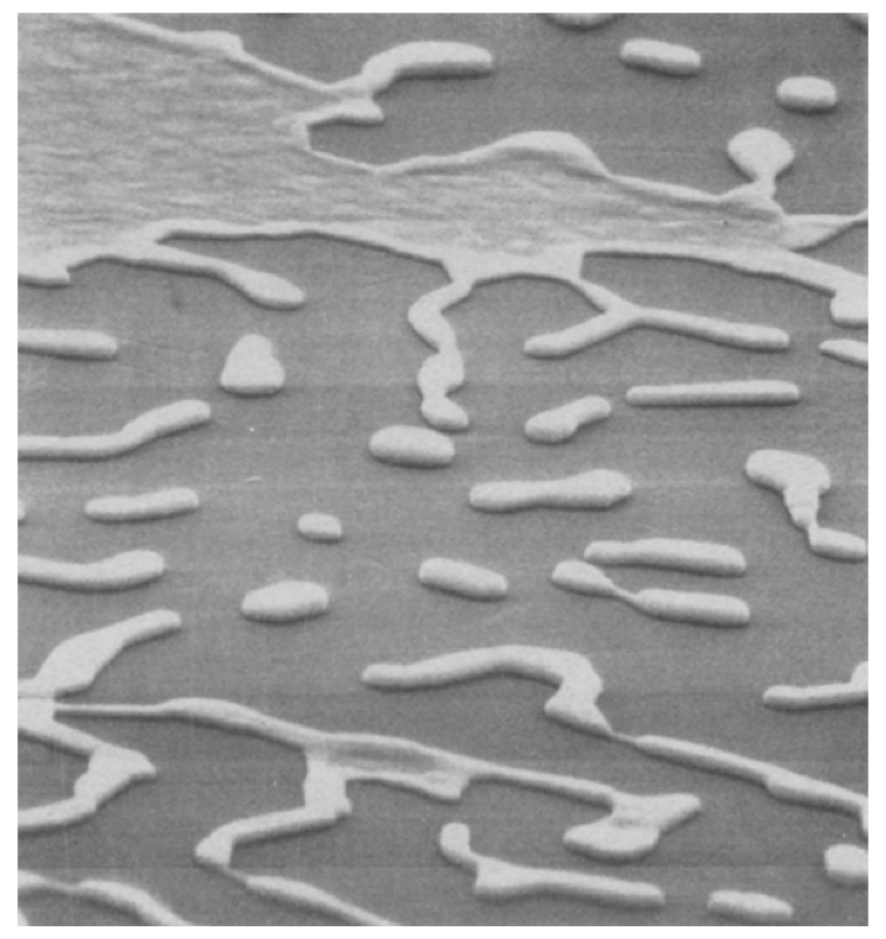
Introduction

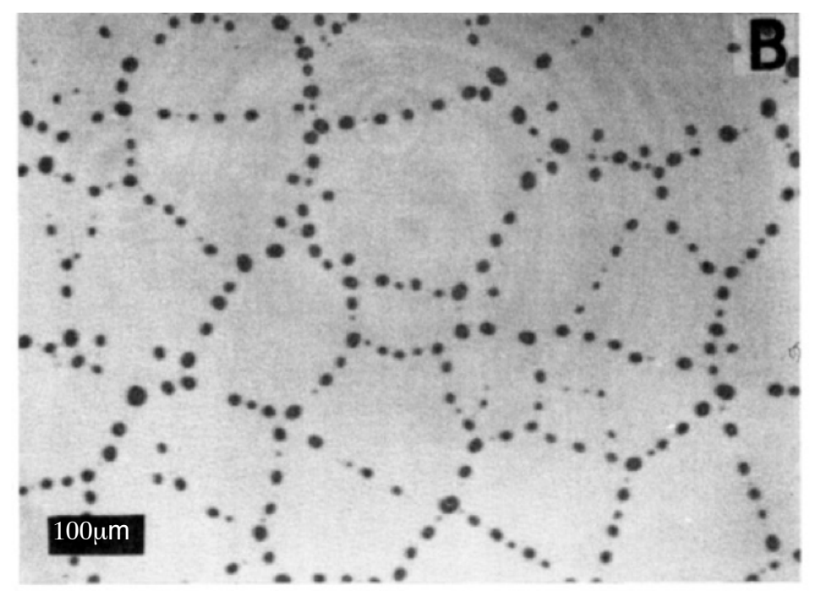
Thin films are the fundamental building blocks for many technologies, including electronic, electrochemical, and optical devices. However, they are unstable against capillary forces. Capillarity (surface tension) drives a process in thin films known as solid-state dewetting: the edges retract, holes can form, and the film breaks up into islands of material with characteristic microstructural features. This process occurs below the film's melting point via capillary-driven surface self-diffusion. Understanding dewetting is critical for the prevention of thermal degradation in micro- and nano-scale systems. For anisotropic materials, it also offers a new avenue for the manufacture of stable, complex, small-scale geometries. We present a variety of theoretical and computational studies to understand and control dewetting microstructures.

Dewetting phenomena

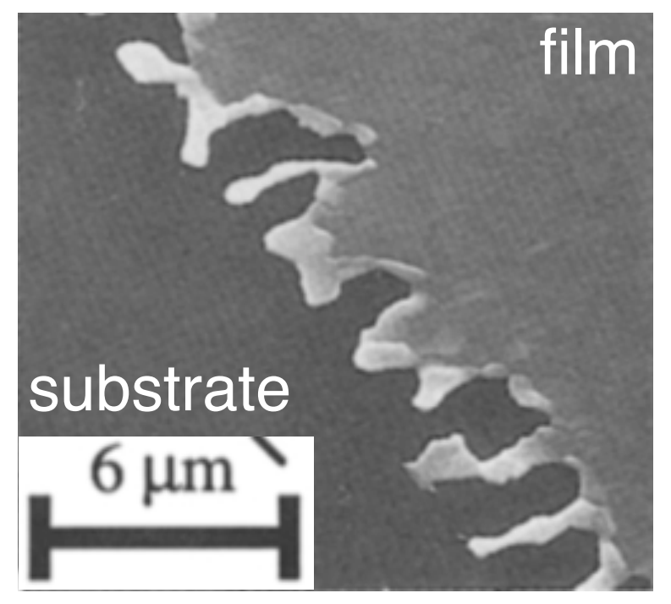


Jiran and Thompson, *J. Elec. Mater.* 19, 1990

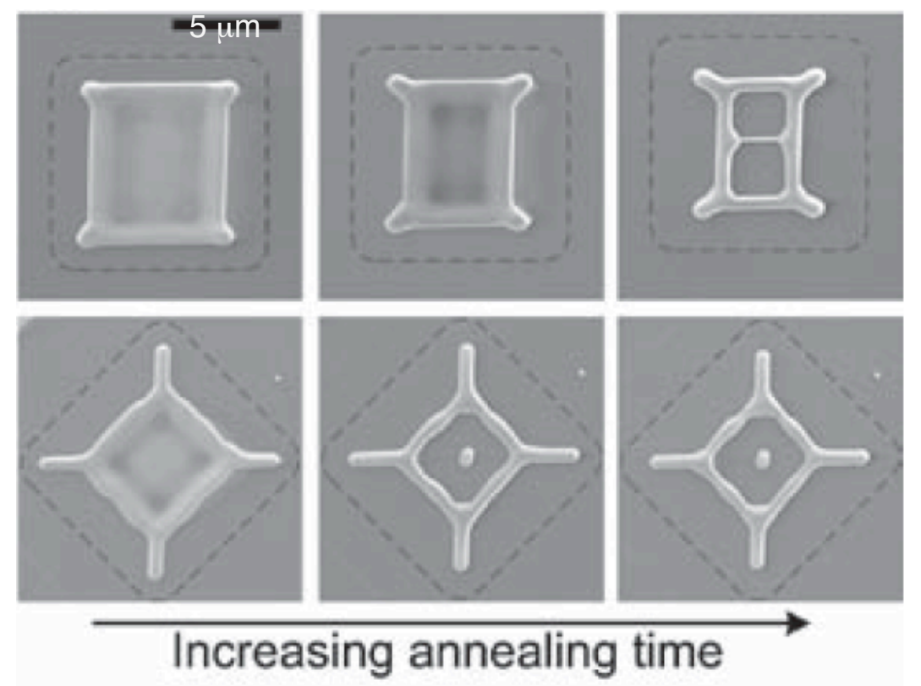
Above: Typical dewetting morphology. Polycrystalline Ag on SiN.



Reiter, *Langmuir,* **9**, 1993

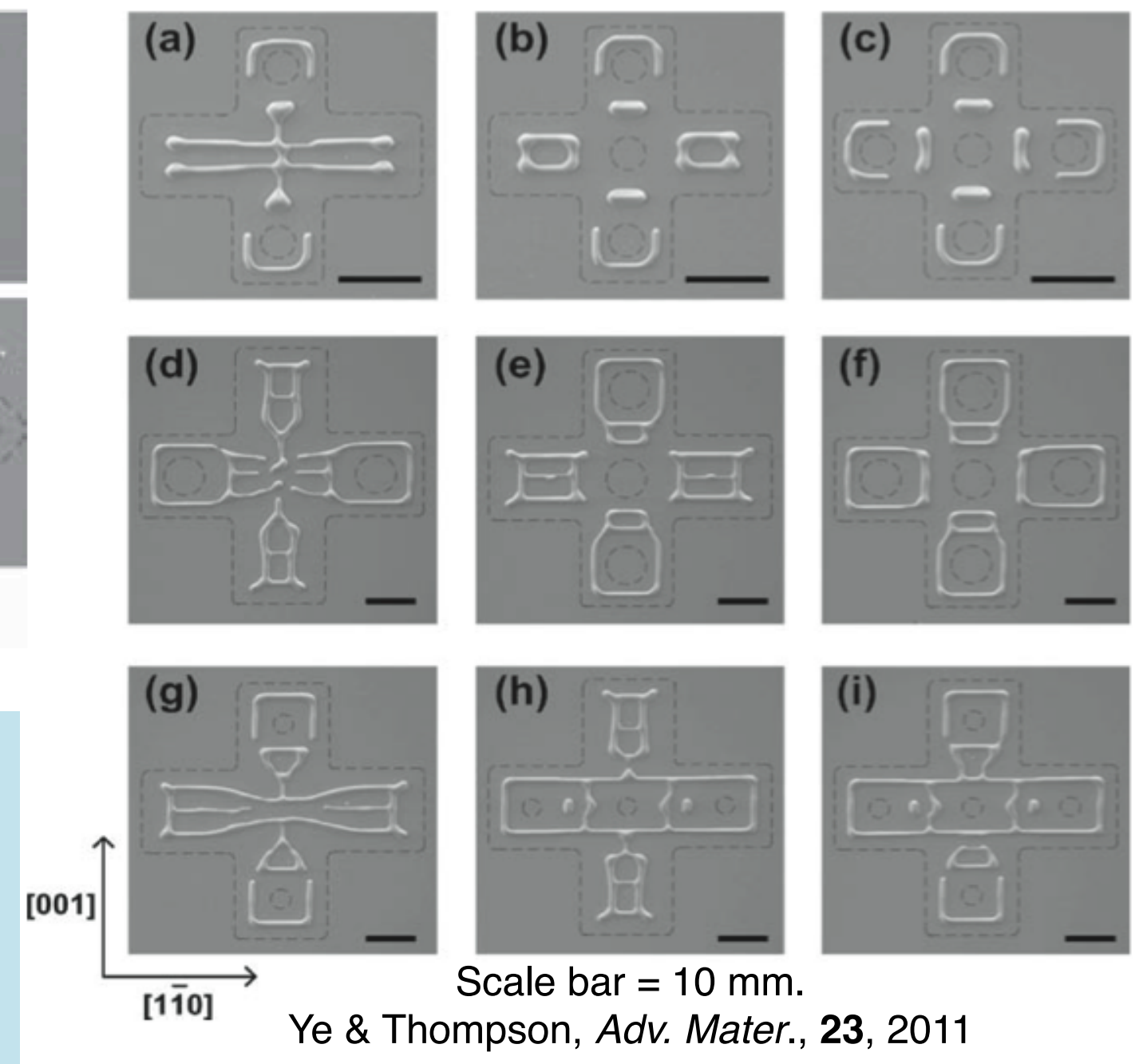


Jiran & Thompson, *Thin Solid Films* 208, 1992



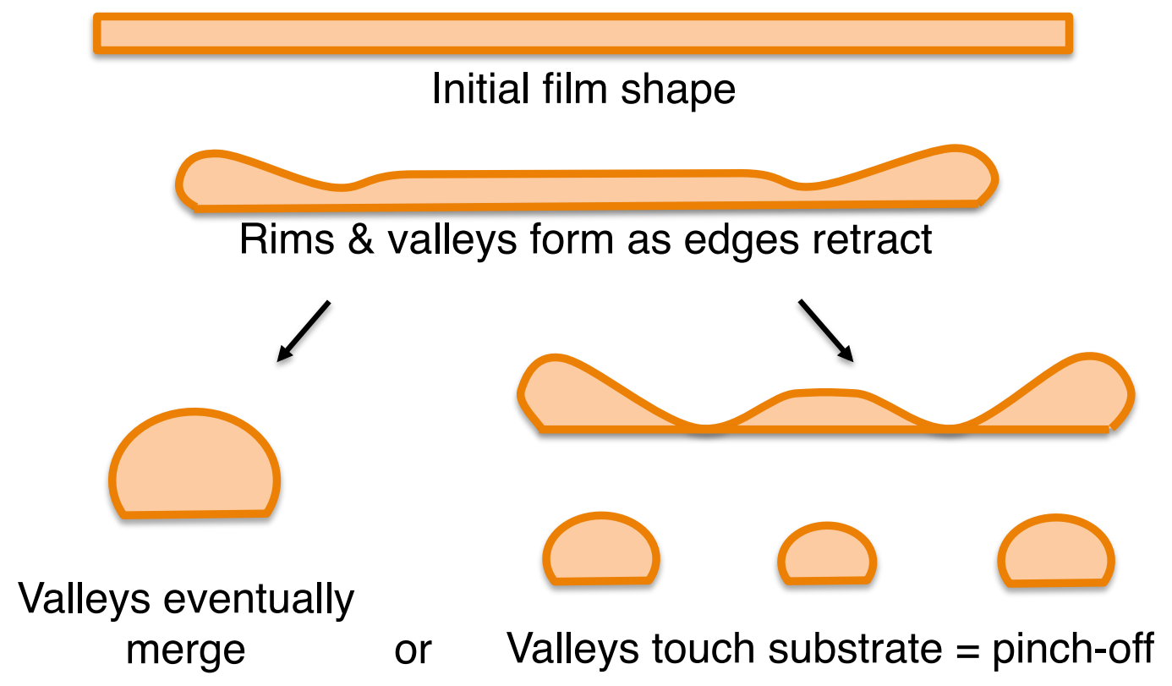
Ye & Thompson, *Adv. Mater.*, **23**, 2011

Above left: after holes form and grow, the thin film material (dark) remaining breaks up by a Rayleigh instability.
Above right: edge retraction and valley formation leads to holes in single crystal films.
Left: The edge of a film undergoes a "fingering instability."

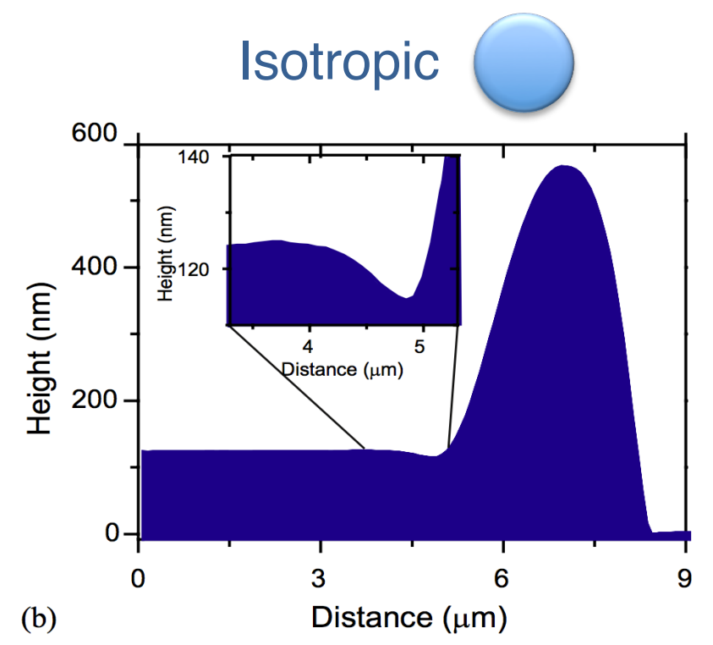


Scale bar = 10 mm.
Ye & Thompson, *Adv. Mater.*, **23**, 2011

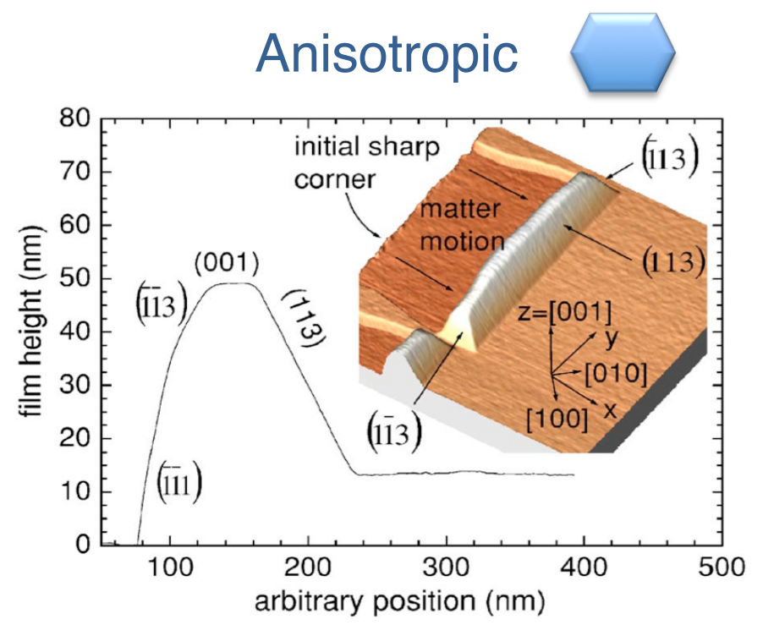
Above: Complex, reproducible morphologies obtained by anisotropic dewetting.



Above: A schematic of an isotropic thin film undergoing dewetting.

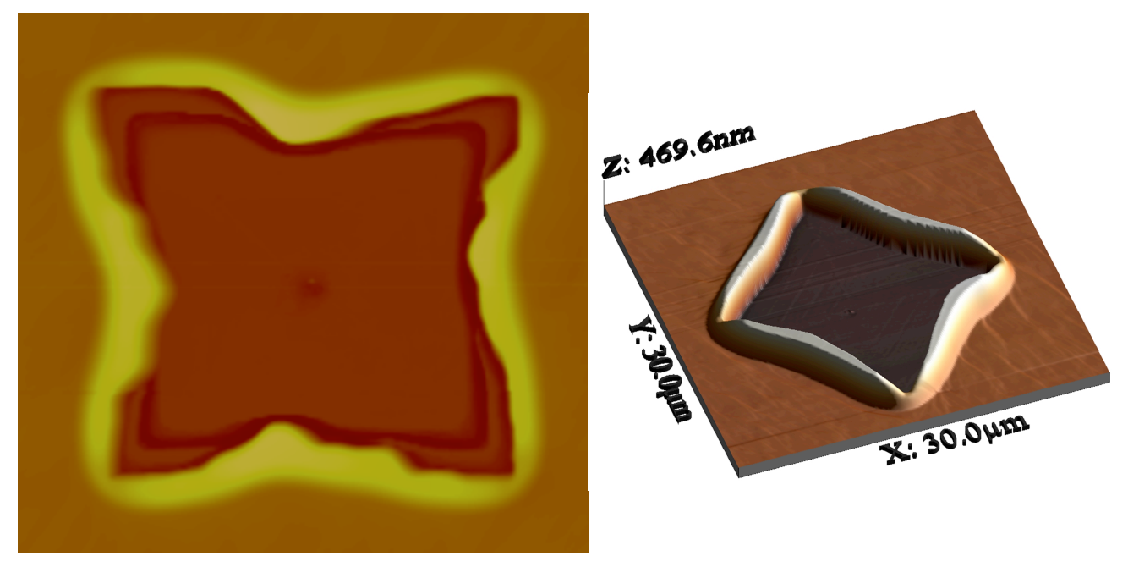


Neto & Jacobs, *Phys. A*, **339**, 2004



Dornel *et al.*, *Phys. Rev. B*, **73**, 2006

Above: Edge profiles for dewetting films. Fully-faceted films do not have a valley, isotropic films do.



Ye and Thompson, *unpublished*

Above: an initially square hole in a single crystal film undergoes a "corner instability."

Governing equations

The chemical potential μ is proportional to the mean curvature κ of the film surface:

$$\mu(s_1, s_2) = \gamma\Omega\kappa(s_1, s_2)$$

where s_1 and s_2 are the arc length coordinates, γ is the surface energy, and Ω is the volume per atom. Using the Nernst-Einstein relation and Fick's law, the non-dimensional velocity of a surface along its normal is

$$v_n = -\Delta_s\kappa(s_1, s_2)$$

where Δ_s is the Laplace-Beltrami operator (i.e., the surface Laplacian). This can be generalized for anisotropic materials.

Mullins, *J. Appl. Phys.*, **28**, 1957

The non-dimensionalized phase field formulation for dewetting is

$$\frac{\partial\phi}{\partial t} = \nabla\cdot((1-\phi^2)\nabla\mu)$$

$$\mu = \phi^3 - \phi - \epsilon^2\Delta\phi$$

where ϕ is the phase field variable, t is time, μ is the chemical potential, and ε is related to the interface width. This is subject to the boundary condition

$$\epsilon\frac{\partial\phi}{\partial\mathbf{n}} + \frac{\sqrt{2}}{2}(\phi^2-1)\cos\theta = 0$$

where **n** is normal to the boundary and θ is the contact angle of the film on the substrate. The periodic boundary condition

$$\frac{\partial\phi}{\partial\mathbf{n}} = 0$$

applies on all other boundaries, and the mass conservation condition

$$\frac{\partial\mu}{\partial\mathbf{n}} = 0$$

applies on all boundaries. This formulation can also be generalized to anisotropic materials.

Jiang *et al.*, *Acta Mat.*, **60**, 2012

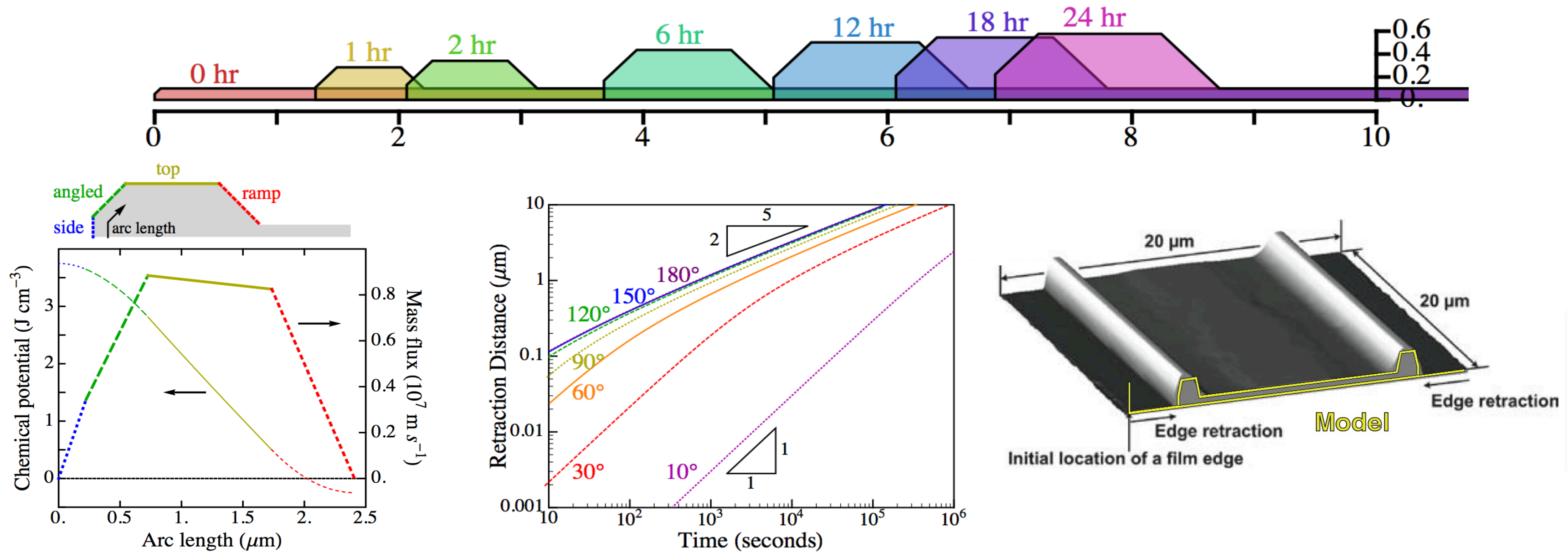
2D highly anisotropic model

Questions: How does strong anisotropy affect edge retraction? Which parameters dominate dewetting behavior?

Method: Explicit interface tracking, material is faceted, analytical in space, explicit forward differencing in time

Results: Consistent with experiments, no valleys are present, predicts a small mass flux towards the rim

Zucker *et al.*, *C. R. Phys.* **14**, 2013
Kim *et al.*, *J. Appl. Phys.* **113**, 2013



Top: Retraction of an example film, similar to Ni at 900°C.
Left: Typical chemical potential and mass flux on the rim.
Center: The dependence of edge retraction distance on the contact angle.
Right: the model is in good quantitative agreement with experiments.

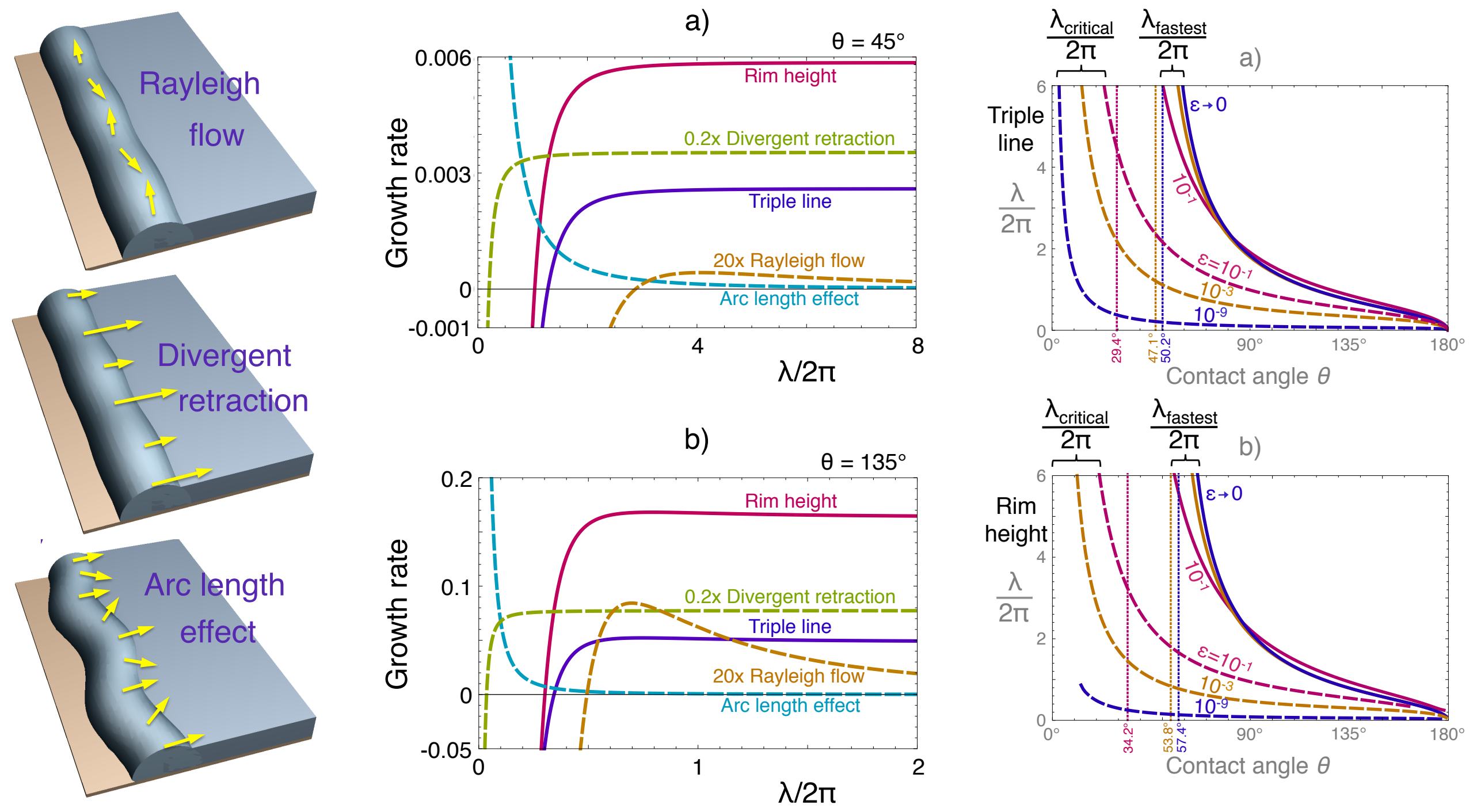
Fingering instability analysis

Questions: What drives the fingering instability? What determines the wavelength?

Method: Perturbation analysis for isotropic material, simplified geometry with a circular rim cross-section

Results: Three drivers of instability contribute: Rayleigh flow, the arc length effect, and divergent retraction. The latter dominates.

Zucker *et al.*, in preparation



Left: the three drivers of the fingering instability.

Center: perturbation growth rates for the three separate drivers, and for the net effect vertically (rim height) and horizontally (triple line position), for two different contact angles.

Right: The critical and fastest-growing perturbation wavelength as a function of perturbation amplitude ε.

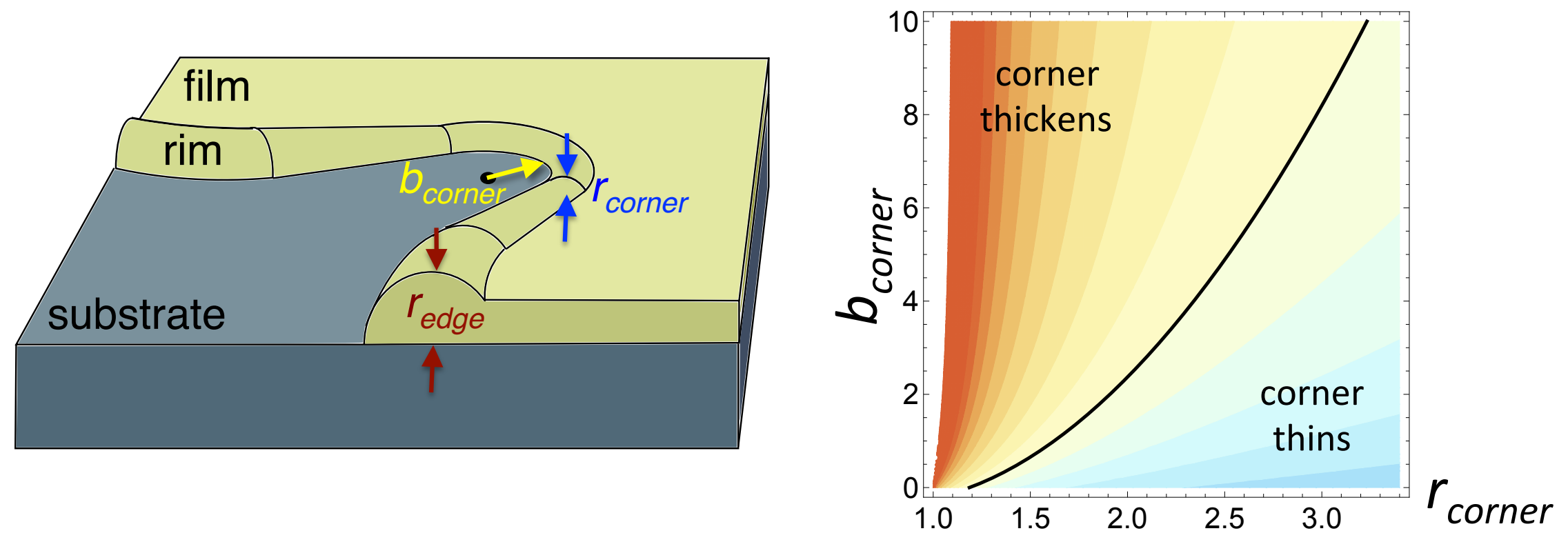
Corner instability analysis

Questions: What causes the corner instability?

Method: Perturbation analysis, anisotropy, simplified geometry

Results: Increasing arc length is the main driver, there is a minimum rim size for instability

Zucker *et al.*, in preparation



Left: schematic geometry, informed by experimental observations.

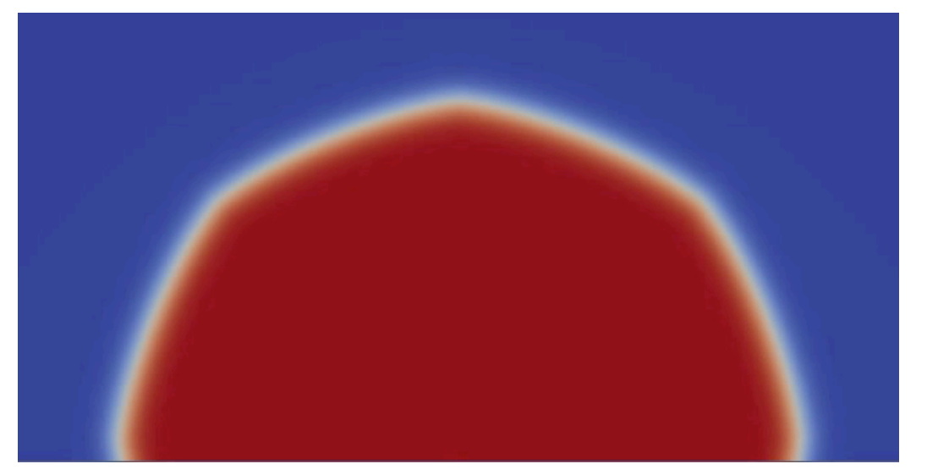
Right: the model predicts that the corner is driven towards a constant rim height and therefore retracts faster than the edges of the hole. This is in excellent agreement with experiments.

Phase field simluation

Questions: Can we simulate dewetting?

Method: AMDiS: finite element, adaptive-mesh, C++ library for PDEs: www.amdis-fem.org

Results: In progress. Verifying the analytical models, simulating the effects of anisotropy



We would like to thank Sebastian Reuther (TU Dresden), Roberto Bergamaschini, and Marco Salvalaglio (U of Milano) for their assistance with AMDiS.

Left: simulated films have all relevant features: rims, valleys, hole formation, pinch-off, and edge instabilities. Future work includes predicting dewetting microstructures.

Right: an anisotropic island of material produced as a result of dewetting in a phase field simulation.