Weierstrass Institute for
Applied Analysis and Stochastics

Leibniz Association

Motion of thin droplets over surfaces

numerical and modeling techniques for moving contact lines

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ICERM Workshop *„Making a Splash - Droplets, Jets and Other Singularities"*

Research Center MATHEON
Mathematics for Key Technologies

ECMath
Einstein Center
for Mathematics Berlin

Outline

1. Setting

2. Droplets on solid planar surfaces (solid substrates)

- *Modeling*
- *Contact angle: regularization vs free boundary*
- *Free boundary approach: numerical algorithm*
- *Examples*

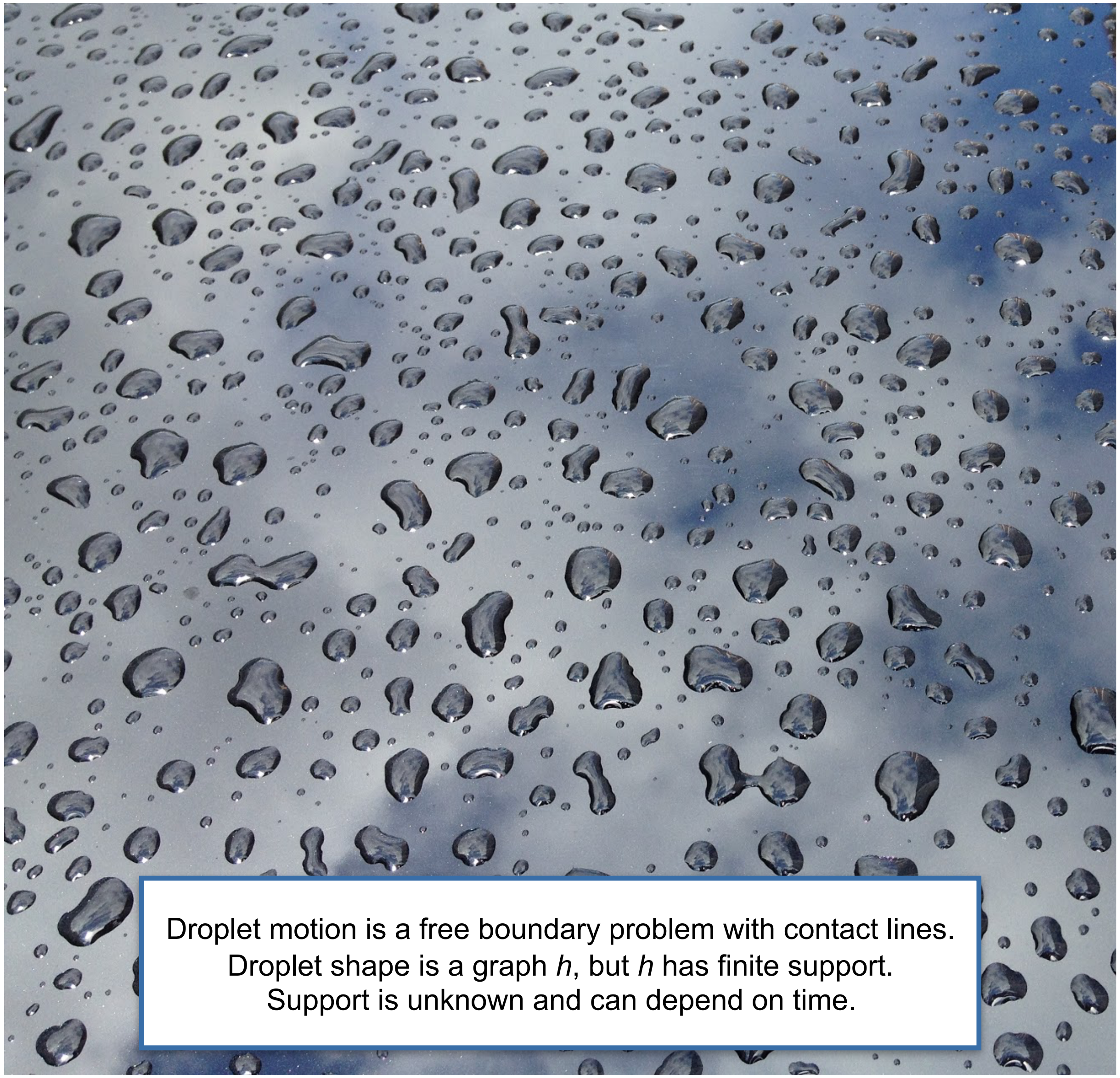
3. Flows over liquid substrates

(discuss extension of the method and compare with experiments)

1. Setting

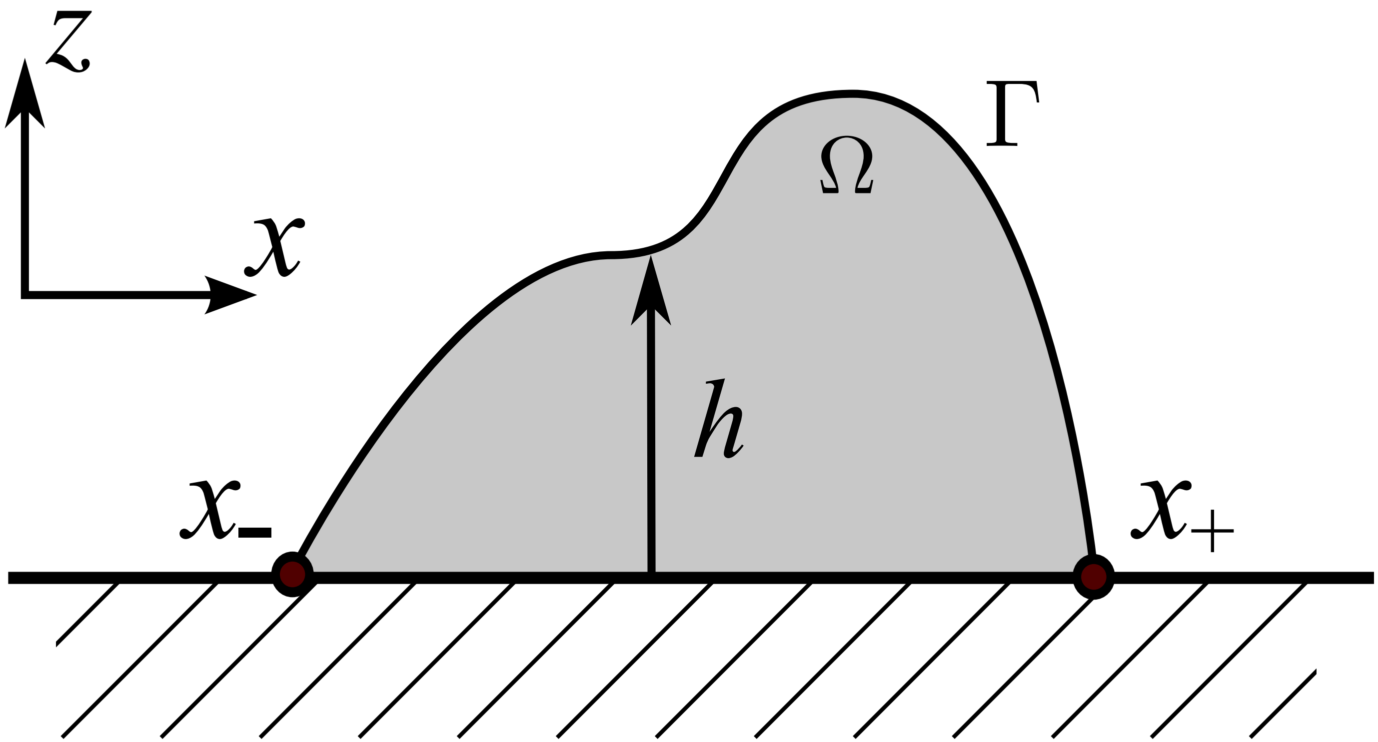
Setting

Setting:
Newtonian liquids
Viscous (Re=0)
partial wetting



Droplet motion is a free boundary problem with contact lines.
Droplet shape is a graph h, but h has finite support.
Support is unknown and can depend on time.

Setting



$$\Omega = \{(x,z) : 0 < z < h(t,x)\}$$

$$\Gamma = \{(x,z) : z = h(t,x) > 0\}$$

2. Droplets on solid planar surfaces

Modeling

Stokes flow with free boundary

$$\left.\begin{aligned} -\nabla p + \mu \nabla^2 \mathbf{u} &= \mathbf{0}, \\ \nabla \cdot \mathbf{u} &= 0, \end{aligned}\right. \qquad \text{in } \Omega$$

$$\left.\begin{aligned} \big(-p\mathbb{I} + 2\mu\mathbb{D}(\mathbf{u})\big)\mathbf{n} &= \sigma\kappa\mathbf{n}, \\ \big(\mathbf{u} - \mathbf{v}_\Gamma\big) \cdot \mathbf{n} &= 0, \end{aligned}\right. \qquad \text{on } \Gamma$$

$$|\nabla h| = \tan\theta, \qquad \text{at } x_\pm$$

Stokes via Helmholtz-Rayleigh variational principle

$$\text{Seek } \mathbf{u} \text{ so that } D(\mathbf{u},\mathbf{v}) = -\langle \operatorname{diff} E, \mathbf{v}\rangle \text{ for all } \mathbf{v}.$$

$$\frac{\mathrm{d}E}{\mathrm{d}t} = \langle \operatorname{diff} E, \mathbf{u}\rangle = -D(\mathbf{u},\mathbf{u}) \leq 0$$

$$D(\mathbf{u},\mathbf{v}) = \int_\Omega \frac{\mu}{2}\mathbb{D}(\mathbf{u}) : \mathbb{D}(\mathbf{v}) + \int_{\{z=0\}} \beta^{-1}\mathbf{u}\cdot\mathbf{v}$$

$$E = \int_\Gamma \sigma$$

Helmholtz (1869), Rayleigh (1873),…,Onsager (1929)

Thin-film limit in a nutshell:

$$D(\mathbf{u},\mathbf{v}) = -\langle \mathrm{diff}\, E, \mathbf{v}\rangle$$

$$D = \int \mathbb{D}(\mathbf{u}) : \mathbb{D}(\mathbf{v}) + \int_{z=0} \beta^{-1} uv$$

$$\approx \int \int_0^h (\partial_z u)(\partial_z v) \mathrm{d}z \, \mathrm{d}x + \int_{z=0} \beta^{-1} uv$$

$$= \int \int_0^h (-\partial_{zz} u) v \mathrm{d}z \, \mathrm{d}x + \int (\partial_z u) v \mathrm{d}x|_0^h + \beta^{-1} uv|_{z=0}$$

$$\langle \mathrm{diff}\, E, \mathbf{v}\rangle = \int \nabla h \nabla \dot{h}_v \mathrm{d}x$$

$$\int \sigma = \int \sigma \sqrt{1 + |\nabla h|^2}\, \mathrm{d}x \approx \int \sigma \left(1 + \tfrac{1}{2} |\nabla h|^2\right) \mathrm{d}x$$

$$= -\int \nabla h \cdot \nabla \left(\nabla \cdot \int_0^h v \mathrm{d}z \right) \mathrm{d}x$$

$$= \int \int_0^h (-\nabla \Delta h) v \mathrm{d}z \, \mathrm{d}x$$

Note: $\dot{h} = \partial_t h$

Conservation of mass + kinematic condition: $$\dot{h} + \nabla \cdot \int_0^h u \, \mathrm{d}z = 0$$

Modeling

For given $h(0,x)$ seek $h(t,x)$ such that

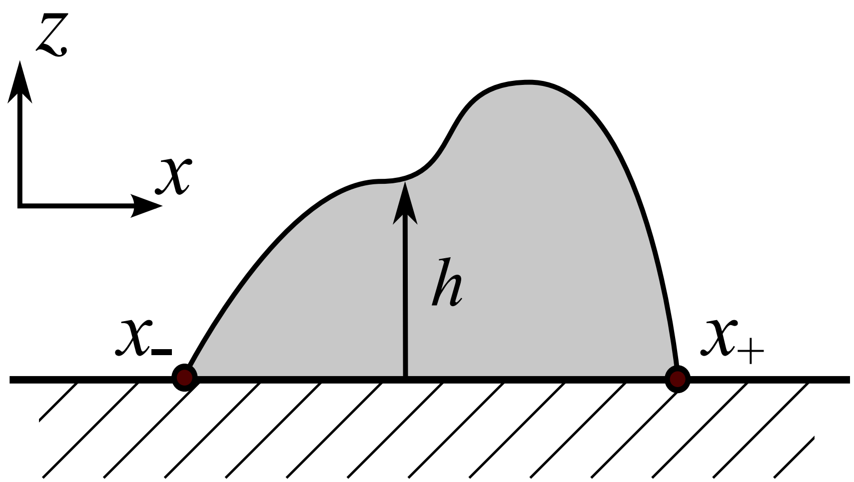
$$\dot{h} - \nabla \cdot \big(m(h)\nabla\pi\big) = 0$$

$$\pi = \frac{\delta E}{\delta h} = -\Delta h, \quad m(h) = |h|^n$$

where $h(t, x_\pm(t)) \equiv 0$

$$|h_x(t, x_\pm)| = \tan\theta$$

$$\dot{x}_\pm = \lim_{x \to x_\pm} \left(\frac{m}{h}\pi_x\right)$$



Existence weak solutions: Bernis & Friedmann (1990)
Positivitiy-preserving schemes: Zhornitskaya and Bertozzi (1999); Grün and Rumpf (2000)
Existence of classical solution in weighted Sobolev spaces: Giacomelli & Knüpfer (2010), Bertsch et al. (2005)

2. Droplets on solid planar surfaces

Contact angle: regularization vs free boundary

Driving energy E and dissipation D

$$E(h) = \int \tfrac{1}{2}|\nabla h|^2 \mathrm{d}x + V(h)$$

$$D(\dot h) = \int m(h)(\nabla \pi)^2$$

$m(h) = |h|^n$

Why is sliding motion singular for no-slip $m(h) = |h|^3$?

$$\dot h + \nabla \cdot \mathbf{j} = 0, \qquad \mathbf{j} = -m\nabla\pi = h\mathbf{v}$$

results in

$$D = \int \frac{\mathbf{j}^2}{m} = \int h^{2-n}|\mathbf{v}|^2$$

so that near a sliding contact line with velocity v_0 and slope α we have

$$D \approx \int_{x_-}^{x_-+\delta} \big(\alpha(x-x_-)\big)^{2-n} v_0^2 + \int_\delta^{x_+} \dots$$

Huh, Scriven. J. Colloid. Interf. Sci. (1971)

Driving energy E and dissipation D

$$E(h) = \int \tfrac{1}{2}|\nabla h|^2 \mathrm{d}x + V(h)$$

$$D(\dot{h}) = \int m(h)(\nabla\pi)^2$$

…with regularization (disjoining pressure)

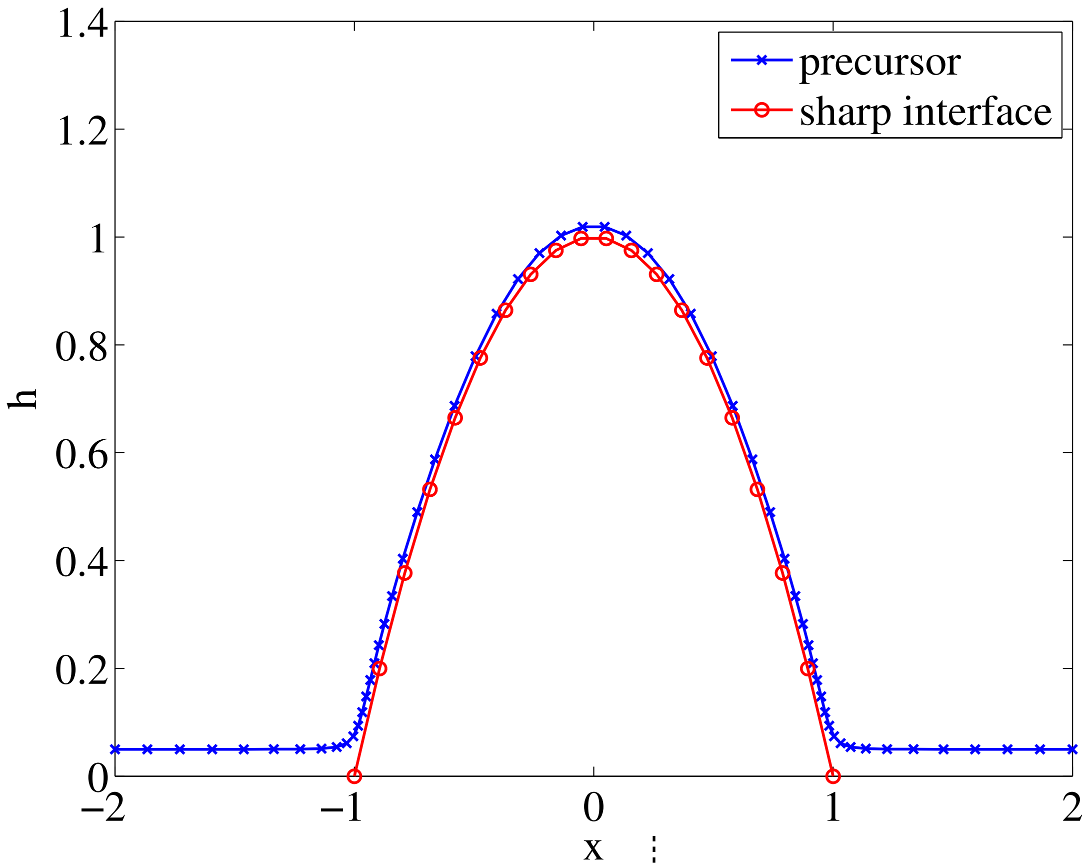
$$V(h) = \sigma_{\mathrm{s}} \int \Phi\left(\tfrac{h}{\varepsilon}\right) \mathrm{d}x$$

$$\xrightarrow{\Gamma-\text{limit}}$$

…or without

$$V(h) = \sigma_{\mathrm{s}}\mu(\{x : h(x) > 0\}) \overset{1d}{=} \sigma|x_+ - x_-|$$

Regularization vs free boundary problem



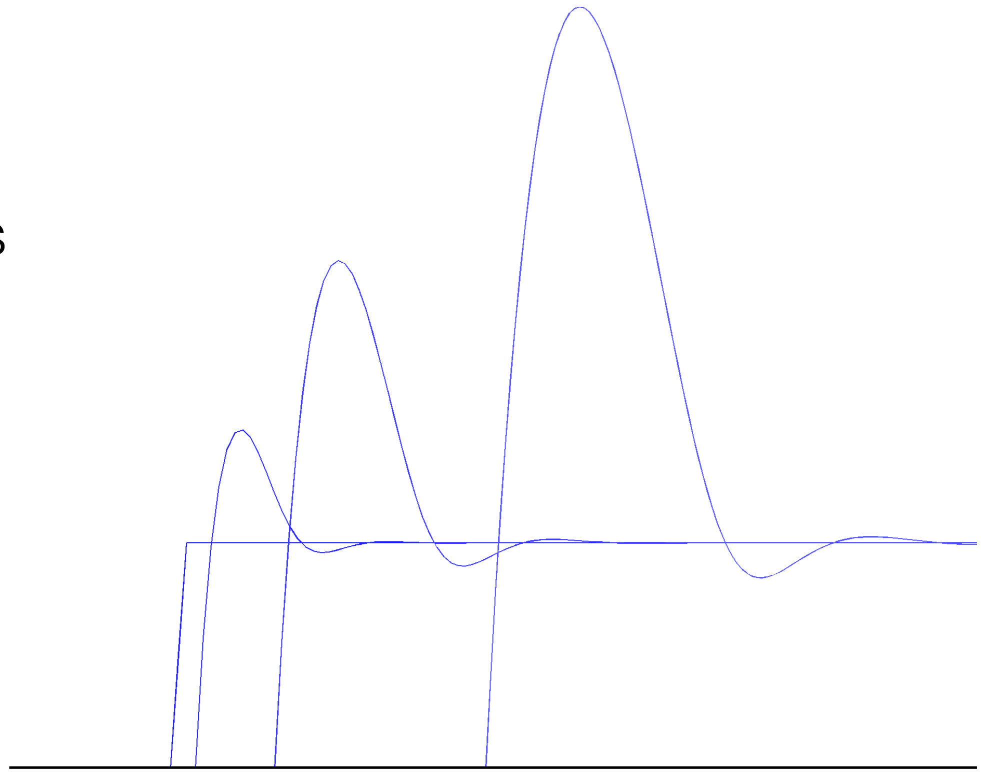
	regularized (global)	free boundary (supported)
+	topological transitions, standard to implement	coarse meshes, stable & accurate
-	refinement near contact line unknowns, stability	no topological transitions free boundary problem

Regularization vs free boundary problem

$$E(h) = \int \frac{1}{2}|\nabla h|^2 \mathrm{d}x + V(h), \qquad V(h) = \sigma_{\mathrm{s}}|x_+ - x_-|$$

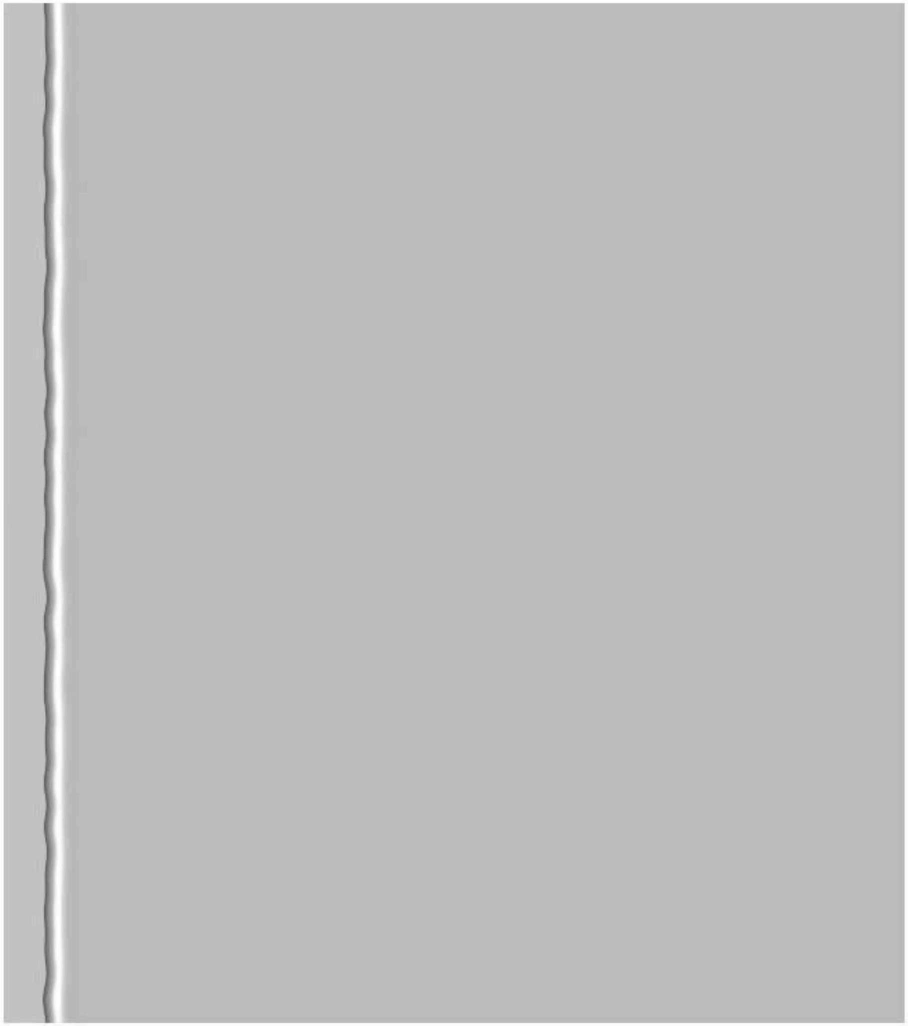
Optimize size of support vs gradients of the solution leads to contact angle

but also drives motion and instabilities

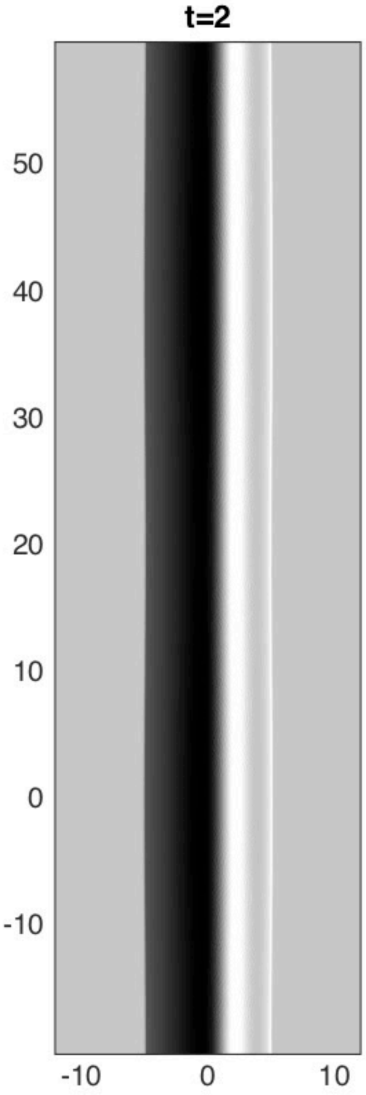


Typical instabilities on solid surfaces

P2 FEM with (heuristic) spatial adaptivity

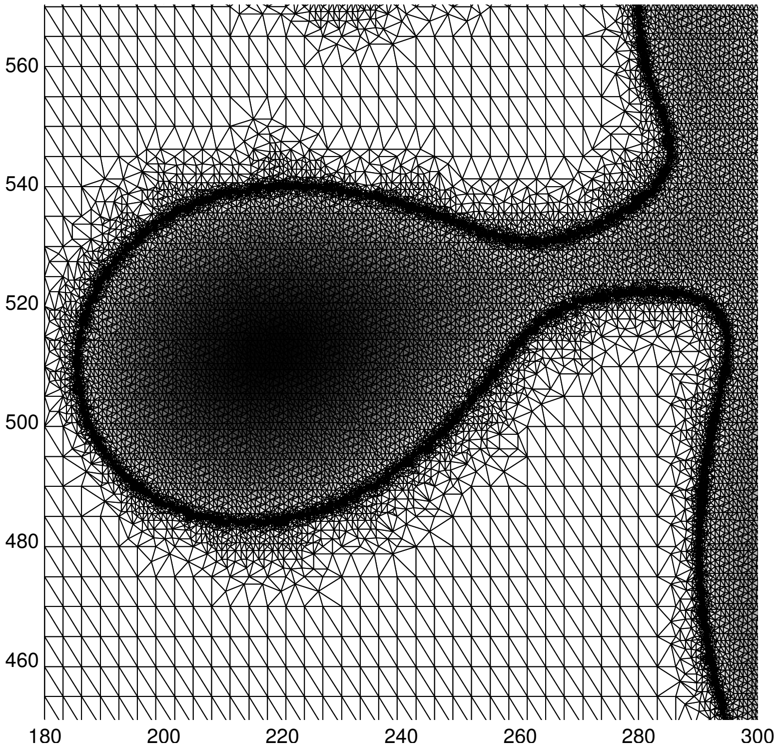


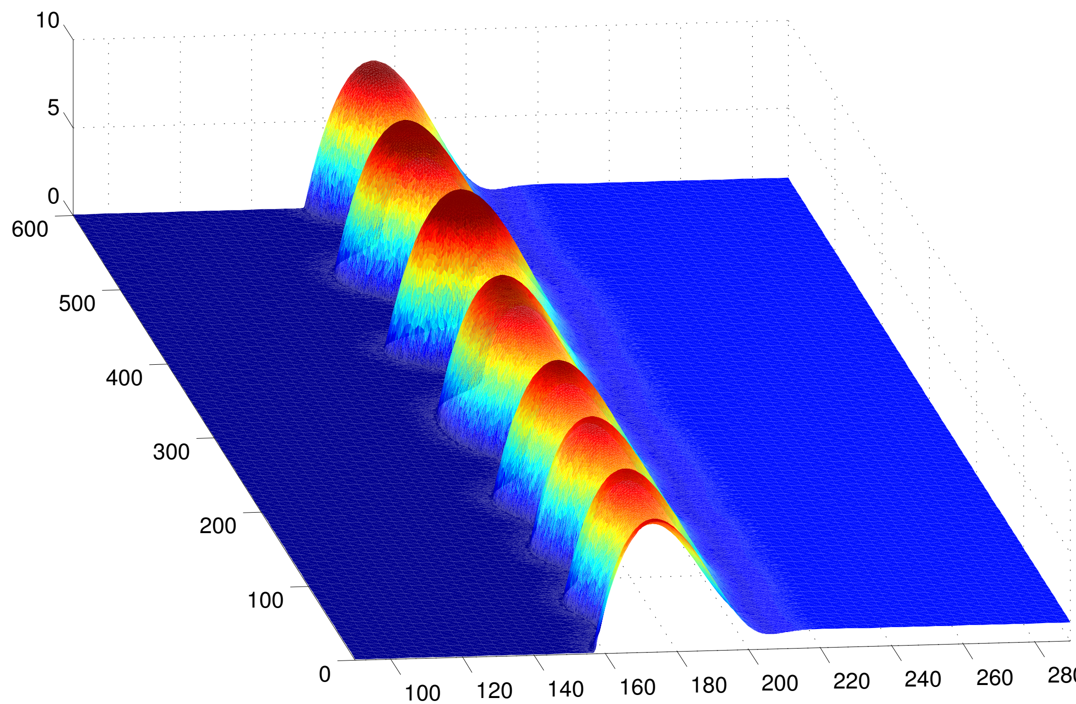
dewetting instability with mobility n=2



Plateau-Rayleigh instability with n=3

Regularization vs free boundary problem





2. Droplets on solid planar surfaces

free boundary approach - algorithm 1D

Algorithm

1. original PDE

$$\dot{h} - (|h|^n \pi_x)_x = 0$$

$$\pi = \frac{\delta E}{\delta h} = -h_{xx}$$

2. weak formulation (discrete in space using linear FEM)

$$\int_{x_-}^{x_+} (\,\dot{h}\phi + |h|^n \pi_x \phi_x)\,\mathrm{d}x = 0$$

$$\int_{x_-}^{x_+} (\,\pi\varphi - \tau\dot{h}_x\varphi_x)\,\mathrm{d}x = \int_{x_-}^{x_+} h_x\varphi_x\,\mathrm{d}x - h_x\phi\big|_{x_-}^{x_+}$$

Note on the handling of time-derivatives

- $h(t+\tau,\cdot) = h(t,\cdot) + \tau\dot{h}$ makes no sense
- ALE (arbitrary Lagrangian-Eulerian) transformation as post-processing

3. define transformation $\psi_{t_0}(t,\cdot) : (x_-(t_0), x_+(t_0)) \to (x_-(t), x_+(t))$

$$\psi_{t_0}(t,x) = x_-(t) + \xi(x)\big(x_+(t) - x_-(t)\big)$$

$$\xi(x) = \frac{x - x_-(t_0)}{x_+(t_0) - x_-(t_0)}$$

4. pull-back of h by ψ_{t_0} gives

$$\boxed{H(t,x) = h\big(t, \psi_{t_0}(t,x)\big)}$$

Algorithm

5. time derivative uniquely decomposes

$$\dot{H} = \dot{h} + \dot{\psi} h_x \quad \xrightarrow[t \approx t_0]{\dot{H}(t, x_\pm(t_0)) \equiv 0} \quad \dot{h}(x) \mapsto \begin{pmatrix} \dot{\psi}(x_-) \\ \dot{H} \\ \dot{\psi}(x_+) \end{pmatrix}$$

$$\dot{x}_\pm = \cancel{\lim_{x \to x_\pm} \left(\frac{m}{h} \pi_x \right)}$$

6. update according to time-derivatives

$$x^{k+1} = x^k + \tau \dot{\psi}(x^k),$$

$$h^{k+1} = h^k + \tau \dot{H},$$

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https://github.com/dpeschka/thinfilm-freeboundary.git (about 120 lines MATLAB proof-of-concept 1D code)

P. Thin-film free boundary problems for partial wetting. J. Comp. Phys. (2015)
P. Numerics of contact line motion for thin films, IFAC PapersOnline (2015)

2. Droplets on solid planar surfaces

free boundary approach - algorithm 2D

Extension to 2D

1. original PDE

$$\dot{h} - \nabla \cdot (|h|^n \nabla \pi) = 0$$

$$\pi = \frac{\delta E}{\delta h}$$

2. weak formulation (discrete in space using linear FEM)

$$\int_\omega (\dot{h}\phi + |h|^n \nabla \pi \cdot \nabla \phi) = 0$$

$$\int_\omega (\pi\psi - \tau \nabla \dot{h} \cdot \nabla \psi = \int_\omega \nabla h \cdot \nabla \psi - \int_{\partial\omega} \psi \partial_\nu h$$

Extension to 2D

3. pull-back and ALE formulation

$$H(t,x) = h(t,y), \qquad \text{where} \quad y = \Psi_t(x)$$

$$\dot H(t,x) = \dot h(t,y) + \dot\Psi_t(x) \cdot \nabla_y h(t,y)$$

4. deformation problem

Use harmonic $\dot\Psi$ with boundary data:

normal part of mapping $\dot\Psi \cdot \nu$:

$$0 = \dot h(t,y) + \dot\Psi_t(x) \cdot \nabla_y h(t,y)$$

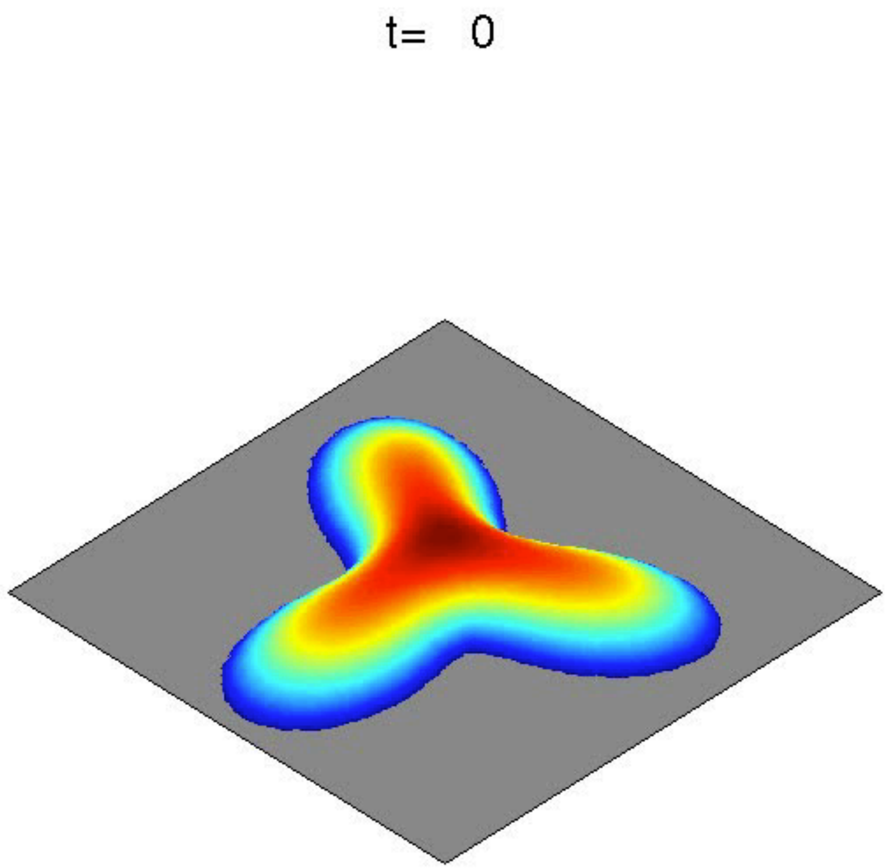
tangential part of mapping $(1 - \nu\nu^\top)\dot\Psi$:

$\Rightarrow$ Deformed meshes are more uniform.

2. Droplets on solid planar surfaces

Examples

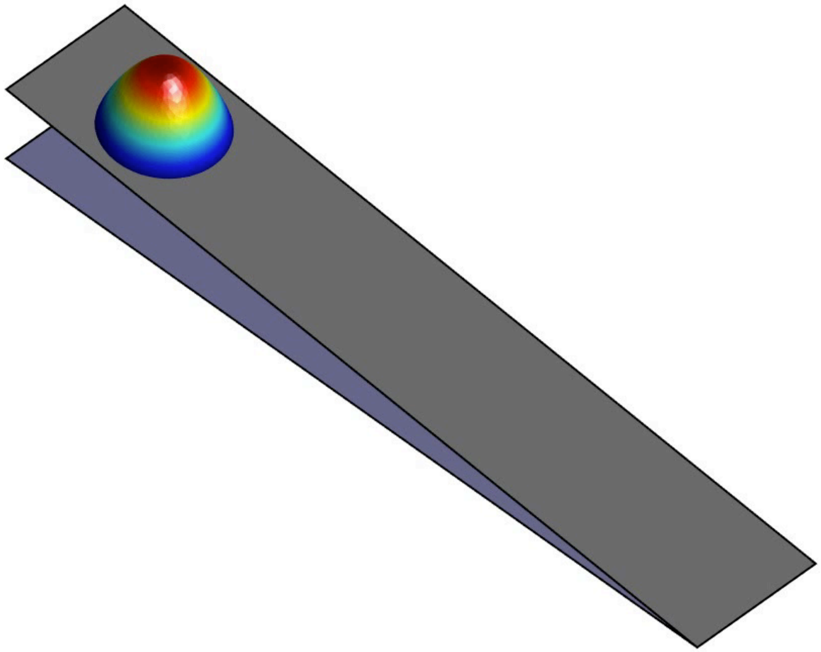
Examples

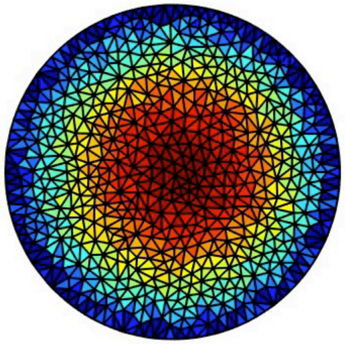


Evolution towards equilibrium $h(x) = \left(\bar{h}\left(1 - \frac{|x-\bar{x}|^2}{R^2}\right)\right)_+$

Examples

Gravity driven motion $$\tilde{E}(h) = E(h) + \int \rho g h(\alpha h + \beta x)\,\mathrm{d}x$$

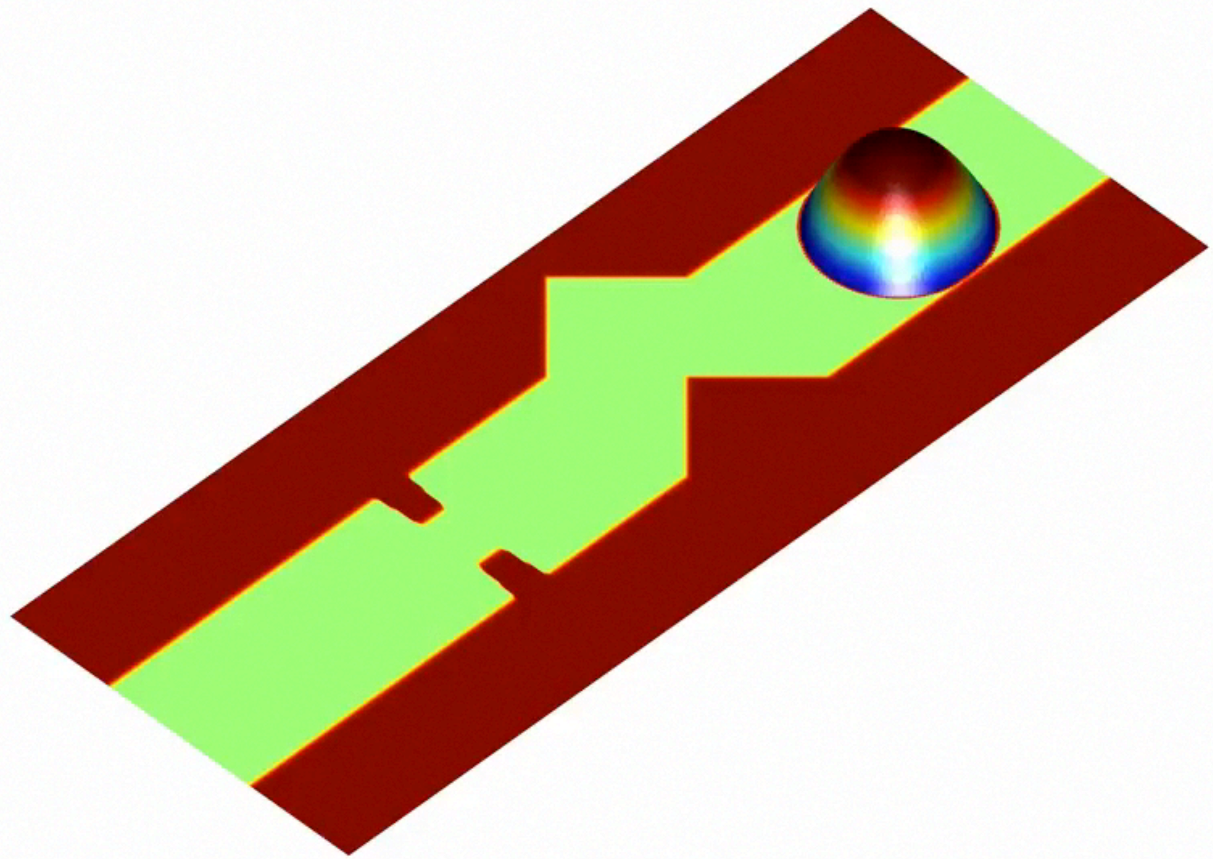




Podgorski, Flesselles, Limat
Schwartz et al.
Eggers/Snoeijer et al.

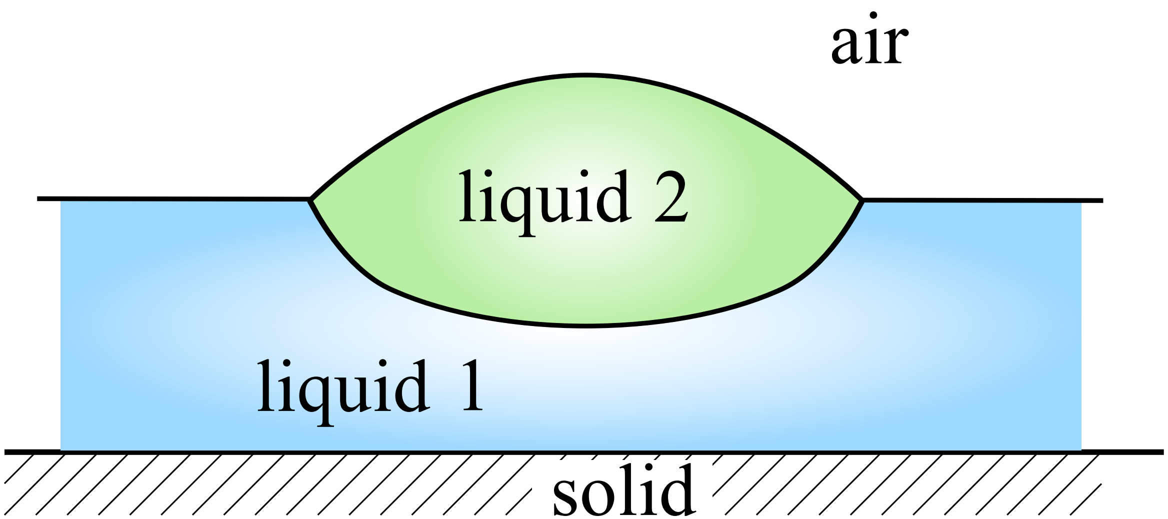
Energetic patterning

$$V(h) = \int_{\omega} \sigma_{\mathrm{s}}(x, y)\,\mathrm{d}x$$

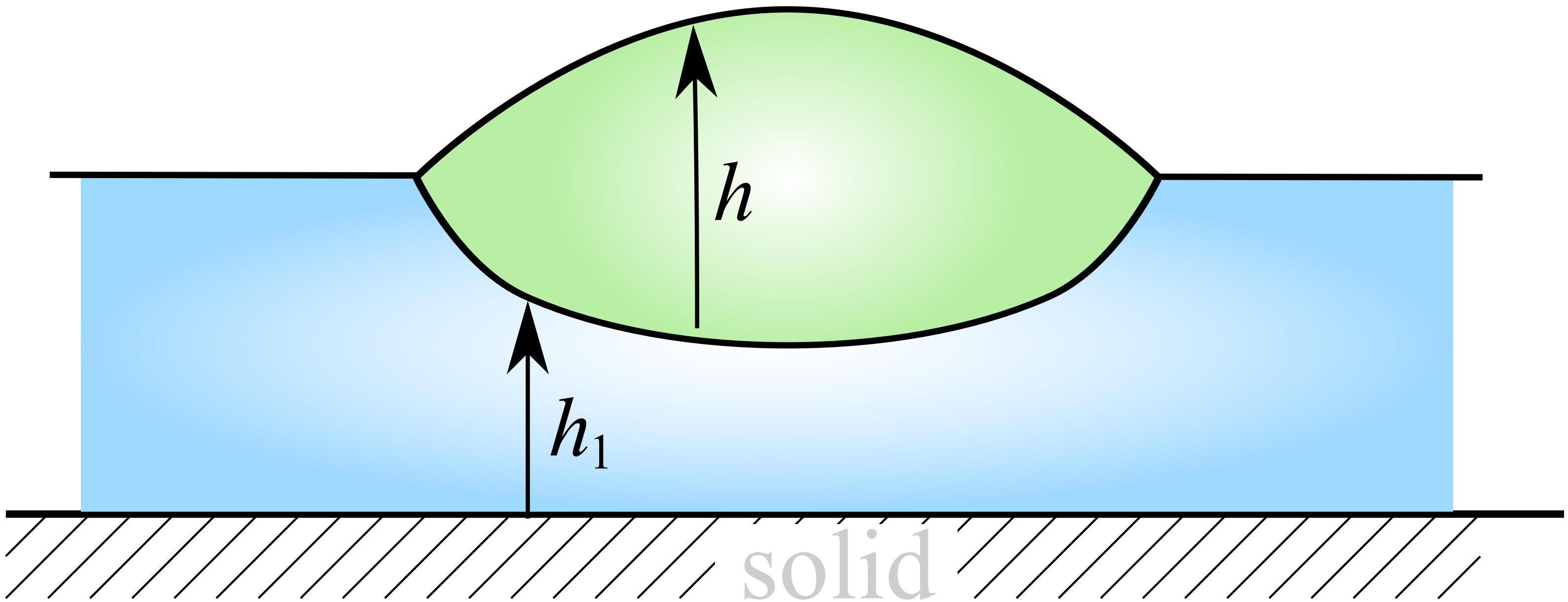


Kondic, Diez. Phys. Fluids (2004)

3. Flows over liquid substrates (1D)



Flow over liquid substrates



$$\Omega_1(t) = \{(x,z) : 0 < z < h_1(t,x)\}$$

$$\Omega_2(t) = \{(x,z) : h_1(t,x) < z < h_1 + h(t,x)\}$$

Outlook

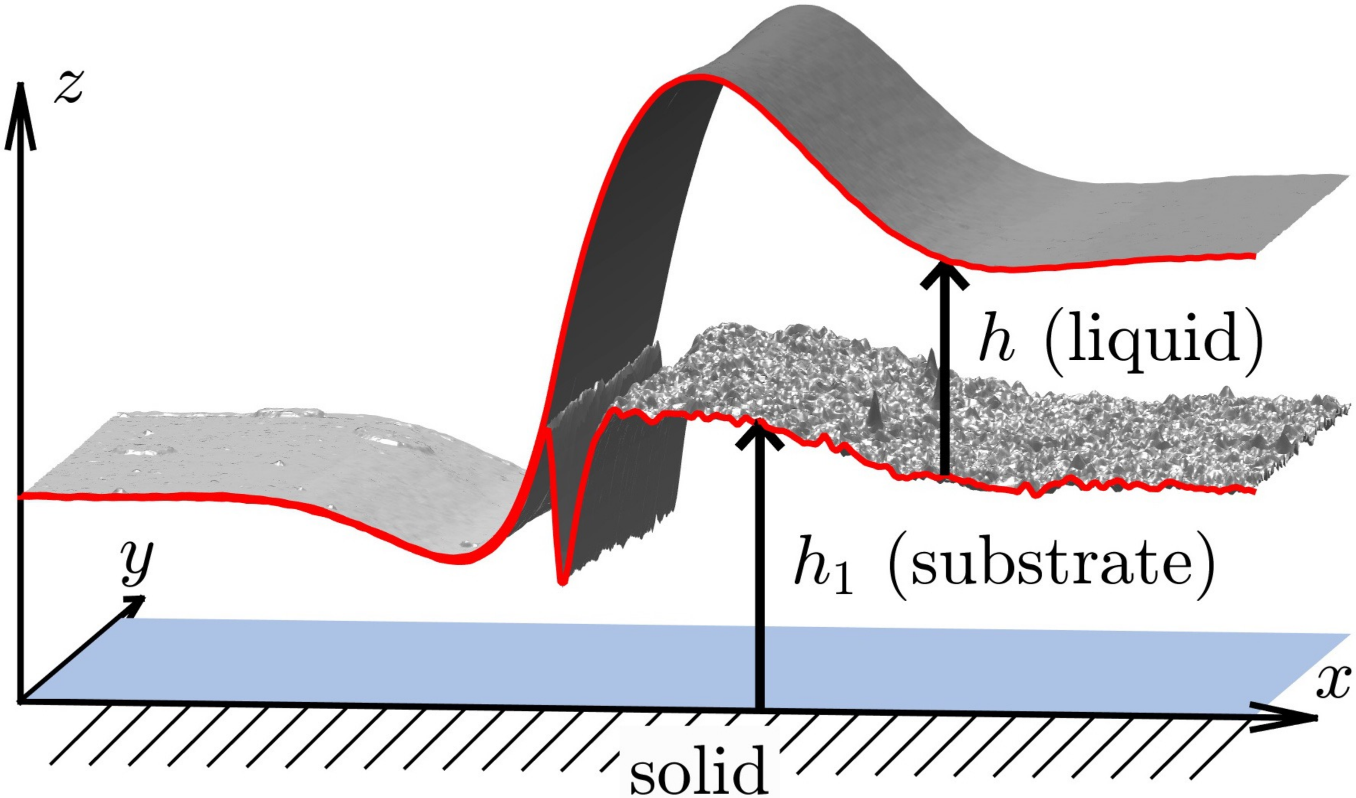
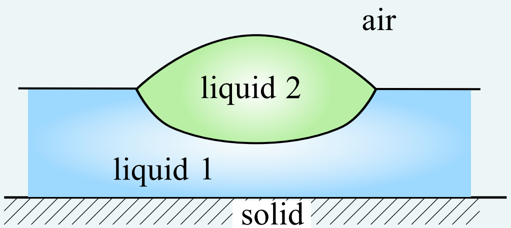


Figure: composed AFM images of liquid polystyrene dewetting on top from an liquid polymethyl methacrylate substrate (R. Seemann, Univ. d. Saarlandes, Saarbrücken, Germany)

$$\partial_t \mathbf{h} - \nabla\cdot\left(M(\mathbf{h})\nabla\boldsymbol{\pi}\right) = \mathbf{0} \qquad \text{in } \omega(t)$$
$$\partial_t h_1 - \nabla\cdot\left(m(h_1)\nabla\pi_1\right) = 0 \qquad \text{in } \Omega\setminus\omega(t)$$



with $\mathbf{h} = (h_1, h)$ and $\boldsymbol{\pi} = (\pi_1, \pi)$ with $\pi_1 = \delta E/\delta h_1$, $\pi = \delta E/\delta h$.

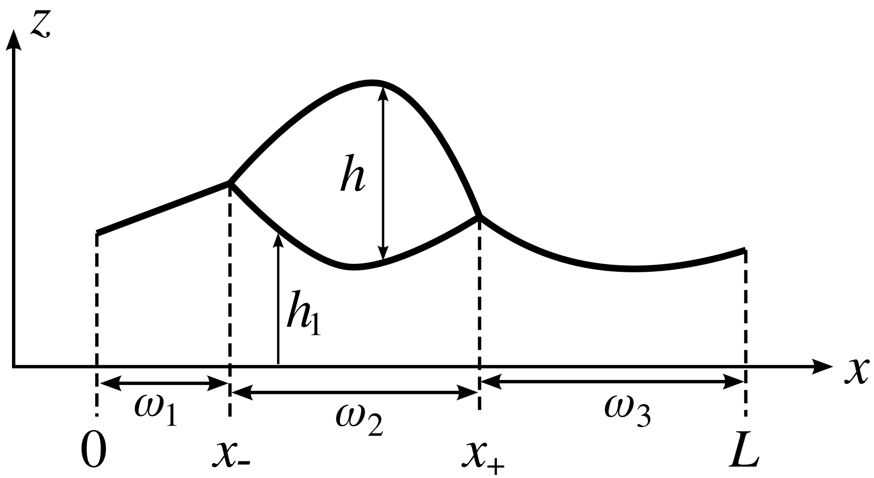
$$m(h_1) = \frac{h_1^3}{3} \qquad M(\mathbf{h}) = \begin{pmatrix} \frac{1}{3}h_1^3 & \frac{1}{2}hh_1^2 \\ \frac{1}{2}hh_1^2 & \frac{\mu}{3}h^3 + h_1h^2 \end{pmatrix}$$

Is also unknown

$$\omega(t) = \{x \in \mathbb{R}^{d-1} : h(t,x) = 0\}$$

Miksis and Kriegsmann, SIAM J. Appl. Math. (2003)
Pototsky, Bestehorn, Merkt, Thiele, Phys. Rev. E (2004)

Flow over liquid substrates



state $$s = \{(h, h_1, x_-, x_+ : 0 < x_- < x_+ < L; 0 \le h, h_1; \ldots\}$$

$$\mathbf{m} = \left(\int h \, \mathrm{d}x, \int h_1 \, \mathrm{d}x \right)$$

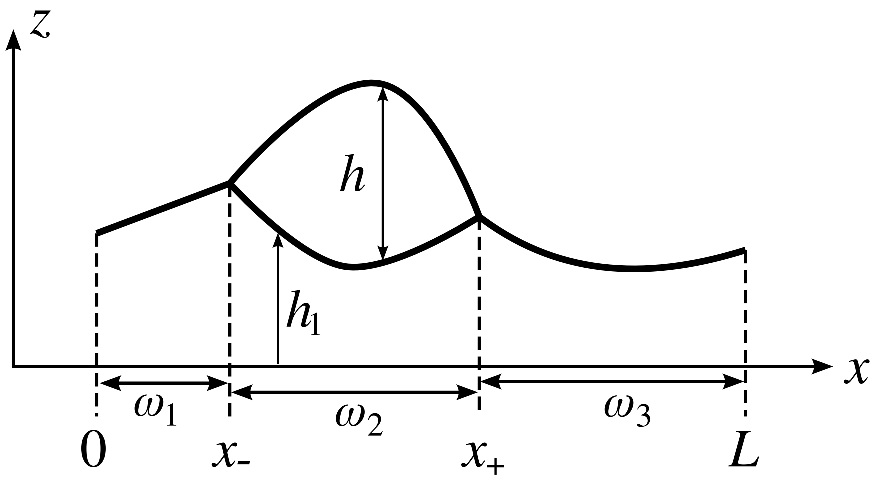
velocity $$u = \{(\dot{h}, \dot{h}_1, \dot{x}_-, \dot{x}_+ : \ldots\}$$

$$\dot{h} + \dot{x}_\pm \cdot \nabla h = 0$$

$$[[\dot{h}_1 + \dot{x}_\pm \cdot \nabla h]] = 0$$

Karapetsas, Craster, Matar, Phys. Fluids (2011)
Huth, Jachalski, Kitavtsev, P. Gradient flow perspective on thin-film bilayer flows. J. Engr. Math. (2014)

Flow over liquid substrates



energetics D,E

$$E(h,h_1) = \int \frac{\sigma}{2}|\nabla h_1|^2 + \frac{1}{2}|\nabla(h_1+h)|^2 \mathrm{d}x + \sigma_s|x_+ - x_-|$$

$$D = \sum_{i,j=1}^{2} \int Q_{ij}\nabla\pi_i \cdot \nabla\pi_j \mathrm{d}$$

constraints C

$$\int_\omega \dot{h}_1\phi_1 + (Q_{11}\nabla\pi_1 + Q_{12}\nabla\pi_2)\nabla\phi_1\,\mathrm{d}x = 0$$

$$\int_{\omega_2} \dot{h}\phi + (Q_{21}\nabla\pi_1 + Q_{22}\nabla\pi_2)\nabla\phi\,\mathrm{d}x = 0$$

$$\dot{h} + \dot{x}_\pm \cdot \nabla h = 0 \qquad [[\dot{h}_1 + \dot{x}_\pm \cdot \nabla h]] = 0$$

minimization problem via Langrange multiplier

$$D(u,v) + \langle v, C^\top\lambda\rangle = -\langle \mathrm{diff}\, E, v\rangle$$

$$\langle q, Cu\rangle = 0$$

Flow over liquid substrates

$$\dot{\xi}(x) := \begin{cases} \dot{x}_- \frac{x}{x_-} & x \in \omega_1 \\ \dot{x}_- \left(1 - \frac{x - x_-}{x_+ - x_-}\right) + \dot{x}_+ \left(\frac{x - x_-}{x_+ - x_-}\right) & x \in \omega_2 \\ \dot{x}_+ \left(1 - \frac{L - x}{L - x_+}\right) & x \in \omega_3 \end{cases}$$

$$H(t,x) = h\big(t, \xi_t(x)\big)$$

$$\dot{h}(t,x) + \dot{\xi}(x) \cdot \nabla h(t,x) = \dot{H}(t,x)$$

$$\dot{h}_1(t,x) + \dot{\xi}(x) \cdot \nabla h_1(t,x) = \dot{H}_1(t,x)$$

$$\xi_{t+\tau}(x) = x + \tau \dot{\xi}(x),$$

$$H(t+\tau, x) = h(t,x) + \tau \dot{H}(t,x),$$

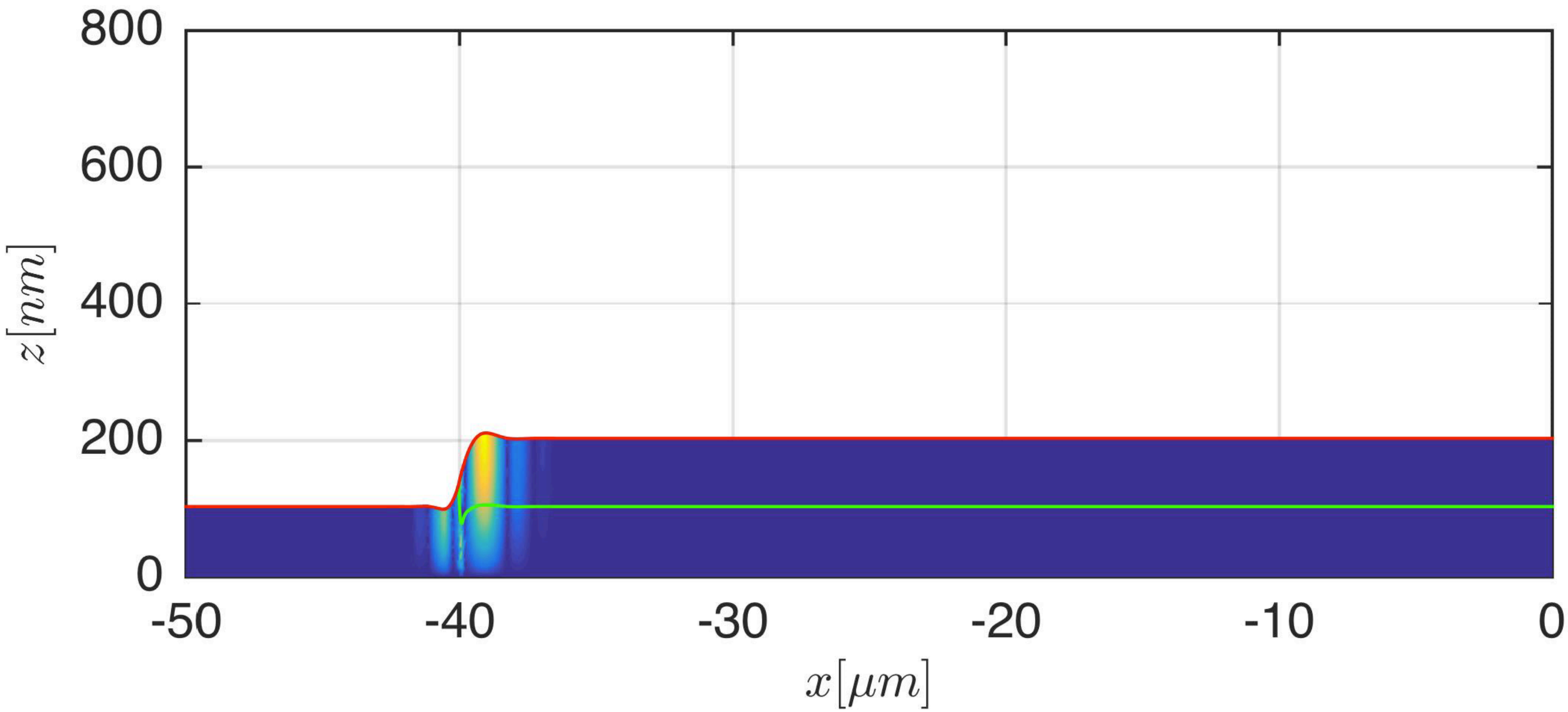
$$H_1(t+\tau, x) = h_1(t,x) + \tau \dot{H}_1(t,x).$$

minimization problem via Langrange multiplier

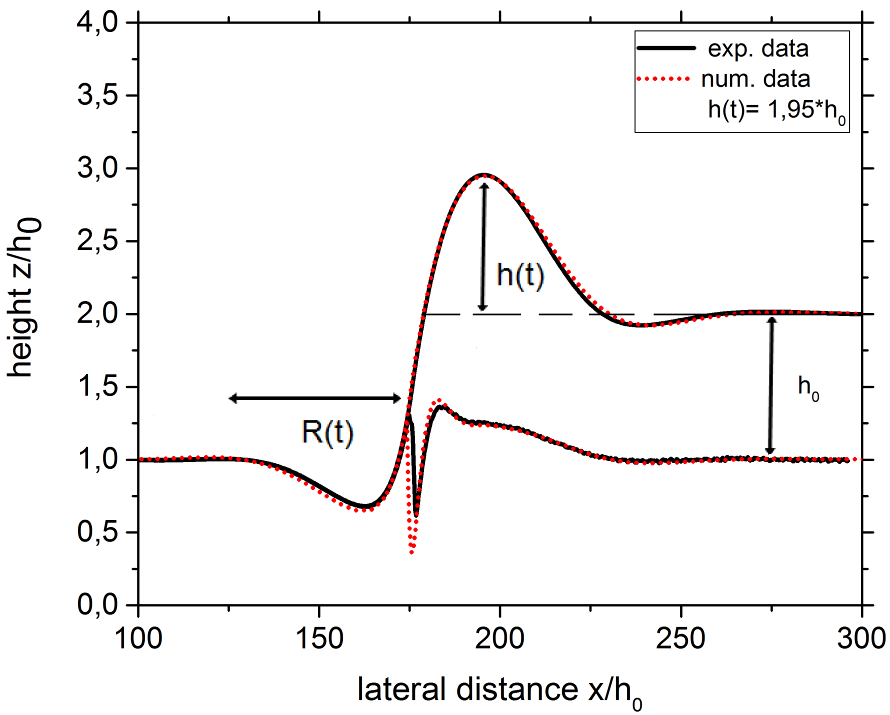
$$D(u,v) + \langle v, C^\top \lambda \rangle = -\langle \mathrm{diff}\, E, v \rangle$$

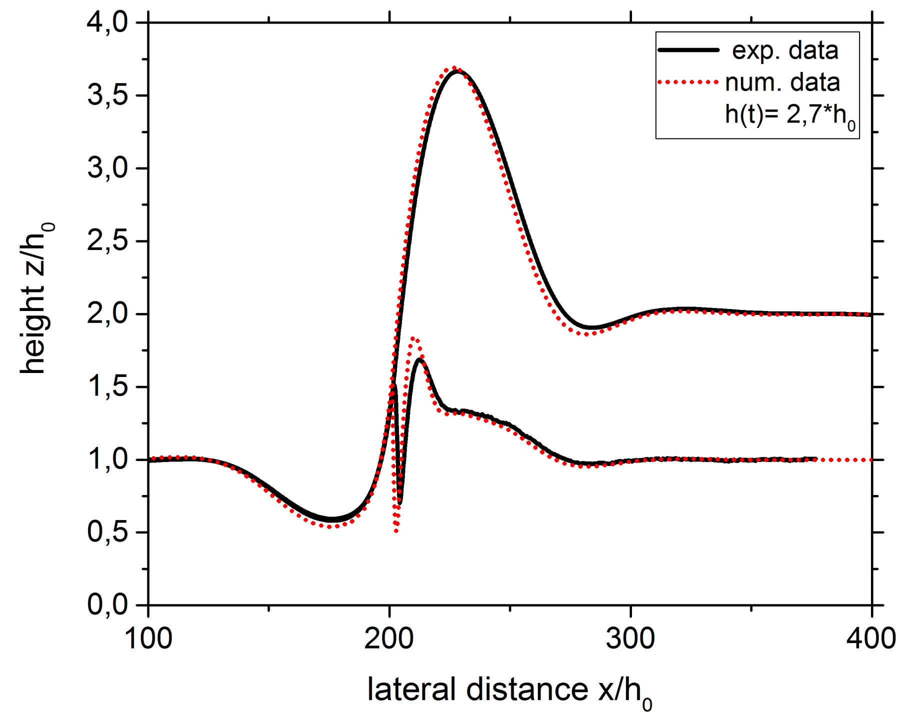
$$\langle q, Cu \rangle = 0$$

Flow over liquid substrates

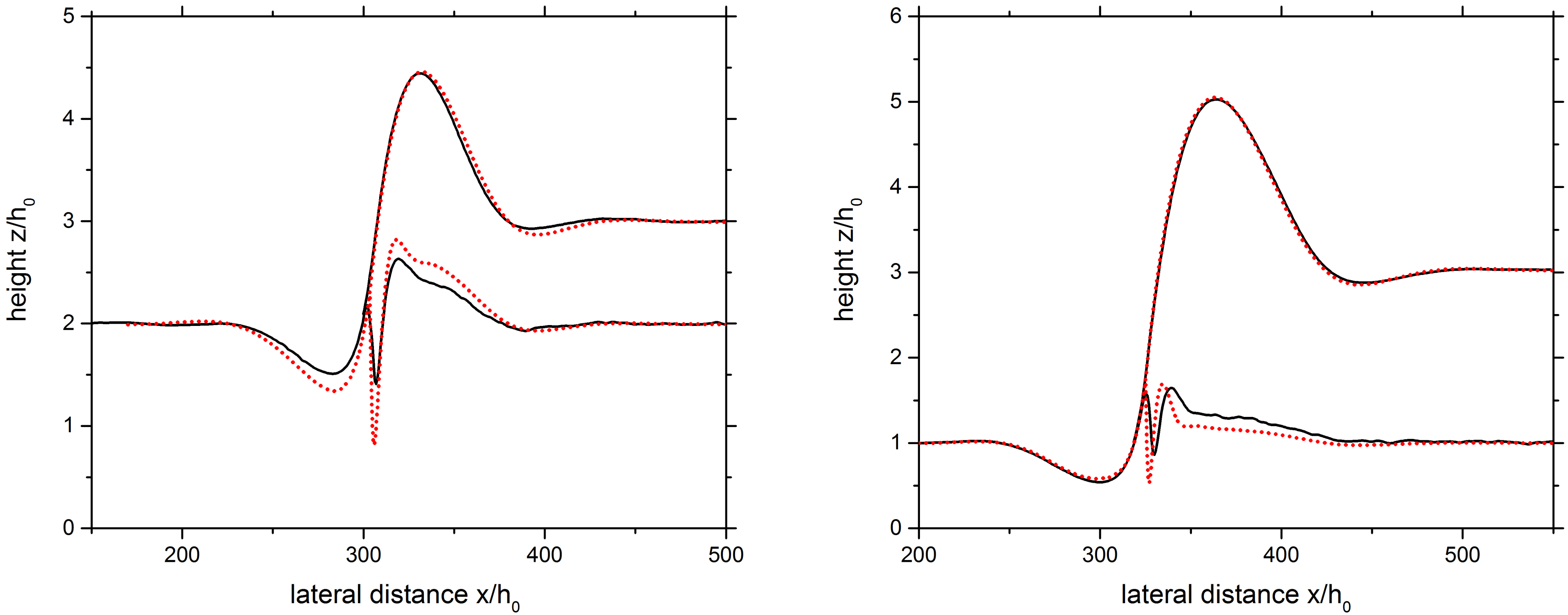


Flow over liquid substrates





Flow over liquid substrates



Summary:

Thin film flows as free boundary problem, where the support set

$$\omega(t) = \{x \in \mathbb{R}^d : h(t,x) > 0\}$$

is unknown and depends on time.

- contact angles naturally in variation formulation
- analysis established (even more natural)
- lack of practical algorithms (so-far)
- extension to bilayer flows works, comparison promising

Outlook:

- higher order algorithms (isoparametric FEM, w. Luca Heltai based on deal.II)
- modeling of contact line physics and better control of contact line motion to better control effects such as contact line hysteresis

Thank you!

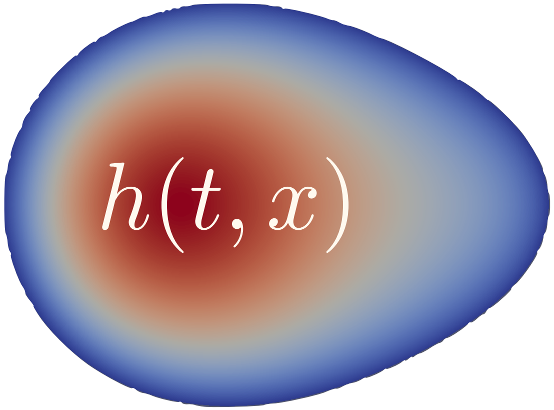
Supported by Transport processes at fluidic interfaces DFG SPP 1506 and ECMath Einstein Center for Mathematics Berlin

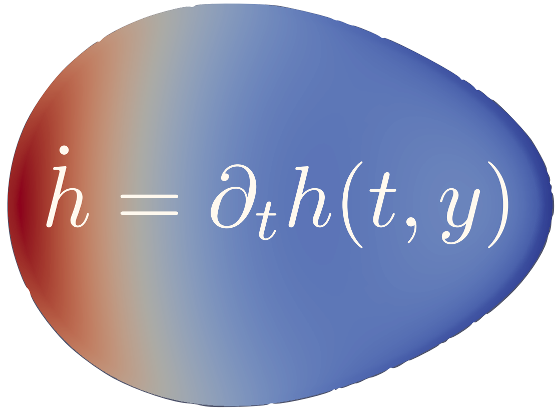
Research Center MATHEON
Mathematics for Key Technologies

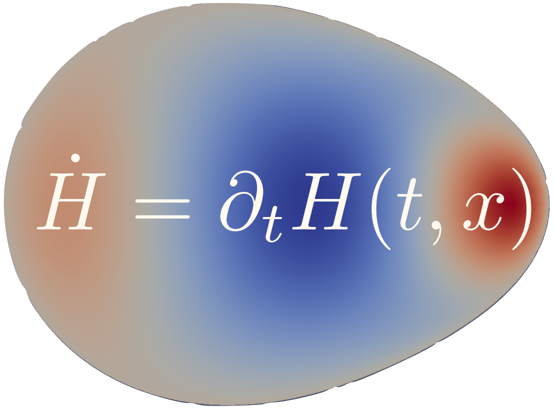
References

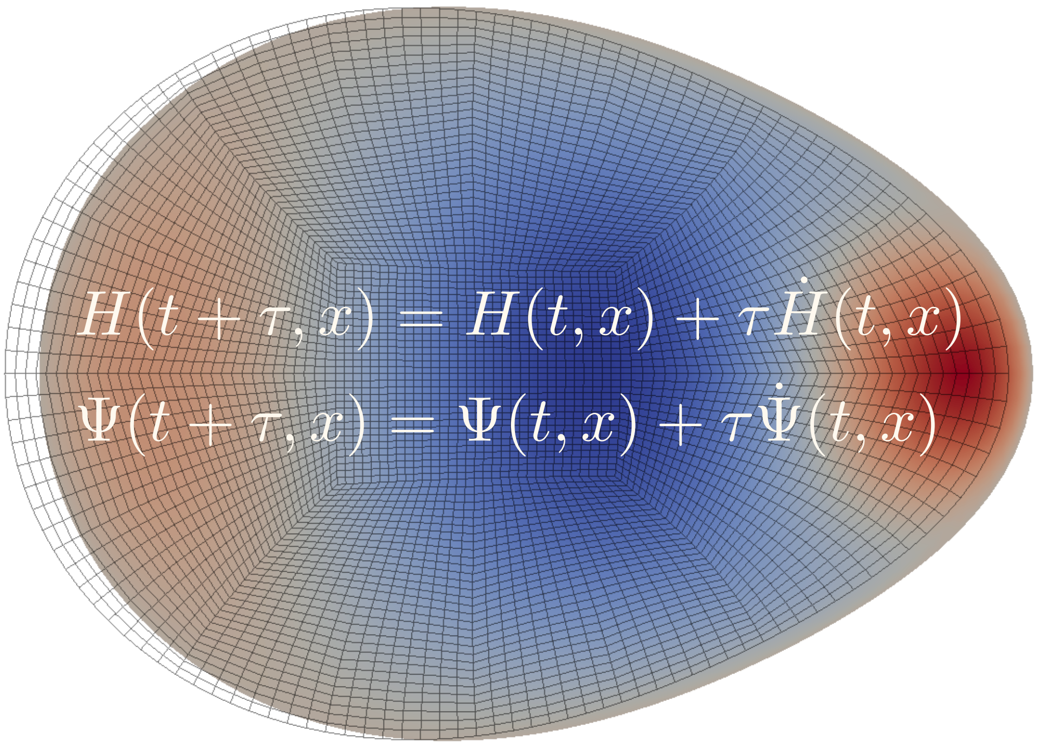
- Bernis & Friedmann 1990 (existence weak sol.)
- Bernis *et al.* 1992 (source type solutions)
- Bertozzi & Pugh 1996 (regularity & long-time behavior)
- Eggers 1997 (breakup & singularities)
- **Review** Oron, Davis, Bankoff 1997
- Otto 1998 (long time existence weak sol. with sharp interface with m(h)=h and nonzero angle)
- Grün, Bertozzi & Witelski 2000 (stationary states and coarsening)
- Grün & Rumpf 2000 (nonegativity preserving schemes)
- Wagner, Münch & Witelski 2005 (New regimes of thin film equations)
- Bertsch, Giacomelli & Karali 2005 (existence of arb. weak sol. with reg. contact angle)
- Giacomelli, Knüpfer & Otto 2008
  (existence & uniqueness! in interp. spaces of weighted Sobolev spaces with zero slope) …

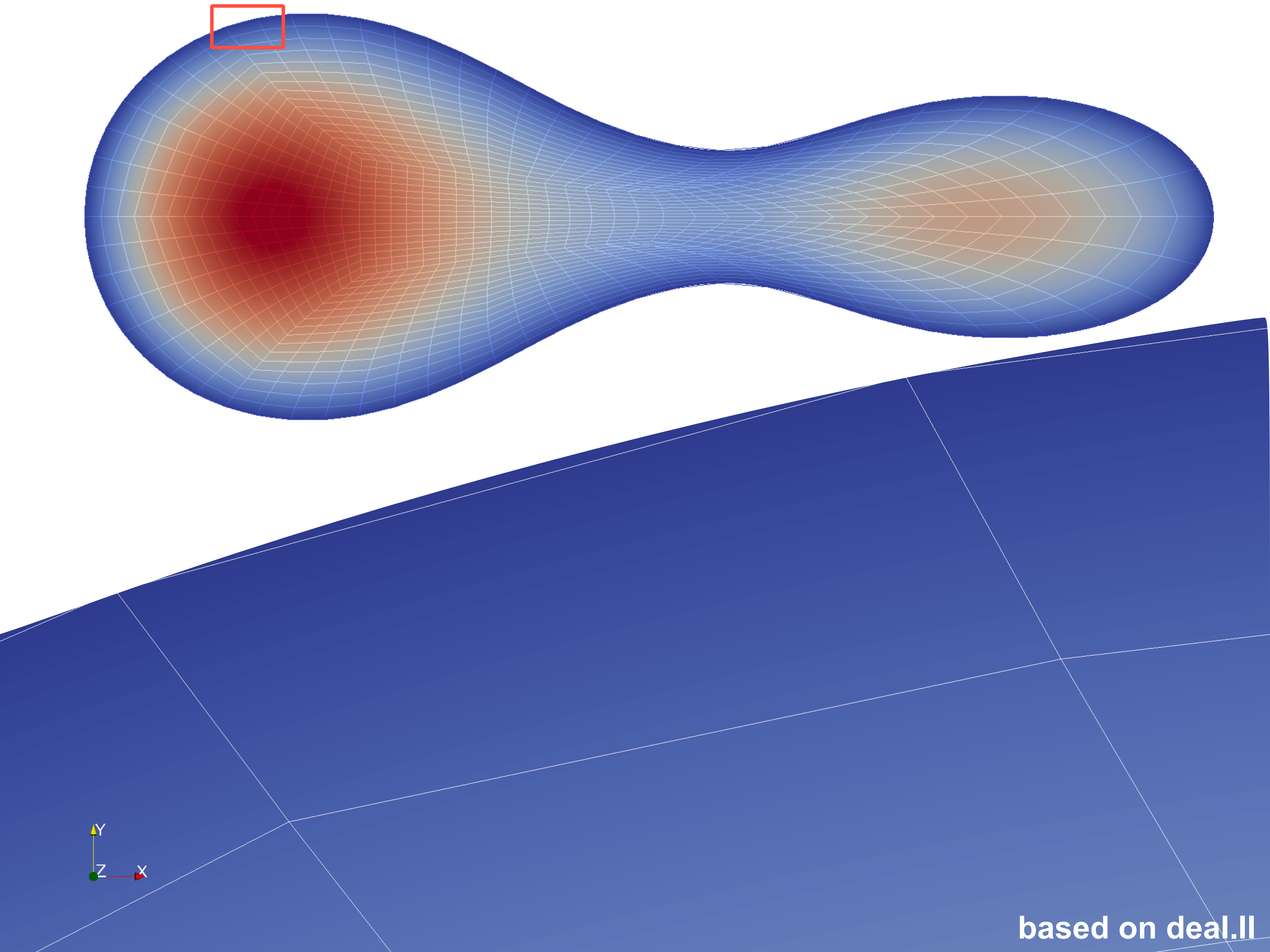
Extension to 2D










Y
Z
X
based on deal.II

Gradient formulation in detail

Dissipation: D

$$D(\mathbf{u},\mathbf{v}) = \int_\Omega \tfrac{1}{2}\tau : (\nabla\mathbf{v} + \nabla\mathbf{v}^\top)\,\mathrm{d}\Omega + \int_{\Gamma_s} \beta^{-1}\mathbf{u}\cdot\mathbf{v}\,\mathrm{d}\Gamma$$
$$= \int_\Omega (-\nabla\cdot\tau)\cdot\mathbf{v}\,\mathrm{d}\Omega + \int_\Gamma (\tau\mathbf{n})\cdot\mathbf{v}\,\mathrm{d}\Gamma + \int_{\Gamma_s} \beta^{-1}\mathbf{u}\cdot\mathbf{v}\,\mathrm{d}\Gamma$$

where $\tau = \mu(\nabla\mathbf{u} + \nabla\mathbf{u}^\top)$.

Energy: $\mathrm{diff}\,E$

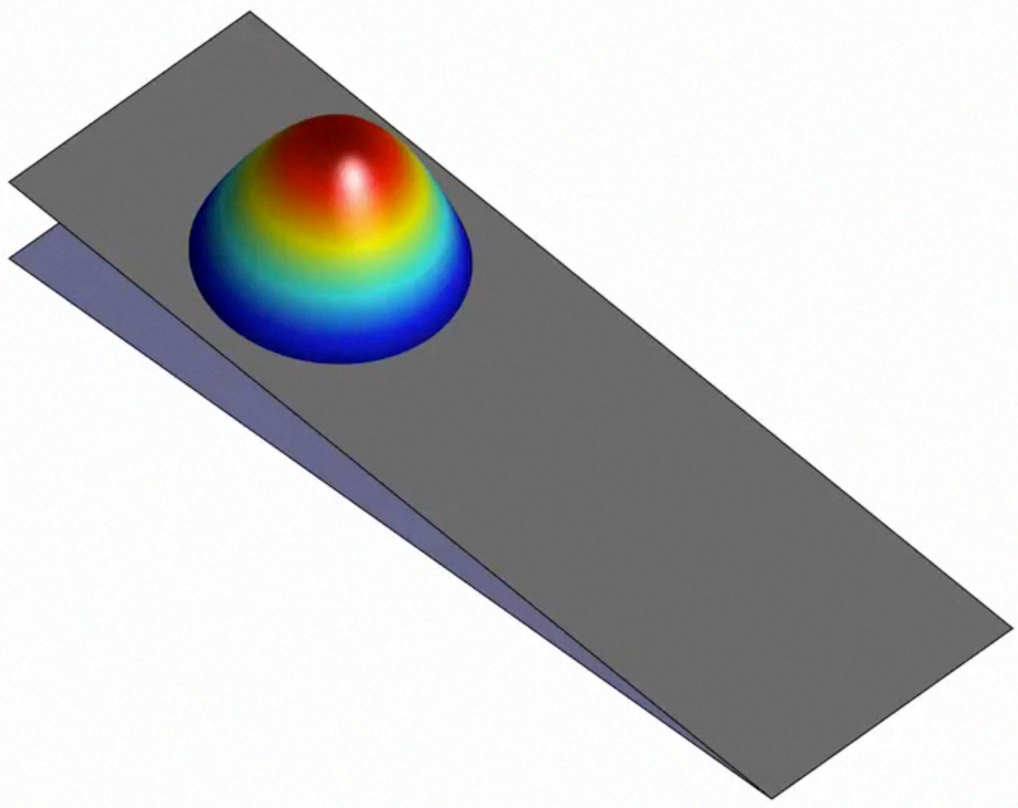
$$\mathrm{diff}\,E[\mathbf{v}] = \sum_f \sigma_f \int_{\Gamma_f} \nabla_{\|}\mathrm{id} : \nabla_{\|}\mathbf{v}$$
$$= -\sum_f \sigma_f \left((d-1)\int_{\Gamma_f} \kappa\mathbf{n}\cdot\mathbf{v} - \int_{\partial\Gamma_f} \mathbf{v}\cdot\mathbf{n}_\Gamma \right)$$

Minimization: $D + \mathrm{diff}\,E = 0$

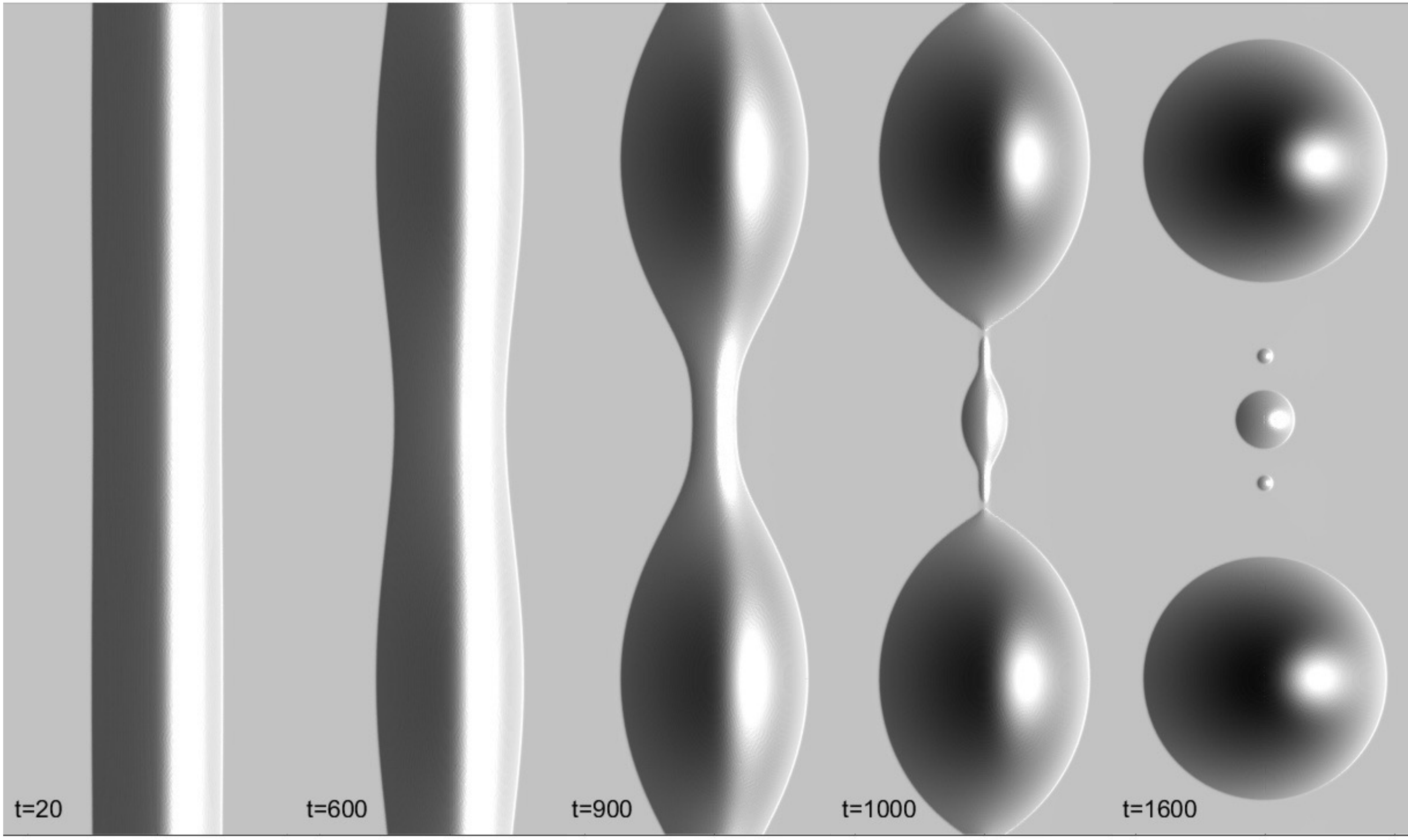
$$-\sum_\alpha \int_{\Omega_\alpha} (\nabla\cdot\tau)\cdot\mathbf{v} + \sum_f \int_{\Gamma_f} \mathbf{v}\cdot\left([[\tau]]\cdot\mathbf{n} - (d-1)\sigma_f\kappa\mathbf{n}\right)$$
$$+ \int_{\Gamma_0} (\mathbf{t}\cdot\tau\mathbf{n} + \beta^{-1}\mathbf{u}\cdot\mathbf{t})(\mathbf{t}\cdot\mathbf{v})\,\mathrm{d}\Gamma + \sum_f \sigma_f \int_{\partial\Gamma_f} \mathbf{v}\cdot\mathbf{n}_{\Gamma_f} = 0$$

Effect of mobility on droplet shape

$$m(h) = |h|^{3+\delta}$$



Regularization vs free boundary problem



Plateau-Rayleigh instability with cubic mobility

„A free boundary problem is a (nonlinear) PDE, whose domain is part of the unknowns“

Examples for free boundary problems:

- flows with free surfaces and interfaces (*e.g.* this talk: dewetting fronts & droplets)
- geometric evolution (mean curvature flow)
- multi-phase problems with phase transitions (Stefan problem)
- fluid-structure interaction, obstacle problems
- …

Algorithm

$$m(h) = |h|^n, \qquad n \in \left(\tfrac{3}{2}, 3\right)$$

Regularity of source type solutions

$$h(t,x) = t^{-\frac{1}{n+4}} H(\xi), \qquad \xi = x t^{-\frac{1}{n+4}}$$

$$H(y) = A^{-\nu/3} y^{\nu}(1 + v(y, y^{\beta})), \qquad y = \xi + 1$$

$$\nu = \frac{3}{n}, \quad A = \nu(\nu-1)(2-\nu), \quad \beta = \frac{\sqrt{-3\nu^2 + 12\nu - 8} - 3\nu + 4}{2}$$

Regularity near the boundary of the support is an issue!

$$\boxed{\begin{aligned} |h_x(t, x_\pm)| &= \tan\theta \\ \dot{x}_\pm &= \lim_{x \to x_\pm} \left(\frac{m}{h}\pi_x\right) \end{aligned}}$$

Gnann, Otto Giacomelli. Eur. J. Appl. Math. (2013)
Bernis et al. ….