Solid-State Dewetting: Modeling & Numerics

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Workshop on Modeling and Simulation of Interface Dynamics in Fluids/Solids and Applications

May 16th, 2018

Outline

1. Introduction
2. What is surface diffusion? – Physics and Mathematics
3. How to model it? – Thermodynamic variation
4. How to numerically solve it? – PFEM
5. How to analyse it? – Onsager's variational principle
6. Summary

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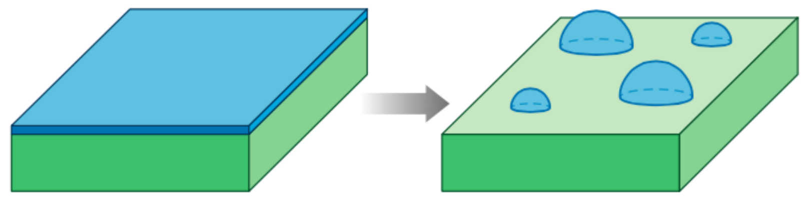
Liquid-State Wetting/Dewetting

Figure: Water droplets in our daily life.

Solid-State Dewetting

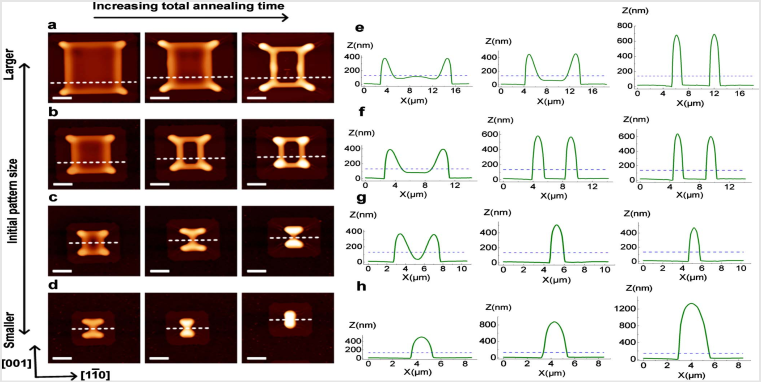
- Most thin films are *metastable* in as-deposited state and *dewet* to form particles (C.V. Thompson, Annu. Rev. Mater. Res., 2012).



- *The dewetting* can occur well *below* the melting temperature of the material, i.e., which is still in the *solid-state*.

Physical Experiments

Dewetting Patterned Films: Ni(110) Square Patches[1]



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[1] J. Ye & C.V. Thompson. PRB, 2010.

Physical Experiments

Dewetting on SOI system:

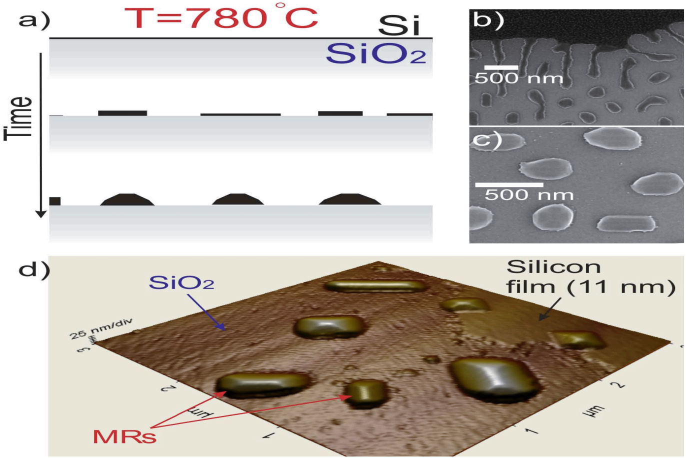
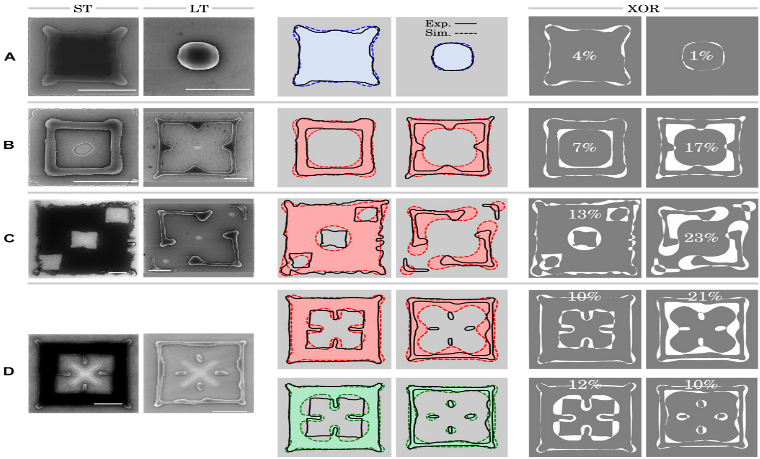


Figure: Abbarchi *et al.*, ACS Nano, 2014.

Physical Experiments

Dewetting of Ultrathin Silicon Films: [2] [3]



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[2] Naffouti *et al.*, Science Advances, 2017.

[3] Jiang-Bao*-Thompson-Srolovitz, Acta Mater., 2012.

Solid-State Dewetting of Thin Films

♠ Intrinsic Physics:

- Is driven by capillarity effects.
- Occurs through surface diffusion controlled mass transport.
- There exist moving contact lines in the thin film - substrate - vapor interface.
- Surface diffusion+Moving Contact Line.

♠ Applications:

- Play an important role in microelectronics processing.
- A common method to produce nano-particles.
- Catalyst for the growth of carbon nanotubes & semiconductor nanowires.

♠ Phenomena Observed from Experiments[4]:

- Pinch-off, Mass-shedding Instability, Geometric Complexity, Corner-induced Instability, Rayleigh Instability...
- Crystalline Anisotropy, Edge Faceting...

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[4] C.V. Thompson. Annu. Rev. Mater. Res., 2012.

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Surface Diffusion – From Physics

- Surface Diffusion: is a general process involving the motion of adatoms, molecules, and atomic clusters (adparticles) at solid material surfaces. (From Wikipedia, the free encyclopedia).
- *The original work* by W.W. Mullins (J. Appl. Phys., 28:333, 1957).
  - **The free energy**: $W = \int_\Gamma \gamma_0 \, d\Gamma$, where Γ is a moving surface.
  - **Chemical potential**: $\mu = \frac{\delta W}{\delta N} = \Omega_0 \frac{\delta W}{\delta \Gamma} = \Omega_0 \gamma_0 \mathcal{H}$, where $\mathcal{H}$ is the mean curvature of the surface.
  - **Constitutive relation**: $\mathbf{J} = -\frac{D_s \nu}{k_B T} \nabla_s \mu$.
  - **Kinematic relation**: $V_n = -\Omega_0 (\nabla_s \cdot \mathbf{J})$
- *Surface Diffusion equation*:

$$V_n = B \Delta_s \mathcal{H}, \quad \text{where} \quad B = \frac{D_s \nu \gamma_0 \Omega_0^2}{k_B T}.$$

Surface Diffusion – From Mathematics

Given a energy (shape) functional, e.g. $W(\Gamma) = \int_\Gamma d\Gamma$, then we can define:

- Mean curvature flow (MCF): (L^2-*Gradient Flow*)

$$V_n = -\mathcal{H},$$

- Surface diffusion flow (SDF): (H^{-1}-*Gradient Flow*)

$$V_n = \Delta_s \mathcal{H}.$$

- MCF and SDF both dissipate the energy functional, and SDF conserves the total volume while MCF does not.
- Analogous to: Allen-Cahn and Cahn-Hilliard equations, but design numerical schemes which can preserve the structure, dissipation and properties seems much more difficult!

Challenges for Modeling & Numerics

(1). Surface Diffusion Problem

- Mean Curvature Flow: $V_n = -\mathcal{H}$, e.g., **Grain growth, Grain boundary migration, Cellular Structure ...**
- Surface Diffusion: $V_n = \Delta_s \mathcal{H}$, e.g., Solid-state dewetting, **Sintering, Self-organized quantum dots ...**

(2). Moving Contact Line Problem

- widely studied in fluids by many researchers
- ... but still **rarely** known for solids.

(3). Multi-Physics, Multi-Scale Problem

- The substrate may be rough and heterogeneous.
- It includes many other **physical effects**, e.g., grain boundary, elasticity, alloy ...

Modeling Techniques

(1). Sharp-Interface Approaches

- Isotropic (*Srolovitz-Safran, JAP, 1986*)
- Weakly anisotropic (*Jiang et al., PRB, 2015*)
- Strongly anisotropic (*Jiang et al., Scripta Mater.,2016*)

(2). Phase-Field Approaches

- Isotropic (*Jiang et al., Acta Mater., 2012.*)
- Weakly anisotropic (*Dziwnik-Munch-Wagner, Nonlinearity, 2017*)
- Strongly anisotropic (still not ?)

(3). Kinetic Monte Carlo Approaches, e.g.,

- *Dufay-Pierre-Louis, Phys. Rev. Lett., 2011.*

(4). Discrete Chemical Potential Approaches, e.g.,

- *Carter-Roosen-Cahn-Taylor, Acta Metall. Mater., 1995.*
- *Klinger-Rabkin, Acta Mater., 2012.*

(5). Other Approaches, e.g., level set, VOF, IBM ... ???

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Total Free Energy in 2D

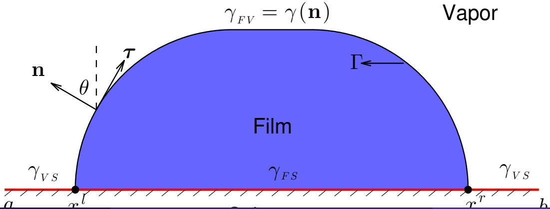
- The total Interfacial Free Energy of the system: (Bao-Jiang*-Srolovitz-Wang, SIAM J. Appl. Math., 2017)

$$W(\Gamma) = \int_\Gamma \gamma(\mathbf{n})\, d\Gamma + \underbrace{(\gamma_{FS} - \gamma_{VS}) \cdot (x_c^r - x_c^l)}_{\textbf{Wall Energy}}$$

$\gamma_{FV} = \gamma(\mathbf{n})$: The energy density of the film-vapor interface,

γ_{VS} : The energy density between the vapor-substrate,

γ_{FS} : The energy density between the film-substrate.



Thermodynamic variation in 2D

- Introduce a family of perturbed curves $\{\Gamma^\epsilon\}$, $\rho \in I = [0,1]$

$$\Gamma^\epsilon = \mathbf{X}(\rho, \epsilon) : I \times [0, \epsilon_0] \to \mathbb{R}^2,$$

- Introduce a smooth deformation vector-field:

$$\mathbf{V}(\rho, \epsilon) = \frac{\partial \mathbf{X}(\rho, \epsilon)}{\partial \epsilon}, \qquad \forall \epsilon \in [0, \epsilon_0],$$

- For the shape functional $W(\Gamma)$, define its first variation with respect to any smooth deformation vector-field V as

$$\delta W(\Gamma; \mathbf{V}) = \lim_{\epsilon \to 0} \frac{W(\Gamma^\epsilon) - W(\Gamma)}{\epsilon}.$$

Thermodynamic variation in 2D

Theorem (First variation)

The first variation of the free energy functional defined above in solid-state dewetting problems with respect to any smooth deformation vector-field $\mathbf{V}$ *is written as* [a]*:*

$$\delta W(\Gamma;\mathbf{V}) = \int_\Gamma \left[-(\partial_s\boldsymbol{\xi})^\perp\cdot\mathbf{n}\right](\mathbf{V}_0\cdot\mathbf{n})\,ds + \Big[(\xi_2-\sigma)(\mathbf{V}_0\cdot\mathbf{e}_1)\Big]\Big|_{s=0}^{s=L},$$

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[a] Jiang-Zhao, submitted, 2018

where $\perp$ represents the clockwise rotation of a vector by 90 degrees, $\boldsymbol{\xi}=(\xi_1,\xi_2)$ is the Cahn-Hoffman vector, $\sigma=\gamma_{VS}-\gamma_{FS}$, and $\mathbf{e}_1=(1,0)$ represents the unit vector along the x-coordinate (or the substrate line). The deformation velocity is denoted as $\mathbf{V}_0=\mathbf{V}(\rho,\epsilon=0)$, and $\mathbf{V}_0\cdot\mathbf{n}$ represents *the deformation velocity along the outer normal direction of the interface*.

Cahn-Hoffman ξ-vector

♠ Cahn-Hoffman $\boldsymbol{\xi}$-vector (D.W. Hoffman and J.W. Cahn, 1974; J.E. Taylor, 1992).

$$\boldsymbol{\xi}(\mathbf{n}) = \nabla\hat{\gamma}(\mathbf{p})\Big|_{\mathbf{p}=\mathbf{n}}, \quad \hat{\gamma}(\mathbf{p}) = |\mathbf{p}|\gamma(\frac{\mathbf{p}}{|\mathbf{p}|}), \quad \forall \mathbf{p} \in \mathbb{R}^3 \backslash \{\mathbf{0}\}.$$

Cahn-Hoffman ξ-vector

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- ◇ Mathematical construction of the **equilibrium shape** : $\boldsymbol{\xi}$-plot.
- ◇ In the isotropic case (i.e., $\gamma(\mathbf{n}) \equiv \gamma_0$), $\boldsymbol{\xi} = \gamma_0\mathbf{n}$.
- ◇ In 2D, it can be expressed as:

$$\boldsymbol{\xi}(\mathbf{n}) = \gamma(\theta)\mathbf{n} - \gamma'(\theta)\boldsymbol{\tau}.$$

- ◇ In 3D, it can be expressed as:

$$\boldsymbol{\xi}(\mathbf{n}) = \gamma(\theta,\phi)\mathbf{n} + \frac{\partial\gamma(\theta,\phi)}{\partial\theta}\boldsymbol{\tau}_{\boldsymbol{\theta}} + \frac{1}{\sin\theta}\frac{\partial\gamma(\theta,\phi)}{\partial\phi}\boldsymbol{\tau}_{\boldsymbol{\phi}},$$

Sharp-interface model in 2D

♠ The variation with respect to Γ, x_c^l and x_c^r

$$\frac{\delta W}{\delta \Gamma} = -(\partial_s \boldsymbol{\xi})^\perp \cdot \mathbf{n}, \quad \frac{\delta W}{\delta x_c^l} = -(\xi_2|_{s=0} - \sigma), \quad \frac{\delta W}{\delta x_c^r} = \xi_2|_{s=L} - \sigma.$$

Sharp-interface model in 2D

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- ◇ Normal velocity for the curve: surface diffusion flow
  - Chemical potential: Gibbs-Thomson relation

$$\mu = \Omega_0 \frac{\delta W}{\delta \Gamma},$$

  - Constitutive relation & Kinematic relation

$$\mathbf{J} = -\frac{D_s \nu}{k_B\, T} \nabla_s \mu, \qquad V_n = -\Omega_0 (\nabla_s \cdot \mathbf{J}) = \frac{D_s \nu \Omega_0^2}{k_B\, T} \partial_{ss} \left[\frac{\delta W}{\delta \Gamma} \right].$$

- ◇ Relaxed contact angle condition: L^2-gradient flow

$$\frac{dx_c^l(t)}{dt} = -\eta \frac{\delta W}{\delta x_c^l}, \qquad \frac{dx_c^r(t)}{dt} = -\eta \frac{\delta W}{\delta x_c^r}.$$

Sharp-interface model in 2D

$$\begin{cases} \partial_t \mathbf{X} = \partial_{ss}\mu\, \mathbf{n}, \quad 0 < s < L(t), \quad t > 0, \\ \mu = -\left(\partial_s \boldsymbol{\xi}\right)^{\perp} \cdot \mathbf{n}, \qquad \boldsymbol{\xi} = \nabla\hat{\gamma}(\mathbf{p})\Big|_{\mathbf{p}=\mathbf{n}}; \end{cases}$$

subject to the boundary conditions:

(i) Contact point condition

$$y(0,t) = 0, \quad y(L,t) = 0, \quad t \geq 0;$$

(ii) Relaxed contact angle condition

$$\frac{dx_c^l}{dt} = \eta(\xi_2|_{s=0} - \sigma), \quad \frac{dx_c^r}{dt} = -\eta(\xi_2|_{s=L} - \sigma), \quad t \geq 0;$$

(iii) Zero-mass flux condition

$$\partial_s\mu(0,t) = 0, \quad \partial_s\mu(L,t) = 0, \quad t \geq 0.$$

The sharp-interface model

♠ Mass conservation

$$A(t) \equiv A(0) = \int_{\Gamma(0)} y_0(s)\partial_s x_0(s)\ ds, \qquad t \geq 0,$$

♠ Energy dissipation

$$\frac{d}{dt}W(t) = -\int_{\Gamma(t)} (\partial_s \mu)^2\ ds - \frac{1}{\eta}\Big[\Big(\frac{dx_c^l}{dt}\Big)^2 + \Big(\frac{dx_c^r}{dt}\Big)^2\Big] \leq 0.$$

Solid-State Dewetting in 3D

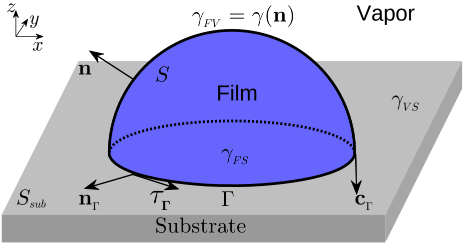

Figure: Solid-state dewetting in 3D

- S: film-vapor interface,
  Γ: moving contact line,
- $\mathbf{n}$: unit normal vector of S,
  $\mathbf{n}_\Gamma$: unit normal vector of Γ,
- $\mathbf{c}_\Gamma$: unit co-normal vector,
- γ_{FV}, γ_{FS} and γ_{VS}: surface energy densities between film/vapor, film/substrate and vapor/substrate interfaces.

♠ The total interfacial energy in 3D[5]:

$$W = W_{\mathrm{f}} + W_{\mathrm{sub}} = \iint_S \gamma(\mathbf{n})\, dS + (\gamma_{FS} - \gamma_{VS}) A(\Gamma).$$

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[5] Bao-Jiang*-Zhao, in preparation, 2018

Thermodynamic variation in 3D

- **Basic Assumptions**:
  (1). γ_{FS}, γ_{VS} are two constants, $\gamma_{FV} = \gamma(\mathbf{n})$;
  (2). The perturbation velocity field $\mathbf{V}$ is smooth;
  (3). The perturbation on the contact line must make the contact line move on the substrate.

- First variation of the substrate energy:

$$\delta W_{\mathrm{sub}}(S;\mathbf{V}) = (\gamma_{FS} - \gamma_{VS}) \int_{\Gamma} \mathbf{V}_0 \cdot \mathbf{n}_{\Gamma}\, d\Gamma.$$

- First variation of the film-vapor interface energy:

$$\delta W_{\mathrm{f}}(S;\mathbf{V}) = \iint_{S} (\nabla_S \cdot \boldsymbol{\xi})(\mathbf{V}_0 \cdot \mathbf{n})\, dS + \int_{\Gamma} \mathbf{V}_0 \cdot \mathbf{c}_{\Gamma}^{\gamma}\, d\Gamma,$$

where $\boldsymbol{\xi}$ is the Cahn-Hoffman vector,
and $\mathbf{c}_{\Gamma}^{\gamma} = (\boldsymbol{\xi} \cdot \mathbf{n})\mathbf{c}_{\Gamma} - (\boldsymbol{\xi} \cdot \mathbf{c}_{\Gamma})\mathbf{n}$.

Sharp-Interface Model in 3D

- The dimensionless sharp-interface model in 3D:

$$\begin{cases} \partial_t \mathbf{X} = (\triangle_S \mu)\,\mathbf{n}, \quad t > 0, \\ \mu = \nabla_S \cdot \boldsymbol{\xi}, \quad \boldsymbol{\xi} = \nabla \hat{\gamma}(\mathbf{p})\Big|_{\mathbf{p}=\mathbf{n}}, \end{cases}$$

- subject to the following Boundary Conditions:

(1). **Contact line condition**:

$$\mathbf{X}_\Gamma \subseteq \text{XOY Plane}, \quad t > 0,$$

(2). **Relaxed contact angle condition**:

$$\partial_t \mathbf{X}_\Gamma = -\eta(\mathbf{c}_\Gamma^\gamma \cdot \mathbf{n}_\Gamma - \sigma)\mathbf{n}_\Gamma, \quad t > 0,$$

where $\sigma = \frac{\gamma_{VS} - \gamma_{FS}}{\gamma_0}$;

(3). **Zero-mass flux condition**:

$$(\nabla_S \mu \cdot \mathbf{c}_\Gamma)\Big|_\Gamma = 0, \quad t > 0.$$

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Numerical Challenges

High order and *nonlinear* geometric PDEs with complex boundary conditions:

- ♠ "Marker-particle" method (explicit finite difference scheme) (Wong *et al.* Acta Mater., 2000; JCP, 2009...)
  1. First update the inner "marker" points by the explicit finite difference scheme;
  2. Then according to the boundary conditions, update the contact points;
  3. Do polynomial interpolation and redistribute the "marker" points uniformly with respect to arc length.

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- ♠ Parametric finite element method for solving geometric PDEs (E.Bansch *et al.*, JCP, 2004; C.M. Elliott *et al.*, Acta Numer., 2005; J.W. Barrett *et al.*, SISC, 2007...)

Variational formulation

- parameterize the curves as

$$\Gamma(t) = \mathbf{X}(\rho, t) : I \times [0, T] \to \mathbb{R}^2,$$

where the time-independent spatial variable $\rho \in I$, and I denotes a fixed reference spatial domain, say $I := [0, 1]$.

- L^2 inner product

$$\left\langle u, v \right\rangle_\Gamma := \int_{\Gamma(t)} u(s)v(s)\, ds = \int_I u(s(\rho, t))v(s(\rho, t))|\partial_\rho \mathbf{X}|\, d\rho.$$

- Functional space

$$H^1_{a,b}(I) = \{u \in H^1(I) : u(0) = a, u(1) = b\}.$$

Variational formulation

- Re-formulate the PDEs [6]:

$$\begin{aligned} \partial_t \mathbf{X} = \partial_{ss}\mu\, \mathbf{n} \quad &\Rightarrow \quad \partial_t \mathbf{X}\cdot\mathbf{n} = \partial_{ss}\mu. \\ \mu = -(\partial_s \boldsymbol{\xi})^{\perp}\cdot\mathbf{n} \quad &\Rightarrow \quad \mu\,\mathbf{n} = -(\partial_s \boldsymbol{\xi})^{\perp}. \end{aligned}$$

Find $\mathbf{X} \in H_{a,b}(I)\times H_0^1(I)$, $\mu \in H^1(I)$ with $a = x_c^l(t) \le x_c^r(t) = b$,

$$\big\langle \partial_t \mathbf{X},\ \varphi\mathbf{n}\big\rangle_\Gamma + \big\langle \partial_s \mu,\ \partial_s\varphi\big\rangle_\Gamma = 0, \quad \forall\, \varphi \in H^1(I),$$

$$\big\langle \mu\mathbf{n},\ \boldsymbol{\omega}\big\rangle_\Gamma - \big\langle \boldsymbol{\xi}^{\perp},\ \partial_s\boldsymbol{\omega}\big\rangle_\Gamma = 0, \quad \forall\, \boldsymbol{\omega} \in H_0^1(I)\times H_0^1(I),$$

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[6] Bao-Jiang*-Wang-Zhao, JCP, 2017

Semi-implicit PFEM

- **Finite element spaces**

$$V^h := \{u \in C(I):\ u|_{I_j} \in P_1, \quad \forall j = 1, 2, \ldots, N\} \subseteq H^1(I),$$
$$\mathcal{V}^h_{a,b} := \{u \in V^h:\ u(0) = a,\ u(1) = b\} \subseteq H^1_{a,b}(I).$$

- **Inner product**

$$\big\langle u,\ v\big\rangle^h_{\Gamma^m} = \frac{1}{2}\sum_{j=1}^{N}\Big|\mathbf{X}^m(\rho_j) - \mathbf{X}^m(\rho_{j-1})\Big|\Big[\big(u\cdot v\big)(\rho_j^-) + \big(u\cdot v\big)(\rho_{j-1}^+)\Big].$$

Find $\mathbf{X}^{m+1} \in \mathcal{V}^h_{a,b} \times \mathcal{V}^h_0$, $\quad \mu^{m+1} \in V^h$ such that

$$\Big\langle \frac{\mathbf{X}^{m+1} - \mathbf{X}^m}{t_{m+1} - t_m},\ \varphi_h \mathbf{n}^m\Big\rangle^h_{\Gamma^m} + \big\langle \partial_s \mu^{m+1},\ \partial_s \varphi_h\big\rangle^h_{\Gamma^m} = 0, \quad \forall\, \varphi_h \in V^h,$$

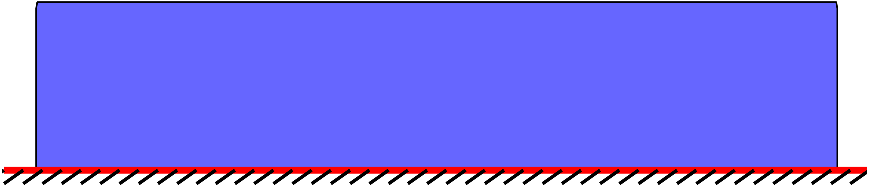
$$\big\langle \mu^{m+1}\mathbf{n}^m,\ \boldsymbol{\omega}_\mathbf{h}\big\rangle^h_{\Gamma^m} - \big\langle [\boldsymbol{\xi}^{m+\frac{1}{2}}]^\perp,\ \partial_s \boldsymbol{\omega}_\mathbf{h}\big\rangle^h_{\Gamma^m} = 0, \quad \forall\, \boldsymbol{\omega}_h \in \mathcal{V}^h_0 \times \mathcal{V}^h_0,$$

◇ Linearization of $\boldsymbol{\xi}$

$$\boldsymbol{\xi}^{m+\frac{1}{2}} = \begin{cases} \gamma(\theta^m)\mathbf{n}^{m+1} - \gamma'(\theta^m)\boldsymbol{\tau}^{m+1}, & \text{if } \gamma = \gamma(\theta), \\ \gamma(\mathbf{n}^m)\mathbf{n}^{m+1} + (\boldsymbol{\xi}^m \cdot \boldsymbol{\tau}^m)\boldsymbol{\tau}^{m+1}, & \text{if } \gamma = \gamma(\mathbf{n}). \end{cases}$$

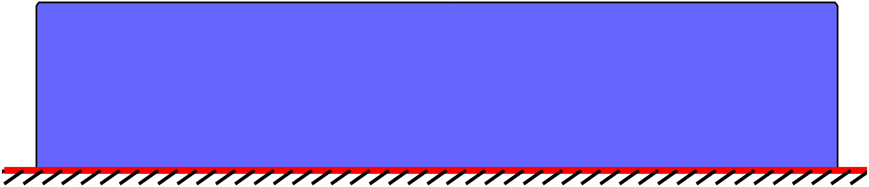
Isotropic, short island

$$L = 5, \beta = 0, \sigma = \cos(3\pi/4)$$



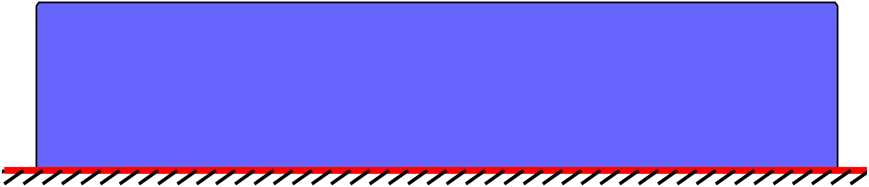
Weakly anisotropic, short island

$$L = 5, m = 4, \beta = 0.06, \sigma = \cos(3\pi/4)$$



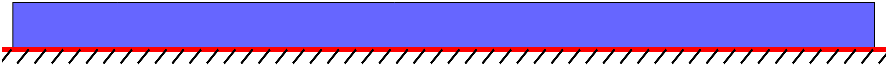
Strongly anisotropic, short island

$$L = 5, m = 4, \beta = 0.2, \sigma = \cos(3\pi/4)$$

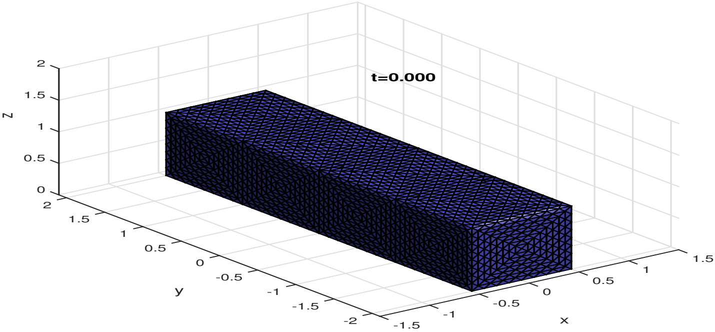


Weakly anisotropic, long island

$$L = 60, m = 4, \beta = 0.06, \sigma = \cos(5\pi/6)$$



Simulation Results, 3D Case



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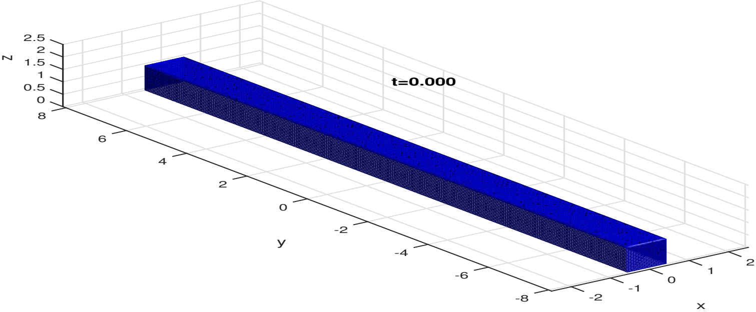

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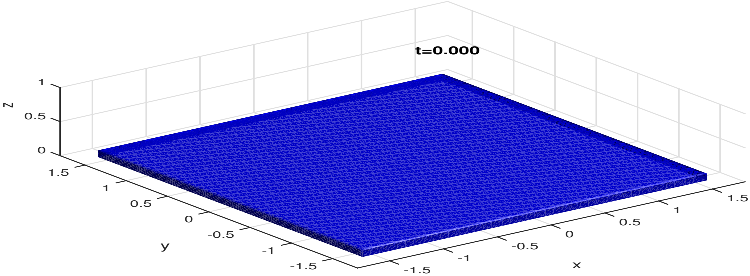

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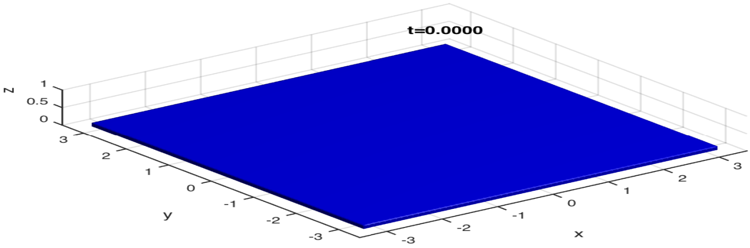
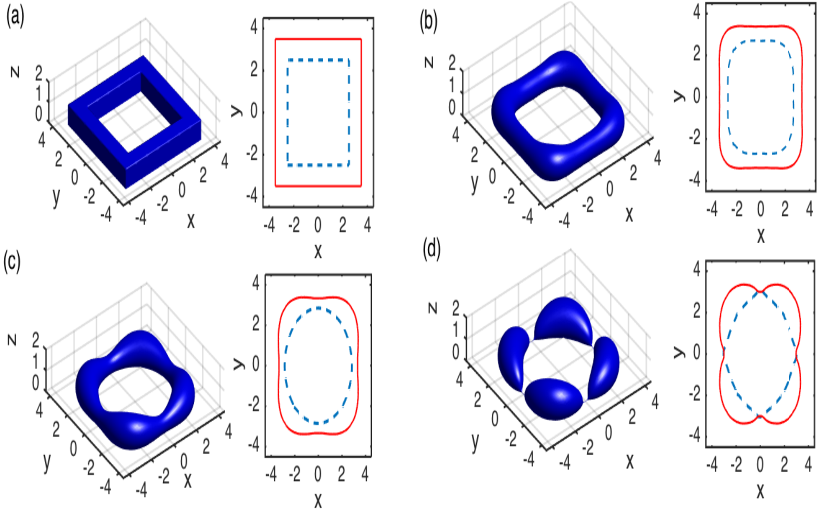
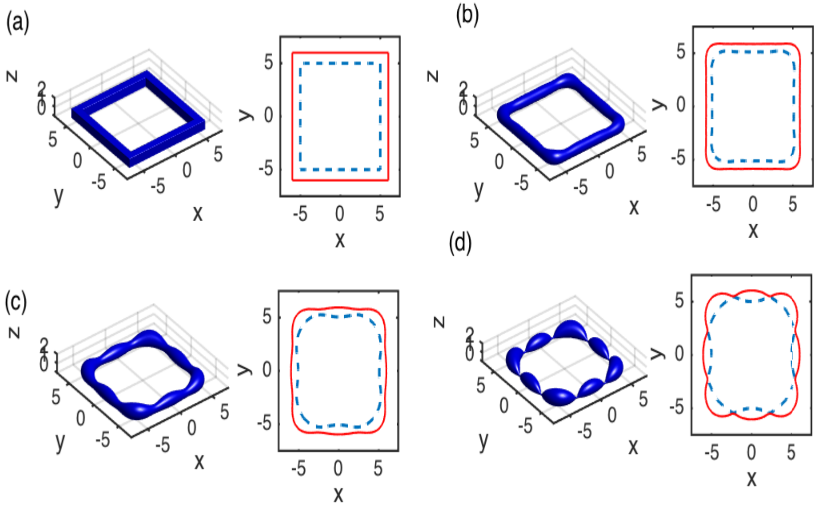


Simulation Results, 3D – Rayleigh Instability



Simulation Results, 3D – Rayleigh Instability



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6. Summary

Onsager's variational Principle

- The Onsager's principle was first proposed by Lars Onsager in his seminal work in 1931, on the reciprocal relations in irreversible processes.
- We consider *an isothermal system*, and let $\alpha = (\alpha_1, \alpha_2, \dots, \alpha_n)$ be a set of state parameters which can be used to determine the non-equilibrium state of a system. It satisfies:

$$\frac{d\alpha_i}{dt} = -\mu_{ij}\frac{\partial W}{\partial \alpha_j},$$

where $W(\alpha)$ is the total free energy of the system, μ_{ij} is the kinetic coefficient.

- *Onsager's reciprocal relation*:

$$\mu_{ij} = \mu_{ji}$$

- There exists ζ_{ij} (friction coefficient), satisfying $\zeta_{ij} = \zeta_{ji}$ and $\zeta_{ik}\mu_{kj} = \delta_{ij}$. So we have:

$$\zeta_{ij}\frac{d\alpha_j}{dt} = -\frac{\partial W}{\partial \alpha_i},$$

- The above equation can be obtained by minimizing the Rayleighian:

$$\mathcal{R}(\alpha, \dot{\alpha}) = \frac{\partial W}{\partial \alpha_i}\dot{\alpha}_i + \frac{1}{2}\zeta_{ij}\dot{\alpha}_i\dot{\alpha}_j,$$

with respect to the rates $\{\dot{\alpha}_i\}$.

Application to Interface Evolution Problems

- Let $\alpha(t) = (\alpha_1(t), \alpha_2(t), \ldots, \alpha_n(t))$ be a set of parameters which can be used to **determine the position of the boundaries**.

$$\mathcal{R} = \dot{W}(\alpha, \dot{\alpha}) + \Phi(\dot{\alpha}, \dot{\alpha}),$$

$$\mathcal{R}(\alpha, \dot{\alpha}) = \frac{\partial W}{\partial \alpha_i}\dot{\alpha}_i + \frac{1}{2}\zeta_{ij}\dot{\alpha}_i\dot{\alpha}_j,$$

- The minimum condition of $\mathcal{R}$ with respect to the rates $\{\dot{\alpha}_i\}$ gives the following kinetic equation:

$$-\frac{\partial W}{\partial \alpha_i} = \zeta_{ij}\dot{\alpha}_j,$$

  which describes a force balance between **the reversible force** $-\frac{\partial W}{\partial \alpha_i}$ and **the dissipative force** $\frac{\partial \Phi}{\partial \dot{\alpha}_i}$ which is linear in the rates $\{\dot{\alpha}_i\}$.

- $\Phi(\dot{\alpha}, \dot{\alpha})$ is the energy dissipation function which is defined as half the change rate of free energy dissipation, and the friction coefficients ζ_{ij} forming a symmetric and positive definite matrix.

Why does a reduced-order variational model work?

- The original surface diffusion equation can actually be derived from Onsager's variational principle
- The rate of change of the free energy $\dot{W}$:

$$\begin{aligned}\dot{W} &= \iint_S \gamma_0\,\mathcal{H}\,v_n\,dS \\ &= \iint_S \gamma_0\,\mathcal{H}\left[-\Omega_0\nabla_s\cdot(\nu\mathbf{V})\right]dS \\ &= \iint_S \mu\left[-\nabla_s\cdot(\nu\mathbf{V})\right]dS \\ &= \iint_S (\nabla_s\mu)\cdot(\nu\mathbf{V})\,dS,\end{aligned}$$

- The dissipation function Φ is defined as:

$$\Phi = \frac{1}{2}\iint_S \frac{k_B T}{D_s}|\mathbf{V}|^2\nu\,dS,$$

  where $k_B T/D_s$ is the friction coefficient for diffusing atoms.
- Defining a Rayleighian $\mathcal{R} = \dot{W} + \Phi$ and minimizing $\mathcal{R}$ with respect to the velocity $\mathbf{V}$, we obtain the constitutive equation for surface diffusion.

A Simple Example: Solid-state dewetting of toroidals

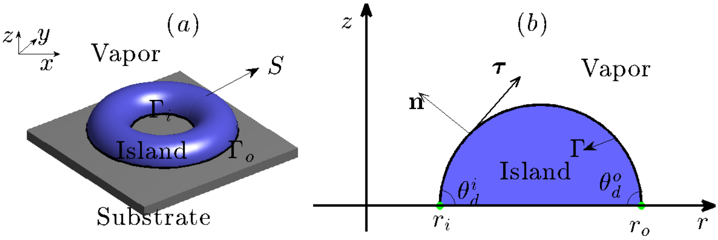

Figure: (a) A schematic illustration of the solid-state dewetting of a toroidal island on a flat, rigid substrate; (b) the cross-section profile (i.e., denoted as Γ) of the toroidal solid island in a cylindrical coordinate system (r, z), with r_i, r_o representing the inner and outer contact points, respectively.

- Axisymmetric geometry: $S = (r(s,t)\cos\varphi,\ r(s,t)\sin\varphi,\ z(s,t))$, $\varphi \in [0, 2\pi]$. (arXiv:1806:08272, 2018)
- A full sharp-interface model has been proposed (arXiv:1711.02402, 2017).

A Reduced Model by the Onsager's Principle

Some assumptions:

- ◇ The cross-section profile of the island is always a circular arc.
- ◇ The contact line does not dissipate any free energy when it moves, i.e., $\theta_i \in [0, \pi]$ is fixed.

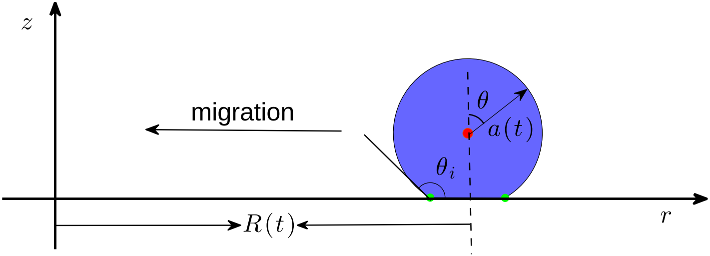

Figure: A cross-section profile of the toroid, where $a := a(t)$ is the radius of the circle, $R := R(t)$ is the distance between the origin of the circle and the z-axis.

A Reduced Model by the Onsager's Principle

- ◇ The cross-section profile can be expressed as:

$$\begin{cases} r(\theta, t) = R(t) + a(t)\sin\theta, \\ z(\theta, t) = a(t)(\cos\theta - \cos\theta_i), \end{cases} \quad \theta \in [-\theta_i, \theta_i].$$

- ◇ The volume is conserved, i.e., $V_0 = \pi R a^2 (2\theta_i - \sin 2\theta_i)$.
- ◇ $\dot{W}$ can be written as:

$$\dot{W} = \gamma_0 \sqrt{\pi V_0 (2\theta_i - \sin 2\theta_i)}\, R^{-\frac{1}{2}} \dot{R}.$$

- ◇ The dissipation function Φ can be written as:

$$\Phi = \frac{k_B T}{D_s \nu \Omega_0^2} \pi R a^3 \dot{R}^2 [g(\theta_i) + \mathcal{O}(\delta^2)],$$

where we assume $\delta = \frac{a}{R}$ is a small parameter, and $g(\theta_i) = \theta_i(2 + \cos 2\theta_i) - \frac{3}{2}\sin 2\theta_i$.

- ◇ Minimization of the Rayleighian $\mathcal{R} = \dot{W} + \Phi$ with respect to the rate $\dot{R}$ yields the kinetic equation:

$$v = -\dot{R}(t) \approx C(\theta_i)\,\frac{B\gamma_0}{V_0},$$

where $B = \frac{D_s \nu\, \Omega_0^2}{k_B T}$ and $C(\theta_i) = \frac{\pi(2\theta_i - \sin 2\theta_i)^2}{2\, g(\theta_i)}$.

Model Validation

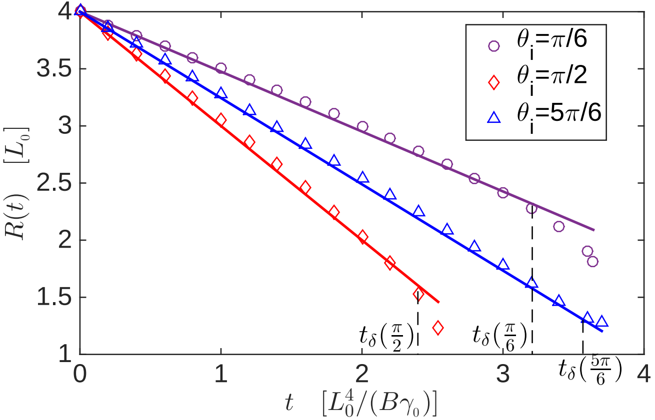

Figure: Comparisons between the numerical results of $R(t)$ by solving the full sharp-interface model and the predicted formula of $R(t)$ by the reduced model, where the symbols (“circles”, “rhombus” and “triangles”) are the numerical results obtained from the solving the full model and the solid lines are the predicted formula under different isotropic Young angles.

Model Validation

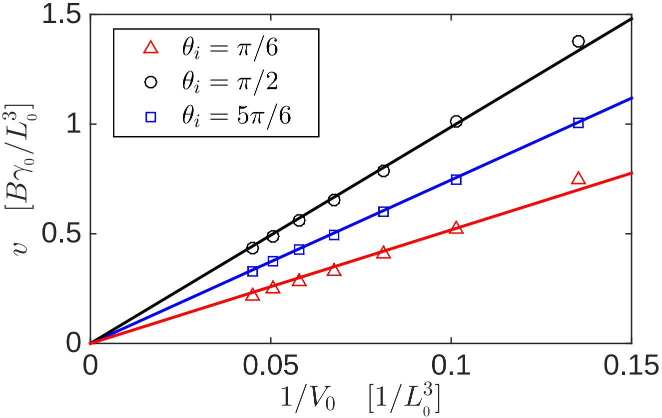

Figure: Comparisons between the numerical migration speed obtained from solving the full sharp-interface model (shown by circles, rhombuses or triangles) and the theoretical migration speed given by the reduced model (shown in solid line) as a function of the initial volume V_0 under different isotropic Young angles $\theta_i = \pi/6, \pi/2$ and $5\pi/6$.

Model Validation

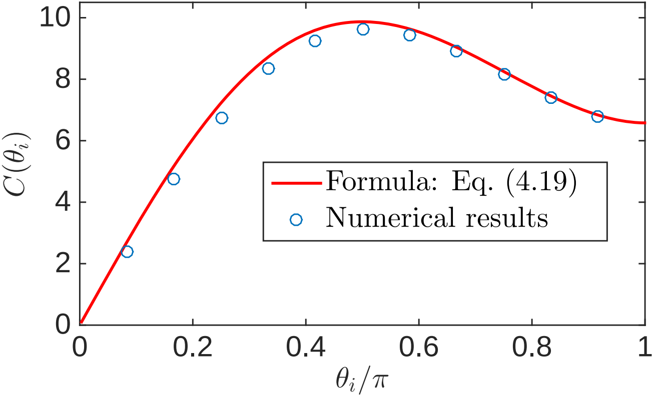

Figure: Comparisons between the fitting values of $C(\theta_i)$ (shown in "circles") obtained by numerically solving the full sharp-interface model and the analytical values given by the reduced model (shown in red solid line).

Outline

1. Introduction
2. What is surface diffusion? – Physics and Mathematics
3. How to model it? – Thermodynamic variation
4. How to numerically solve it? – PFEM
5. How to analyse it? – Onsager's variational principle
6. Summary

Summary and On-going Works

Summary:

- Propose thermodynamic variation for understanding the models.
- Propose a parametric FEM for solving sharp-interface models.
- Demonstrate that the Onsager's principle is a powerful tool for describing and approximating surface diffusion controlled problems.

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- Propose thermodynamic variation for understanding the models.
- Propose a parametric FEM for solving sharp-interface models.
- Demonstrate that the Onsager's principle is a powerful tool for describing and approximating surface diffusion controlled problems.

On-going Works:

- ◇ Include more physical effects (e.g., elasticity and grain boundary) into the models.
- ◇ Reduced Models by using Onsager principle & Mathematical analysis.
- ◇ Develop accurate and efficient numerical methods for solving 3D solid-state dewetting problems.
- ◇ Compare with experiments & guide new experiments.

Thank You for Your Attention!