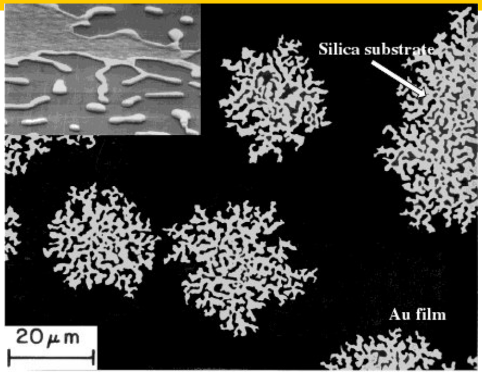
Modeling and Simulation for Solid-State Dewetting Problems



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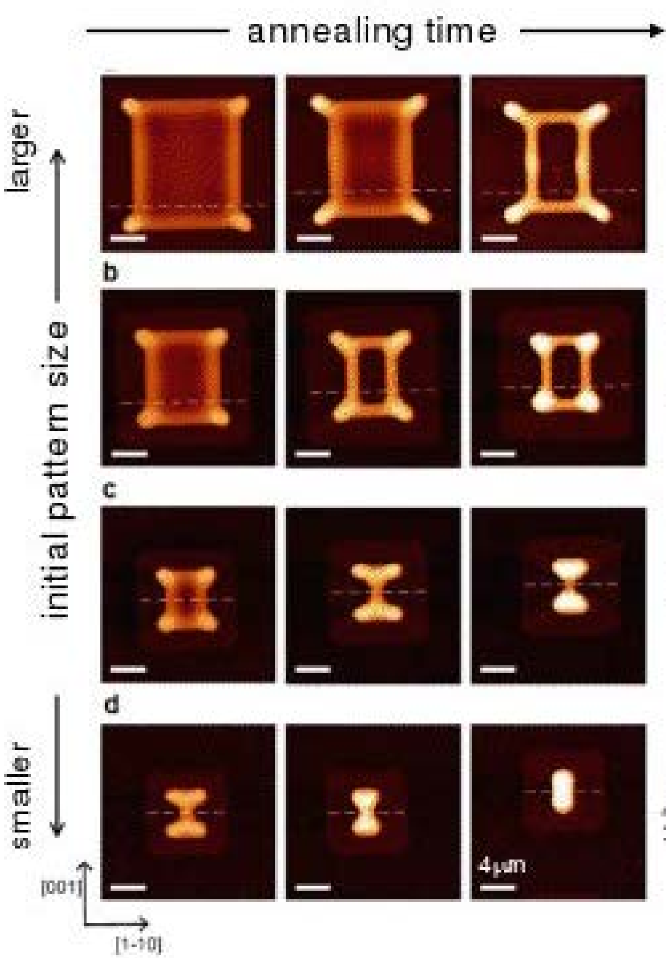
Outline

- Motivation
- Thermodynamic variation & equilibrium
- Sharp Interface models (SIMs)
  - Isotropic/weak anisotropic surface energy
  - Numerical methods & results in 2D
  - Extension to strong/cusped surface energy, to curved substrate & to 3D
- Phase field/Diffuse interface model
  - Mathematical model
  - Numerical methods & results in 3D
- Conclusion & future works

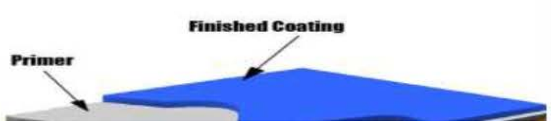
Patterned Ni(110) square patche

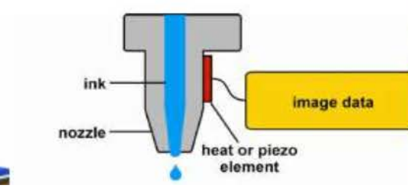


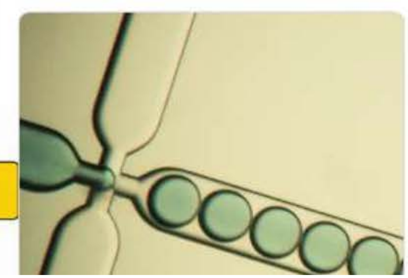
Interfaces are ubiquitous in nature and our daily life





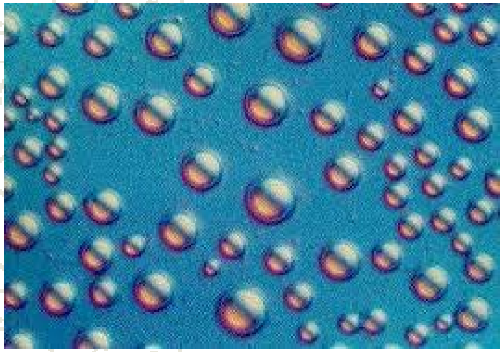


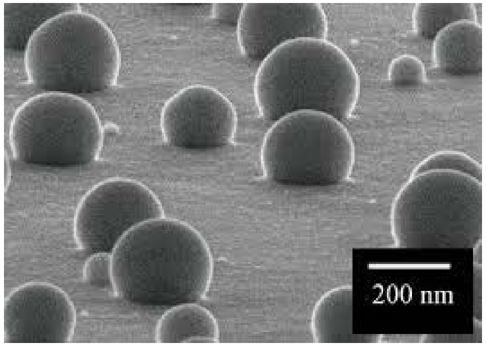


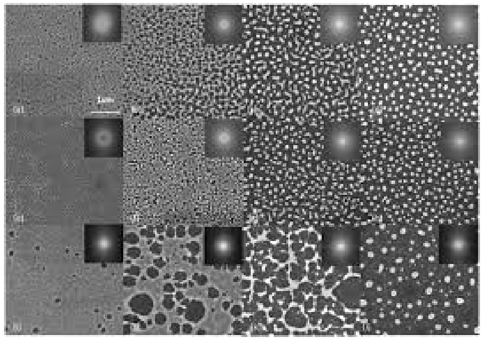
Wetting / Dewetting in Fluid Mechanics

- **Wetting**– spread of a liquid on a substrate

- **Dewetting** – the rupture of a thin liquid film on a substrate

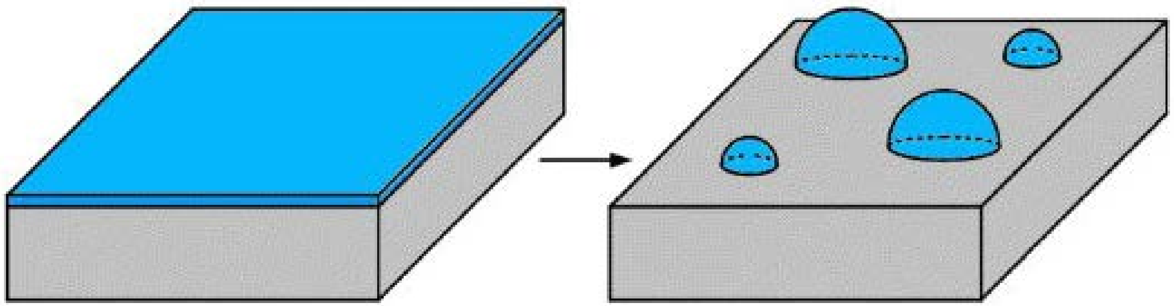




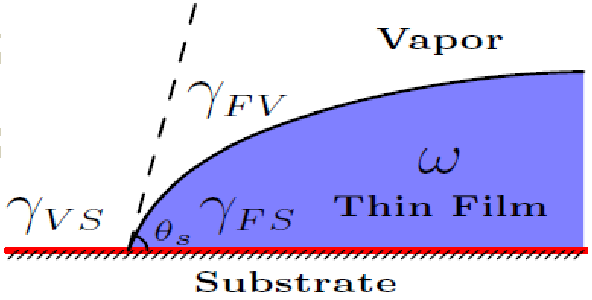


Solid-State Dewetting of Thin Films

- Most thin films are metastable in as-deposited state & dewet to form particles



- This occurs when the temperature is high enough for surface self diffusion, which can be well below the film's melting temperature

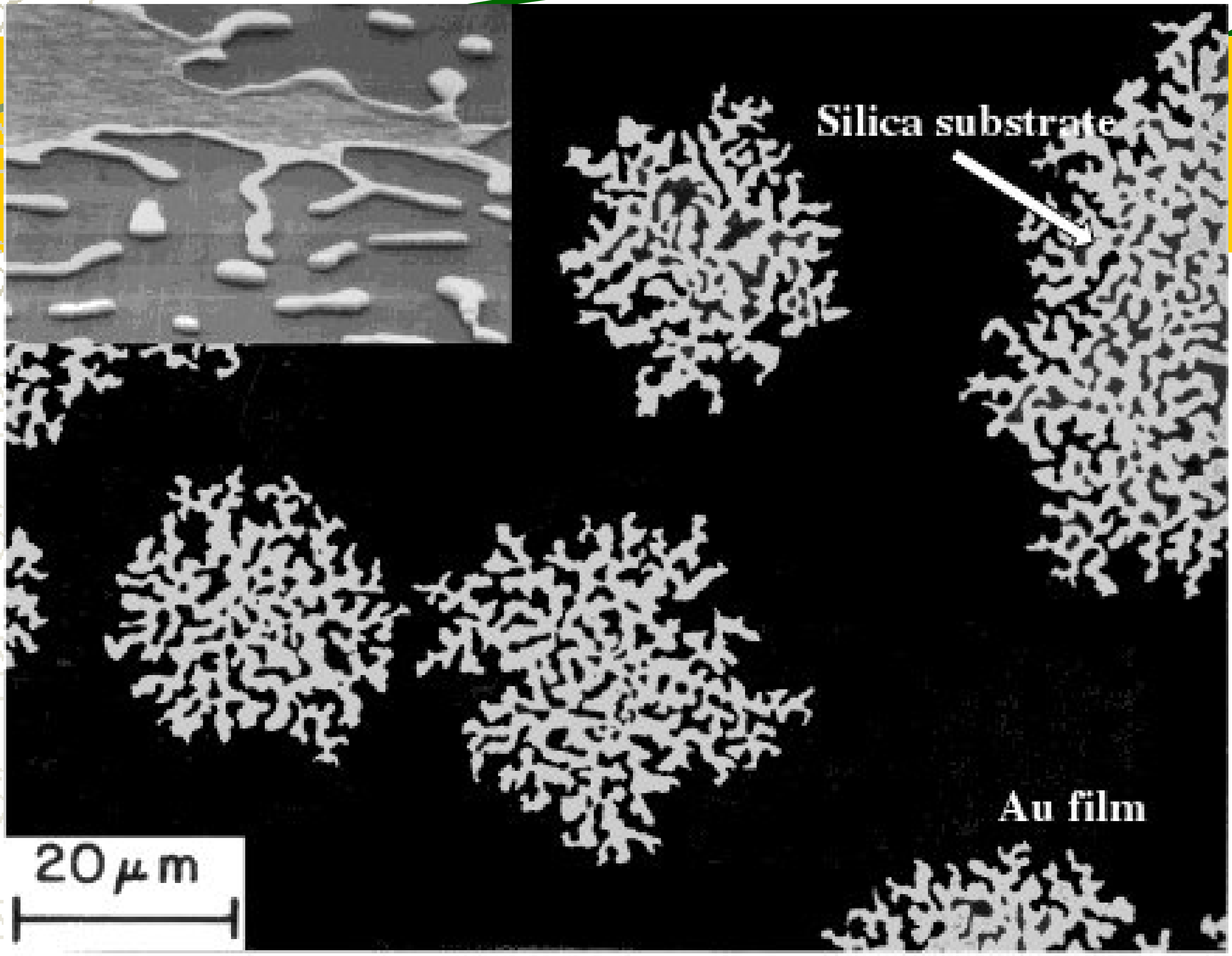


$$\gamma_{VS} = \gamma_{FS} + \gamma_{FV}\cos(\theta) \text{ -- Young, 1805}$$

γ_{VS} : substrate free surface energy

γ_{FV} : film free surface energy

γ_{FS} : film-substracte interface energy



Dewetting on a flat substrate

[1] E. Jiran & C. V. Thompson, Journal of Electronic Materials, 19 (1990), pp. 1153-1160.

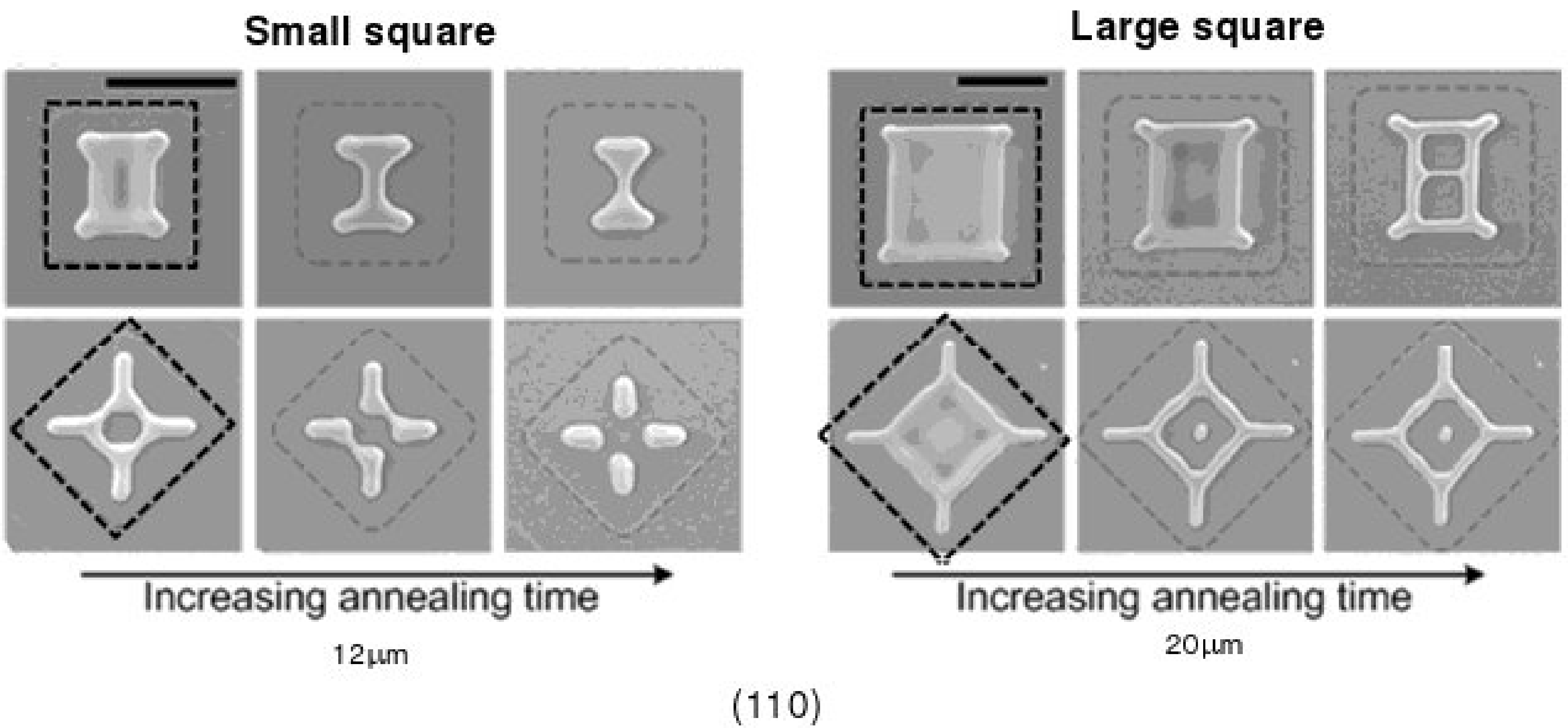
[2] E. Jiran & C. V. Thompson, Thin Solid Films, 208 (1991), pp. 23-28.

Solid-State Dewetting Problems

- Solid-state dewetting
  - Is driven by capillarity effects
  - Occurs through surface diffusion controlled mass transport
  - Belongs to capillarity-controlled interface/surface evolution problems
  - Surface diffusion + contact line migration
- Applications of dewetting of thin films
  - Play an improtant role in micorelectronics processing
  - A common method to produce nanoparticles
  - Catalyst for the growth of carbon nanotubes & semiconductor nanowires
- Recent experiments -- [1]
  - Geometric complexity, capillarity-driven instabilities, faceting
  - Crystalline anisotropy, corner-induced instabilities, pinch-off, ….
- Wetting/dewetting in fluids: TZ Qian, XP Wang&P Sheng; W. Ren&W E, ...

[1] J. Ye & C.V. Thompson, Acta Mater. 23 (2011), 1567; 59 (2011), 582; Appl. Phys. Lett. 97 (2010), 071904

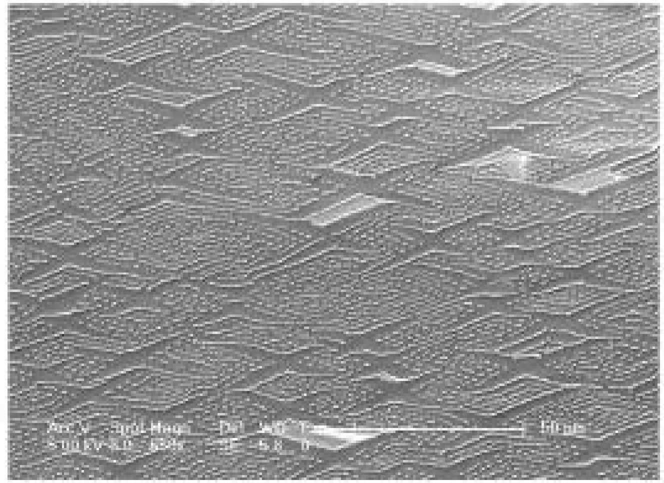
Effect of Size & Orientation of Pattern

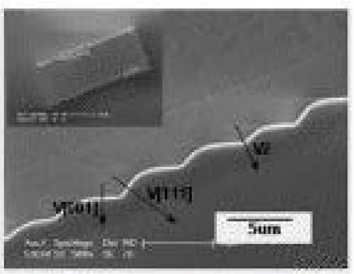


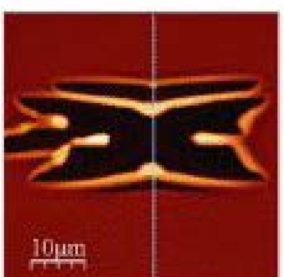
[1] J. Ye & C.V. Thompson, Adv. Mater., 23 (2011), 1567.

Dewetting of Patterned Single-Crystal Films

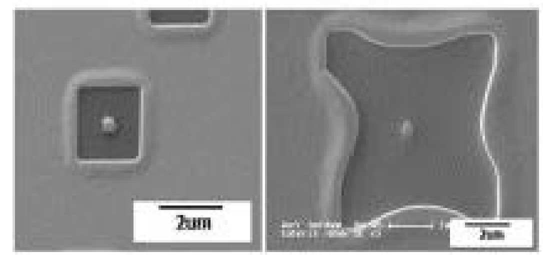
Complex pattern formation

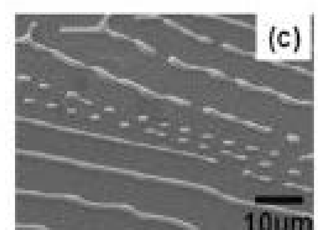






- Edge faceting
- Corner instability
- Mass shedding instability
- Rayleigh-like instability

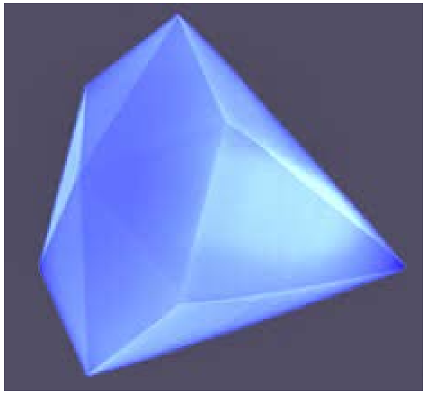




Equilibria of a Droplet (or Anisotropic Particle)

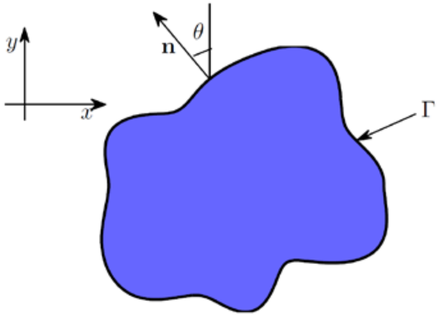
- A droplet (or solid particle--crystal)





- Equilibrium shape

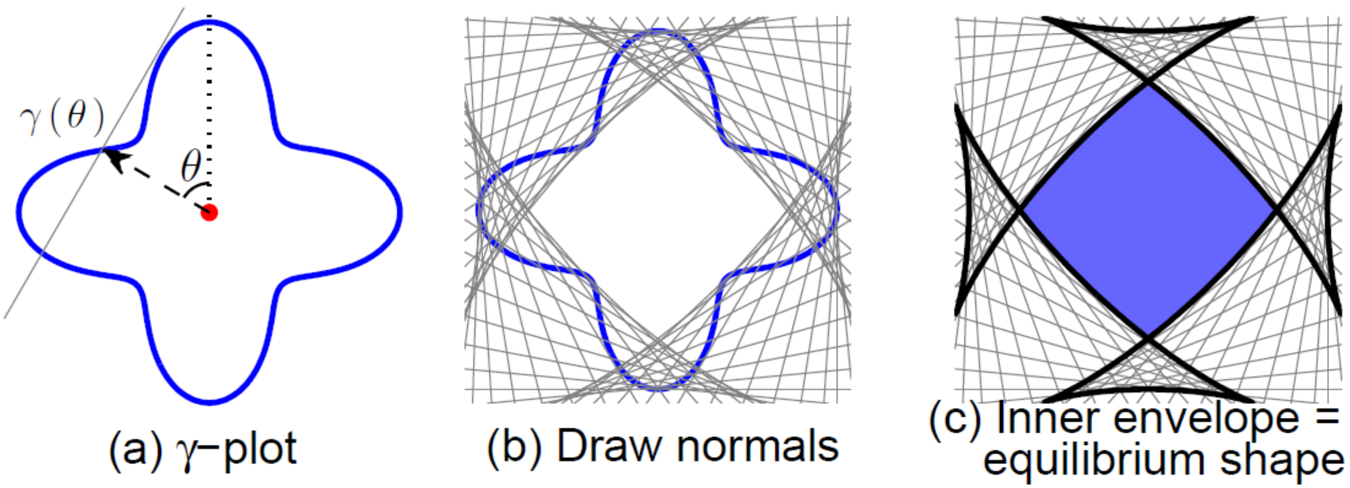
$$\min_{\Omega} W = \int_{\Gamma} \gamma(\theta)\, d\Gamma \quad \text{with} \quad |\Omega| = \text{const}$$



$\gamma := \gamma(\theta)$ (or $\gamma(\vec{n})$): surface energy density

Equilibria of a Droplet (or Anisotropic Particle)

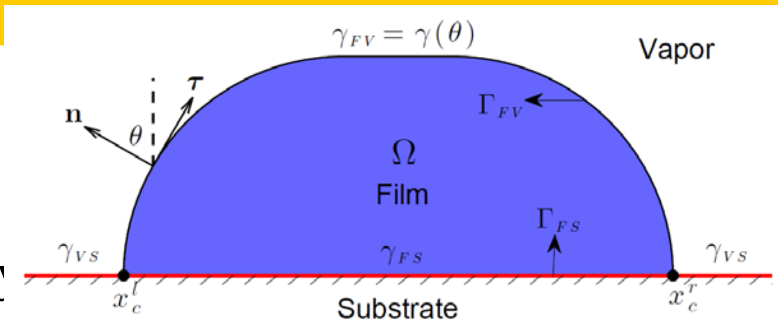
- Wulff construction – G. Wulff, 1901' $\gamma(\theta) = 1 + 0.3\cos(4\theta)$



- Justified by geometric measure theory– J.E. Taylor, 74';I. Fonseca &S. Muller, 91'

$$\begin{cases} x(\theta) = -\gamma(\theta)\sin\theta - \gamma'(\theta)\cos\theta, \\ y(\theta) = \gamma(\theta)\cos\theta - \gamma'(\theta)\sin\theta, \end{cases} \qquad 0 \le \theta \le 2\pi$$

Equilibria of a Particle (Droplet) on Substrate



- Total interfacial free energy

γ_{FV} -- film-vapor energy; γ_{SV} -- substrate-vapor energ

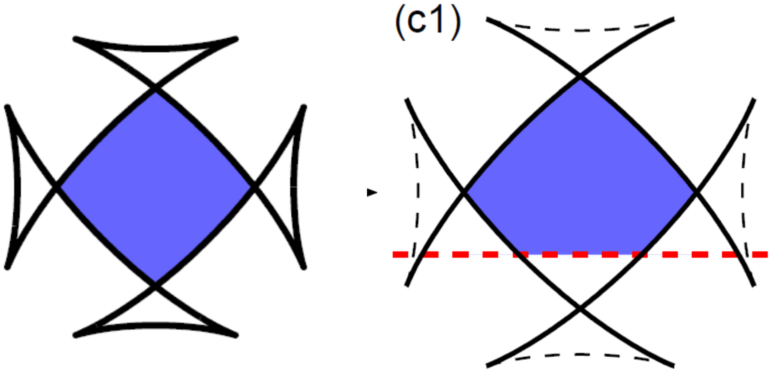
$\gamma(\theta)$-- interfacial energy (density)

$$W(\omega) = \int_\Gamma \gamma(\theta)\, d\Gamma + \underbrace{\gamma_{FS}\,|\Sigma_{FS}| + \gamma_{VS}\,|\Sigma_{VS}|}_{\text{Wall Energy}}$$

- Equilibrium configuration:

$$\min W(\omega)$$

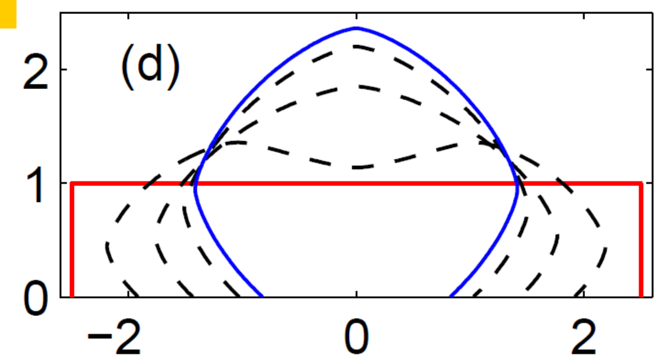
$$\text{subject to} \quad \int_\omega d\omega = \text{constant}$$



- Winterbottom construction – W. Winterbottom, 1967'

$\gamma(\theta) = 1 + 0.3\cos(4\theta)$

Models and Methods for Dynamical Evolution



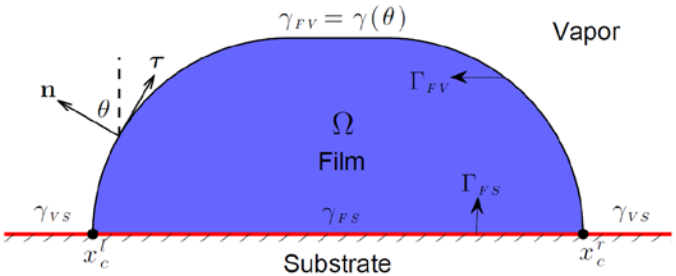
- Sharp interface model
  - Isotropic case
    - Model & power law --- Srolovitz & Safran JAP 86'
    - Marker particle method ---- Wong, Voorheers & Miksis 00'; Du etc JCP 10'; ….
  - Anisotropic (weakly and strongly) case
    - Model via thermodynamical variation – Wang etc PRB 15'; Jiang etc 16', …
    - Parametric finite element method (PFEM) -- Bao, Jiang, Wang &Zhao, 16'
- Kinetic Monte Carlo method – Dufay & Pierre-Lious PRL 11'; Pierre-Louis etc, EPL&PRL 09
- Discrete surface chemical potential method –Dornel etc. PRB 06'; Klinger etc 12
- Phase field model ---Jiang, Bao Thompson & Srolovitz, Acta Mater. 12'
- Crystalline formulation method – Cahn & Taylor 94'; Cater etc 95'; Kim etc 13'; Zucker etc 13'; Roosen etc 94'&98'; …..

Thermodynamic Variation

$\Gamma: \quad \vec{X}(s) = (x(s), y(s))^T \qquad s$ -- arclength

- Total interfacial free energy

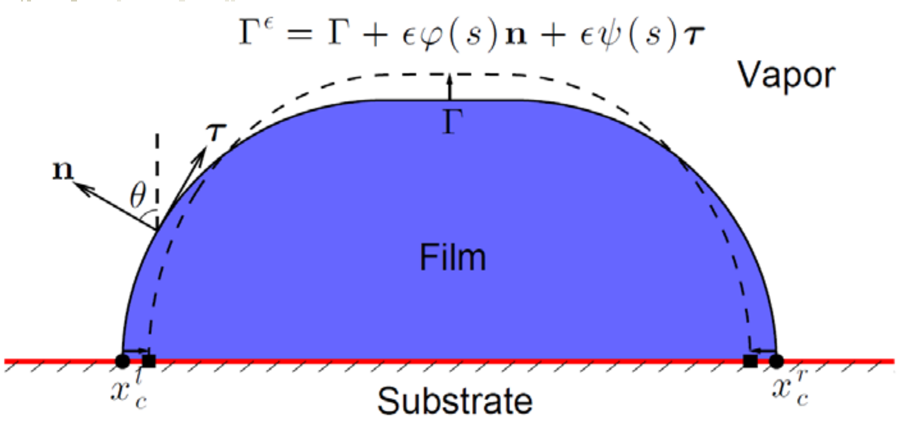
$$W = \int_{\Gamma} \gamma_{FV}(\theta) d\Gamma + (\gamma_{FS} - \gamma_{VS})(x_c^r - x_c^l)$$



  – Isotropic/anisotropic surface energy

$$\gamma_{FV} = \gamma(\theta) = 1 + \beta \cos(m\theta), \qquad m = 2, 3, 4, 6$$

- Thermo-dynamical variation: $\psi(s)$ is arbitrary & $\int_0^L \varphi(s) ds = 0$



$$\Gamma^{\varepsilon} : (x^{\varepsilon}(s), y^{\varepsilon}(s))$$
$$= (x(s) + \varepsilon u(s), y(s) + \varepsilon v(s))$$

$$u(s) = x_s(s)\psi(s) - y_s(s)\varphi(s)$$
$$v(s) = x_s(s)\varphi(s) + y_s(s)\psi(s)$$
$$v(0) = v(L) = 0 \quad \& \quad |A(\Gamma^{\varepsilon}) - A(\Gamma)| \leq C_0\, \varepsilon^2$$

Thermodynamic Variation

- Calculate first variation of the energy functional

$$W=\int_{\Gamma}\gamma_{FV}(\theta)d\Gamma+(\gamma_{FS}-\gamma_{VS})(x_c^r-x_c^l)$$

$$W^{\varepsilon}=\int_{\Gamma^{\varepsilon}}\gamma_{FV}(\theta^{\varepsilon})d\Gamma^{\varepsilon}+(\gamma_{FS}-\gamma_{VS})[(x_c^r+\varepsilon u(L,t))-(x_c^l+\varepsilon u(0,t))]$$

$$\left.\frac{dW^{\varepsilon}}{d\varepsilon}\right|_{\varepsilon=0}=\lim_{\varepsilon\to 0}\frac{W^{\varepsilon}-W}{\varepsilon}=\int_0^L\big(\gamma(\theta)+\gamma''(\theta)\big)\kappa\varphi ds+f(\theta_c^r)u(L)-f(\theta_c^l)u(0)$$

$$\mu:=\frac{\delta W}{\delta\Gamma}=(\gamma(\theta)+\gamma''(\theta))\kappa,\quad \frac{\delta W}{\delta x_c^r}=f(\theta)\big|_{\theta=\theta_c^r},\quad \frac{\delta W}{\delta x_c^l}=-f(\theta)\big|_{\theta=\theta_c^l}$$

$$f(\theta):=\gamma(\theta)\cos\theta-\gamma'(\theta)\sin\theta-\gamma_0\cos\theta_i,\quad \cos\theta_i=\sigma:=\frac{\gamma_{VS}-\gamma_{FS}}{\gamma_0}$$

[1] Wang, Jiang, Bao & Srolovitz, Phys. Rev. B, 15'; Bao, Jiang, Srolovitz & Wang, SIAP, 17'

Equilibrium in 2D

- Equilibrium shape

$$\mu(s) := \widetilde{\gamma}(\theta)\kappa(s) = [\gamma(\theta) + \gamma''(\theta)]\,\kappa(s) \equiv C, \qquad a.e. \quad s \in [0, L],$$
$$f(\theta;\sigma) = \gamma(\theta)\cos\theta - \gamma'(\theta)\sin\theta - \sigma = 0, \qquad \theta = \theta_a^l, \theta_a^r,$$

- Calculate second variation

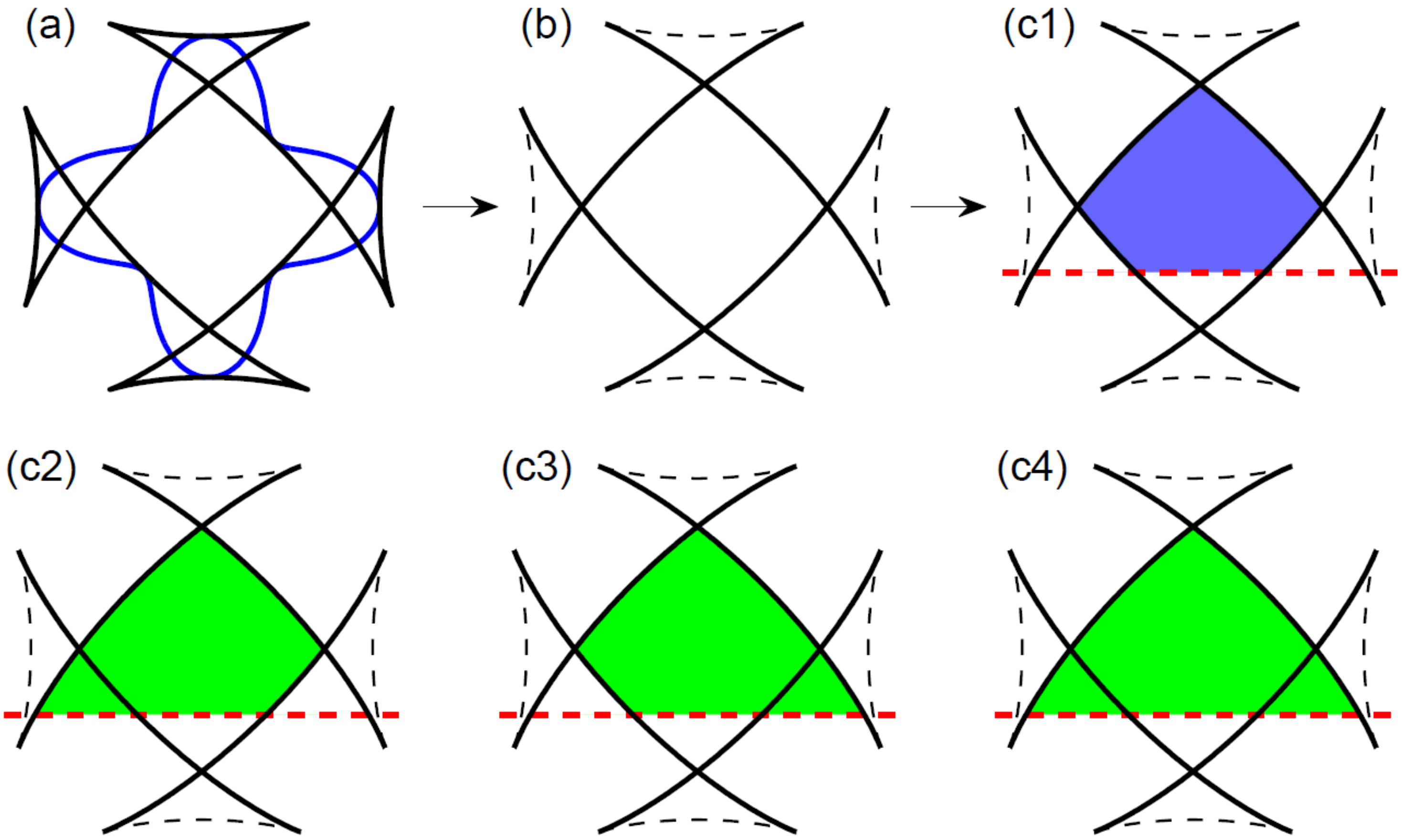
$$\delta^2 W(\Gamma;\varphi,\psi) := \left.\frac{d^2 W^{\varepsilon}}{d\varepsilon^2}\right|_{\varepsilon=0} = \int_0^L \big(\gamma(\theta) + \gamma''(\theta)\big)(\varphi_s - \kappa\psi)^2\, ds$$

- Stable equilibrium shape $\tilde{\gamma}(\theta) := \gamma(\theta) + \gamma''(\theta) \geq 0$

[1] Bao, Jiang, Srolovitz & Wang, SIAP, 2017.

Generalized Winterbottom Construction

(Bao, Jiang, Srolovitz & Wang, SIAP, 17')

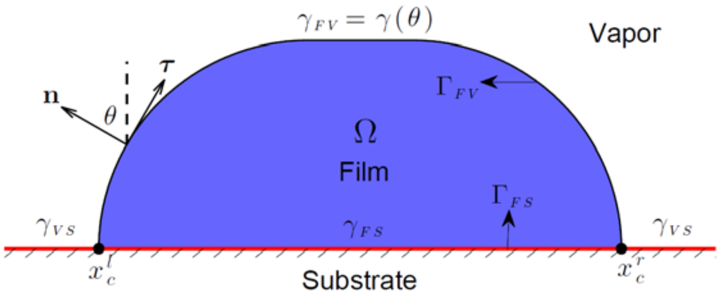


$$\gamma(\theta) = 1 + 0.3\cos(4\theta)$$

Sharp Interface Model

(Isotropic/Weakly Anisotropic Surface Energy)

$\Gamma: \quad \vec{X}(s,t) = (x(s,t), y(s,t))^T \qquad s$ -- arclength



- The Model (Wang, Jiang, Bao, Srolovitz, PRB 15')

$$\frac{\partial \vec{X}(s,t)}{\partial t} = V_n \vec{n}, \qquad \text{with} \qquad V_n = B\frac{\partial^2 \mu}{\partial s^2}$$

$$\mu(\theta) = (\gamma(\theta) + \gamma''(\theta))\kappa$$

θ_c^r --Dynamical contact angle

θ_i----Isotropic Young contace angle

- Boundary conditions
  - Contact point condition (BC1): $y(x_c^r, t) = 0$
  - Relaxed contact angle condition (BC2)[1]: $\frac{dx_c^r(t)}{dt} = -\eta\frac{\delta W}{\delta x_c^r} = -\eta f(\theta_c^r),$
  - Zero-mass flux condition (BC3): $\partial_s \mu(x_c^r, t) = 0$
- Anisotropic Young equation $\eta \to \infty$

$$\gamma(\theta)\cos\theta - \gamma'(\theta)\sin\theta - \gamma_0\cos\theta_i = 0 \overset{\gamma(\theta)\equiv\gamma_0}{\Rightarrow} \cos\theta = \cos\theta_i$$

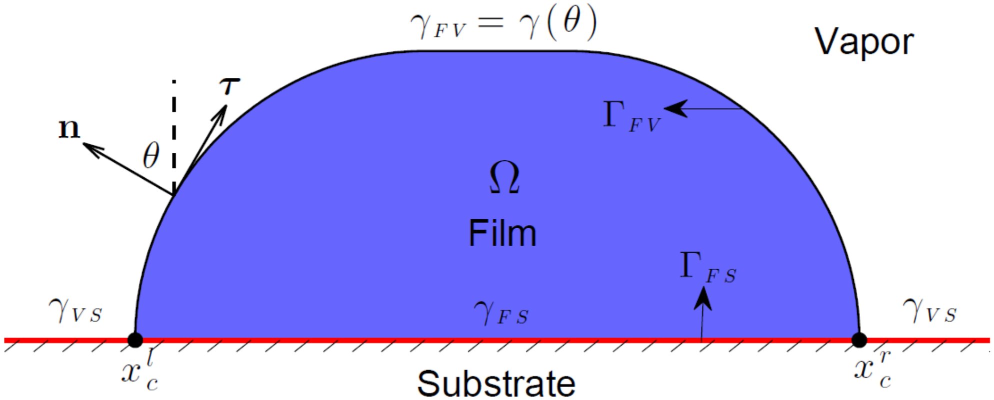
[1] Ren, E, Phys. Fluid, 2007&2011; Ren, Hu & E, Phys Fluid, 2010.

Dynamical Properties

- Area (Mass) conservation

$$A(t) = \int_0^{L(t)} y \partial_s x \, ds$$

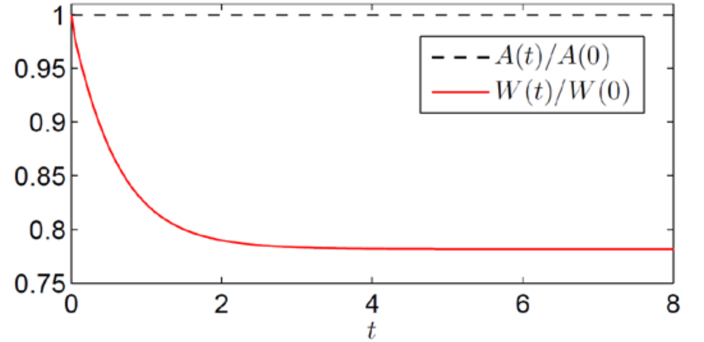
$$\Rightarrow A(t) \equiv A(0)$$



- Energy dissipation

$$W(t) = \int_0^{L(t)} \gamma_{FV}(\theta) ds + (\gamma_{FS} - \gamma_{VS})(x_c^r - x_c^l)$$

$$\Rightarrow W'(t) \leq 0$$



Parametric Finite Element Method (PFEM)

$$\frac{\partial \vec{X}(s,t)}{\partial t} = V_n \vec{n}, \quad \text{with} \quad V_n = B\frac{\partial^2 \mu}{\partial s^2}$$

- Isotropic/weakly anisotropic case $\mu(\theta) = (\gamma(\theta) + \gamma''(\theta))\kappa$
  - Mathematical model and variational form

$$(\partial_t \vec{X}(s,t)) \bullet \vec{n} = B\partial_{ss}\mu \qquad \int_C (\partial_t \vec{X}(s,t)) \bullet \vec{n}\, \phi dp + \int_C B\partial_s \mu\, \partial_s \phi dp = 0, \quad \phi \in H_0^1$$

$$\mu(\theta) = (\gamma(\theta) + \gamma''(\theta))\kappa \Leftrightarrow \qquad \int_C [\mu(\theta) - (\gamma(\theta) + \gamma''(\theta))\kappa]\varphi dp = 0, \qquad \varphi \in H^1$$

$$\kappa\vec{n} = -\partial_{ss}\vec{X}(s,t) \qquad \int_C \kappa\vec{n} \bullet \vec{\eta} dp + \int_C \partial_s \vec{X}(s,t) \bullet \partial_s \vec{\eta} dp = 0, \qquad \vec{\eta} \in (H_0^1)^2$$

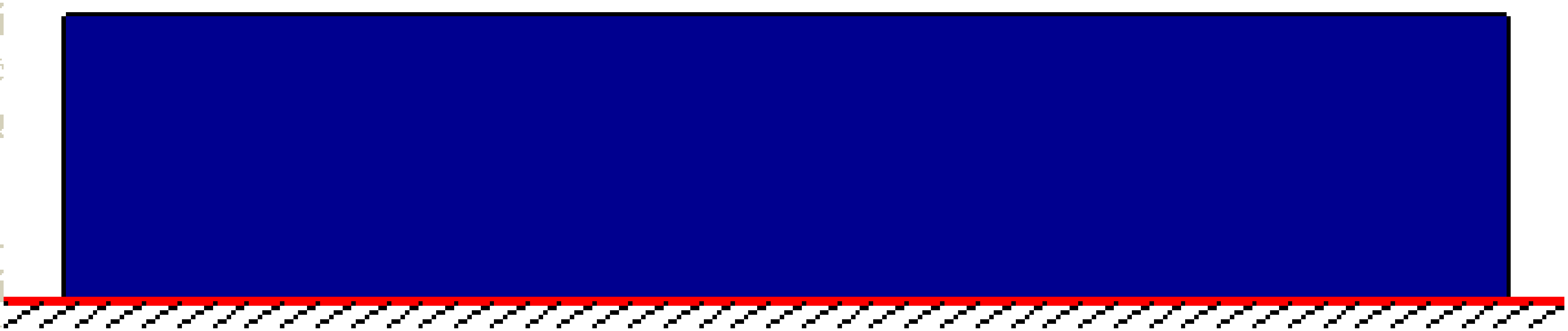
  - Boundary conditions $y(x_c^r, t) = 0, \qquad \frac{dx_c^r(t)}{dt} = -\eta f(\theta_c^r)$
  - Finite element discretization via piecewise polynomials
- Ref: [1] J.W. Barratt, H. Garcke & R. Nurnberg, J. Comput. Phys., 2007; SISC (2007); 2012.

  [2] W. Bao, W. Jiang Y. Wang & Q. Zhao, JCP, 2017.

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

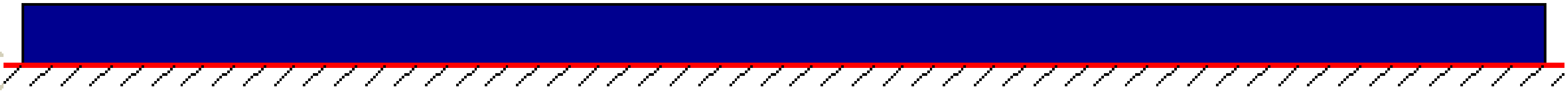
Isotropic surface energy and short

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



Weakly anisotropic surface energy & short

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



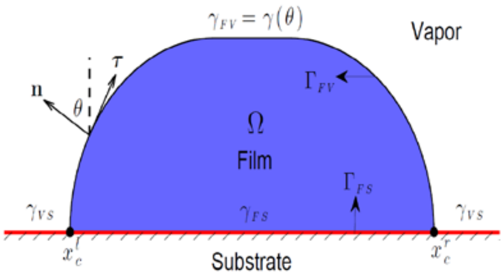
Weakly anisotropic surface energy & long for pinch-off

Extension to Strongly Anisotropic Case

$$\tilde{\gamma}(\theta) := \gamma(\theta) + \gamma''(\theta) < 0 \qquad \Gamma: \quad \vec{X}(s) = (x(s), y(s))^T \qquad s \text{ -- arclength}$$

- Regularized interfacial free energy

$$W_\varepsilon = \int_\Gamma \gamma_{FV}(\theta)\, d\Gamma + (\gamma_{FS} - \gamma_{VS})(x_c^r - x_c^l) + \frac{\varepsilon^2}{2}\int_\Gamma \kappa^2 \, d\Gamma$$



- Calculate first variation of the energy functional

$$\mu_\varepsilon := \frac{\delta W_\varepsilon}{\delta \Gamma} = (\gamma(\theta) + \gamma''(\theta))\kappa - \varepsilon^2 \left[\partial_{ss}\kappa + \frac{\kappa^3}{2} \right],$$

$$\frac{\delta W_\varepsilon}{\delta x_c^r} = f_\varepsilon(\theta)\Big|_{\theta=\theta_c^r}, \quad \frac{\delta W_\varepsilon}{\delta x_c^l} = -f_\varepsilon(\theta)\Big|_{\theta=\theta_c^l}, \qquad \cos\theta_i = \sigma := \frac{\gamma_{VS} - \gamma_{FS}}{\gamma_0}$$

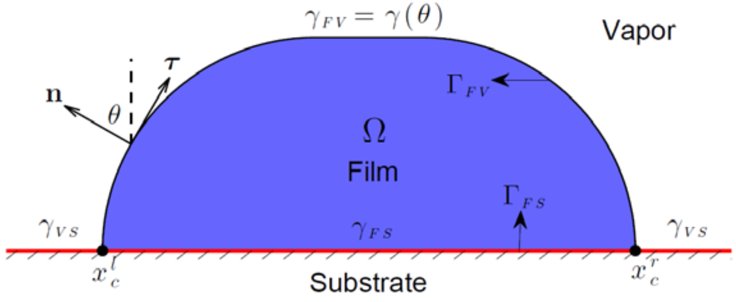
$$f_\varepsilon(\theta) := \gamma(\theta)\cos\theta - \gamma'(\theta)\sin\theta - \gamma_0 \cos\theta_i + \varepsilon^2 \left[\frac{\kappa(\theta)^2}{2}\cos\theta - \partial_s \kappa(\theta)\sin\theta \right]$$

[1] J. Lowengrub, A. Voigt, ….
[2] Jiang, Wang, Zhao, Srolovitz & Bao, Script. Mater., 16'; Bao, Jiang, Srolovitz & Wang, SIAP,17'

Sharp Interface Model

(for Strongly Anisotropic Surface Energy)

$\Gamma: \quad \vec{X}(s,t) = (x(s,t), y(s,t))^T \qquad s$ -- arclength



- The Model (Jiang, Wang, Zhao, Srolovitz, Bao, 16')

$$\frac{\partial \vec{X}(s,t)}{\partial t} = V_n \, \vec{n}, \qquad \text{with} \qquad V_n = B\frac{\partial^2 \mu_\varepsilon}{\partial s^2}$$

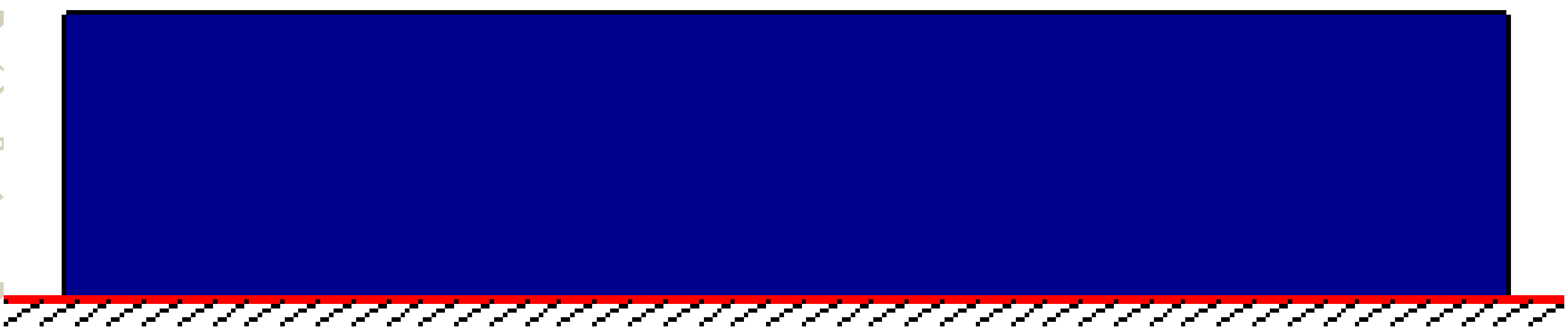
$$\mu_\varepsilon(\theta) = (\gamma(\theta) + \gamma''(\theta))\kappa - \varepsilon^2 \left[\partial_{ss}\kappa + \frac{\kappa^3}{2} \right]$$

θ_c^r --Dynamical contact angle

θ_i----Isotropic Young contace angle

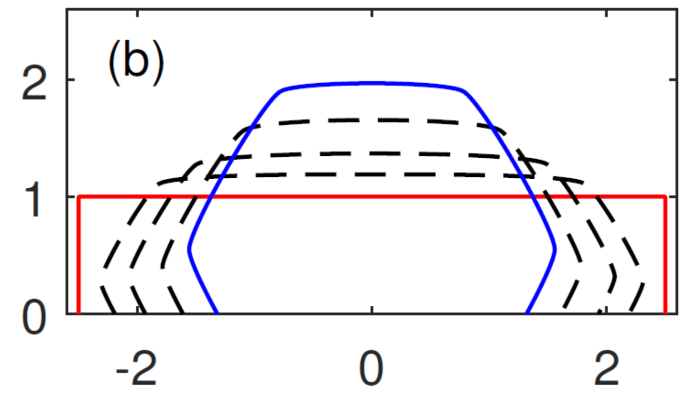
- Boundary conditions
  - Contact point condition (BC1): $y(x_c^r, t) = 0$
  - Relaxed contact angle condition (BC2): $\frac{dx_c^r(t)}{dt} = -\eta \frac{\delta W}{\delta x_c^r} = -\eta f_\varepsilon(\theta_c^r),$
  - Zero-curvature condition (BC3): $\kappa(x_c^r, t) = 0$
  - Zero-mass flux condition (BC4): $\partial_s \mu(x_c^r, t) = 0$

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



Strongly anisotropic surface energy & short

Extension to Cusped Surface Energy



- Cusped surface energy (or not smooth)

$$\gamma(\theta) \notin C^2 \Rightarrow \gamma_\delta(\theta) \in C^2$$

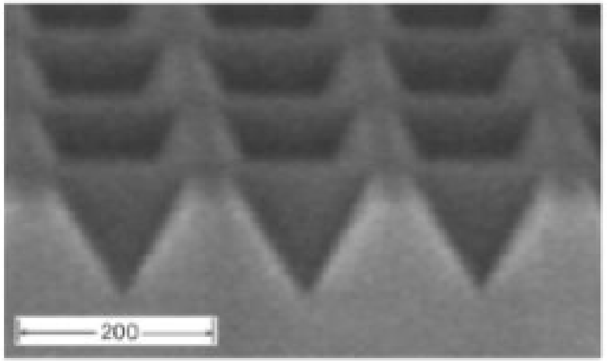
  – A typical example

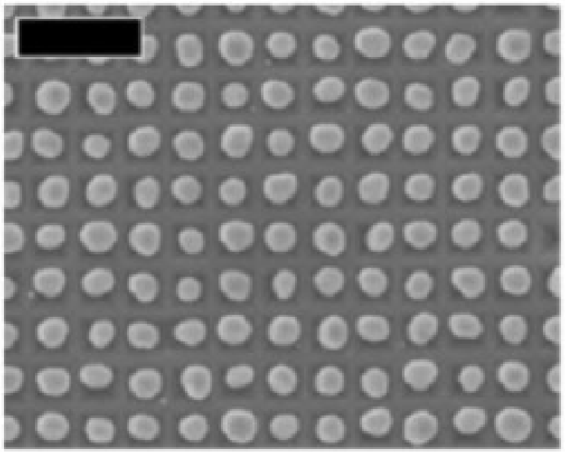
$$\gamma(\theta) = |\cos\theta| + |\sin\theta| \Rightarrow \qquad (0 < \delta \ll 1)$$

$$\gamma_\delta(\theta) = \sqrt{\delta^2 + (1-\delta^2)\cos^2\theta} + \sqrt{\delta^2 + (1-\delta^2)\sin^2\theta}$$

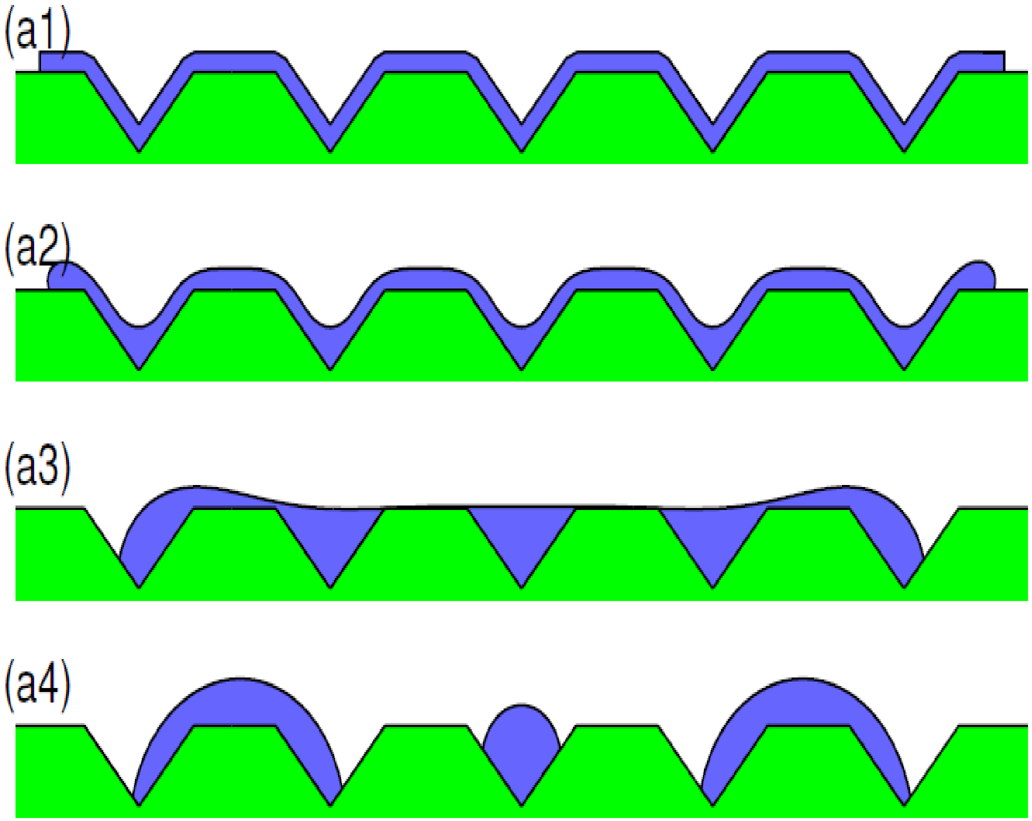
- Regularize (or smooth) the surface energy density
  – Adapt the sharp interface model for $\gamma_\delta(\theta)$

Extension to Curved Substrate





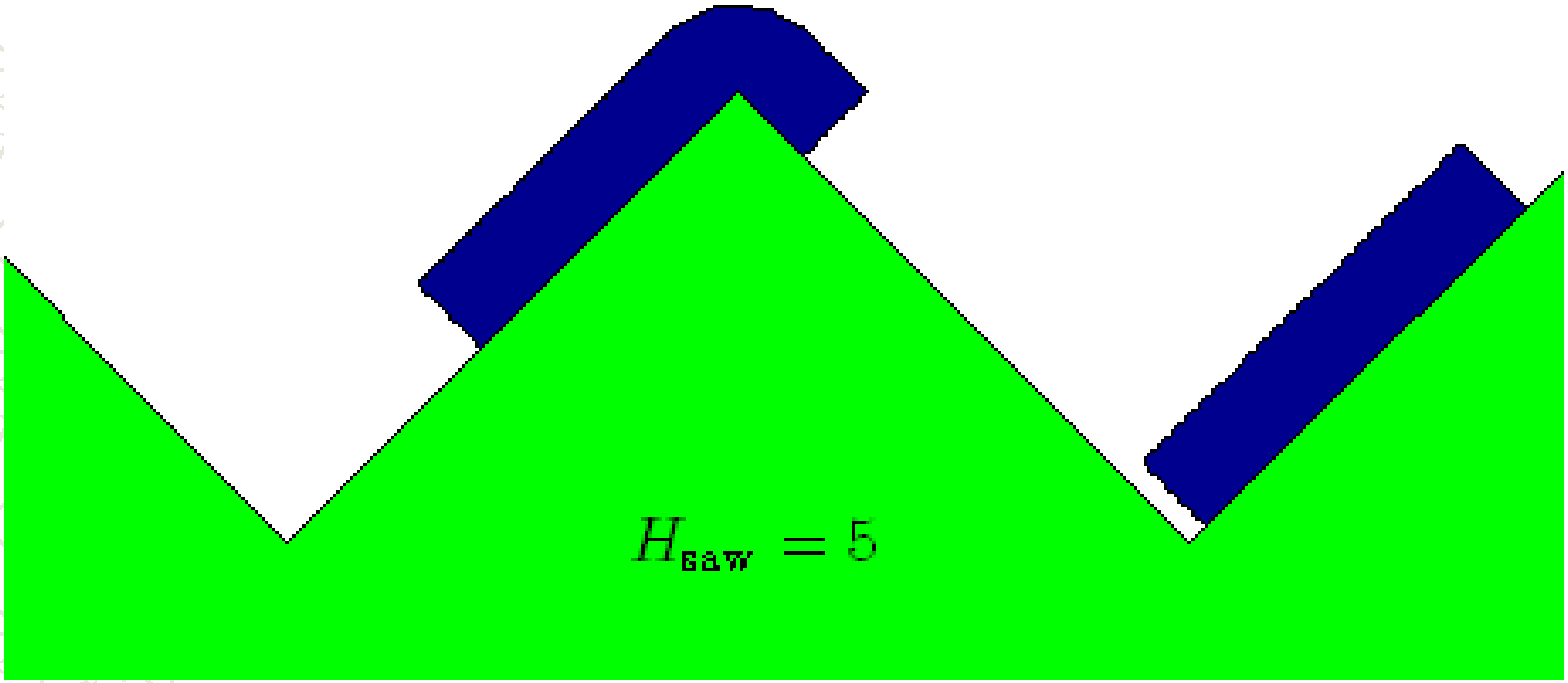
Experiment (Ye, et. Al, PRB, 10')



Simulation

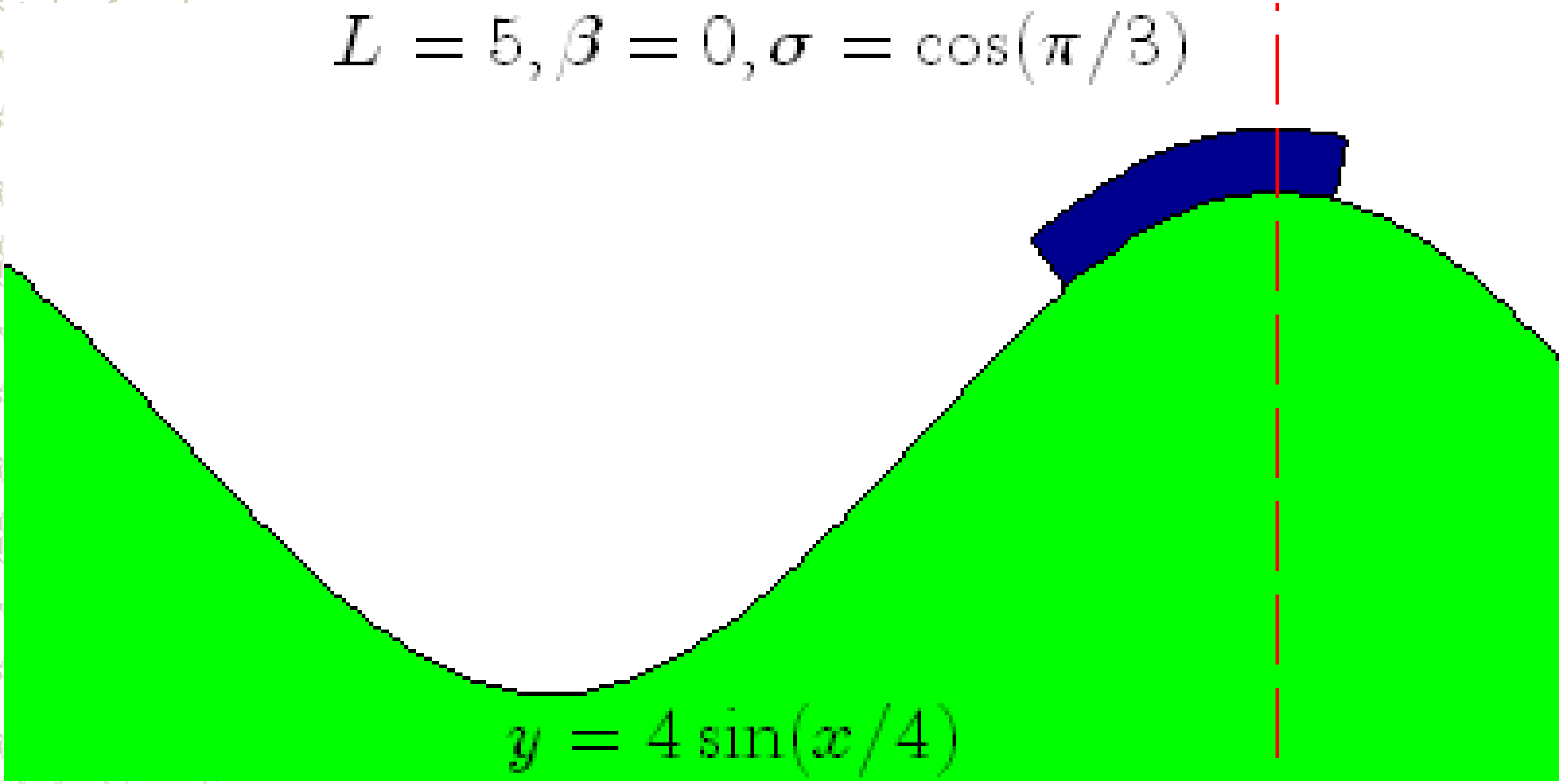
Extension to Curved Substrate

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



Extension to Curved Substrate --Small Particle Migration

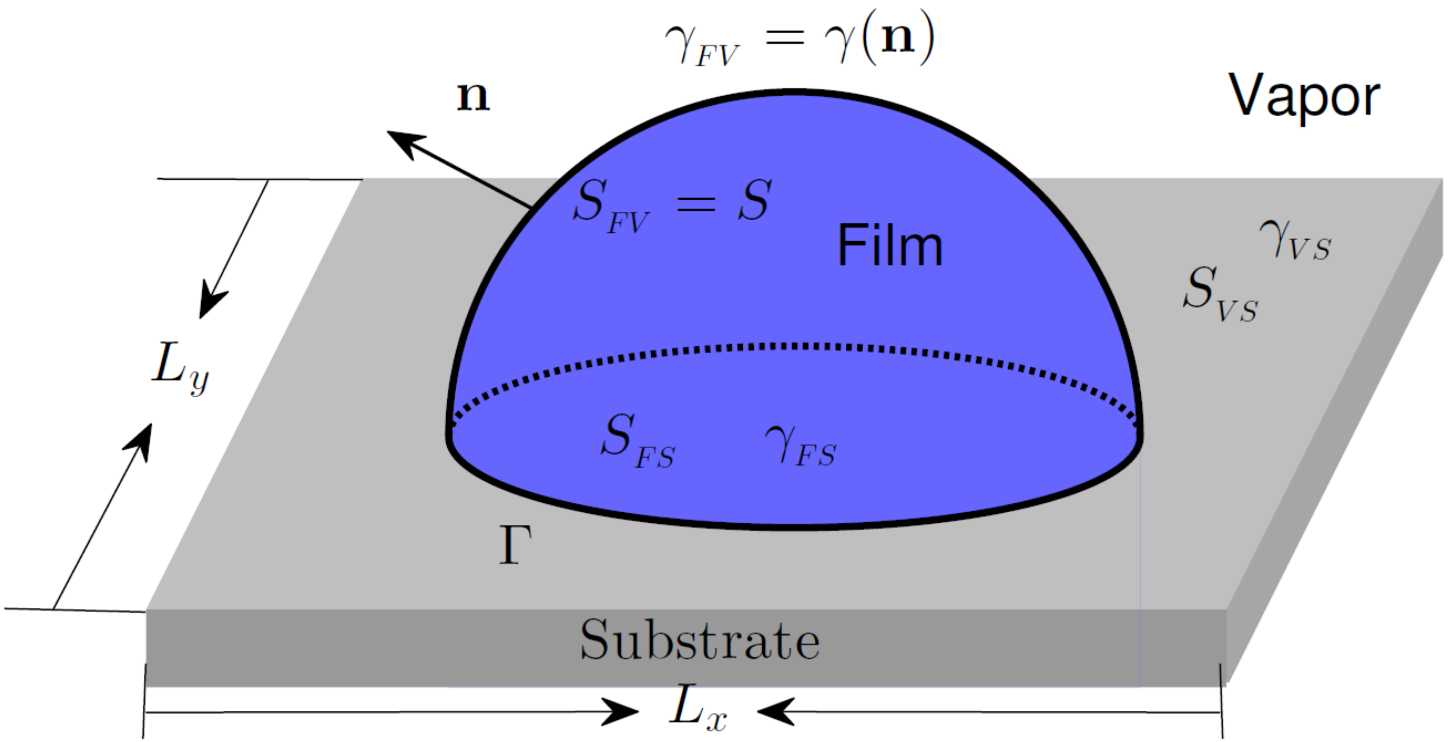
(Jiang, Wang, Srolovitz & Bao, 18')



$$\Gamma(s) = (x(s), y(s)) \text{ with } \kappa := \kappa(s) \Rightarrow v(s) \propto C(\sigma)\kappa'(s)$$

A variation approach via Onsager's principle– Jiang, Wang, Qian, Srolovitz, Bao, 18'

Extension to Three Dimension (3D)



- Total interfacial energy

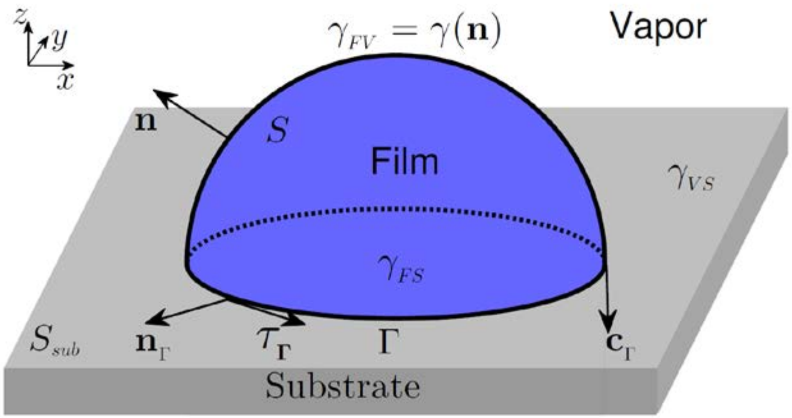
$$W = W_I + W_S = \iint_S \gamma(\mathbf{n})\, dS + (\gamma_{FS} - \gamma_{VS})A(\Gamma),$$

Extension to 3D

(Bao, Jiang & Zhao, 18')

$$S: \quad \vec{X}(u,v,t) = (x(u,v,t), y(u,v,t), z(u,v,t))^T$$

- Main ideas -- thermodynamic variation, shape derivatives & Cahn-Hoffman \xi-vector
- The sharp interface model



$$\partial_t \mathbf{X} = \Delta_S \mu\, \mathbf{n}, \qquad t > 0,$$

$$\mu = \nabla_S \cdot \boldsymbol{\xi}, \qquad \boldsymbol{\xi} = \nabla \hat{\gamma}(\mathbf{n}),$$

– Cahn-Hoffmann \xi-vector

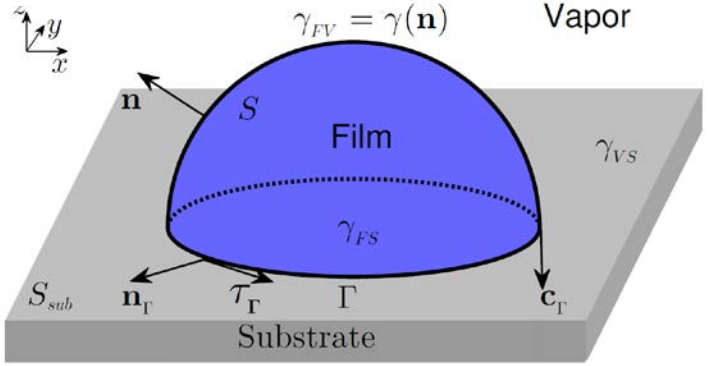
$$\gamma(\mathbf{n}) : S^2 \to \mathbb{R} \qquad \hat{\gamma}(\mathbf{p}) = |\mathbf{p}|\gamma(\frac{\mathbf{p}}{|\mathbf{p}|}), \quad \forall \mathbf{p} \in \mathbb{R}^3 \backslash \{\mathbf{0}\}$$

$$\boldsymbol{\xi}(\mathbf{n}) = \nabla \hat{\gamma}(\mathbf{n})$$

Extension to 3D

(Bao, Jiang & Zhao, 18')

$\mathbf{c}_\Gamma^\gamma = (\boldsymbol{\xi}\cdot\mathbf{n})\mathbf{c}_\Gamma - (\boldsymbol{\xi}\cdot\mathbf{c}_\Gamma)\mathbf{n},\ \mathbf{n}_\Gamma = \frac{1}{\sqrt{n_1^2+n_2^2}}(n_1, n_2, 0),\ \sigma = \frac{\gamma_{VS}-\gamma_{FS}}{\gamma_0},$

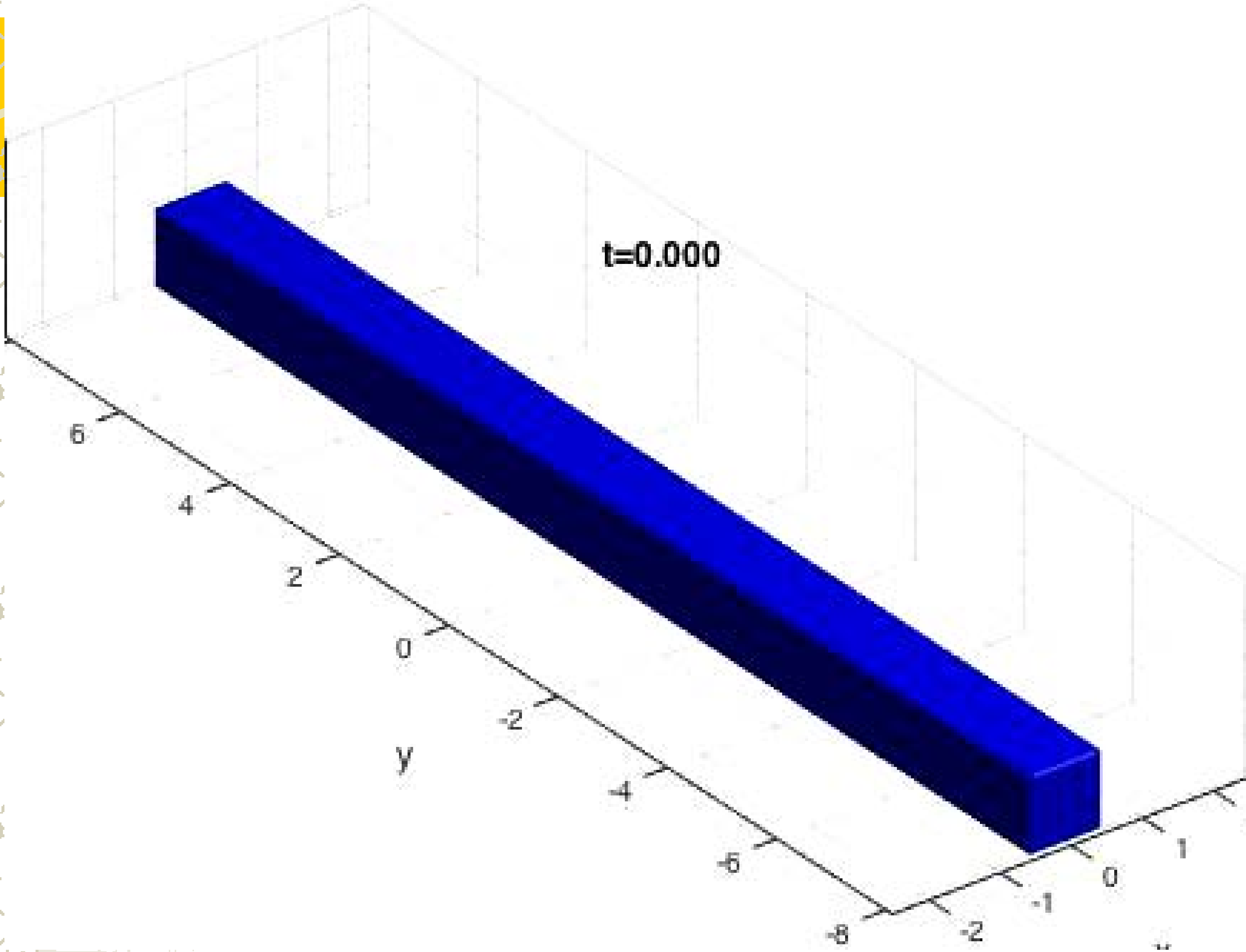


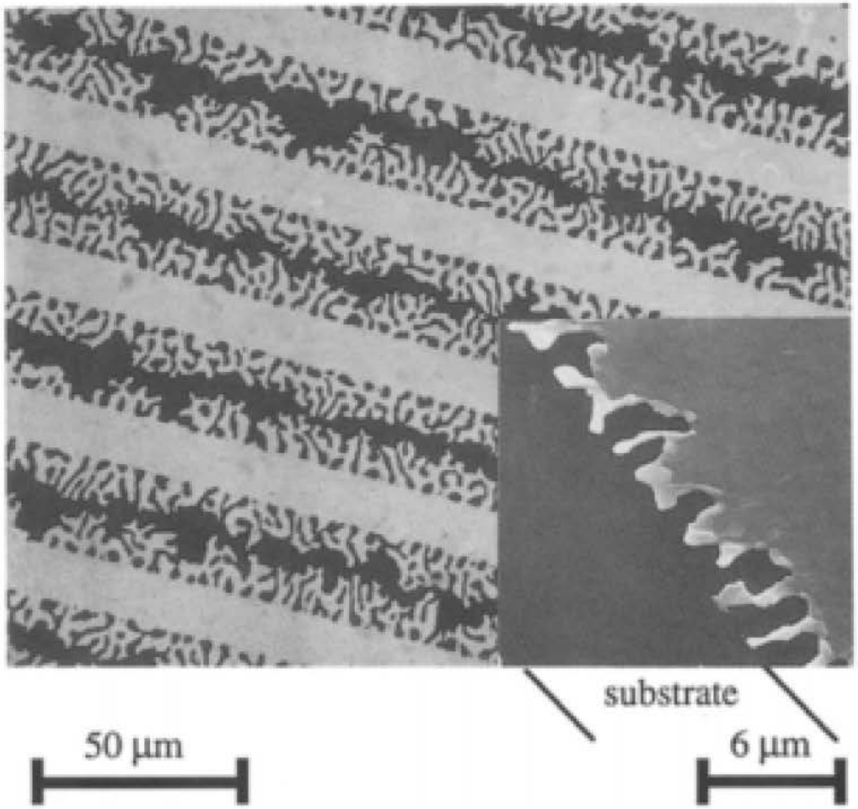
- Boundary conditions
  - Contact line condition $\Gamma \subset S_{sub}$
  - Relaxed contact angle condition

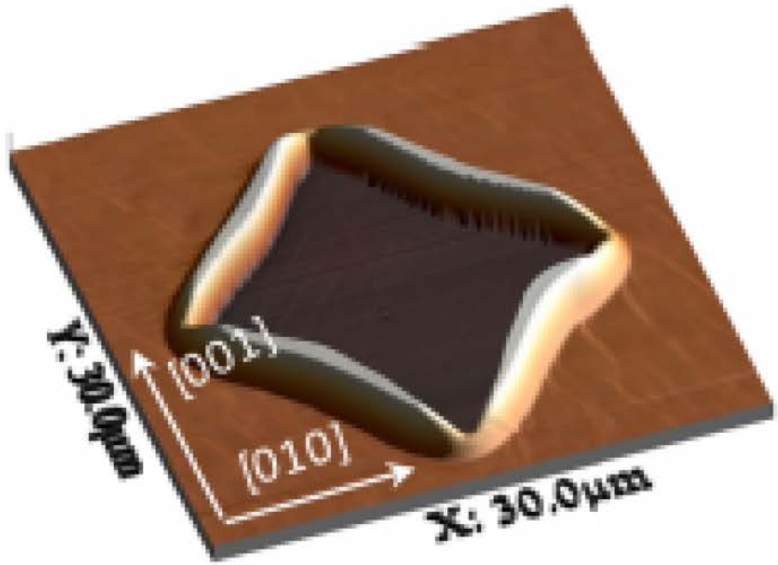
$$\partial_t \mathbf{X}_\Gamma = -\eta\left[\mathbf{c}_\Gamma^\gamma \cdot \mathbf{n}_\Gamma - \sigma\right]\mathbf{n}_\Gamma$$

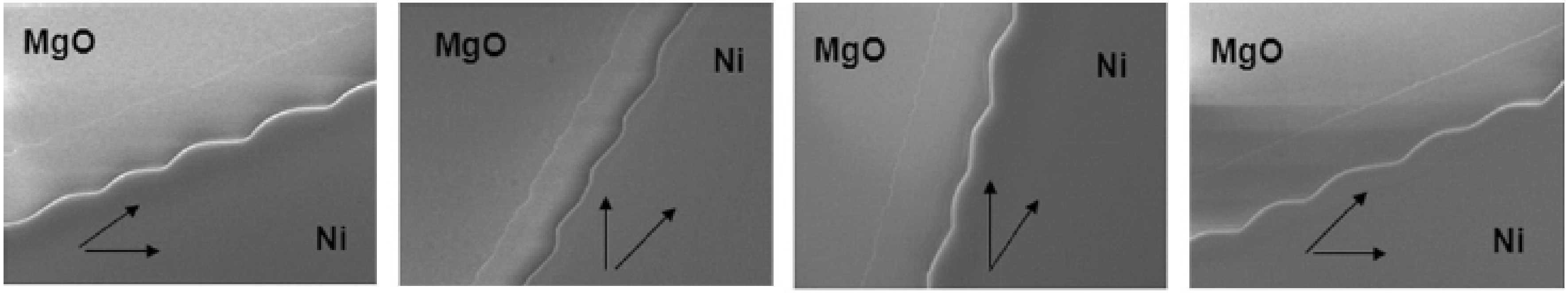
  - Zero-flux condition $(\mathbf{c}_\Gamma \cdot \nabla_S \mu)\big|_\Gamma = 0,$
- For isotropic case, $\mu = H$ -- mean curvature

- Volume conservation & energy dissipation
- Parameter finite element (PFEM) method

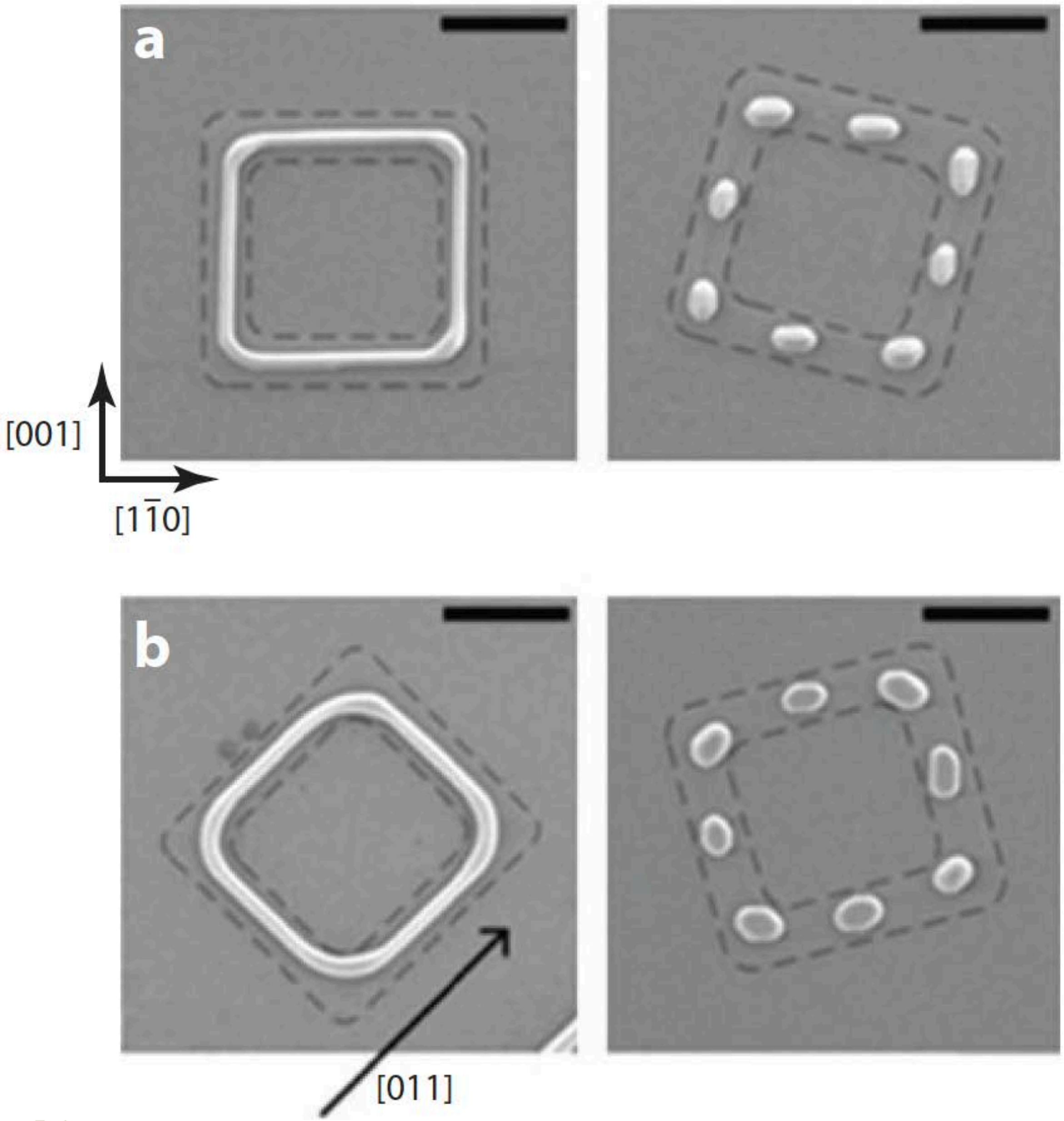

t=0.000
6
4
2
0
-2
-4
-6
-8
y
-2
-1
0
1



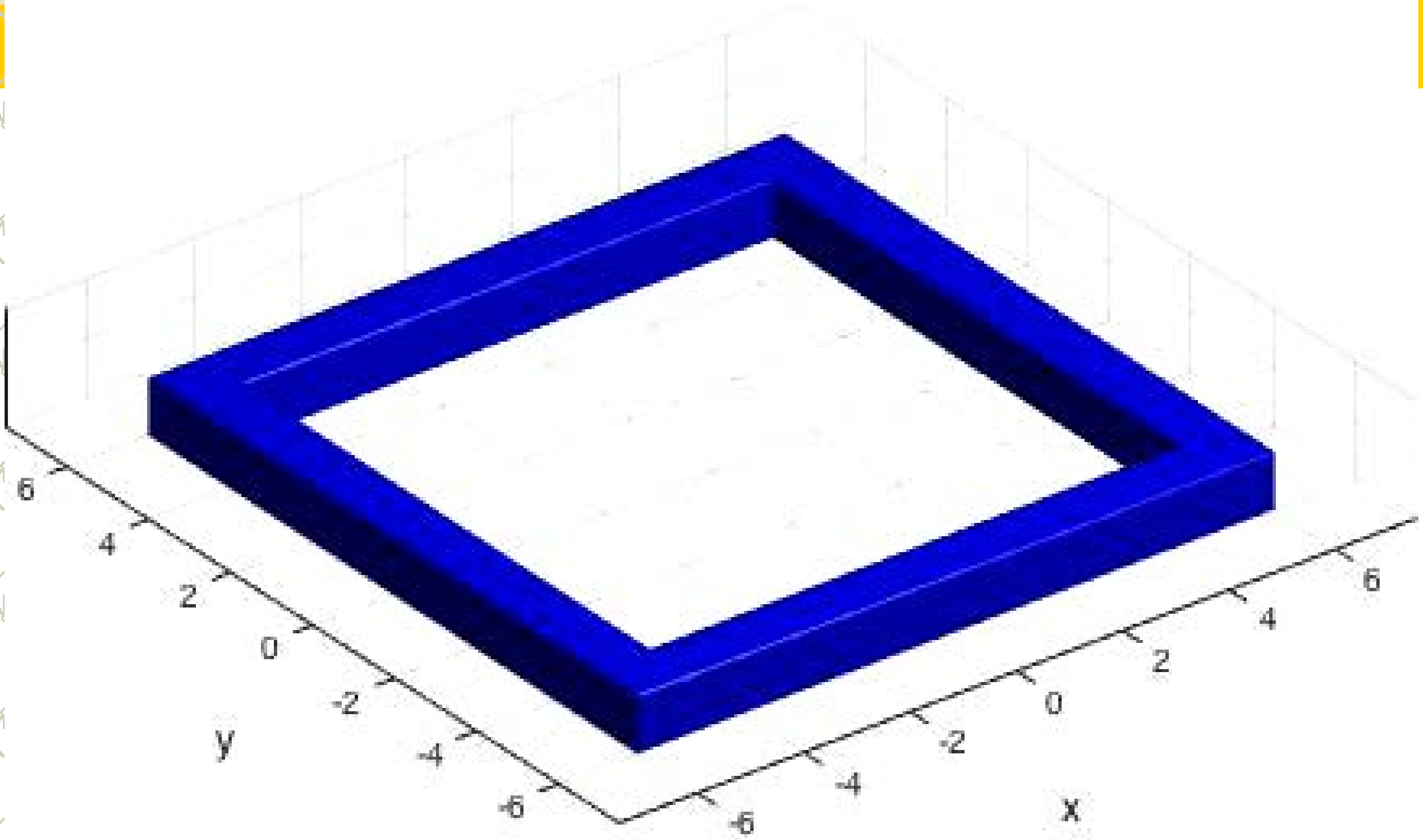




[1] J. Ye & C.V. Thompson, Acta Mater. 23 (2011), 1567; 59 (2011), 582; Appl. Phys. Lett. 97 (2010), 071904



[1] J. Ye & C.V. Thompson, Acta Mater. 23 (2011), 1567; 59 (2011), 582; Appl. Phys. Lett. 97 (2010), 071904



6
4
2
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-2
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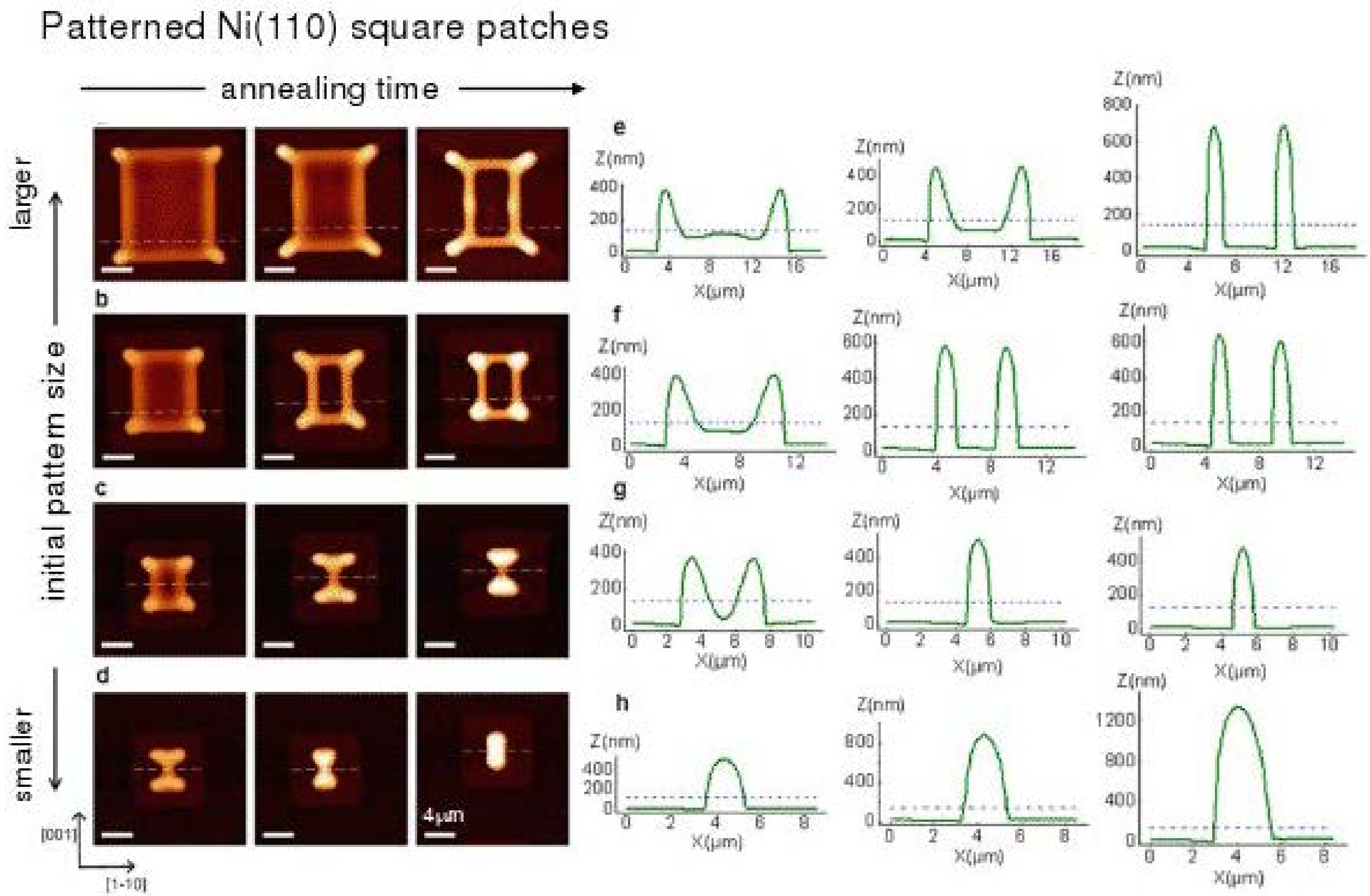
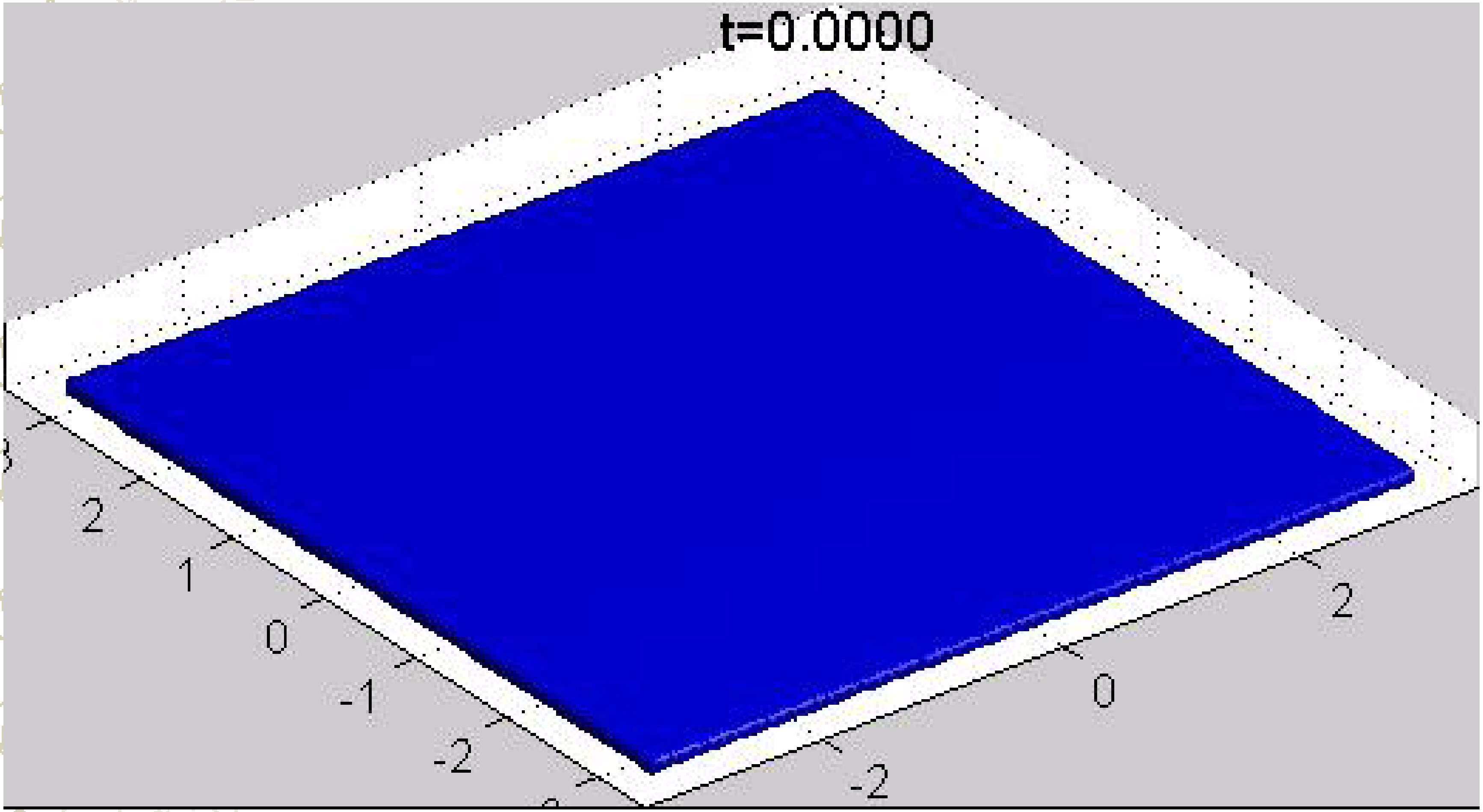
-6
y
-6
-4
-2
0
2
4
6
x

Dewetting Patterned Films

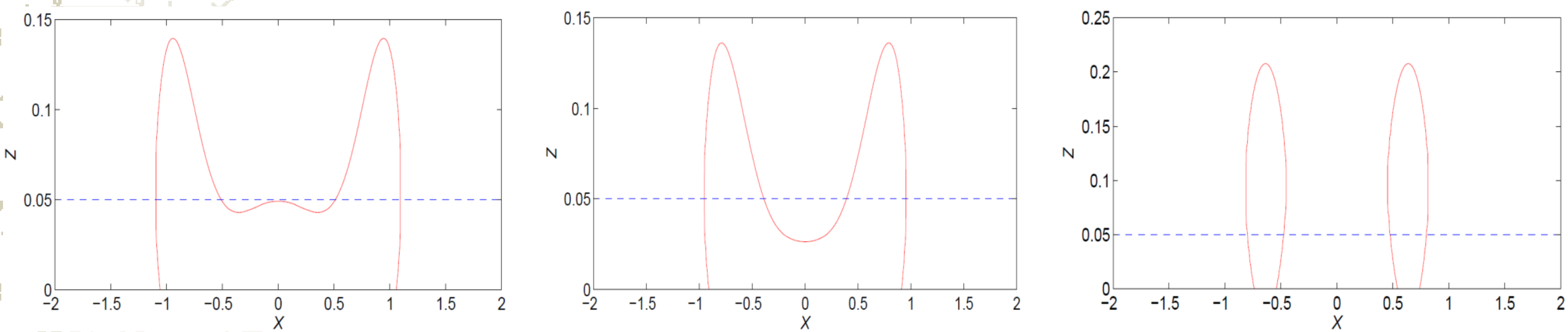


[1] J. Ye & C.V. Thompson, Phys. Rev. B, 82 (2010), 193408.

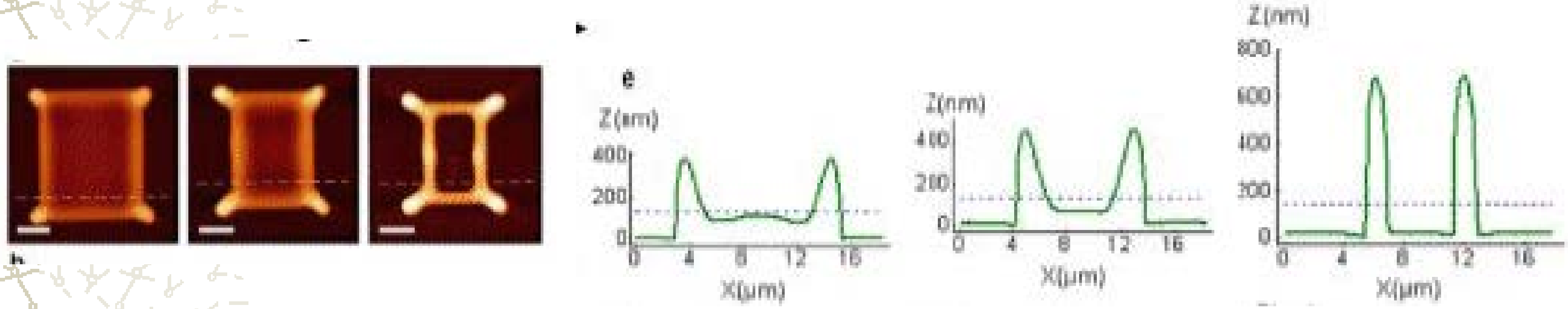
Solid-state dewetting in 3D via SIM



3D Results – Comparison with Experiment



Computational Results



Experimental Results

Phase Field/Diffuse Interface Model

- Phase field models --- S. Allen & J.W. Cahn (1975—79), J.W. Cahn & J. E. Hilliard (1958); G.J. Fix (1983), J.S. Langer (1986), L.Q. Chen (2002); C.M. Elliott, X.F. Chen, J. Shen, Q. Du, J. Lowengrub, A. Voigt, Q. Wang, G. Forest, XB Feng, PW Zhang, A.A. Lee A. Munch & E. Suli, …
  - Introduce a phase function& write down the energy
  - By variation – Allen-Cahn or Cahn-Hilliard equations
  - Applications in materials simulation: solidification; viscous fingering; fracture; solid-state nucleation; dislocation dynamics, …….
- Advantages
  - Interface/surface capturing method – Eulerian coordinates
  - Easy extension from 2D to 3D
  - Naturally capture complex topological changes

Problem Set-up

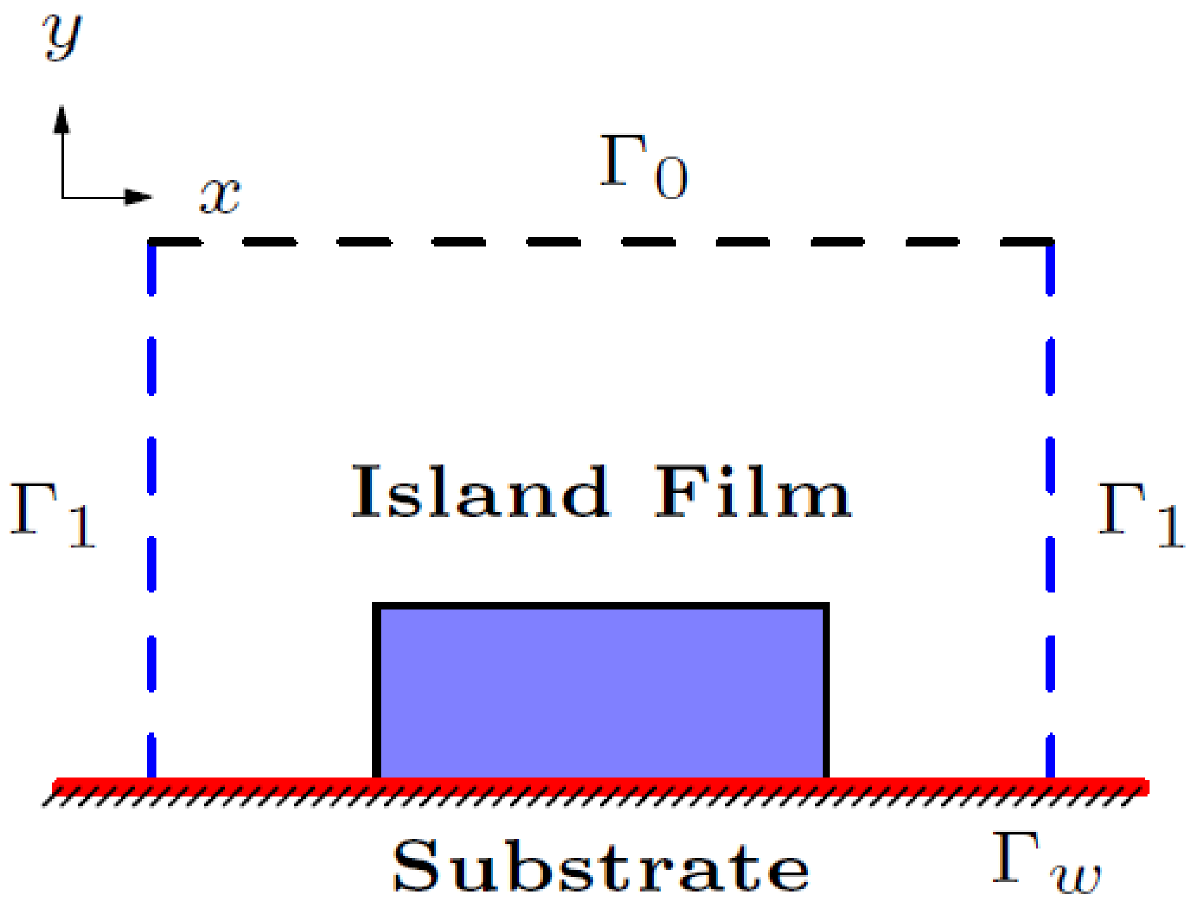


Figure: A schematical illustration of the problem set-up.

Phase Field Model

- Introduce the phase function $\phi := \phi^{\varepsilon}(\vec{x},t) \in [-1,1]$

$$\phi = \begin{cases} \approx 1 \ \& \in (0,1] & \text{thin film phase} \\ \approx -1 \ \& \in [-1,0) & \text{vapor phase} \\ = 0 & \text{interface/surface} \end{cases} \qquad \varepsilon \text{ --interface width}$$

- Total free energy-- isotropic surface energy

$$\tanh\left(\frac{d_s(\vec{x},\Gamma)}{\varepsilon}\right)$$

$$W^{\varepsilon} = \underbrace{W_{FV}^{\varepsilon}}_{\text{Film-vapor phase energy}} + \underbrace{W_{W}^{\varepsilon}}_{\text{Wall energy}}$$

$$= \int_{\Omega} \underbrace{f_{FV}}_{\text{Film-vapor energy density}} d\Omega + \int_{\Gamma_W} \underbrace{f_W}_{\text{Wall energy density}} d\Gamma_W$$

- Energy minimization problem:

$$\min W^{\varepsilon} \qquad \text{subject to BCs}$$

Phase Field Model

- Film/vapor phase energy – Ginzburg-Landau free energy

$$f_{FV}(\phi,\nabla\phi) = \underbrace{\lambda}_{\text{mixing constant}}\left[\frac{\varepsilon}{2}\underbrace{|\nabla\phi|^2}_{\text{film/vapor energy}} + \frac{1}{\varepsilon}\underbrace{F(\phi)}_{\text{interface energy}}\right], \qquad F(\phi) = (\phi^2 - 1)^2$$

  - Convergence – L. Modica & S. Mortola, Boll. Un. Mat. Ital. A, 14 (1977), 526.

$$\text{If } \lambda = \frac{3\sqrt{2}}{4}\gamma_{FV} \quad \Rightarrow \quad W_{FV}^{\varepsilon} \to \gamma_{FV}\,|\Sigma_{FV}|$$

- Wall energy must satisfy
  - At homogeneous vapor phase $\phi = -1 \Rightarrow f_W(\phi) = \gamma_{VS}$ & $f_W^{'}(\phi) = 0$
  - At homogeneous film phase $\phi = 1 \Rightarrow f_W(\phi) = \gamma_{FS}$ & $f_W^{'}(\phi) = 0$

$$f_W(\phi) = \frac{\gamma_{VS} + \gamma_{FS}}{2} - \frac{\phi(3-\phi^2)}{4}(\gamma_{VS} - \gamma_{FS})$$

Phase Field Model

- Cahn-Hilliard equation with function-dependent mobility

$$\begin{cases} \dfrac{\partial \phi}{\partial t} = \nabla \bullet (M \nabla \mu) \\ \mu = \phi^3 - \phi - \varepsilon^2 \Delta \phi \end{cases} \quad \text{in} \quad \Omega \quad \text{with} \quad M = (1-\phi^2)^2$$

- With BCs(Jiang, Bao, Thompson & Srolovitz, Acta Mater., 12')
  - On wall boundary Γ_W $\quad \varepsilon \dfrac{\partial \phi}{\partial \vec{n}} + \dfrac{f_W^{'}(\phi)}{\lambda} = 0 \quad \& \quad \cos\theta_s = \dfrac{\gamma_{VS} - \gamma_{FS}}{\gamma_{FV}}$

$$\varepsilon \frac{\partial \phi}{\partial \vec{n}} + \frac{\sqrt{2}}{2}(\phi^2 - 1)\cos\theta_s = 0 \qquad \& \qquad \frac{\partial \mu}{\partial \vec{n}} = 0$$

  - On other boundaries $\Gamma_0 \cup \Gamma_1 = \partial\Omega \setminus \Gamma_W \; \dfrac{\partial \phi}{\partial \vec{n}} = 0 \quad \& \quad \dfrac{\partial \mu}{\partial \vec{n}} = 0$

- Recent debate on proper mobility for surface diffusion – A.A. Lee, A. Munch & E Suli, APL 15' & 16'; A. Voigt, APL 16', …..

Numerical Method

- Stabilized semi-implicit method

$$\frac{\phi^{n+1}-\phi^{n}}{\tau}=A\varepsilon^{2}\Delta^{2}(\phi^{n}-\phi^{n+1})+S\Delta(\phi^{n+1}-\phi^{n})+\nabla\bullet(M^{n}\nabla\mu^{n})$$

$$\mu^{n}=(\phi^{n})^{3}-\phi^{n}-\varepsilon^{2}\Delta\phi^{n},\qquad M^{n}=1-(\phi^{n})^{2}$$

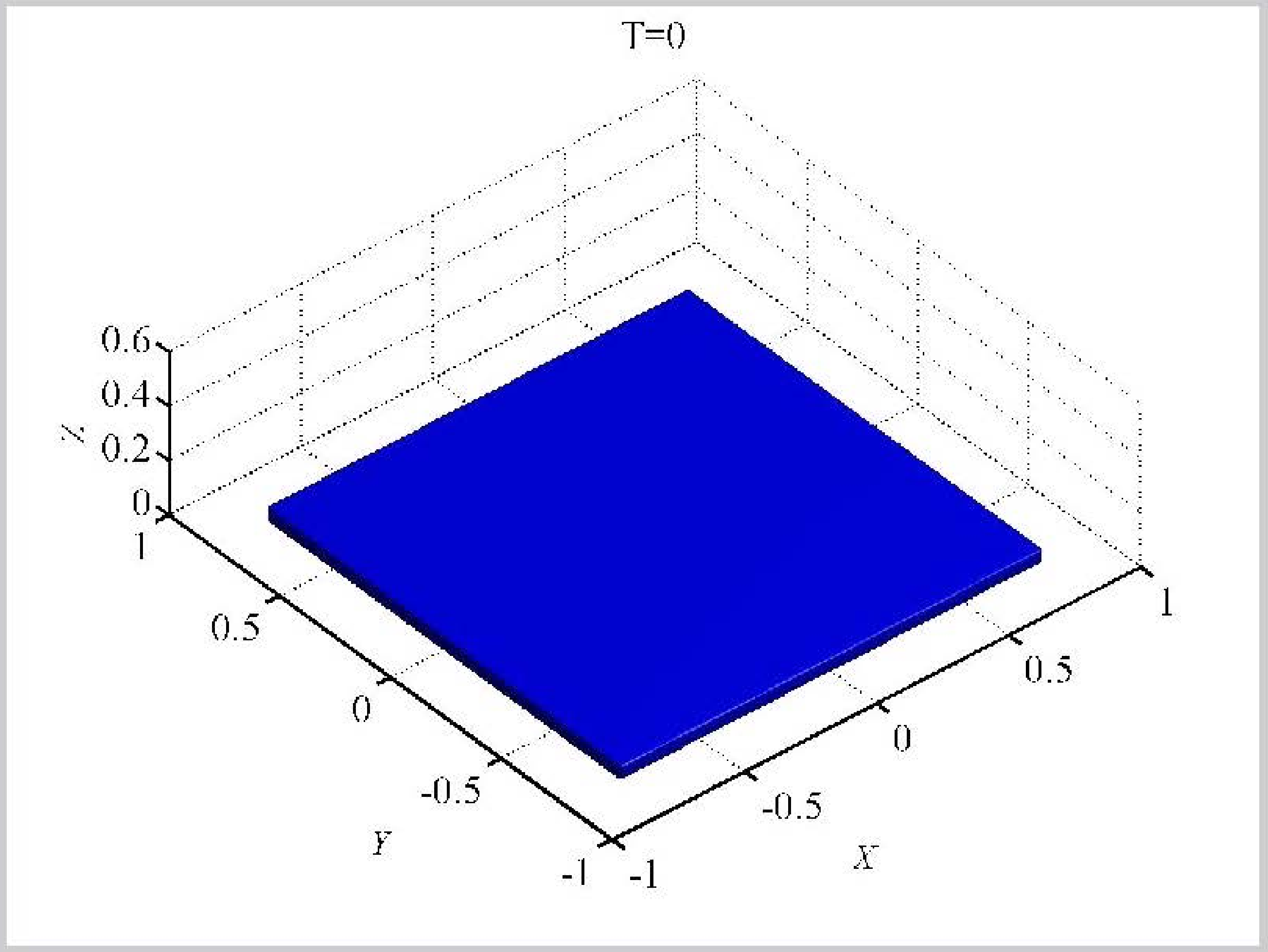
  - *A* and *S* are two stabilizing constants
  - *x*-direction by cosine pseudospectral method
  - *y*-direction by finite difference/finite element
  - 1st order in time & can be upgraded to 2nd
  - Can be solved efficiently at every step & Extension to 3D is easy

[1] W. Jiang, W. Bao, C.V. Thompson &D. J. Srolovitz, Acta Mater. 60(2012), 5578.

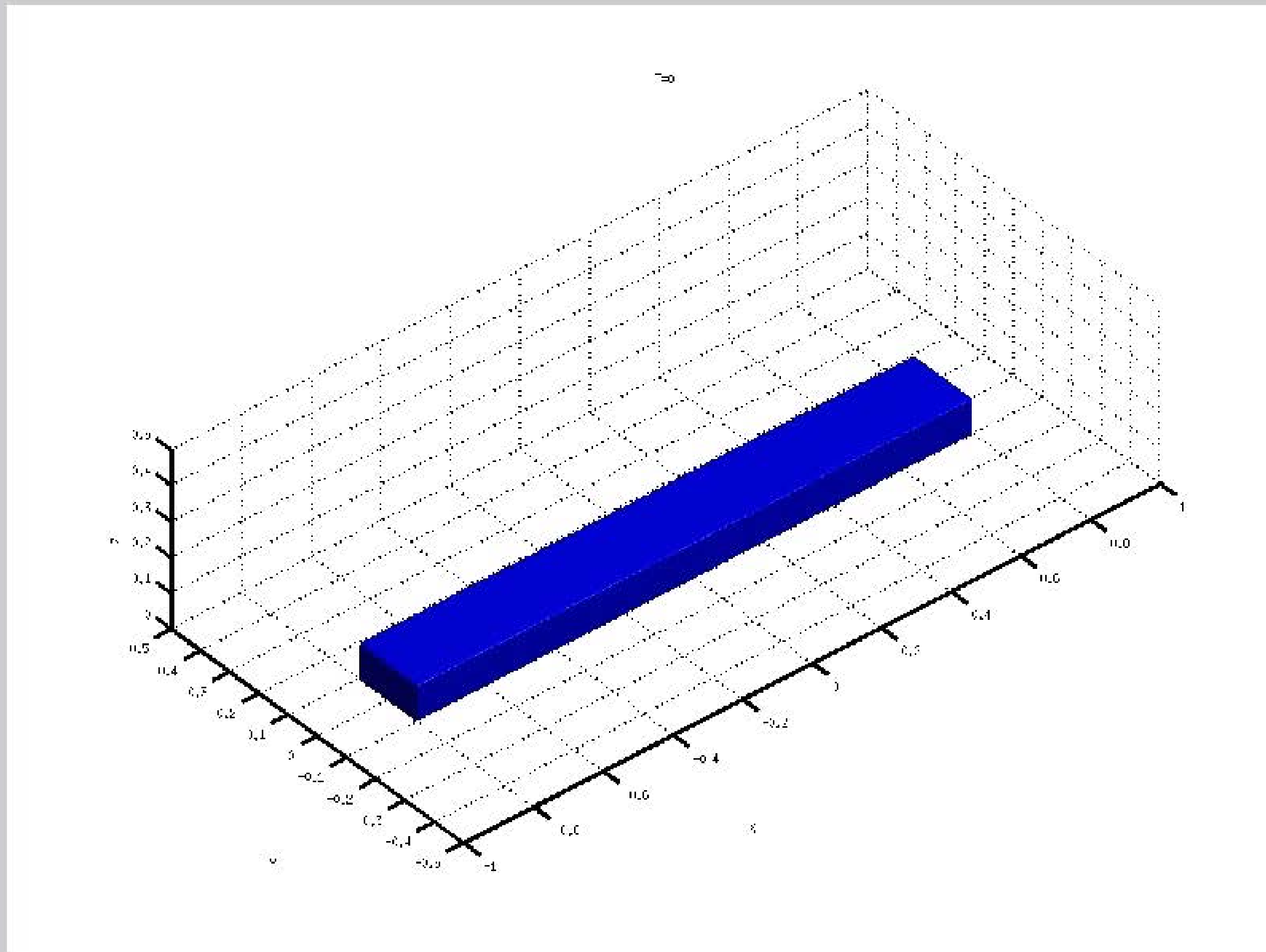
[2] J.Z. Zhu, L.Q. Chen, J Shen & V. Tikare, Phys. Rev. E, 60 (1999), 3564.

[2] J. Shen & X.F. Yang, SIAM J. Sci. Comput., 32 (2010), 1159.

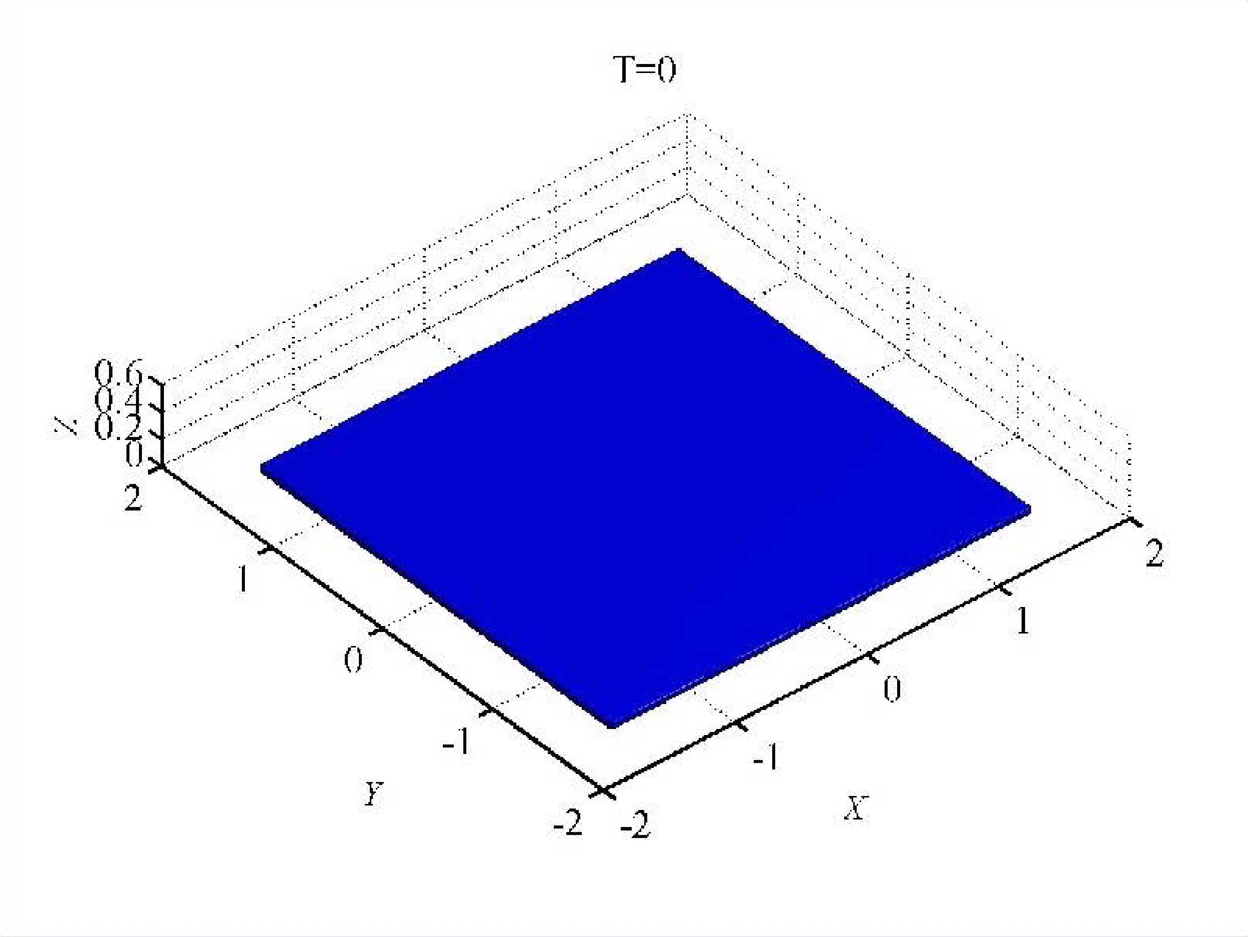
3D Results I -- Square $\theta_s = 5\pi / 6$



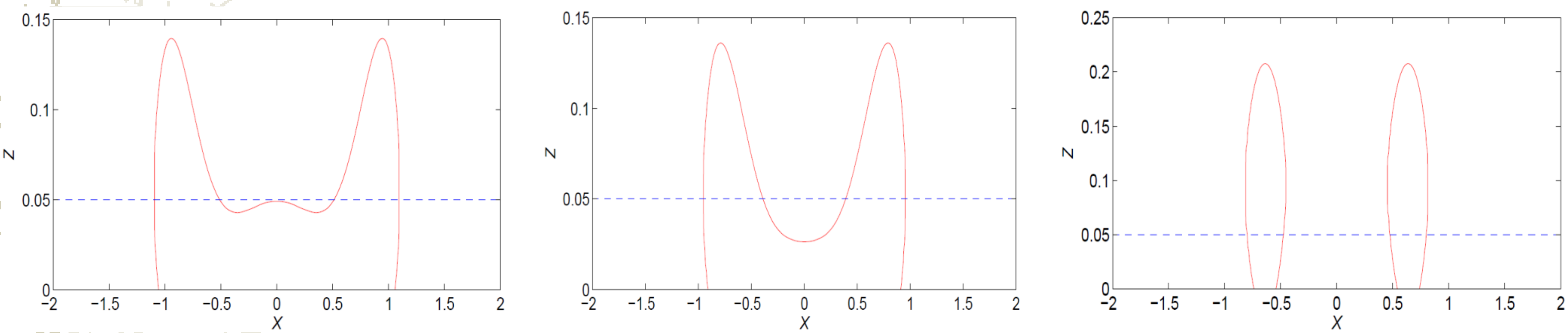
3D Results II – Pinch-off $\theta_s = 5\pi / 6$



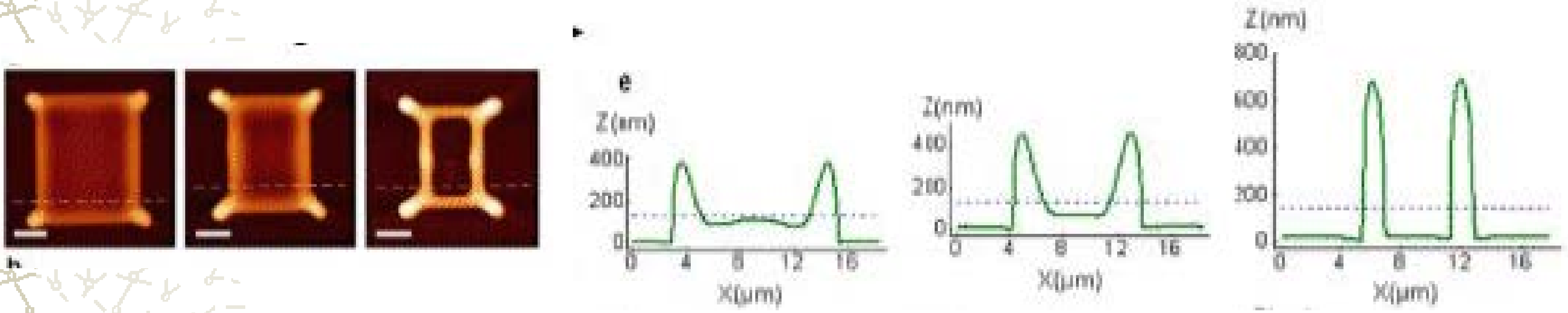
3D Results III – Large-Thin Square



3D Results – Comparison with Experiment



Computational Results



Experimental Results

Conclusion & Future Works

- Conclusion
  – Propose sharp interface model (SIM) for anisotropic surface energy
  – Propose phase field model – Cahn-Hilliard Eq. with function-dependent mobility
  – Design stable, efficient & accurate numerical methods
  – Test parameters regimes & simulate 2D & 3D results
  – Compare qualitatively with experimental / asymptotic results
- Future works
  – Phase field model with anisotropic surface energy
  – Investigate role of different surface energy anisotropy & regularization
  – Develop adaptive & parallel numerical method
  – Mathematical analysis of the models
  – Compare with experiments quantitatively & guide new experiments