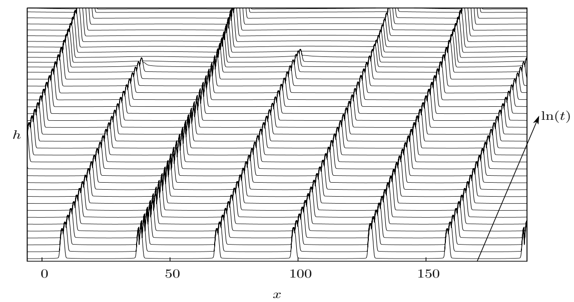
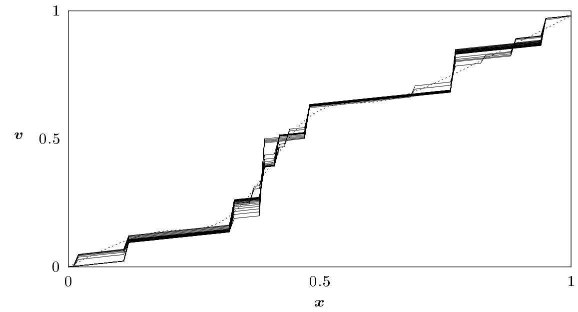
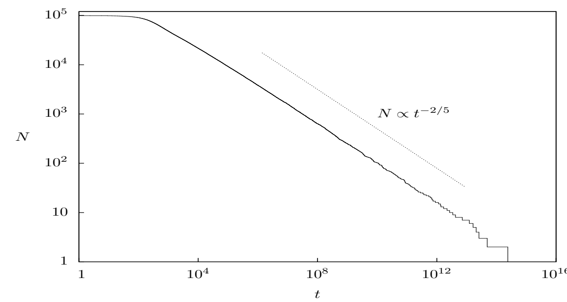
Coarsening: transient and self-similar dynamics in 1-D







Michael Gratton (Northwestern) and Thomas Witelski (Duke)

- **Cahn-Hilliard PDE** models: destabilizing 2nd, regularizing 4th order terms
- Image processing (**Perona-Malik**), materials science (**Ostwald ripening**), fluid dynamics (**Dewetting**), granular materials, ...
- Short-time behavior: linear instability "spinodal decomposition" or "roughening" $\longrightarrow$ formation of arrays of near-equilibrium **localized structures**
- Long-time behavior: **Coarsening** – Energetically favored gradient flow process of re-grouping into fewer structures
  - Coarsening Dynamical System (CDS): **ODEs** for interacting structures
  - Long-long-time behavior: Stationary distribution (**LSW** theory) and statistical scaling law for coarsening (**Kohn-Otto** rate bounds)
  - Short-long-time: Transient dynamics controlled by spatial structure of IC's

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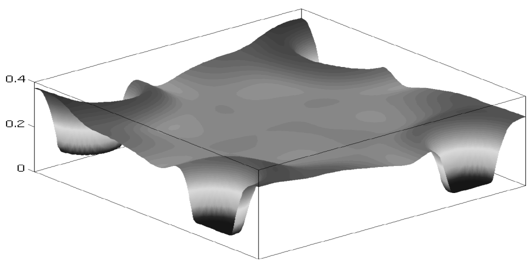
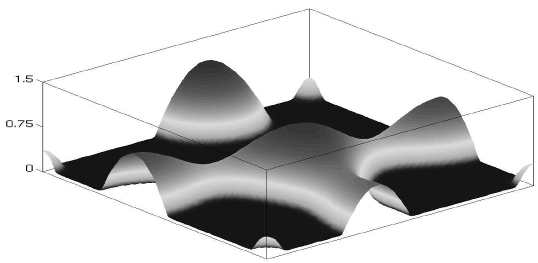
[submitted to Physica D] **[supported by NSF DMS CAREER 0239125]**

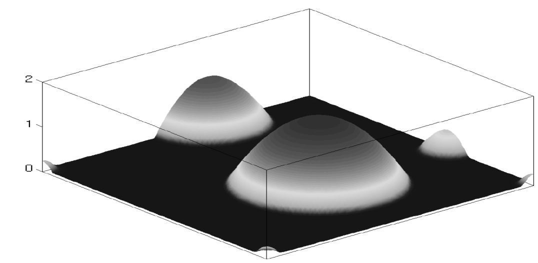
Dewetting: The instability of coatings of viscous fluids on solid surfaces.

Very undesirable for most applications **(painting, printing, microfluidic devices)**.

- Lubrication models of flows of viscous fluid films on hydrophobic solids.
- Multi-scale dynamics controlled by *materials properties*.
- **Dewetting**: pattern forming instabilities – break-up of layers into droplets





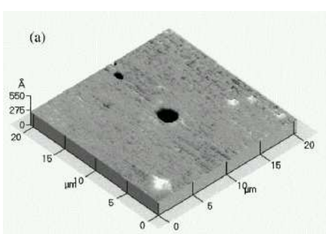


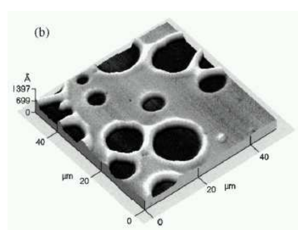
- Long-time behavior: **Coarsening** – re-grouping into fewer larger droplets

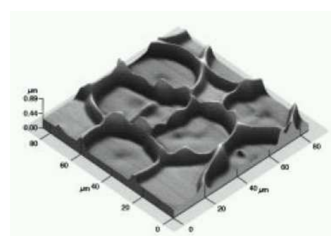
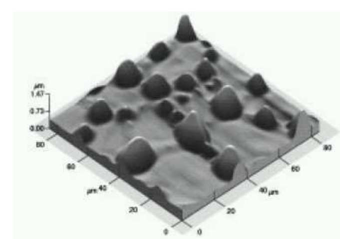
Observed non-uniformities in experiments for polymer films on SiO

Holes $\longrightarrow$ complex patterns **(fractals?)** $\longrightarrow$ polygonal ridges $\longrightarrow$ droplets

Increasing time $\longrightarrow$

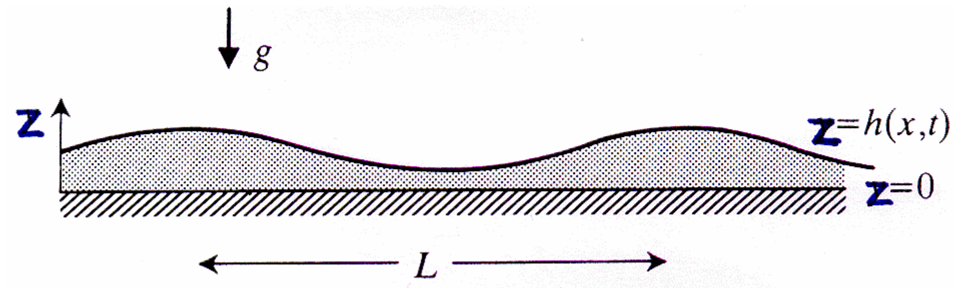




[P. F. Green et al, 2001]

Classical lubrication models for thin viscous films



Fluid volume: $\quad 0 \le x, y \le L \qquad 0 \le z \le H(x,y,t)$

- Navier-Stokes eqns: velocity field $\vec{\mathbf{u}}$ for viscous incompressible flow

$$\mathrm{Re}\frac{D\vec{\mathbf{u}}}{Dt} = -\nabla p + \nabla^2 \vec{\mathbf{u}} \qquad \nabla \cdot \vec{\mathbf{u}} = 0$$

- Low Reynolds number creeping flow limit: Stokes flow

$$\vec{0} = -\nabla p + \nabla^2 \vec{\mathbf{u}} \qquad \nabla \cdot \vec{\mathbf{u}} = 0$$

- Asymptotics in the aspect ratio: $\delta = H/L \to 0$
- Boundary conditions at $z = 0$ and $z = h(x,y,t)$

The Reynolds lubrication equation in 1-D

$$\boxed{\frac{\partial h}{\partial t} = \frac{\partial}{\partial x}\left(h^3 \frac{\partial p}{\partial x}\right)}$$

$h = h(x,t)$: film thickness

$J = -h^3 \partial_x p$: mass flux

$p = p[h]$: dynamic pressure

Contributions to the pressure

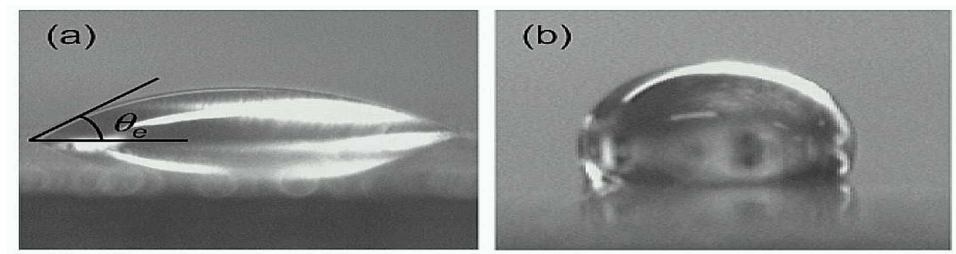
$$\boxed{p \equiv \Pi(h) - \frac{\partial^2 h}{\partial x^2}}$$

1. Surface tension of the free surface (linearized curvature of h)
2. Fluid-solid intermolecular forces: chemical properties of the solid and fluid

   Wetting/non-wetting interactions described by a potential $U(h) \Rightarrow$

   disjoining pressure: $\boxed{\Pi(h) = \frac{dU}{dh}}$

   ex: Scotchgard, Teflon, TurtleWax,...



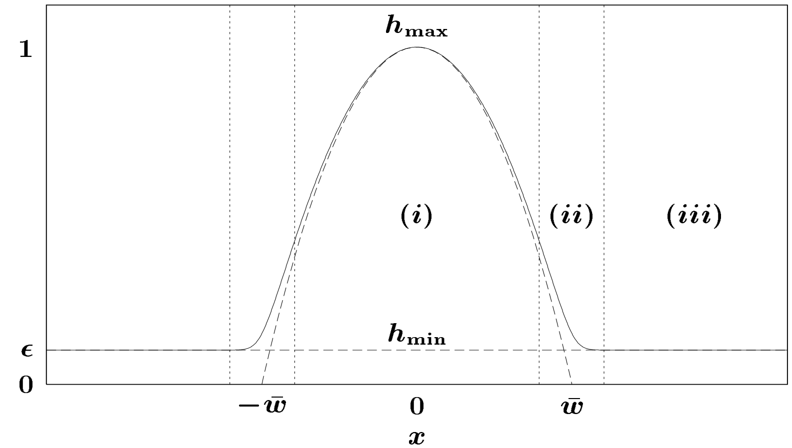
The dewetting model:

$$\boxed{\frac{\partial h}{\partial t} = \frac{\partial}{\partial x}\left(h^3 \frac{\partial}{\partial x}\left[\Pi(h) - \frac{\partial^2 h}{\partial x^2}\right]\right)} \qquad \Pi(h) = \frac{1}{\epsilon}\left(\frac{\epsilon}{h}\right)^3\left[1 - \left(\frac{\epsilon}{h}\right)^n\right]$$

- $n \geq 1$
- $\epsilon > 0$: thickness scale for equilibrium "Ultra-thin film" (UTF), $h(x,t) \geq \epsilon$
- Cahn-Hilliard-like model with degenerate mobility
- early stage instabilities called "spinodal dewetting"

1-D Equilibrium fluid droplets: localized steady states $h = H(x)$

$p[H] = \mathrm{const}(\bar{p}) \implies$ Phase plane



$$\boxed{\frac{d^2H}{dx^2} = \Pi(H;\epsilon) - \bar{p}}$$

Droplet: homoclinic solution

Asymptotic structure for $\epsilon \to 0$

(i) Droplet core: parabolic profile for $|x| \ll \bar{w}$

$$\boxed{H(x) \sim \tfrac{1}{2}\bar{p}(\bar{w}^2 - x^2)}$$

(ii) Contact line: matching region $|x| \sim \bar{w}$ determines width and contact angle

$$\left.\frac{dH}{dx}\right|_{x\to-\bar{w}} = A = \sqrt{2|\Pi'(\epsilon)|} \qquad \boxed{\bar{w}(\bar{p}) = \frac{A}{\bar{p}}}$$

Droplet mass $\sim$ mass of core region

$$\bar{m}(\bar{p}) = \int_{-\bar{w}}^{\bar{w}} H(x)\,dx \sim \frac{2A^3}{3\bar{p}^2}$$

Dynamics of a single near-equilibrium droplet on $-L \le x \le L$

Initial condition: $h(x,0) = H(x;\bar{p})$

Boundary conditions: small mass fluxes imposed

$$J(-L) = \sigma \tilde{J}_- \qquad J(L) = \sigma \tilde{J}_+$$

- Fluxes set a slow timescale, $\sigma \ll 1$: $\boxed{\tau = \sigma t}$
- Fluxes drive the slow evolution of the droplet. They change the droplet's position $X(\tau)$ and pressure (mass) $P(\tau)$:

$$h(x,t) = \boxed{H(x - X(\tau); P(\tau))} + \sigma h_1(x,\tau) + O(\sigma^2)$$

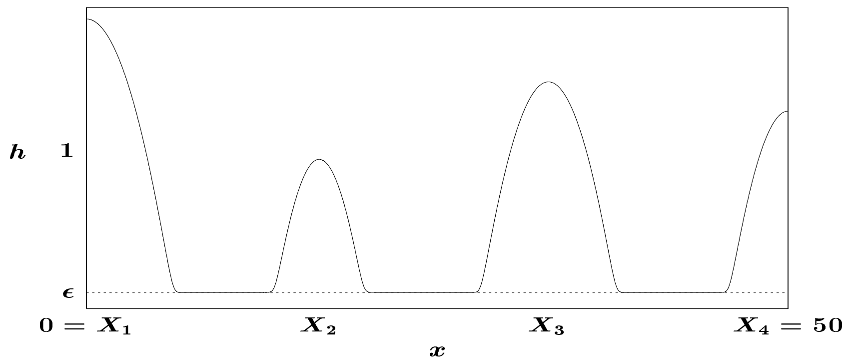
- Equations for droplet evolution derived from the solvability conditions for the $h_1(x,\tau)$ eqn:

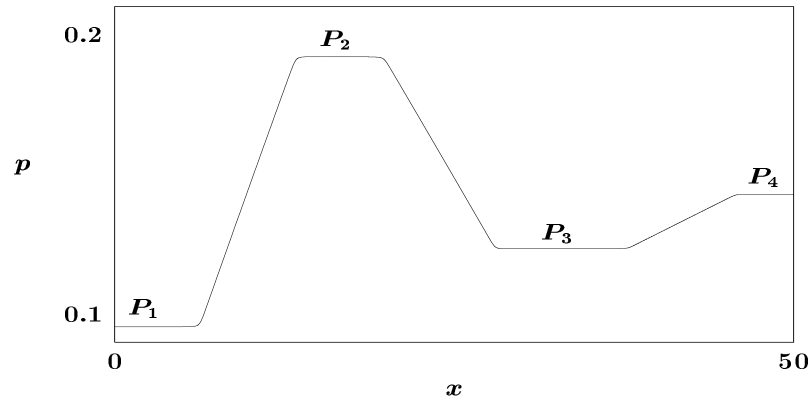
$$\boxed{\frac{dP}{dt} = C_P(P)(J_+ - J_-) \qquad \frac{dX}{dt} = -C_X(P)(J_+ + J_-)}$$

- The coefficient functions $C_P(P), C_X(P)$ are given by integrals of the equilibrium droplet $H(x;P)$.

Dynamics of arrays of droplets

- Widely separated droplets: each has a locally constant pressure
- Differences in the pressures will generate fluxes through the UTF





- Can define a potential fcn, $J = -\partial_x V(p)$
- The flux between droplet k and its neighbor, $k+1$:

$$J_{k,k+1} = -\frac{V(P_{k+1}) - V(P_k)}{[X_{k+1} - \bar{w}(P_{k+1})] - [X_k + \bar{w}(P_k)]}$$

- Evolution equations for an array of N droplets, $k = 1, 2, \cdots, N$, are

$$\frac{dP_k}{dt} = C_P(P_k)(J_{k,k+1} - J_{k-1,k}) \qquad \frac{dX_k}{dt} = -C_X(P_k)(J_{k,k+1} + J_{k-1,k})$$

Nonlinear system with nearest-neighbor coupling

"Interacting particle system" with finite-time singularities:

some $P_k \to \infty$ ($\mathrm{Mass}_k \to 0$) – droplet collapse

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[Glasner and Witelski: PRE 2003, Physica D 2005]

Coarsening Dynamical System (CDS) =

Near-equilibrium ODE system + Coarsening rules

- For $t < t_1$

$$\left\{ \frac{dX_k}{dt} = \cdots \qquad \frac{dP_k}{dt} = \cdots \right\} \qquad k = 1, 2, \cdots, N$$

- At $t = t_1^-$, the soln of ODEs satisfies a *detection condition*. Stop the ODEs. Remove collapsed drop. Create new IC's at $t = t_1^+$ for remaining $N-1$ drops via *coarsening rules* for collision or collapse:

$$\left\{ X_k(t_1^+) = \mathcal{X}[X_k, P_k; t_1^-] \quad P_k(t_1^+) = \mathcal{P}[X_k, P_k; t_1^-] \right\} \quad k = 1, \cdots, N-1$$

- For $t_1^+ \le t < t_2$

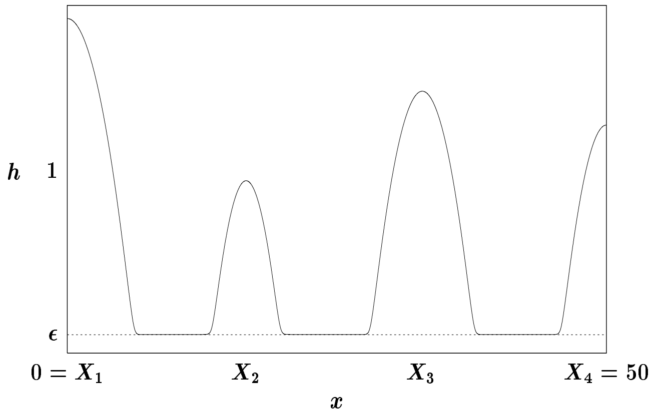
$$\left\{ \frac{dX_k}{dt} = \cdots \qquad \frac{dP_k}{dt} = \cdots \right\} \qquad k = 1, 2, \cdots, N-1$$

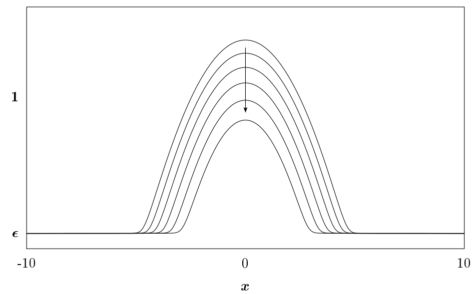
- At $t = t_2^-$ another coarsening event detected...
(rinse and repeat, $N \to N-1 \to N-2 \to \cdots$)

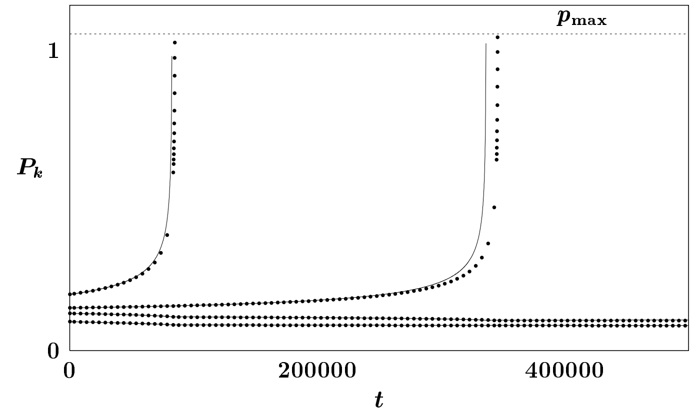
Coarsening via droplet collapse:

Collapse: when $\text{Mass}_k \to 0$ ($P_k \to \infty$) for one drop

Initial Conditions **Evolution of Pressures**







[PDE results (dots), ODE model (curves)]

$$\frac{dP_k}{dt} \approx \frac{P_k^4}{L} \qquad \longrightarrow \qquad P_k(t) \propto ([T_c - t]/L)^{-1/3}$$

Heuristic scaling argument: statistics for $N \gg 1$

L: mean spacing between drops on fixed domain, $L = O(1/N)$

P_0: typical drop pressure, $P_0 = O(M_0^{-1/2})$

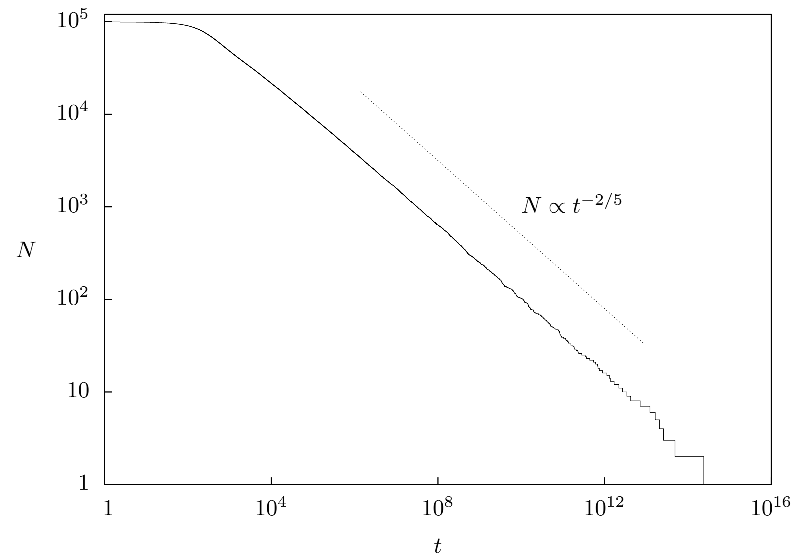
M_0: typical mass $M_0 = O(1/N)$

T_c: critical collapse time, $T_c = O(P_0^{-3} L)$

Assume independent, uncorrelated events:

$$\frac{1}{N}\frac{dN}{dt} = -\frac{1}{T_c} \qquad \longrightarrow \qquad \boxed{N(t) = O(t^{-2/5})}$$

Coarsening statistics



Upper bound on coarsening rate* can be rigorously established via two estimates on the energy of the system **[Kohn and Otto 2002]**

PDE conserves mass and dissipates energy

$$E = \int U(h) + \tfrac{1}{2} h_x^2 \, dx = O(N^{1/2}) \qquad \frac{dE}{dt} = -\int h^3 p_x^2 \, dx \le 0$$

[Otto, Rump, and Slepcev 2006]: Thin film PDE $\longrightarrow N(t) \geq O(t^{-2/5})$

[Esedoglu et al]: Coarsening in image processing PDE models

Alternative approach to analysis: reduce PDE to CDS and study dynamics of the CDS, ala **[Dai and Pego 2005]**

Simplified CDS model

$$J_{k,k+1} = -\frac{V(P_{k+1}) - V(P_k)}{[X_{k+1} - \bar{w}(P_{k+1})] - [X_k + \bar{w}(P_k)]} \approx -\frac{V(P_{k+1}) - V(P_k)}{X_{k+1} - X_k}$$

- Dilute limit (narrow drops): $\Delta X \gg \bar{w}$
- Define separations: $L_k \equiv X_k - X_{k-1}$
- No-drift limit: $dX_k/dt \to 0$ so $X_k \approx$ const (until deletion)
- Focus on droplet masses: $P_k \to M_k^{-1/2}$

$$\boxed{\frac{dM_k}{dt} = \frac{M_{k+1}^{-1/2} - M_k^{-1/2}}{L_{k+1}} - \frac{M_k^{-1/2} - M_{k-1}^{-1/2}}{L_k}}$$

- Conserves mass

$$\frac{d}{dt}\left(\sum M_k\right) = 0$$

- Same gradient flow, $E = O(N^{1/2})$

$$E = \sum M_k^{1/2} \qquad \frac{dE}{dt} = -\tfrac{1}{2}\sum \frac{\left(M_{k+1}^{-1/2} - M_k^{-1/2}\right)^2}{L_{k+1}} \leq 0$$

Even-more-Simplified CDS model

$$\frac{dM_k}{dt} = \frac{M_{k+1}^{-1/2} - M_k^{-1/2}}{L_{k+1}} - \frac{M_k^{-1/2} - M_{k-1}^{-1/2}}{L_k}$$

- Replace individual separations by average $L_* = L_{total}/N$

$$\frac{dM_k}{dt} = \frac{M_{k+1}^{-1/2} - 2M_k^{-1/2} + M_{k-1}^{-1/2}}{L_*}$$

- Replace neighbors by average (mean field pressure)

$$M_*^{-1/2} = \frac{1}{N} \sum_k M_k^{-1/2}$$

$$\boxed{\frac{dM_k}{dt} = \frac{2}{L_*} \left(M_*^{-1/2} - M_k^{-1/2} \right)}$$

Discrete mean field model **(DMF)**

- Discrete collapse events change $N(t)$ and L_* (piecewise const)
- $M_*(t)$ changes continuously
- Global coupling through M_*, no "nearest neighbor" preference

Lifshitz-Slyozov-Wagner (LSW) continuous mean field model

- Evolution of the distribution of drop sizes: $\phi(m,t)$

$$N(t) = \int_0^\infty \phi(m,t)\,dm \qquad M_{total} = \int_0^\infty m\phi(m,t)\,dm$$

- Conservation law for droplets for $m > 0$

$$\frac{\partial \phi}{\partial t} + \frac{\partial}{\partial m}\left[u(m)\phi\right] = 0$$

Flux from DMF model

$$u(m) = \frac{dm}{dt} = \frac{2}{L_*}\left(\frac{1}{\sqrt{m_*}} - \frac{1}{\sqrt{m}}\right)$$

Mean-field averages

$$\frac{1}{\sqrt{m_*}} = \frac{1}{N}\int_0^\infty \frac{1}{\sqrt{m}}\phi(m,t)\,dm \qquad L_* = \frac{L_{total}}{N}$$

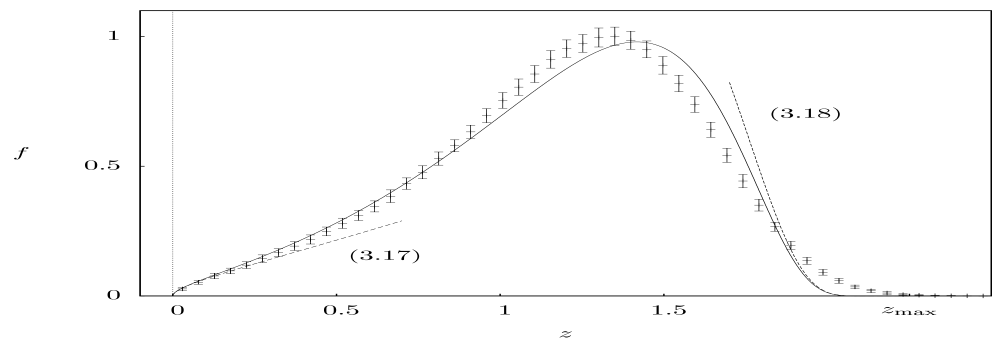
A nonlocal model...

- Discrete collapse events are "averaged out"

  m_*, L_*, N all evolve continuously

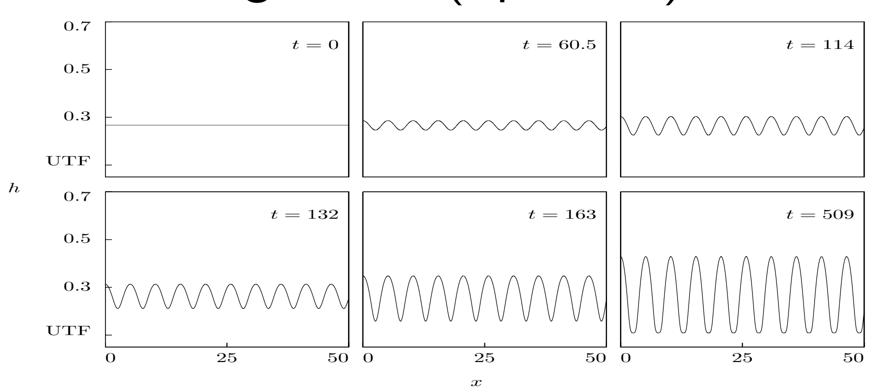
LSW model (continued) Stationary distribution – self-similar solution for ϕ

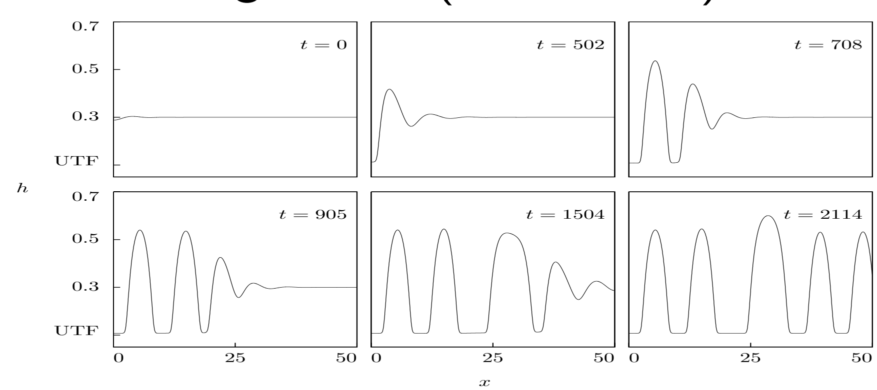
$$\phi(m,t) = t^{-4/5} f(\eta) \qquad \eta = \frac{m}{t^{2/5}} \qquad N(t) = O(t^{-2/5})$$



- Issues about uniqueness/convergence **[Niethammer and Pego]**
- Generally good agreement with CDS simulations for $t \to \infty$ asymptotics
- But, too long for comparison with real experiments (polymer films)
- Want to study transient coarsening behaviors: Are there different transients due to differences in IC $\phi_0(m)$ from the two dewetting instability mechanisms?

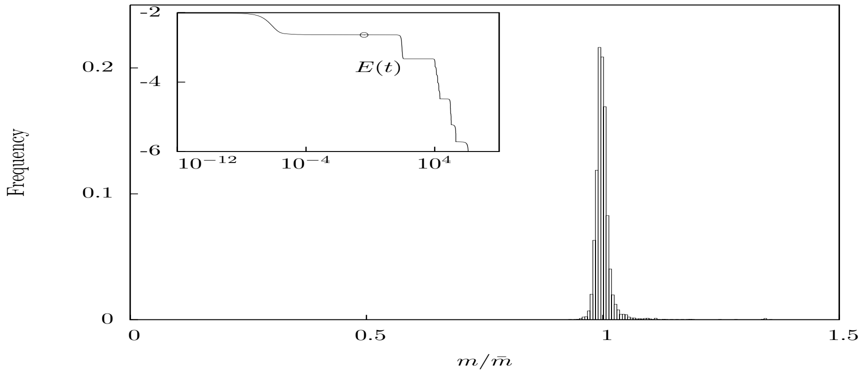
Homogeneous (Spinodal) vs. Heterogeneous (Nucleation)

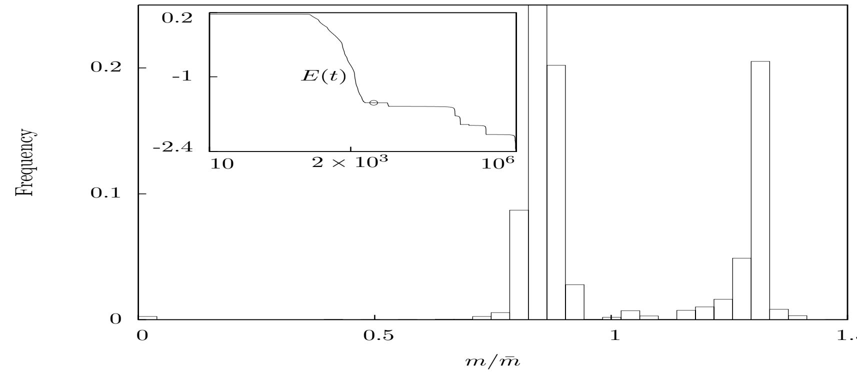




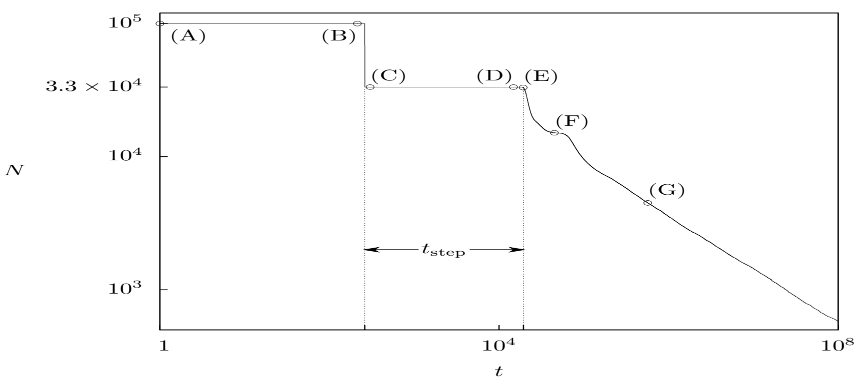
Transient coarsening: Initial distributions $\phi_0(m)$

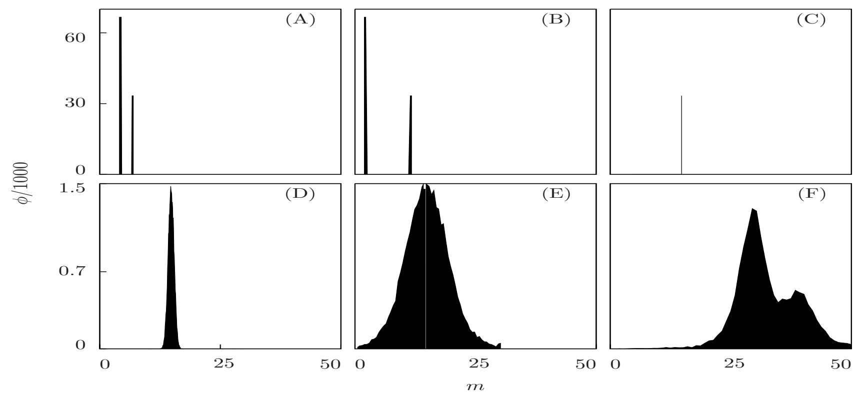
Homogeneous (Spinodal) vs. Heterogeneous (Nucleation)





Simulated nucleated data: CDS simulation



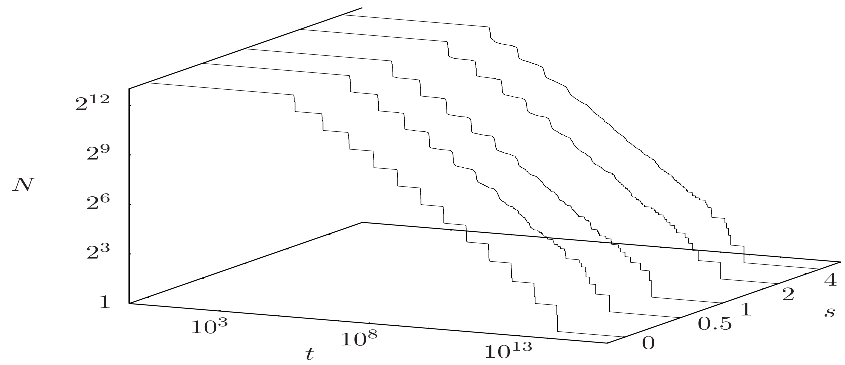


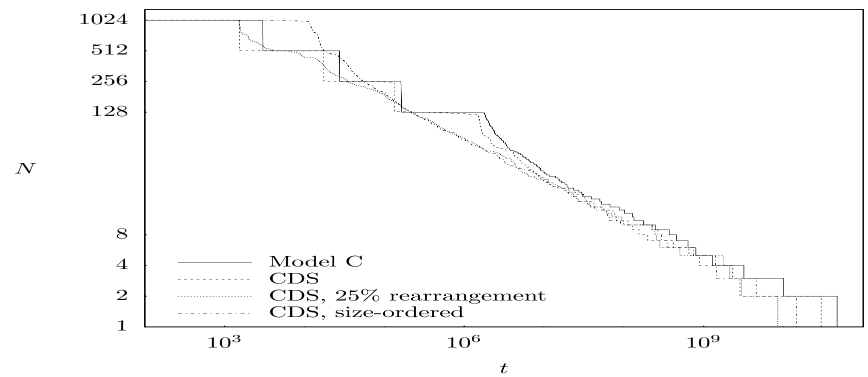
Staircasing behavior

- Concentrations in the mass distribution
- No collapses (plateau) while support of ϕ bounded away from $m = 0$
- An avalanche (cliff) when a concentration propagates to $m = 0$

Transient coarsening: Staircasing behavior

- Observed with CDS, DMF, and other studies of image processing models
- A "sustainable" transient: can recur, depends on population structure





- Sub- and Super-coarsening rates for plateaus/cliffs in staircasing is compatible with the Kohn-Otto bound on **average** coarsening rate