Universality Classes in Coarsening

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19 May 2006

Universality Classes in Coarsening

Coarsening . . .

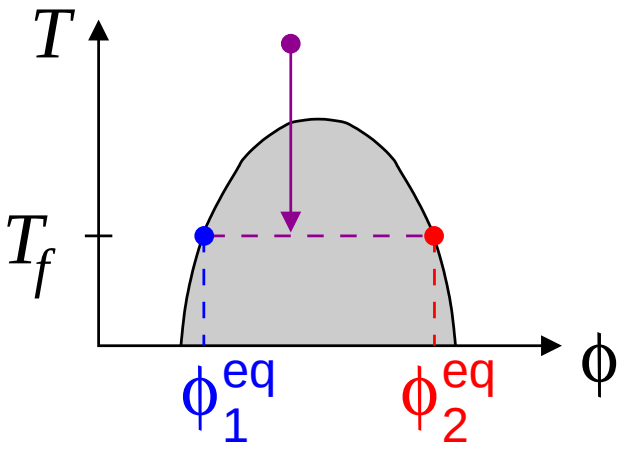
is a nonequilibrium relaxational process in which the characteristic length scale grows with time.

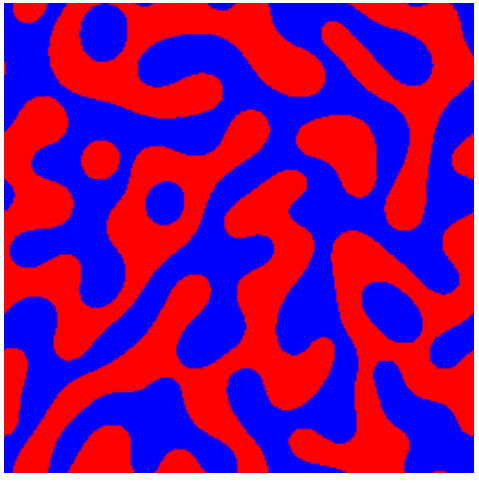
Many examples in nature:

- binary alloys
- polycrystals
- magnetic domains
- binary fluids
- epitaxy
- salad dressing
- polymer blends
- soap froths
- colloids
- liquid crystals
- faceted surfaces
- and more . . .

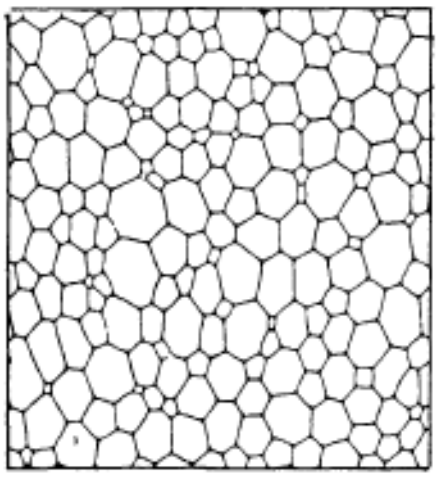
Phase Ordering Dynamics (binary alloys, polymer blends)

- Rapid quench into the forbidden region of a phase diagram
- system responds locally by equilibrating into one of the two phases
- leads to equilibrated domains separated by costly interface
- dissipative dynamics gives coarsening

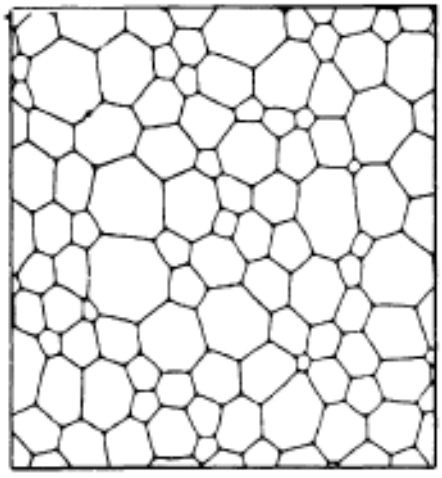




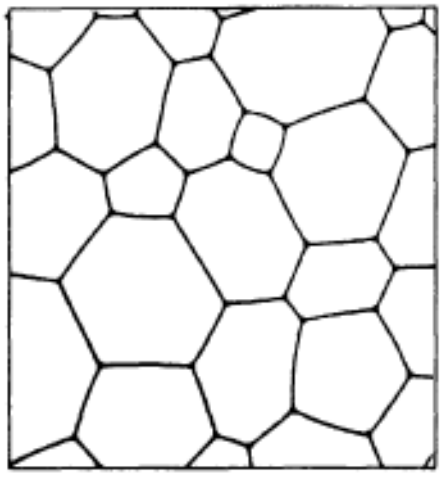
2D Dry Soap Froth



$t = 1.95$ h



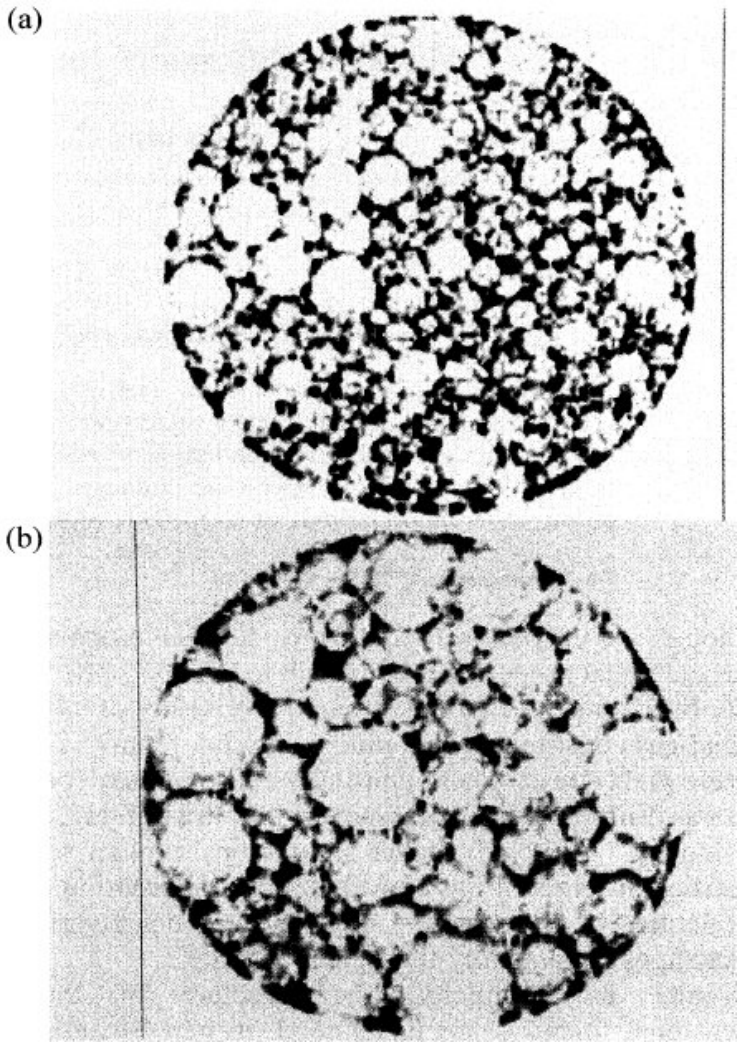
$t = 21.5$ h



$t = 166$ h

Glazier, Gross, and Stavans, *Phys. Rev. A* **36**, 306 (1987).

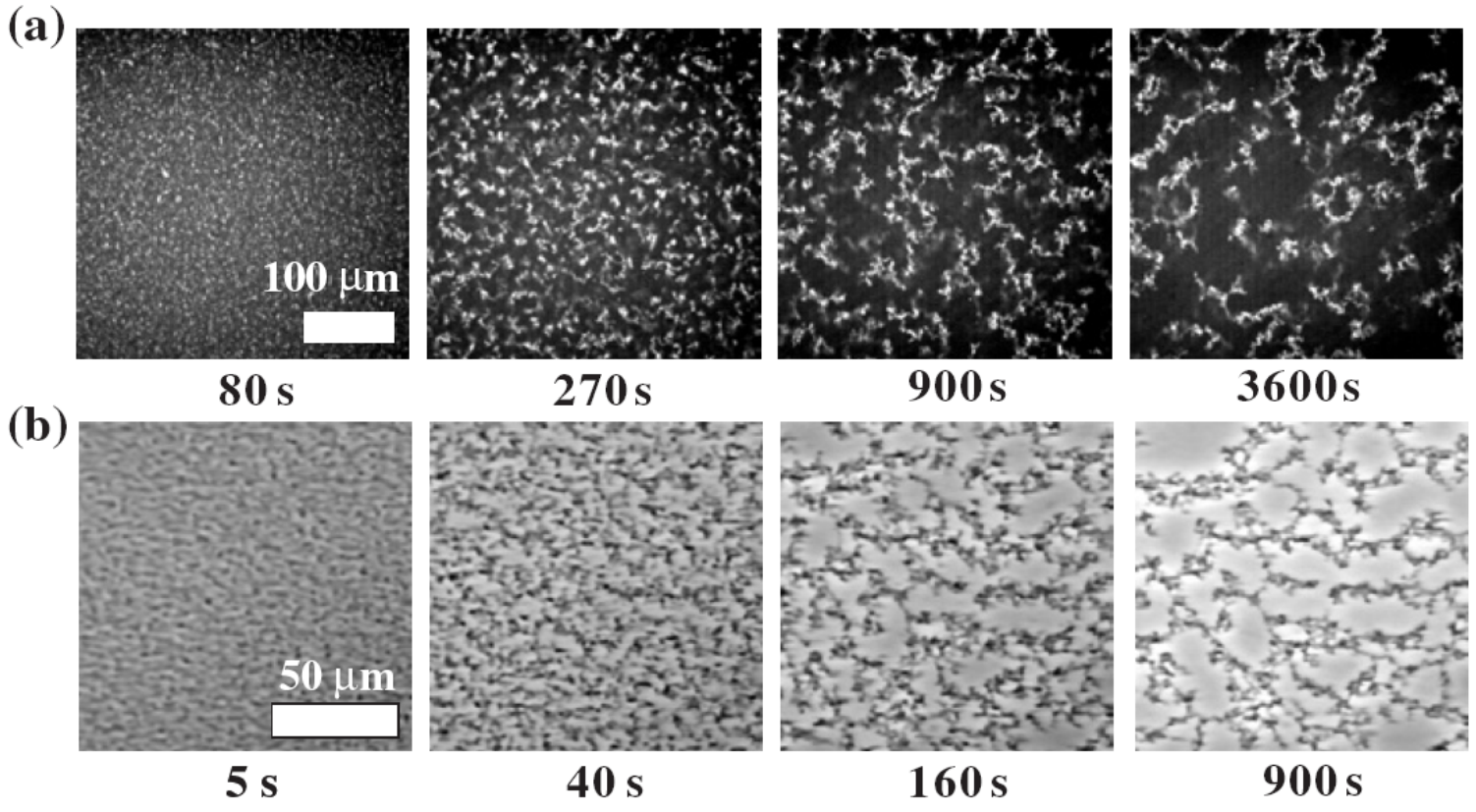
3D Wet Soap Froth



Magnetic Resonance Imaging

Gonata *et al.*, *Phys. Rev. Lett.* **75**, 573 (1995).

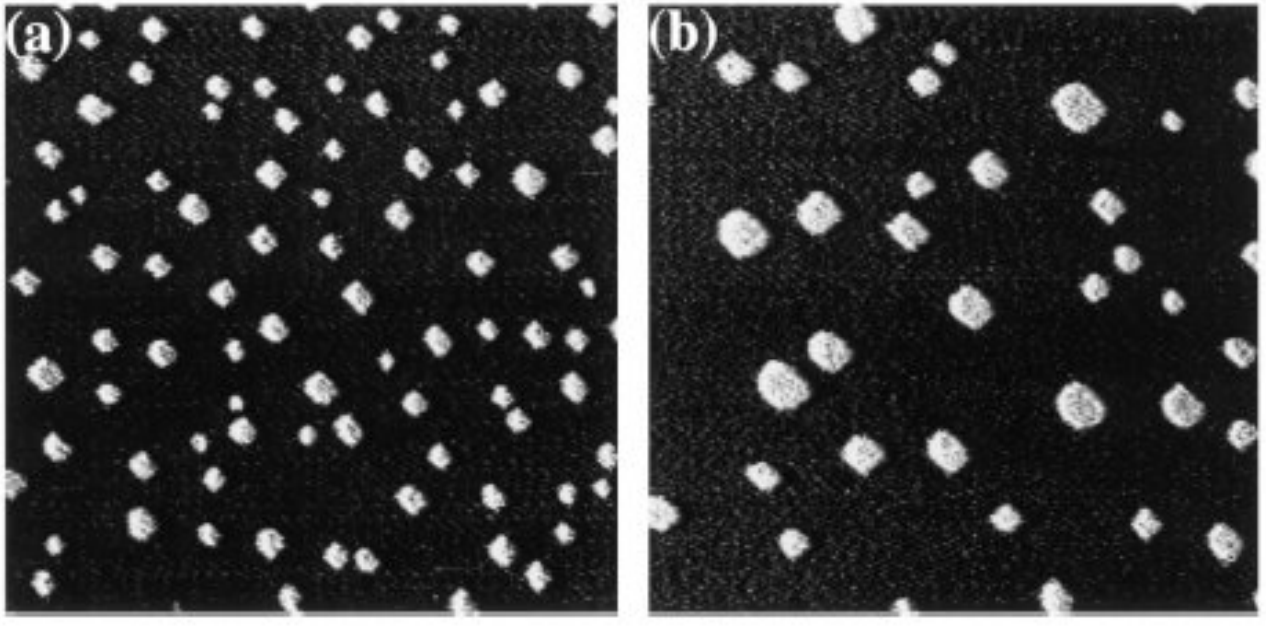
(a) Colloidal Suspension and (b) Polymer Solution



Tanaka, Nishikawa, and Koyama, *J. Phys. Cond. Matt.* **17**, L143 (2005).

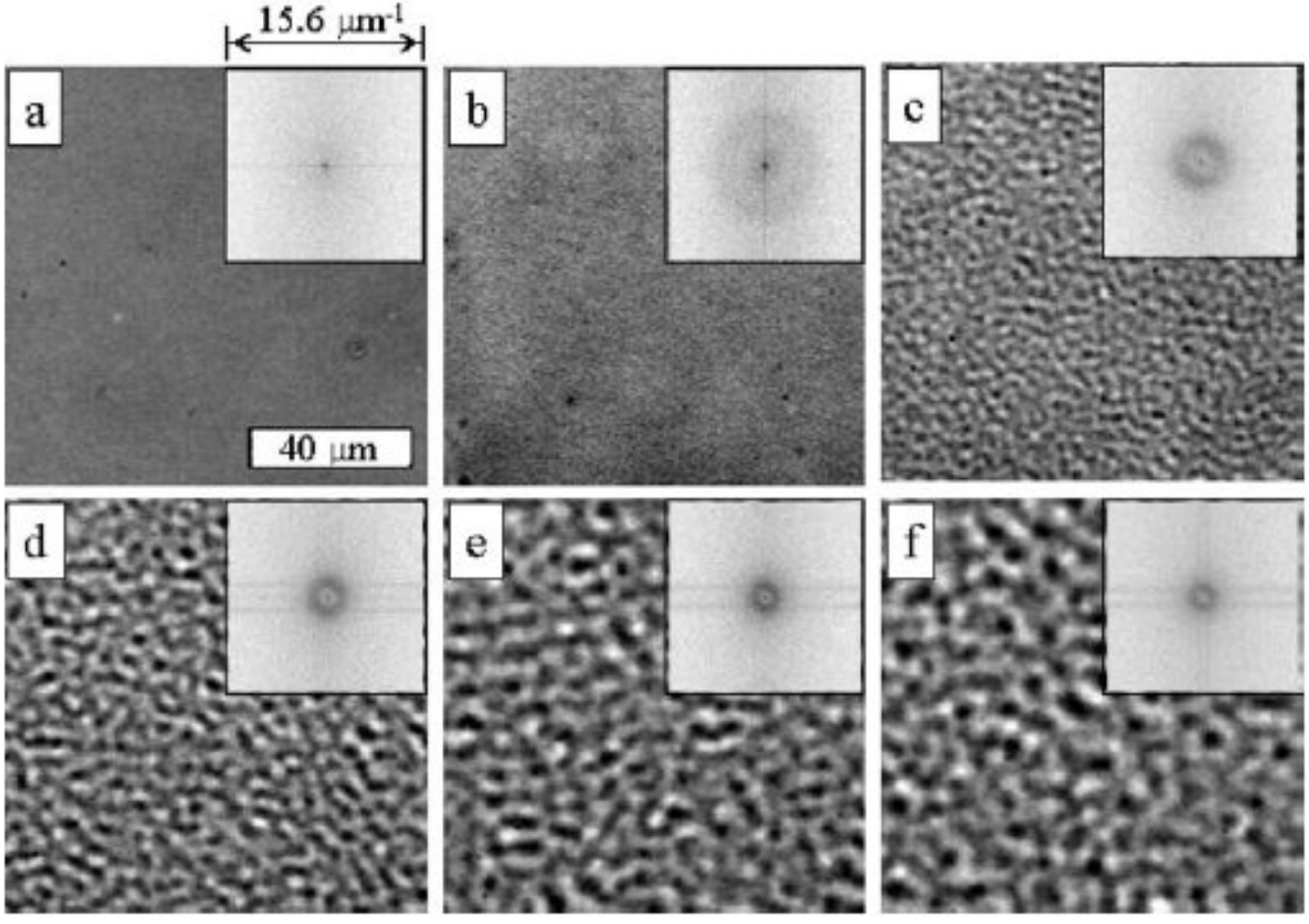
Homoepitaxial Islands

Cu on a Cu(100) surface



Pai *et al.*, *Phys. Rev. Lett.* **79**, 3210 (1997).

Random Copolymers – PEH/PEB

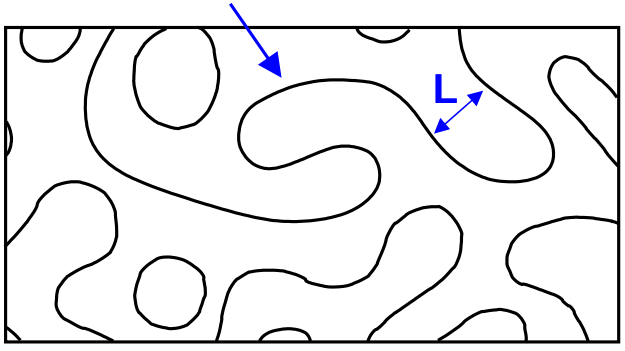


Shimizu *et al.*, *Polymer* **45**, 7061 (2004).

Why is coarsening so common?

Requirements:

- Excess free energy stored in stable, local defects (e.g., domain walls): $F - F_{eq} \propto \rho_{\text{def}}$



- Dissipation: $\dfrac{dF}{dt} < 0 \quad \Rightarrow \quad \dfrac{d\rho_{\text{def}}}{dt} < 0$

Result: growing characteristic length $L(t)$

Basic Features of Coarsening

Sharp defects defect size ξ fixed, so for asymptotically late times $L(t) \gg \xi \Leftrightarrow$ sharp-defect limit.

Self-similarity domain structure statistically invariant when rescaled by $L(t)$.

Implies correlation function scaling
$C(\mathbf{r}, t) = f\big(\mathbf{r}/L(t)\big)$

Power law growth characteristic scale $L \sim t^{\alpha}$

Universality exponent α determined by only a few general features: conservation laws and nature of order parameter

Coarsening Models I: Kinetic Ising Models

Lattice of spins $s_i = \pm 1$, with hamiltonian $H = -J \sum_{\langle ij \rangle} s_i s_j$

Spins initially random ($T_i = \infty$). Quench at time $t = 0$ to $T < T_c$.

Glauber Dynamics

- spins flip with probability determined by energy $\Rightarrow$ nonconserved order parameter.

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Glauber Dynamics

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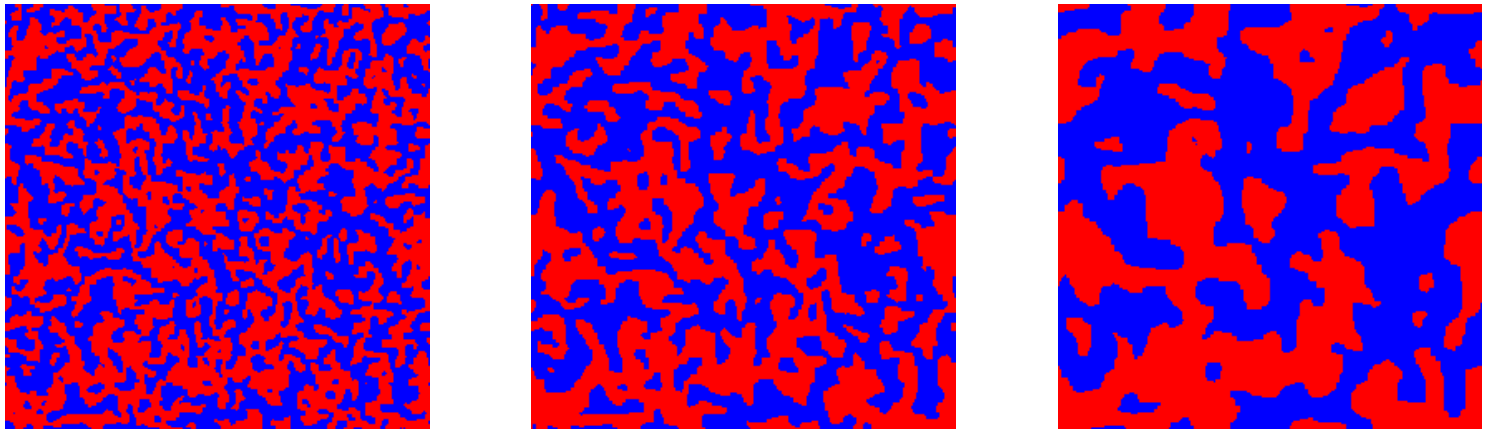
Kawasaki Dynamics

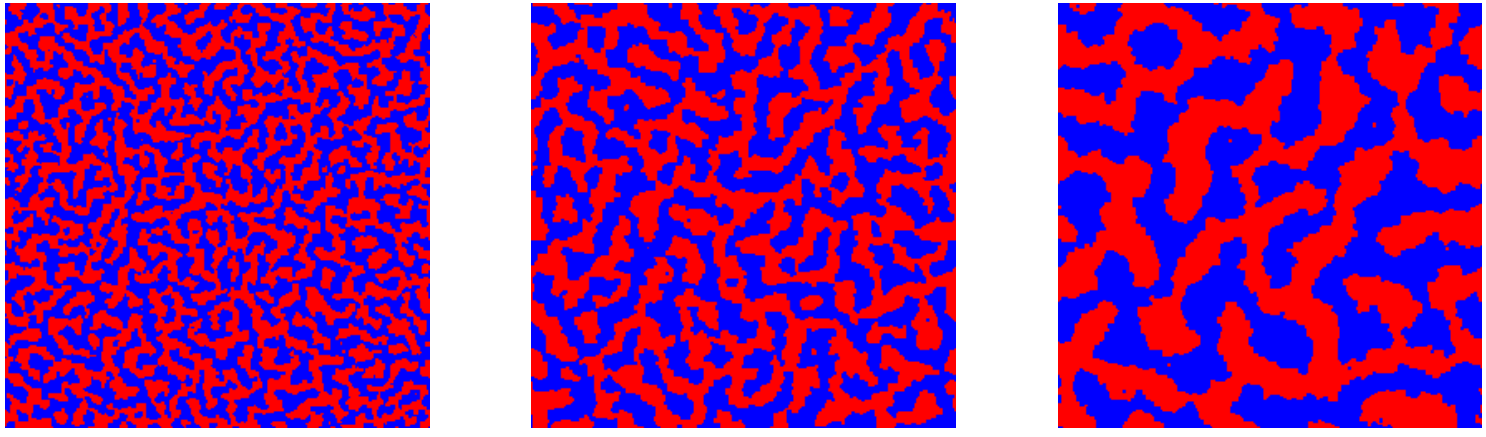
- neighboring spins exchanged $\Rightarrow$ conserved OP.
- volume fraction ϵ additional parameter
- appropriate for binary mixtures: $\uparrow =$ Fe, $\downarrow =$ Al.

Kinetic Ising Models

Glauber (spin flip): nonconserved OP $\longrightarrow L \sim t^{1/2}$



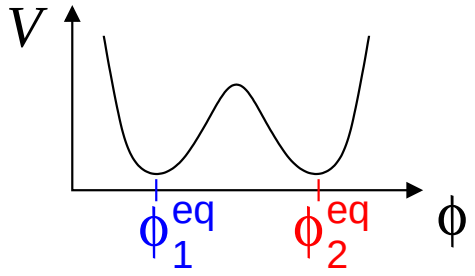
Kawasaki (spin exchange): conserved OP $\longrightarrow L \sim t^{1/3}$



Coarsening Models II: Phase Field Models

Field $\phi(\mathbf{x}, t)$ describes local concentration. Free energy functional:

$$F[\phi] = \int d^d x \{ \tfrac{1}{2}(\nabla\phi)^2 + V(\phi) \}$$



Allen-Cahn equation

Nonconserved OP: $\dfrac{\partial\phi}{\partial t} = -\dfrac{\delta F}{\delta\phi} \quad \Rightarrow \quad \dfrac{\partial\phi}{\partial t} = \nabla^2\phi - V'(\phi)$

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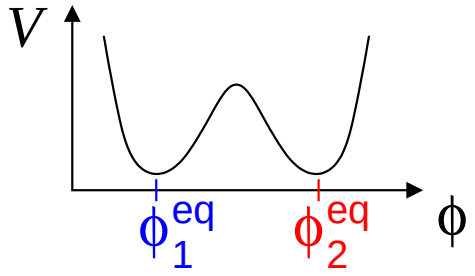

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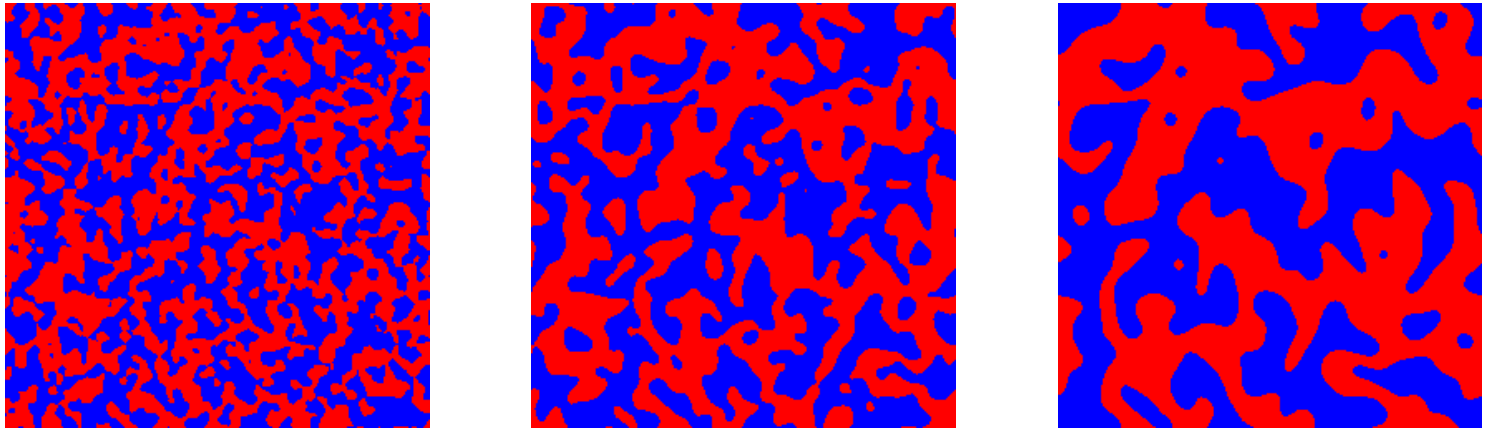
Cahn-Hilliard equation

Conserved OP: $\dfrac{\partial\phi}{\partial t} = -\nabla\cdot\mathbf{J}$ and $\mathbf{J} = -\nabla\dfrac{\delta F}{\delta\phi}$

$$\Rightarrow \frac{\partial\phi}{\partial t} = -\nabla^2[\nabla^2\phi - V'(\phi)]$$

Phase Field Models

Allen-Cahn eq: nonconserved OP $\longrightarrow L \sim t^{1/2}$



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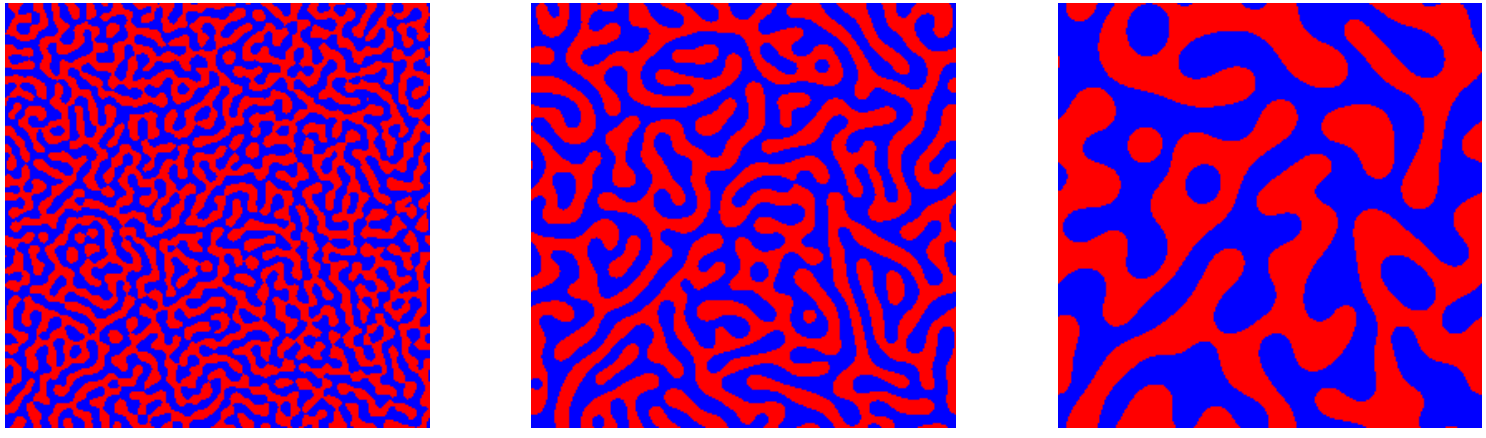
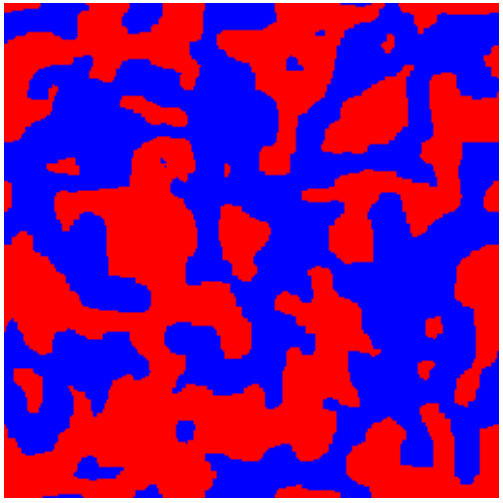


Universality?

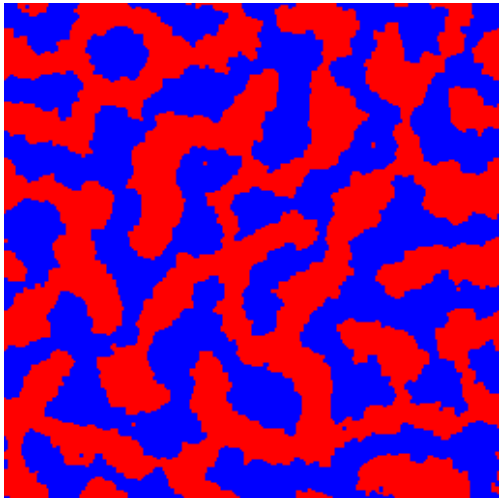
$L \sim t^{1/2}$

Glauber

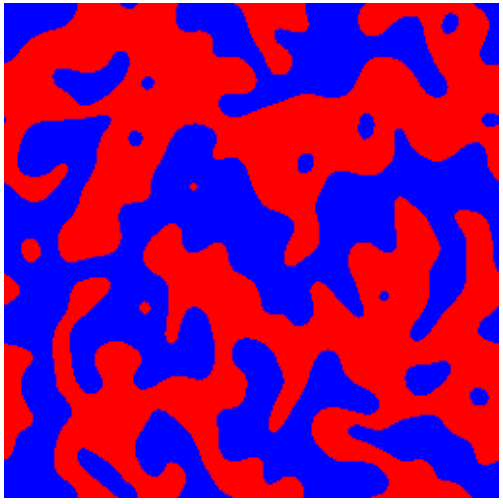


$L \sim t^{1/3}$

Kawasaki



Allen-
Cahn



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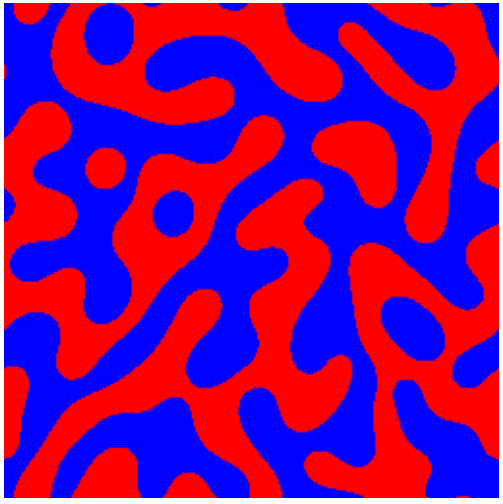
Hilliard



Universality Classes in Coarsening

Theoretical Challenge

- Systems generically evolve into a self-similar scaling state with universal power law growth. This demands explanation!
- Characterizing this asymptotic scaling state, i.e. finding universality classes, a starting point for analysis of real systems.

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Renormalization Group Scenario

- Critical-like behavior suggests a dynamical RG fixed point controlling the asymptotic dynamics.
- Believed to be a $T = 0$ fixed point based on irrelevance of Langevin additive noise ($\propto \sqrt{T}$) for phase field models.
- Not (yet) tractable — a strong-coupling fixed point.

How do we proceed?

Two Routes to Progress

Lifshitz-Slyozov Theory ('58)

- Exact, nontrivial solution of conserved OP coarsening in dilute $\epsilon \longrightarrow 0$ limit (isolated droplets) for all $d \geq 2$.
- Derives scaling state, demonstrates its universality.
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Assume Scaling: then derive consequences.

- Huse ('86) argued COP $t^{1/3}$ extends to all ϵ.
- Bray $T = 0$ RG fixed point scenario ('89) also gives $t^{1/3}$.
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So why am I here talking about universality classes?

Bray-Rutenberg energy scaling predicts growth exponent ($L \sim t^{\alpha}$) universality classes:

- α depends only on conservation law and nature of order parameter
- does not depend on spatial dimension d, volume fraction ϵ, or microscopic details

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Conventional wisdom: correlation function $C(\mathbf{r}, t)$ or structure factor $S(\mathbf{k}, t)$ has same universality as the growth exponent.

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We have recently learned that this is not true!

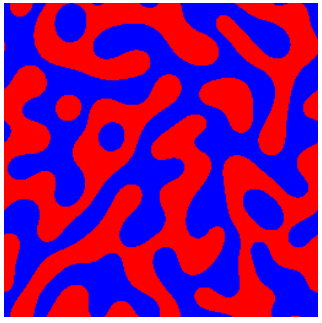
Distinct Universality (for conserved scalar OP)

Quantities that affect the correlation function but not the growth exponent:

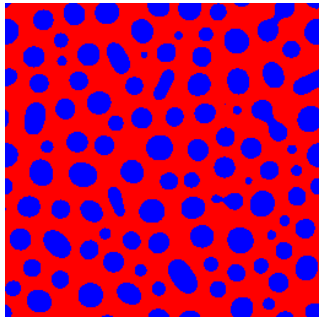
Trivial

- volume fraction ϵ
- spatial dimension d

. . . everyone knew that already.



$\epsilon = 1/2$



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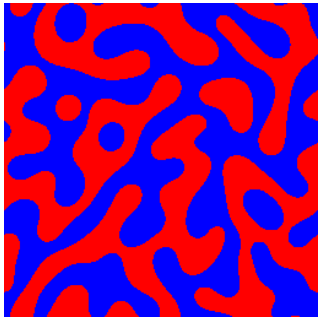

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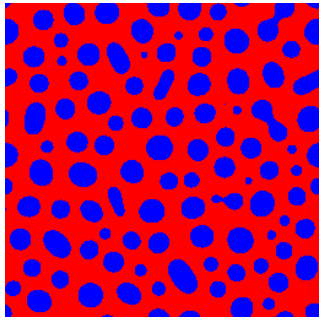
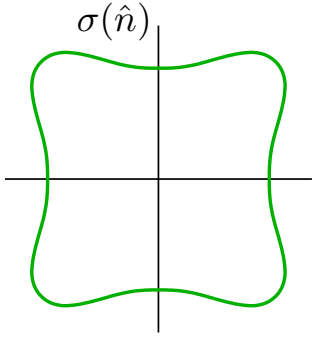


$\epsilon < 1/2$

Less Trivial

- anisotropic surface tension $\sigma(\hat{n})$ (e.g. Ising model)



exact Lifshitz-Slyozov solution for dilute coarsening

[BVL & Rutenberg '99; Gildner, Fowler, and BVL '06]

Questions

- Does the scaled correlation function have any universality?
- If so, what are its universality classes?
- Which quantities belong to which universality classes?

  For example: higher order correlation functions, curvature distribution, autocorrelation exponents, persistence exponents, amplitudes, ...

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My proposed answer:

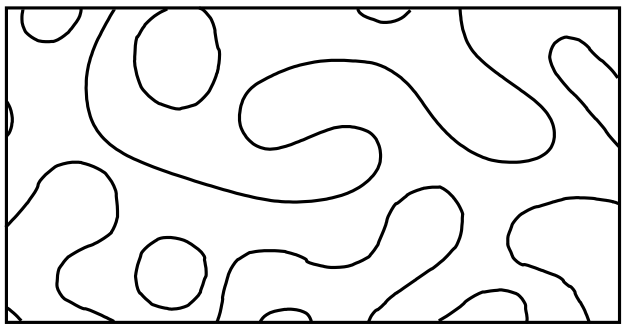
Growth exponents are a special case. For general universality look at the sharp defect dynamics.

Universality Classes in Coarsening

Asymptotic Defect Trajectories

What are the dynamical rules for the interfaces?

For a given domain configuration, e.g.



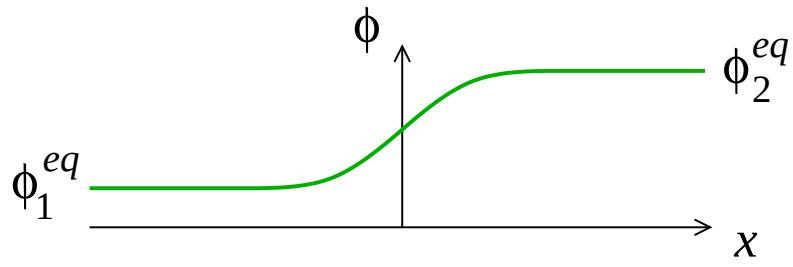
what is the sequence of future domain configurations?

We can take advantage of late-time asymptotia to reduce the original field dynamics to simpler sharp defect dynamics.

Example: conserved scalar OP with isotropic σ

Step 1. Surface Tension

Consider a flat interface at $x = 0$ with b.c. as shown:



Equilibrium concentration profile given by

$$0 = \mu(\mathbf{x}) = \frac{\delta F}{\delta \phi(\mathbf{x})} = V'(\phi) - c\nabla^2\phi + \dots$$

Solution $\phi_{int}(x)$ gives free energy per unit interface:

$$\sigma \equiv F[\phi_{int}(x)]/A$$

For curved interfaces, $\sigma(\kappa) = \sigma + O(\kappa)$

Example: conserved scalar OP with isotropic σ

Step 2. Bulk Mobility

- In bulk $\phi \approx \phi_1^{eq}$, so local chemical potential proportional to the supersaturation:

$$\mu(\mathbf{x}) = V''(\phi_1^{eq})\big(\phi(\mathbf{x}) - \phi_1^{eq}\big) + O(1/L^2)$$

- Asymptotic current: $\mathbf{J} = -M(\phi)\nabla\mu = -M(1)V''(1)\nabla\phi$

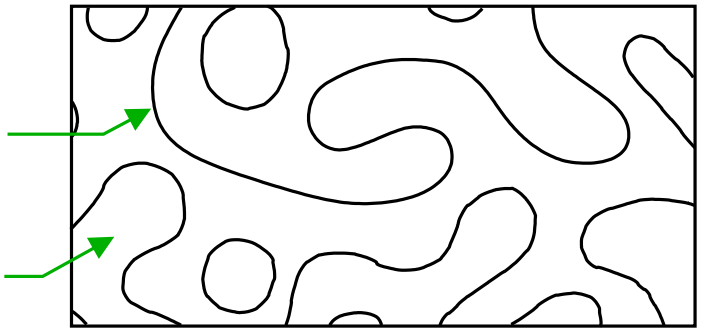
- Gives diffusion equation in bulk: $\partial_t\phi = -\nabla\cdot\mathbf{J} \sim D\nabla^2\phi$. Same equation for $\mu(\mathbf{x})$.

- Diffusion field equilibrates in domains of size L in time $t_{eq} \sim L^2$

Example: conserved scalar OP with isotropic σ

Step 3. Gibbs-Thomson at interfaces:

$$\mu(\mathbf{x}) = \frac{\sigma}{\Delta\phi_{eq}}\kappa(\mathbf{x}) + O(\kappa^2)$$



Step 4. Quasistatic in bulk: $\nabla^2\mu = 0$ since diffusion field equilibrates faster than interfaces move.

Determines $\mu(\mathbf{x})$ everywhere!

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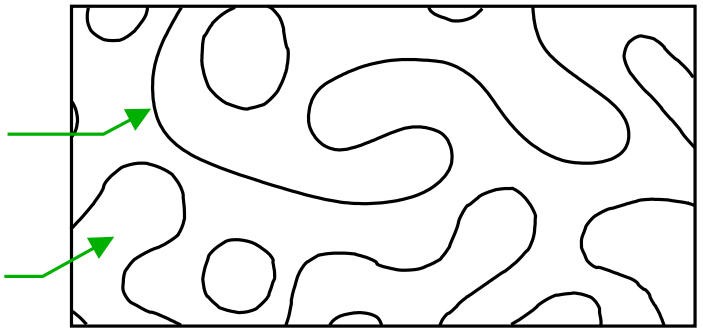

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Step 5. Interface velocity determined by bulk flux to interface:

$$\Delta\phi_{eq} v(\mathbf{x}) = \hat{n}\cdot(\mathbf{J}_+ - \mathbf{J}_-) \Rightarrow v(\mathbf{x}) = \frac{M_1\,\hat{n}\cdot\nabla\mu_1 - M_2\,\hat{n}\cdot\nabla\mu_2}{\Delta\phi_{eq}}$$

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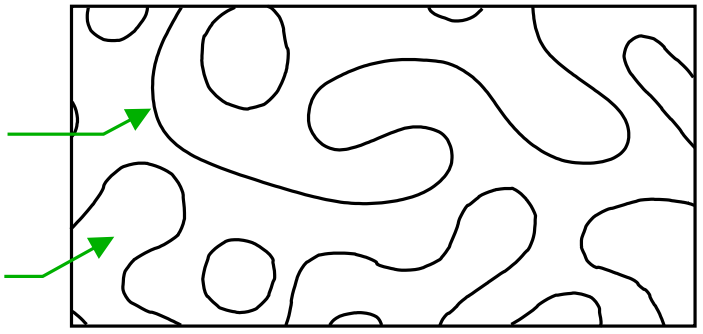

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Huse: $v \sim \dot{L},\ \nabla\mu \sim 1/L^2$

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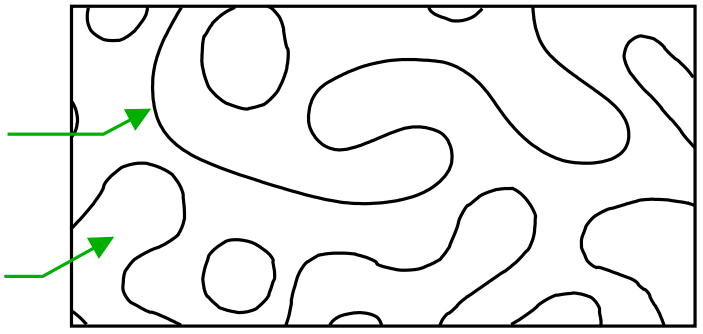

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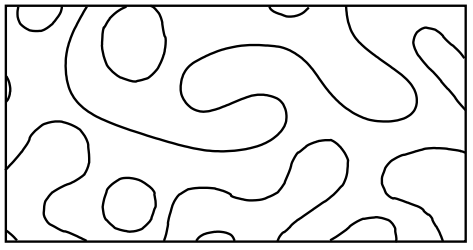
Huse: $v \sim \dot{L},\ \nabla\mu \sim 1/L^2 \Rightarrow \dot{L} \sim 1/L^2 \Rightarrow L \sim t^{1/3}$

Example: conserved scalar OP with isotropic σ

Take case of equal bulk mobilities: $M_1 = M_2 = M$.



- For all such systems $v(\mathbf{x})$ same at each point along the interface, up to an overall factor $M\sigma/(\Delta\phi_{eq})^2$.
- All systems will evolve through the same sequence of configuration: they have the same defect trajectories.
- In rescaled time $\tau = \frac{M\sigma}{(\Delta\phi_{eq})^2}t$, all systems evolve identically!
- If $M_1 \neq M_2$, the above still hold for all systems with the same ratio M_1/M_2.

Universality Classes in Coarsening

Domain Morphology Distribution

Domain structure governed by a domain morphology distribution $P[t; \phi(\mathbf{x}, t)]$ with asymptotic scaling form $P[\phi(\mathbf{x}/L)]$.

- $P[\phi]$ (DMD) contains all equal time properties of coarsening system, e.g.

$$C(\mathbf{x}, t) = \int \mathcal{D}\phi \, P[\phi] \, \phi(0, t)\phi(\mathbf{x}, t)$$

- Ockham (circa 1300) suggests that any correlation function universality comes from DMD universality.
- **Note:** $P[\phi]$ + asymptotic defect trajectories provides full specification of scaling state.

Domain Morphology Universality

Conjecture: DMD has same universality as the defect trajectories.

Wrong if

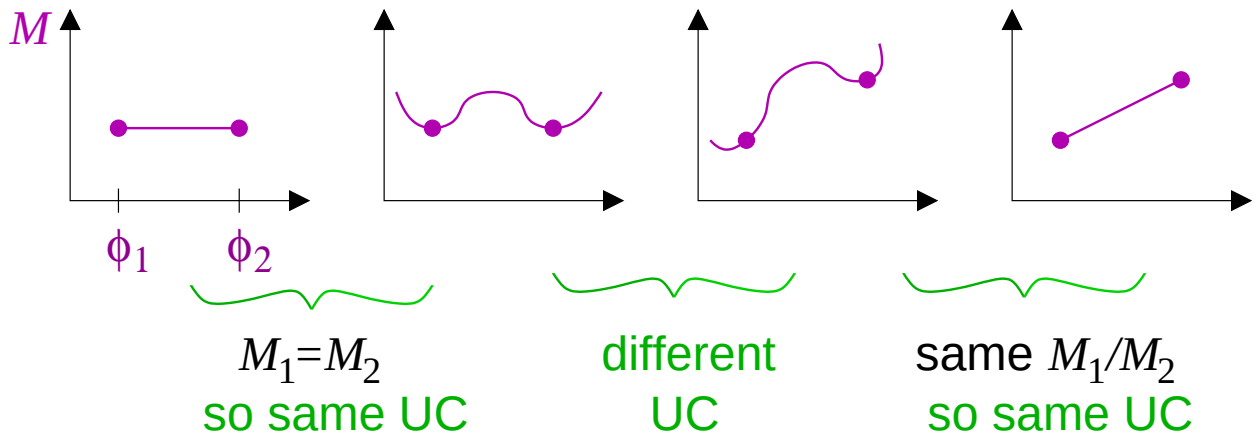
- different trajectories can lead to the same DMD (superuniversal)
- different DMD possible from same trajectories (history dependent)

Consequence: analysis of which factors determine trajectories $\Rightarrow$ prediction of universality classes for the DMD and the entire asymptotic state.

Corollary: in rescaled time growth law $L \sim A\tau^{\alpha}$ is determined by the DMD $\Rightarrow$ the growth law amplitude should have the same universality as the correlation function.

Predicted Universality Classes — conserved scalar OP

- anisotropic $\sigma(\hat{n})$ modifies $\mu(\mathbf{x})$ at interface, so trajectories and DMD depend on $\sigma(\hat{n}, T)$.
- field-dependent mobility $M(\phi)$, specifically the ratio $M(\phi_1^{eq})/M(\phi_2^{eq})$.



- volume fraction ϵ and spatial dimension d.

DMD universality determines correlation function, growth law amplitude, persistence exponents, ...

Some Extra Bits

- Time scaling can be determined experimentally by evaporating isolated drops
- Defect trajectory analysis determines physical length and time scales in CDS simulations [Oono & Puri '88].
- Growth law amplitudes extracted from published Cahn-Hilliard and CDS data agree
- Exact solution in dilute limit [Rutenberg & BVL '99] shows persistence exponent depends on $\sigma(\hat{n}) \Rightarrow$ DMD universality class
- Powerful new method for generating sharp defect dynamics [Watson '06]

Universality Classes in Coarsening

Universality Classes in Coarsening

What's new to learn about late stage coarsening?

- The growth law exponent and the correlation function do not have the same universality (FACT)
- The growth law amplitude and the correlation function do have the same universality, determined by the DMD (OCKHAM + ANALYSIS)
- These universality classes apply to the complete asymptotic scaling state, and might be determined defect trajectories (CONJECTURE)

It is time to look!

Future Work

- It is time to look! Simulations in progress with Andrew Rutenberg, Jaime Wallace, Phil Marquis, James Miante, and David Enrico
- Generalize defect trajectory analysis (vector order parameter, liquid crystals, hydrodynamics, facets, froths, . . .). With Steven Watson.
- Autocorrelation exponent? Is it superuniversal like the growth exponent?