Domain Structure in Coarsening Dynamics

Ben Vollmayr-Lee
Bucknell University

November 20, 2012
Institute for Theoretical Physics, University of Göttingen

Domain Structure in Coarsening Dynamics

- Andrew Rutenberg, Dalhousie University
- Melinda Gildner, Bucknell $\rightarrow$ UPenn
- Will Rosenbaum, Reed $\rightarrow$ Indiana
- Fawntia Fowler, Reed $\rightarrow$ Stanford
- Sohei Yasuda, Bucknell $\rightarrow$ Purdue
- Kate VanNess, Bucknell

NSF REU grant PHY-0552790

Domain Structure in Coarsening Dynamics

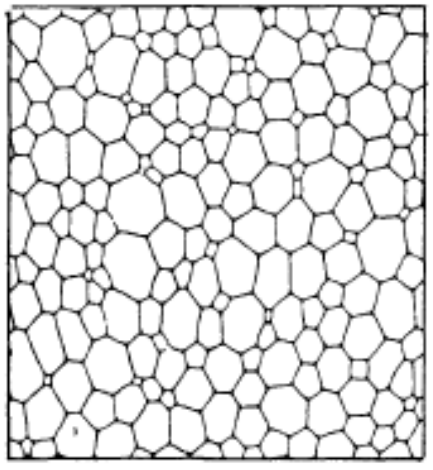
Coarsening . . .

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

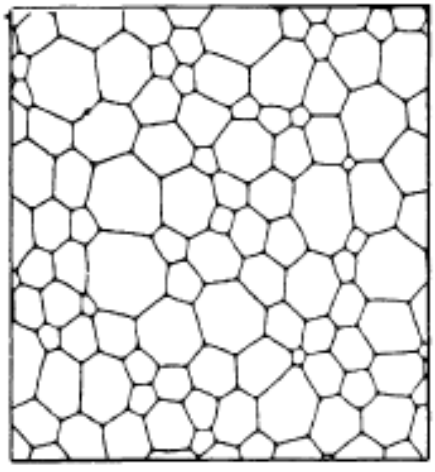
Many examples in nature:

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

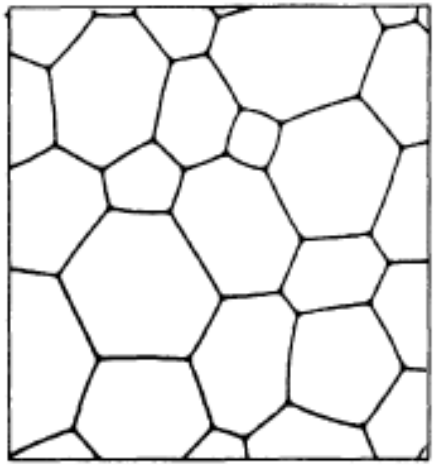
2D Dry Soap Froth



$t = 1.95$ h



$t = 21.5$ h

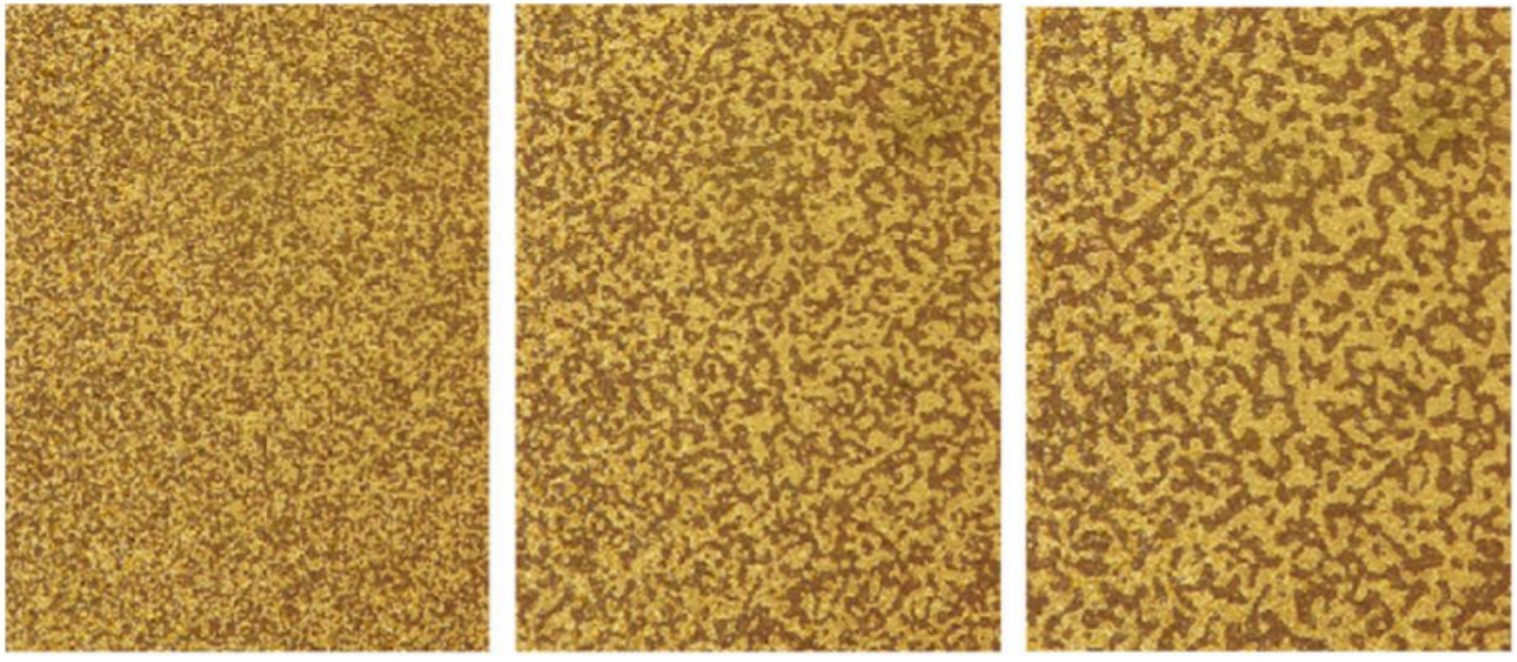


$t = 166$ h

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

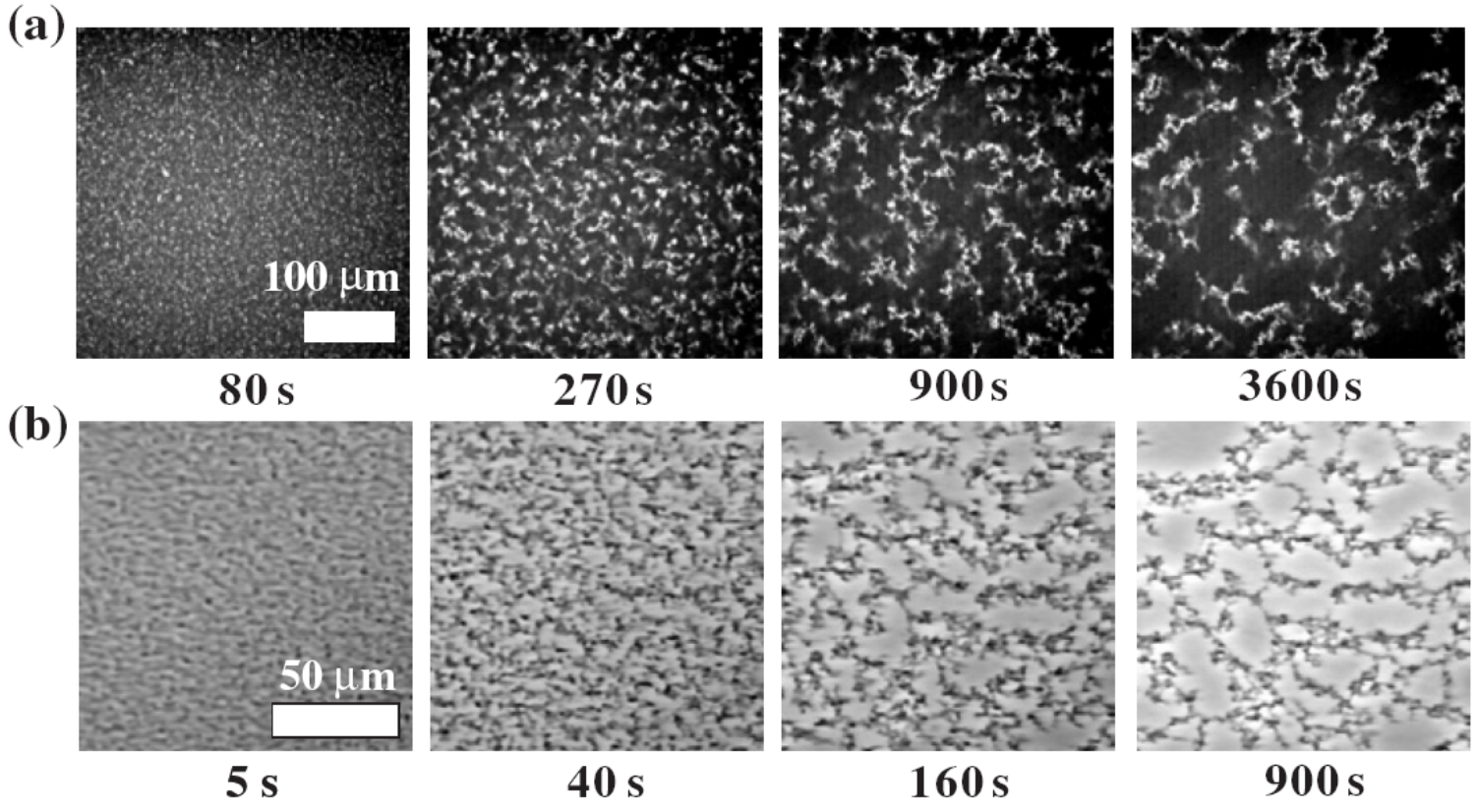
Self-similarity!

Chiral Liquid Crystals



Sicilia, *et al.*, *Phys. Rev. Lett.* **101**, 197801 (2008).

(a) Colloidal Suspension and (b) Polymer Solution

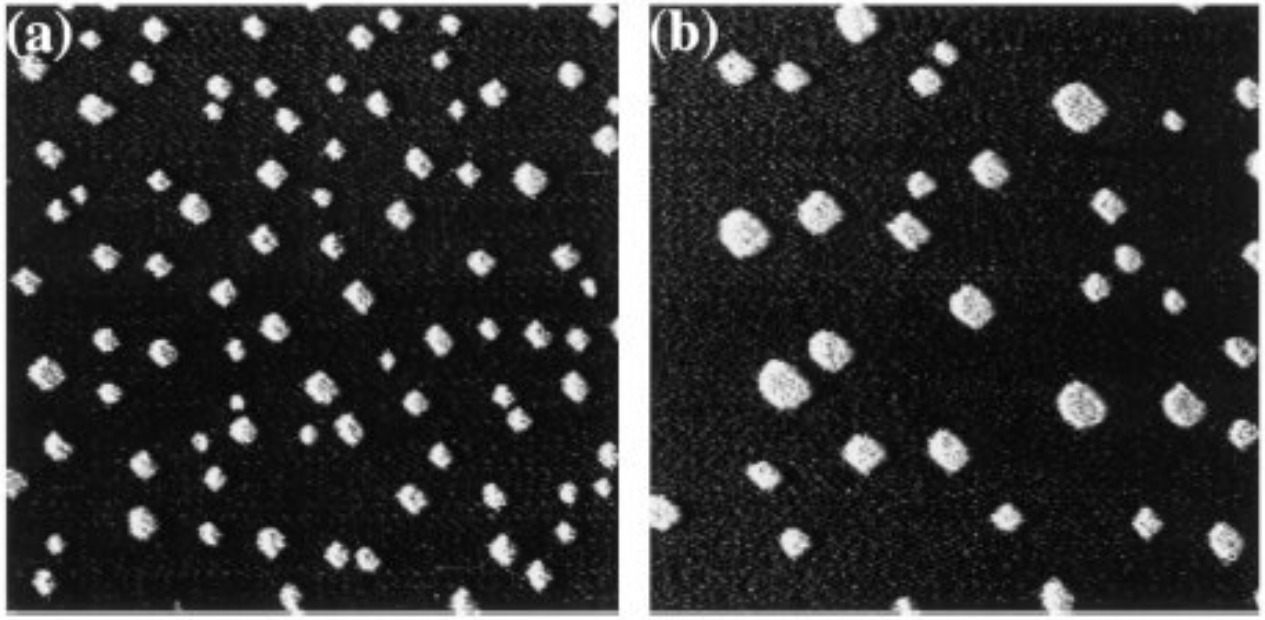


Tanaka, Nishikawa, and Koyama, *J. Phys. Cond. Matt.* **17**, L143 (2005).

Universality!

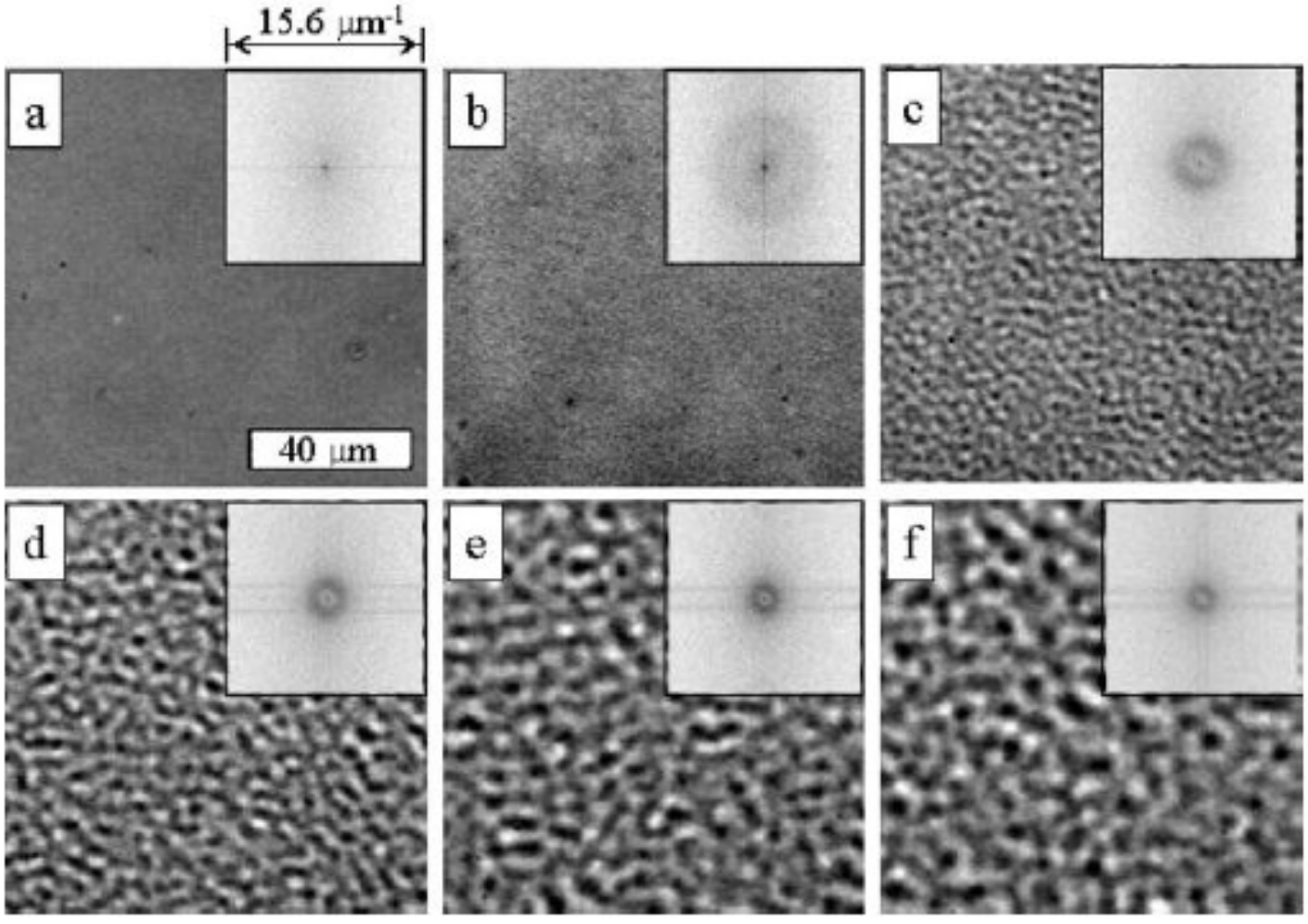
Homoepitaxial Islands

Cu on a Cu(100) surface



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

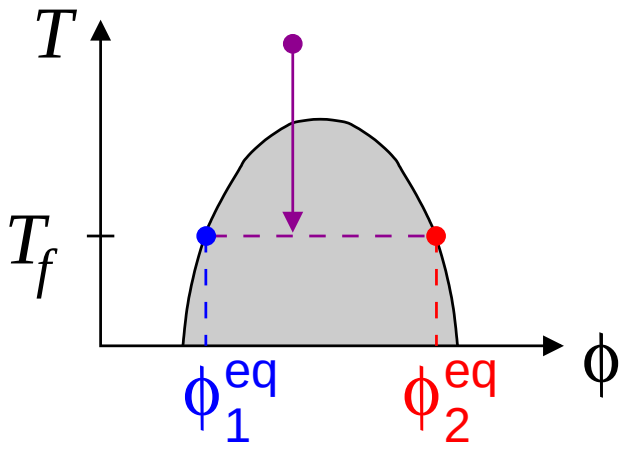
Random Copolymers – PEH/PEB

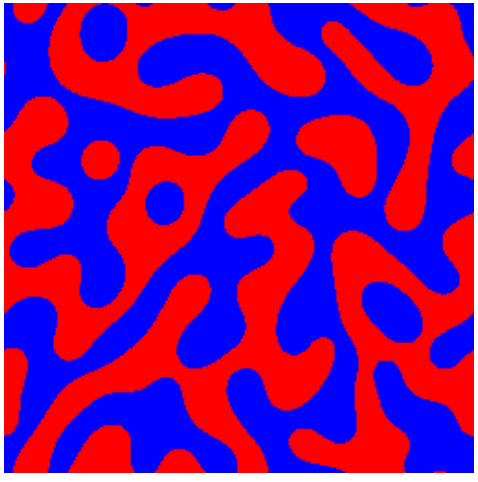


Shimizu *et al.*, *Polymer* **45**, 7061 (2004).

Phase Ordering Dynamics (binary alloys, polymer blends)

- Rapid quench into the forbidden region of a phase diagram
- system equilibrates locally into either ϕ_1^{eq} or ϕ_2^{eq}





$\leftrightarrow$
$L(t)$

- $F - F_{\text{eq}} \propto$ amount of interface
- dissipative dynamics ($dF/dt \leq 0$) gives coarsening

Basic Features of Coarsening

Sharp defects

defect size ξ fixed, asymptotically $L(t) \gg \xi$

Self-similarity

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

$\Rightarrow \quad C(\mathbf{r}, t) = f\big(\mathbf{r}/L(t)\big)$

Power law growth

characteristic scale $L \sim t^{\alpha}$

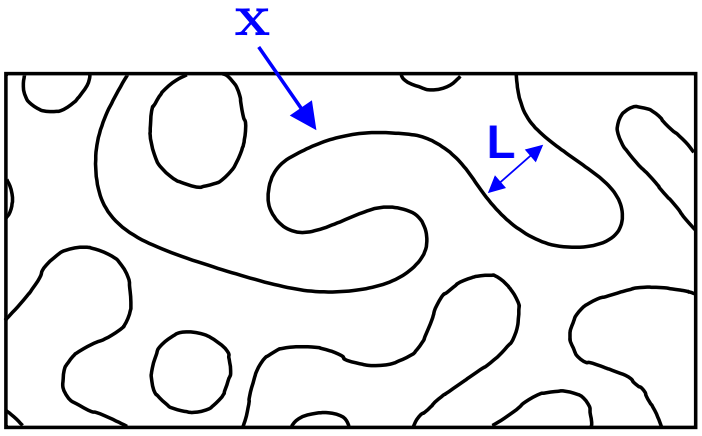
Universality

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

Growth Exponent via Dynamical Scaling Hypothesis

Conserved order parameter:

- $\mu(\mathbf{x}) = \dfrac{\sigma}{\Delta\phi}\kappa(\mathbf{x}) \sim 1/L$
- $\mathbf{J} \sim \nabla\mu \sim 1/L^2$
- $v \propto [J] \sim 1/L^2$

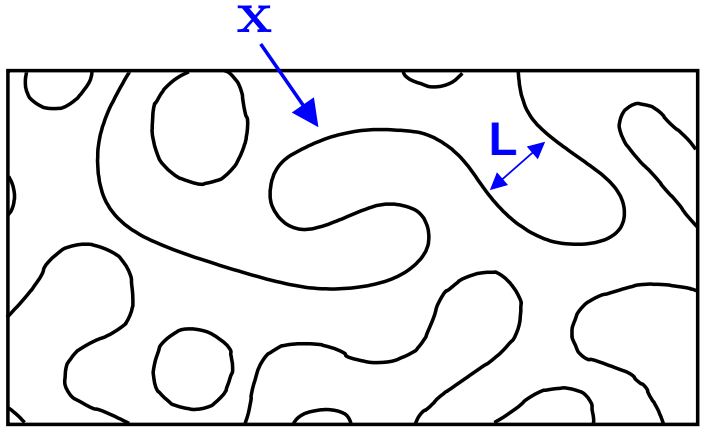


But $v \sim \dot{L}$, so $\dot{L} \sim 1/L^2 \quad \Rightarrow \quad L \sim t^{1/3}$ [Huse '86]

Growth Exponent via Dynamical Scaling Hypothesis

Conserved order parameter:

- $\mu(\mathbf{x}) = \dfrac{\sigma}{\Delta\phi}\kappa(\mathbf{x}) \sim 1/L$
- $\mathbf{J} \sim \nabla\mu \sim 1/L^2$
- $v \propto [J] \sim 1/L^2$



But $v \sim \dot{L}$, so $\dot{L} \sim 1/L^2 \quad \Rightarrow \quad L \sim t^{1/3}$ [Huse '86]

Bray-Rutenberg Energy Scaling ['94]

Generalized to surface, line, or point defects with and without conservation laws.

Dynamical Scaling $\Rightarrow$ Growth Exponent Universality Classes

But We Can't Prove Dynamical Scaling

- Scaling can be derived in a few special cases: LS theory and ABCS theory (and some exact solutions in 1D).
- But so far no RG calculation for coarsening has been found!

Nevertheless,

Dynamical Scaling $\Rightarrow$ Growth Exponent Universality Classes

Domain Structure in Coarsening Dynamics

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 to $T < T_c$ ...

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 to $T < T_c$ ...

Glauber Dynamics

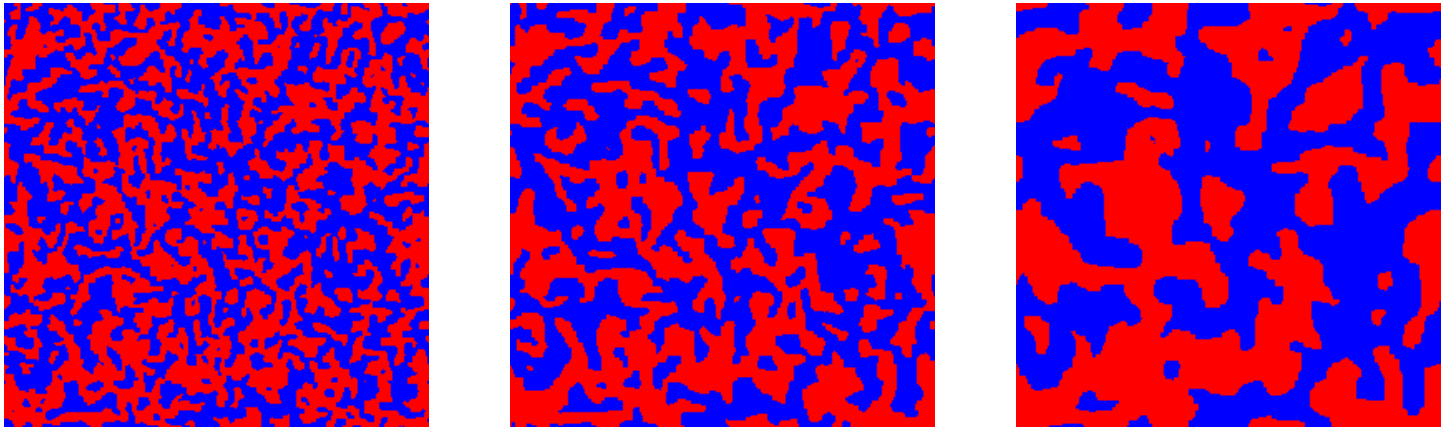
- spins flip with probability determined by energy
  $\Rightarrow$ nonconserved OP.

Kawasaki Dynamics

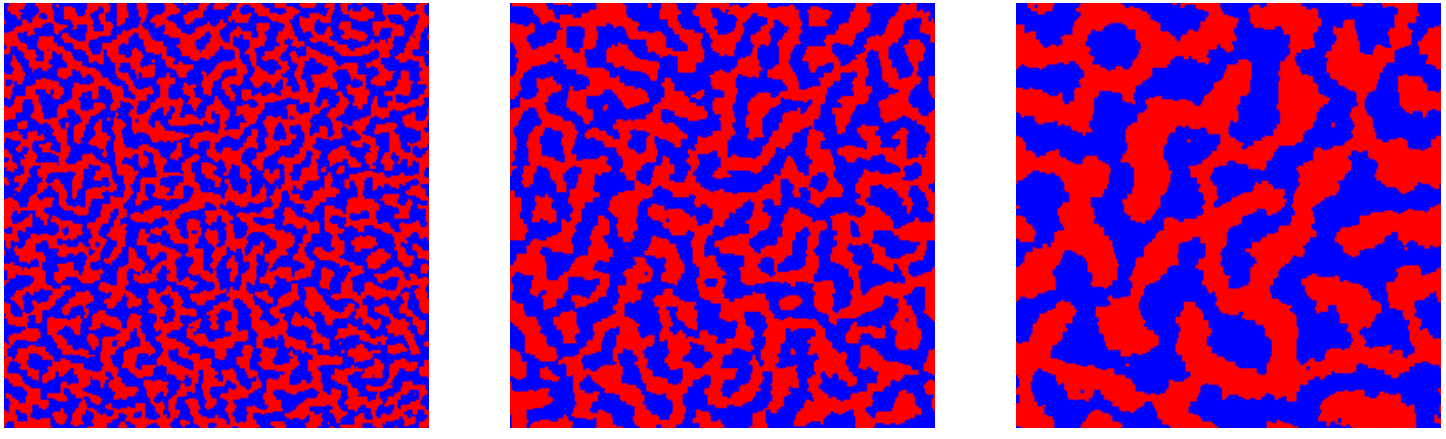
- neighboring spins exchanged $\Rightarrow$ conserved OP.
- additional parameter $\epsilon =$ fraction of spins up
- appropriate for binary mixtures: $\uparrow =$ Cu, $\downarrow =$ Ni.

Kinetic Ising Models

Glauber: spin flip = nonconserved OP $\Rightarrow L \sim t^{1/2}$



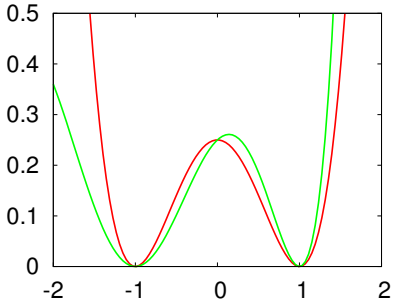
Kawasaki: spin exchange = conserved OP $\Rightarrow L \sim t^{1/3}$



Coarsening Models II: Phase Field Models

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

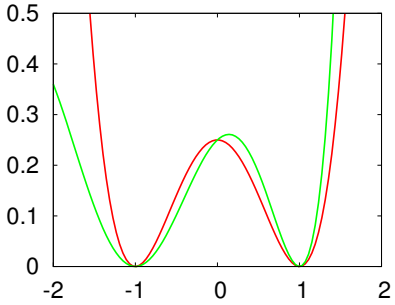
$$F[\phi] = \int d^d x \left\{ \tfrac{1}{2}(\nabla\phi)^2 + V(\phi) \right\}$$



Coarsening Models II: Phase Field Models

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

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



Allen-Cahn equation

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

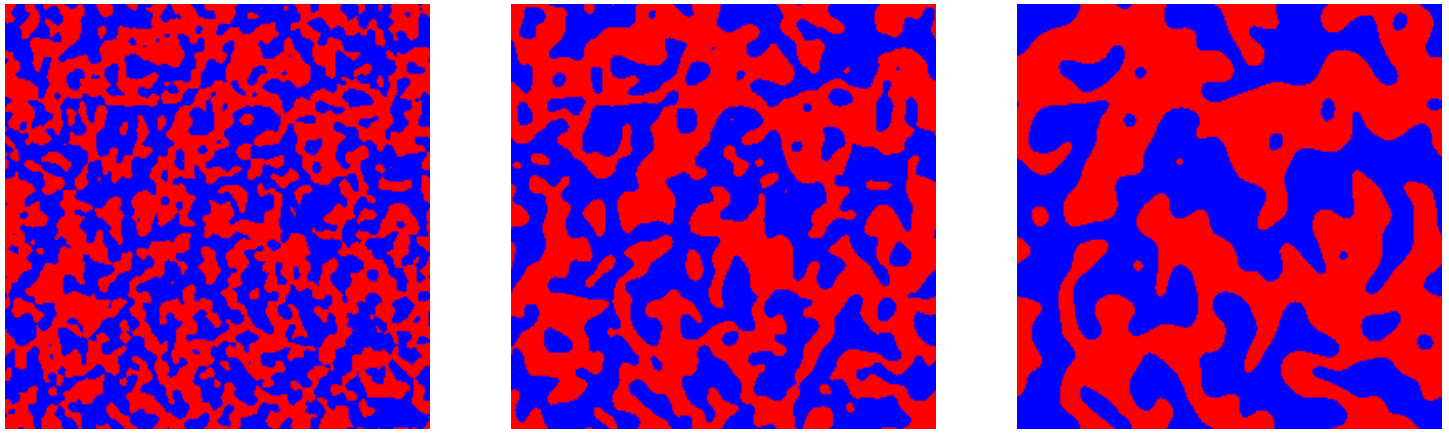
Cahn-Hilliard equation

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

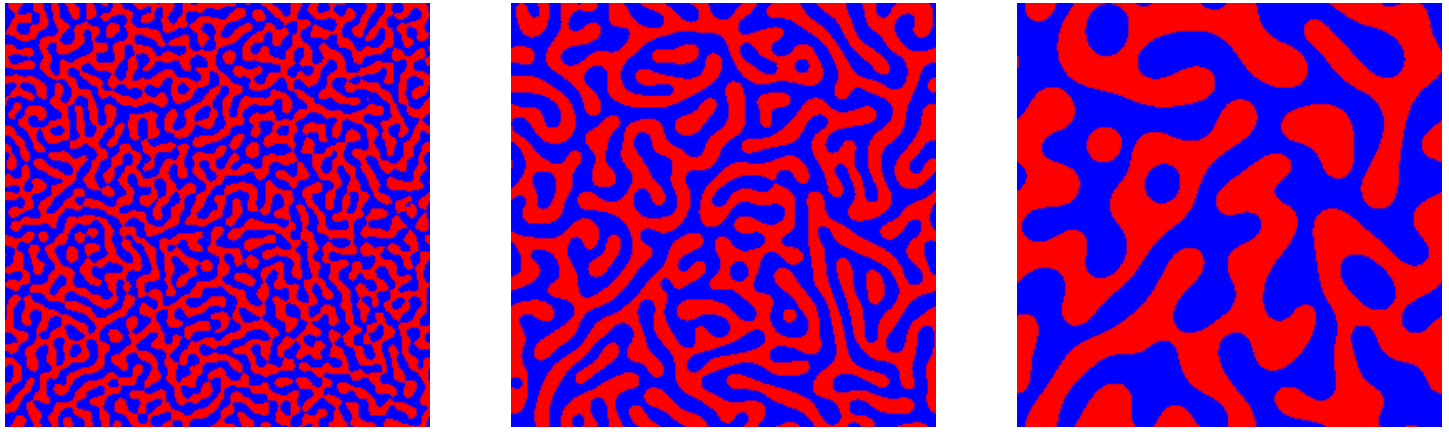
$$\Rightarrow \frac{\partial\phi}{\partial t} = -\nabla^2[\nabla^2\phi - V'(\phi)]$$

Phase Field Models

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



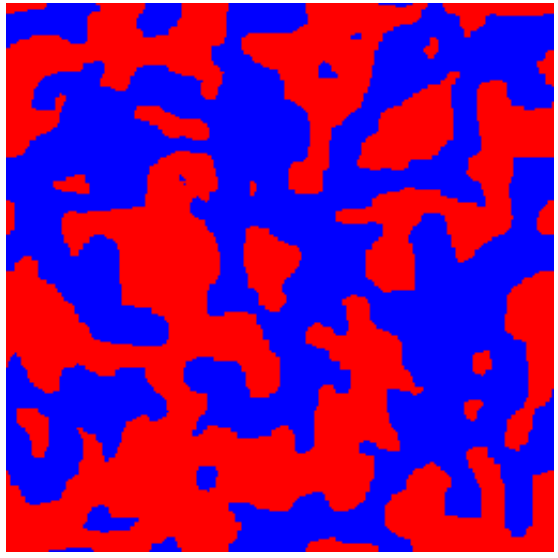
Cahn-Hilliard: conserved OP $\Rightarrow L \sim t^{1/3}$

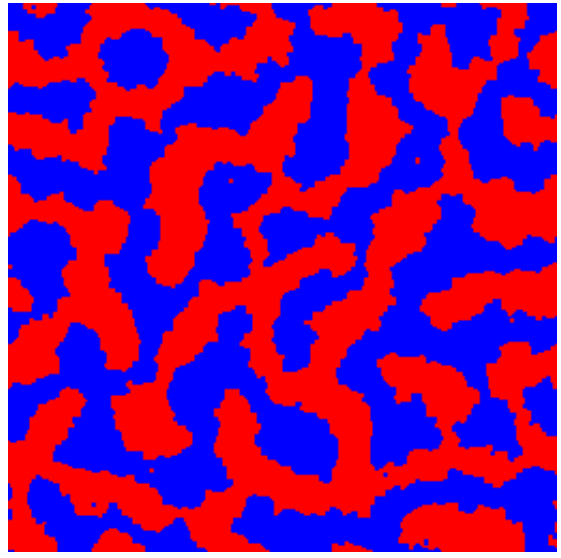


Nonconserved: $L \sim t^{1/2}$

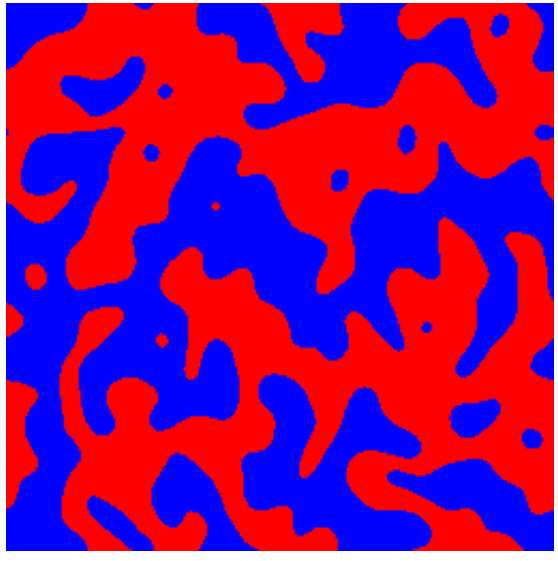
Conserved: $L \sim t^{1/3}$

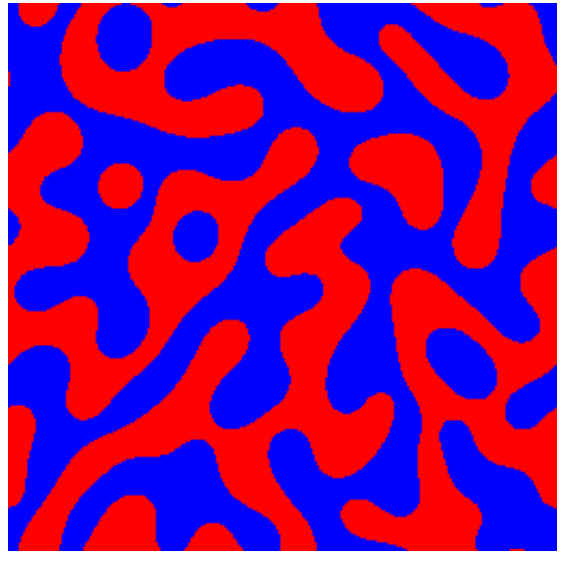
Ising





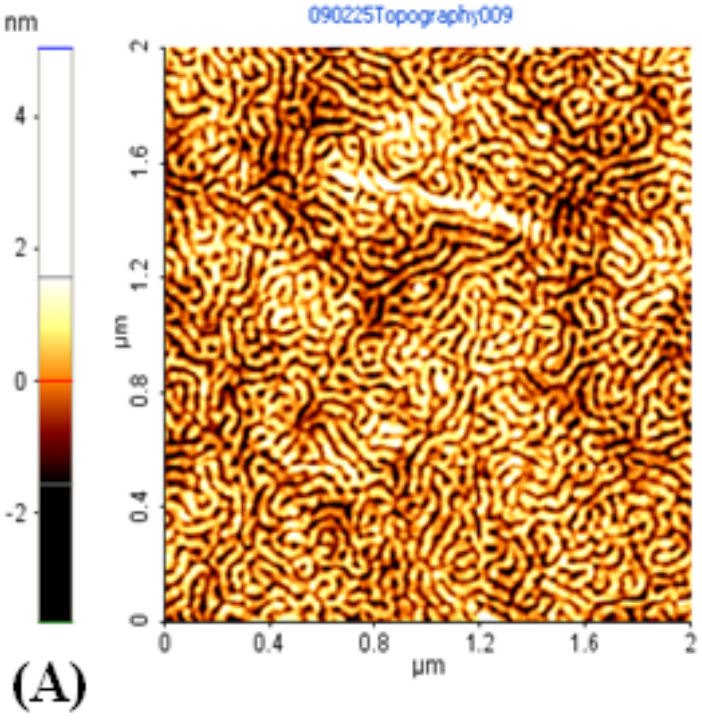
Phase Field



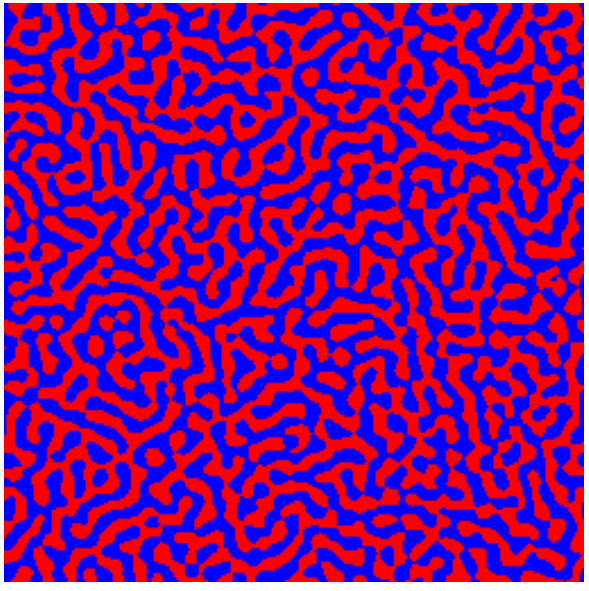


Compare to Experiment

Polymer Blend AFM Image



Cahn-Hilliard Simulation



O'Mahony, *et al.* in *Thermodynamics — Systems in Equilibrium and Non-Equilibrium*, Moreno-Piraján, Ed. (2011).

Is the Domain Structure Universal?

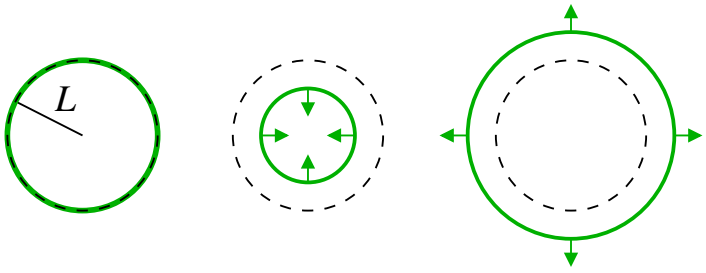
- It is for equilibrium criticality (percolation, Ising model, etc.)
- Determined by RG calculations of correlations, cluster size distributions, etc, but we don't have an RG fixed point in hand.
- The domain structure is universal for some special cases: LS theory and ABCS theory ...

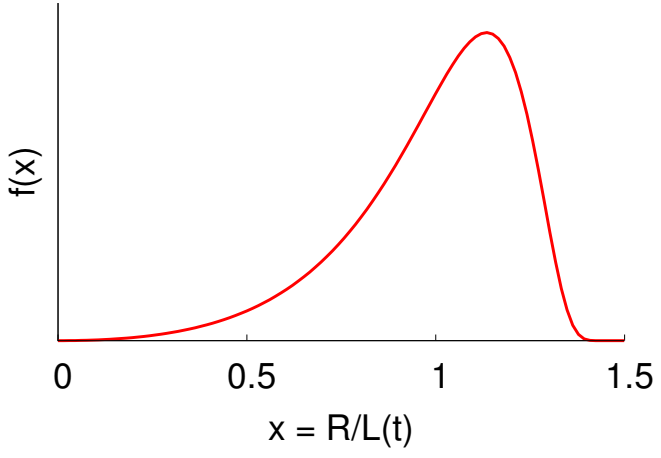
Lifshitz-Slyozov ['58]: applies in dilute limit $\epsilon \to 0$

- Isolated drops of A in supersaturated matrix of B
- Large drops grow, small drops shrink
- Original derivation of $L \sim t^{1/3}$
- Produces scaling drop size distribution

$$n(R,t) = \frac{1}{L(t)^4} f\left(\frac{R}{L(t)}\right)$$

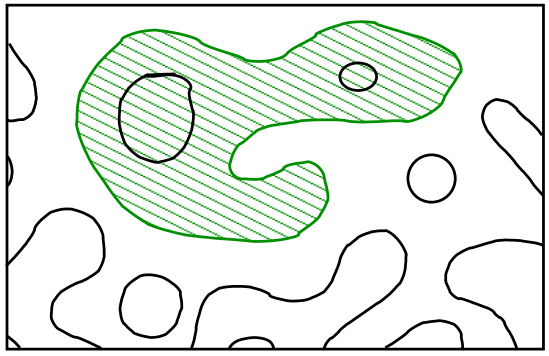
- Universal!





Arenzon, Bray, Cugliandolo, & Sicilia, PRL (2007)

For nonconserved OP in $d = 2$:



- curvature driven interfaces: $v = \frac{\lambda}{2\pi}\kappa$
- Hull areas decay as

$$\frac{dA_h}{dt} = -\oint_{\mathcal{P}} v\, dl = -\frac{\lambda}{2\pi}\oint_{\mathcal{P}} \kappa\, dl = -\lambda$$

- Initial distribution given by percolation [Cardy-Ziff '03]

Result:

$$n(A_h, t) = \frac{1}{4\pi\sqrt{3}}(A_h + \lambda t)^{-2} = \frac{1}{t^2} f(A_h/t)$$

Scales as $L \sim \sqrt{A_h} \sim t^{1/2}$ and universal

Is the Domain Structure Universal?

- It is for equilibrium criticality (percolation, Ising model, etc.)
- Determined by RG calculations of correlations, cluster size distributions, etc, but we don't have an RG fixed point in hand.
- The domain structure is universal for some special cases: LS theory and ABCS theory . . .
- . . . so it is often assumed that the structure is universal, with the same universality classes as the growth exponent.

Is the Domain Structure Universal?

- It is for equilibrium criticality (percolation, Ising model, etc.)
- Determined by RG calculations of correlations, cluster size distributions, etc, but we don't have an RG fixed point in hand.
- The domain structure is universal for some special cases: LS theory and ABCS theory . . .
- . . . so it is often assumed that the structure is universal, with the same universality classes as the growth exponent.

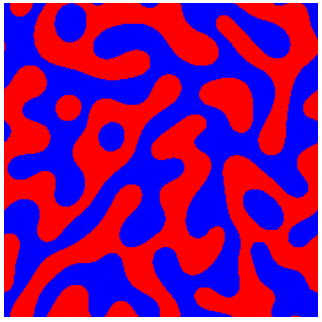
Not True!

Distinct Universality (for conserved scalar OP)

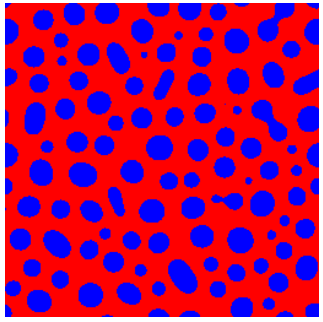
Quantities that affect the domain structure but not the growth exponent:

Trivial

- volume fraction ϵ
- spatial dimension d



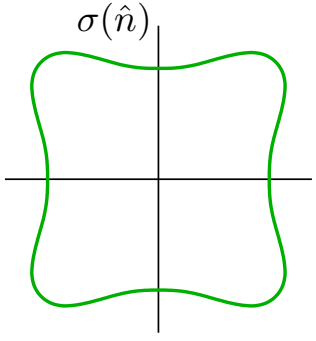
$\epsilon = 1/2$



$\epsilon < 1/2$

Less Trivial

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



exact Lifshitz-Slyozov solution for dilute coarsening

[BVL & Rutenberg PRL '99; Gildner, Rosenbaum, Fowler, and BVL, *in prep.*]

So What Does Determine the Structure?

Proposal: Memory Erasure Hypothesis

- Asymptotically, the system loses memory of initial short-range correlations in the structure.
- Eventually, the structure will be determined solely by the asymptotic dynamics of the defects.

Combined:

Dynamic Scaling Hypothesis $\Rightarrow$ Growth Exponent

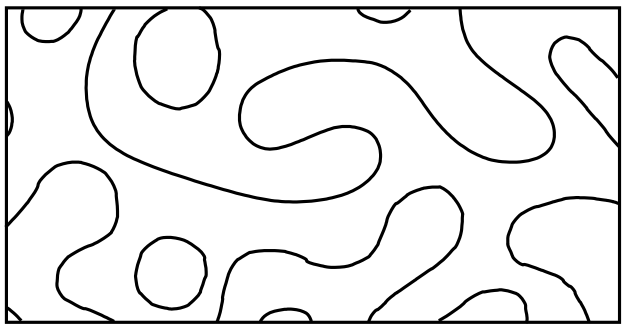
Memory Erasure Hypothesis $\Rightarrow$ Domain Structure

Domain Structure in Coarsening Dynamics

Asymptotic Defect Dynamics

What are the dynamical rules for the interfaces?

For a given domain configuration, e.g.



how will it evolve? What is the sequence of future domain configurations?

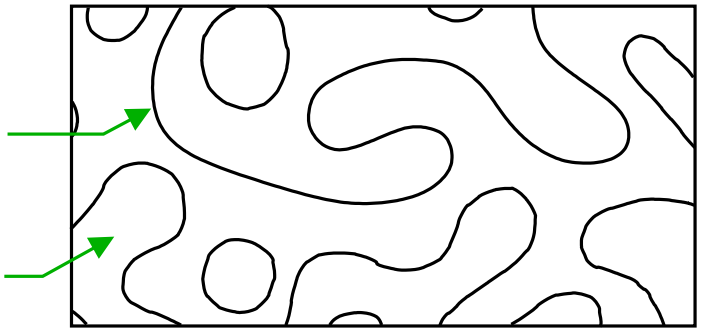
Use late-time asymptotia to reduce to simpler sharp defect dynamics.

Example: conserved scalar OP with isotropic σ

Gibbs-Thomson at interfaces:

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

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

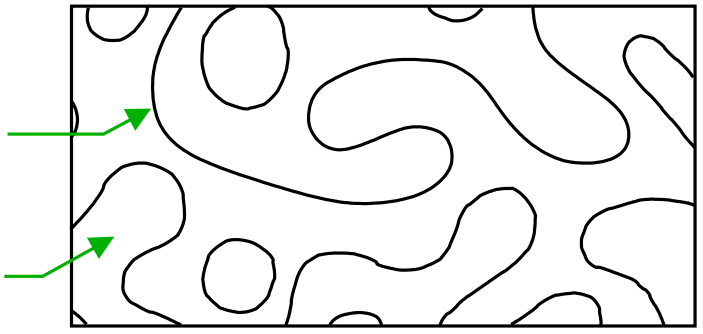


Determines $\mu(\mathbf{x})$ everywhere!

Example: conserved scalar OP with isotropic σ

Gibbs-Thomson at interfaces:

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



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

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

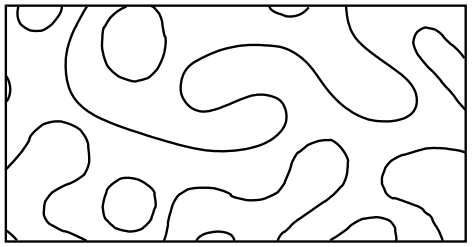
Current driven by chemical potential gradient: $\mathbf{J} \sim -M(\phi)\nabla\mu$

Interface velocity determined by bulk flux to interface:

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

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 configurations: they have the same defect dynamics.
- In universal 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 sharing the same ratio $R_M = M_1/M_2$.

Predicted Structure Universality Classes — conserved OP

- anisotropic surface tension modifies $\mu(\mathbf{x})$ at interface, so structure depends on $\sigma(\hat{n}, T)$.
- Mobility ratio $R_M = M(\phi_1^{eq})/M(\phi_2^{eq})$.
- volume fraction ϵ and spatial dimension d.
- and nothing more!

Highly constrained growth exponent is *superuniversal*.

Most quantities follow domain structure universality classes: correlation functions, growth law amplitude, persistence exponents, etc.

Domain Structure in Coarsening Dynamics

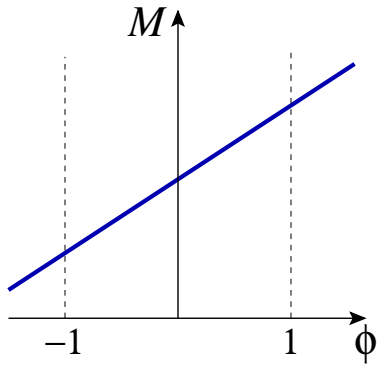
Coarsening with Asymmetric Mobility

Cahn-Hilliard with Field-Dependent Mobility:

$$\frac{\partial \phi}{\partial t} = \nabla \cdot \left\{ M(\phi) \nabla \left(V'(\phi) - \nabla^2 \phi \right) \right\}$$

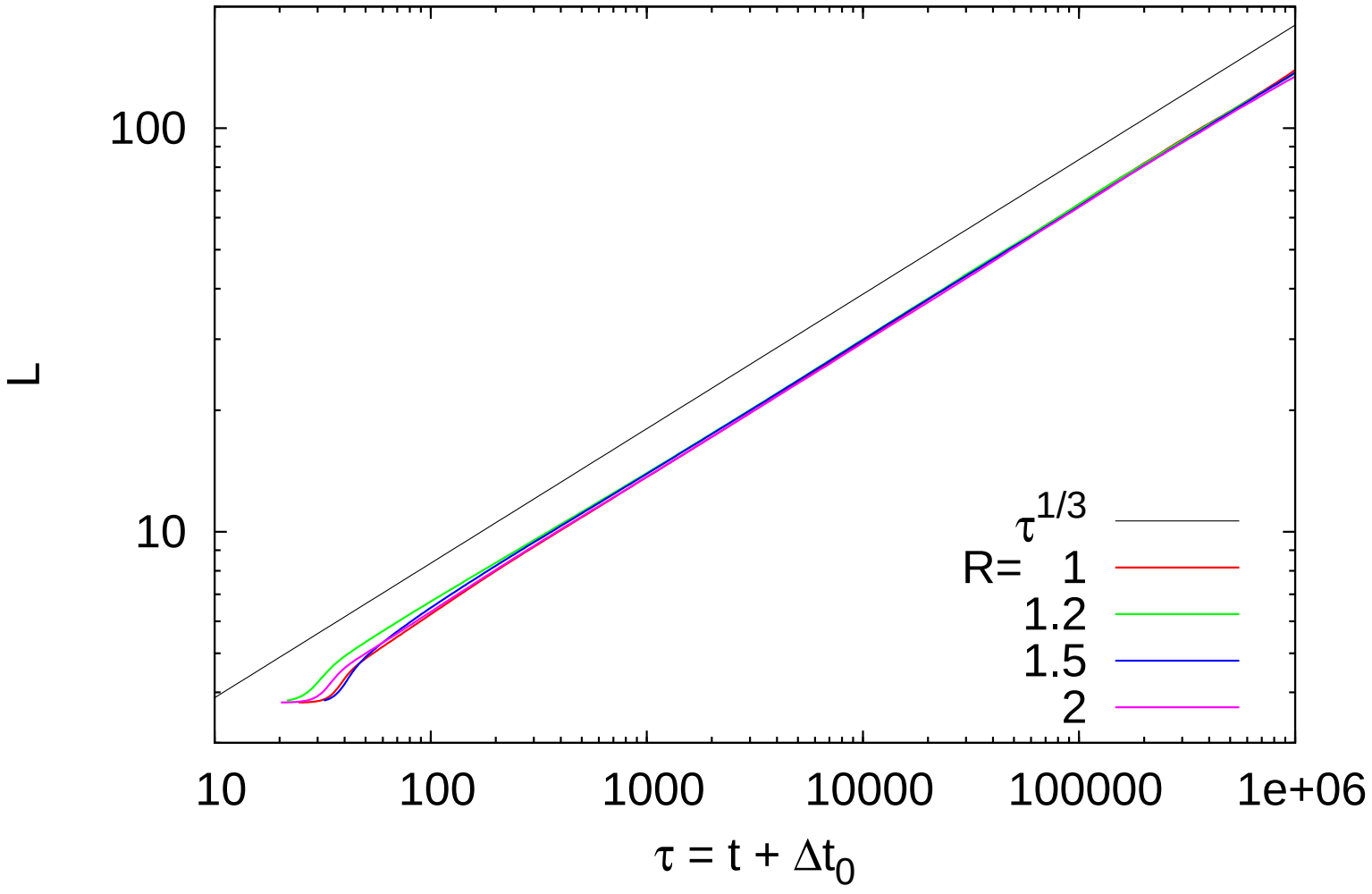
Mobility $M = 1 + m\phi$, with $0 \leq m \leq 1$

$$R_M \equiv \frac{M(1)}{M(-1)} = \frac{1+m}{1-m}$$



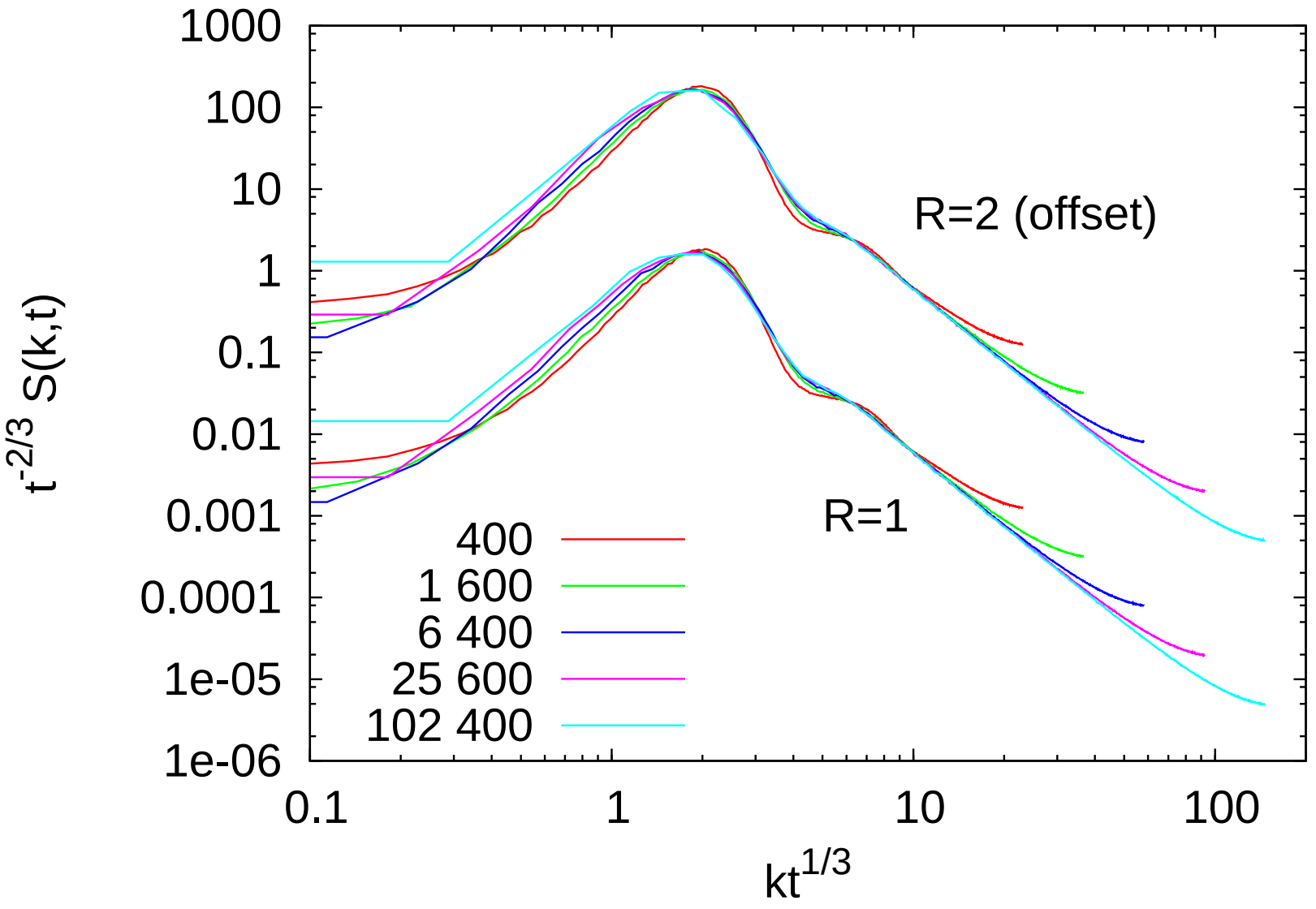
Simulation 2-D lattice, from 256^2 up to 4096^2, using stable semi-implicit methods that allows $\Delta t \sim t^{2/3}$ as the interfaces slow down, $v \sim t^{-2/3}$ [Eyre '98, BVL & Rutenberg, '03]

Growth Law [Yasuda, BVL, & Rutenberg, *in prep.*]

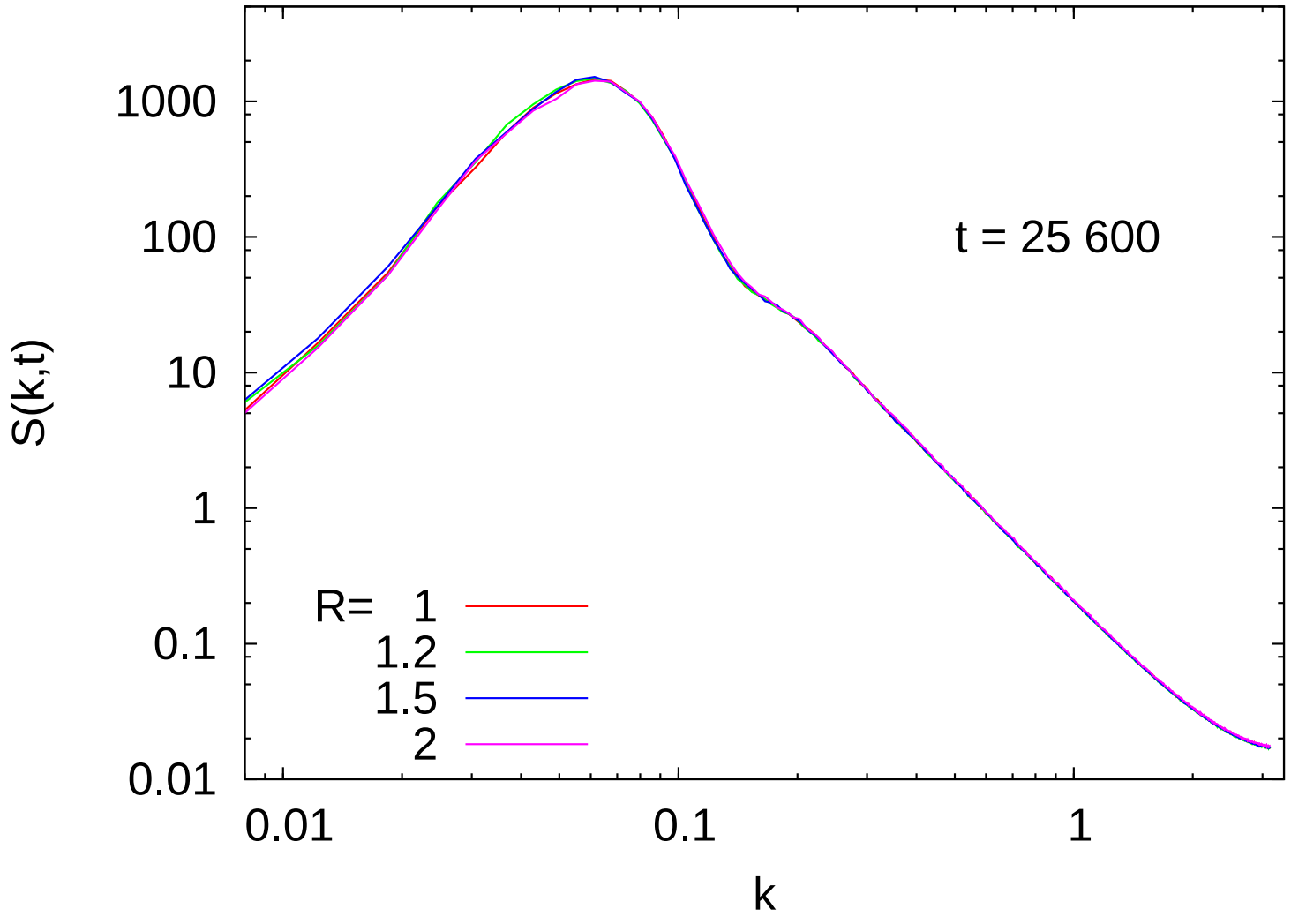


Best fit exponent 0.3336_2. No discernible R_M dependence.

Structure Factor Scaling

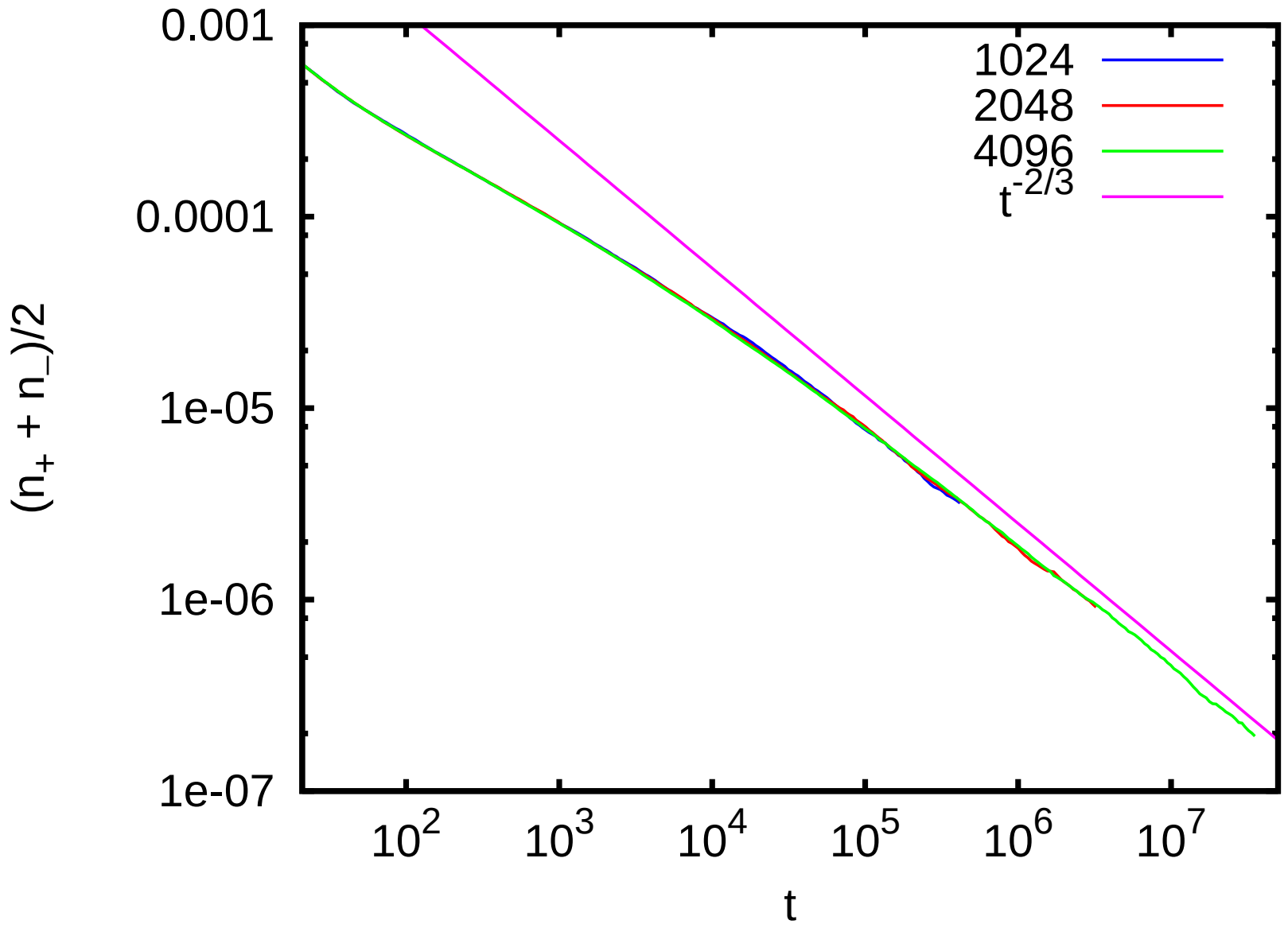


Structure Factor for various R



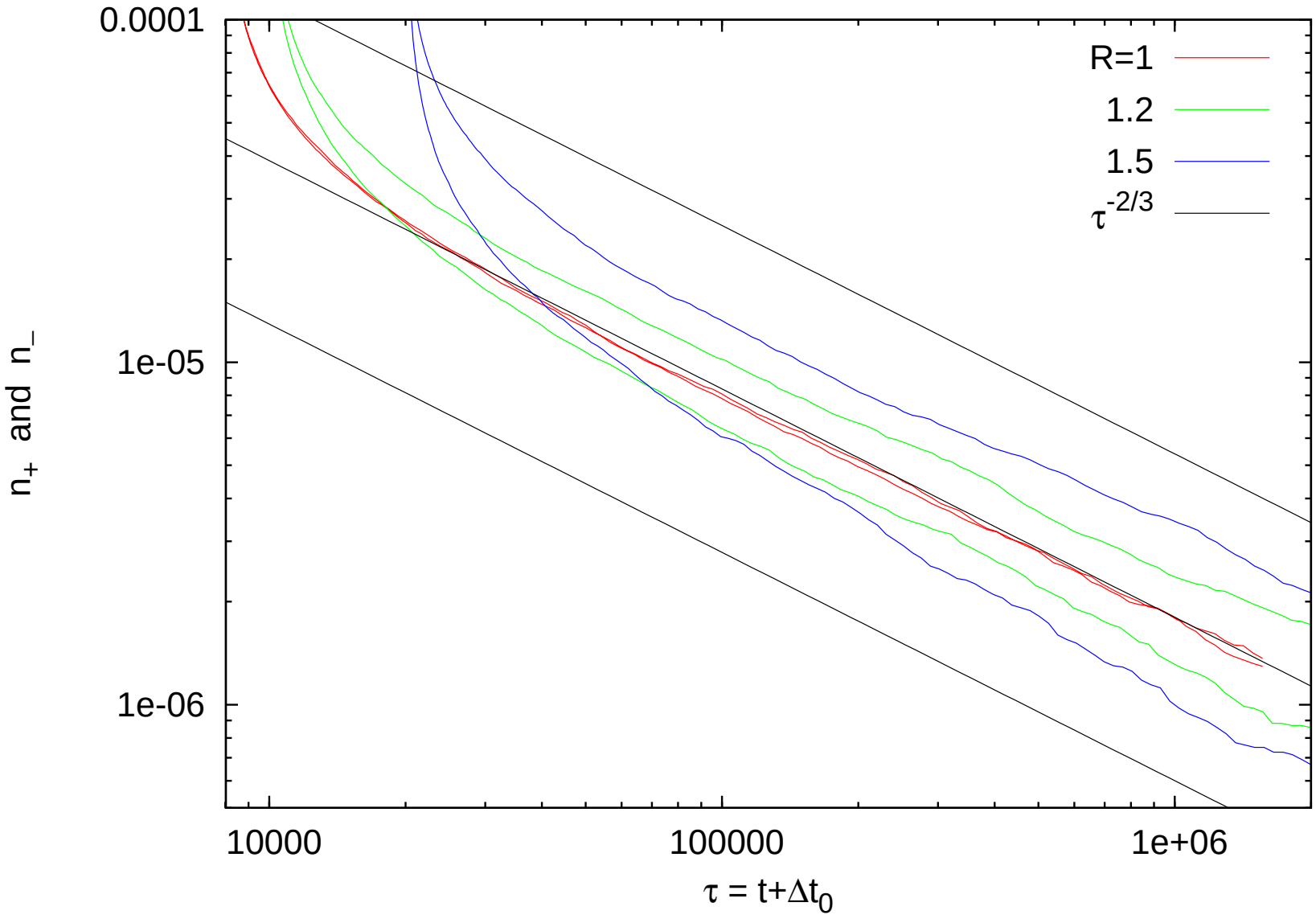
No discernible dependence on R_M! What's going on?

Cluster Density, $R = 1$



Discard percolating clusters. $N_{min} = 3$

Cluster Density for Various R

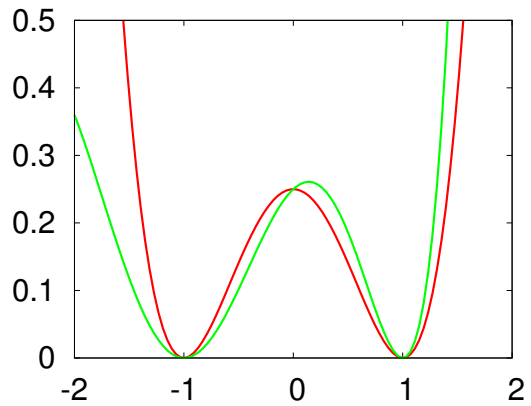


Different Types of Asymmetry

So asymmetric mobility creates an asymmetric domain number.
Maybe this would happen for any asymmetry in the CH equation?

Consider an asymmetric potential:

$$V = \tfrac{1}{4}(1-\phi^2)^2(1+c\phi)^2$$

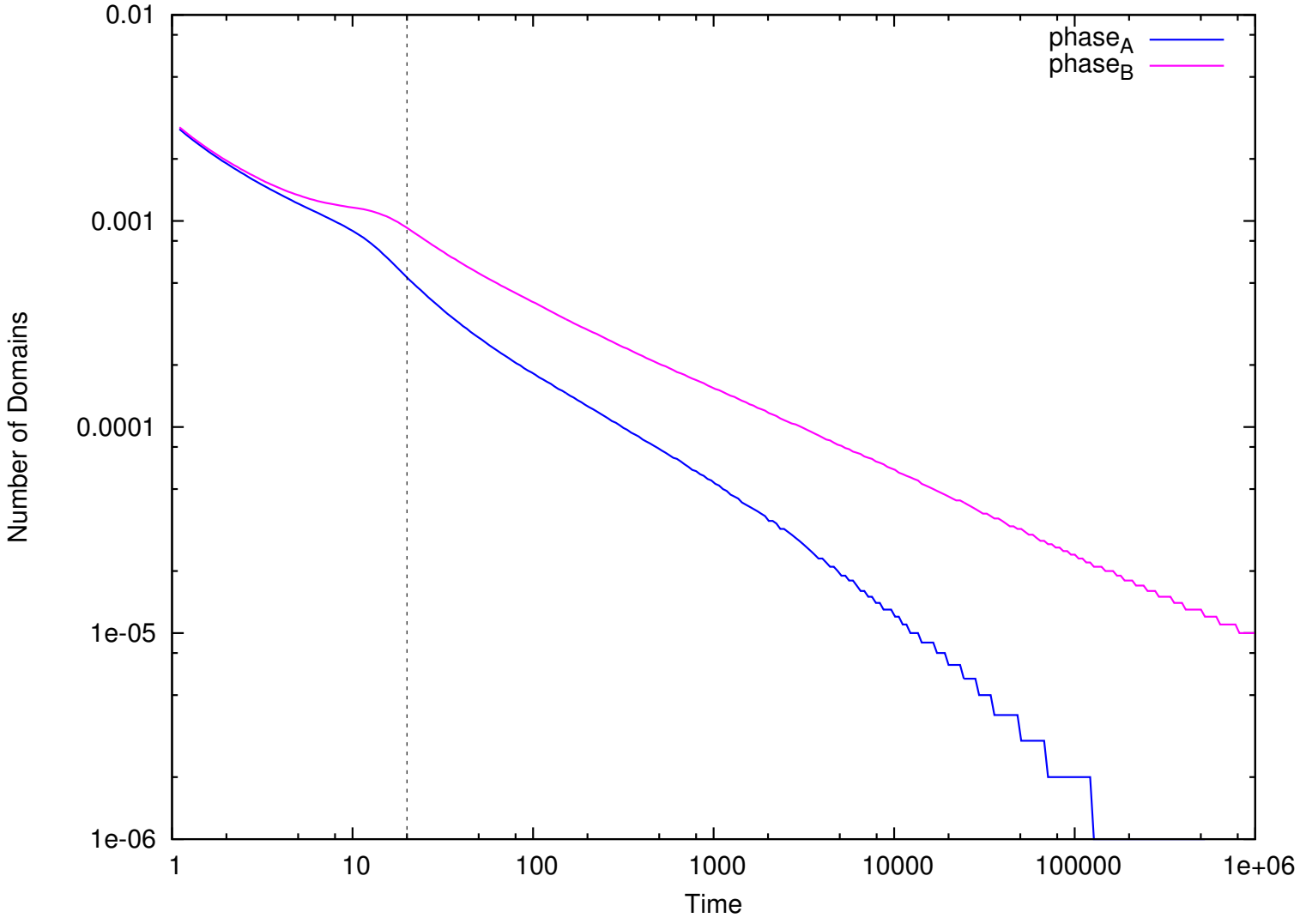


Evolve via CH eq:

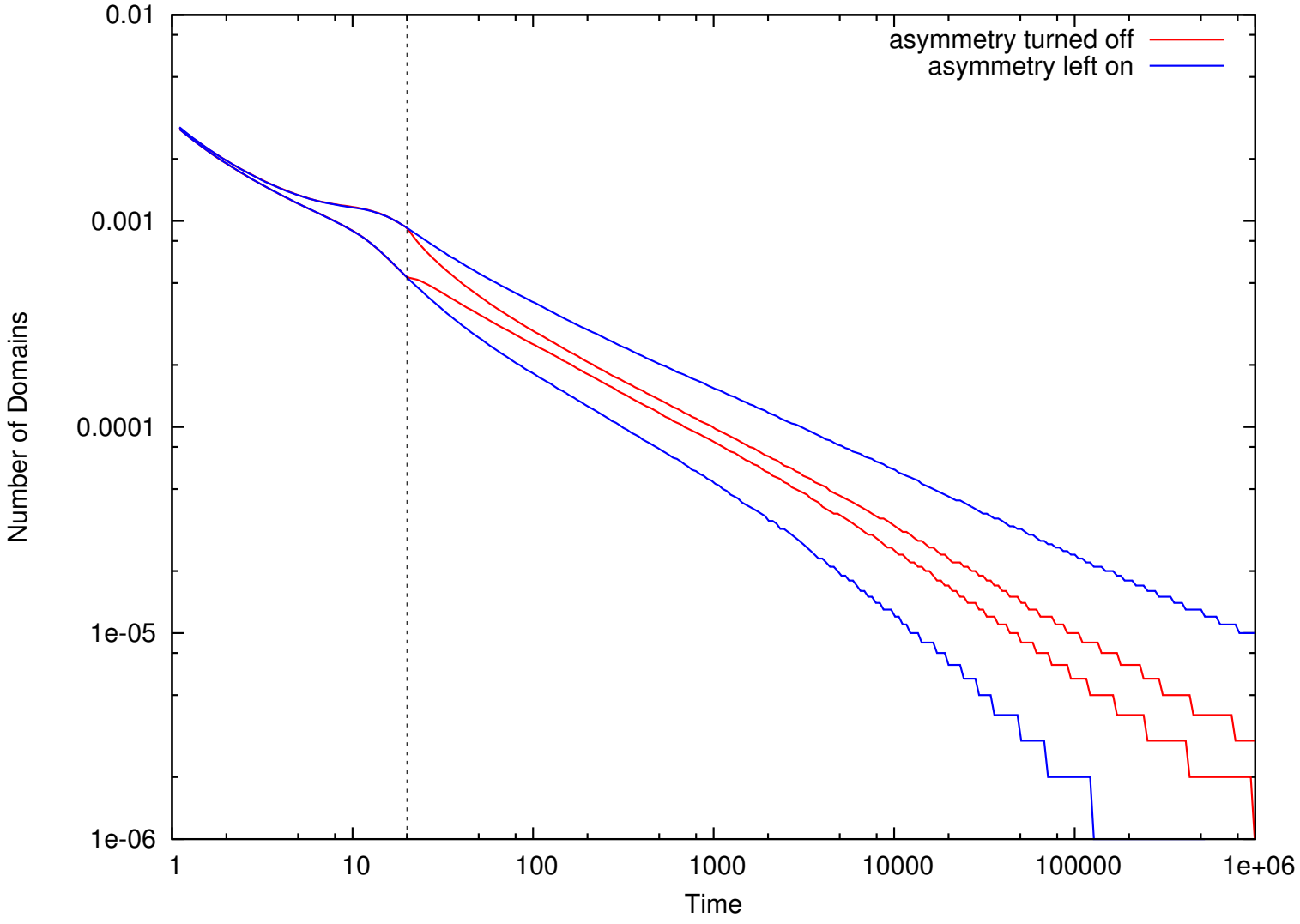
$$\dot{\phi} = M\nabla^2\Big(-\nabla^2\phi + V_a'(\phi)\Big)$$

Define asymmetry parameter $R_V = V''(1)/V''(-1)$:

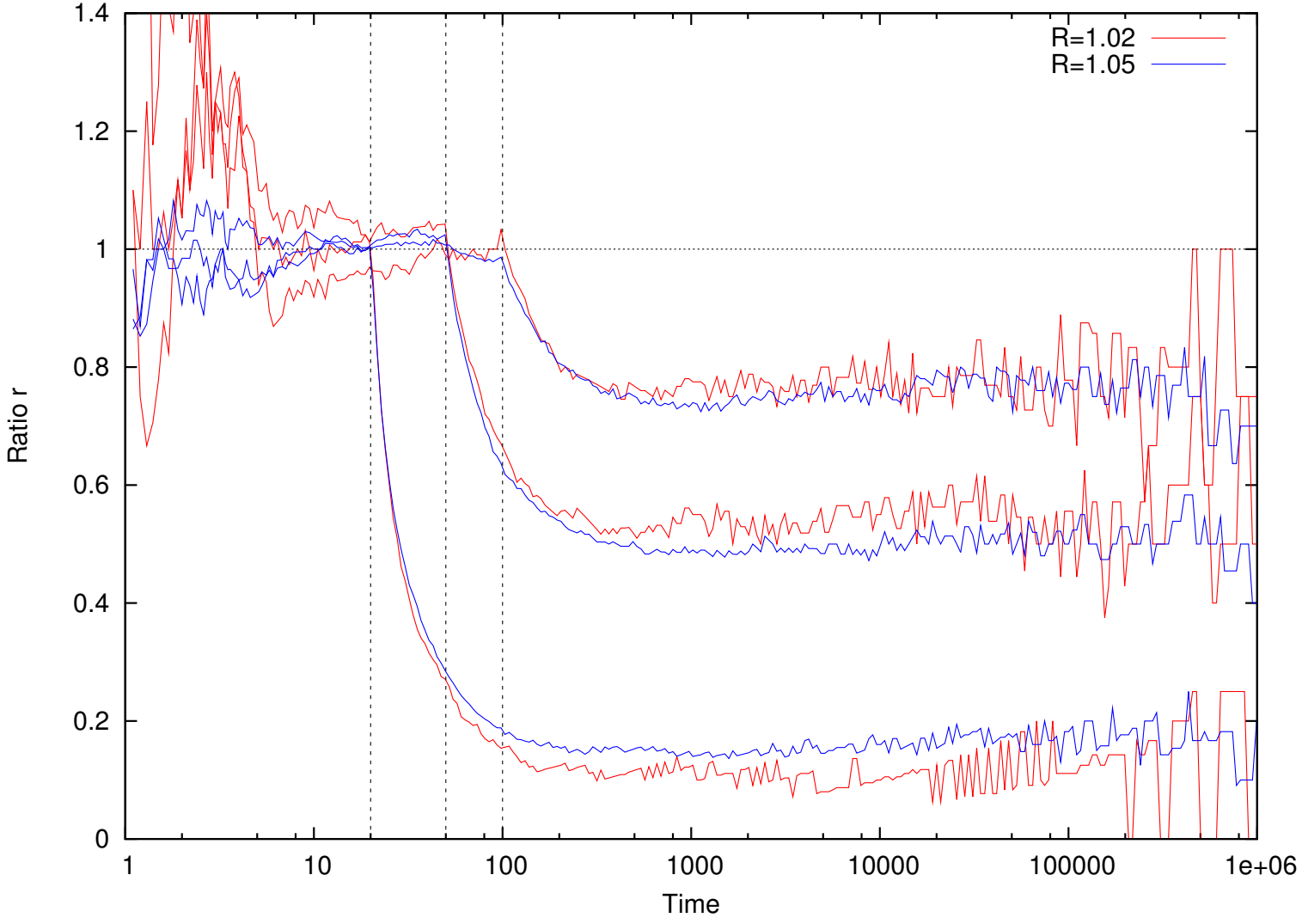
Asymmetric Potential [VanNess & BVL, *in prep.*]



Asymmetric Potential: Switched Off



Asymmetric Potential: Ratio of Off versus On



Domain Structure in Coarsening Dynamics

Conclusions

- The growth law exponent and the domain structure do not have the same universality.
- The growth law amplitude and the structure do have the same universality (or lack thereof)
- Memory erasure hypothesis says structure gets universality from the asymptotic dynamics.
- Numerical tests of the asymmetric Cahn-Hilliard equation challenge the MEH.
- Structure factor is not a sensitive measure — need to look at domain number.

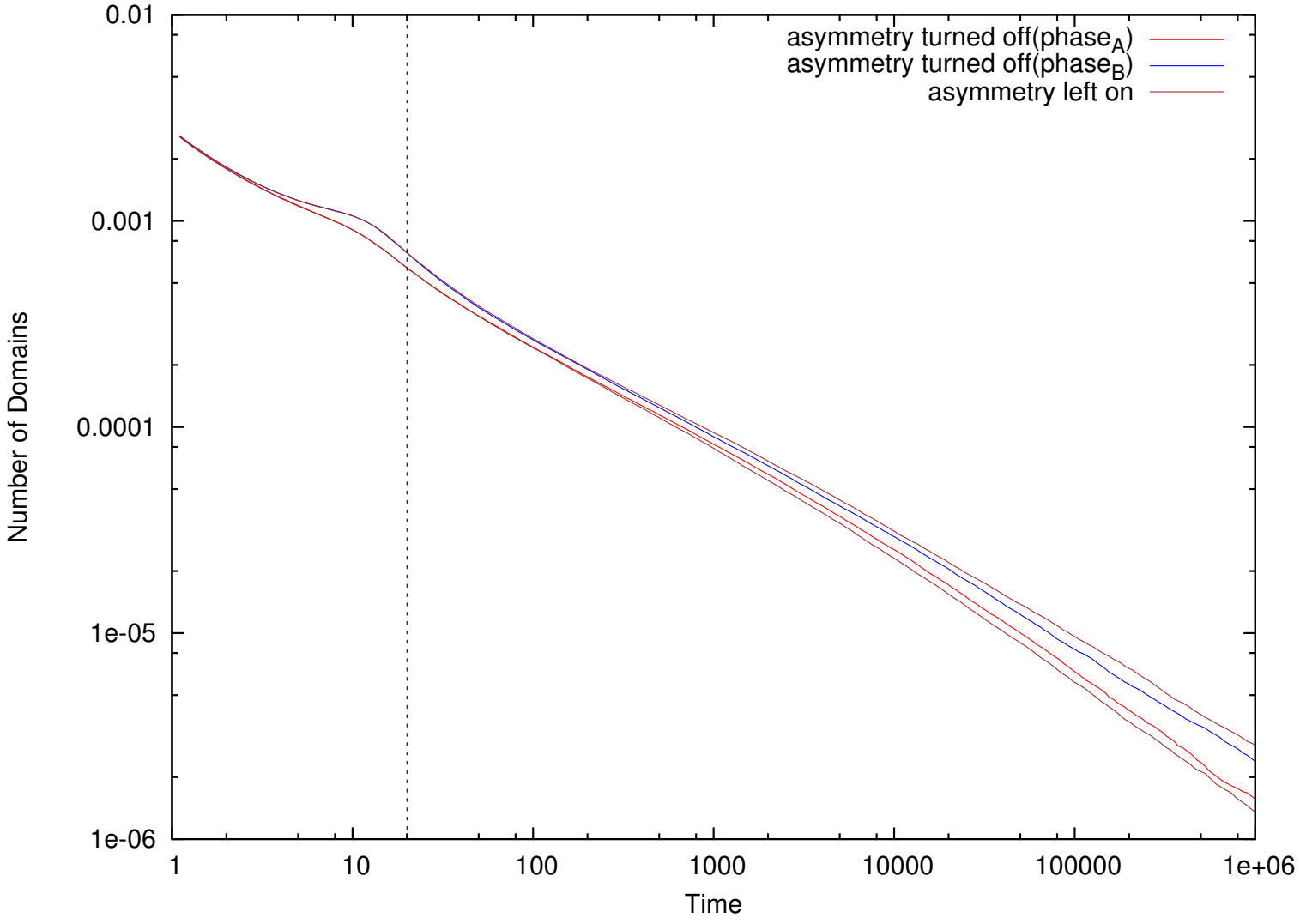
Future Work

- Still worth more testing of memory erasure hypothesis.
- Generalize defect dynamics analysis (vector order parameter, liquid crystals, hydrodynamics, facets, froths, . . .).
- For numerical tests, we need larger system sizes to push runs to later times.
- Generalizing ABCS theory:

  Should percolation apply for initial hull distributions with conserved dynamics? Our numerical data do not support this.

Thanks!

Kate's Plots: Mobility Switched Off



Kate's Plots: Mobility Ratio