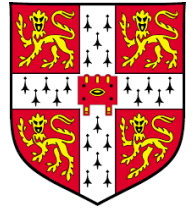
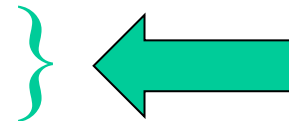


# Active Phase Separation



- Phase Separation in Passive Systems
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  - Stead-State Entropy Production

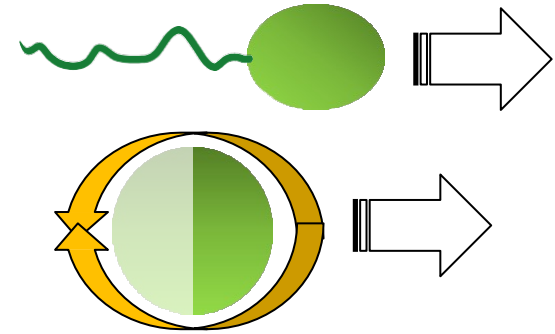


# Active Colloids / Micro-Organisms

Self propulsion via local drive mechanism  
steady-state entropy production

Examples: **Bacteria**

**Autophoretic colloids**



Pt-coated Janus particles bathed in fuel ( $\text{H}_2\text{O}_2$ )

*JR Howse et al, PRL 99 048102 (2007)*

Janus particles in binary solvent + laser heating

*I Buttinoni et al, PRL 110 238301 (2013)*

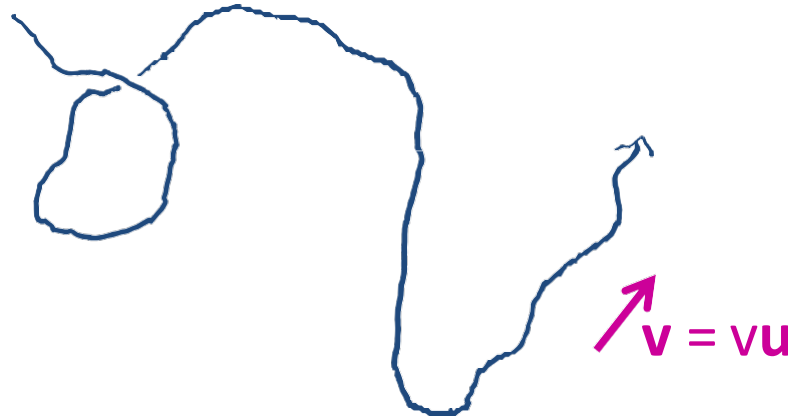
Each particle surfs a gradient of its own production

Cross-interactions:

*R Golestanian PRL 108 038303 (2012) et seq*

# Simplest Model of Active Motion: ABPs

single particle



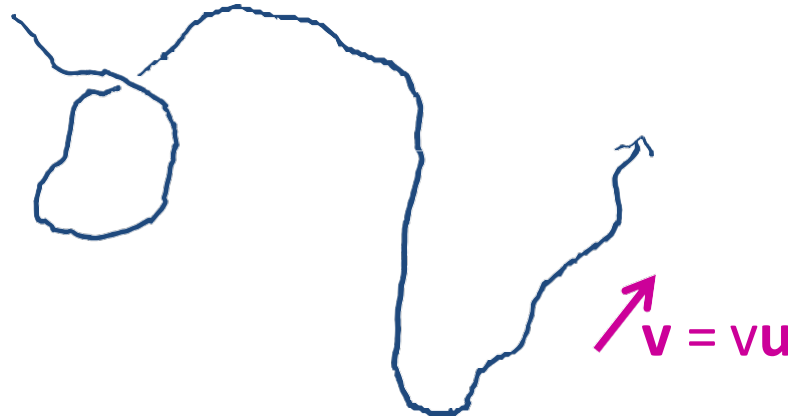
persistent Brownian motion  
speed  $v = v_0$ , rotational diffusivity  $D_r$   
orientation  $\mathbf{u}$

## Alternatives:

- Run-and-Tumble Particles ( $\mathbf{u}$  reassigns with rate  $\alpha$ )
- Active Ornstein-Uhlenbeck Particles
- various lattice models

## Simplest Model of Active Motion: ABPs

single particle



persistent Brownian motion  
speed  $v = v_0$ , rotational diffusivity  $D_r$   
orientation  $\mathbf{u}$

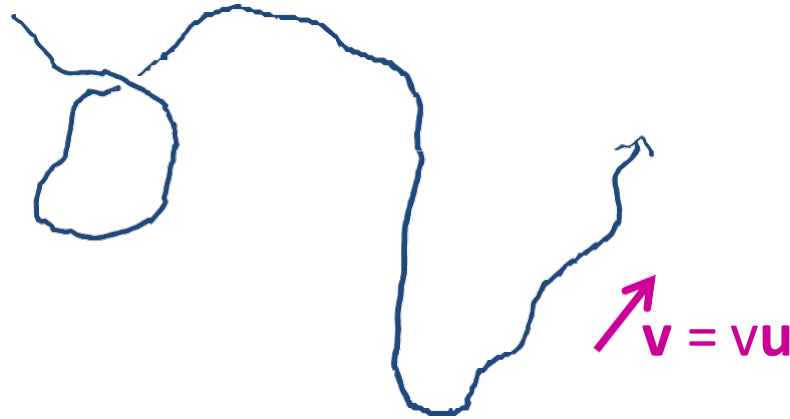
persistence length  $\gamma = v_0 \tau$

rotational relaxation time:  $\tau = ((d - 1)D_r)^{-1}$

defined via  $\langle \mathbf{u}(t) \cdot \mathbf{u}(t') \rangle = \exp[-|t - t'|/\tau]$

## Simplest Model of Active Motion: ABPs

single particle



persistent Brownian motion  
speed  $v = v_0$ , rotational diffusivity  $D_r$   
orientation  $\mathbf{u}$

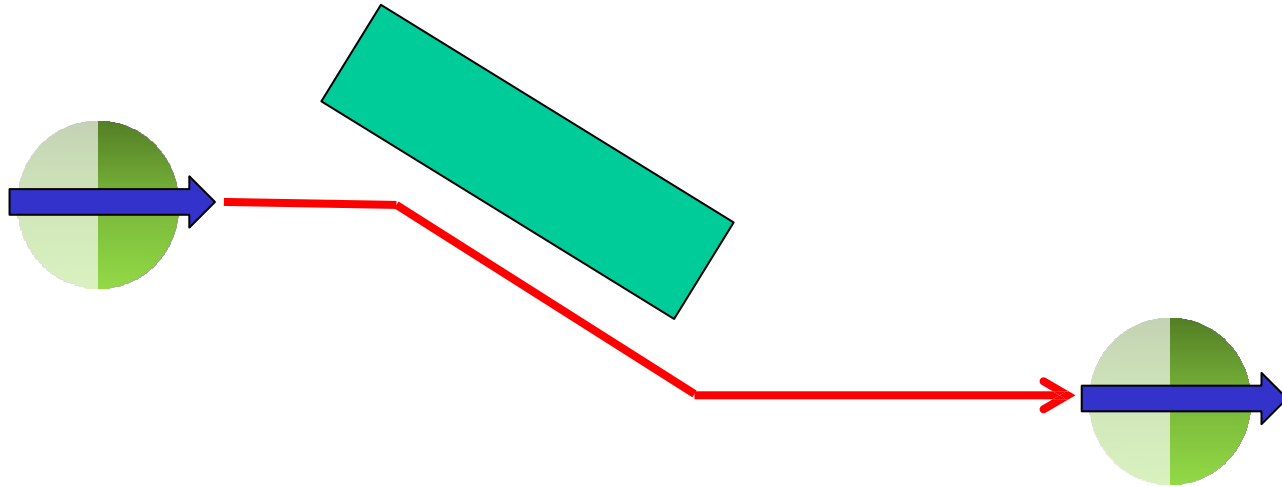
Coarse Grain to large scales  $\Rightarrow$  Random walk

$$D = \frac{v^2 \tau}{d}$$

$\cong$  simple Brownian motion with TRS if  $v$ ,  $\tau$  constant

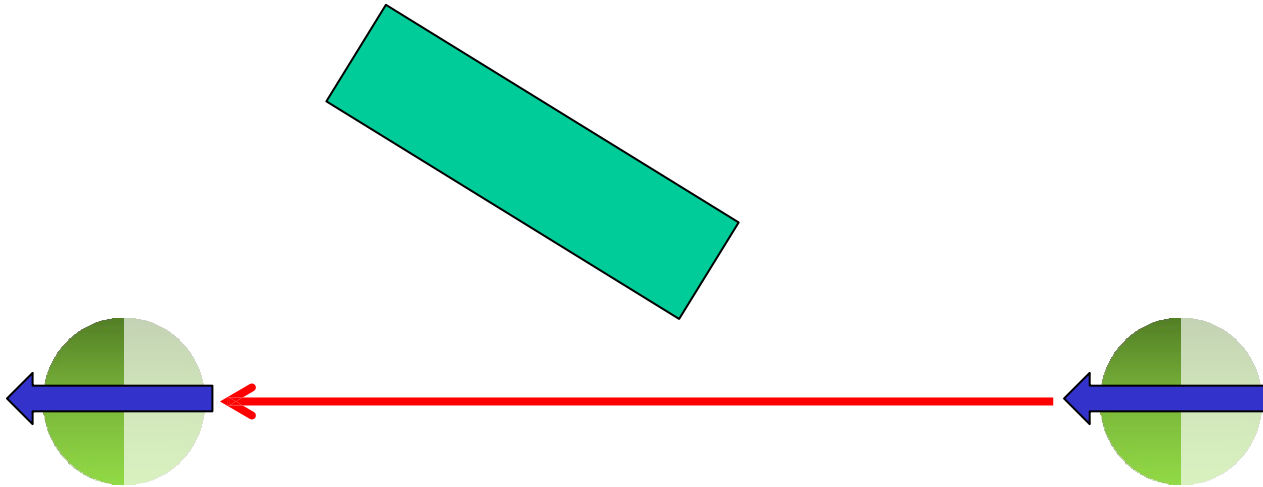
# TRS-Breaking Made Manifest

## 1. Interaction with obstacles

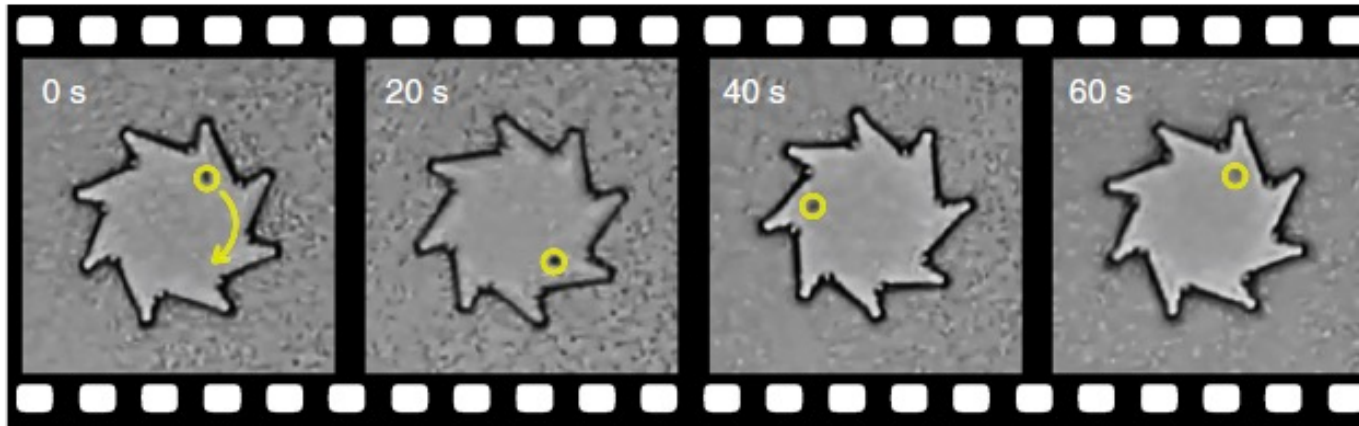


# TRS-Breaking Made Manifest

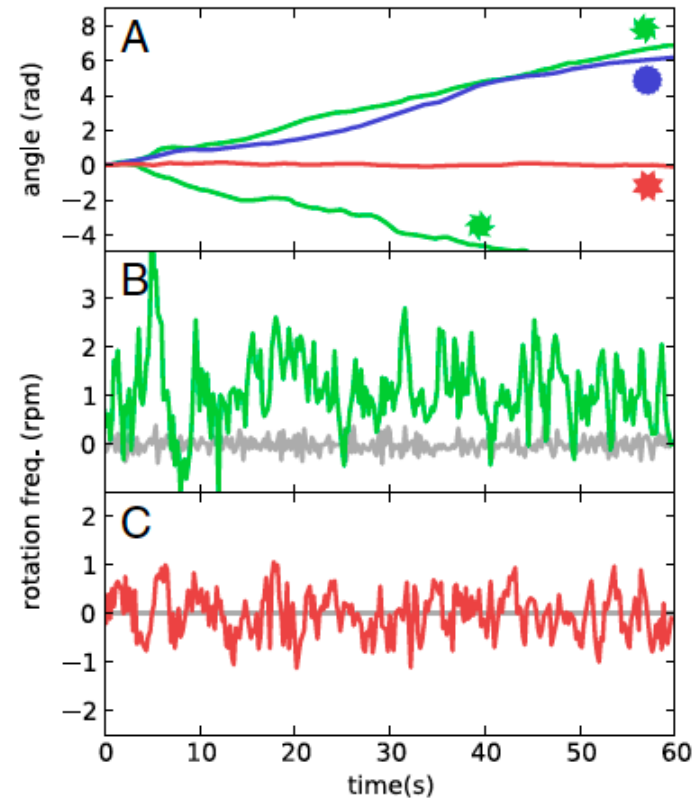
## 1. Interaction with obstacles



# Manifestations of TRS Breaking



Passive obstacle:  
rotor in bacterial bath



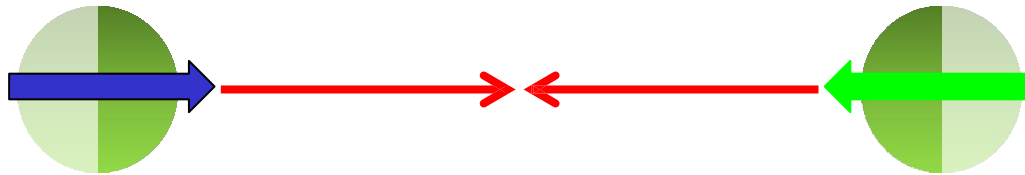
*R. di Leonardo et al, PNAS 2009*



# TRS-Breaking Made Manifest

1. Interaction with obstacles

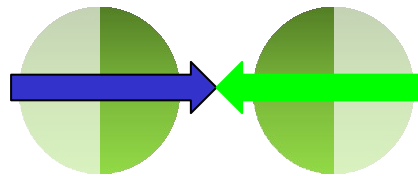
1. Interactions among particles



# TRS-Breaking Made Manifest

1. Interaction with obstacles

1. Interactions among particles

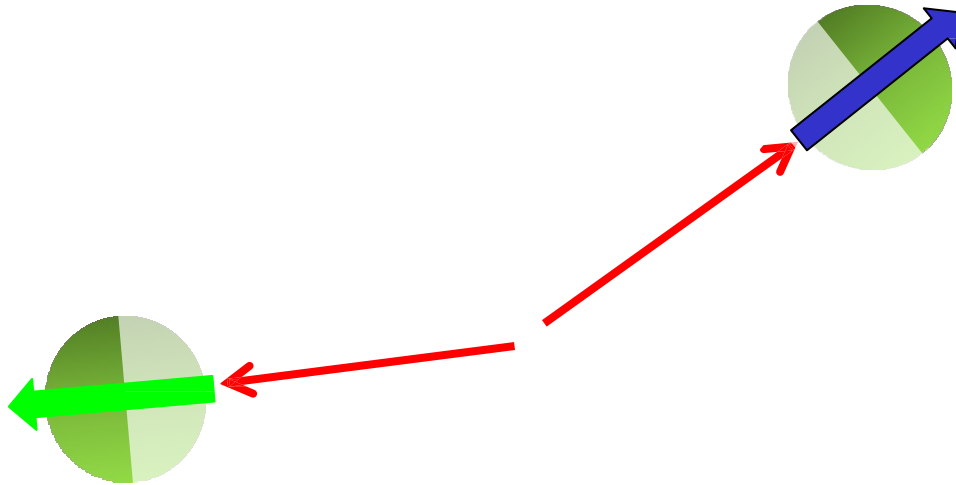


long pause  
to rotate

# TRS-Breaking Made Manifest

1. Interaction with obstacles

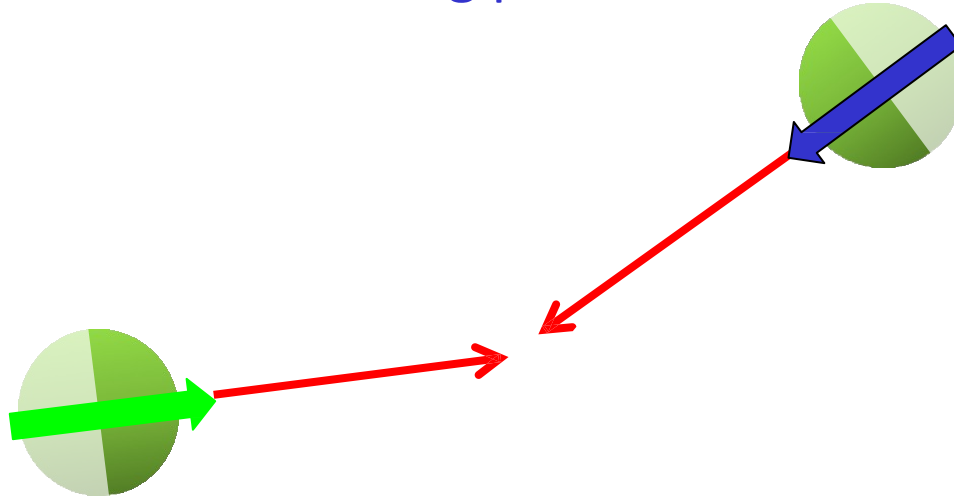
1. Interactions among particles



# TRS-Breaking Made Manifest

1. Interaction with obstacles

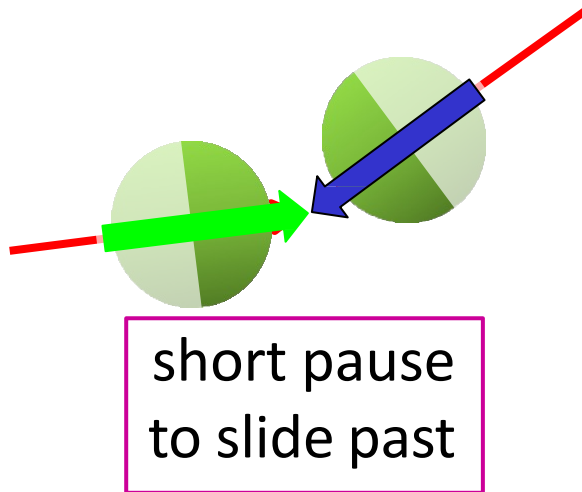
1. Interactions among particles



# TRS-Breaking Made Manifest

1. Interaction with obstacles

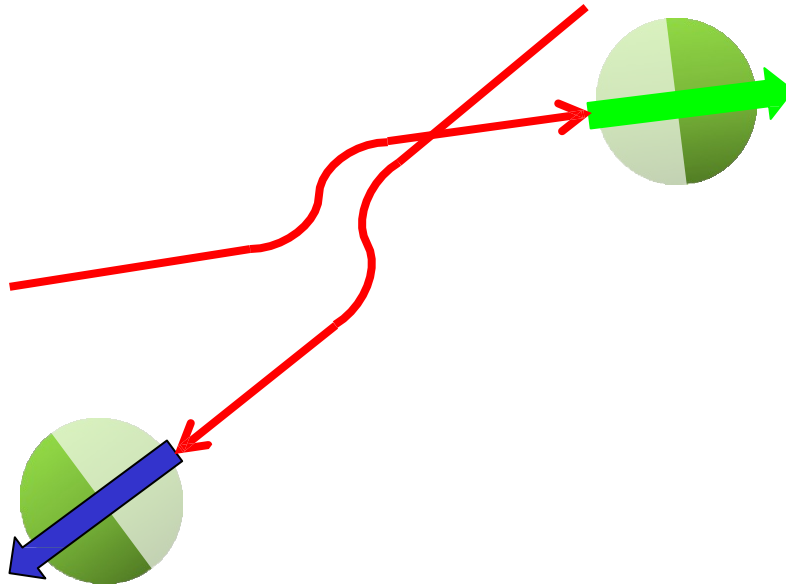
1. Interactions among particles



# TRS-Breaking Made Manifest

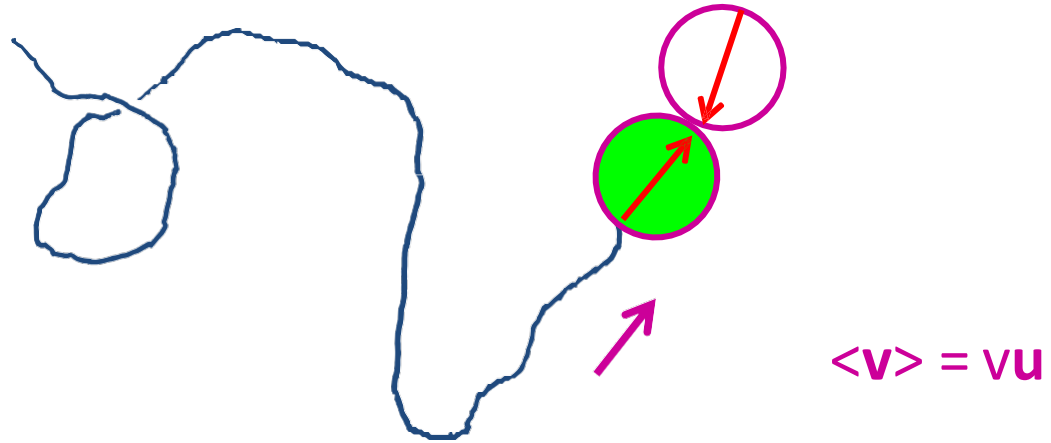
1. Interaction with obstacles

1. Interactions among particles



## Simplest Model of Active Motion: ABPs

many particles



persistent Brownian motion

speed  $v \neq v_0$ , rotational diffusivity  $D_r$  orientation  $\mathbf{u}$

### Main ABP physics: Collisional slowing down

$\approx$  density-dependent swim speed  $v[\rho] < v_0$  = 'Quorum Sensing'

*Y Fily and M C Marchetti PRL 108 235702 (2012) MEC and J  
Tailleur EPL 101 20010 (2013)*

# Manifestations of TRS Breaking



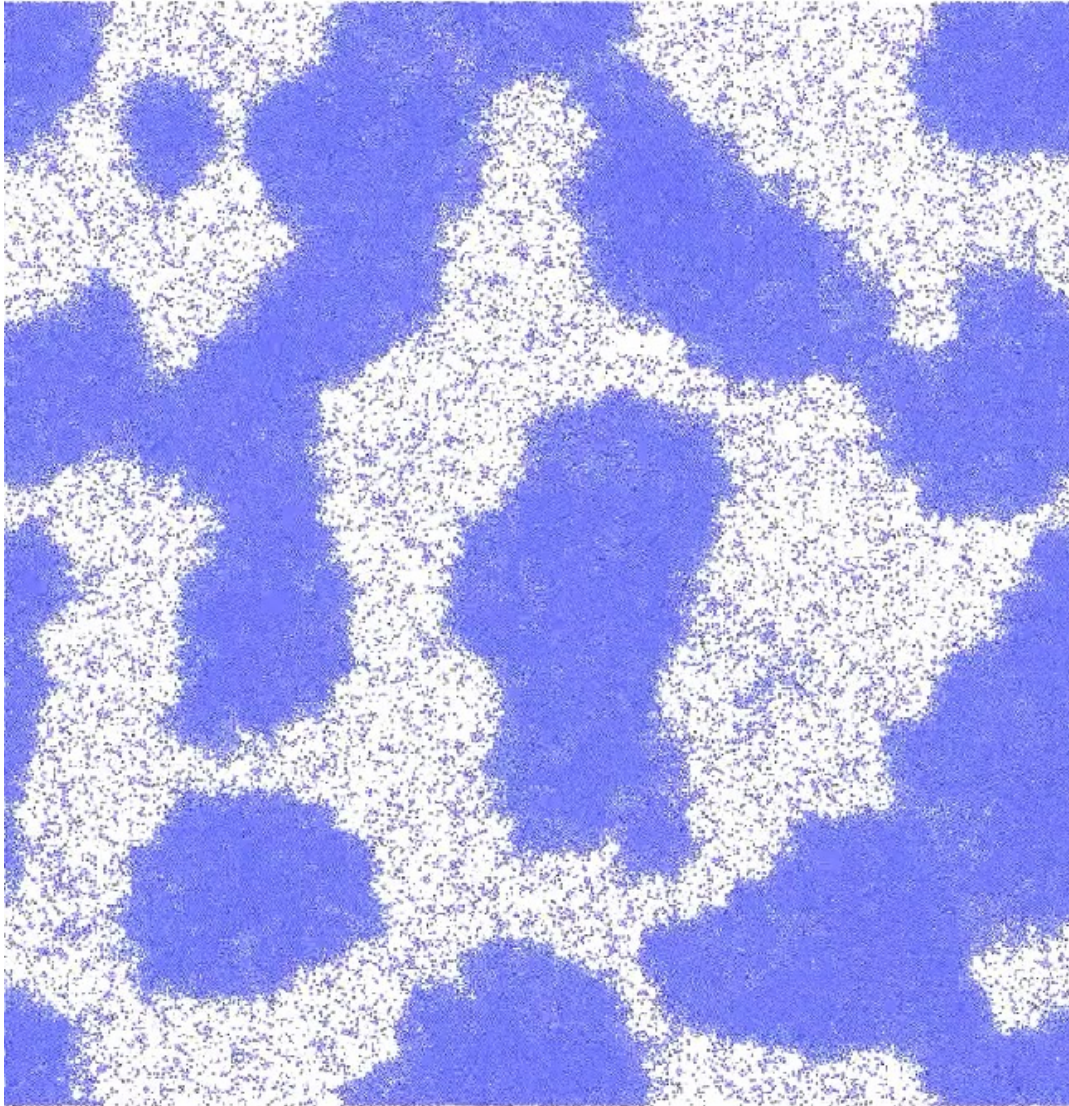
Motility-Induced  
Phase Separation  
(MIPS)

Janus particles in peroxide, light activated catalysis

*J Palacci et al, Science 2013*



# Motility-Induced Phase Separation (MIPS)



Motility-Induced  
Phase Separation  
(MIPS)

purely repulsive ABPs

*MEC + J Tailleur,  
Ann Rev CMP 2015*

*movie: J Stenhammar*

# MIPS: Phase Separation without TRS

Particles accumulate where they move slowly

Move slowly where they are dense: +ve feedback

Confirmed by simulations of repulsive ABPs

## Experiments:

MIPS seen, but complicated by phoretic, hydrodynamic... interactions

Also seen: microphase separation = **cluster phases** in steady state

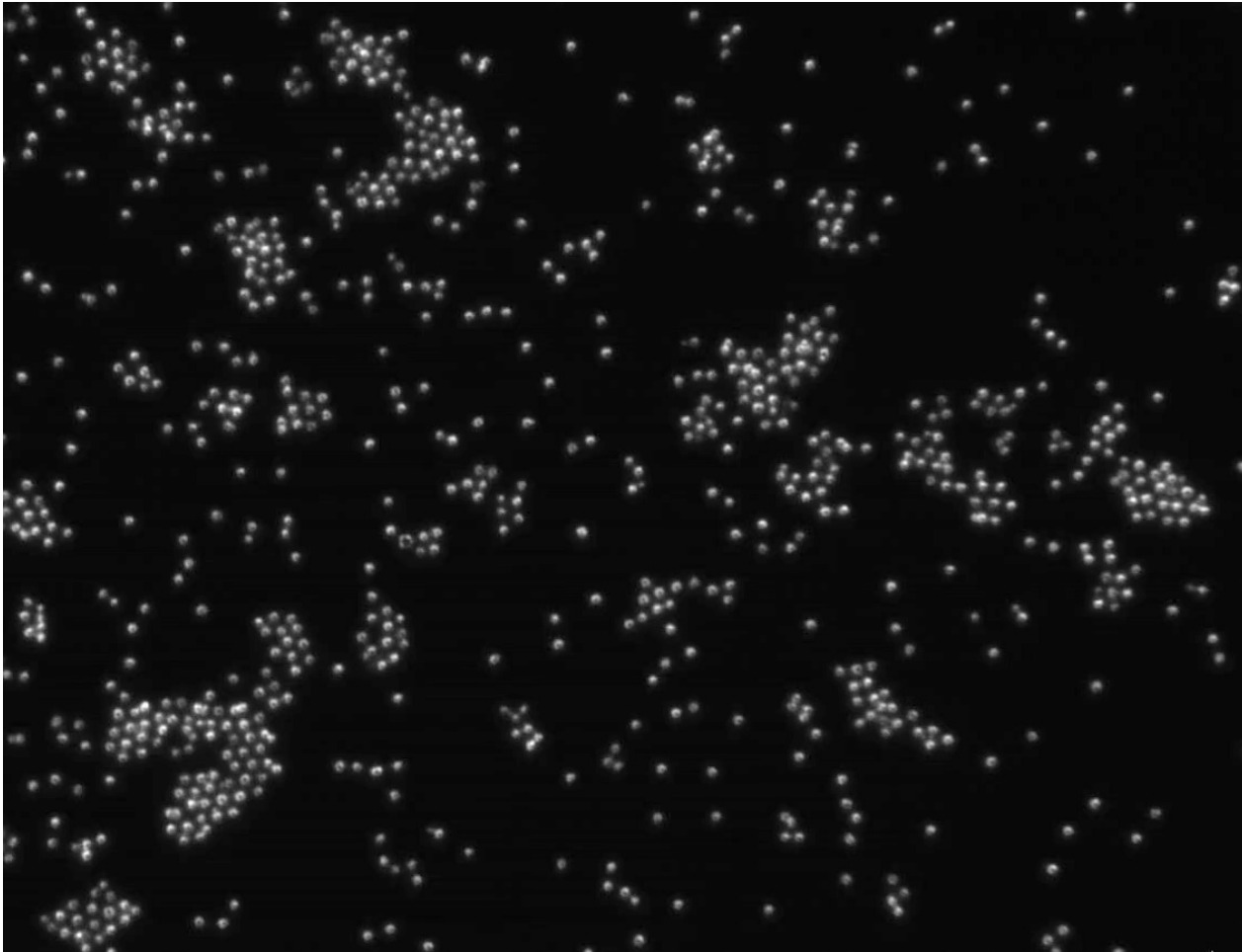
*MEC + J Tailleur PRL 2008, EPL 2013, Y Fily et al PRL 2012*

*J Stenhammar et al PRL 2014, I Theurkauff et al PRL 2012*

*I Buttinoni et al, PRL 2013, J Palacci et al, Science 2013*

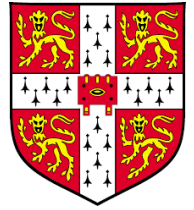
*J Schwarz-Linek et al, PNAS 2012*

# Cluster Phases

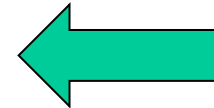


*Movie: F Ginot et al, Nat Comms 2018*

# Active Phase Separation



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# Field Theory of Phase Separation

$$\dot{\phi} = -\nabla \cdot \mathbf{J}$$

$$\mathbf{J} = -\nabla \mu + \sqrt{2D} \Lambda$$

$$\mu = a\phi + b\phi^3 - \kappa \nabla^2 \phi$$

$\Lambda$  = unit white noise

$D = k_B T M$

$M = 1$  mobility

## MODEL B

Model B has Time Reversal Symmetry:  
Forward and backward movies are statistically identical  
once steady state is achieved

# Field Theory of Phase Separation

$$\dot{\phi} = -\nabla \cdot \mathbf{J}$$

$$\mathbf{J} = -\nabla \mu + \sqrt{2D} \Lambda$$

$$\mu = a\phi + b\phi^3 - \kappa \nabla^2 \phi$$

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$D = k_B T M$

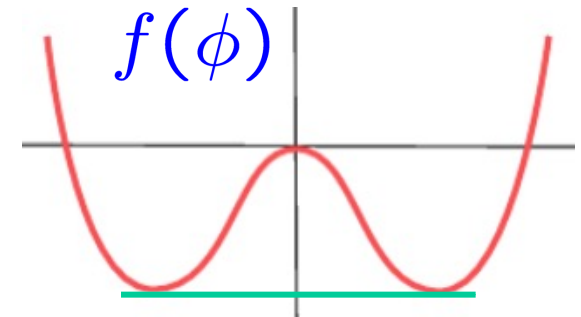
$M = 1$  mobility

## MODEL B

$$\mu = \delta \mathcal{F} / \delta \phi$$

$$\mathcal{F} = \int dV [f(\phi) + \frac{\kappa}{2} (\nabla \phi)^2]$$

$$f(\phi) = \frac{a}{2} \phi^2 + \frac{b}{4} \phi^4$$



phase equilibria:  
common tangent

$$\mu_1 = \mu_2$$

$$P_1 = P_2$$

where  $P = \mu\phi - f$

# Active Field Theory of Phase Separation

$$\dot{\phi} = -\nabla \cdot \mathbf{J}$$

$$\mathbf{J} = -\nabla \hat{\mu} + \sqrt{2D\Lambda}$$

$$\hat{\mu} = a\phi + b\phi^3 - \kappa \nabla^2 \phi + \lambda (\nabla \phi)^2$$

## ACTIVE MODEL B

$$\lambda (\nabla \phi)^2 \neq \delta \mathcal{F} / \delta \phi \text{ for any } \mathcal{F}$$

= **minimal** violation of TRS

This form of TRS violation can be derived microscopically...

e.g. via  $\mathbf{v} = \mathbf{v}(\rho_{\text{local}})$

*E Tjhung et al PRX 2018*

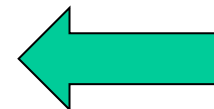
*R Wittkowski et al, Nat Comms 2014*



# Active Phase Separation



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# Active Field Theory of Phase Separation

**MIPS:** uncommon tangent construction

$$\hat{\mu}_1 = \hat{\mu}_2$$

$$\hat{\mu}_1\phi_1 - f_1 \neq \hat{\mu}_2\phi_2 - f_2$$

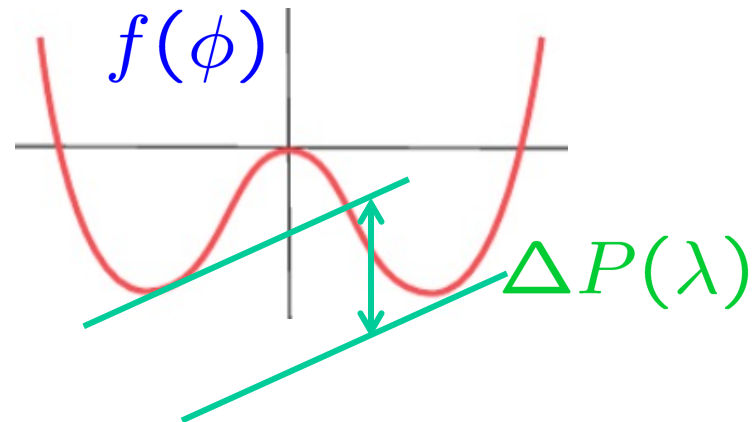
## ACTIVE MODEL B

Calculation Method:

(a) 1D interface

(b)  $J = 0$  requires  $\partial_x \hat{\mu} = 0 \Rightarrow \hat{\mu}_1 = \hat{\mu}_2$

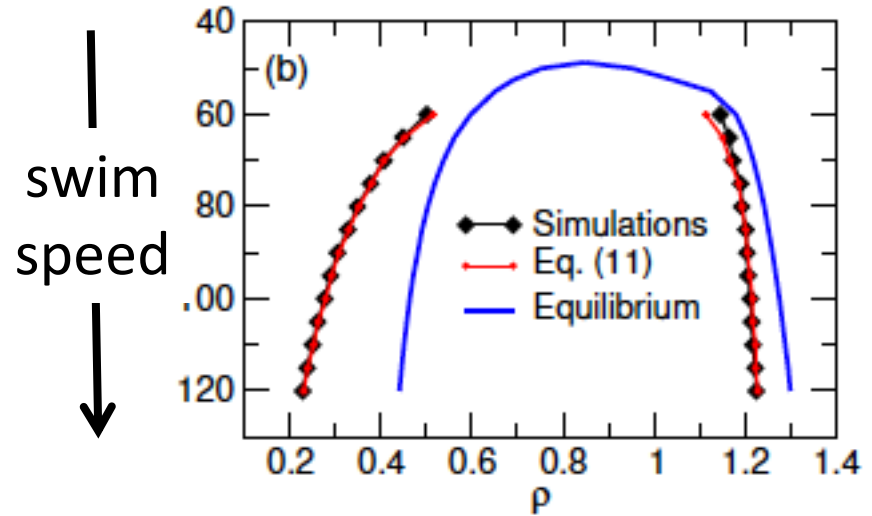
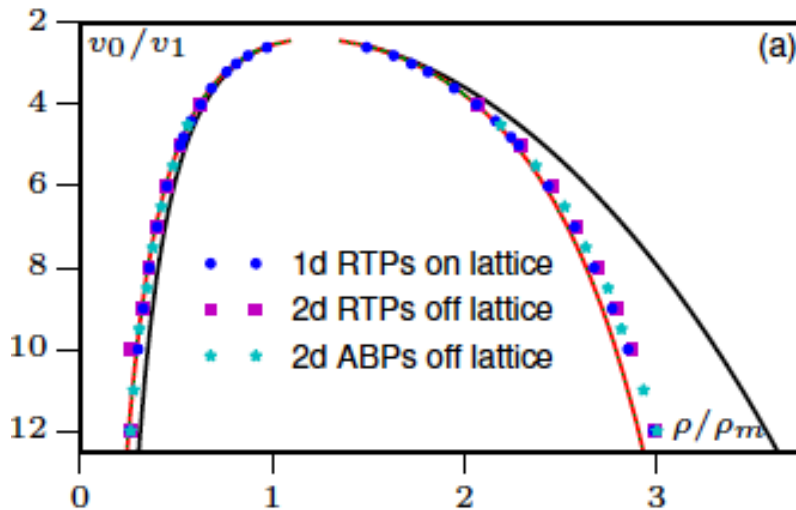
(c) seek  $J = 0$  solutions with  $\partial_x \phi = 0$  at  $x = \pm\infty$



*R Wittkowski et al, Nat Comms 2014*

*A Solon et al, PRE 2018*

# Active Field Theory of Phase Separation



swim  
speed

**Anomalous Phase Coexistence: seen for particle-based models**

$v(\rho)$  model: density-dependent swim speed

ABPs with hard-core collisions: similar outcomes, different analysis

*A Solon et al, PRE 2018*

## Active Model B

$$\dot{\phi} = -\nabla \cdot \mathbf{J}$$

$$\mathbf{J} = -\nabla \hat{\mu} + \sqrt{2D} \Lambda$$

$$\hat{\mu} = a\phi + b\phi^3 - \kappa \nabla^2 \phi + \lambda (\nabla \phi)^2$$

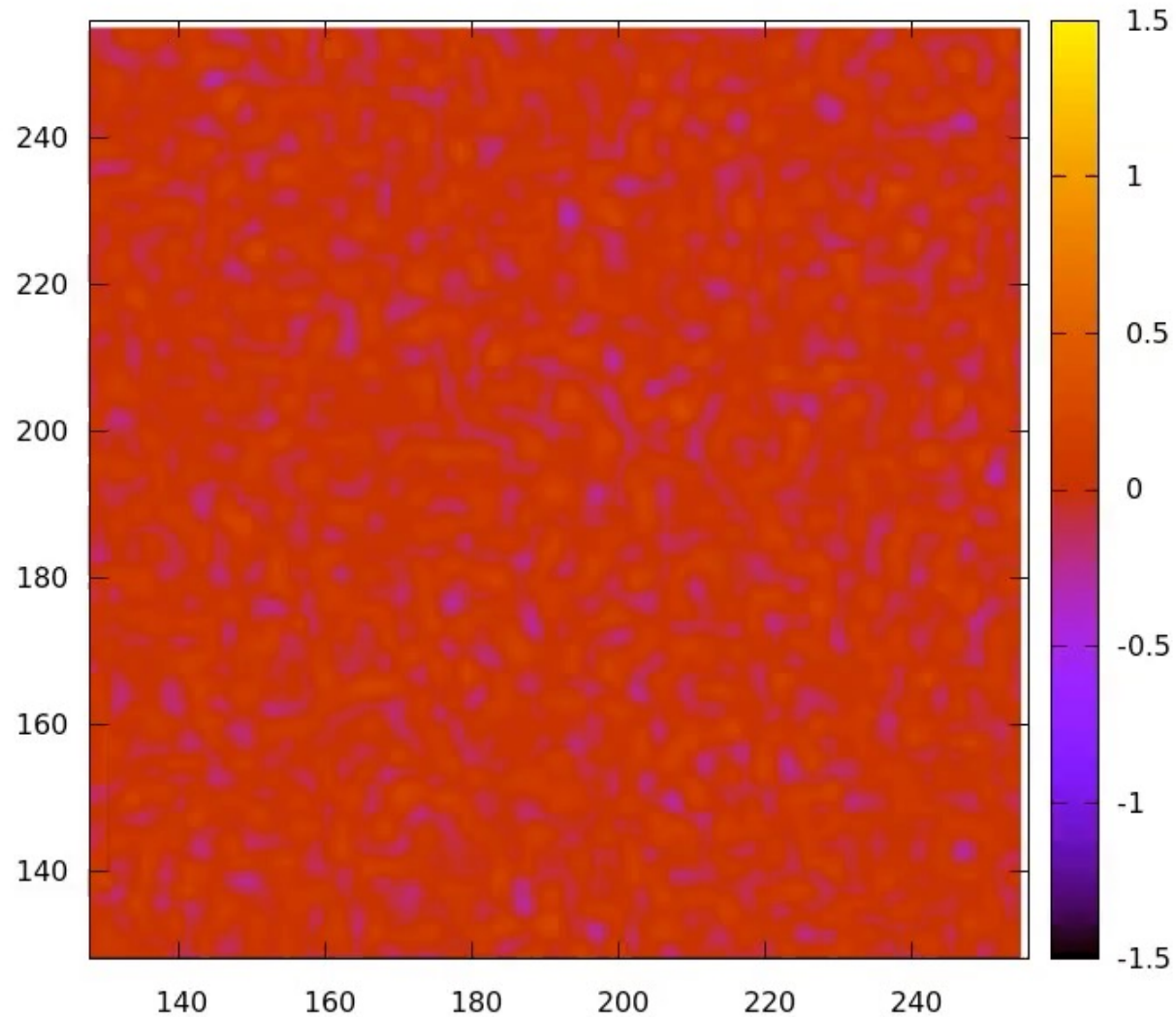
Anomalous Phase Coexistence ✓

Otherwise similar to passive phase separation

- Spinodal, Nucleation+ Growth regimes
- Late stage coarsening law  $L(t) \propto t^{1/3}$

# Phase Separation Kinetics: Active Model B

## Spinodal decomposition



## Active Model B

$$\dot{\phi} = -\nabla \cdot \mathbf{J}$$

$$\mathbf{J} = -\nabla \hat{\mu} + \sqrt{2D} \Lambda$$

$$\hat{\mu} = a\phi + b\phi^3 - \kappa \nabla^2 \phi + \lambda (\nabla \phi)^2$$

Anomalous Phase Coexistence ✓

Otherwise similar to passive phase separation

- Spinodal, Nucleation+ Growth regimes
- Late stage coarsening law  $L(t) \propto t^{1/3}$

|                         |                                                              |
|-------------------------|--------------------------------------------------------------|
| <b>Absent features:</b> | bubble / cluster phases<br>TRS violation in stationary state |
|-------------------------|--------------------------------------------------------------|

## Active Model B

$$\dot{\phi} = -\nabla \cdot \mathbf{J}$$

$$\mathbf{J} = -\nabla \hat{\mu} + \sqrt{2D\Lambda}$$

$$\hat{\mu} = a\phi + b\phi^3 - \kappa \nabla^2 \phi + \lambda (\nabla \phi)^2$$

What's missing?

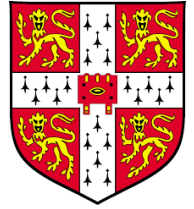
Anomalous Phase Coexistence

Otherwise similar to passive phase separation

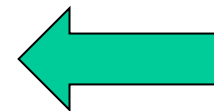
- Spinodal, Nucleation+ Growth regimes
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**Absent features:** bubble / cluster phases  
TRS violation in stationary state

# Active Phase Separation



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## Active Model B+

$$\dot{\phi} = -\nabla \cdot \mathbf{J}$$

$$\mathbf{J} = -\nabla \hat{\mu} + \sqrt{2D}\mathbf{\Lambda} + \zeta [(\nabla^2 \phi) \nabla \phi - \frac{1}{2} \nabla (\nabla \phi)^2]$$

$$\hat{\mu} = a\phi + b\phi^3 - \kappa \nabla^2 \phi + \lambda (\nabla \phi)^2$$

Expansion of active currents to order  $\nabla^3, \phi^2$

$\zeta$  and  $\lambda$  terms same order, separately break TRS

$\zeta$  term vanishes for flat interface: no effect on bulk coexistence

What does the  $\zeta$  term do?

*C. Nardini et al, PRX 2017*

*E. Tjhung et al, PRX 2018*

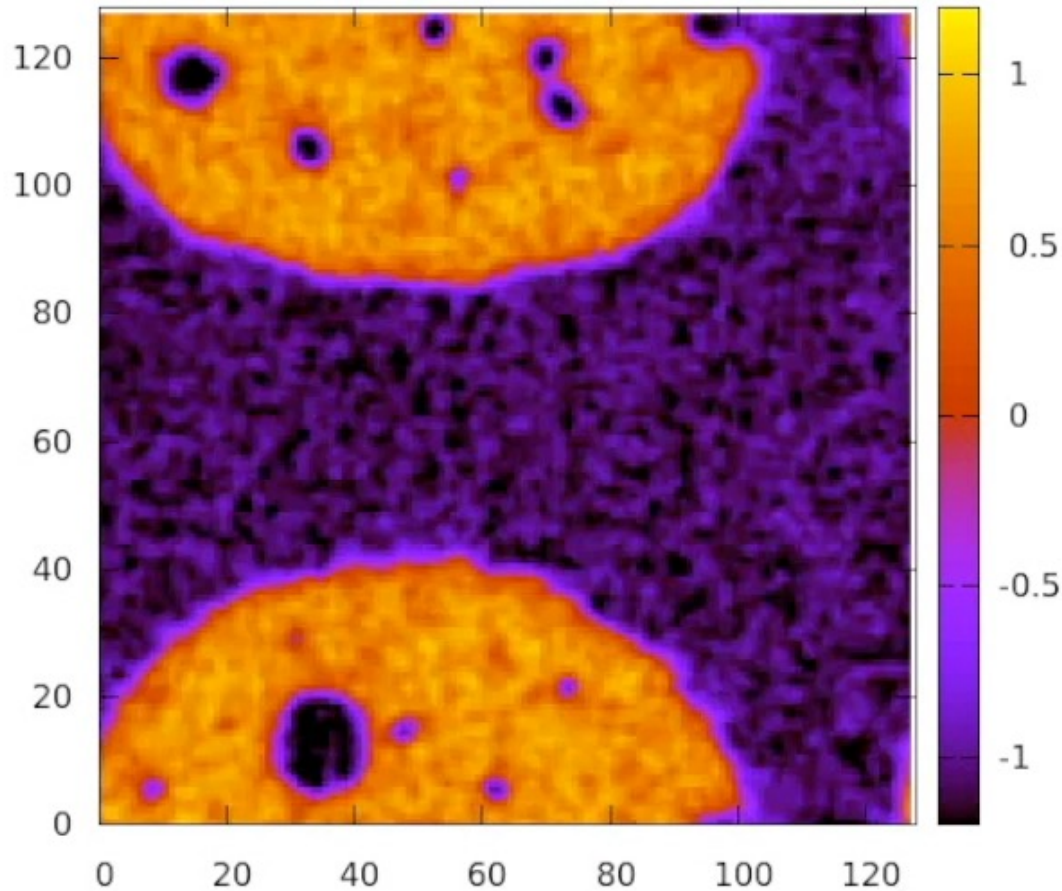


# Bubbles and Clusters

## Active Model B+

$t = 13020$

$\zeta > 0$



*E. Tjhung et al, PRX 2018*

## Active Model B+

$$\dot{\phi} = -\nabla \cdot \mathbf{J}$$

$$\mathbf{J} = -\nabla \hat{\mu} + \sqrt{2D}\mathbf{\Lambda} + \zeta [(\nabla^2 \phi) \nabla \phi - \frac{1}{2} \nabla (\nabla \phi)^2]$$

$$\hat{\mu} = a\phi + b\phi^3 - \kappa \nabla^2 \phi + \lambda (\nabla \phi)^2$$

*C. Nardini et al, PRX 2017*

*E. Tjhung et al, PRX 2018*

## Active Model B+

$$\dot{\phi} = -\nabla \cdot \mathbf{J}$$

$$\mathbf{J} = -\nabla \hat{\mu} + \sqrt{2D\Lambda} \mathbf{J}_{\zeta}$$

$$\hat{\mu} = a\phi + b\phi^3 - \kappa \nabla^2 \phi + \lambda (\nabla \phi)^2$$

Dealing with the  $\zeta$  current:

$$\mathbf{J}_{\zeta} = -\nabla \mu_{\zeta} + \underbrace{\nabla \times \mathbf{H}}$$

Leaves nonlocal chemical potential:

$$\mu_{\zeta}(\mathbf{r}) = - \int d\mathbf{r}' \frac{\nabla \cdot \mathbf{J}_{\zeta}}{4\pi |\mathbf{r} - \mathbf{r}'|}$$

invisible for  
 $\phi$  dynamics

Coulomb integral:  
dipole density  
 $\propto$  curvature

# Active Model B+

$$\dot{\phi} = -\nabla \cdot \mathbf{J}$$

$$\mathbf{J} = -\nabla \hat{\mu} + \sqrt{2D\Lambda} + \mathbf{J}_{\zeta}$$

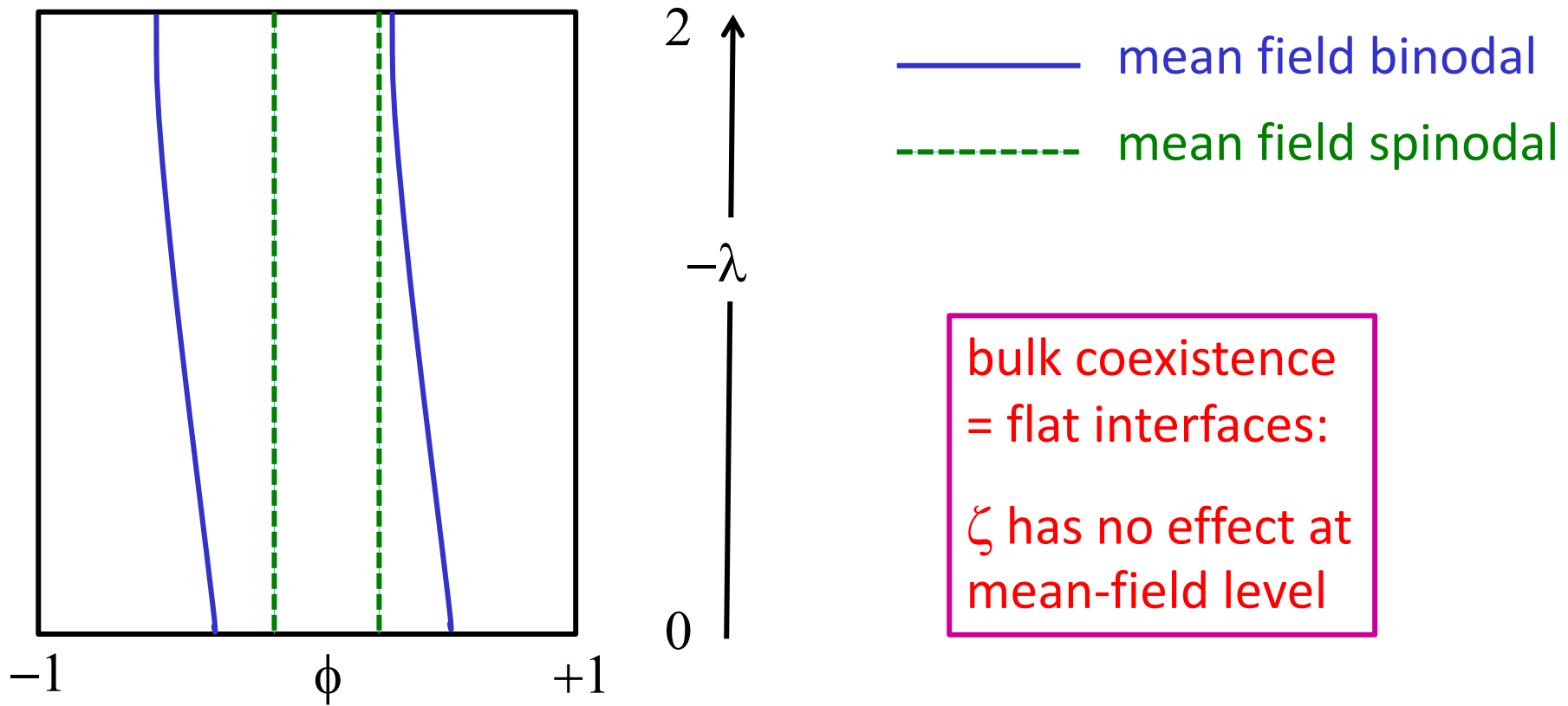
- $\zeta$  term changes the way chemical potential varies with interfacial curvature
- dramatic consequences for phase separation... whenever droplets involved

$$\mu_{\zeta}(\mathbf{r}) = - \int d\mathbf{r}' \frac{\nabla \cdot \mathbf{J}_{\zeta}}{4\pi |\mathbf{r} - \mathbf{r}'|}$$

Coulomb integral:  
dipole density  
 $\propto$  curvature

# Bubbles and Clusters

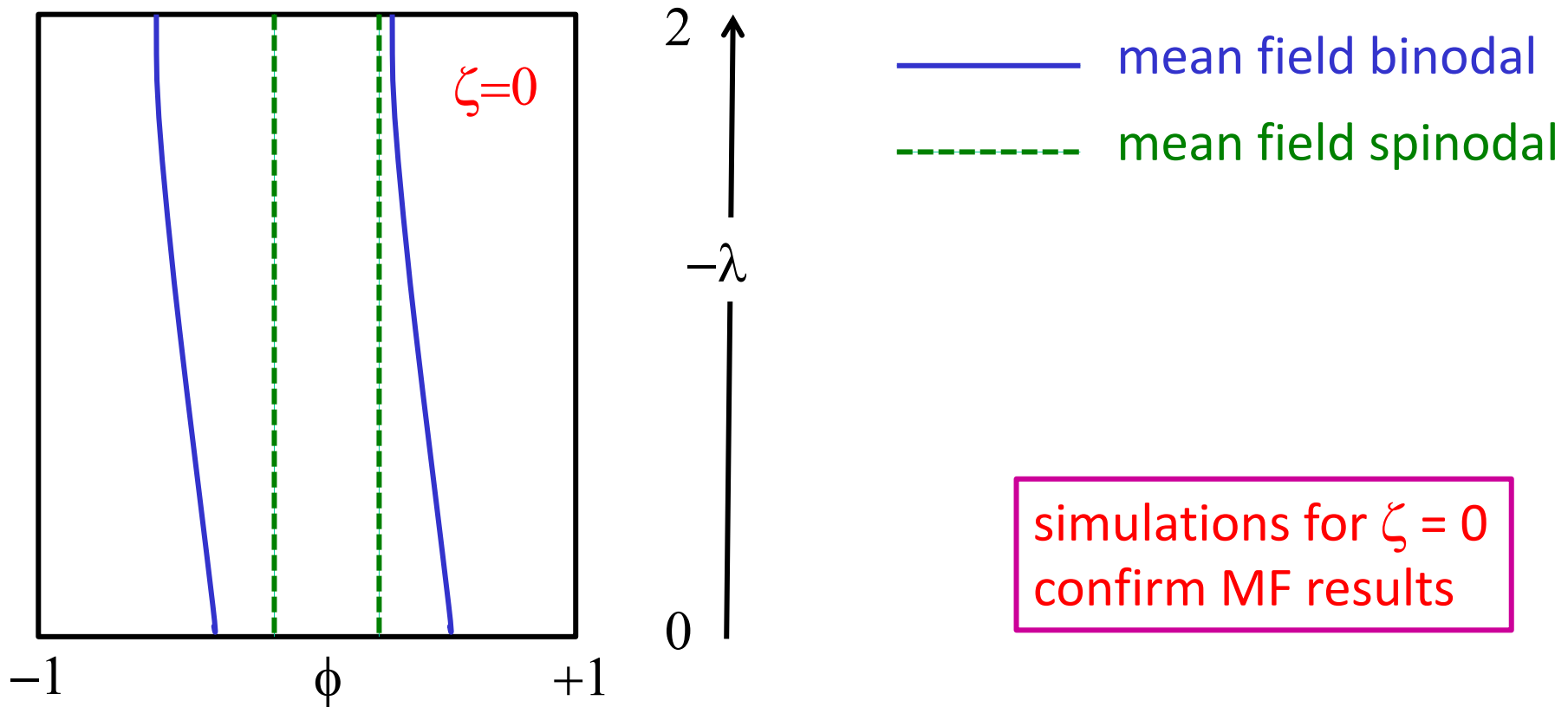
Active Model B+ schematic phase diagram at fixed  $a, b, \kappa, D$



*E. Tjhung et al., PRX 8, 031080 (2018)*

# Bubbles and Clusters

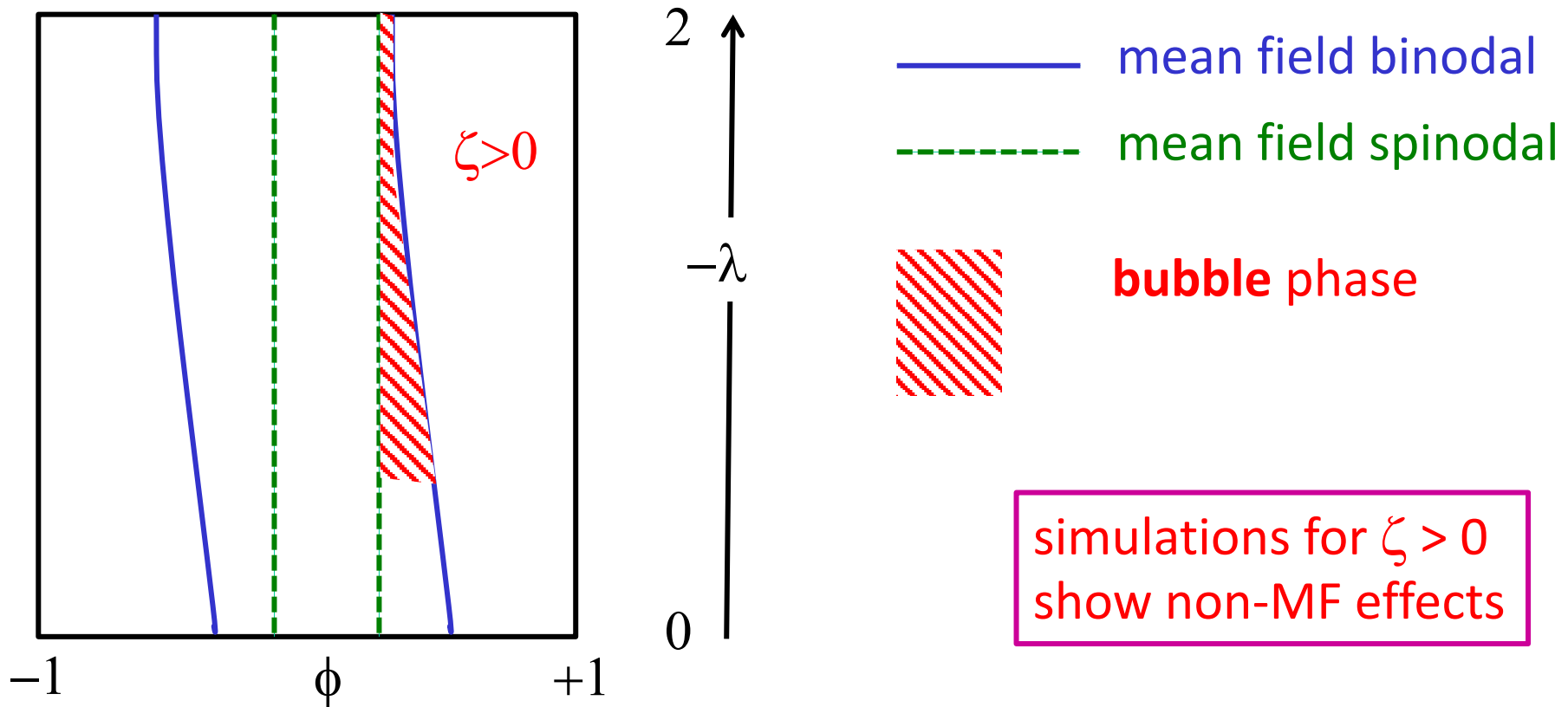
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*E. Tjhung et al., PRX 8, 031080 (2018)*

# Bubbles and Clusters

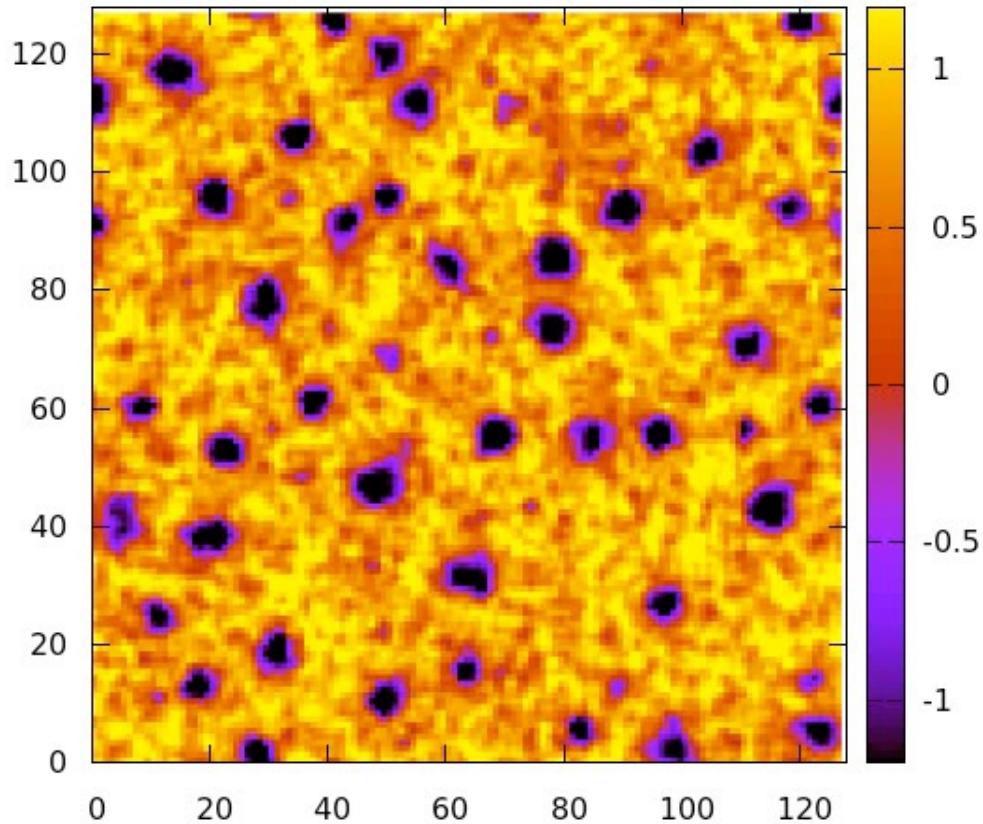
Active Model B+ schematic phase diagram at fixed  $a, b, \kappa, D$



*E. Tjhung et al., PRX 8, 031080 (2018)*

# Bubbles and Clusters

## Active Model B+



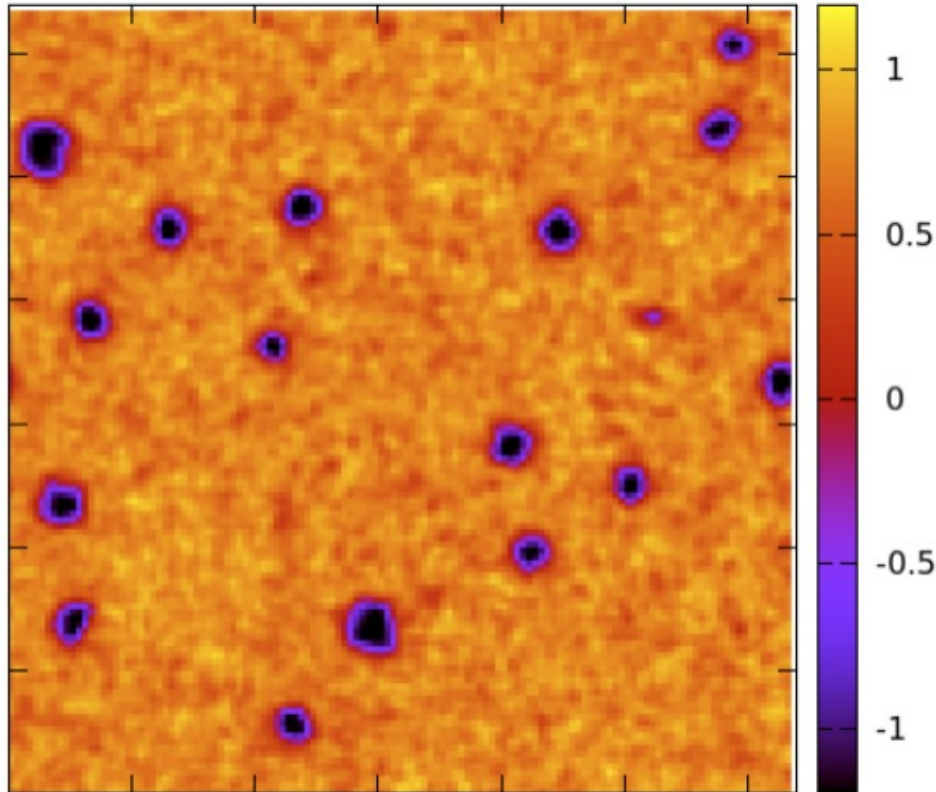
**bubble phase**

*E. Tjhung et al, PRX 2018*



# Bubbles and Clusters

## Active Model B+



**bubble** phase

symmetry of model:

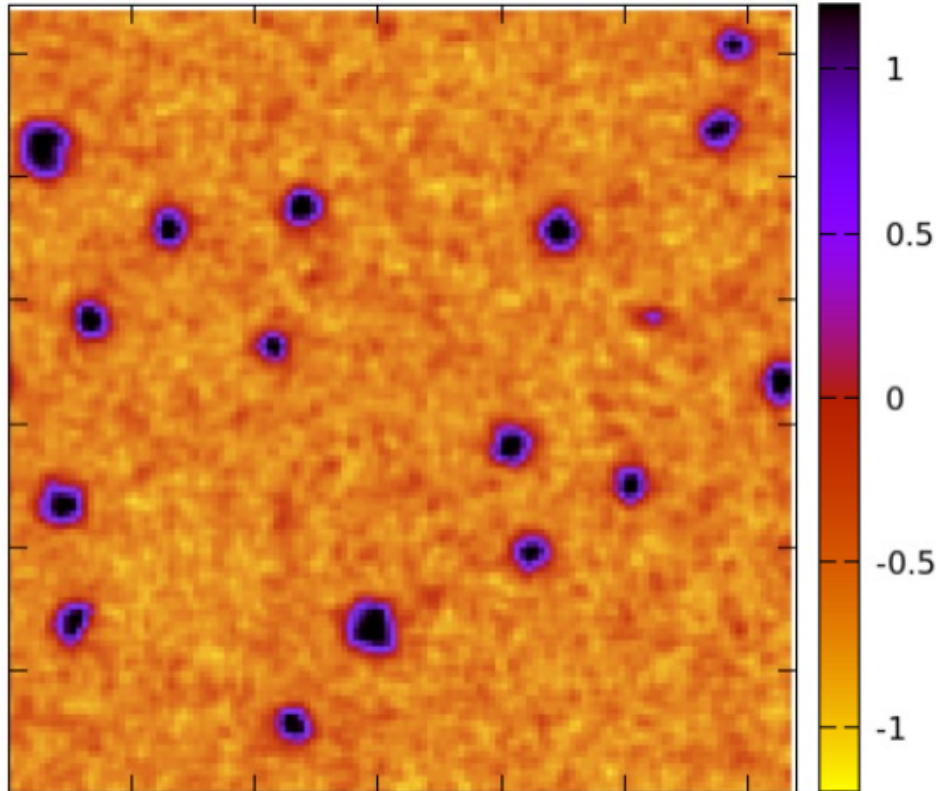
$$(\zeta, \lambda, \phi) \rightarrow -(\zeta, \lambda, \phi)$$

**bubbles**  $\rightarrow$  **clusters**

*E. Tjhung et al, PRX 2018*

# Bubbles and Clusters

## Active Model B+



**cluster phase**

symmetry of model:

$$(\zeta, \lambda, \phi) \rightarrow -(\zeta, \lambda, \phi)$$

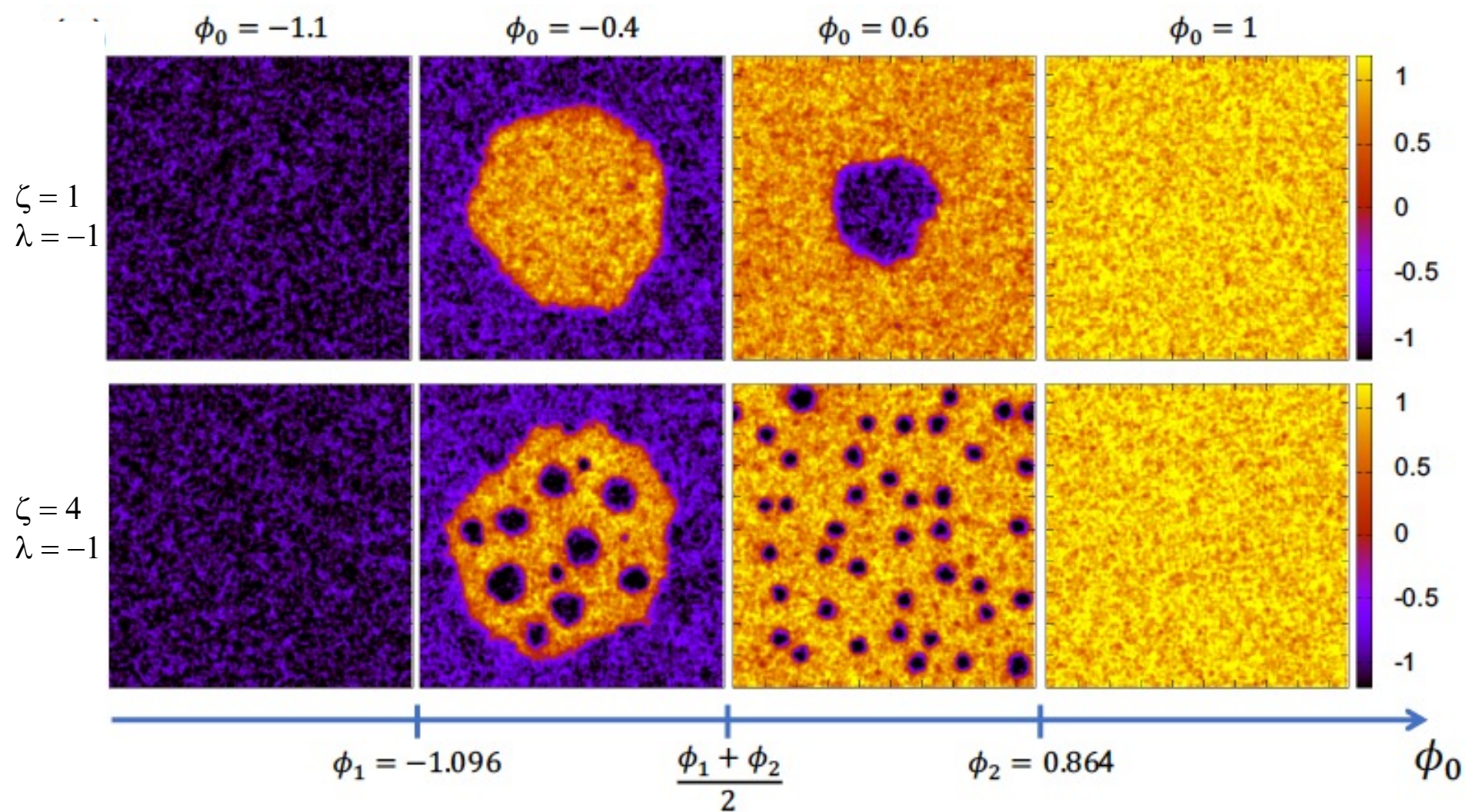
**bubbles  $\rightarrow$  clusters**

ABPs: bubbles ( $\zeta > 0$ )

Colloid experiments:  
clusters ( $\zeta < 0$ )

*E. Tjhung et al, PRX 2018*

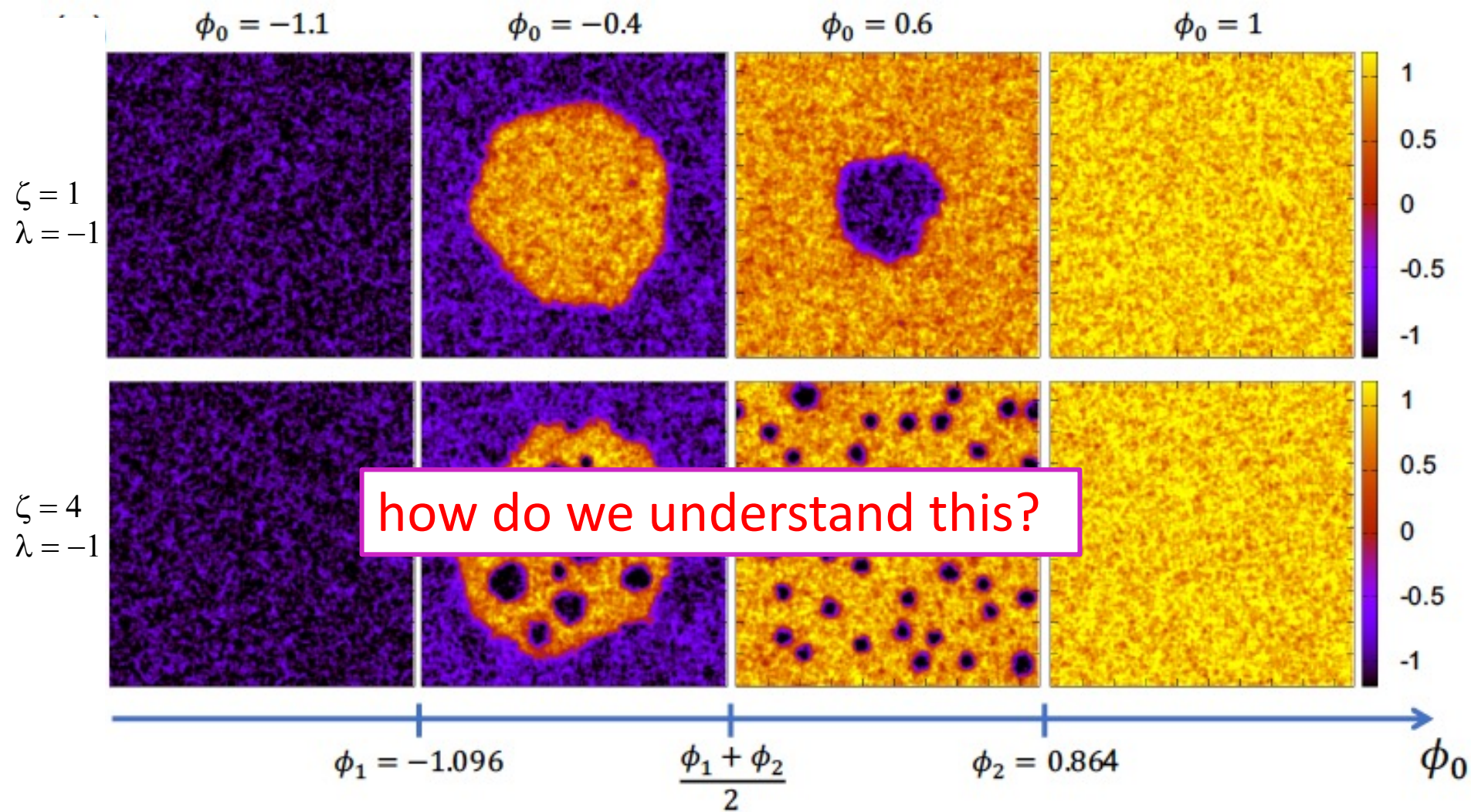
# Phase Diagram: Microphase Separation



*E. Tjhung et al, PRX 2018*



# Phase Diagram: Microphase Separation



*E. Tjhung et al, PRX 2018*

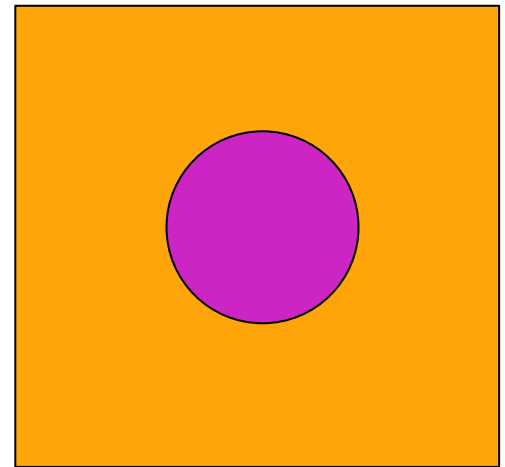
# Ostwald Ripening: Passive Case

$\mu$  raised at curved interface:  $\mu_I(R) = \frac{\gamma(d-1)}{\phi_B R}$

( $\gamma$  = tension,  $\phi_B$  = binodal,  $R$  = radius)

Current at surface:  $J(R) = -\nabla\mu(R) = \frac{\mu_\infty - \mu_I(R)}{R^2}$

Single droplet coexists with vapour at  $\mu_\infty = \mu_l(R)$  in finite system



# Ostwald Ripening: Passive Case

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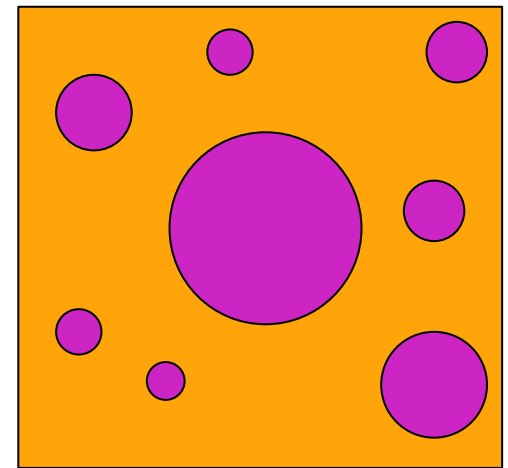
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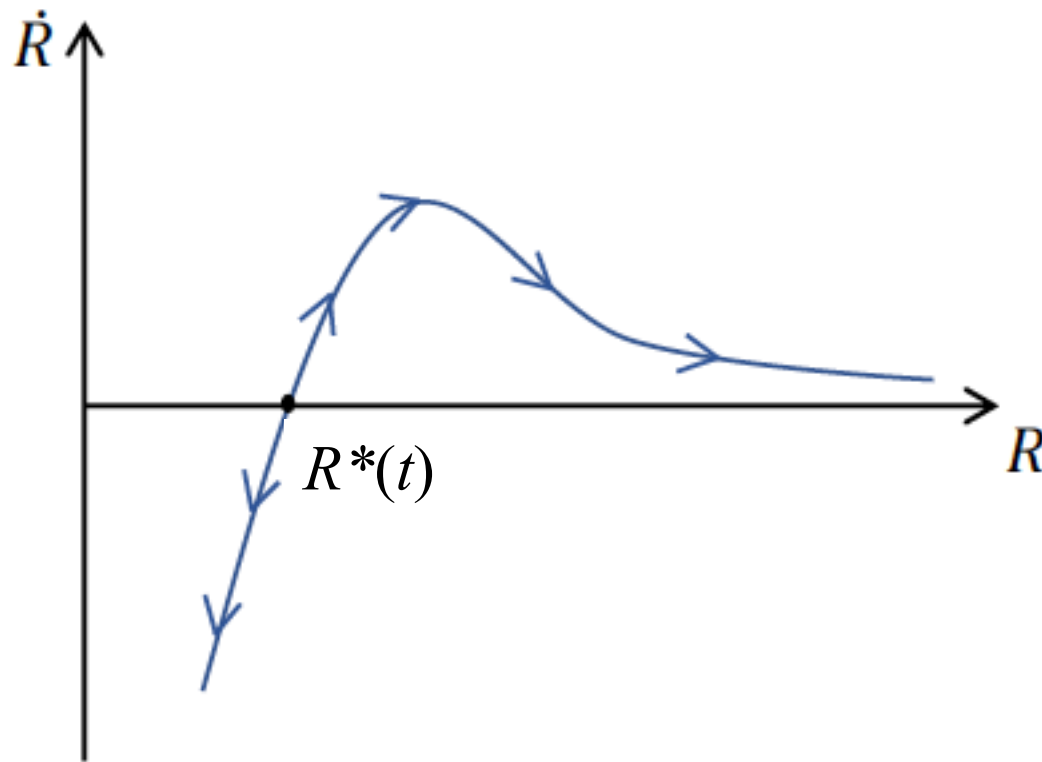
Many drops:  $\mu_\infty$  set by mean size  $R^*$

$$\dot{R} = -\frac{J(R)}{2\phi_B} \propto \frac{\gamma}{R} \left[ \frac{1}{R^*(t)} - \frac{1}{R} \right]$$



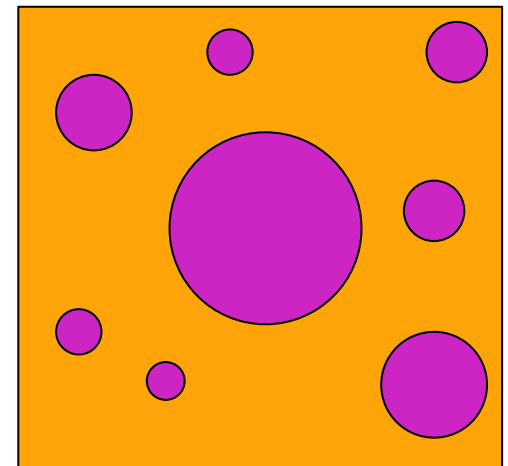
*A. Bray, Adv. in Phys. 1994*

# Ostwald Ripening: Passive Case



- large droplets always grow at cost of small
- phase separation always completes
- noise is irrelevant (T=0 fixed point)

$$\dot{R} = -\frac{J(R)}{2\phi_B} \propto \frac{\gamma}{R} \left[ \frac{1}{R^*(t)} - \frac{1}{R} \right]$$



# Ostwald Ripening: Active Effects

at curved interface  $\mu_I(R) = \frac{\gamma(d-1)}{\phi_B R} + \mu_\zeta(R^+)$

Nonlocal  $\zeta$  contribution:

$$\mu_\zeta(\mathbf{r}) = -\zeta \int d\mathbf{r}' \frac{\nabla \cdot [(\nabla^2 \phi) \nabla \phi - \frac{1}{2} \nabla (\nabla \phi)^2]}{4\pi |\mathbf{r} - \mathbf{r}'|}$$

Discontinuous across curved surface

$$\mu_\zeta(R^+) \propto \frac{\zeta}{R}$$

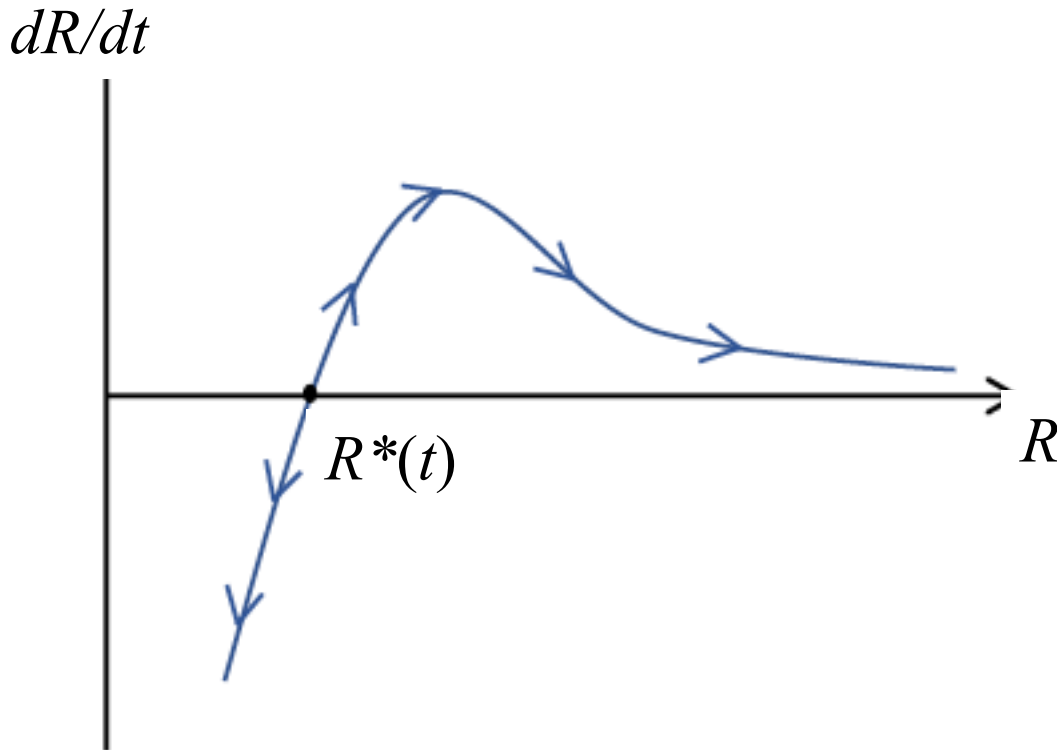
Sign of effective tension set by  $\zeta$

Reverse Ostwald regime emerges at high activity

*E. Tjhung et al, PRX 2018*



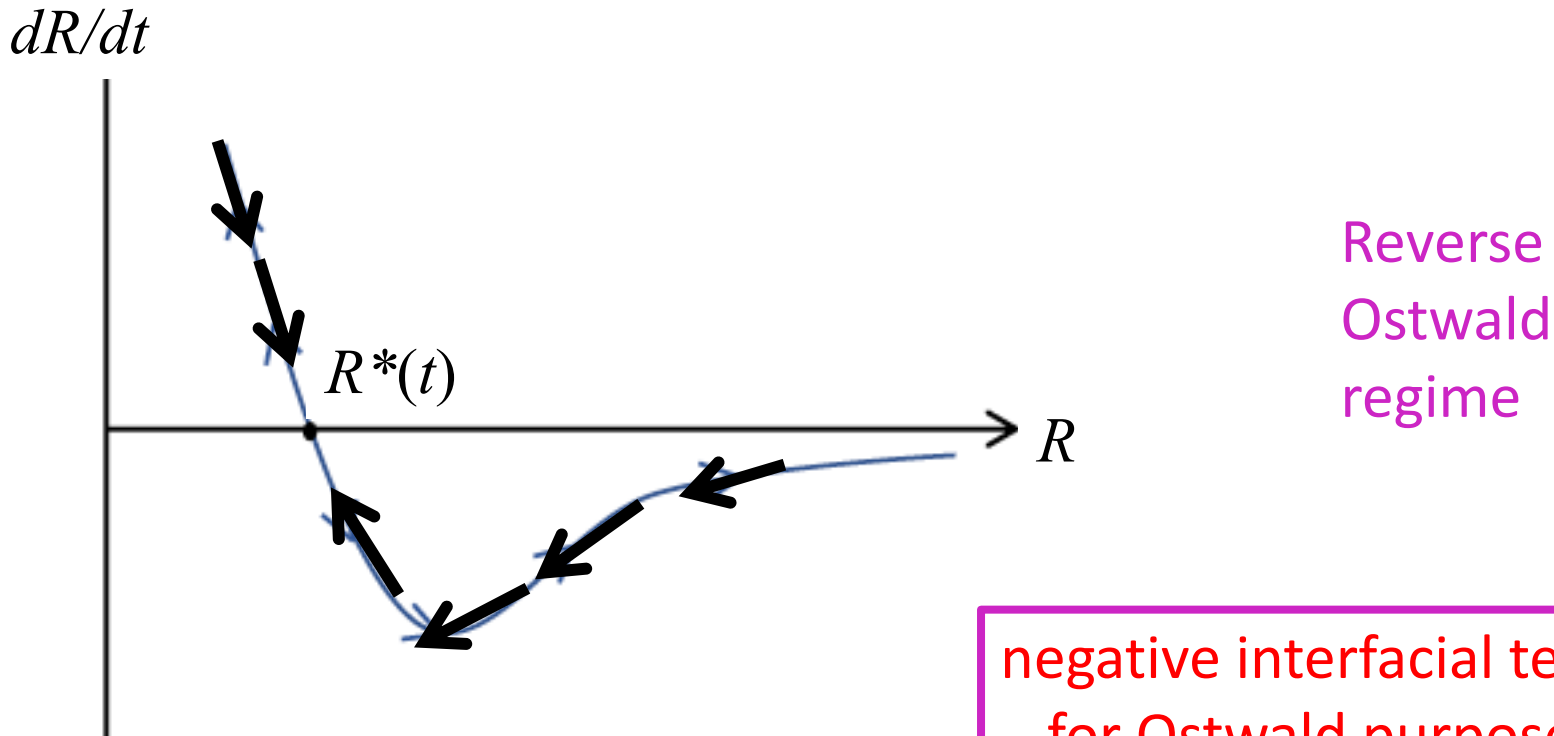
# Ostwald Ripening: Active Effects



Normal  
Ostwald  
regime

*A. Bray, Adv. in Phys. 1994*

# Ostwald Ripening: Active Effects

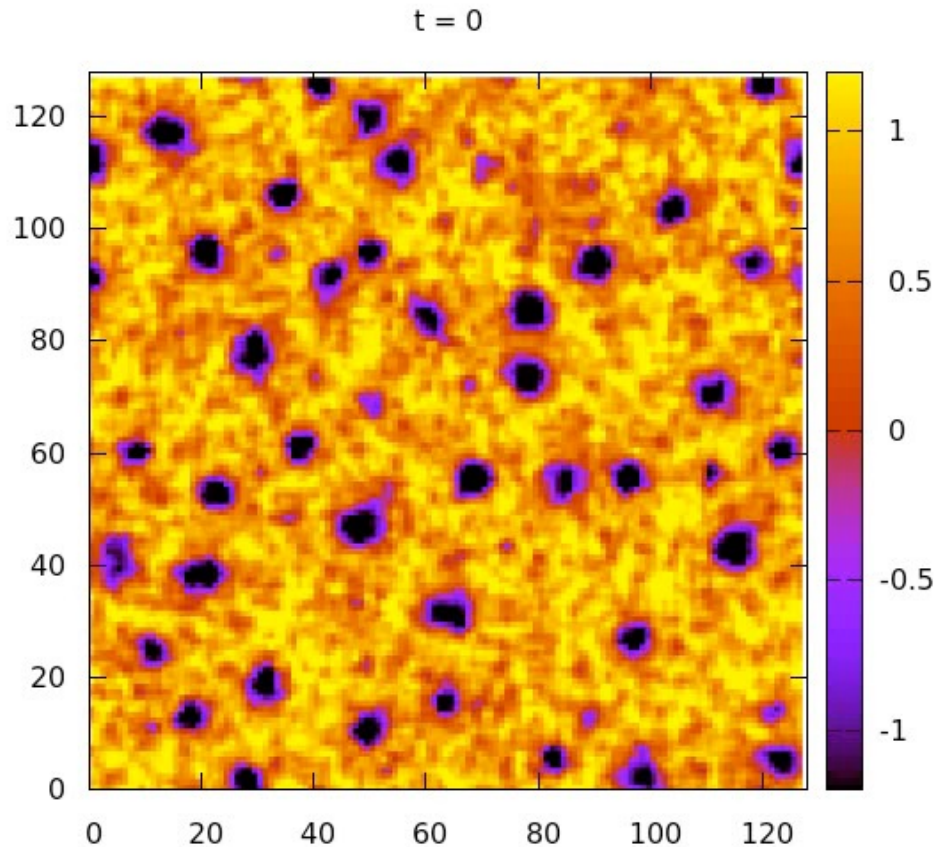


negative interfacial tension  
...for Ostwald purposes only  
single stable fixed point at  $R^*$

# Reverse Ostwald Regime

Without noise: any initial size distribution converges to its own  $R^*$

With noise:



*E. Tjhung et al, PRX 2018*

# Reverse Ostwald Regime

Without noise: any initial size distribution converges to its own  $R^*$

With noise:

- Droplets form via nucleation
- Destroyed by coalescence
- Reverse Ostwald pulls towards uniform size
- Balance of these 3 fixes mean size
- Noise ( $T$ ) no longer irrelevant
- Steady-state life cycle manifestly breaks TRS

*E. Tjhung et al, PRX 2018*

# Reverse Ostwald Regime: Role of Interfacial Tension

The 'Ostwald tension'  $\gamma_o$  is defined via

$$\dot{R} = \frac{J}{2\phi_B} = \frac{\gamma_o}{2\phi_B^2} \left[ \frac{1}{R^*} - \frac{1}{R} \right]$$

Microphase separation arises for  $\gamma_o < 0$

In systems with DB, the Ostwald tension coincides with

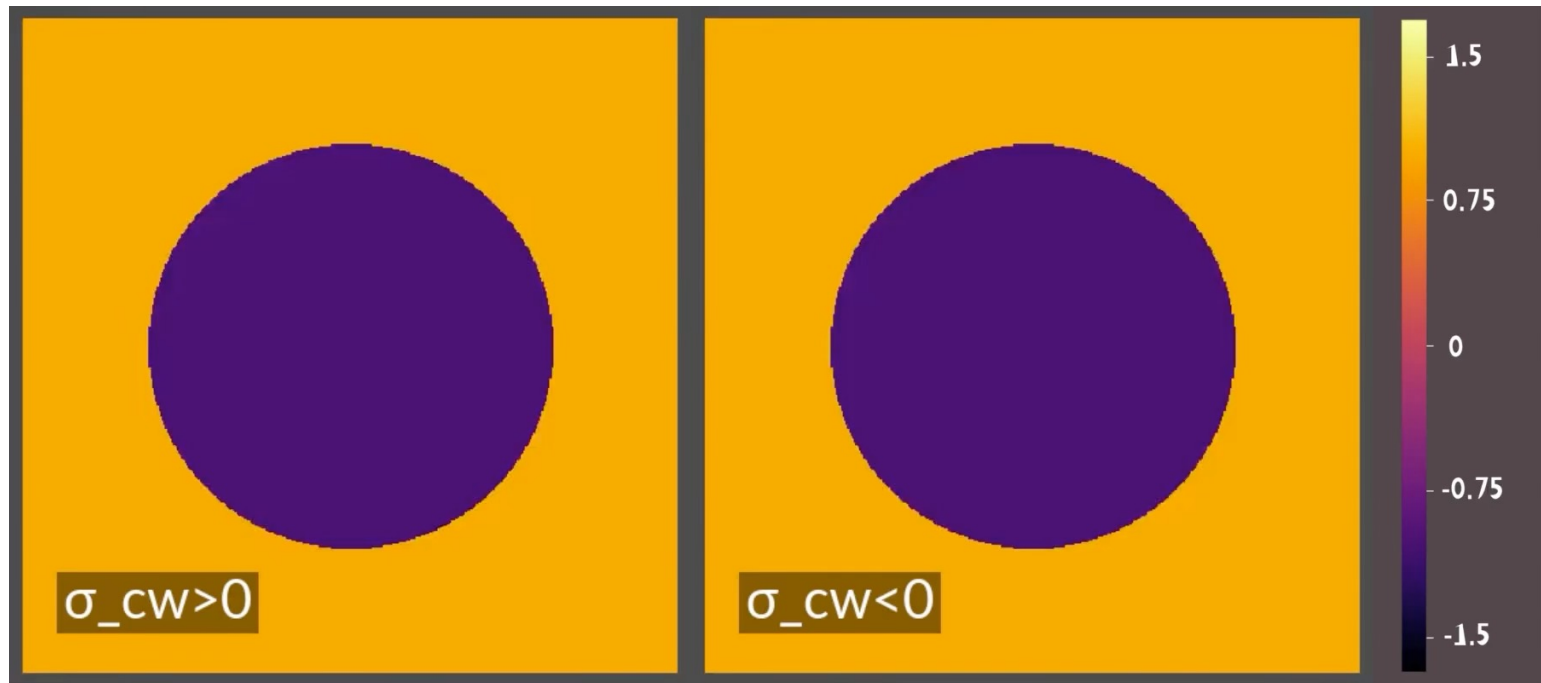
- Capillary tension:  $\gamma < 0 \Rightarrow$  divergent fluctuations  $\langle |h_{\mathbf{q}}|^2 \rangle = \frac{k_B T}{\gamma q^2}$
- Excess free energy per unit area:  $\gamma < 0 \Rightarrow$  phases remix

Without DB, these definitions are inequivalent

# Capillary Interfacial Tension $\gamma_{cw}$

Becomes negative at high enough activity  $\zeta$

Reverse Ostwald:  $\gamma_{cw} > 0 > \gamma_o$       Higher activity:  $0 > \gamma_{cw} > \gamma_o$



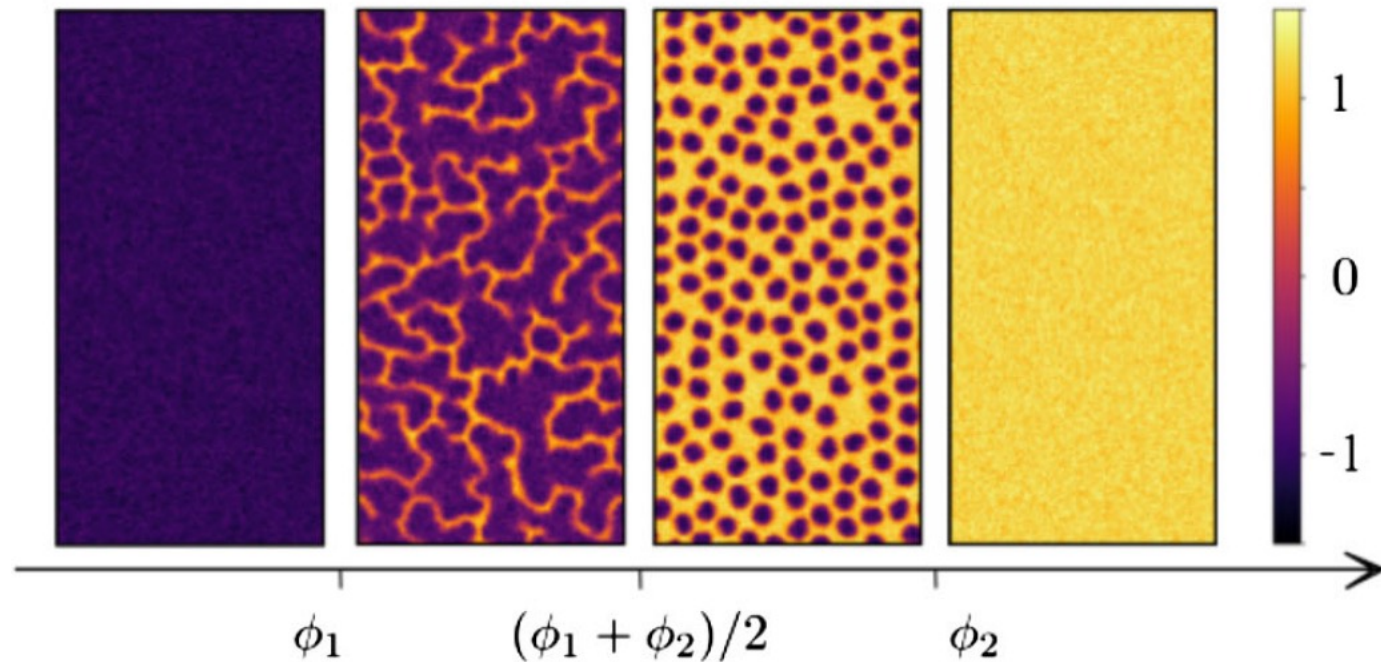
*G Fausti et al, PRL 2021*

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phase diagram:  
now features  
'active foam'



*G Fausti et al, PRL 2021*

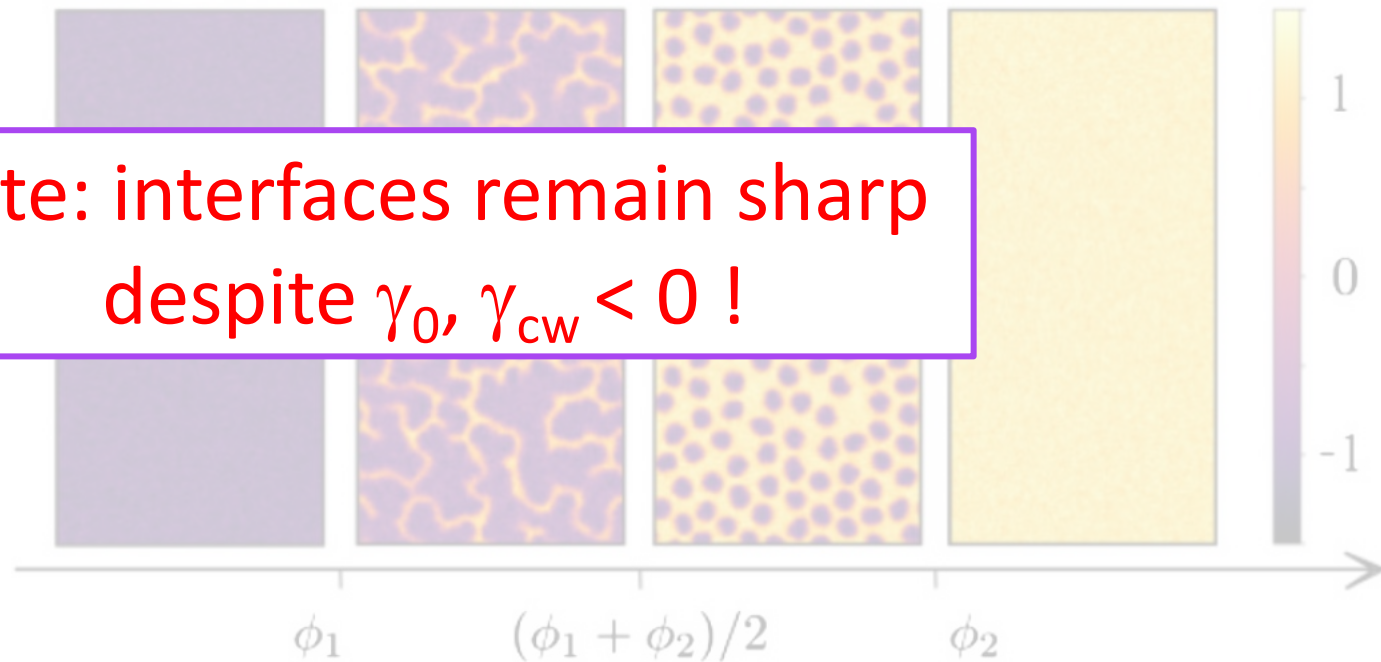
# Capillary Interfacial Tension $\gamma_{cw}$

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phase diagram:  
now feature  
'active foam'

**Note: interfaces remain sharp  
despite  $\gamma_o, \gamma_{cw} < 0$  !**





## Summary: Model B+

### Strengths:

Generically shows anomalous coexistence

Generically admits microphase separation (clusters/bubbles/foam)

Allows explicit calculations of these effects

Allows clean simulations including noise terms

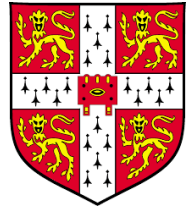
### Weakness:

Agnostic of microscopic mechanisms

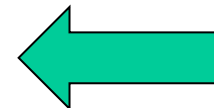
BUT:  $\zeta$ ,  $\lambda$  terms **are** derivable from microscopics in various cases

*E. Tjhung et al, PRX 2018*

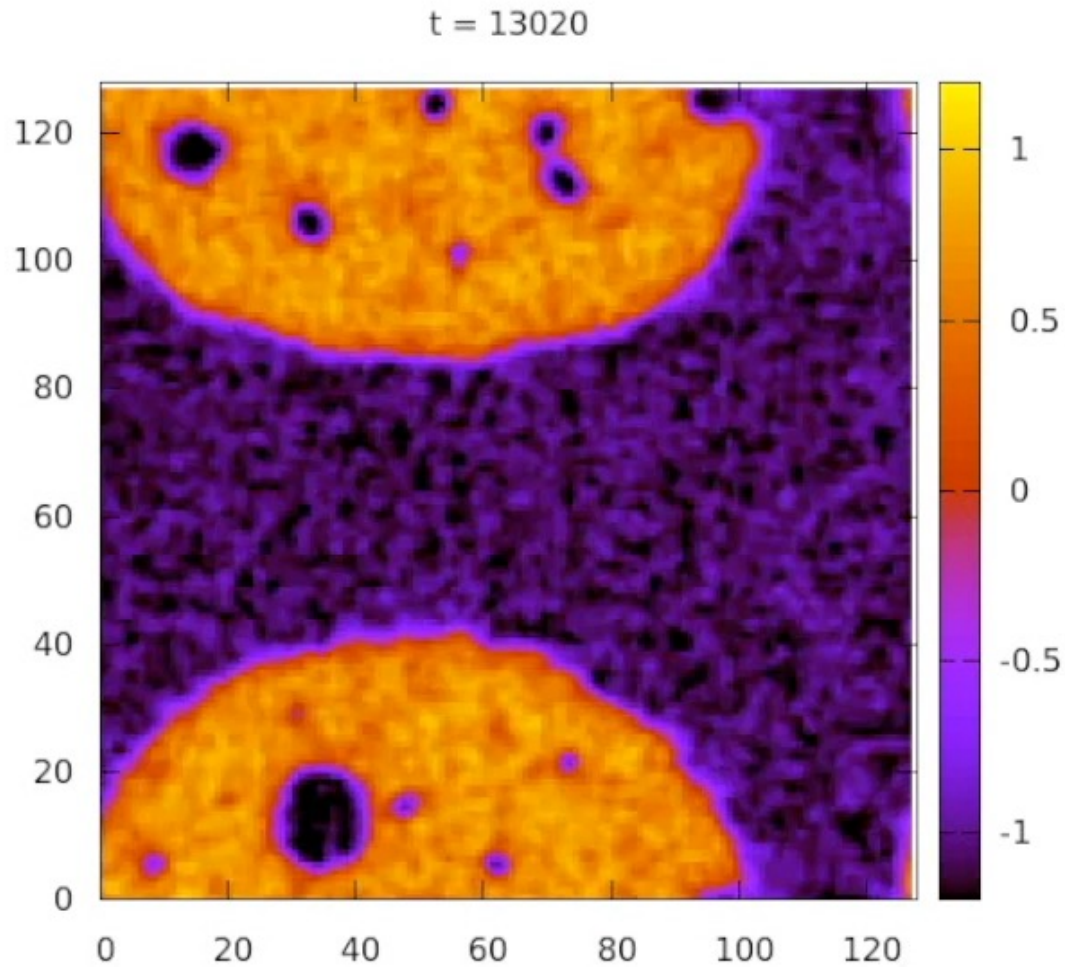
# Active Phase Separation



- Phase Separation in Passive Systems
  - Thermodynamics of the Density Field
  - Diffusive Time Evolution: Model B
  - Phase Separation Kinetics
- Active Particles
  - Broken Time Reversal Symmetry
  - Macroscopic Consequences
- Adapting Model B for Active Matter
  - Phase Coexistence
  - Microphase Separation
  - Stead-State Entropy Production

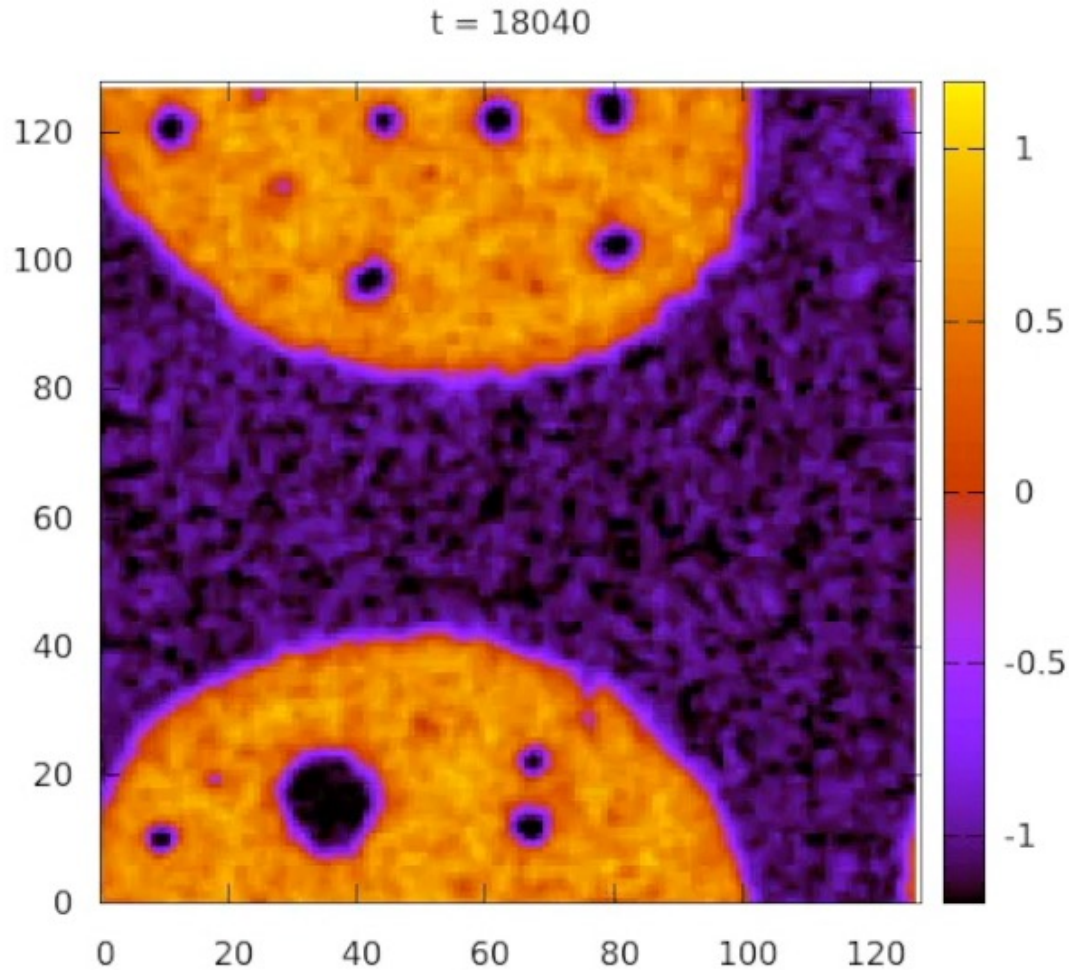


# Active Matter: The Arrow of Life



Active Model B+

# Active Matter: The Arrow of Life



Time-Reversed  
Steady State:  
**not the same**

Active Model B+

# Quantification via Stochastic Thermodynamics

In steady state system attached to heat bath

$$\frac{dS}{dt} = \frac{1}{t_2 - t_1} \log \left( \frac{\text{Prob}(\text{forward sequence})}{\text{Prob}(\text{backward sequence})} \right)$$

$dS/dt$  **directly quantifies** the unlikelihood of reverse processes

*Review: U Seifert, Repts Prog Phys 2012*

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$dS/dt$  **directly quantifies** the unlikelihood of reverse processes

- $dS/dt$  depends on scale of observation
- Field theories are coarse-grained  $\Rightarrow$   
our  $dS/dt$  quantifies “visible” irreversibility only
- = **Informatic** Entropy Production Rate (IEPR)
- IEPR calculable from path integral

## IEPR: Active Model B

$$\nabla^{-1}\dot{\phi} = \mathbf{J} = -\nabla(\mu_E + \mu_{NE}) + (2D)^{1/2}\Lambda$$

$$\ln P[\phi] = -\frac{1}{4D} \int_0^\tau dV dt |\nabla^{-1}\dot{\phi} + \nabla(\mu_E + \mu_{NE})|^2$$

$$\ln(P_f/P_b) = \frac{1}{D} \int_0^\tau dV dt \left( \nabla^{-1}\dot{\phi} \nabla \mu_E + \nabla^{-1}\dot{\phi} \nabla \mu_{NE} \right) \\ - \Delta F$$

steady state:  $\frac{dS}{dt} = \lim_{\tau \rightarrow \infty} \frac{[\text{this}]}{\tau} = -\frac{1}{D} \int \langle \dot{\phi} \mu_{NE} \rangle dV$

## IEPR: Active Model B

$$\frac{dS}{dt} = \int \sigma(\mathbf{r}) dV$$

steady state **local** entropy production rate density

$$\sigma(\mathbf{r}) = \langle \hat{\sigma}(\mathbf{r}) \rangle$$

$$\hat{\sigma}(\mathbf{r}) = -\lambda \dot{\phi} (\nabla \phi)^2 / D$$

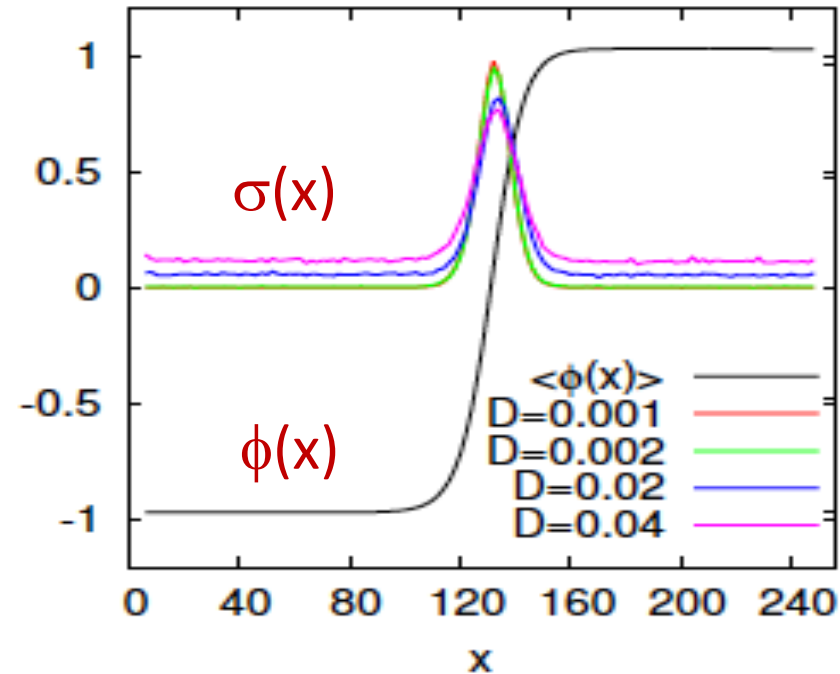
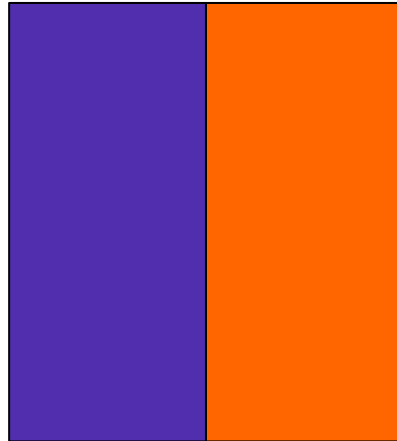
$$\doteq \nabla(a\phi + b\phi^3 - \kappa \nabla^2 \phi + \lambda (\nabla \phi)^2) \cdot \nabla(\lambda (\nabla \phi)^2) / D$$

Global IEPR calculable from stationary measure  $P[\phi(\mathbf{r})]$



# IEPR as Irreversibility Measure

Active Model B

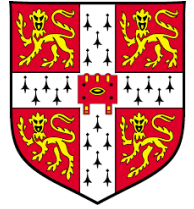


$\sigma \propto D^1$  bulk phases,  $\propto D^0$  at interface

Generic scalings of IEPR density  $\sigma$  for coarse-grained fields:

- $D^{-1}$ : dynamics breaks TRS at deterministic level
- $D^0$ : TRS broken by leading-order fluctuations
- $D^{1,2,\dots}$ : only broken at higher order

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