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Convegno Informale di Fisica Teorica

# Spinodal decomposition of a binary fluid mixture with inhomogeneous temperature

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# Summary

- **Brief introduction**
  - **Dynamic model for binary fluids**
  - **The Lattice Boltzmann Model (LBM)**
  - **Numerical results**
  - **Conclusions**
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# Why this research?

- Innumerable physical phenomena in which the temperature is not constant (boiling processes, droplet motion under thermal gradient <sup>(1)</sup>, phase separation <sup>(2)</sup>)
- Theoretical framework and numerical scheme necessary to study these systems
- Recent studies show the possibility to control the morphology of coarsening domains by exploiting thermal diffusion properties and proper heat sources<sup>(3)</sup>

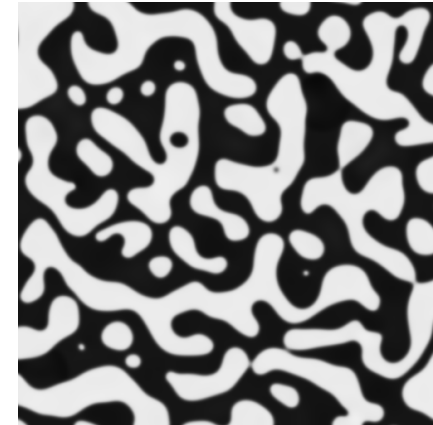
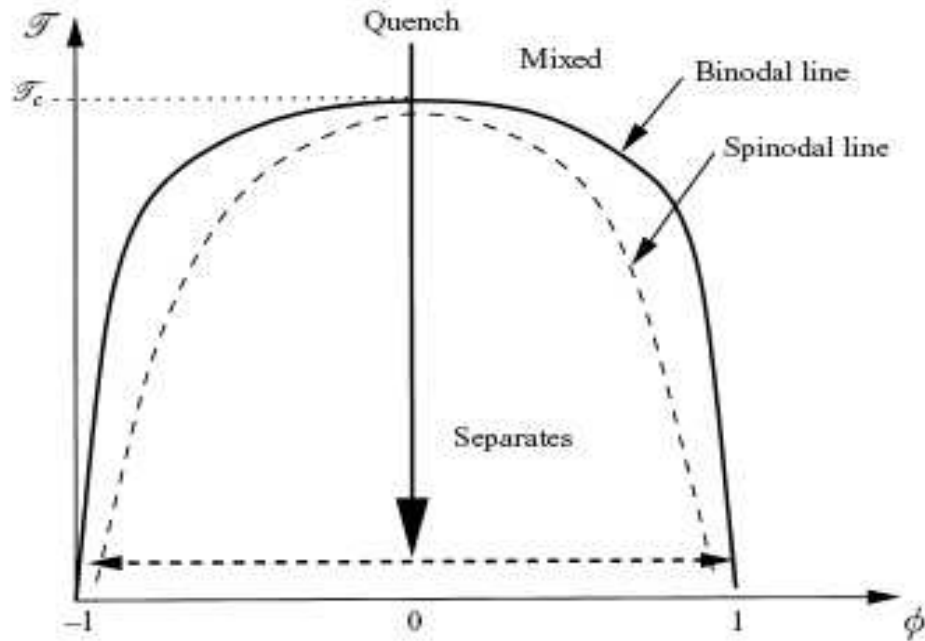
<sup>(1)</sup> *A. Onuki, Phys.Rev.Lett, 94,054501, 2005*

<sup>(2)</sup> *G. Gonnella, A. Lamura, and V. Sofonea, Phys.Rev.E, 76, 036703 2007*

<sup>(3)</sup> *A.P.Krekhov and L.Kramer, Phys.Rev. E, 70, 061801, 2004*

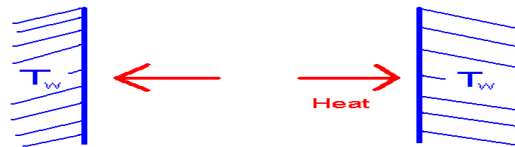
# What about the phase separation?

- Quenching from above the critical point



$R \sim t^\alpha$        $\alpha$  is the “Universal” growth exponent

- How do the morphology of domains and the bulk temperature evolve?



# Dynamic equations for binary fluids

$$\begin{aligned}\partial_t n &= -\partial_\alpha (n u_\alpha) \\ \partial_t \varphi &= -\partial_\alpha (\varphi u_\alpha) - 2\partial_\alpha \mathcal{J}_\alpha^D \\ n(\partial_t u_\alpha + u_\beta \partial_\beta u_\alpha) &= -\partial_\beta (\Pi_{\alpha\beta} - \sigma_{\alpha\beta}) \\ \partial_t e &= -\partial_\alpha (e u_\alpha) - (\Pi_{\alpha\beta} - \sigma_{\alpha\beta}) \partial_\alpha u_\beta - \partial_\alpha \mathcal{J}_\alpha^Q\end{aligned}$$

$$\begin{aligned}\vec{u}(\vec{r}, t) \\ n(\vec{r}, t) \\ \varphi(\vec{r}, t) \\ e(\vec{r}, t)\end{aligned}$$



**Velocity Field**  
**Total Density**  
**Concentration Field**  
**Total Internal Energy Density**

$$\sigma_{\alpha\beta} = \eta(\partial_\alpha u_\beta + \partial_\beta u_\alpha - \partial_\gamma u_\gamma 2\delta_{\alpha\beta}/d) + \zeta \partial_\gamma u_\gamma \delta_{\alpha\beta} \rightarrow \text{Dissipative Stress Tensor}$$

$\Pi_{\alpha\beta} \rightarrow$  **Reversible Stress Tensor**

$\vec{\mathcal{J}}^D, \vec{\mathcal{J}}^Q \rightarrow$  **Diffusion and Heat Current**

**What are the expressions for  $\Pi_{\alpha\beta}$  and  $\vec{\mathcal{J}}^D, \vec{\mathcal{J}}^Q$  ?**

# Pressure tensor and currents

$$\Pi_{ij} = \left( p - M\varphi\nabla^2\varphi - M\frac{|\vec{\nabla}\varphi|^2}{2} - T\varphi\vec{\nabla}\varphi \cdot \vec{\nabla}\left(\frac{M}{T}\right) \right) \delta_{ij} + M\partial_i\varphi\partial_j\varphi$$

$$p = -\psi + n\partial\psi/\partial n + \varphi\partial\psi/\partial\varphi$$

$$\psi = AnT + \left[ \frac{n-\varphi}{2} \ln\left(\frac{n-\varphi}{2}\right) + \frac{n+\varphi}{2} \ln\left(\frac{n+\varphi}{2}\right) \right] + \frac{\lambda n}{4} \left(1 - \frac{\varphi^2}{n^2}\right)$$

$$T_c = \lambda/2 \quad \varphi_{\pm} = \pm\sqrt{3n^2(\lambda/2k_B T - 1)}$$

$$\mu = -T \left[ \frac{\partial(ns)}{\partial\varphi} - \frac{1}{T} \frac{\partial e_B}{\partial\varphi} + \vec{\nabla} \cdot \left( \frac{M}{T} \vec{\nabla}\varphi \right) \right]$$

Chemical Potential

$$\vec{J}^D = -\mathcal{L}_{11} \vec{\nabla}\left(\frac{\mu}{T}\right) + \mathcal{L}_{12} \vec{\nabla}\left(\frac{1}{T}\right)$$

$$\mathcal{L}_{11} > 0, \mathcal{L}_{22} > 0, \mathcal{L}_{11}\mathcal{L}_{22} - \mathcal{L}_{12}\mathcal{L}_{21} > 0$$

$$\vec{J}^Q = -\mathcal{L}_{21} \vec{\nabla}\left(\frac{\mu}{T}\right) + \mathcal{L}_{22} \vec{\nabla}\left(\frac{1}{T}\right)$$

$$\mathcal{L}_{11} = Tl_{11}, \mathcal{L}_{22} = T^2l_{22} \quad l_{11}, l_{22} \text{ constant}$$

See de Groot and Mazur

In an homogeneous system  $l_{22}$  is the heat conductivity

*G. Gonnella, A. Lamura, A. Piscitelli, J.P.A, 41,105001,2008*

*A. Onuki, Phys.Rev.Lett, 94,054501, 2005*

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# Lattice Boltzmann Model (LBM)

## What is it?

- An algorithm to solve numerically the fluid equations

## Where is LBM used?

- Multi-component fluid mixtures
- Non-ideal fluids
- Complex fluids (Liquid Crystal)
- Microfluidics

## Advantages 😊

- Simple implementation
- Parallelization
- Boundary conditions

## Disadvantages ☹️

- Spurious velocities
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# LBM with a forcing term

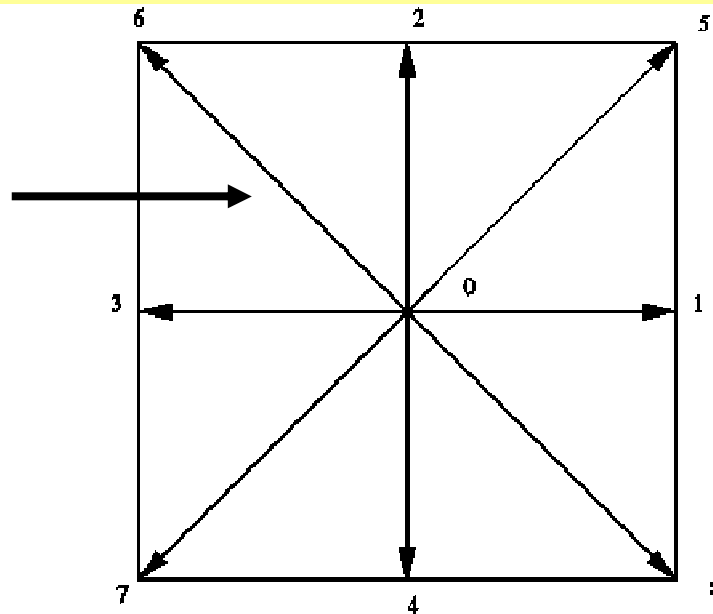
## Evolution Equation

$$f_i(\mathbf{r} + \mathbf{e}_i \Delta t_{LB}, t + \Delta t_{LB}) - f_i(\mathbf{r}, t) = -\frac{\Delta t_{LB}}{\tau} [f_i(\mathbf{r}, t) - f_i^{eq}(\mathbf{r}, t)] + \Delta t_{LB} F_i$$

## Density and Momentum

$$n = \sum_i f_i \quad n\mathbf{u} = \sum_i f_i \mathbf{e}_i + \frac{1}{2} \mathbf{F} \Delta t_{LB},$$

$e_i$



$D_2Q_9$  Model



# LBM with a forcing term (2)

## Equilibrium distribution function

$$f_i^{eq}(\mathbf{r}, t) = \omega_i n \left[ 1 + \frac{\mathbf{e}_i \cdot \mathbf{u}}{c_s^2} + \frac{\mathbf{u}\mathbf{u} : (\mathbf{e}_i \mathbf{e}_i - c_s^2 \mathbf{I})}{2c_s^4} \right]$$

$$\omega_0 = 4/9, \quad \omega_i = 1/9 \quad i = 1, 4 \quad \omega_i = 1/36 \quad i = 5, 8$$

## Forcing term

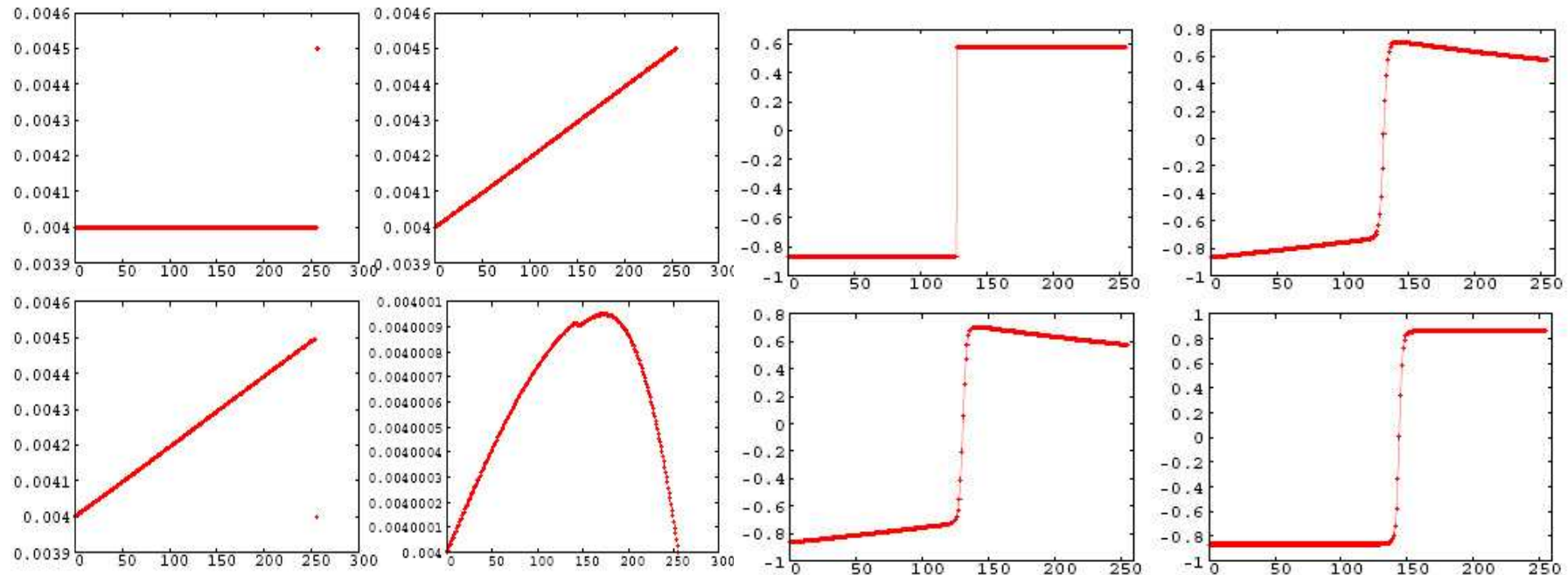
$$F_i = \omega_i \left[ A + \frac{\mathbf{B} \cdot \mathbf{e}_i}{c_s^2} + \frac{\mathbf{C} : (\mathbf{e}_i \mathbf{e}_i - c_s^2 \mathbf{I})}{2c_s^4} \right]$$

- The continuity and the Navier-Stokes equations are recovered in the continuum limit
- The convection-diffusion and the energy equations are implemented by using a finite-difference scheme

# Interface profile (with hydrodynamics)

Temperature

Concentration



The left wall is kept at  $T=0.8$  while the right wall is at  $T=0.9$ .

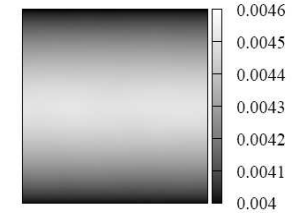
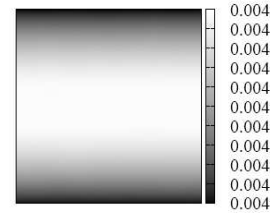
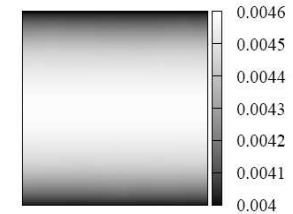
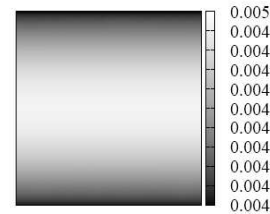
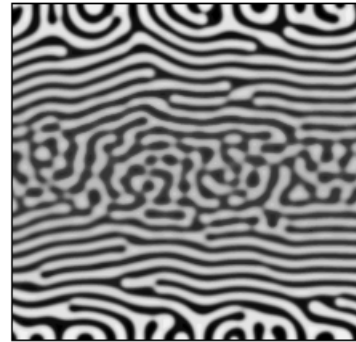
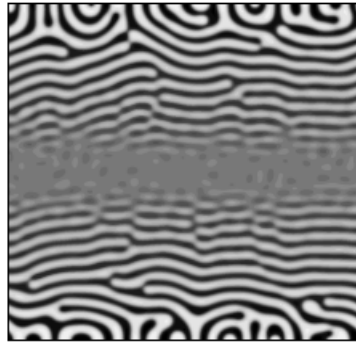
The system equilibrates with the right wall which is then set to the temperature of the left wall. (The temperature is expressed in  $T_c$  units, with  $T_c=5 \cdot 10^{-3}$ )

$$\psi = AnT + \left[ \frac{n - \varphi}{2} \ln\left(\frac{n - \varphi}{2}\right) + \frac{n + \varphi}{2} \ln\left(\frac{n + \varphi}{2}\right) \right] + \frac{\lambda n}{4} \left(1 - \frac{\varphi^2}{n^2}\right)$$

$$T_c = \lambda/2$$

$$\varphi_{\pm} = \pm \sqrt{3n^2(\lambda/2k_B T - 1)}$$

# Phase separation with no hydrodynamics



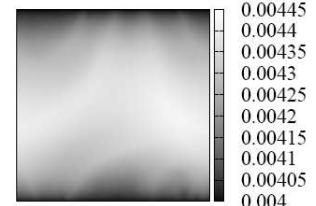
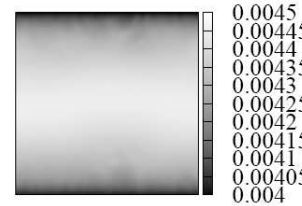
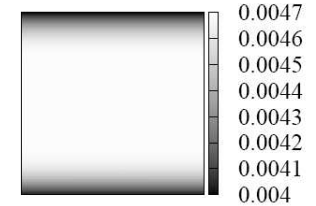
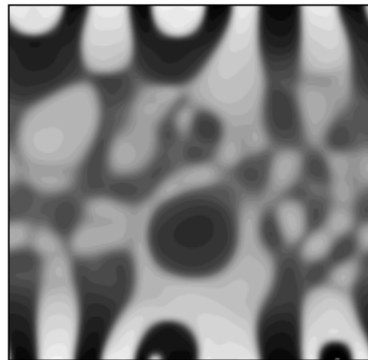
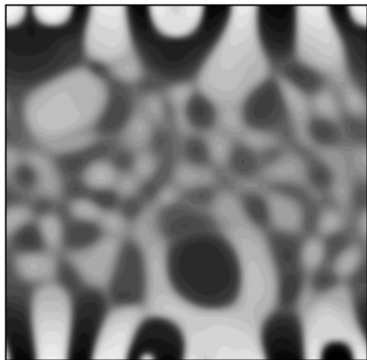
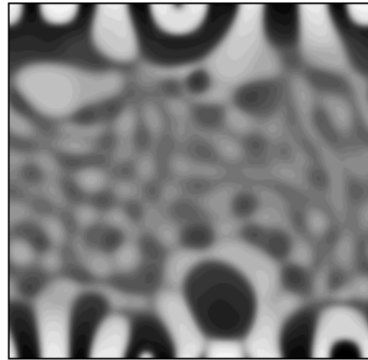
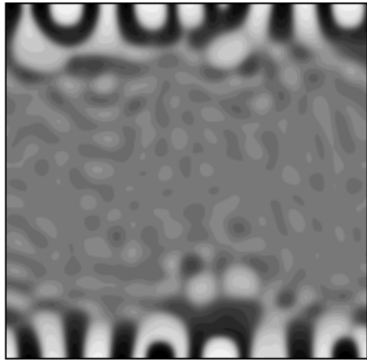
Concentration field for a system of size  $512 \times 512$  with  $T_u = 4 \cdot 10^{-3}$ ,  $T_l = 4 \cdot 10^{-3}$  and  $T_c = 5 \cdot 10^{-3}$  at times  $t = 75 \cdot 10^4, 225 \cdot 10^4, 375 \cdot 10^4, 500 \cdot 10^4$ . The mixture composition is symmetric (50% species A and 50% species B). The heat diffusion constant is  $10^{-2}$ .

Temperature field for a system of size  $512 \times 512$  with  $T_u = 4 \cdot 10^{-3}$ ,  $T_l = 4 \cdot 10^{-3}$  and  $T_c = 5 \cdot 10^{-3}$  at times  $t = 75 \cdot 10^4, 225 \cdot 10^4, 375 \cdot 10^4, 500 \cdot 10^4$ . The mixture composition is symmetric (50% species A and 50% species B). The heat diffusion constant is  $10^{-2}$ .

- Neutral wetting is imposed on the boundaries
- A lamellar pattern is observed

$T=0.8$  at both walls  
(in  $T_c$  units)

# Phase separation with hydrodynamics



Temperature field for a system of size  $256 \times 256$  with  $T_u = 4 \cdot 10^{-3}$ ,  $T_l = 4 \cdot 10^{-3}$  and  $T_c = 5 \cdot 10^{-3}$  at times  $t = 1 \cdot 10^3, 1 \cdot 10^5, 8 \cdot 10^5, 34 \cdot 10^5$ . The mixture composition is symmetric (50% species A and 50% species B). The heat diffusion constant is  $10^{-2}$ .

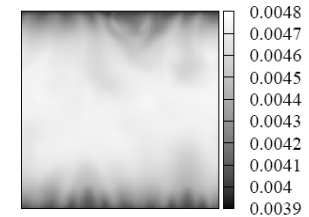
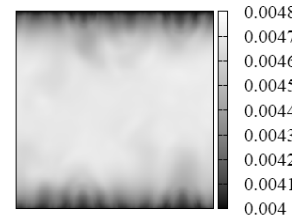
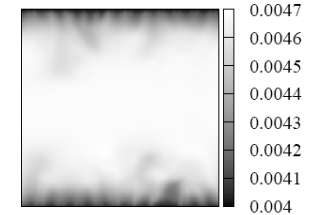
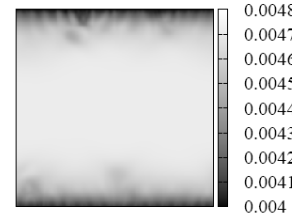
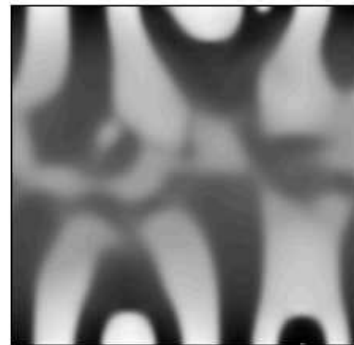
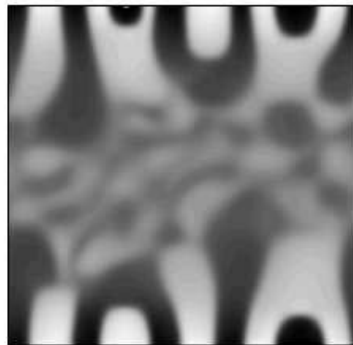
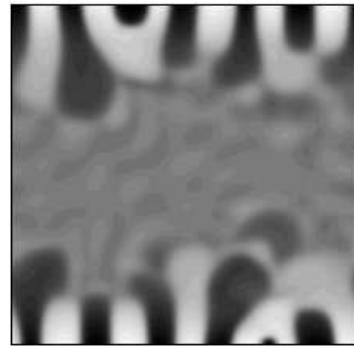
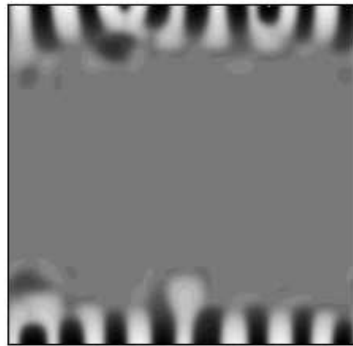
Concentration field for a system of size  $256 \times 256$  with  $T_u = 4 \cdot 10^{-3}$ ,  $T_l = 4 \cdot 10^{-3}$  and  $T_c = 5 \cdot 10^{-3}$  at times  $t = 8 \cdot 10^5, 11 \cdot 10^5, 13 \cdot 10^5, 15 \cdot 10^5$ . The mixture composition is symmetric (50% species A and 50% species B). The heat diffusion constant is  $10^{-2}$ .

$$\tau = 1$$

Phase separation seems to occur with two scales

$T=0.8$  at both walls (in  $T_c$  units)

# Phase separation with hydrodynamics (2)



Concentration field for a system of size  $256 \times 256$  with  $T_u = 4 \cdot 10^{-3}$ ,  $T_l = 4 \cdot 10^{-3}$  and  $T_c = 5 \cdot 10^{-3}$  at times  $t = 15 \cdot 10^5, 30 \cdot 10^5, 40 \cdot 10^5, 50 \cdot 10^5$ . The mixture composition is symmetric (50% species A, 50% species B). The heat diffusion constant is  $10^{-3}$ .

Temperature field for a system of size  $256 \times 256$  with  $T_u = 4 \cdot 10^{-3}$ ,  $T_l = 4 \cdot 10^{-3}$ ,  $T_c = 5 \cdot 10^{-3}$  at times  $t = 15 \cdot 10^5, 30 \cdot 10^5, 40 \cdot 10^5, 50 \cdot 10^5$ . The mixture composition is symmetric (50% species A, 50% species B). The heat diffusion constant is  $10^{-3}$ .

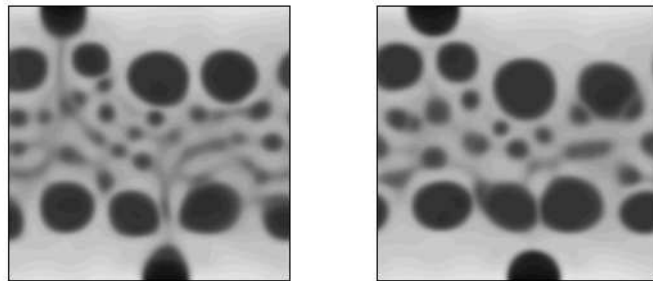
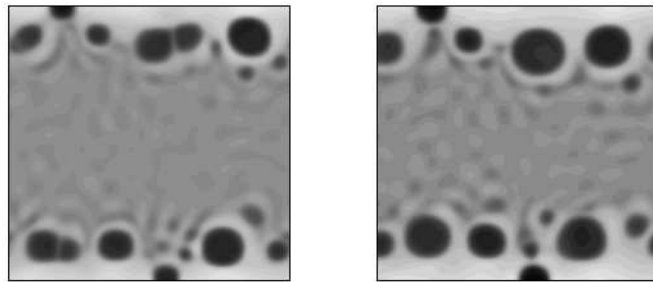
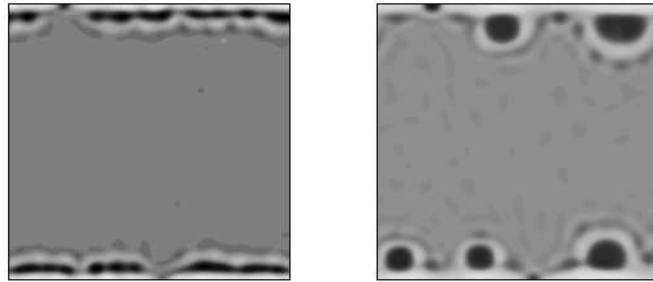
$$\tau = 1$$

- Phase separation seems to occur with two length scales
- Thermal diffusion is ten times lower than the previous case

$T=0.8$  at both walls  
(in  $T_c$  units)

# Phase separation with asymmetric composition 55%-45%

With hydrodynamics

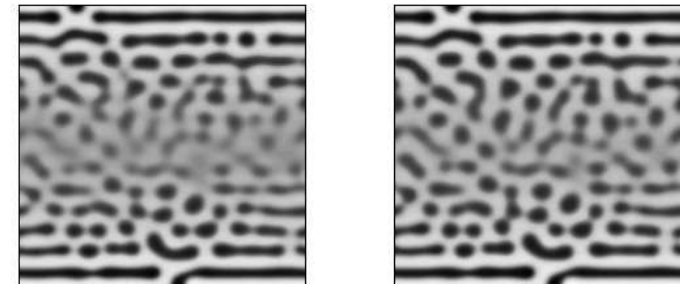
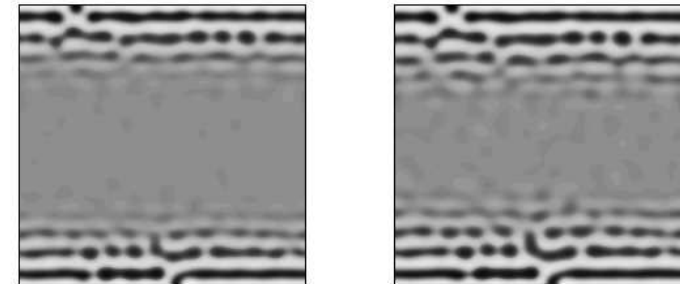
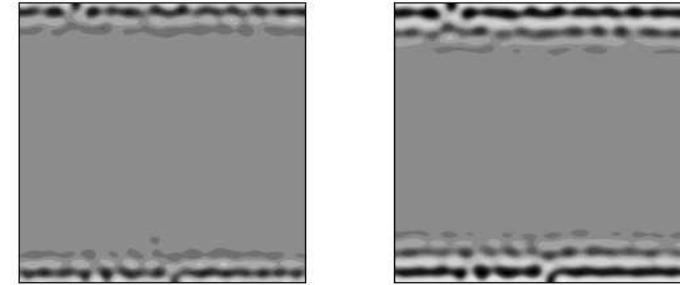


$T=0.8$  at both walls  
(in  $T_c$  units)

Lamellar pattern without hydrodynamics

$\tau = 1$

Without hydrodynamics



Concentration field for a system of size  $256 \times 256$  with  $T_u = 4 \cdot 10^{-3}$ ,  $T_l = 4 \cdot 10^{-3}$  and  $T_c = 5 \cdot 10^{-3}$  at times  $t = 4 \cdot 10^5, 6 \cdot 10^5, 9 \cdot 10^5, 11 \cdot 10^5, 16 \cdot 10^5, 18 \cdot 10^5$ . The mixture composition is asymmetric (55% species A, 45% species B). The heat diffusion constant is  $10^{-2}$ .

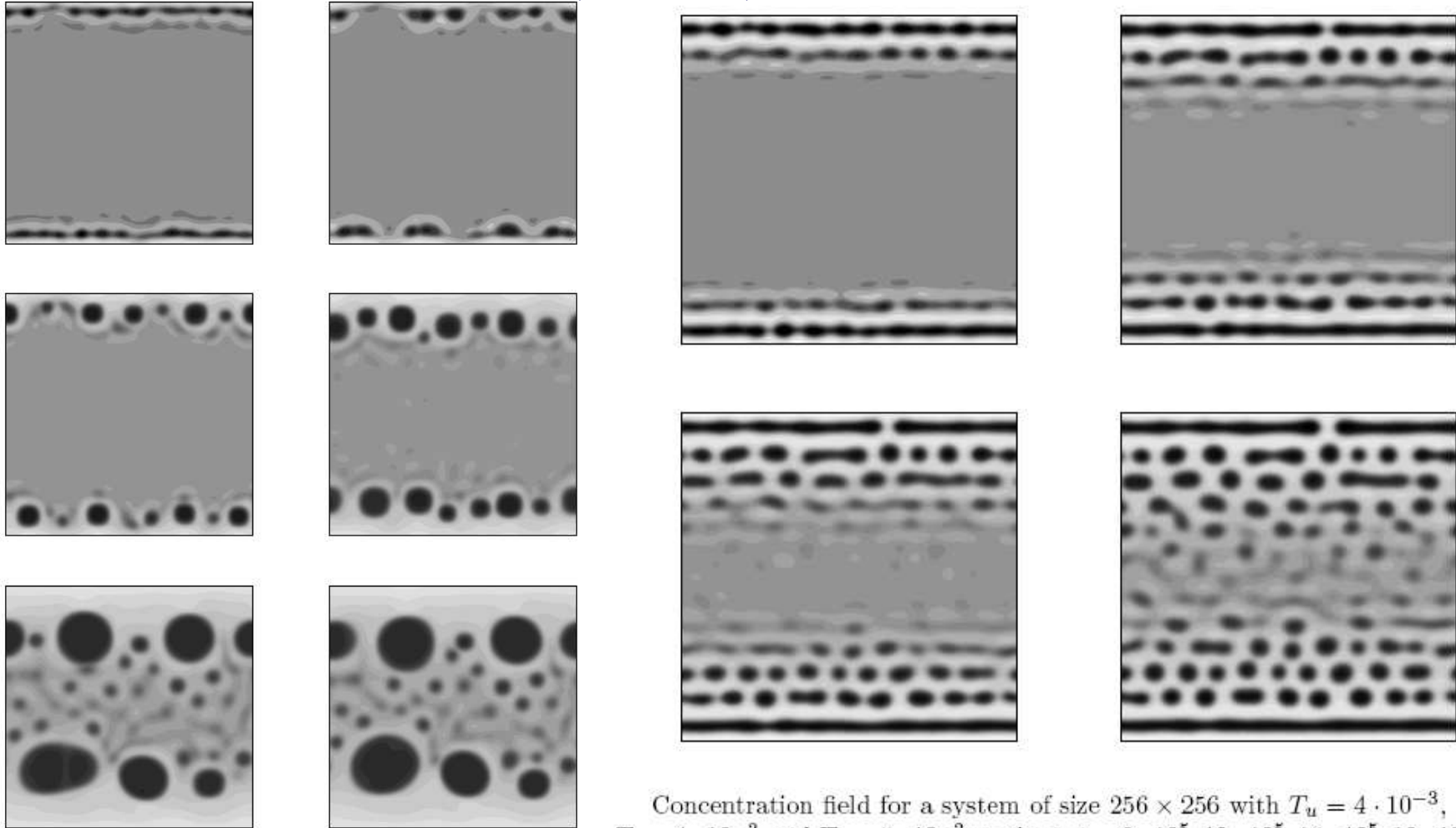
Concentration field for a system of size  $256 \times 256$   $T_u = 4 \cdot 10^{-3}$ ,  $T_l = 4 \cdot 10^{-3}$  and  $T_c = 5 \cdot 10^{-3}$  at times  $t = 4 \cdot 10^5, 6 \cdot 10^5, 9 \cdot 10^5, 11 \cdot 10^5, 16 \cdot 10^5, 18 \cdot 10^5$ . The mixture composition is asymmetric (55% species A, 45% species B). The heat diffusion constant is  $10^{-2}$ .

# Phase separation with asymmetric composition 62%-38%

With hydrodynamics

$T=0.8$  at both walls  
(in  $T_c$  units)

Without hydrodynamics



Concentration field for a system of size  $256 \times 256$  with  $T_u = 4 \cdot 10^{-3}$ ,  
 $T_l = 4 \cdot 10^{-3}$  and  $T_c = 5 \cdot 10^{-3}$  at times  
 $t = 4 \cdot 10^5, 5 \cdot 10^5, 7 \cdot 10^5, 10 \cdot 10^5, 18 \cdot 10^5, 19 \cdot 10^5$ . The mixture composition  
 is asymmetric (62% species A, 38% species B). The heat diffusion constant  
 is  $10^{-2}$ .

Concentration field for a system of size  $256 \times 256$  with  $T_u = 4 \cdot 10^{-3}$ ,  
 $T_l = 4 \cdot 10^{-3}$  and  $T_c = 5 \cdot 10^{-3}$  at times  $t = 8 \cdot 10^5, 12 \cdot 10^5, 15 \cdot 10^5, 19 \cdot 10^5$ .  
 The mixture composition is asymmetric (62% species A, 38% species B).  
 The heat diffusion constant is  $10^{-2}$ .

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# Conclusions

- At high viscosities lamellar pattern can be observed
  - At low viscosities two scales can be observed during the phase separation with symmetric composition
  - At low viscosities a more rounded shape of the domains is favoured by hydrodynamics during the phase separation with asymmetric composition
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END

**Thank you for your attention**

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