Active dumbbells

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Work in collaboration with

D. Loi & S. Mossa (Grenoble, France, 2007-2009) and

G. Gonnella, P. Di Gregorio, G.-L. Laghezza, A. Lamura, A. Mossa & A. Suma

(Bari & Trieste, Italia, 2013-2015)

Palma de Mallorca, España, 2017

Plan

5 lectures & 2 exercise sessions

1. Introduction

2. Active Brownian dumbbells

3. Effective temperatures

4. Two-dimensional equilibrium phases

5. Two-dimensional collective behaviour of active systems

Fifth lecture

Plan

5 lectures & 2 exercise sessions

1. Introduction
2. Active Brownian dumbbells
3. Effective temperatures
4. Two-dimensional equilibrium phases
5. **Two-dimensional collective behaviour of active systems**

Results & questions

- Brand new Bernard & Krauth two step transition scenario

  Liquid (1st order) Hexatic (BKTHNY) Solid

  confirmed for hard and soft passive disks

- Passive molecules ?
- Active disks and molecules ?

- Mobility induced phase transition for purely repulsive interactions *vs.*

  just an extension of the Bernard & Krauth passive system scenario

Plan

1. The result : new phase diagram
2. The interacting dumbbells model
3. Passive case
4. Active case
5. Discussion of

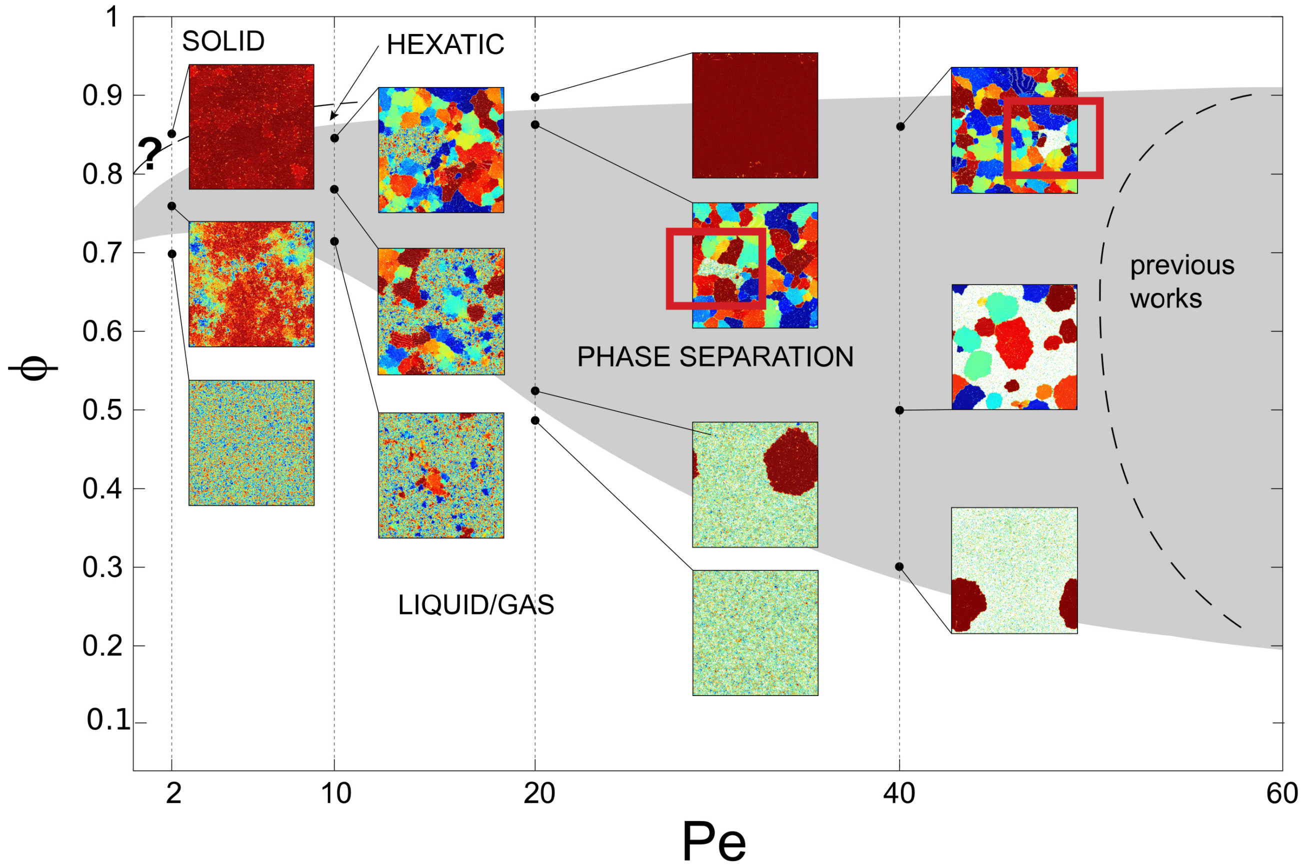
   Mobility induced phase transition for purely repulsive interactions

   vs.

   just an extension of the Bernard & Krauth passive system scenario

Phase diagram

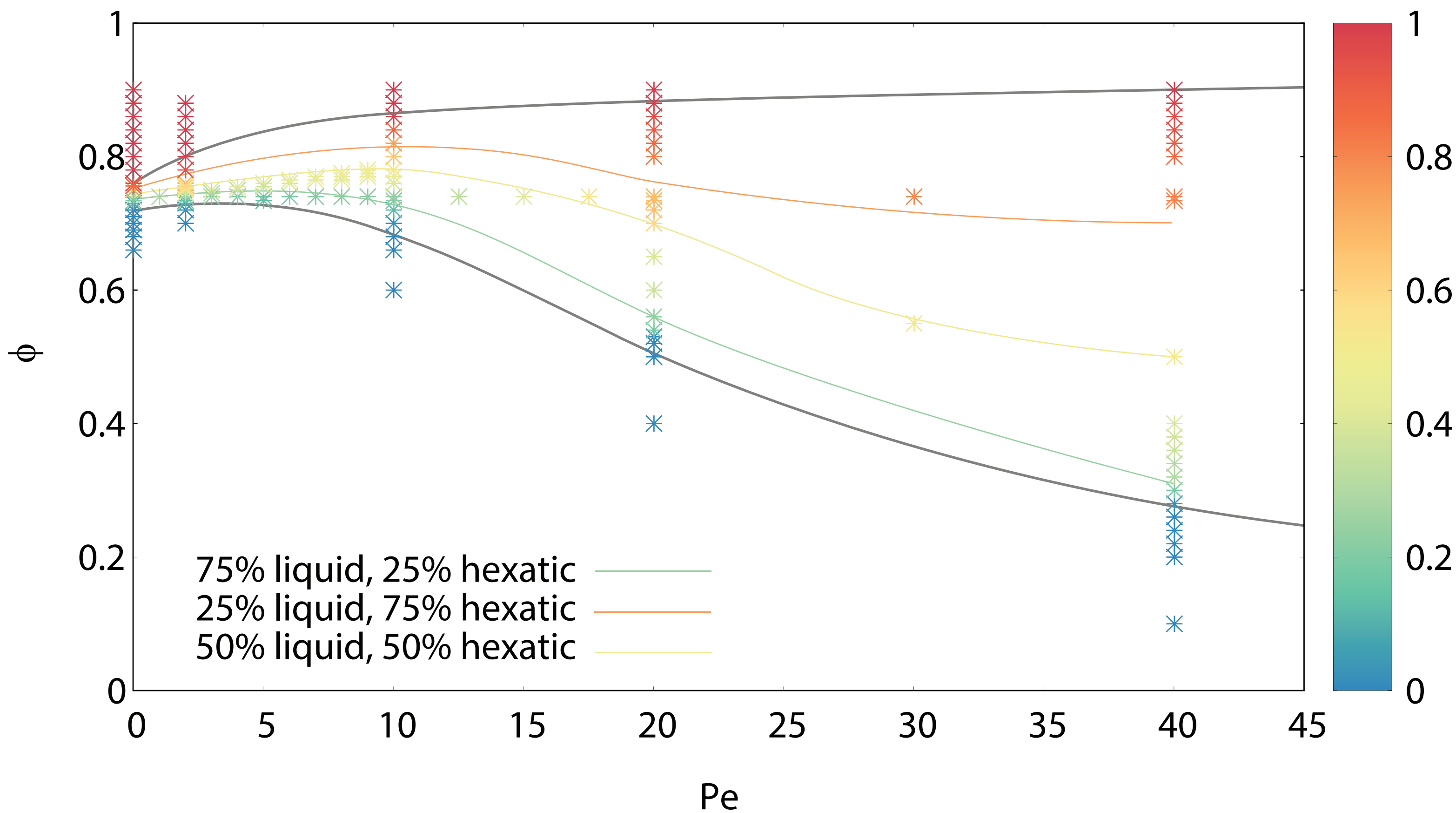
Active dumbbells



$T = 0.05$

Coexistence region

& lines of constant proportion



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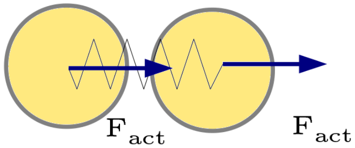
Interacting active dumbbells

Many-body interacting system



Two spherical atoms with diameter $\sigma_{\rm d}$ and mass $m_{\rm d}$

Massless spring modelled by a finite extensible non-linear elastic (fene) force between the beads i and j belonging to the same dumbbell, $\mathbf{F}_{\rm fene} = -\dfrac{k(\boldsymbol{r}_i - \boldsymbol{r}_j)}{1 - r_{ij}^2/r_0^2}$,

with an additional repulsive contribution (WCA) to avoid colloidal overlapping.

Polar active force along the main molecular axis $\boxed{\mathbf{F}_{\rm act} = F_{\rm act}\ \hat{\mathbf{n}}}$

Purely repulsive interaction between colloids in different molecules.

Langevin modelling of the interaction with the embedding fluid:

isotropic viscous forces, $-\gamma \boldsymbol{v}_i$, and independent noises, $\boldsymbol{\eta}_i$, on the beads.

Particles with shape

e.g., a diatomic molecule or a dumbbell

$$\begin{aligned} m_{\mathrm{d}}\ddot{\boldsymbol{r}}_i(t) &= -\gamma\dot{\boldsymbol{r}}_i(t) + \mathbf{F}_{\mathrm{pot_i}}(\boldsymbol{r}_i, \boldsymbol{r}_{i+1}) + \boldsymbol{\eta}_i \\ m_{\mathrm{d}}\ddot{\boldsymbol{r}}_{i+1}(t) &= -\gamma\dot{\boldsymbol{r}}_{i+1}(t) + \mathbf{F}_{\mathrm{pot_{i+1}}}(\boldsymbol{r}_i, \boldsymbol{r}_{i+1}) + \boldsymbol{\eta}_{i+1} \end{aligned}$$

with $\mathbf{F}_{\mathrm{pot}} = \mathbf{F}_{\mathrm{wca}} + \mathbf{F}_{\mathrm{fene}}$, $V = V_{\mathrm{wca}} + V_{\mathrm{fene}}$ and

$$V_{\mathrm{wca}}(\boldsymbol{r}_i, \boldsymbol{r}_{i+1}) = \begin{cases} V_{LJ}(r_{i,i+1}) - V_{LJ}(r_c) & r < r_c \\ 0 & r > r_c \end{cases}$$

$$V_{LJ}(r) = 4\epsilon\left[\left(\frac{\sigma}{r}\right)^{2n} - \left(\frac{\sigma}{r}\right)^{n}\right] \qquad r_c = 2^{1/n}\sigma_{\mathrm{d}} = \sigma$$

Active dumbbell

Control parameters

Number of dumbbells N and box volume S in two dimensions:

packing fraction $\phi = \dfrac{\pi \sigma_{\rm d}^2 N}{2S}$

Energy scales:

Active work $2\sigma_{\rm d}\, F_{\rm act}$

thermal energy $k_B T$

Péclet number $\text{Pe} = \dfrac{2F_{\rm act}\sigma_{\rm d}}{k_B T}$

Active force $Lv \mapsto \sigma_{\rm d}\, F_{\rm act}/\gamma$

viscous force $\nu \mapsto \gamma\sigma_{\rm d}^2/m_{\rm d}$

Reynolds number $\text{Re} = \dfrac{m_{\rm d} F_{\rm act}}{\sigma_{\rm d}\gamma^2}$

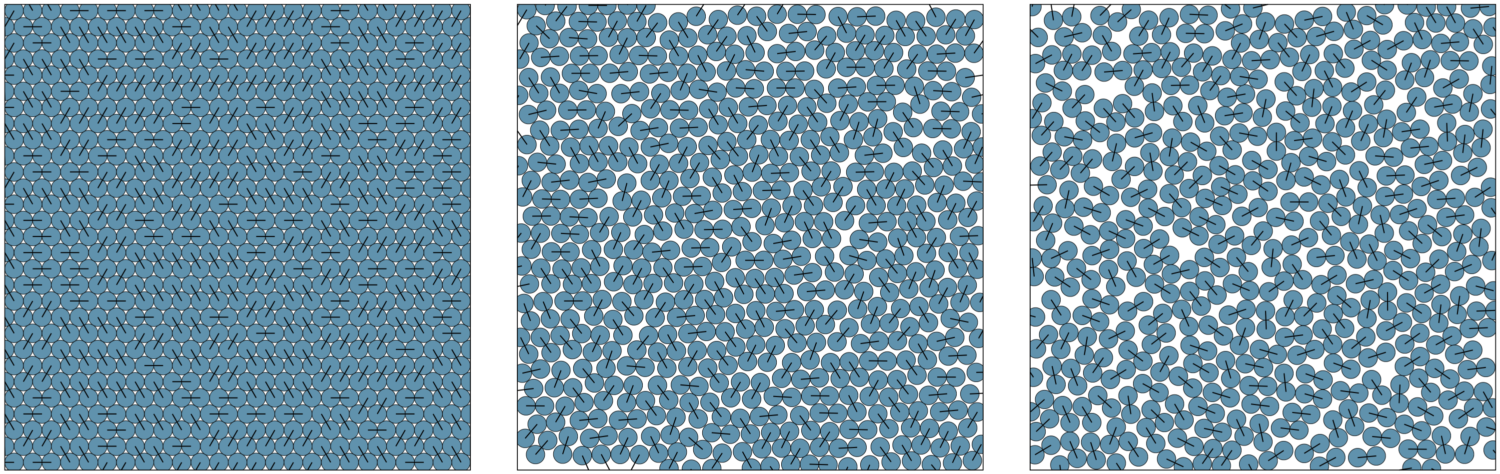
$\text{Pe} \in [0, 40]$ $\qquad$ $\text{Re} < 10^{-2}$

We keep the parameters in the harmonic (fene) and Lennard-Jones (repulsive) potential fixed. Stiff molecule limit: vibrations frozen.

Interest in the ϕ, $F_{\rm act}$ and $k_B T$ dependencies.

Initial conditions

Three cases



Crystal Hexatic order Random

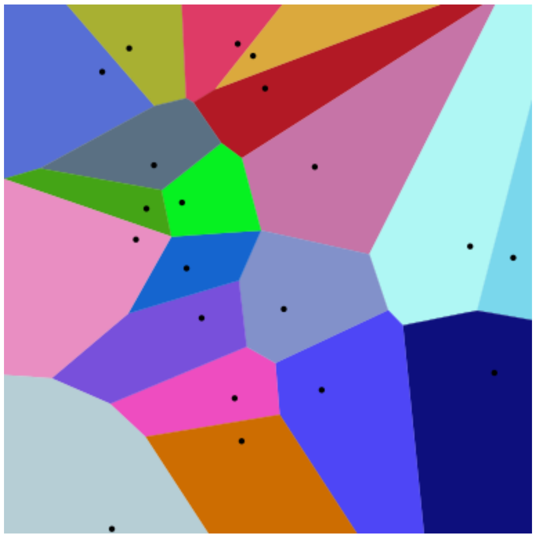
with the desired ϕ

Observables

Voronoi tessellation

A Voronoi diagram is induced by a set of points, called sites, that in our case are the centres of the dumbbell beads.

The plane is subdivided into faces that correspond to the regions where one site is closest.



Focus on the central light-green face

All points within this region are closer to the dot within it than to any other dot on the plane

The region has five neighbouring cells from which it is separated by an edge

The grey zone has six neighbouring cells

Observables

Voronoi tessellation

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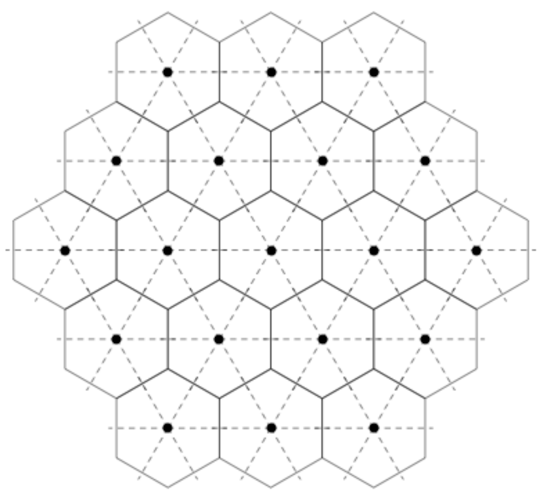


With dashed lines, the triangular lattice

The vertices are the sites

Each site has six nearest neighbours

The angles of the edges of the triangular lattice are

$$\theta_{ij} = 2\pi j/6$$

The hexagonal lattice is the Voronoi tessellation

Observables

Local density

For each bead, i the first estimate of the local density ϕ_i^{Vor} is the ratio between its surface and the area A_i^{Vor} of its Voronoi region:

$$\phi_i^{\mathrm{Vor}} = \frac{\pi \sigma_{\mathrm{d}}^2}{4 A_i^{\mathrm{Vor}}}$$

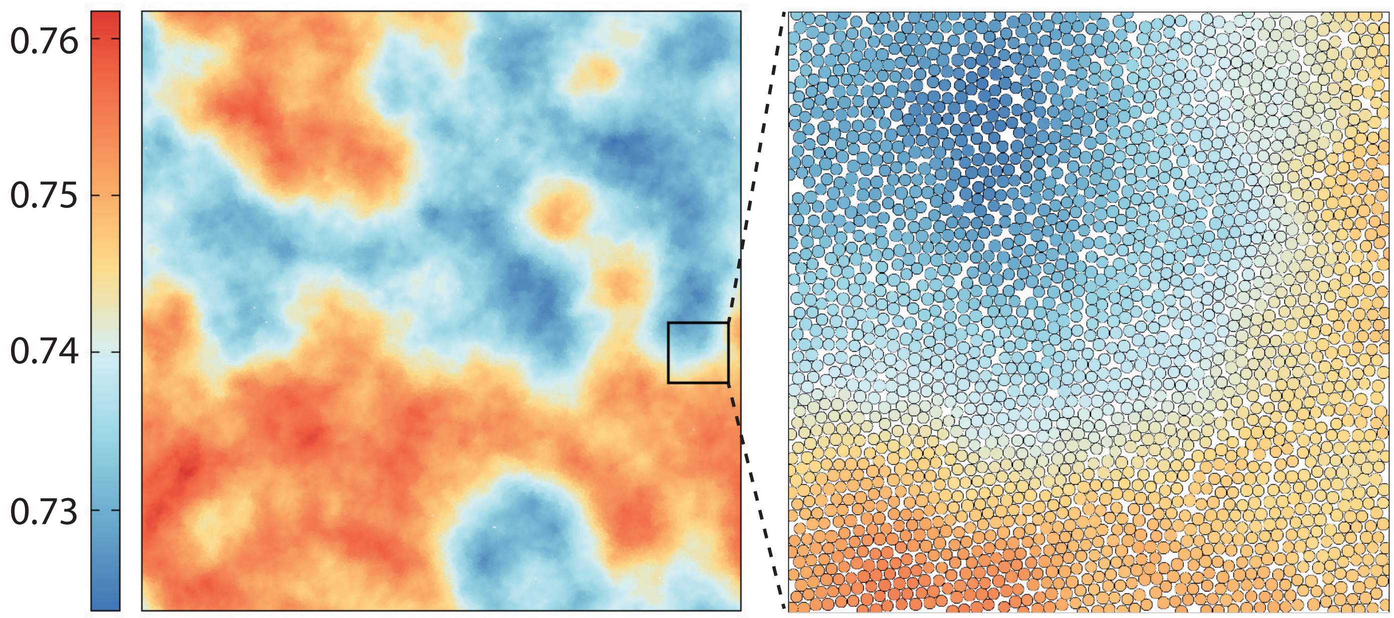
We next coarse-grain this value by averaging the single-bead densities ϕ_i^{Vor} over a disk $S_R^{(i)}$ with radius R

$$[[\phi_i]] \equiv \sum_{i \in S_R^{(i)}} \phi_i^{\mathrm{Vor}} / (\pi R^2)$$

Visualisation: each bead is painted with the colour of its coarse-grained local density value, $[[\phi_i]]$, denser in red, looser in blue.

Observables

Local density colour map - an example



More on this figure later

Observables

Positional order

The (fluctuating) local particle number density

$$\rho(\boldsymbol{r}_0) = \sum_{i=1}^{N} \delta(\boldsymbol{r}_0 - \boldsymbol{r}_i)$$

with normalisation $\int d^d\boldsymbol{r}_0\, \rho(\boldsymbol{r}_0) = N$. In a homogeneous system $\rho(\boldsymbol{r}_0) = N/V$.

The density-density correlation function $C(\boldsymbol{r}+\boldsymbol{r}_0, \boldsymbol{r}_0) = \langle \rho(\boldsymbol{r}+\boldsymbol{r}_0)\rho(\boldsymbol{r}_0)\rangle$ that, for homogeneous (independence of $\boldsymbol{r}_0$) and isotropic ($\boldsymbol{r} \mapsto |\boldsymbol{r}| = r$) cases, is simply $C(\boldsymbol{r}+\boldsymbol{r}_0, \boldsymbol{r}_0) = C(r)$.

The double sum in $C(\boldsymbol{r}+\boldsymbol{r}_0, \boldsymbol{r}_0) = \langle \sum_{ij} \delta(\boldsymbol{r}+\boldsymbol{r}_0 - \boldsymbol{r}_i)\delta(\boldsymbol{r}_0 - \boldsymbol{r}_j)\rangle$ has contributions from $i = j$ and $i \neq j$: $C_{\mathrm{equal}} + C_{\mathrm{diff}}$

Observables

Positional order

The density-density correlation function

$$C(\boldsymbol{r}+\boldsymbol{r}_0,\boldsymbol{r}_0) = \langle \rho(\boldsymbol{r}+\boldsymbol{r}_0)\rho(\boldsymbol{r}_0)\rangle = \sum_{ij}\langle \delta(\boldsymbol{r}+\boldsymbol{r}_0-\boldsymbol{r}_i)\delta(\boldsymbol{r}_0-\boldsymbol{r}_j)\rangle$$

is linked to the structure factor

$$S(\boldsymbol{q}) \equiv \frac{1}{N}\langle \tilde{\rho}(\boldsymbol{q})\tilde{\rho}(-\boldsymbol{q})\rangle = \frac{1}{N}\langle \sum_{i=1}^{N}\sum_{j=1}^{N} e^{-\mathrm{i}\boldsymbol{q}\cdot(\boldsymbol{r}_i-\boldsymbol{r}_j)}\rangle$$

by

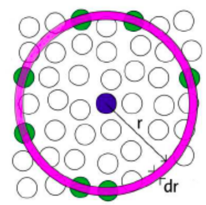
$$N\,S(\boldsymbol{q}) \;=\; \int d^d\boldsymbol{r}_1 \int d^d\boldsymbol{r}_2\, C(\boldsymbol{r}_1,\boldsymbol{r}_2)\, e^{-\mathrm{i}\boldsymbol{q}\cdot(\boldsymbol{r}_1-\boldsymbol{r}_2)}$$

Observables

Positional order

In isotropic cases, i.e. liquid phases, the pair correlation function

$\frac{N}{V}\, g(r) =$ average number of particles at distance r from a tagged particle at $\boldsymbol{r}_0$



is linked to the structure factor

$$S(\boldsymbol{q}) = \frac{1}{N}\langle \sum_{i=1}^{N} \sum_{j=1}^{N} e^{-\mathrm{i}\boldsymbol{q}\cdot(\boldsymbol{r}_i - \boldsymbol{r}_j)} \rangle$$

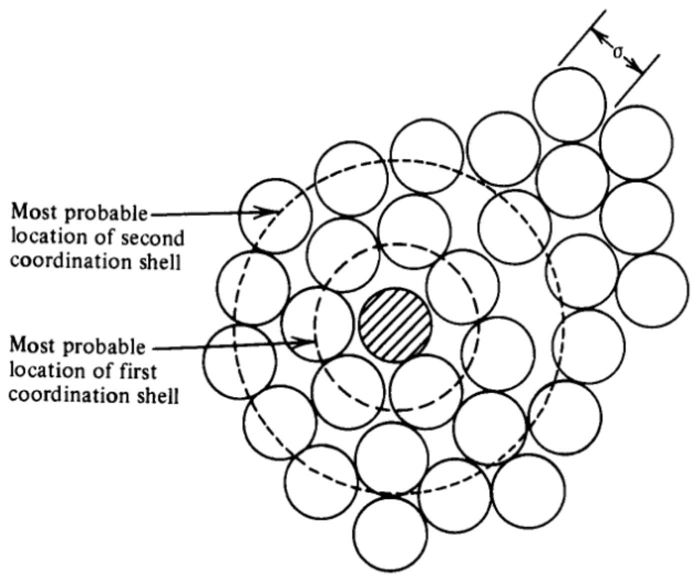
by

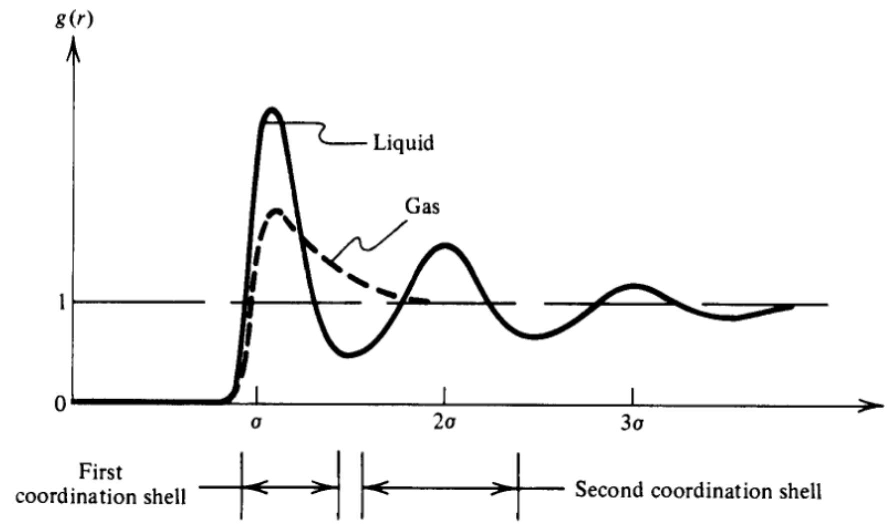
$$S(\boldsymbol{q}) = 1 + \frac{N}{V} \int d^d\boldsymbol{r}\; g(r)\, e^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}}$$

Peaks in $g(r)$ are related to peaks in $S(q)$. The first peak in $S(q)$ is at $q_0 = 2\pi/\Delta r$ where Δr is the distance between peaks in $g(r)$ (that is close to the inter particle distance as well).

Observables

Liquid

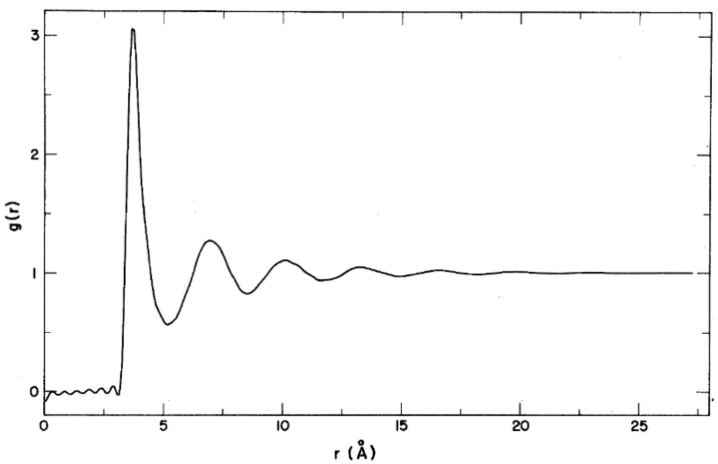


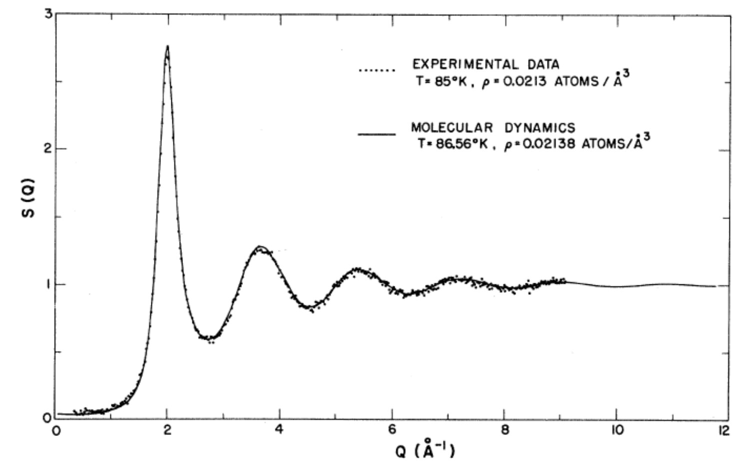


"Introduction to Modern Statistical Mechanics", **Chandler** (OUP)

Observables

Experiments & simulations of liquids





Inter-peak distance in $g(r)$ is $\Delta r \simeq \sigma \simeq 3\text{Å}$

Position of the first peak in $S(q)$ is at $q_0 \simeq 2\pi/\Delta r \simeq 2\ \text{Å}^{-1}$

"Structure Factor and Radial Distribution Function for Liquid Argon at 85K",

Yarnell, Katz, Wenzel & König, Phys. Rev. Lett. 7, 2130 (1973)

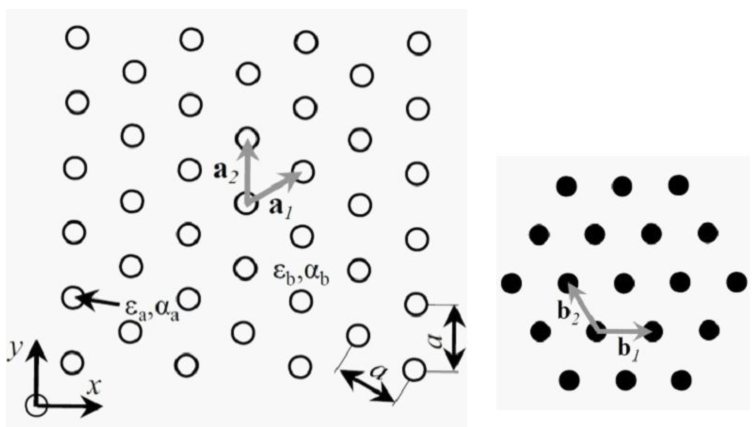
Observables

Structure factor for crystals

$\boldsymbol{r}_i$ and $\boldsymbol{r}_j$ are the positions of the beads i and j and $\boldsymbol{q}$ is the wave-vector :

$$S(\boldsymbol{q}) = \frac{1}{N} \sum_{ij} \langle e^{\mathrm{i}\boldsymbol{q}\cdot(\boldsymbol{r}_i - \boldsymbol{r}_j)} \rangle$$

Visualisation: $2d$ representation in the (q_x, q_y) plane, Bragg peaks.



(a) (b)

Triangular lattice in real space Hexagonal lattice in reciprocal space

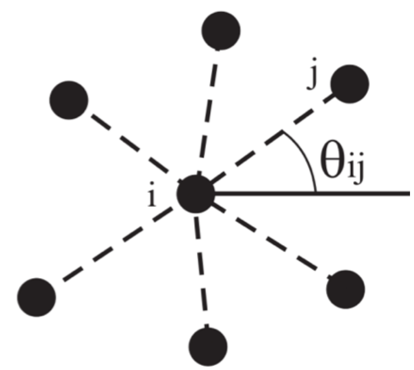
Voronoi cell Brillouin zone

Observables

Hexatic order

The local hexatic fluctuating order

$$\psi_{6i} = \frac{1}{N^i_{\rm nn}} \sum_{j=1}^{N^i_{\rm nn}} e^{6{\rm i}\theta_{ij}}$$



with $N^i_{\rm nn}$ the number of nearest (Voronoi) neighbours of bead i and θ_{ij} the angle between the segment that connects i with its neighbour j and the x axis.

For beads placed on the vertices of a triangular lattice, each bead has six nearest-neighbours, $j = 1, \ldots, 6$, the angles are $\theta_{ij} = 2\pi j/6$ and $\psi_{6i} = 1$ for all i.

measures orientational order

Observables

Hexatic order

The local hexatic fluctuating order

$$\psi_{6i} = \frac{1}{N^i_{\rm nn}} \sum_{j=1}^{N^i_{\rm nn}} e^{6\mathrm{i}\theta_{ij}}$$

We also look at the average of the modulus and modulus of the average

$$2N\,\psi_6 = \left| \sum_{i=1}^{N} \psi_{6i} \right| \qquad\qquad 2N\,\Gamma_6 = \sum_{i=1}^{N} |\psi_{6i}|$$

and the correlation functions

$$g_6(r) = \frac{\sum_{ij} [\langle \psi^*_{6i} \psi_{6j} \rangle] \Big|_{r_{ij}=r}}{[\langle |\psi_{6i}|^2 \rangle]}$$

Note that the normalisation is site independent

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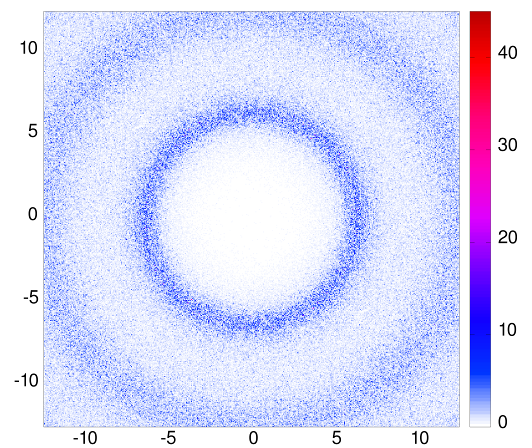
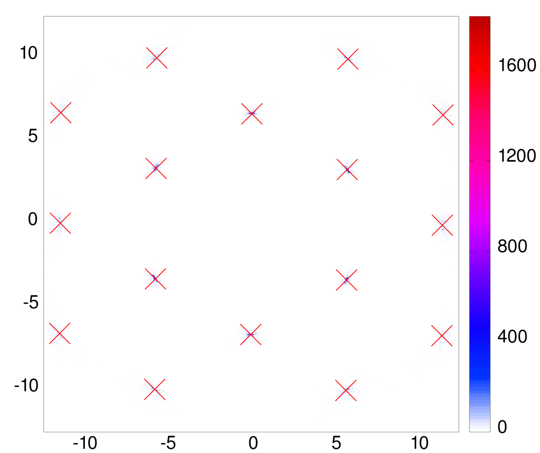
Passive system

Structure factor - very low and very high density

$\phi = 0.66$



$\phi = 0.76$



Liquid

Solid

Bragg peaks

Primitive vectors

$$\boldsymbol{q}_1 = \frac{4\pi}{a\sqrt{3}} \left(\frac{\sqrt{3}}{2}, -\frac{1}{2} \right)$$

$$\boldsymbol{q}_2 = \frac{4\pi}{a\sqrt{3}} (0, 1)$$

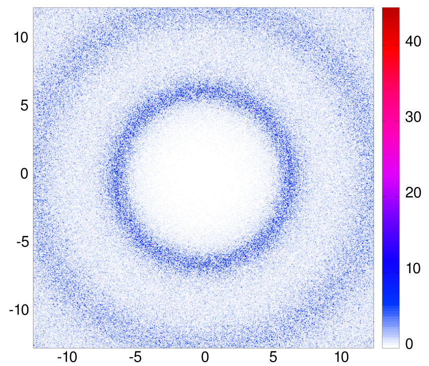
Unit of length

$$a = \left(\frac{\pi}{2\sqrt{3}\phi} \right)^{1/2} \sigma_{\rm d}$$

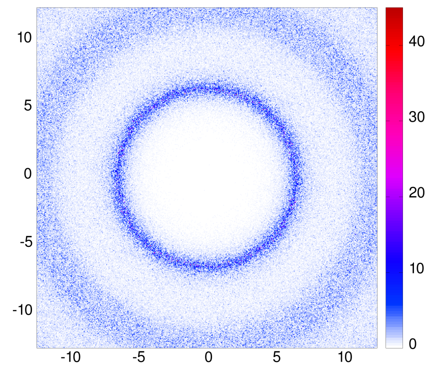
Passive system

Structure factor - progressive increase in density

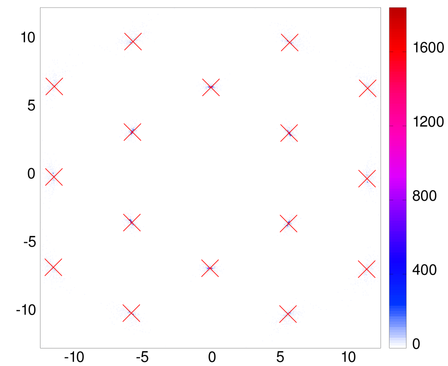
$\phi = 0.66$
(liquid)



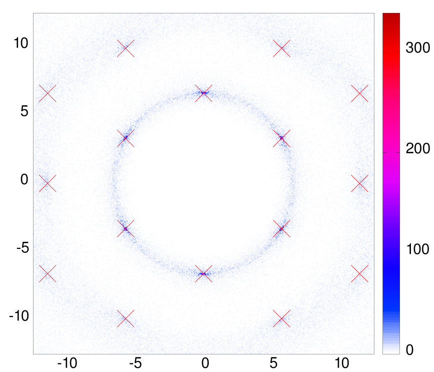
$\phi = 0.72$
(liquid)



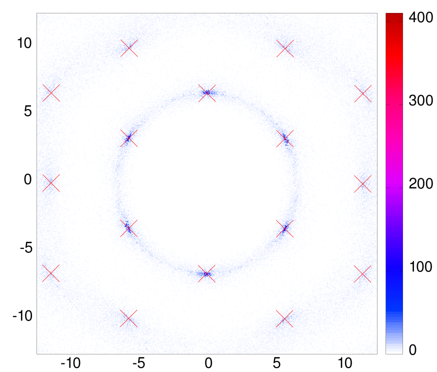
$\phi = 0.76$
(solid)



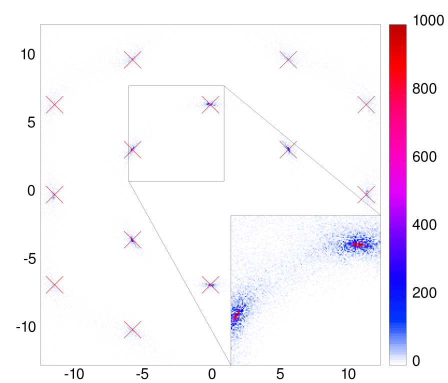
$\phi = 0.734$
(co-existence)



$\phi = 0.74$
(co-existence)

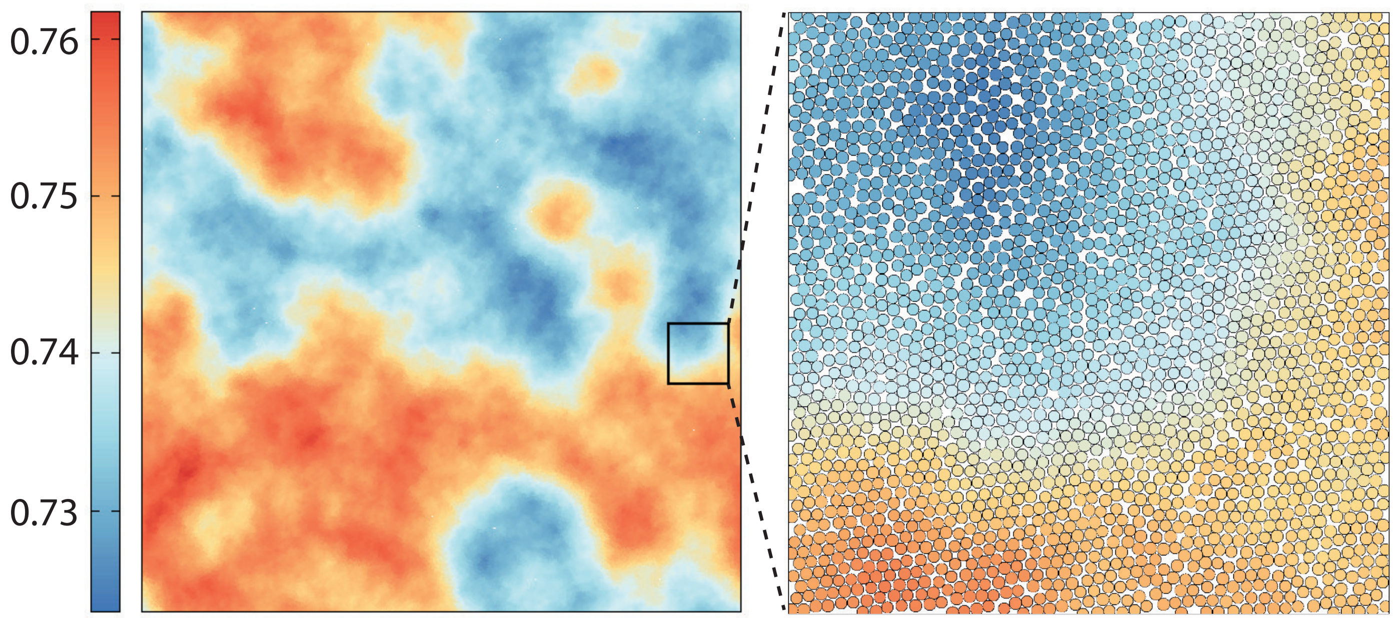


$\phi = 0.75$
(co-existence)



Observables

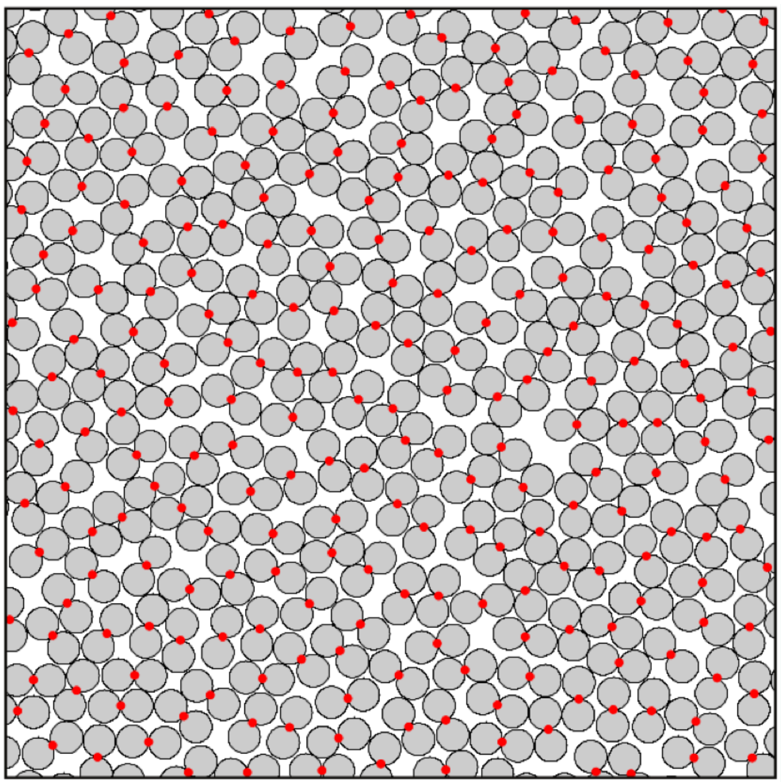
Local density colour map in the co-existence region



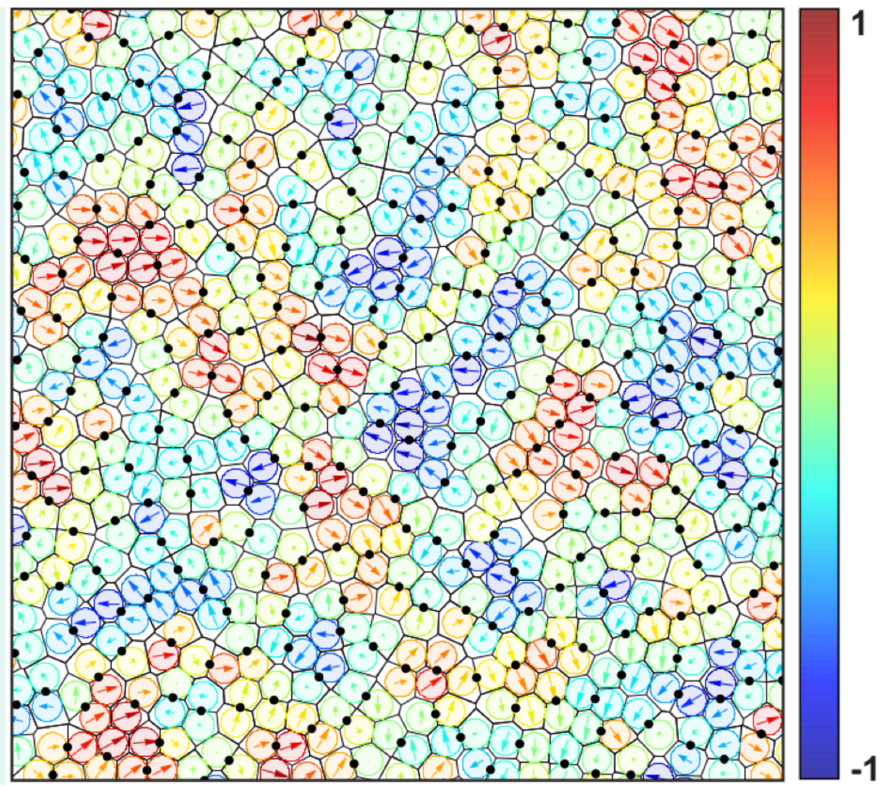
Zoom over an interface

Passive system

Hexatic order parameter



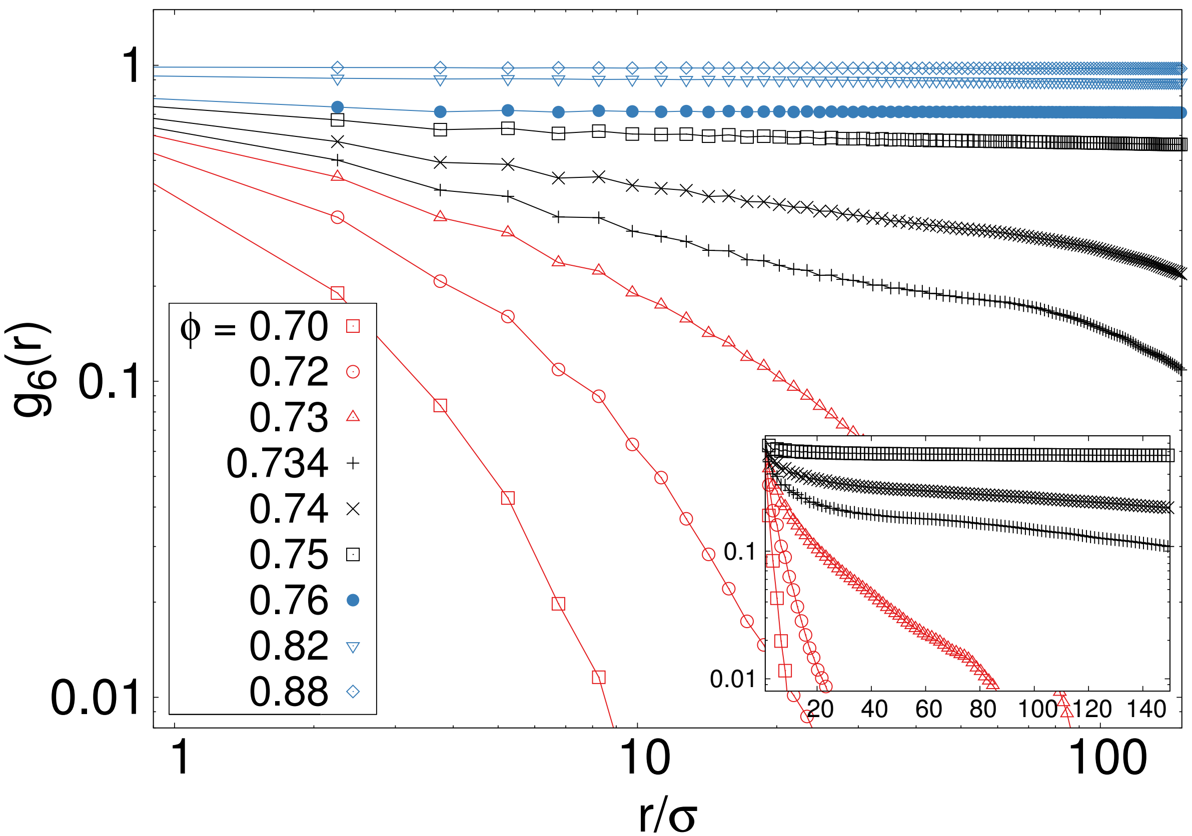
Dumbells



Hexatic local vector

Passive system

Hexatic correlation function



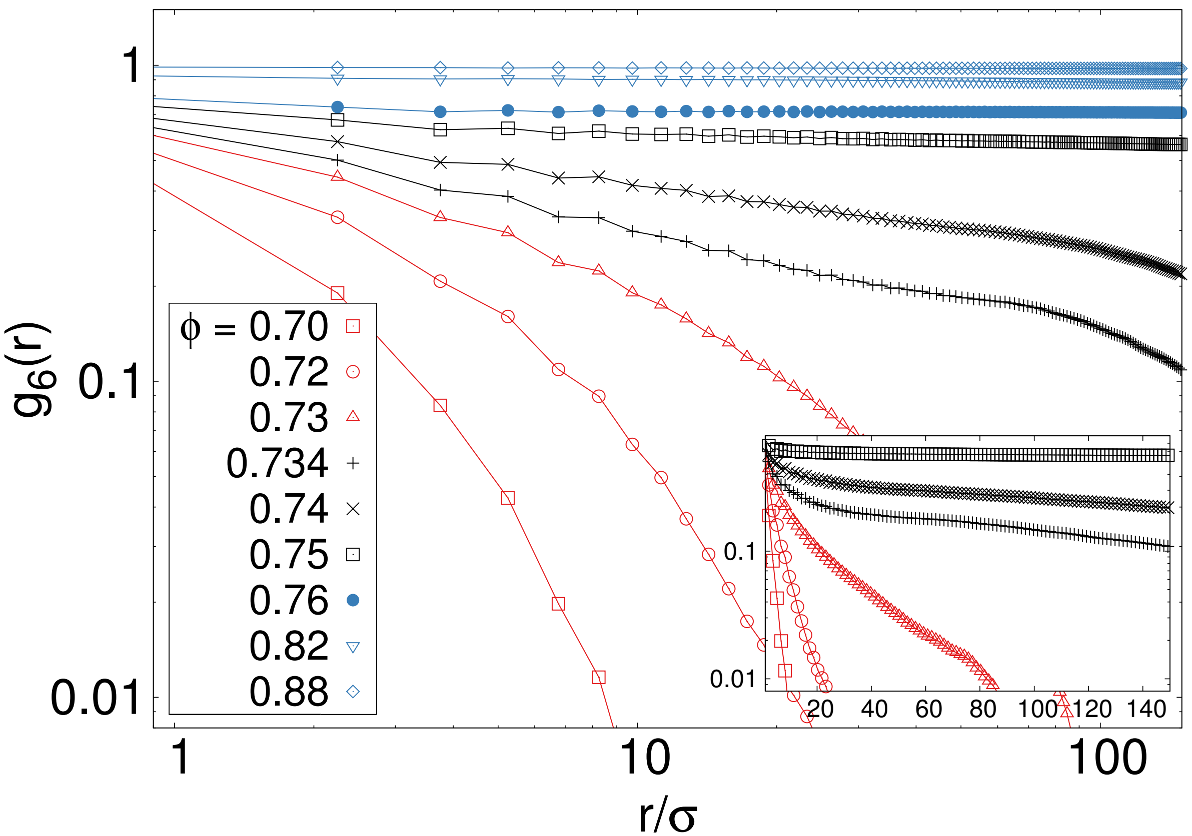
Solid

Hexatic

Liquid

Passive system

Hexatic correlation function



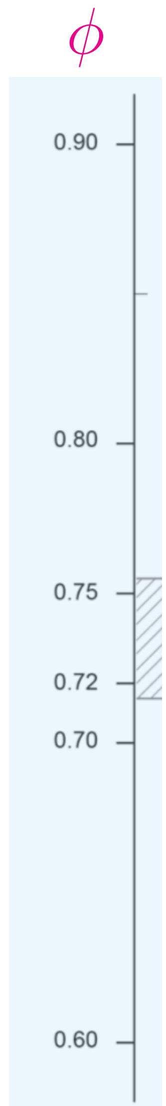
Saturation

Algebraic

Exponential

Passive system

Phase diagram



ϕ_{CP}

ϕ_{RCP}

solid

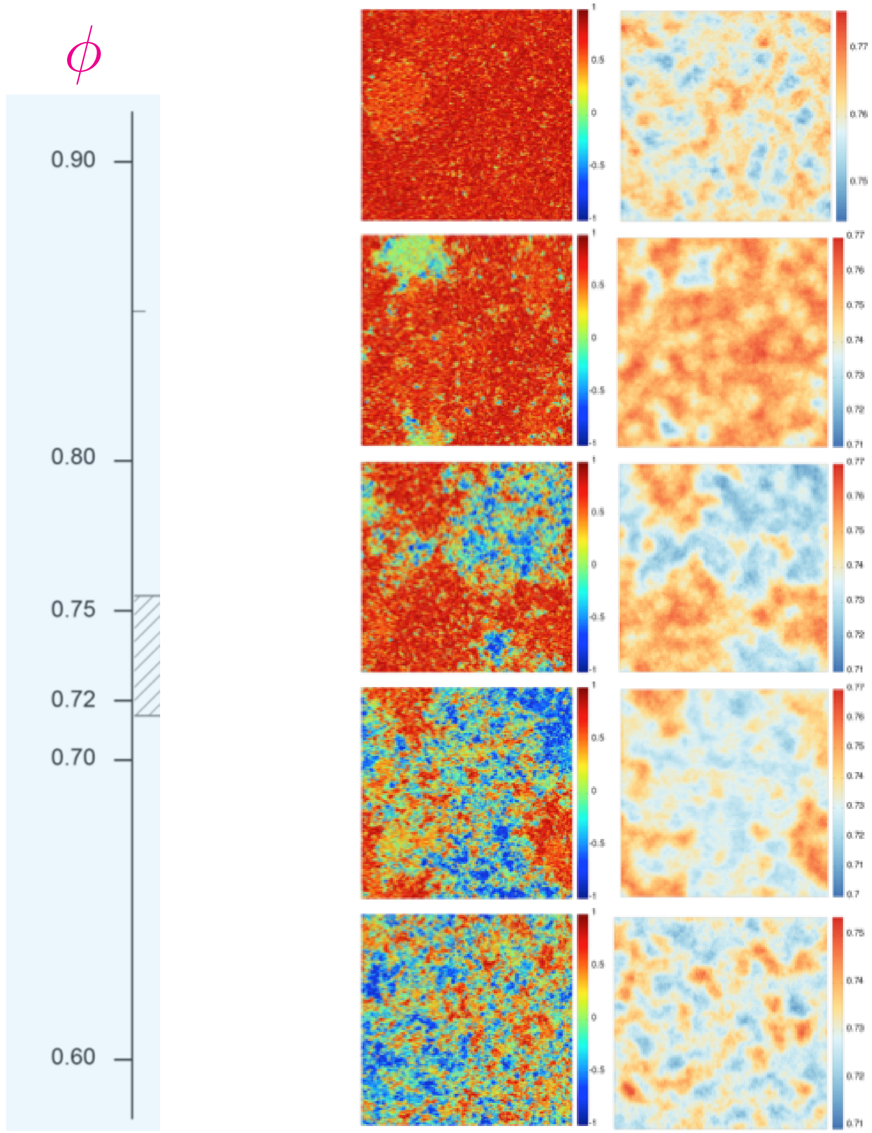
hexatic

co-existence   Really ?

liquid

Passive system

Phase diagram



1st column

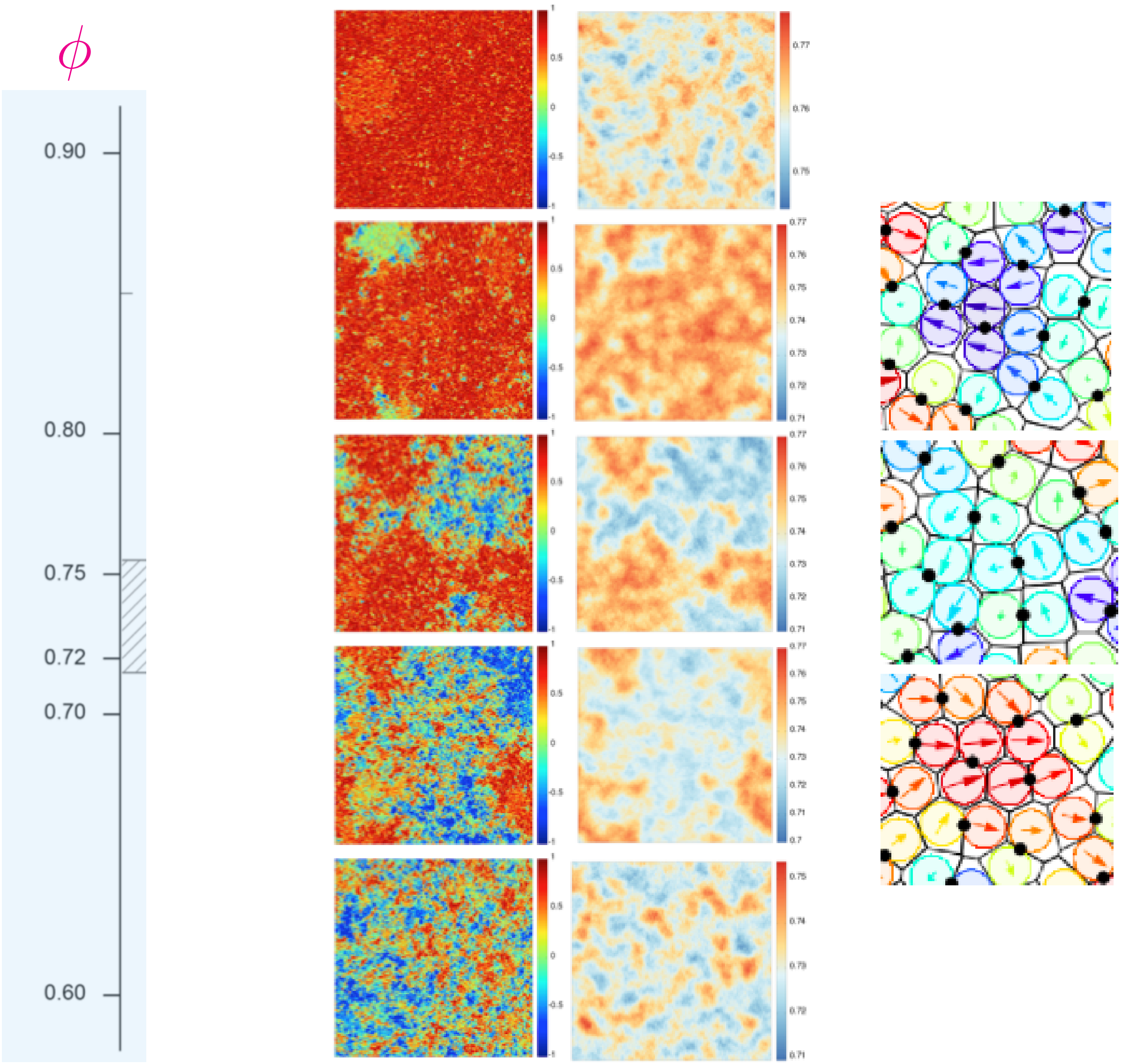
Local hexatic ψ_{6i}

2nd column

Local density $[[\phi_i]]$

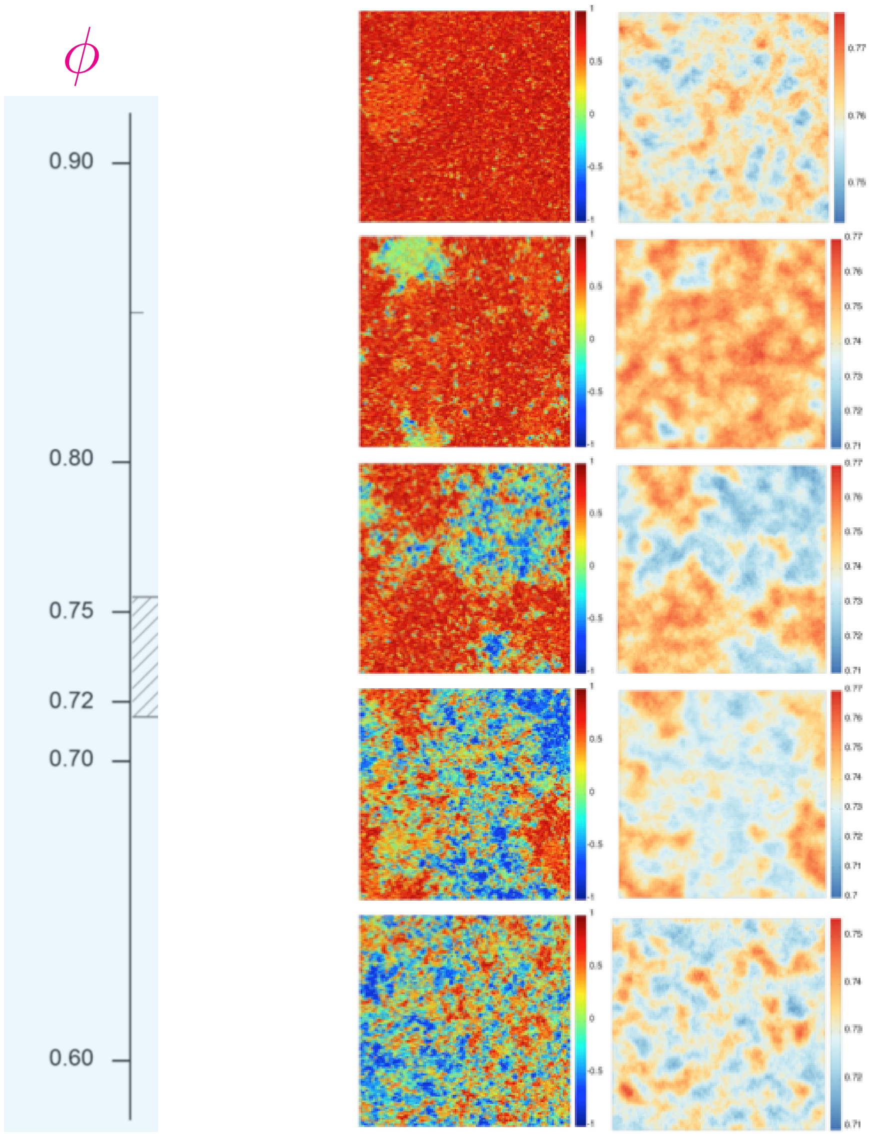
Passive system

Phase diagram



Passive system

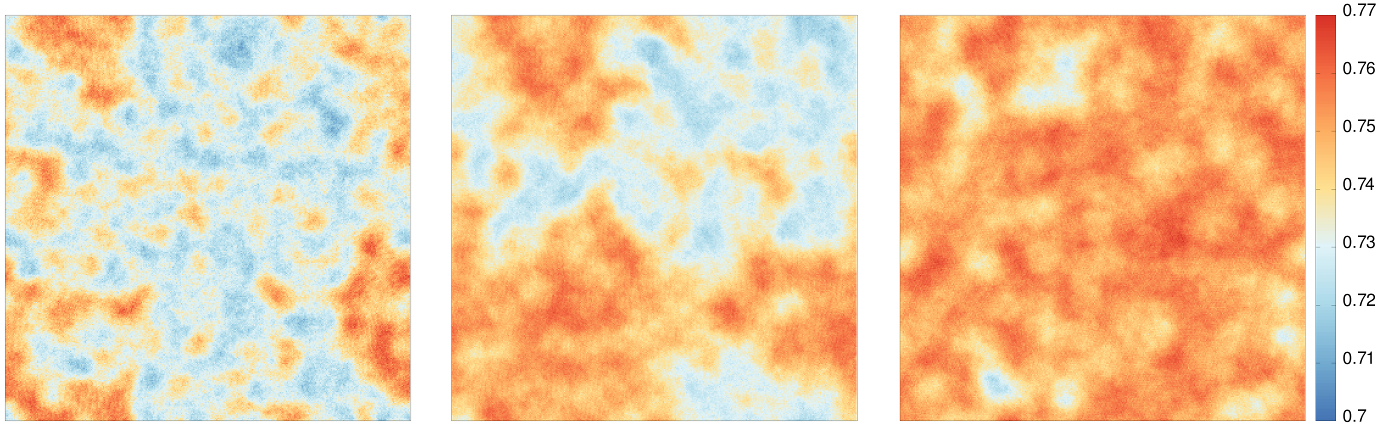
Phase diagram



Spatial correlation between regions of high density and regions of large absolute value of the local hexatic order parameter

Passive system

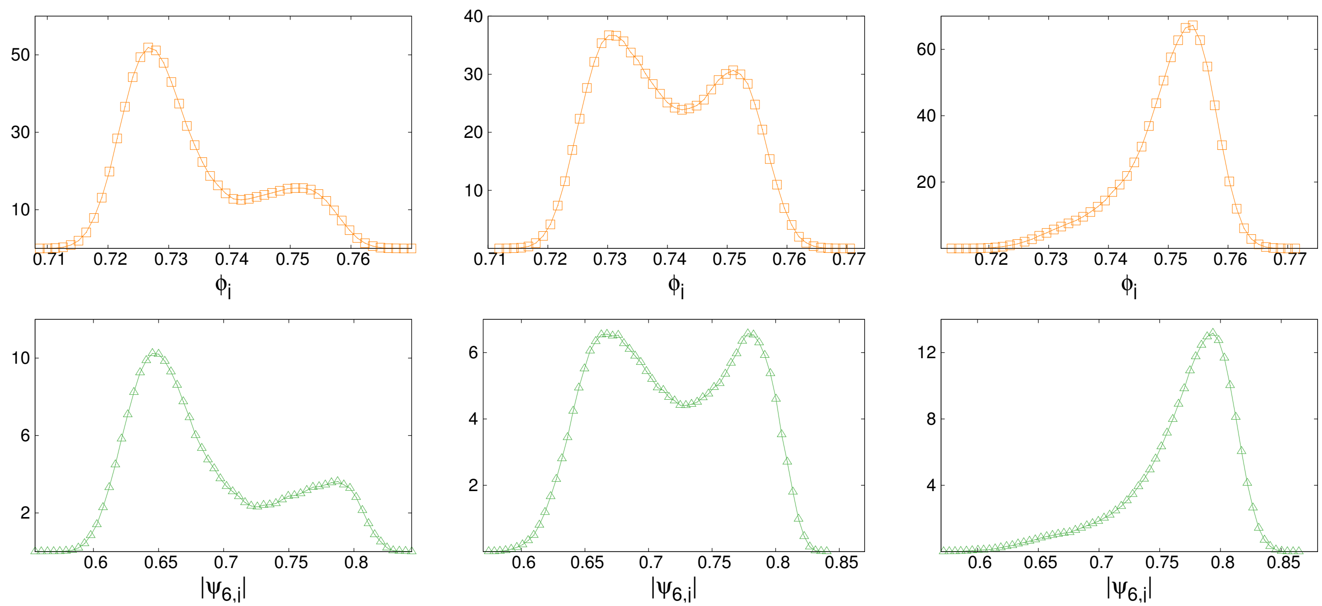
Local density & local hexatic parameter



$\phi = 0.734$ (co-existence)

$\phi = 0.74$ (co-existence)

$\phi = 0.75$ (upper limit of co-existence)



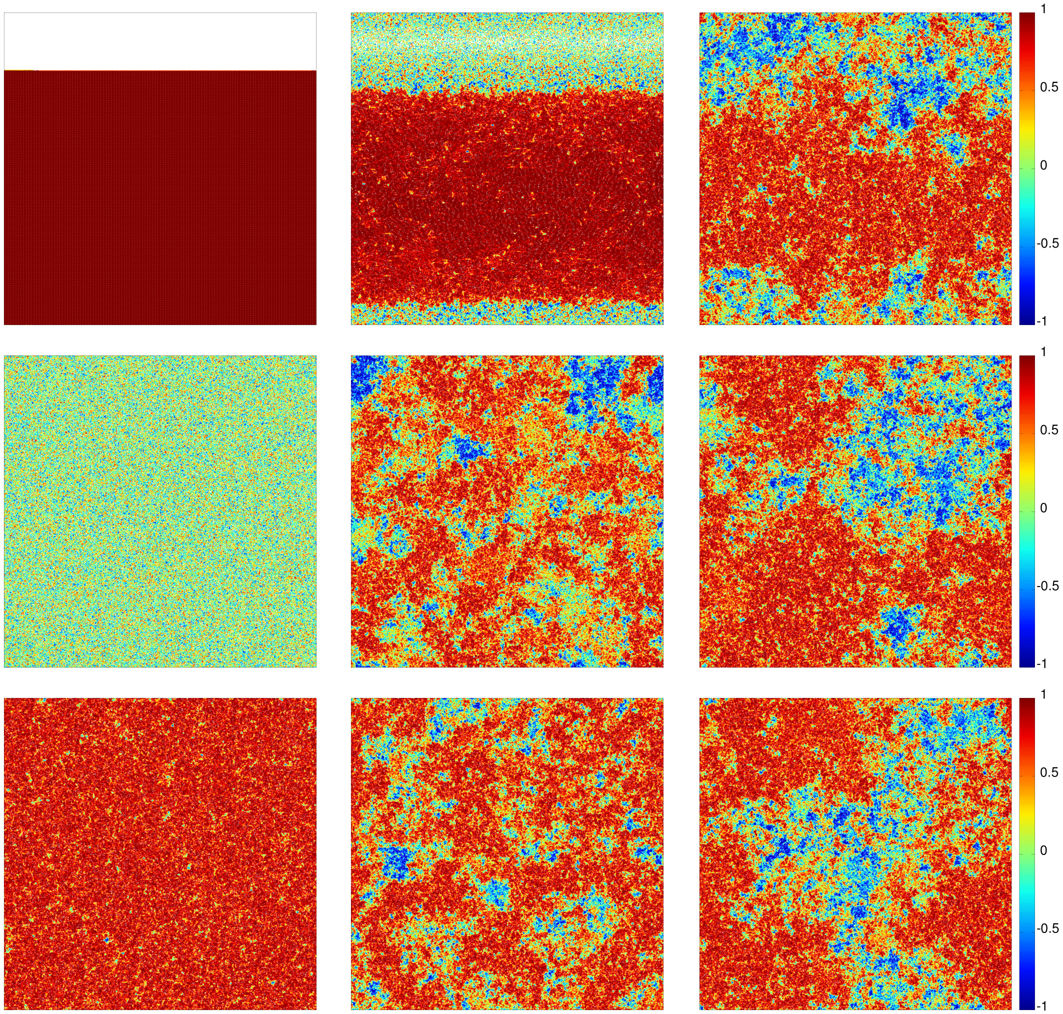
Passive system

Co-existence region : independence of the initial conditions

crystal

random

hexatic



$t = 0$ t_1 t_2

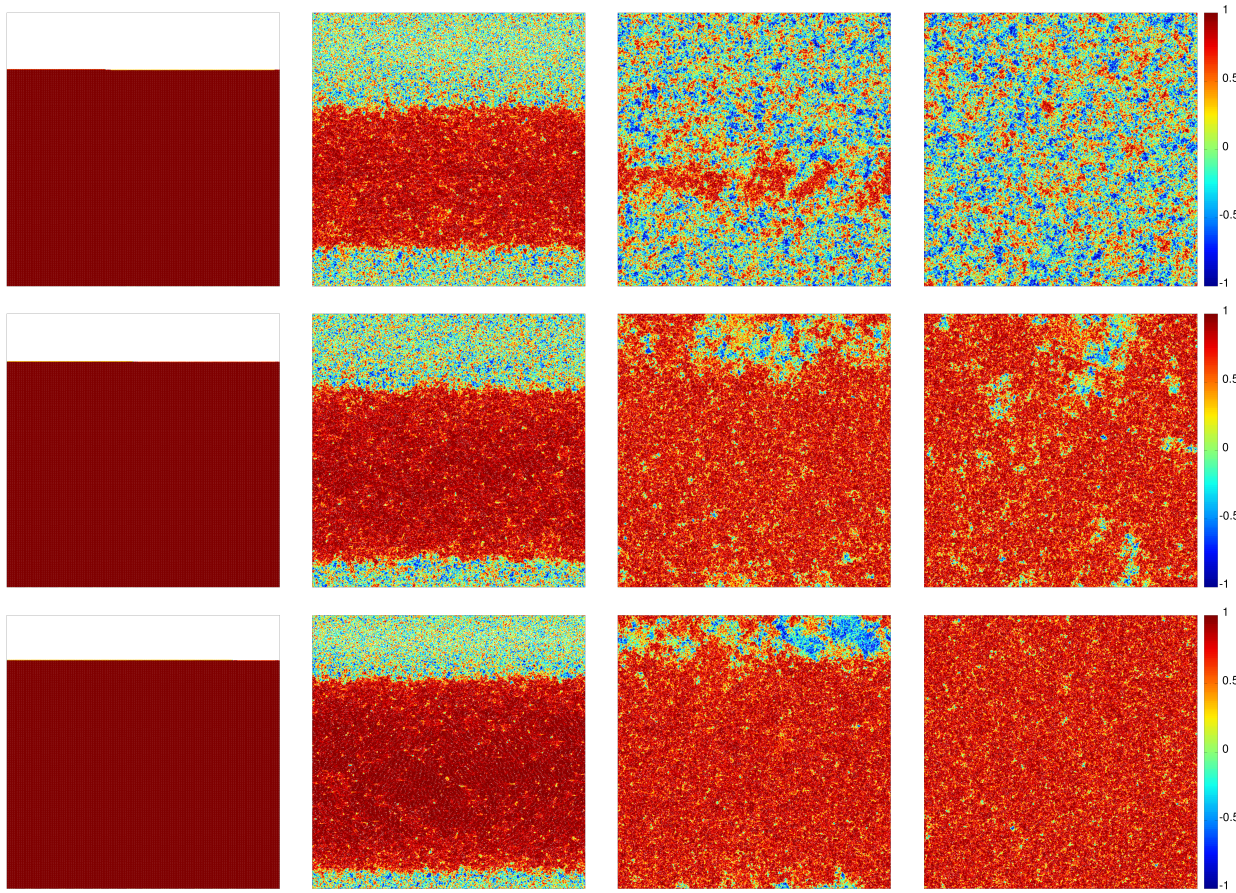
Passive system

Dynamics: Below, in and above the co-existence region

$\phi = 0.72$

$\phi = 0.75$

$\phi = 0.76$



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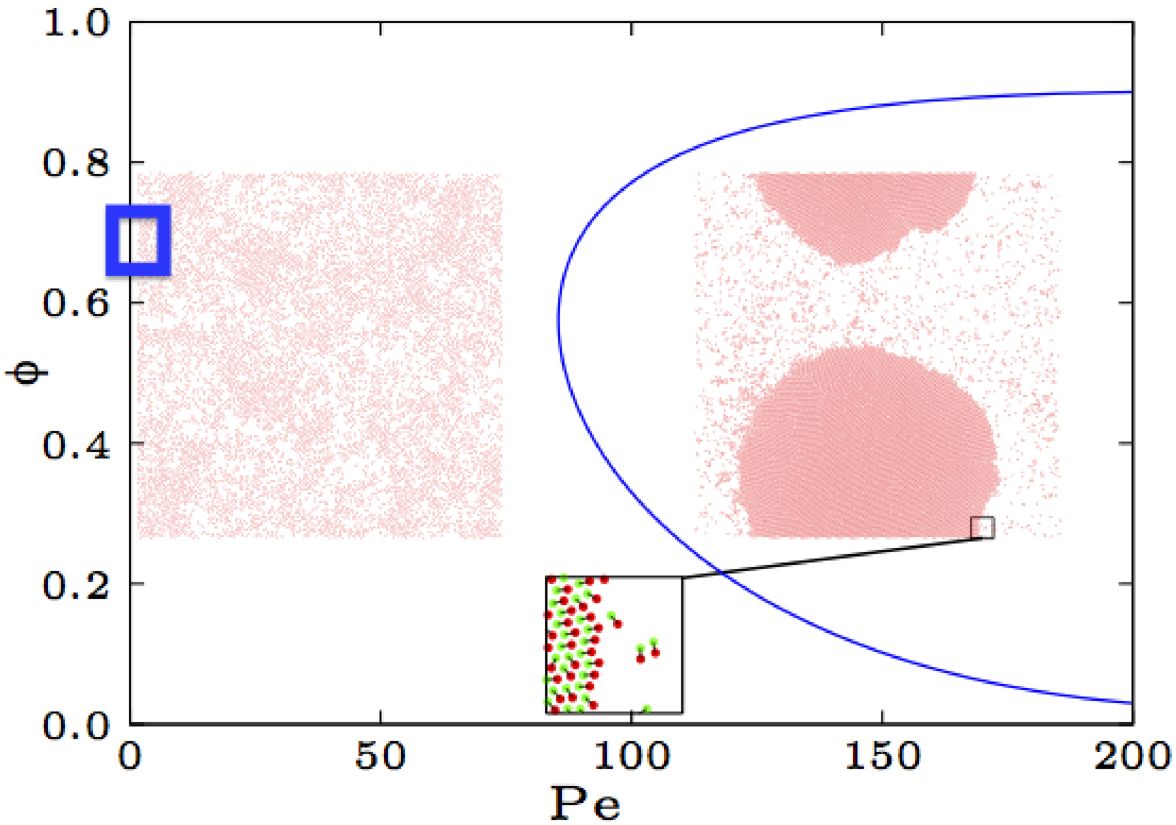
Active system

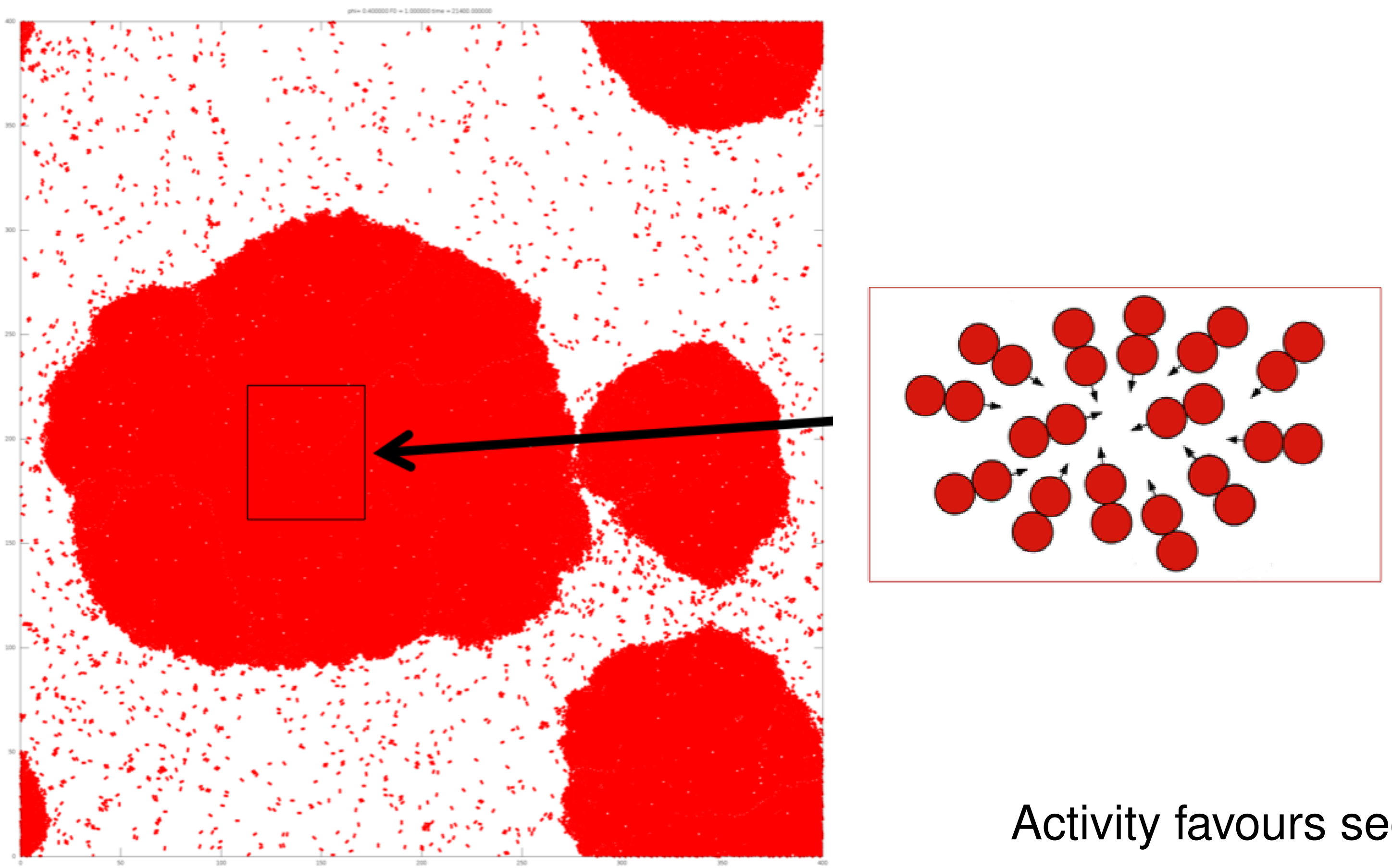
OLD phase diagram & new result



Connection between the two extremes ?

Active system

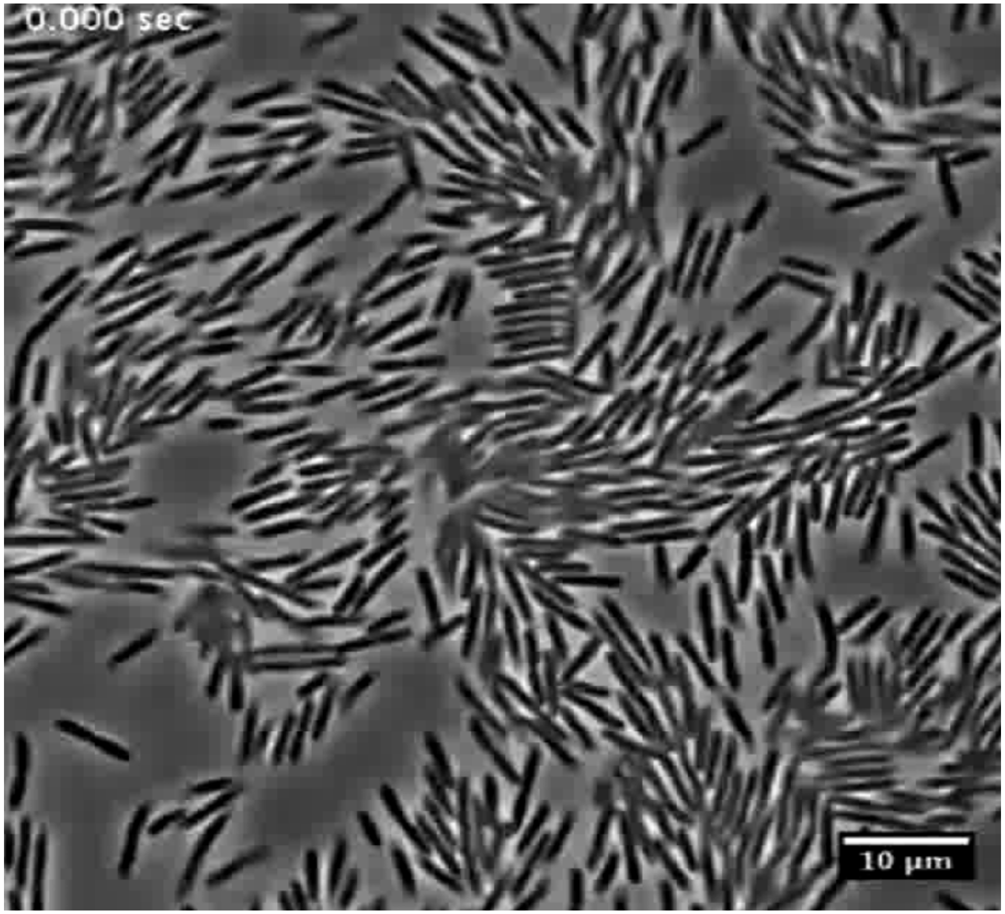
Active mechanism for segregation



Activity favours segregation

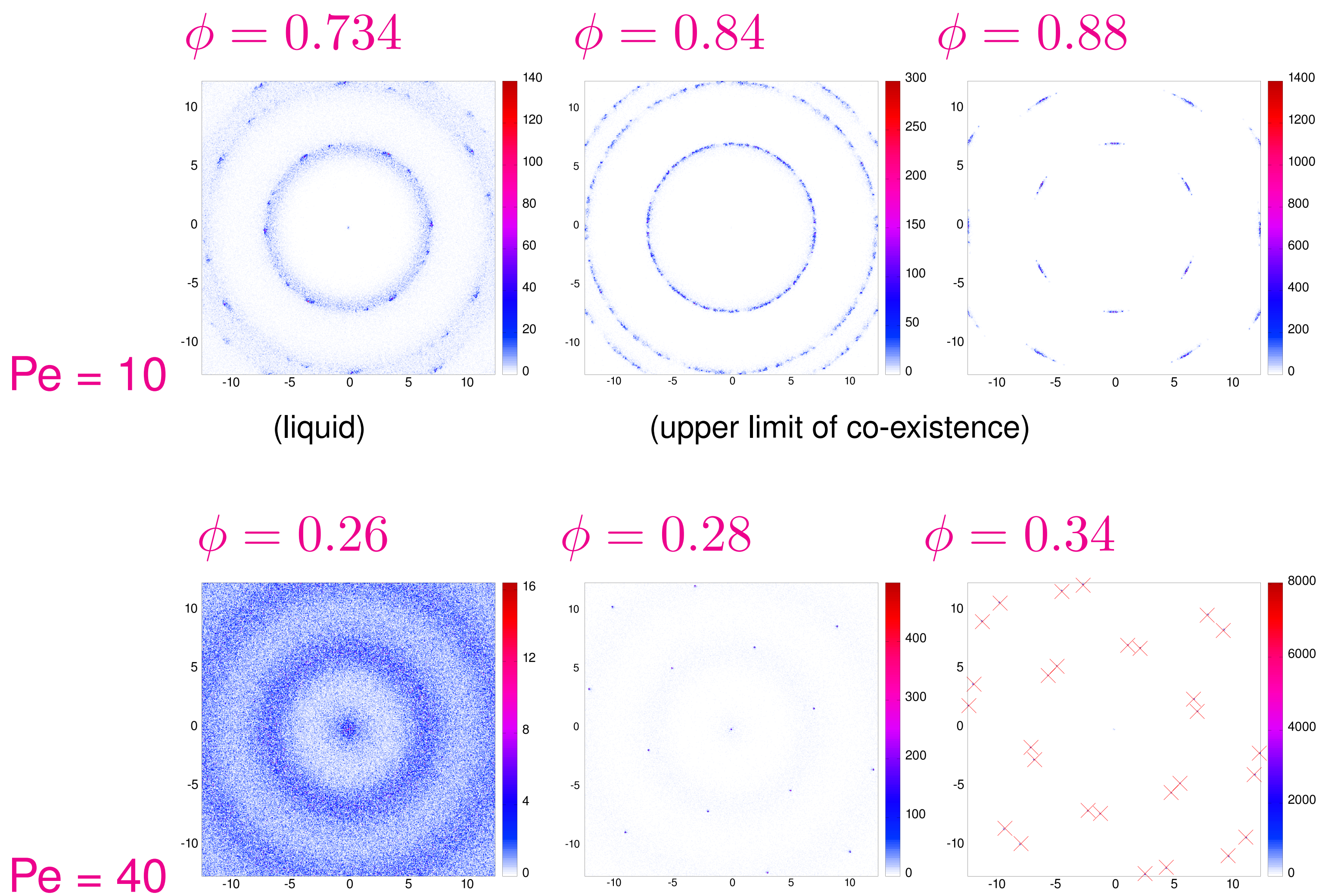
Active system

Mechanism for segregation



Active system

Structure factor Pe = 10 & Pe = 40



Pe = 10: (liquid) — (upper limit of co-existence)

Pe = 40: (liquid) — (lower limit of co-existence

Dynamics

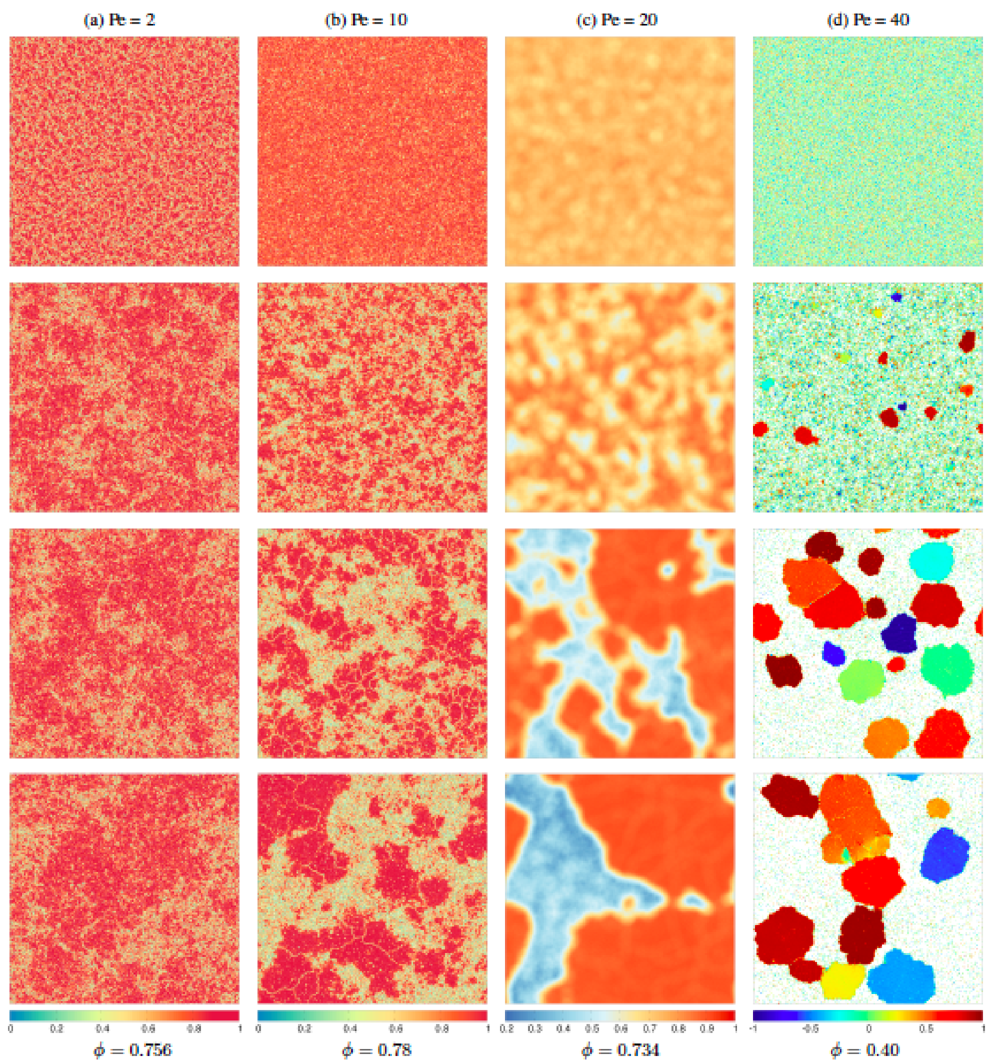
$\phi = 0.756$ **and Pe =** 2 **(co-existence)**

Dynamics

$|\psi_{6i}|$ at $\phi = 0.74$ and Pe = 10 (co-existence)

Active coarsening

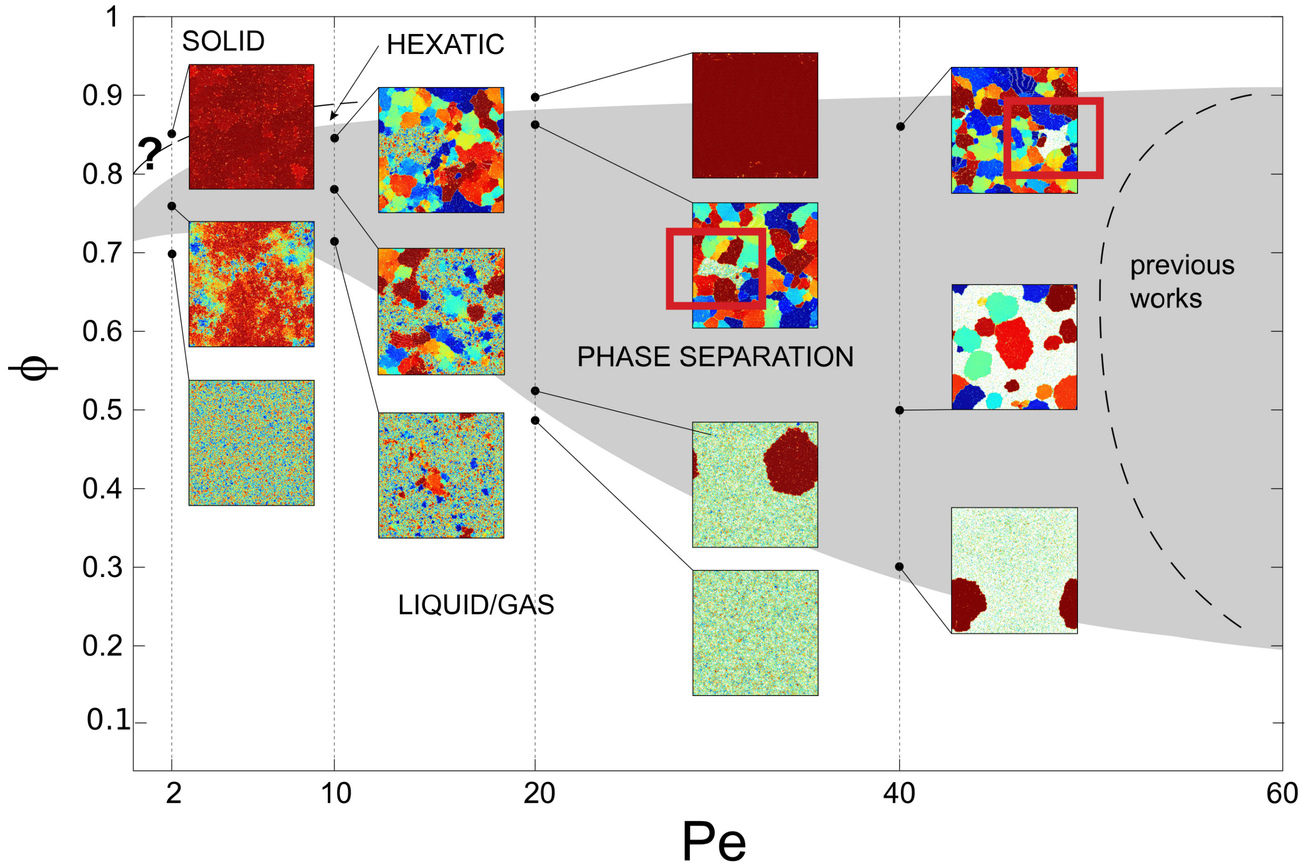
at lower limit of coexistence



$|\psi_{6i}|$ $|\psi_{6i}|$ $[[\phi_i]]$ ψ_{6i}

Phase diagram

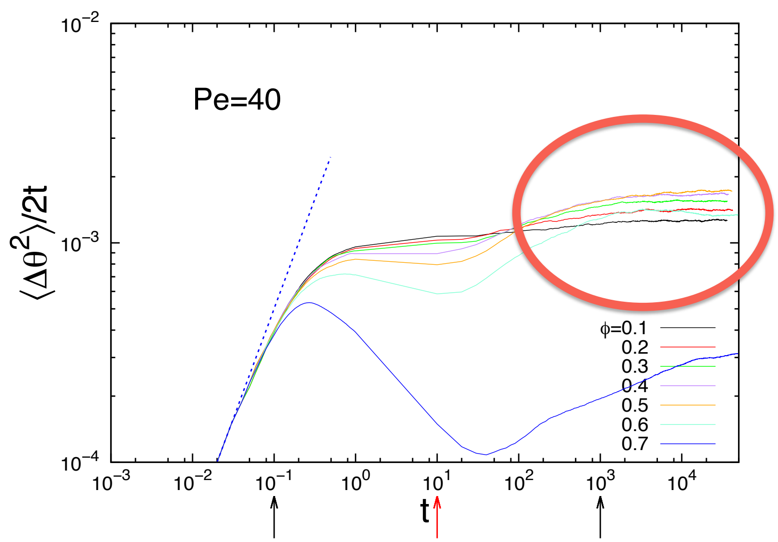
Active dumbbells

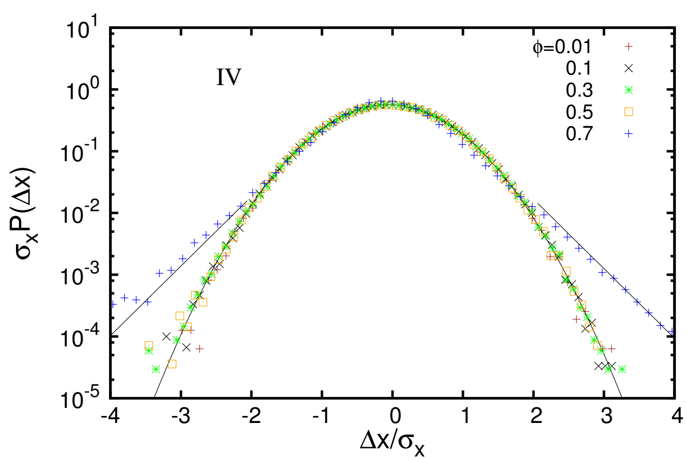


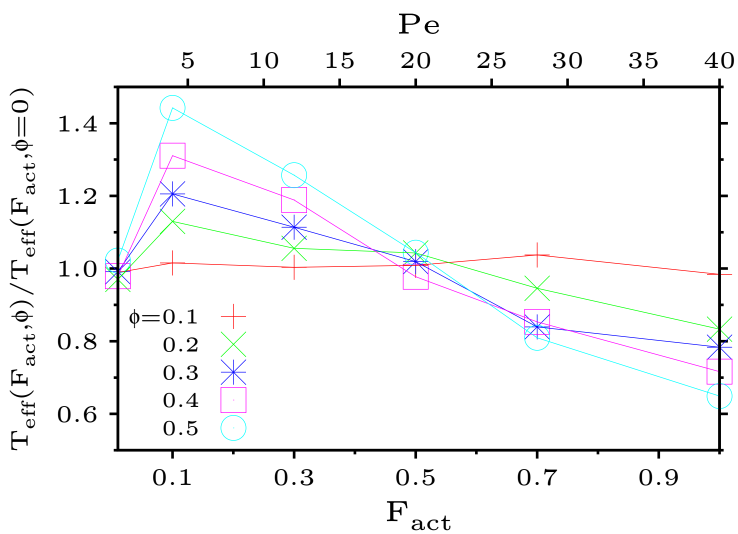
$T = 0.05$

Discussion

To understand better

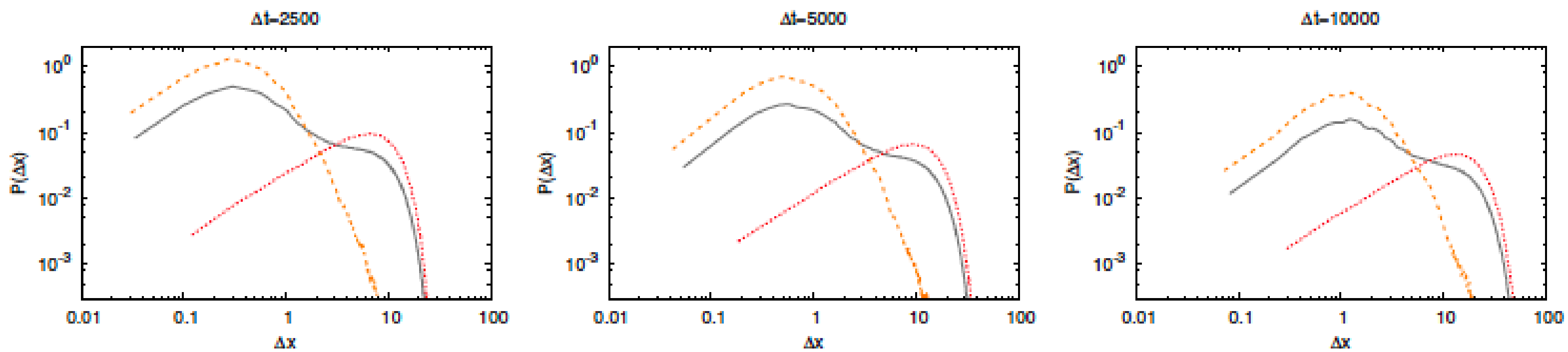






Discussion

Two populations in co-existence region



Pe = $10, \phi = 0.78$ at $t = 2500,\ 5000,\ 10000$

The averaged hexatic modulus is computed for each particle on a radius of 10 $\sigma_{\rm d}$ around the particle itself, and a particle is considered to be inside a cluster only if this value is greater than 0.75. Only such particles were taken into account in the red peak on the right.

In black : all dumbbells

Discussion

Some things to do

- Confirm this picture for active hard and soft disks.
- Understand how to define a meaningful pressure.
- Investigate the dynamics taking into account the heterogeneity of the co-existence region.
- Revisit the effective temperature measurements.