Active Matter in two dimensions

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Work in collaboration with

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L. Carenza (Bari & Istanbul)
A. Suma (Trieste, Philadelphia & Bari)
D. Levis & I. Pagonabarraga (Barcelona & Lausanne)

Leipzig, 2023

Active matter

Definition

Active matter is composed of large numbers of active "agents", which consume energy and thus move or exert mechanical forces.

Due to the energy consumption, these systems are intrinsically out of thermal equilibrium.

Homogeneous energy injection (not from the borders, *cfr.* shear).

Coupling to the environment (bath) allows for dissipation

Active matter

Realisations & modelling

- Wide range of scales: macroscopic to microscopic

  Natural examples are birds, fish, cells, bacteria.

  Artificial realisations are Janus particles, asymmetric grains, toys, etc.

- Embedding spaces in $3d$, $\boxed{2d}$ and $1d$.

- Modelling: very detailed to coarse-grained or schematic:

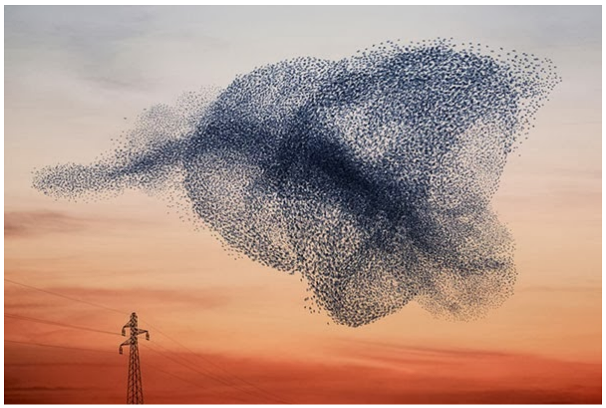
  — microscopic or *ab initio* with focus on active mechanism,

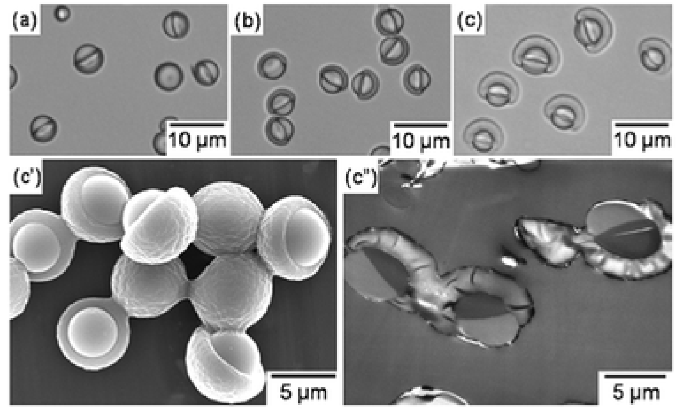
  — *mesoscopic*, just forces that do not derive from a potential,

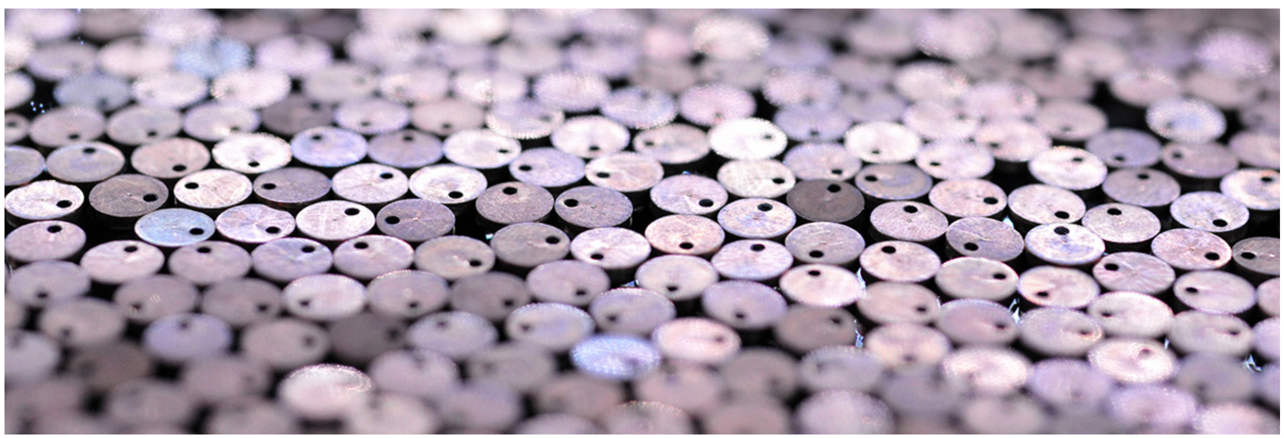
  — *Cellular automata* like in the Vicsek model.

Active matter

Natural & artificial systems







Experiments & observations **Bartolo et al.** Lyon, **Bocquet et al.** Paris, **Cavagna et al.** Roma, **di Leonardo et al.** Roma, **Dauchot et al.** Paris, just to mention some Europeans

Active matter

Realisations & modelling

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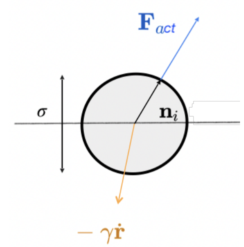
Active Brownian Disks in $2d$

(Overdamped) Langevin equations (the standard model)



Active force $\mathbf{F}_{\rm act}$ along $\mathbf{n}_i = (\cos\theta_i, \sin\theta_i)$

$$m\ddot{\mathbf{r}}_i + \gamma\dot{\mathbf{r}}_i = F_{\rm act}\mathbf{n}_i - \nabla_i \sum_{j(\neq i)} U_{\rm Mie}(r_{ij}) + \boldsymbol{\xi}_i \; , \qquad \dot{\theta}_i = \eta_i \; ,$$

$\mathbf{r}_i$ position of ith particle & $r_{ij} = |\mathbf{r}_i - \mathbf{r}_j|$ inter-part distance,

$U_{\rm Mie}$ short-range **repulsive** Mie potential, over-damped limit $m \ll \gamma$

$\boldsymbol{\xi}$ and η zero-mean Gaussian noises with

$\langle \xi_i^a(t)\, \xi_j^b(t')\rangle = 2\gamma k_B T \delta_{ij}^{ab}\delta(t-t')$ and $\langle \eta_i(t)\, \eta_j(t')\rangle = 2D_\theta\delta_{ij}\delta(t-t')$

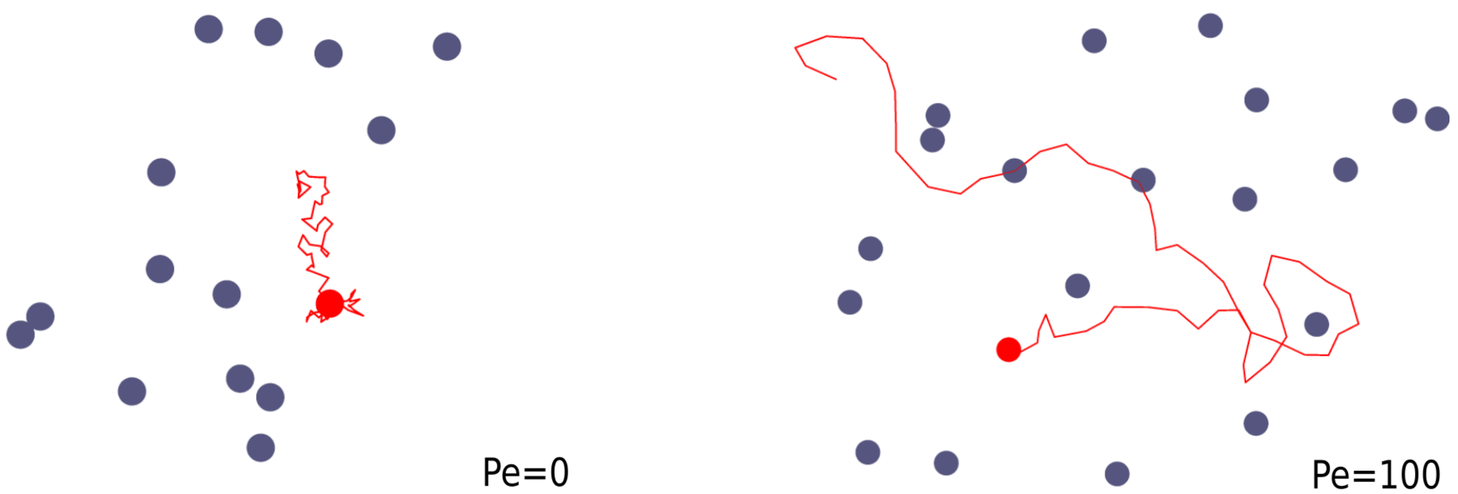
The units of length, time and energy are given by σ, $\tau_p = D_\theta^{-1}$ and ε

$D_\theta = 3k_BT/(\gamma\sigma^2)$ controls persistence, $\gamma/m = 10$ and $k_BT = 0.05$

Péclet number Pe = $F_{\rm act}\sigma/(k_BT)$ measures activity and $\phi = \pi\sigma^2 N/(4S)$

Active Brownian disks

The typical motion of particles in interaction



The active force induces a persistent random motion due to

$$\langle \mathbf{F}_{\rm act}(t) \cdot \mathbf{F}_{\rm act}(t') \rangle \propto F_{\rm act}^2 \, e^{-(t-t')/\tau_p}$$

with $\tau_p = D_\theta^{-1}$

Active Brownian disks

Questions – à la Statistical Physics

- Pe - ϕ Phase diagram
- Mechanisms for phase transitions.
  - Topological defects.
  - Dynamics across phase transitions.
- Motility Induced Phase Separation.

- Influence of particle shape, *e.g.* disks *vs.* dumbbells.

Active Brownian disks

Questions – à la Statistical Physics

- Pe - ϕ Phase diagram – start from solid and dilute
- Mechanisms for phase transitions.
  - Topological defects.
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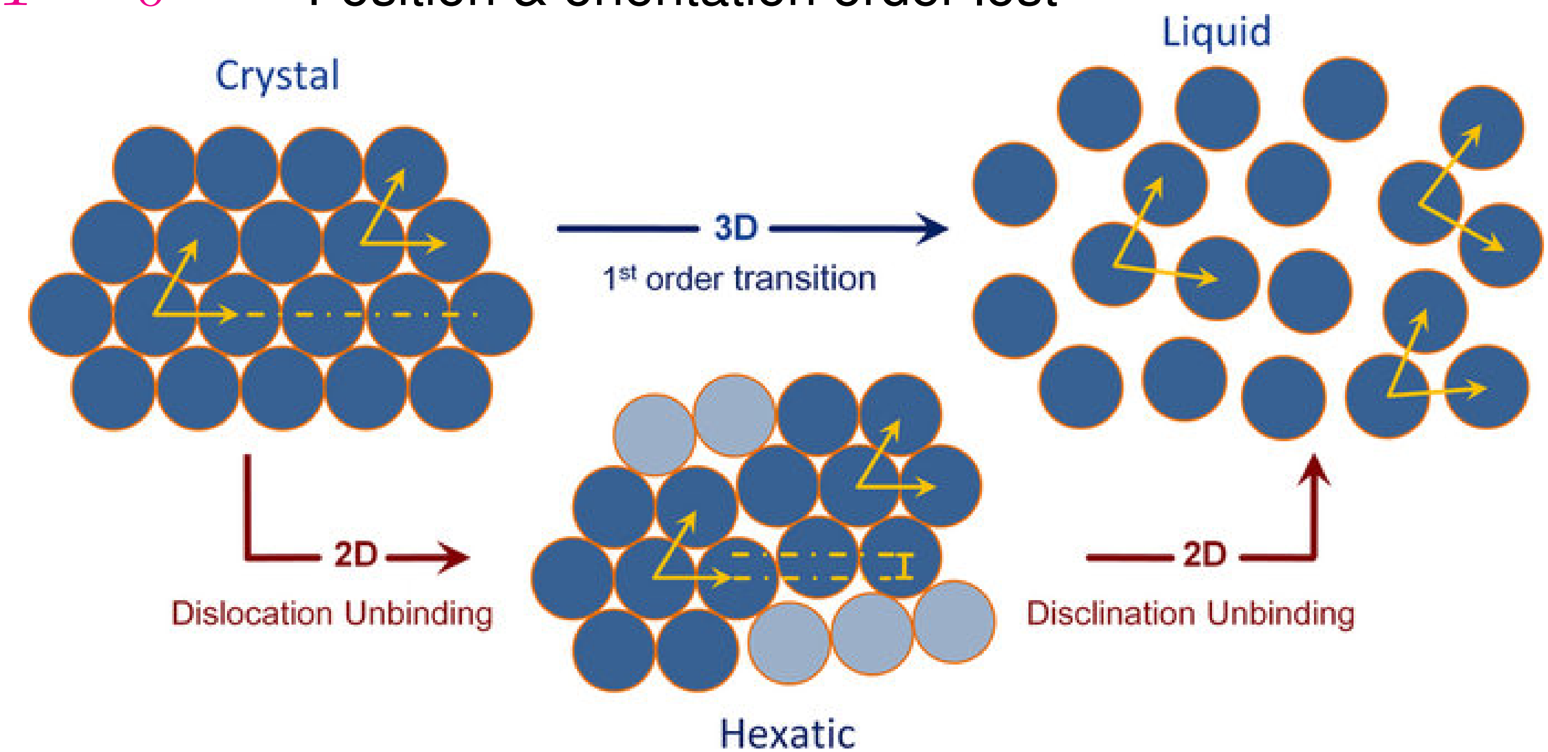
- Influence of particle shape, *e.g.* disks *vs.* dumbbells.

Freezing/Melting

Two step route in passive Pe = 0 $2d$ systems

$T = 0$ Position & orientation order lost

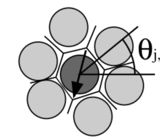


Orientation order preserved also lost

Image from **Pal, Kamal & Raghunathan, Sc. Rep. 6, 32313 (2016)**

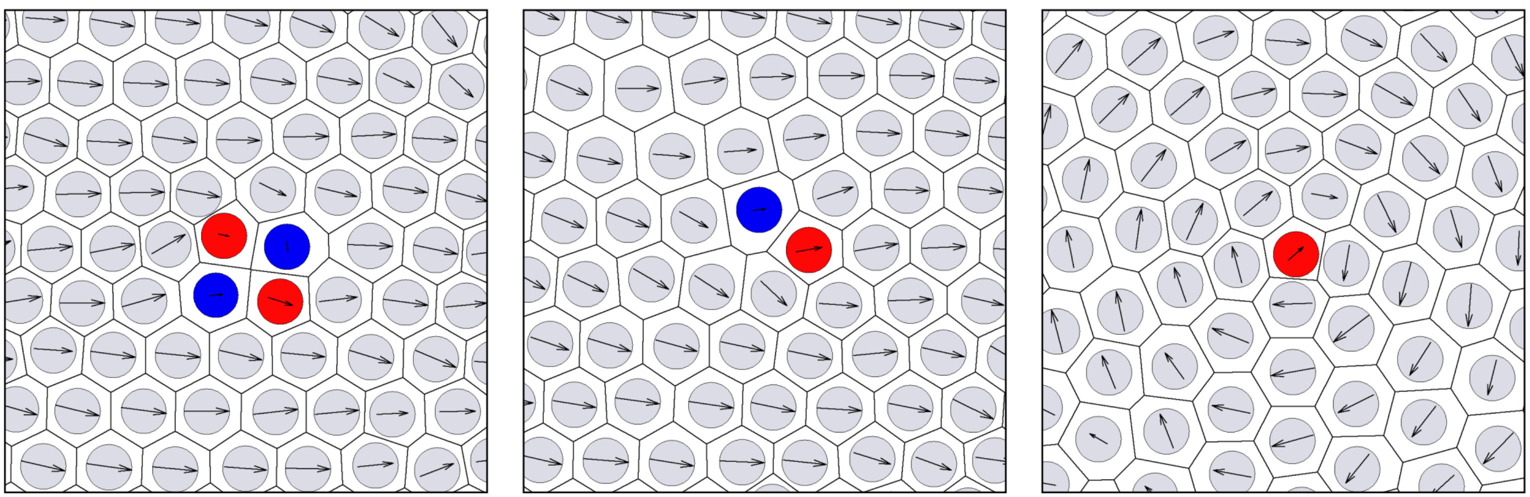
Freezing/Melting - arrows

Hexatic (orientational) order parameter $\psi_{6j} = \frac{1}{nn_j} \sum_{k=1}^{nn_j} e^{i6\theta_{jk}}$



ϕ ←

SOLID	HEXATIC	LIQUID
LR orient.	**QLR** orient.	**SR** orient.
QLR transl.	**SR** transl.	**SR** transl.



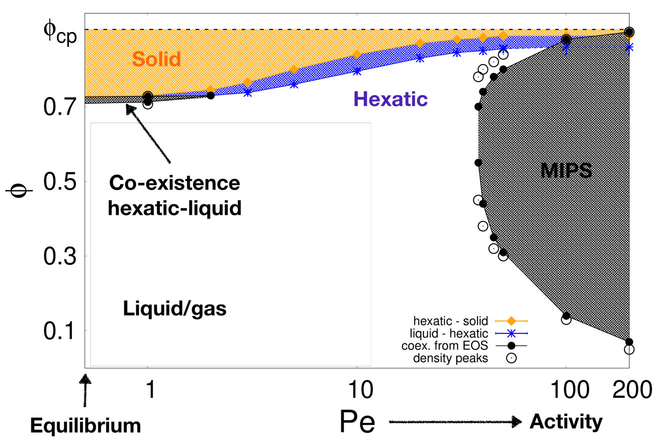
arrows oriented (LR) less oriented (QLR) order lost (SRL)

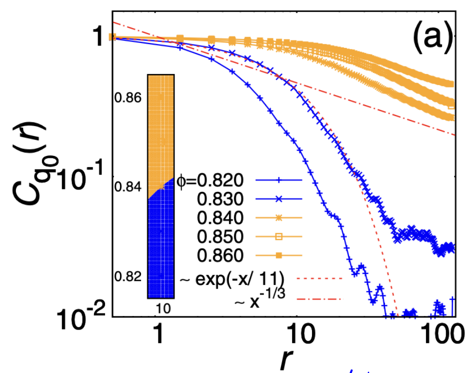
● five neighbours ● seven neighbours

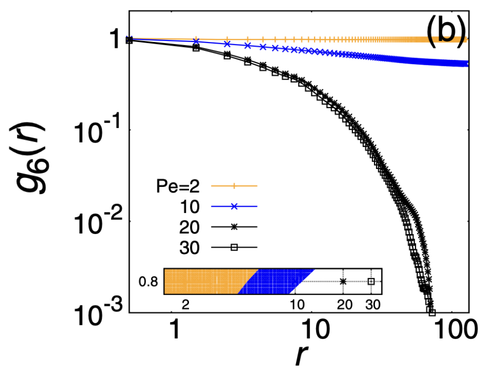
Voronoi tessellation

Phase Diagram

Solid, hexatic, liquid, co-existence and MIPS







$r^{-\eta}$ $e^{-r/\xi}$ ct $r^{-\eta_6}$ e^{-r/ξ_6}

First order **liquid** - **hexatic** transition & co-existence at low Pe from

- Pressure $P(\phi, \text{Pe})$ (EoS)
- Distributions of local densities ϕ_i and hexatic order parameter $|\psi_{6i}|$

Phases characterized by

- Translational correlations $C_{q_0}(r)$ & orientational order correlations $g_6(r)$

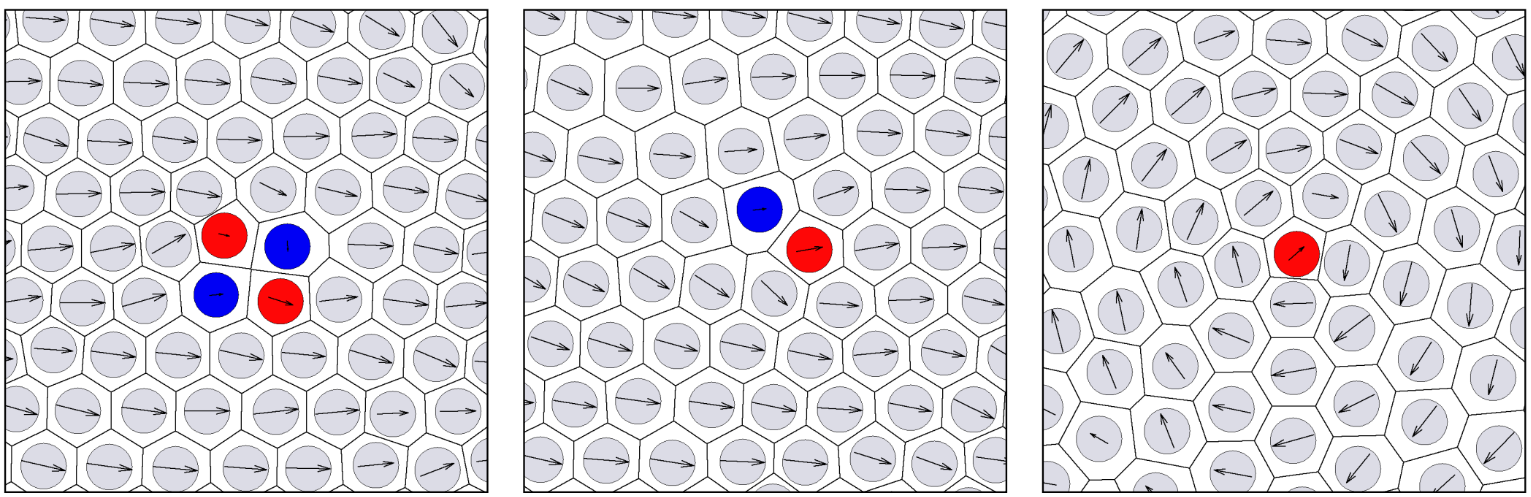
Digregorio, Levis, Suma, LFC, Gonnella & Pagonabarraga, PRL 121, 098003 (2018)

Freezing/Melting - defects

Mechanisms in $2d$ passive systems

SOLID	HEXATIC	LIQUID
LR orient.	**QLR** orient.	**SR** orient.
QLR transl.	**SR** transl.	**SR** transl.

ϕ ← (arrow pointing toward SOLID)



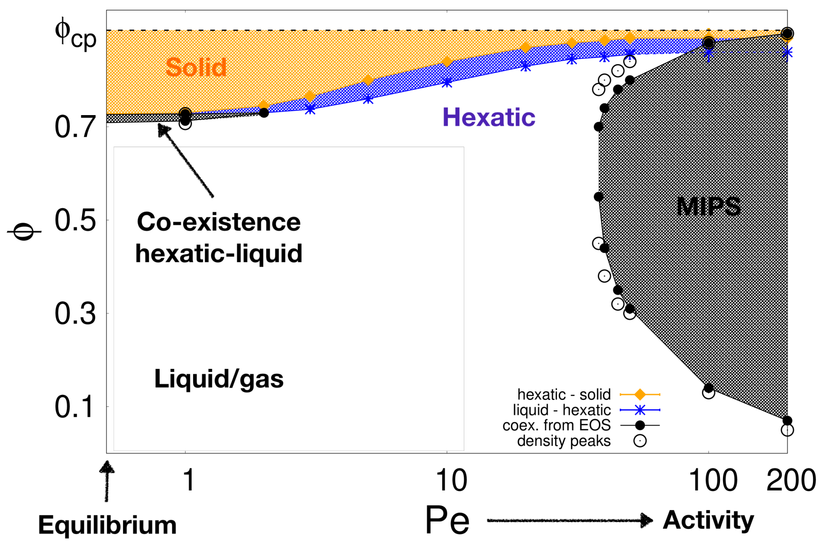
Confined dislocations	dislocations unbind	disclinations unbind

● five neighbours (red) ● seven neighbours (blue)

Voronoi tessellation

Active Brownian disks

Phase diagram with solid, hexatic, liquid, co-existence and MIPS



Different from BKTHN picture

1st order **hexatic-liquid** close to Pe = 0

KT-HNY **solid-hexatic** dislocation unbinding

disclination unbinding in liquid

percolation of defect clusters in liquid

Pressure $P(\phi, \text{Pe})$ (EOS), correlations $C_{q_0}(r)$, $g_6(r)$, and distributions of ϕ_i, $|\psi_{6i}|$

defect identification & counting

Digregorio, Levis, Suma, LFC, Gonnella & Pagonabarraga, PRL 121, 098003 (2018)

Digregorio, Levis, LFC, Gonnella & Pagonabarraga, Soft Matter 18, 566 (2022)

Topological defects

Summary of results

- **Solid** - **hexatic** à la BKT-HNY even quantitatively (ν value) and independently of the activity (Pe) *Universality*

- **Hexatic** - **liquid** very few disclinations and not even free

  Breakdown of the BKT-HNY picture for all Pe (even zero)

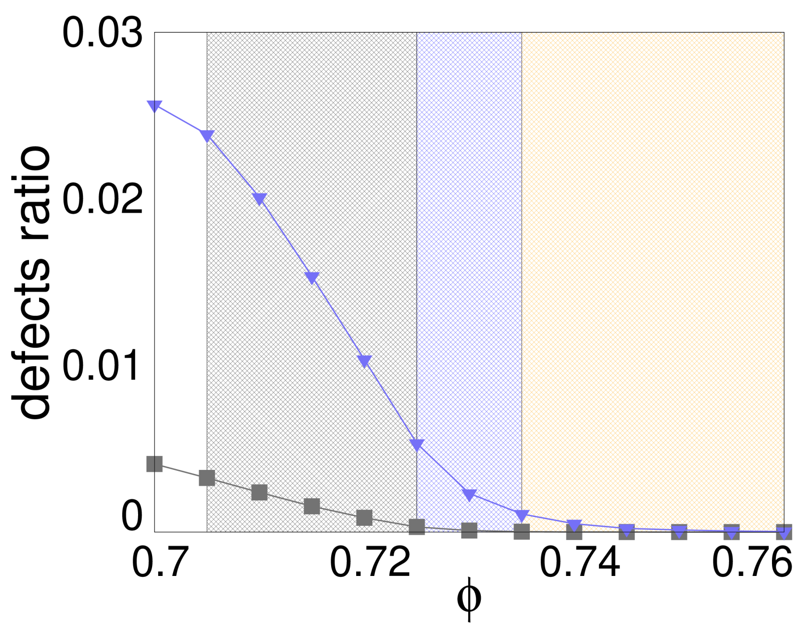
- Close to, but in the liquid, **percolation** of *clusters of defects* with properties of uncorrelated critical percolation ($d_{\rm f}, \tau$)

- In **MIPS**, network of defects on top of the interfaces between hexatically ordered regions, interrupted by the *gas bubbles in cavitation*

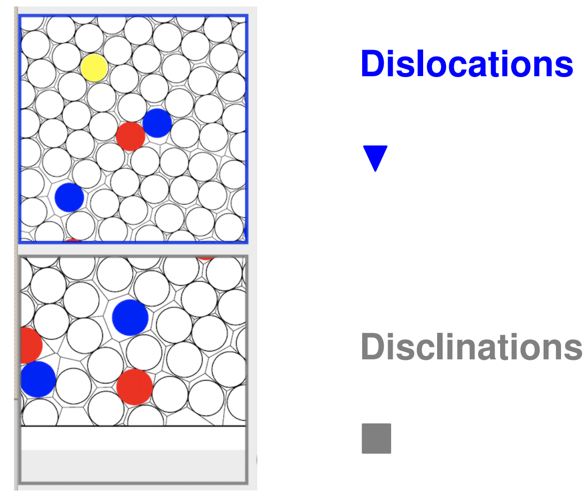
Digregorio, D Levis, LF Cugliandolo, G Gonnella, I Pagonabarraga, Soft Matter 18, 566 (2022)

Mechanisms

Unbinding of dislocations & disclinations ?



5 & 7 neighbors



Dislocations ▼ unbind at the **solid** - **hexatic** transition as in BKT-HNY theory

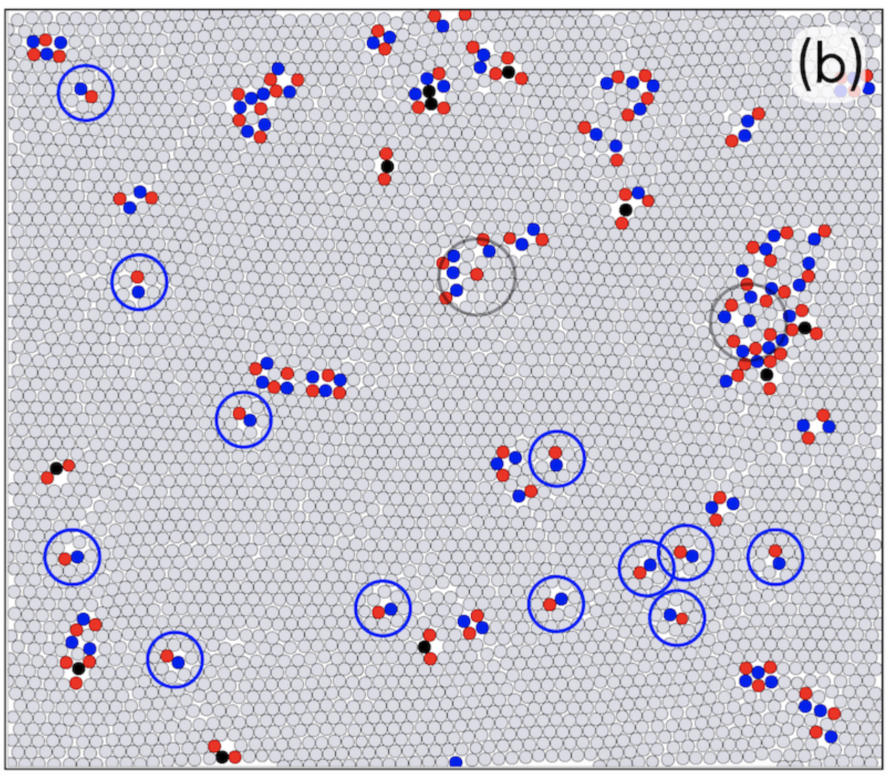
$$\rho_{dislocations} \sim a \ \exp\left[-b\left(\frac{\phi_c}{\phi_c-\phi}\right)^{\nu}\right] \qquad \nu \sim 0.37 \ \forall \text{ Pe}$$

Disclinations ■ unbind when the **liquid** appears in the co-existence region

Digregorio et al. Soft Matter 18, 566 (22) ; experiments Han, Ha, Alsayed & Yodh, PRE 77, 041406 (08)

Disclinations

At the hexatic - liquid transition ϕ_l at all Pe



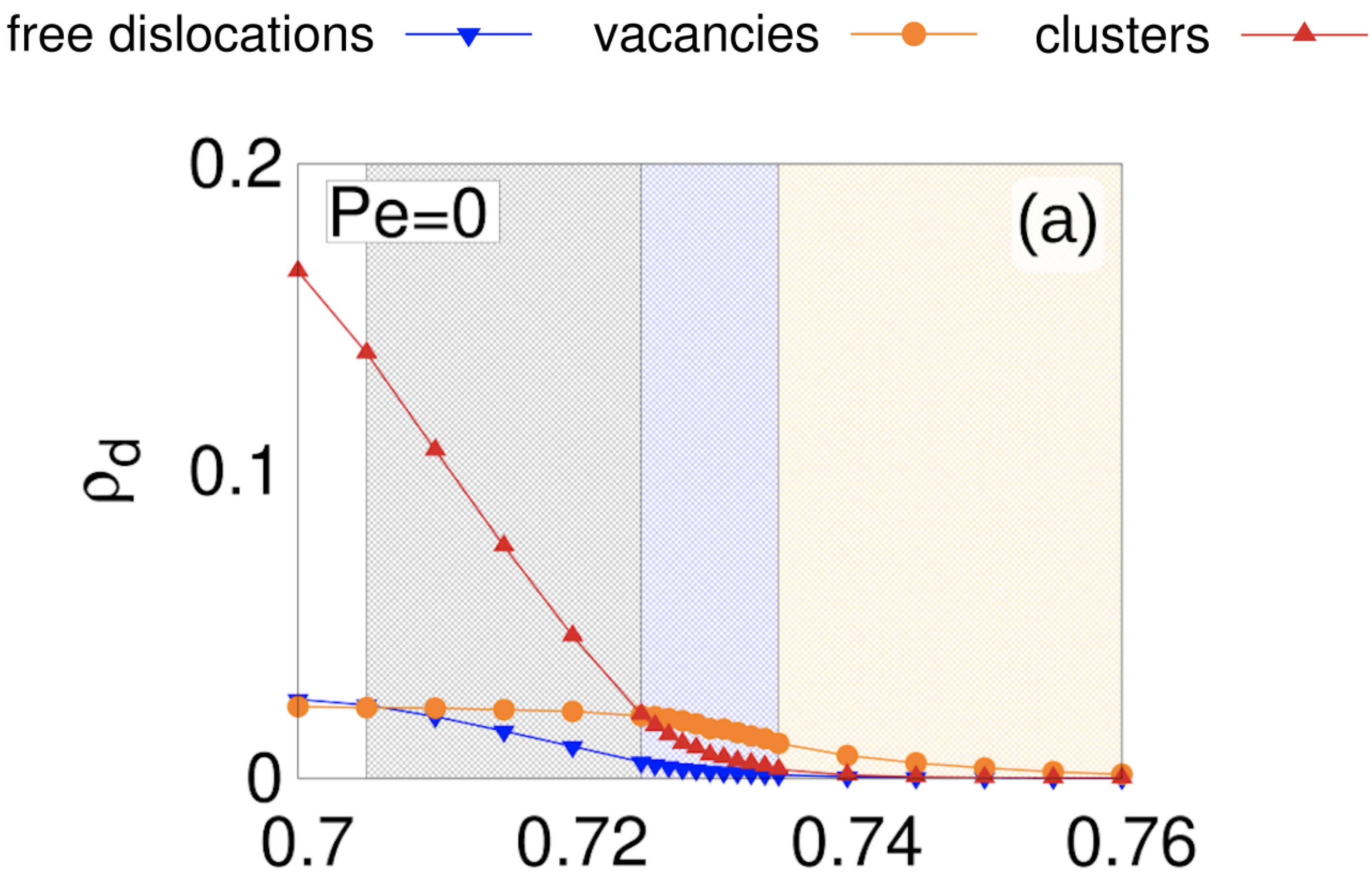
dislocations

disclinations

Very few disclinations, and always very close to other defects, so **not free**

Clusters

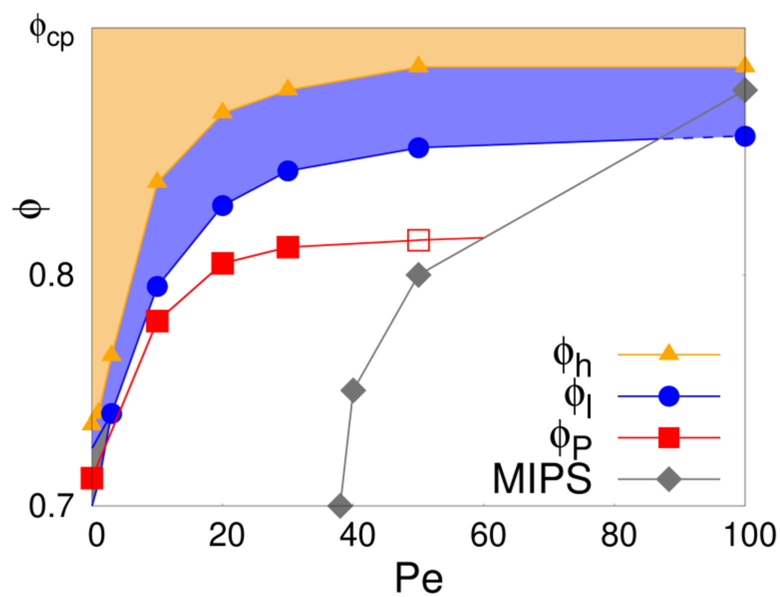
Close to the hexatic - liquid transition



As soon as the liquid appears in co-existence, **defects in clusters dominate**

Clusters

Percolation of defect clusters : the critical curve



Critical percolation with

fractal properties $d_{\rm f} \sim 1.9$ and

corresponding algebraic size distribution $\tau \sim 2.05$

With some coarse-graining the **percolation curve** moves upward towards the **hextic**-**liquid** critical one.

Do they coincide ?

Active Brownian disks

Questions – à la Statistical Physics

- Pe - ϕ Phase diagram.
- Mechanisms for phase transitions.
  - Topological defects.
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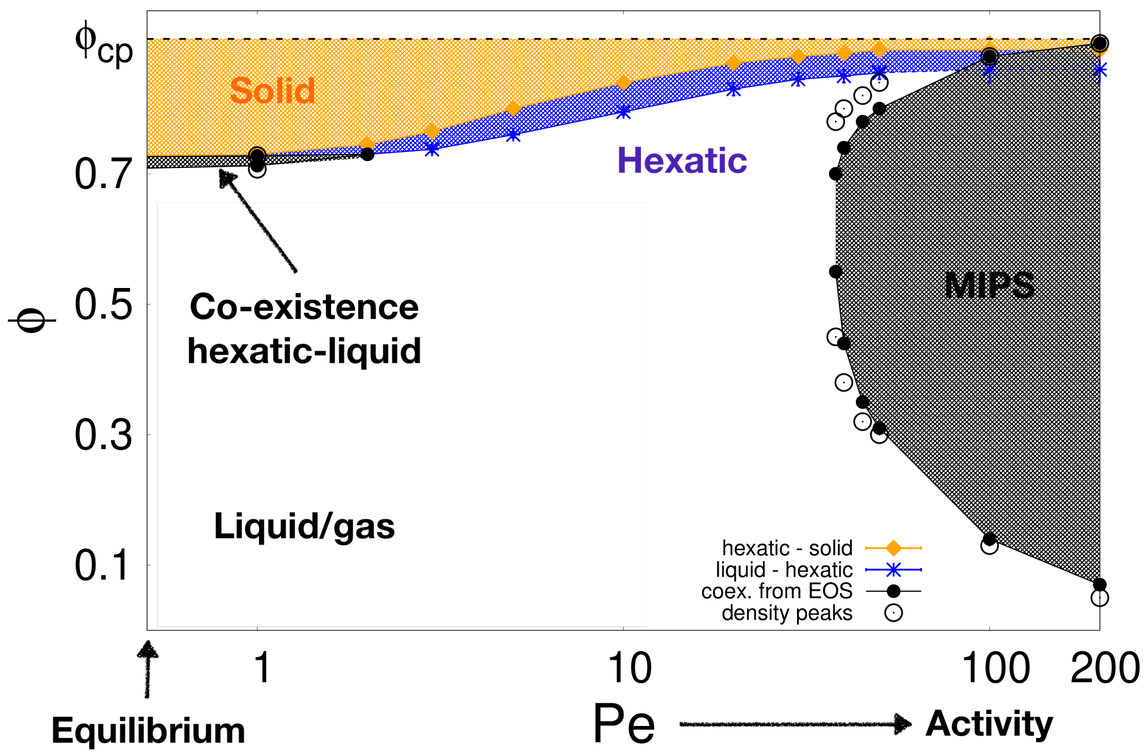
- Motility Induced Phase Separation.

- Influence of particle shape, *e.g.* disks *vs.* dumbbells.

Active Brownian disks

Phase diagram with solid, hexatic, liquid, co-existence and MIPS



Motility induced

phase separation (MIPS)

gas & dense

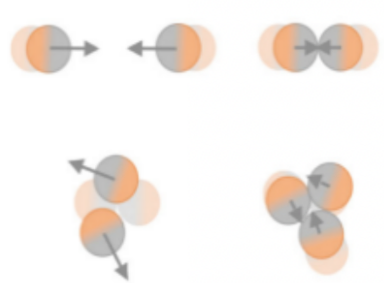
Cates & Tailleur
Ann. Rev. CM 6, 219 (2015)
Farage, Krinninger & Brader
PRE 91, 042310 (2015)

Pressure $P(\phi, \text{Pe})$ (EOS), correlations $G_T(r)$, $G_6(r)$, and distributions of ϕ_i, $|\psi_{6i}|$

Digregorio, Levis, Suma, LFC, Gonnella & Pagonabarraga, PRL 121, 098003 (2018)

Motility Induced Phase Separation

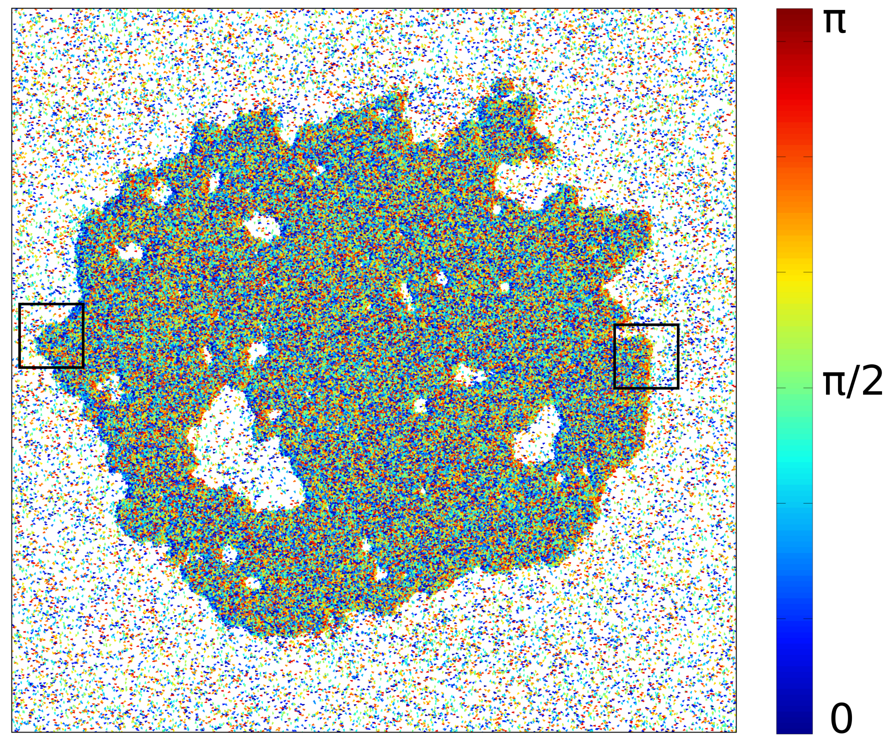
The basic mechanism



Particles collide heads-on

and cluster even in the

absence of attractive forces

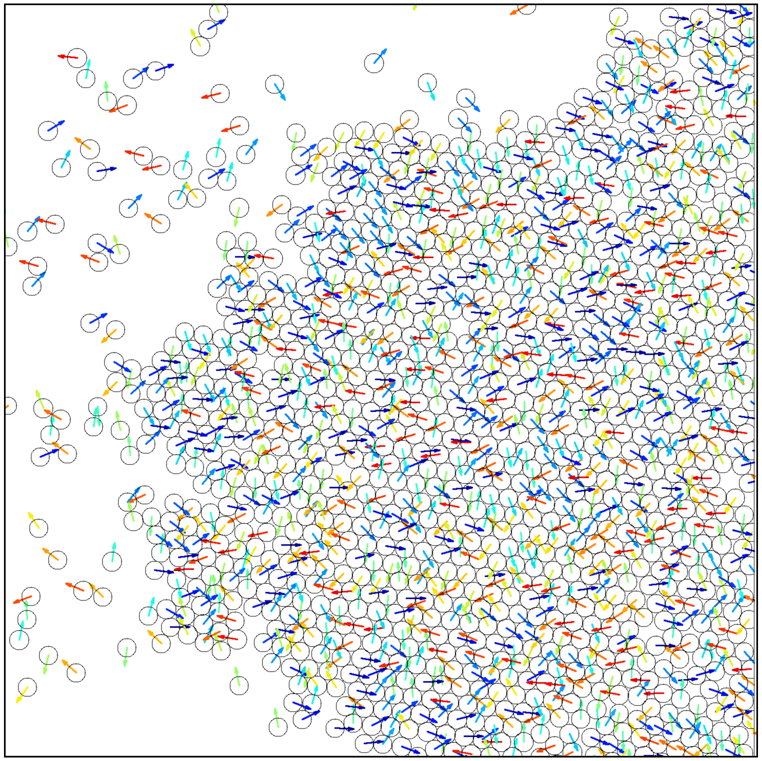


$\longrightarrow$ blue 0 **$\longleftarrow$ red π**

The colours indicate the direction along which the particles are pushed by the active force $\boldsymbol{F}_{\mathrm{act}}$

MIPS

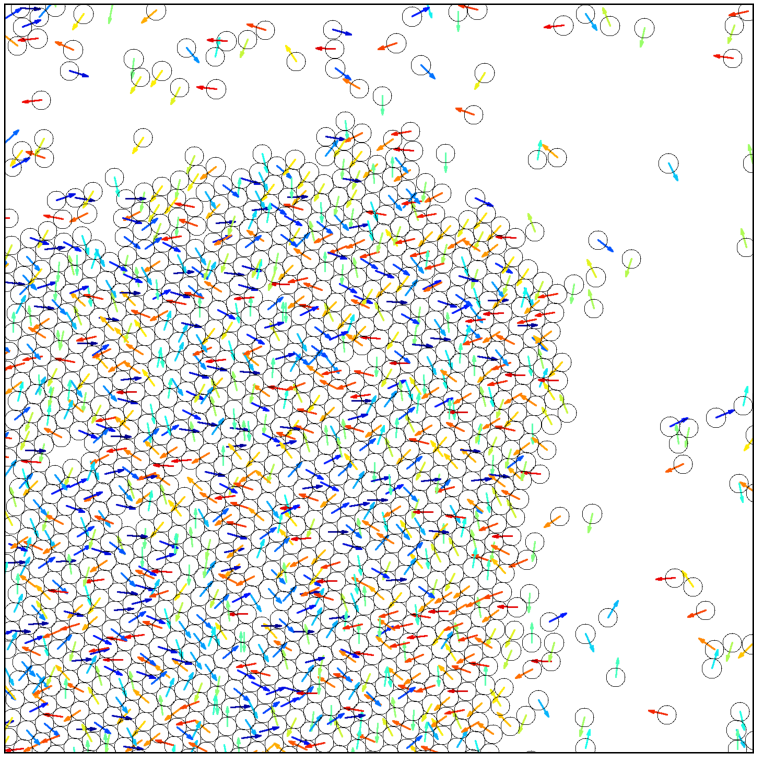
Particle orientation at the borders



Zoom over **left border** $\rightarrow 0$

MIPS

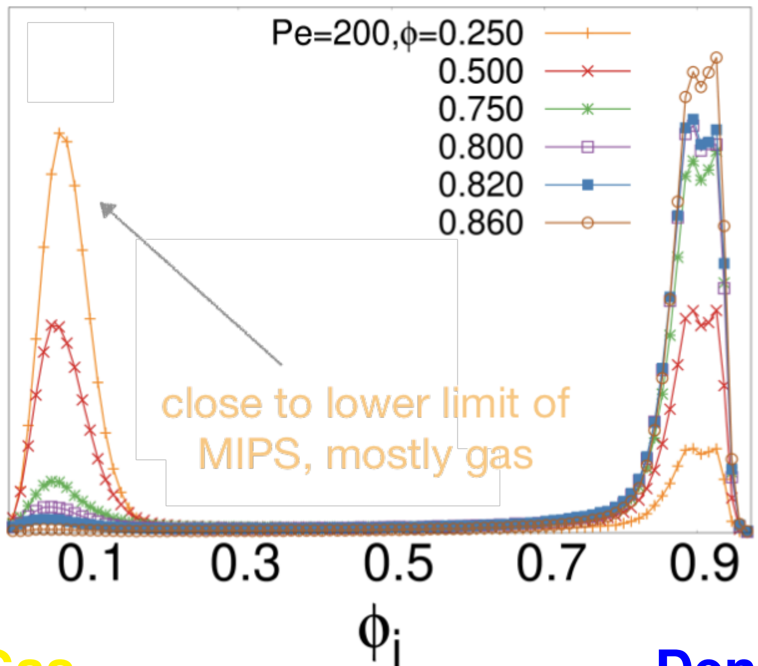
Particle orientation at the borders

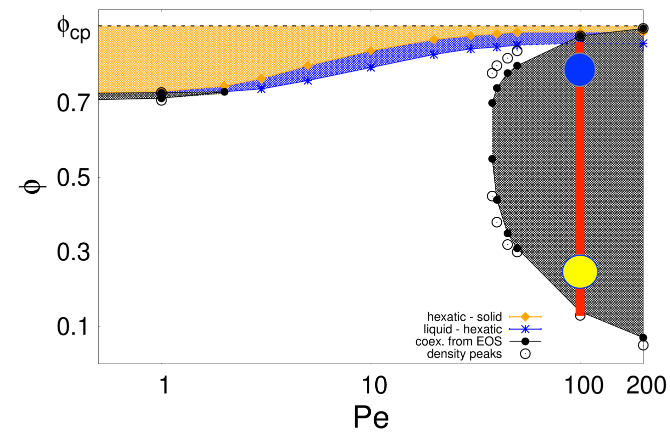


Zoom over **right border** $\leftarrow \pi$

MIPS

Local density distributions - dense & gas



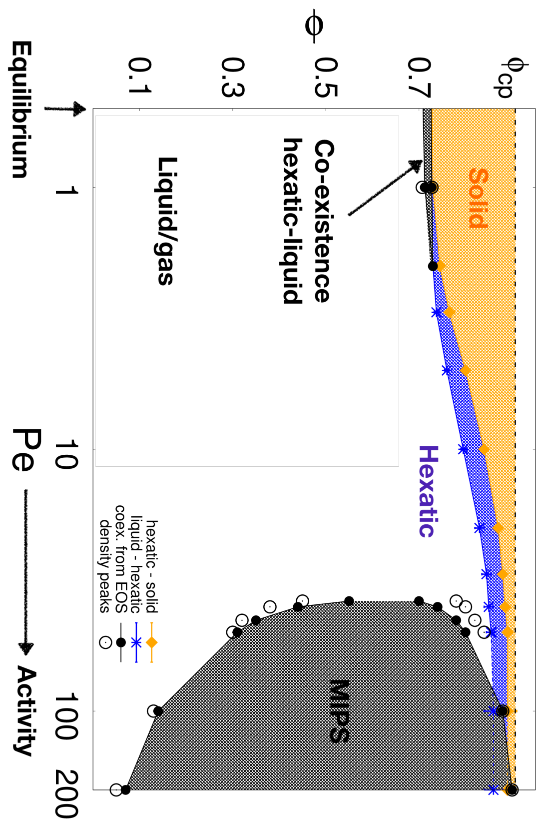


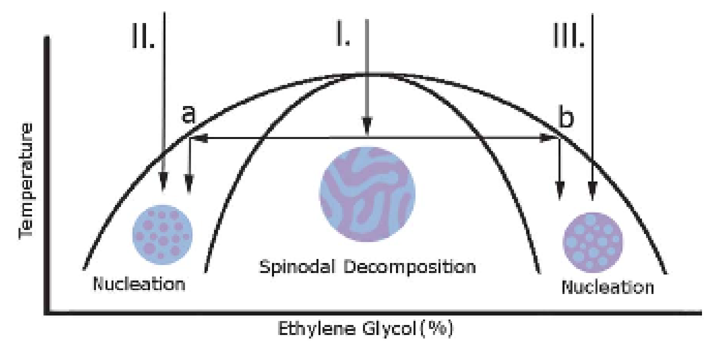
Gas Dense

The position of the peaks does not change while changing the global packing fraction ϕ but their relative height does. Transfer of mass from gas to **dense** component as ϕ increases

MIPS

Is it just a conventional phase separation ?



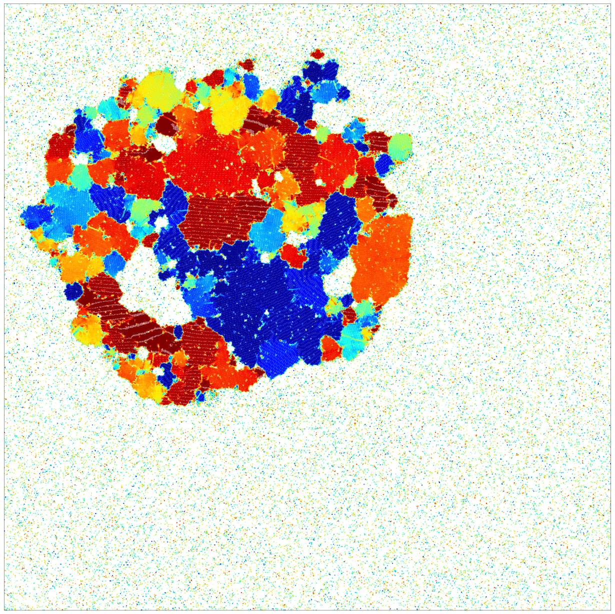


Similar to phase separation with percentage of system covered by dense and gas phases determined by a level rule ?

Cates & Tailleur 12

The dense phase

Hexatic patches, defects, bubbles



Dense/dilute separation[1]

For low packing fraction ϕ

a single round droplet

Growth[2] of clusters[3] with a mosaic

of hexatic orders[3] with

gas bubbles[2,4,5] & defects[6]

[1] **Cates & Tailleur, Annu. Rev. Cond. Matt. Phys. 6, 219 (2015)**

[2] **Caporusso, Digregorio, Levis, LFC & Gonnella, PRL 125, 178004 (2020)**

[3] **Caporusso, Digregorio, LFC, Gonnella, Levis & Suma, PRL 131, 068201 (2023)**

[4] **Tjhung, Nardini & Cates, PRX 8, 031080 (2018)**

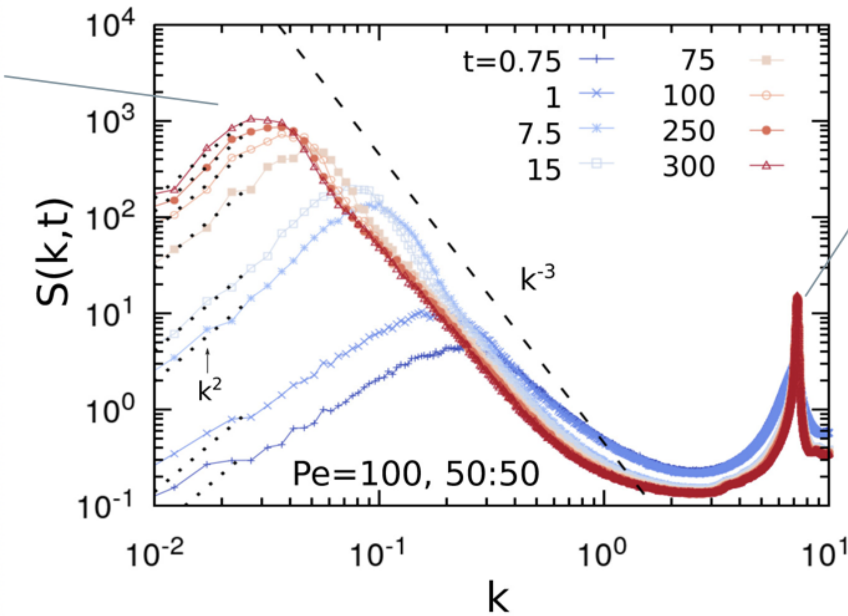
[5] **Shi, Fausti, Chaté, Nardini & Solon, PRL 125, 168001 (2020)**

[6] **Digregorio, Levis, LFC, Gonnella & Pagonabarraga, Soft Matter 18, 566 (2022)**

Structure

Dynamic structure factor ⇒ growing length of dense component

I peak:
system average
size



II peak:
particles' packing

$$S(\mathbf{k},t) = \sum_{i=1}^{N} e^{i\mathbf{k}(\mathbf{r_i}-\mathbf{r_j})}$$

related to Fourier transform of
the pair correlation function

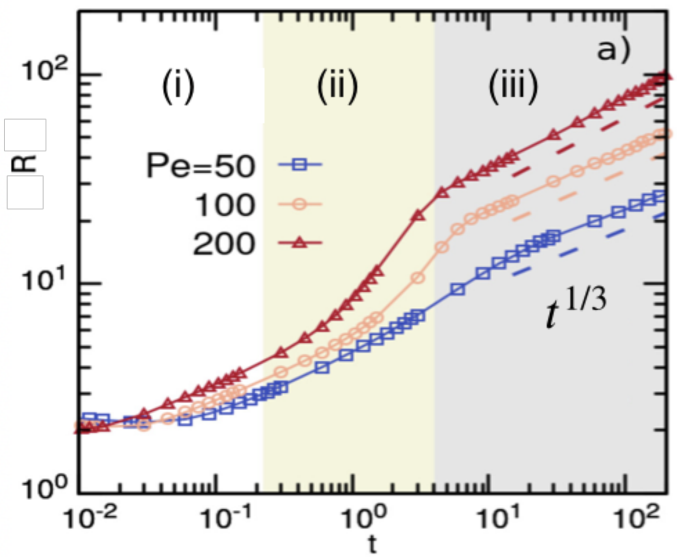
$$k_{\mathrm{I}}(t) \propto R^{-1}(t)$$

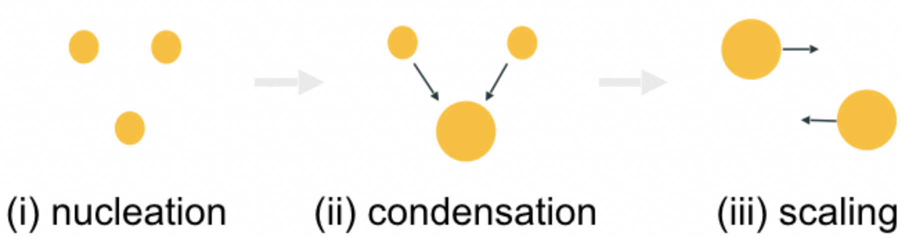
No sign of fractality here. Porod's law $S(k) \sim k^{-(d+1)}$ for compact domains with sharp interfaces

The growth law

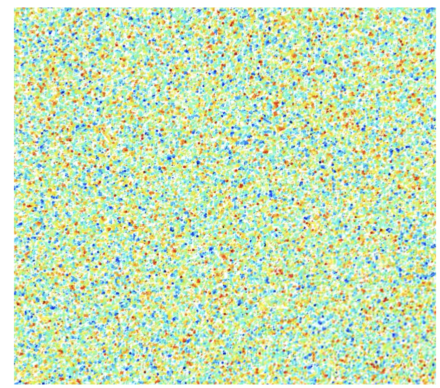
Growing length of the dense component and regimes

Different Pe





Movie



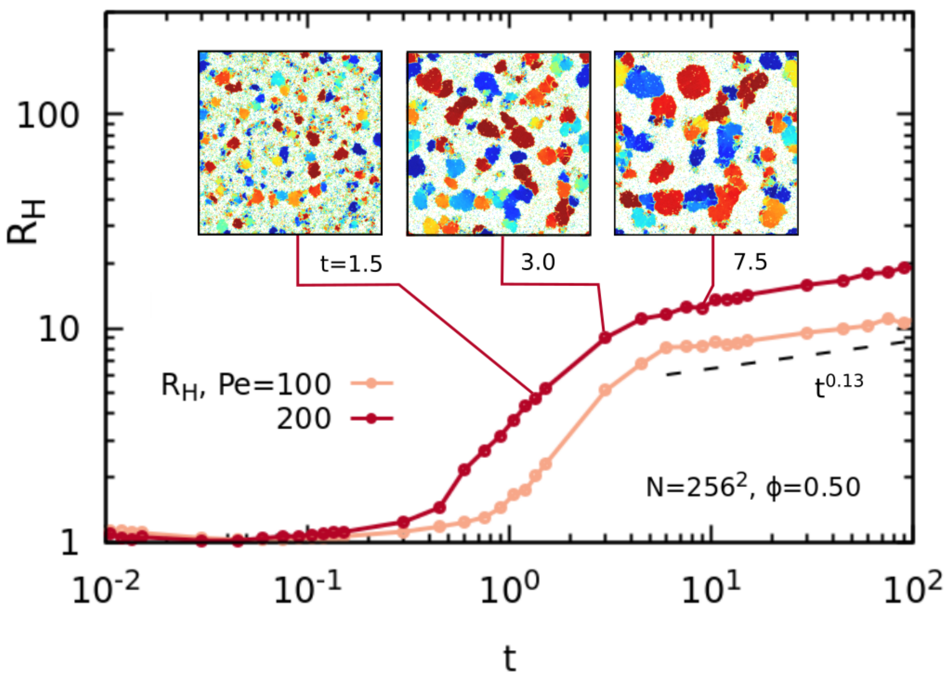
$R(t) \to aL$

In scaling regime $t^{1/3}$ like in **Lifshitz-Slyozov-Wagner**, scalar phase separation.

More about it & asymptotic value later

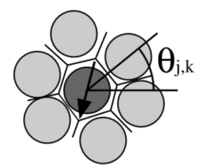
Local hexatic order

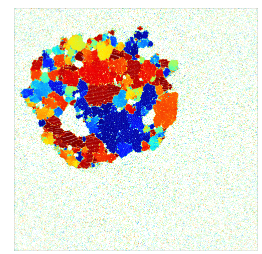
Growing length of the orientational order – regimes



Local hexatic order parameter

$$\psi_{6j} = \frac{1}{nn_j} \sum_{k=1}^{nn_j} e^{i6\theta_{jk}}$$





Full hexatically ordered small clusters

Larger clusters with several orientational order within

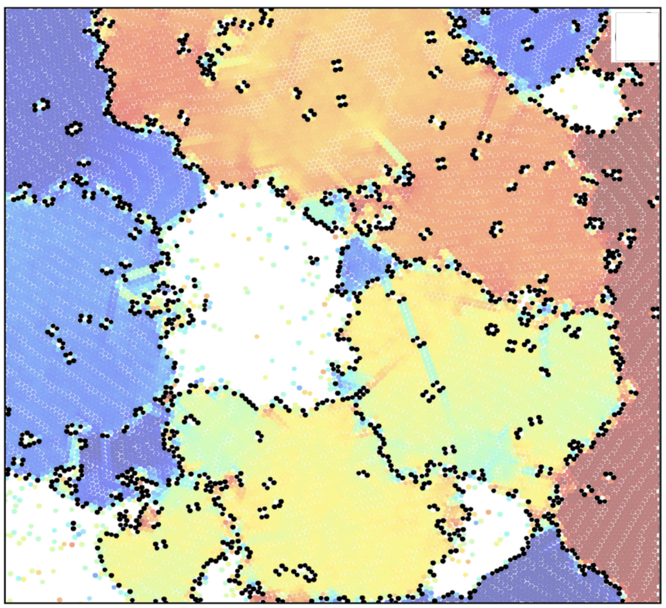
$R_H \sim t^{0.13}$ in the scaling regime and $R_H \to R_H^s \ll L$

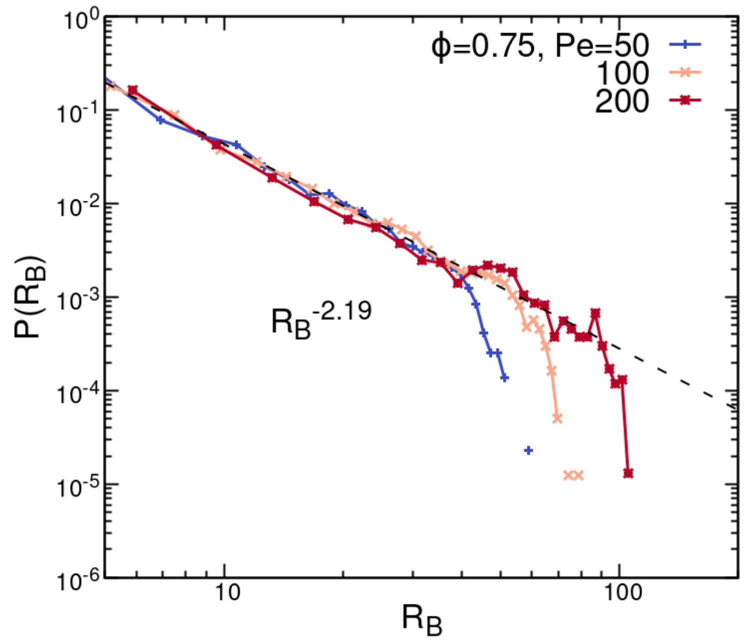
Similar to pattern formation, e.g.

Vega, Harrison, Angelescu, Trawick, Huse, Chaikin & Register, PRE 71 061803 (2005)

Bubbles in cavitation

At the internal interfaces bubbles pop up





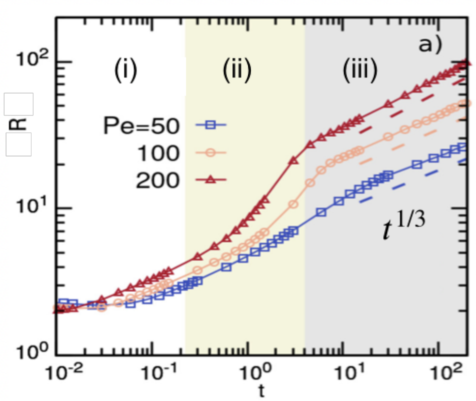
Bubbles appear and disappear at the interfaces between hexatic patches

Algebraic distribution of bubble sizes with a Pe-dependent exponential cut-off

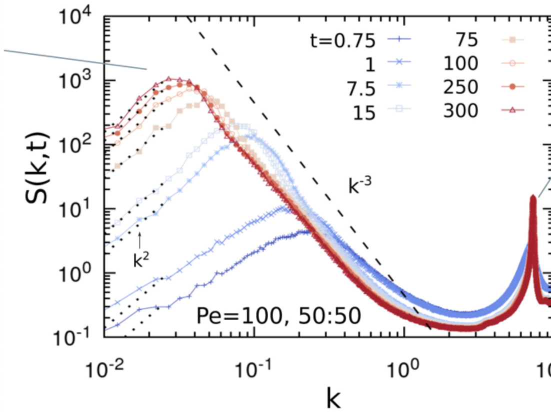
Growth of the dense phase

Scaling of the structure factor and growth regimes

Different Pe



I peak: system average size



II peak: particles' packing

$$S(\mathbf{k},t) = \sum_{i=1}^{N} e^{i\mathbf{k}(\mathbf{r}_i-\mathbf{r}_j)}$$

related to Fourier transform of the pair correlation function

$$S(k,t) \sim R(t)^d \, f(kR(t))$$

$$R(t) \sim t^{1/3}$$

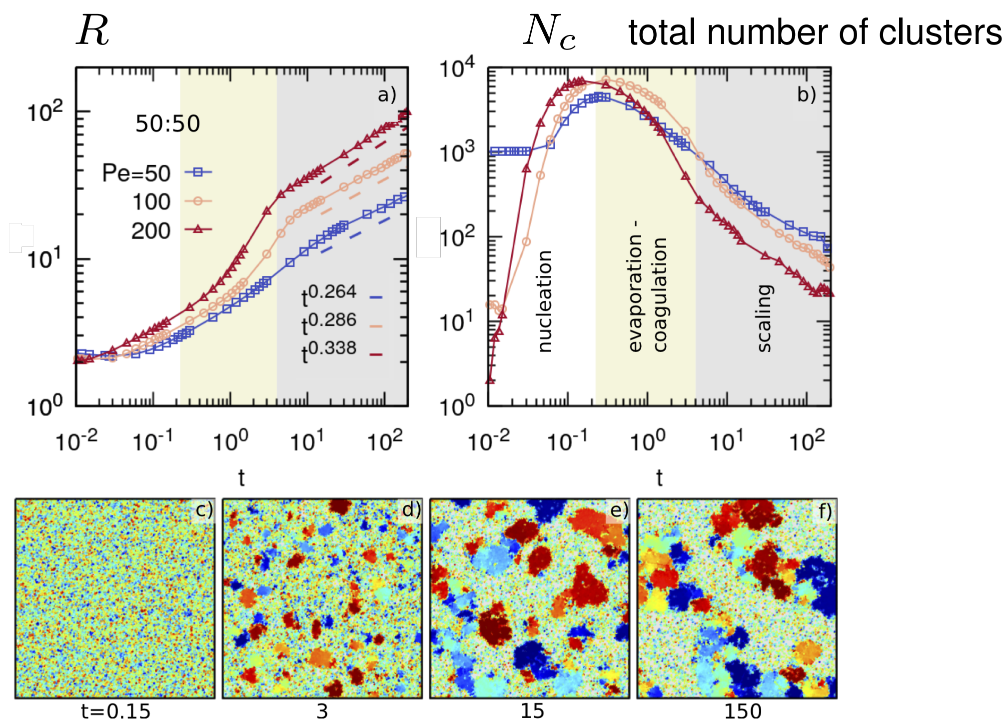
In the **scaling regime** $t^{1/3}$ like in **Lifshitz-Slyozov-Wagner**, scalar phase separation

Ostwald ripening small cluster evaporate and large ones capture gas particles

but is it just that ?

Growth of the dense phase

Focus on the clusters



Nucleation Evap/coag Scaling

On the averaged scaling regime:

Redner, Hagan & Baskaran, PRL 110, 055701 (2013)

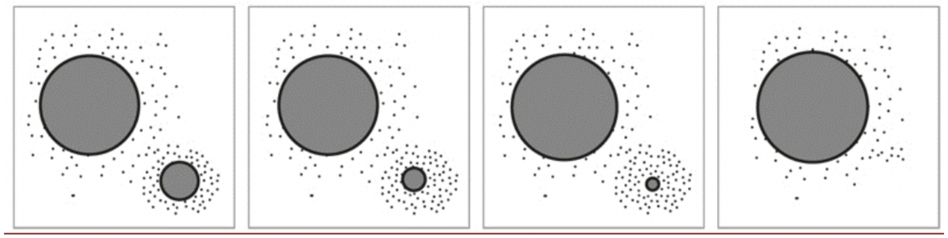
Stenhammar, Marenduzzo, Allen & Cates, Soft Matter 10, 1489 (2014)

Caporusso, Digregorio, Levis, LFC & Gonnella, PRL 125, 178004 (2020)

Beyond?

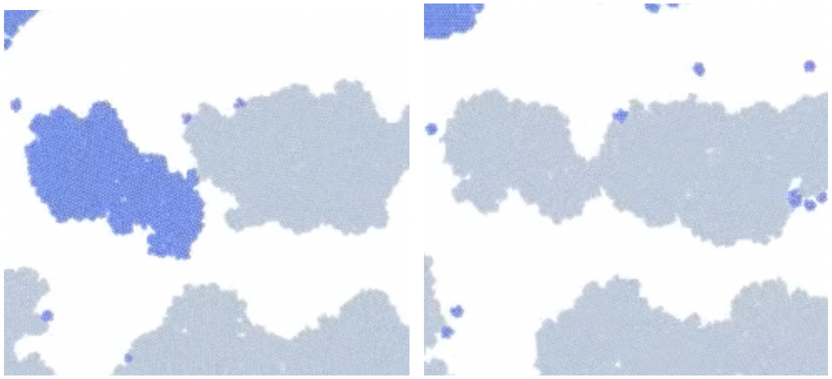
Goals :

1. Is it like the one undergone by a system of **passive attractive particles** ?



Ostwald ripening

2. Other **mechanisms** for the growth process ?

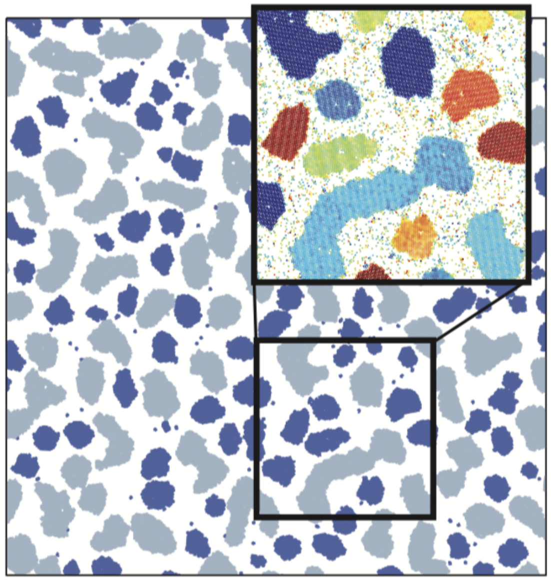


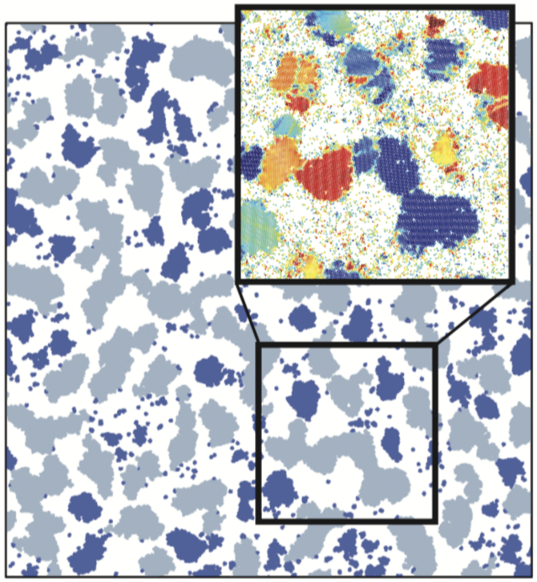
Cluster-cluster aggregation

Dense clusters

Instantaneous configurations (DBSCAN)

Passive / **Active**





The Mie potential is not truncated in the passive case $\Rightarrow$ attractive

Parameters are such that $R(t)$ is the same

Colors in the zoomed box indicate orientational order

Caporusso, LFC, Digregorio, Gonnella, Levis & Suma, PRL 131, 068201 (2023)

Dense clusters

Visual facts about the instantaneous configurations

Similarities

- Large variety of shapes and sizes (masses)

  Co-existence of

  small regular (**dark blue**) and large elongated (**gray**) clusters

Differences

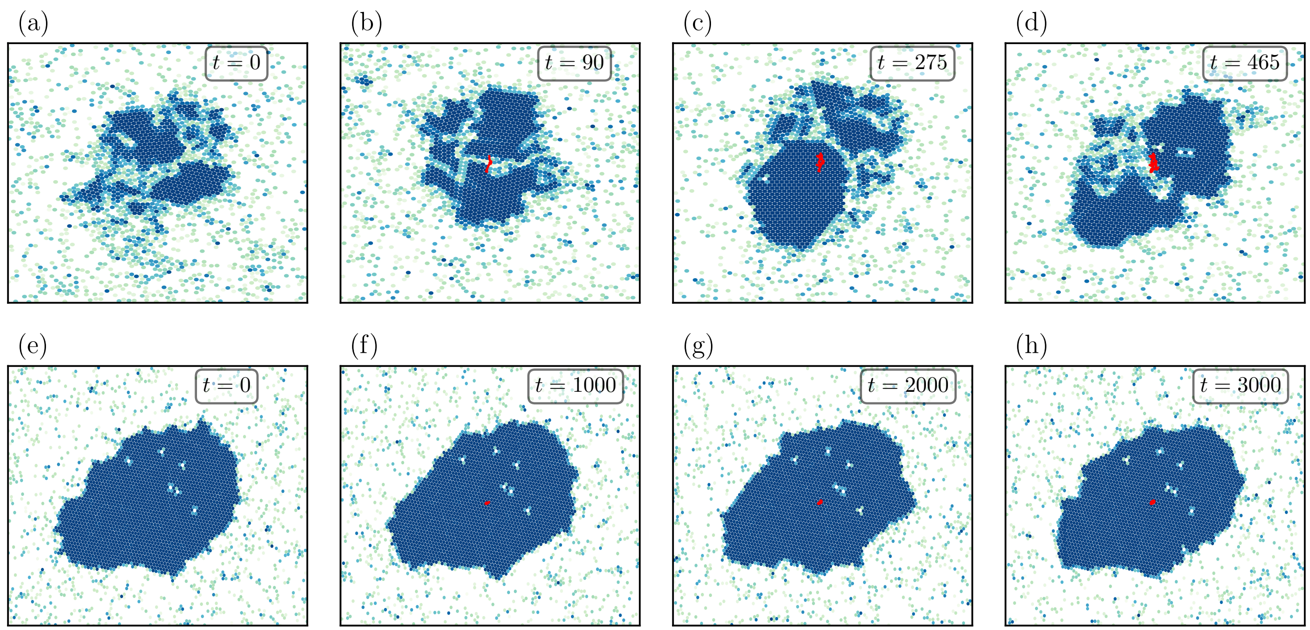
- Rougher interfaces in active
- Homogeneous (passive) vs. heterogeneous (active) orientational order within the clusters

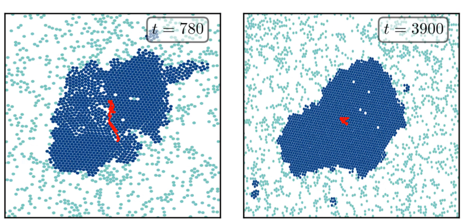
Cluster dynamics

Tracking of individual cluster motion - video

Active

Passive





In **red** the center of mass trajectory

Active is much faster than passive

Dense clusters

Visual facts about the cluster dynamics

In both cases, **Ostwald ripening** features

- small clusters evaporate
- gas particles attach to large clusters

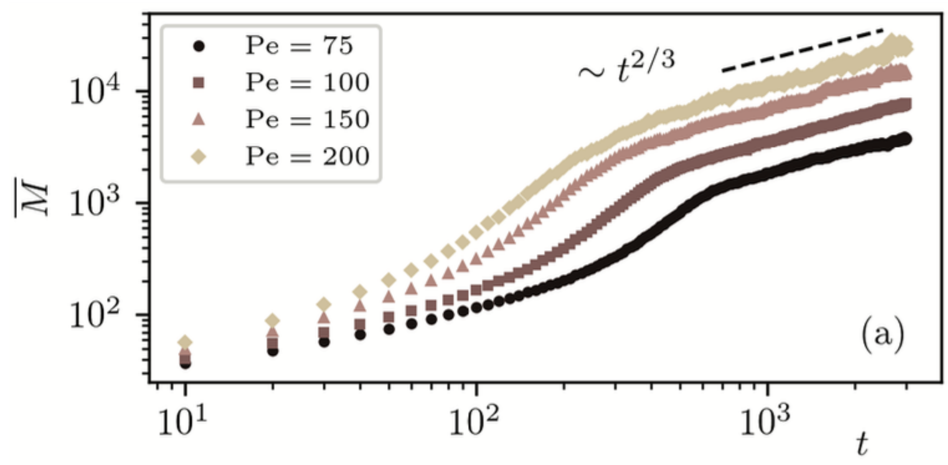
In the **active system**

- clusters displace much more & sometimes aggregate
- they also break & recombine

like in **diffusion limited cluster-cluster aggregation**

Dense clusters

Averaged mass $\overline{M} \equiv N_c^{-1}(t) \sum_{\alpha=1}^{N_c(t)} M_\alpha(t) \sim t^{2/3}$



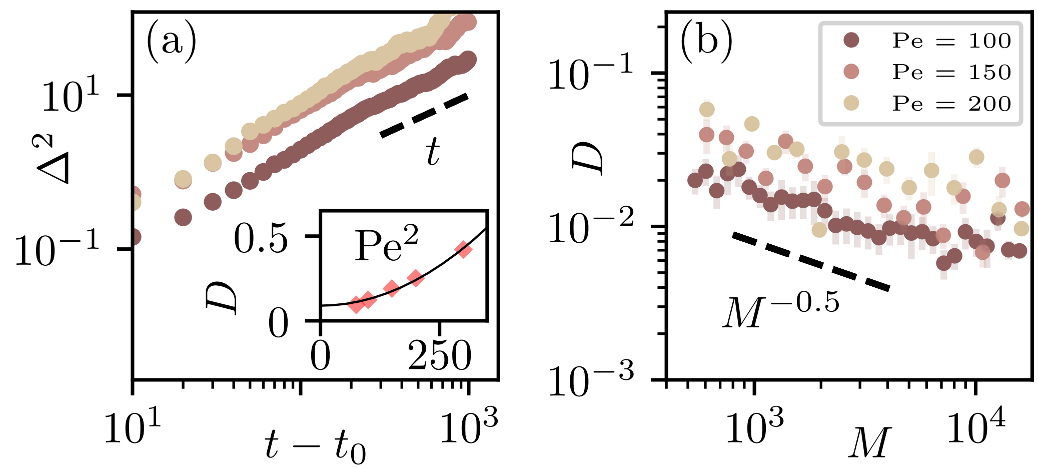
Same three regimes as in R from the structure factor

Clusters' dynamics origin ?

Active cluster evolution

Mean Square Displacement : diffusion

Average over all clusters **Mass dependence**



$$\Delta_k^2(t, t_0) = [\mathbf{r}^{(k)}_{\rm c.o.m.}(t) - \mathbf{r}^{(k)}_{\rm c.o.m.}(t_0)]^2 \sim 2d\, D(M_k, {\rm Pe})\,(t - t_0)$$

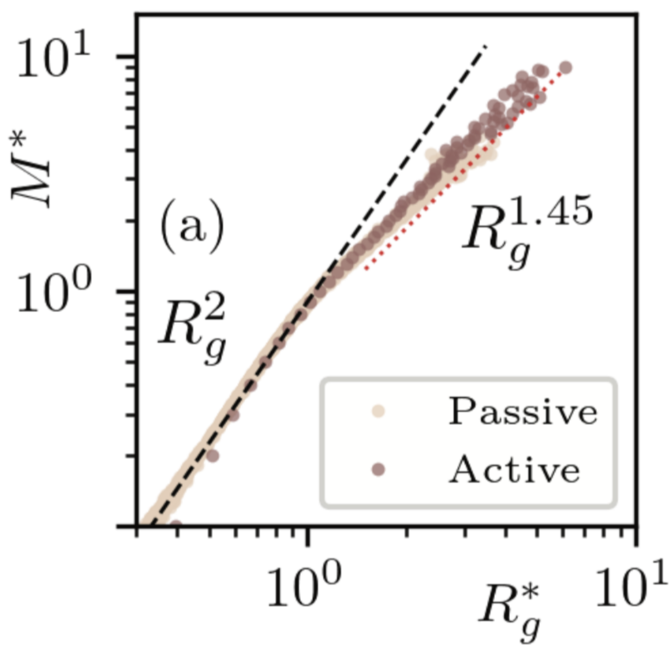
A sum of random forces yields $D \sim M^{-1}$

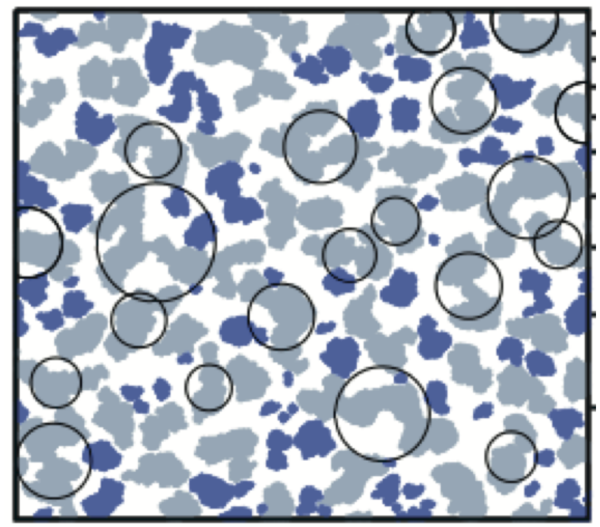
Passive tracer in a dilute active bath $D \sim R^{-1} \sim M^{-1/2}$ Solon & Horowitz (22)

Passive & very heavy isolated active clusters behave as $D \sim M^{-1}$

Geometry

Scatter plots : small regular – large fractal





Cluster mass $M^*(t) = \dfrac{M_k(t)}{\overline{M}(t)}$ Gyration radius $R_g^*(t) = \dfrac{R_{g\,k}(t)}{\overline{R_g}(t)}$

Data sampled in the scaling regime $t = 10^3 - 10^5$ every 10^3 time steps

$\overline{M}(t) = \dfrac{1}{N_c(t)} \sum_{k=1}^{N_c(t)} M_k(t)$ and $N_c(t)$ the total number of clusters at time t

Cluster-cluster aggregation

Extended Smoluchowski argument

From $\overline{R}_g \sim t^{1/z}$ and using $D(M) \sim M^{-\alpha}$

Smoluchowski eq. $\Rightarrow z = d_f(1+\alpha) - (d - d_w)$

Regular clusters $M < \overline{M}$

$d_f = d = d_w = 2$

$\alpha = 0.5$

$z = 2(1+0.5) = 3$

Fractal clusters $M > \overline{M}$

$d_f = 1.45, d = 2$ and $d_w \sim 2$

$\alpha = 0.5$ in the bulk

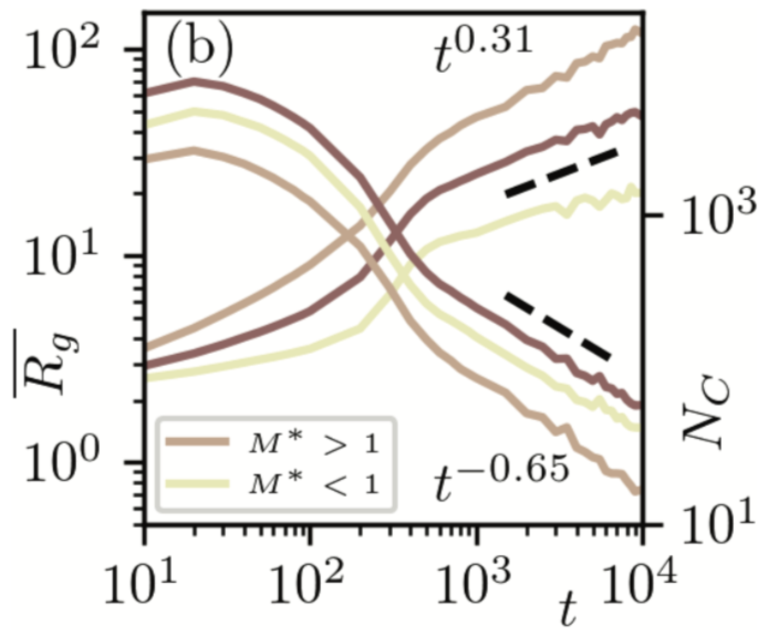
$z = 1.45(1+0.5) = 2.18 < 3$

Reviews on the application of fractals to colloidal aggregation

R. Jullien, Croatia Chemica Acta 65, 215 (1992) P. Meakin, Physica Scripta 46, 295 (1992)

Regular vs fractal clusters

Radius of gyration and number



regular $z \gtrsim 3$ | fractal $z < 3$ | average $z = 1/0.31 \sim 3$

More | Less | All

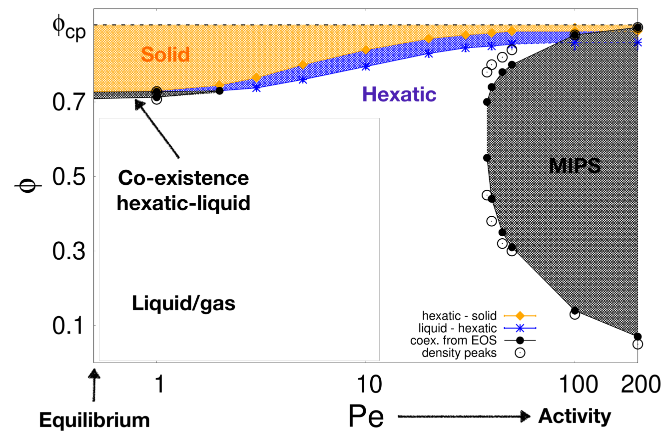
Dominate

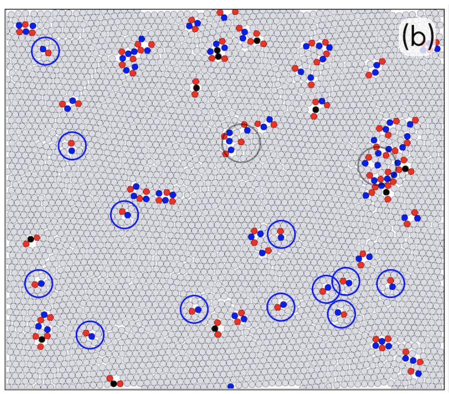
Results I

We established the full

phase diagram of ABPs

solid, **hexatic**, **liquid** & **MIPS**





We clarified the role played by point-like

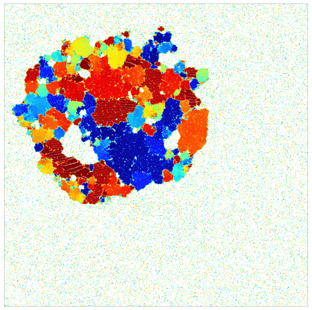
(**dislocations** & **disclinations**)

and **clustered** defects in

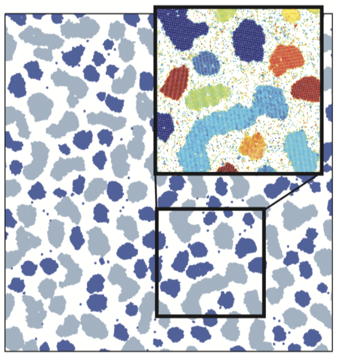
passive & active $2d$ models.

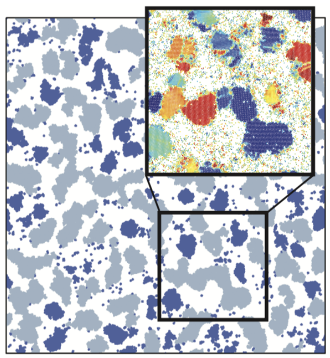
In **MIPS**

Micro *vs.* macro : hexatic patches & bubbles



Results II





Difference between

Passive

Active

growth

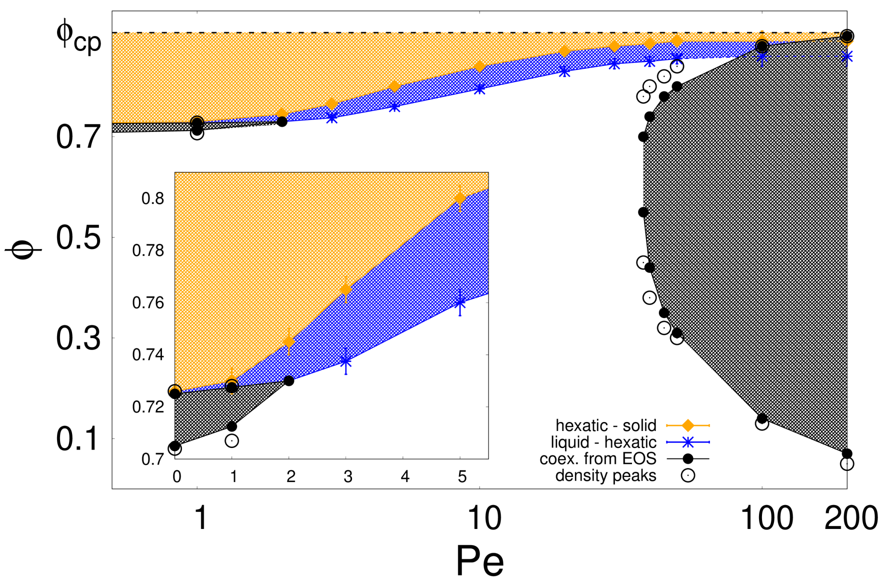
Ostwald ripening & cluster-cluster diffusive aggregation in active case
cluster-cluster aggregation almost not present in passive

Co-existence of regular and fractal clusters

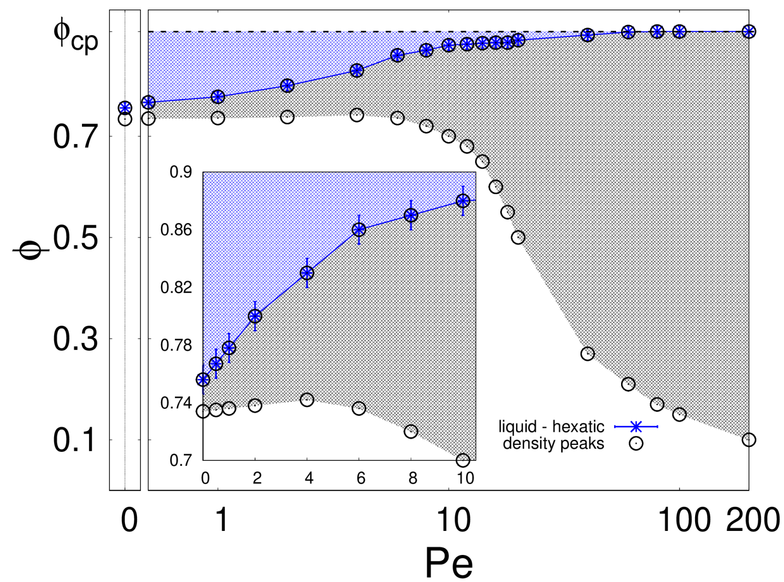
Heterogeneous orientational order in large active clusters

Beyond disks

Phase diagrams & plenty of interesting facts



Disks



Dumbbells

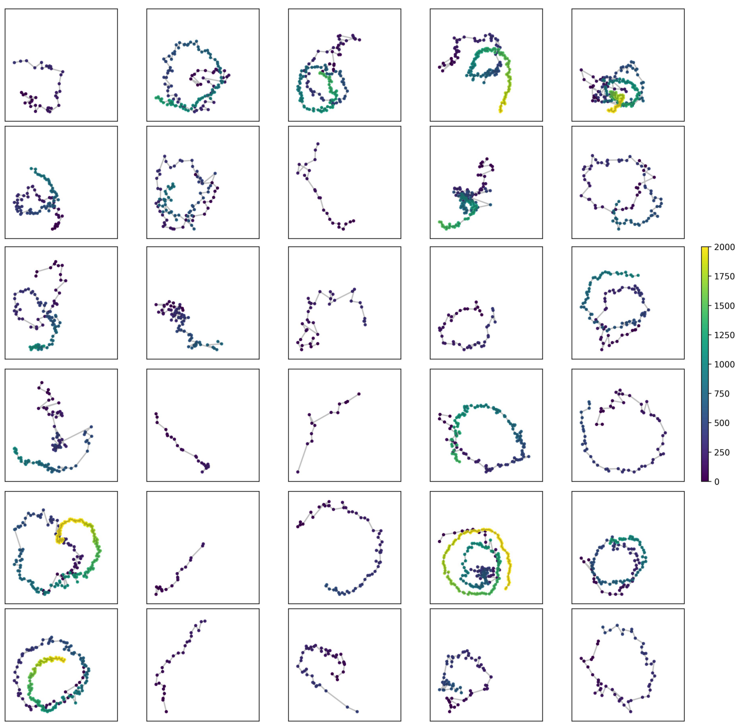
LFC, Digregorio, Gonnella & Suma, Phys. Rev. Lett. 119, 268002 (2017)

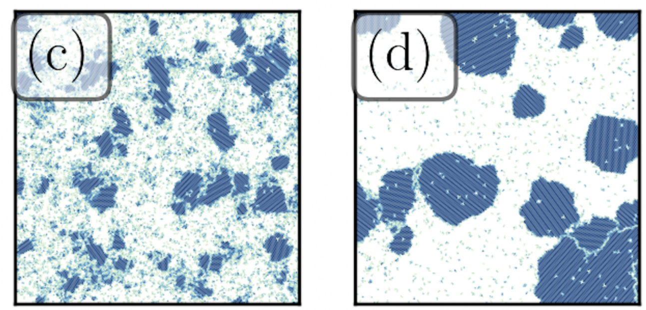
Dumbbell clusters

Trajectories

$$r = M R_g \frac{F^{\perp}_{\rm act}}{T_{\rm act}}$$

The radius of the c.o.m. trajectory





Non-vanishing :

active torque $T_{\rm act}$ & force $F_{\rm act}$

Rotation instead of ABP diffusion

Video

Caporusso, Negro, Suma, Digregorio, Carenza, Digregorio, Gonnella & LFC, Soft Matter (2024)

Caporusso, LFC, Digregorio, Gonnella & Suma, in preparation

Extras

Cluster-cluster aggregation

Extended Smoluchowski argument

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Smoluchowski eq. $\Rightarrow z = d_f(1+\alpha) - (d - d_w)$

Regular clusters $M < \overline{M}$

$d_f = d = d_w = 2$

$\alpha = 0.5$

$z = 2(1 + 0.5) = 3$

Fractal clusters $M > \overline{M}$

$d_f = 1.45$, $d = 2$ and $d_w \sim 2$

if, instead, $\boxed{\alpha = 1}$

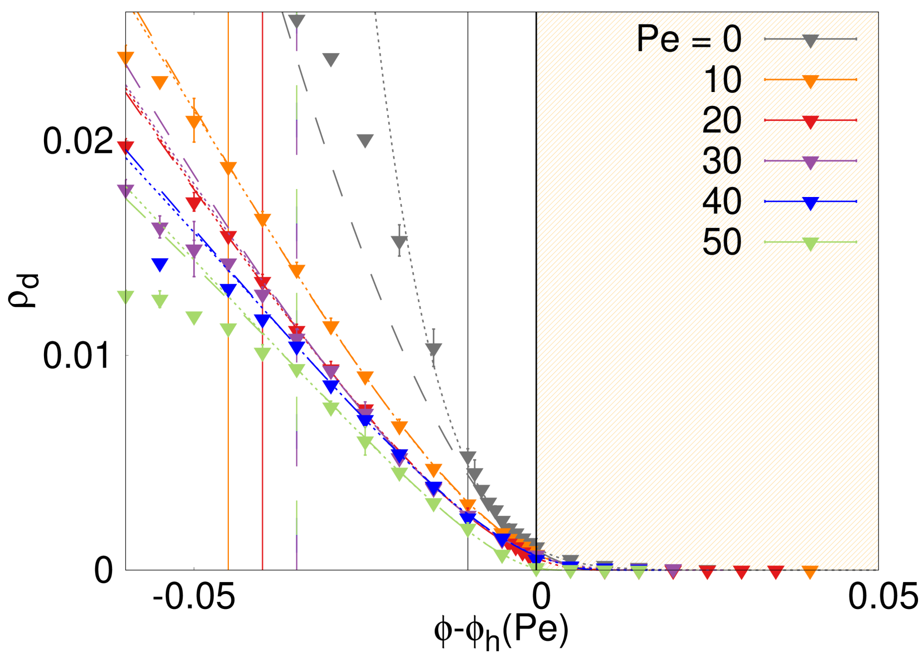
$z = 1.45(1+1) \sim 3$

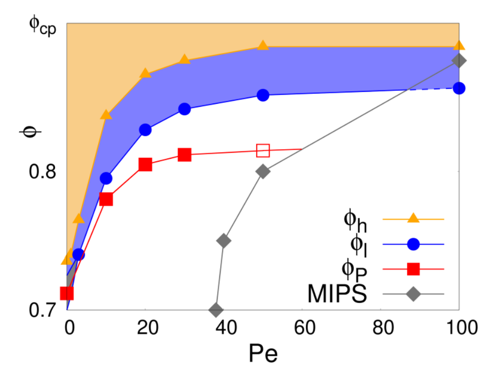
Reviews on the application of fractals to colloidal aggregation

R. Jullien, Croatia Chemica Acta 65, 215 (1992) P. Meakin, Physica Scripta 46, 295 (1992)

Dislocations

At the solid-hexatic transition for all Pe $\nu = 0.37$ Universality





Four (ϕ_c, ν, a, b dotted) vs. three ($\phi_c, \nu = 0.37, a, b$ dashed) parameter fits on data in the hexatic & solid phases only. Criteria to support $\nu = 0.37$:

– χ^2 *cfr.* **Batrouni et al** for $2d$XY

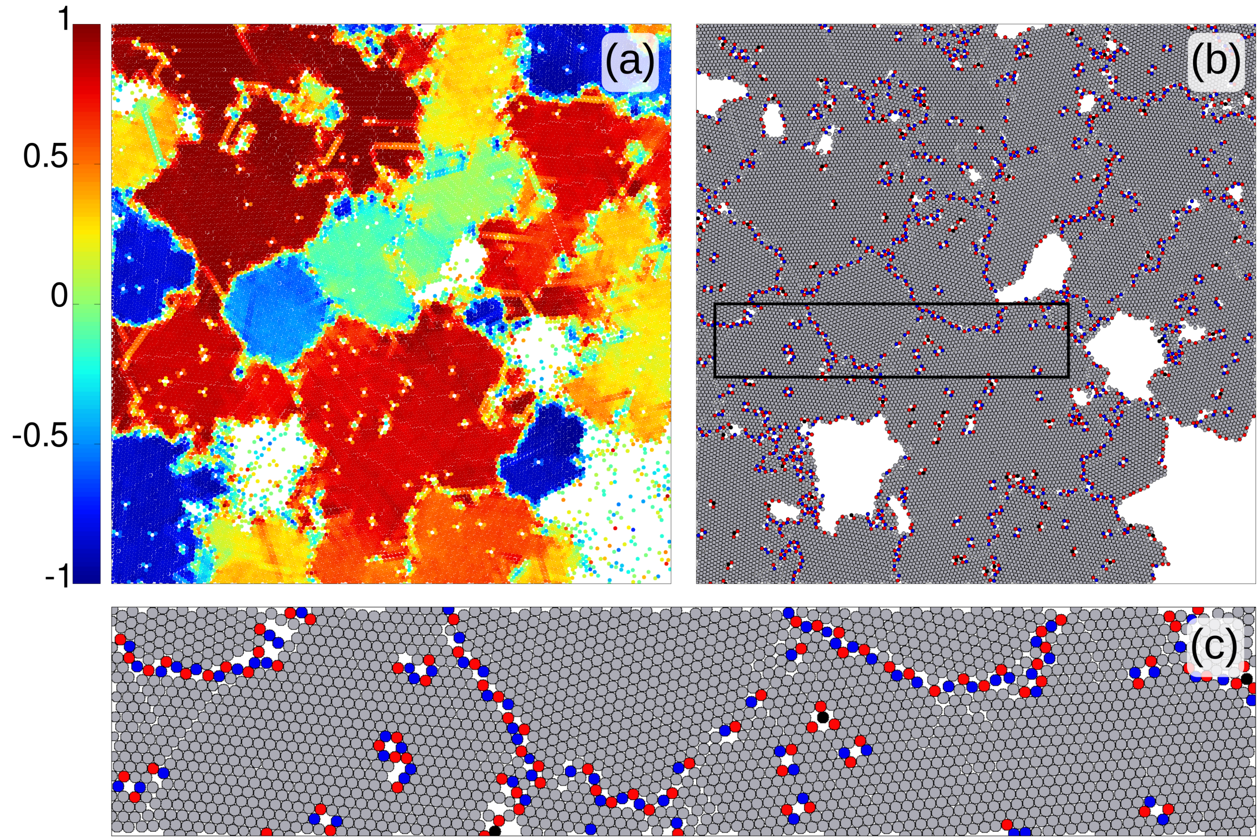
– not crazy values for a, b but crazy values for ν if let to be fitted

– difference between ϕ_c and ϕ_h erased by coarse-graining

Interfaces

Clusters of defects – mostly along hexatic-hexatic interfaces

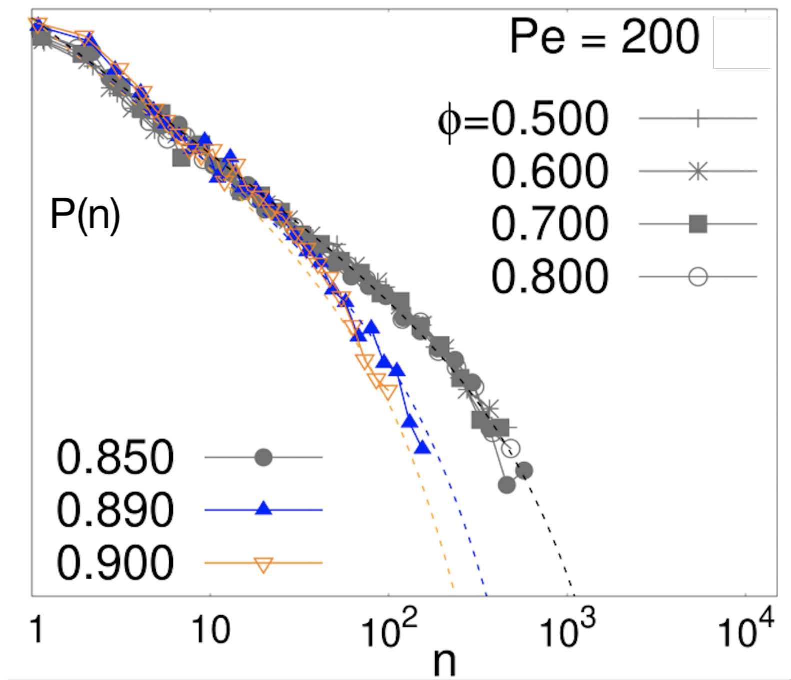
Hexatic order map | Defects



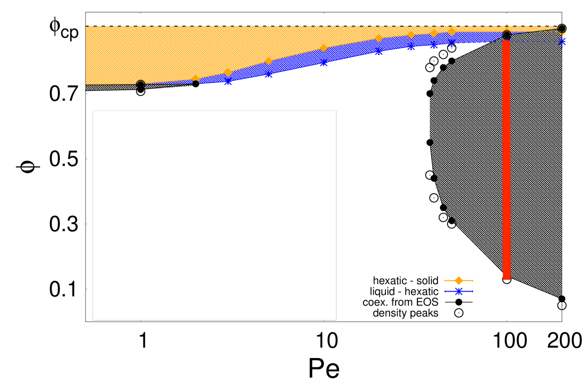
Zoom over the rectangular selection

Clusters of defects

Size distribution - Finite size cut-off



$$P(n) \simeq n^{-\tau} e^{-n/n^*}$$



Independence of ϕ at fixed Pe within MIPS

$n^* \sim 30, 50, 200$ in the **solid**, **hexatic** and **MIPS**, respectively, and $\tau \sim 2.2$