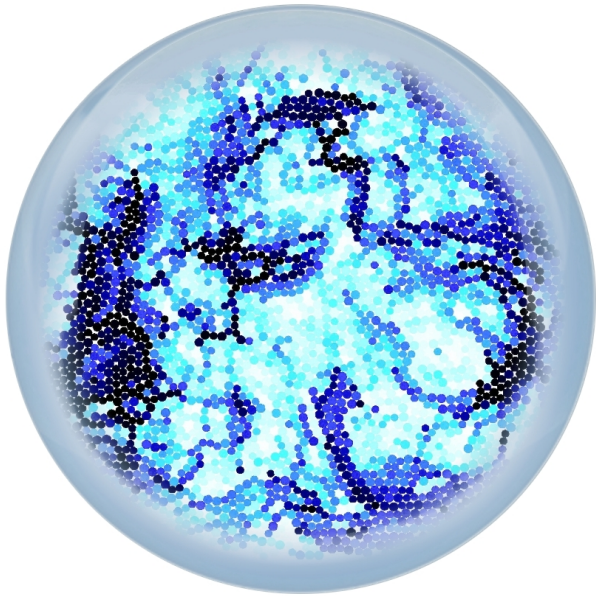


# Dynamics and Structure close to the Glass and Jamming transitions

Experiments and simulations



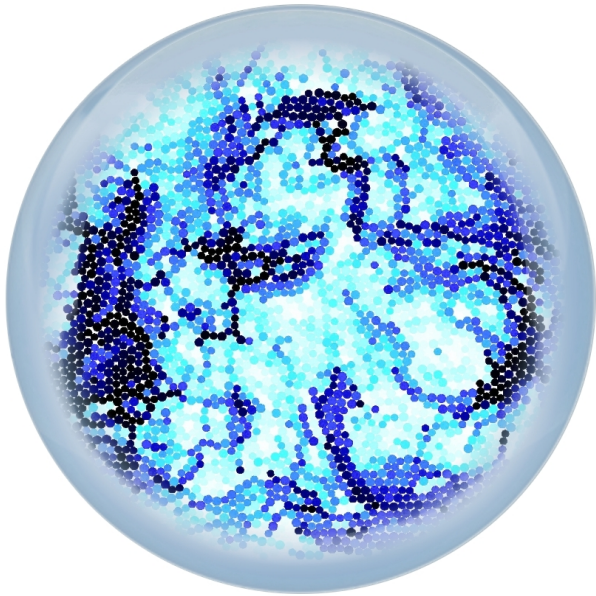
Présentée par :  
*Raphaël Candelier*

Directeur de thèse :  
*Olivier Dauchot (SPEC)*

En collaboration étroite avec :  
*Giulio Biroli (IPhT)*

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Experiments and simulations



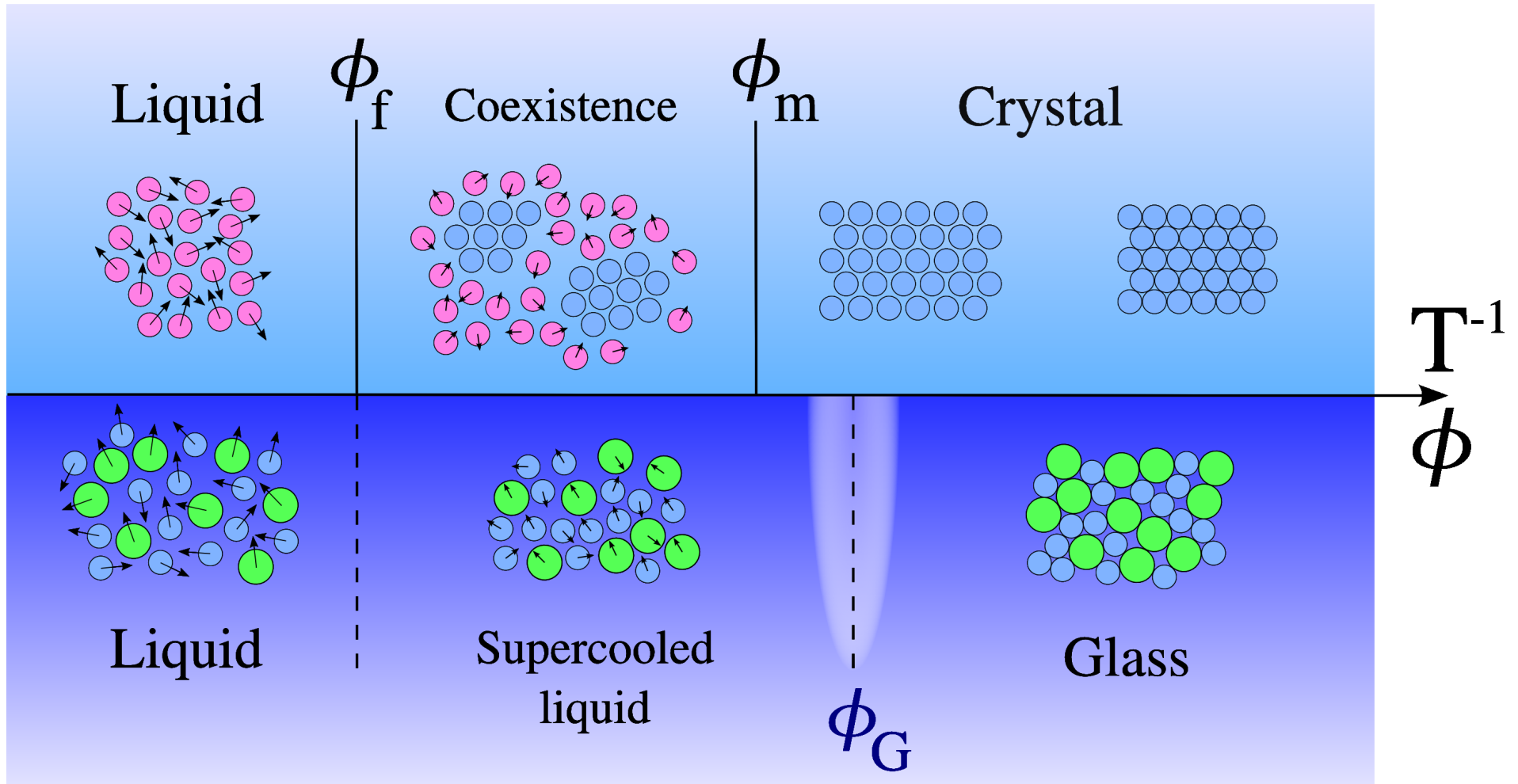
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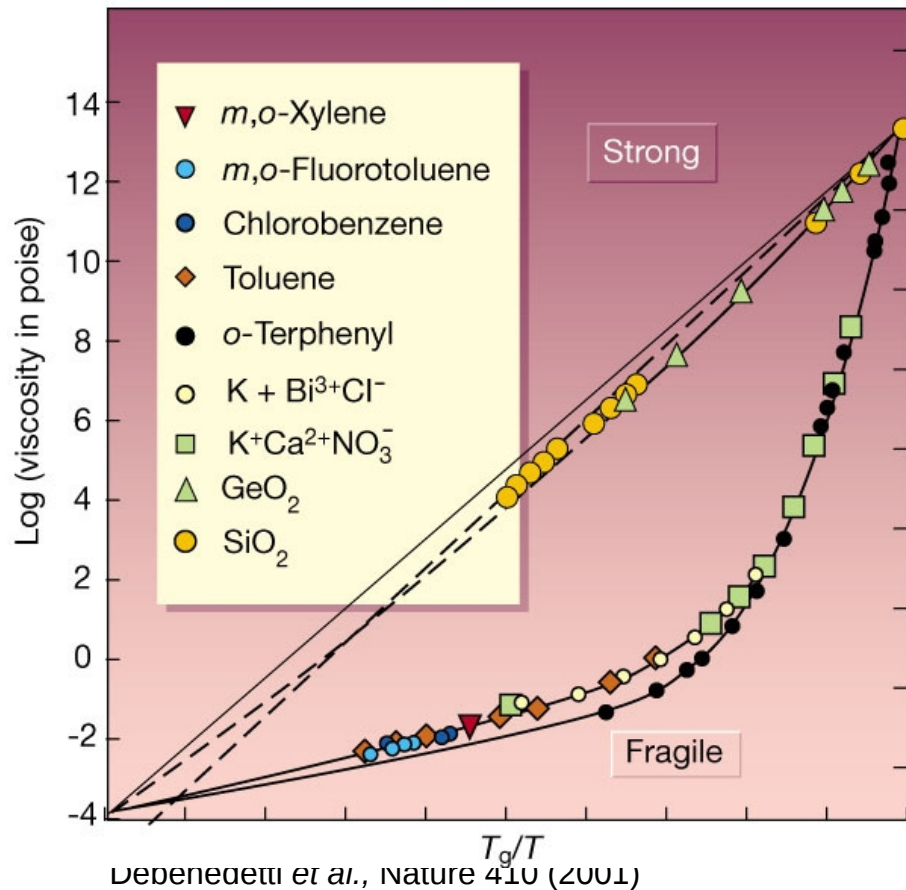
# The glass transition at a glance

From liquid to crystal ...



... or to amorphous solid

# The glass transition in a nutshell (1)



Viscosity increase :

- *Exponentially*  
→ « strong » glass
- *More than exponentially* :  
→ « fragile » glass

**Super-Arrhenius behavior : increasing activation energies suggest collective effects**

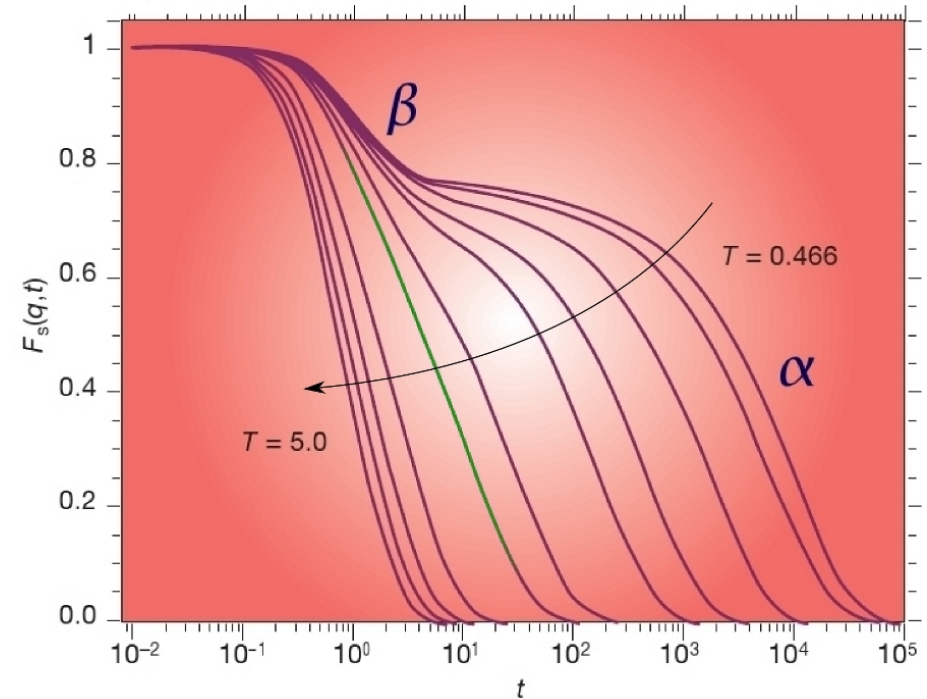
# The glass transition in a nutshell (2)

Intermediate scattering function (« structure factor »):

$$F_s(\vec{k}, t, T) \equiv \left\langle \frac{1}{N} \sum_{j=1}^N e^{i\vec{k} \cdot [\vec{r}_j(t) - \vec{r}_j(0)]} \right\rangle$$

Steeply growing timescale :  $\tau_\alpha$

Debenedetti et al., Nature 410 (2001)



The late relaxation is well-fitted by stretched exponentials

**This suggests the superposition of several relaxation times**

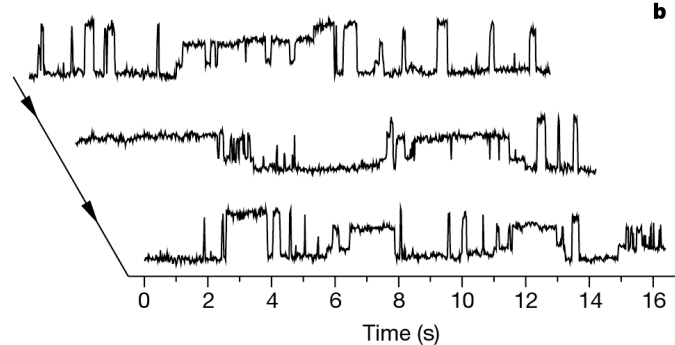
**→ heterogeneous dynamics**



# Burning questions on supercooled matter (1)

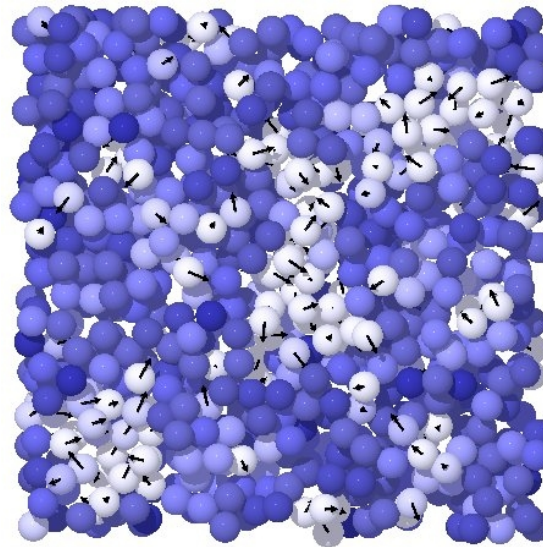
Indeed, dynamical correlations have been observed in several systems :

Liquids



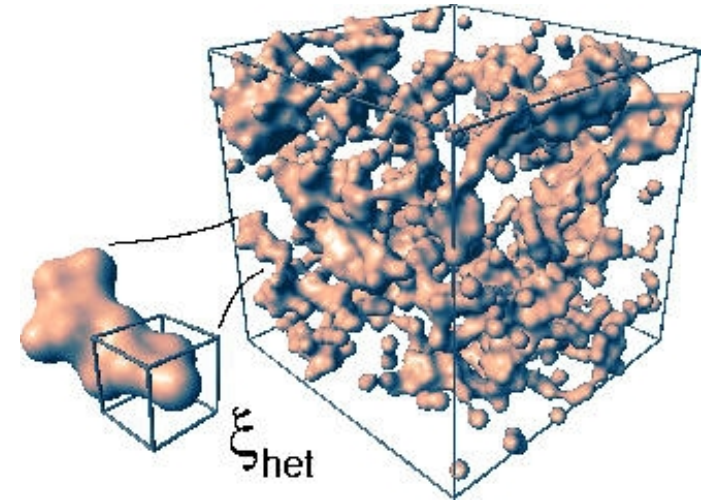
Vidal Russel *et al.*, Nature 408, 695 (2000)

Colloidal gels



Credits : Maria Kilfoil, McGill University

Hard sphere simulations



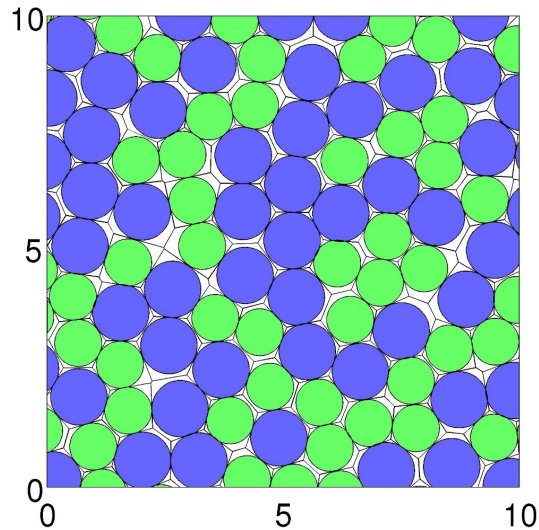
Credits : Graf & Heuer , Max-Planck-Institut

- Where do these correlations come from ?
- How do they evolve with thermodynamic parameters ( $T$  or  $\phi$ ) ?

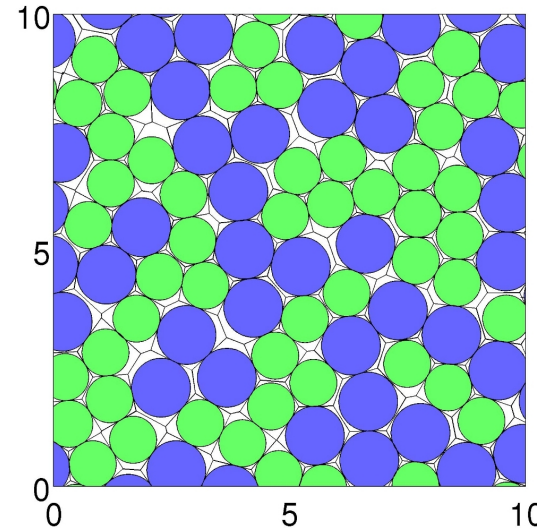


# Burning questions on supercooled matter (2)

Structure of a  
supercooled  
system →



Structure of a  
← glass



▶  $\phi$

**No trivial static signature of the transition  
in the structure of the material**

- What kind of static property rules the dynamics ? (if any ...)

# Roadmap

## Systems

- Why granular media ?
- Systems investigated during my thesis



## Part 1 : Heterogeneous relaxations

- From granular media to Lennard-Jones liquids

## Part 2 : The Building Blocks of Dynamical Heterogeneities

- From cage jumps to avalanches, a non conserved facilitation
- Evolution of dynamical facilitation with  $\phi$
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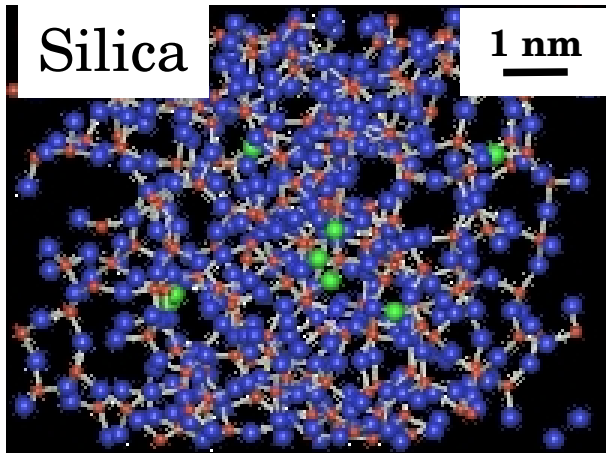
## Part 3 : Relation to Structure ?

- No clear signature of the dynamics in the structure
- More on simulated liquids : the isoconfigurational ensemble

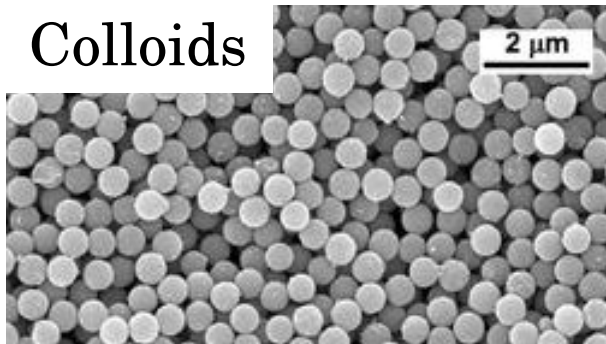
## Conclusion and perspectives



# Why meso/macroscale particles ?



Credits : JM Zuo, University of Illinois



Credits : ETH Zurich - D-MATL



T

1) Easy to locate and track

2) *Phenomenological* similarities with glasses were previously reported

3) Very different :

equilibrated supercooled liquid

versus

out of equilibrium dissipative steady state

→ If the analogy is possible, then it is not only a question of equilibrium measure

T,  $\phi$

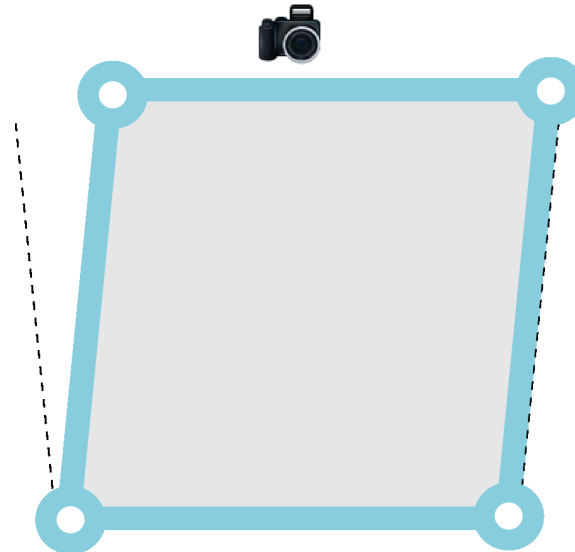
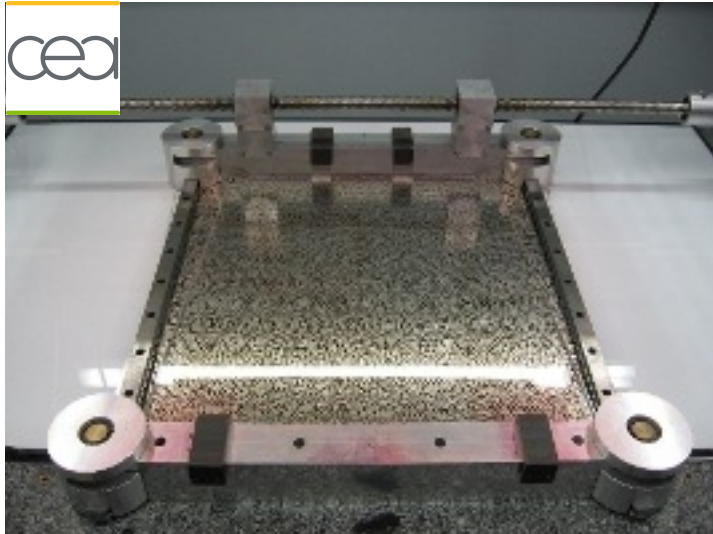
$\phi$

# Systems

Cyclic shear exp.

Fluidized bed exp.

LJ simulation



**2D**

quasi-static

*cyclic shear*

$(\theta_{\max} = \pm 5^\circ)$

8300 metallic cylinders with 2 sizes  $\rightarrow$  *no cristallisation*

Constant packing fraction during shear ( $\phi = 0.84$ )

Evolution from one cycle to another

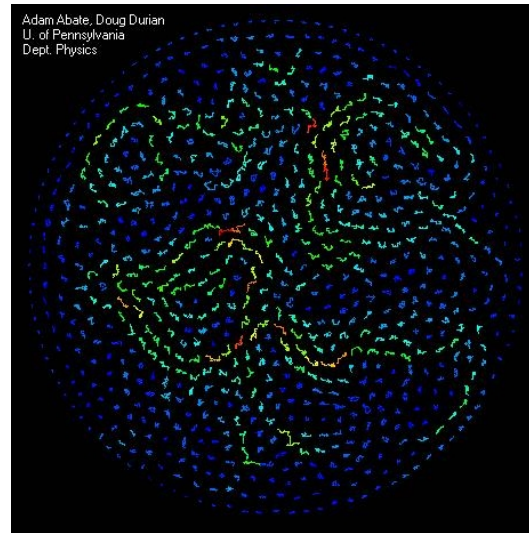
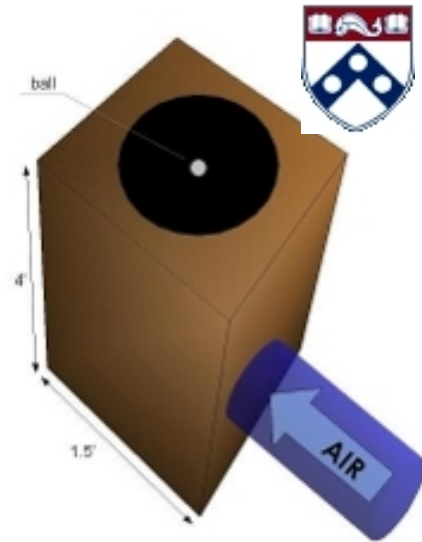
$\rightarrow$  Marty *et. al.* Phys. Rev. Lett. 94, 1 (2005)

# Systems

Cyclic shear exp.

Fluidized bed exp.

LJ simulation



**2D**

*Upflow of air*

( $v = 500$  to  $545$  cm.s<sup>-1</sup>)

Credits : Abate & Durian, Penn University

1790 to 1975 bidisperse metallic balls

6 packing fractions from  $\phi = 0.758$  to  $0.802$

Framerate : 12Hz

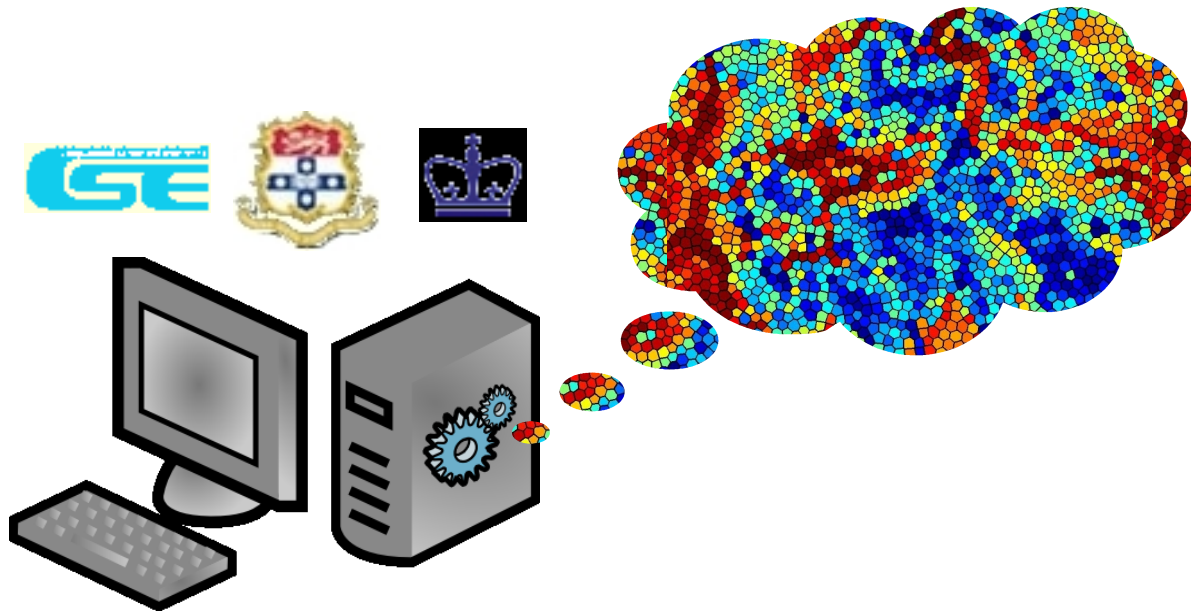
→ [Abate et. al. Phys. Rev. E 74, 3 \(2006\)](#)

# Systems

Cyclic shear exp.

Fluidized bed exp.

LJ simulation



2D

*Molecular Dynamics*

Non-additive  
Lennard-Jones liquid

5760 bidisperse particles softly interacting with non-additive potential ( $\sigma_{ss}=1$ ,  $\sigma_{bb}=1.4$  but  $\sigma_{sb}=1.1$ )  $\Rightarrow$  no microcrystallization

One density  $d = 0.719$

$\rightarrow$  [Widmer-Cooper et. al. Nat. Phys. 4, 711 \(2008\)](#)

# Roadmap

## Systems

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## Conclusion and perspectives

# An heterogeneous relaxation (1)

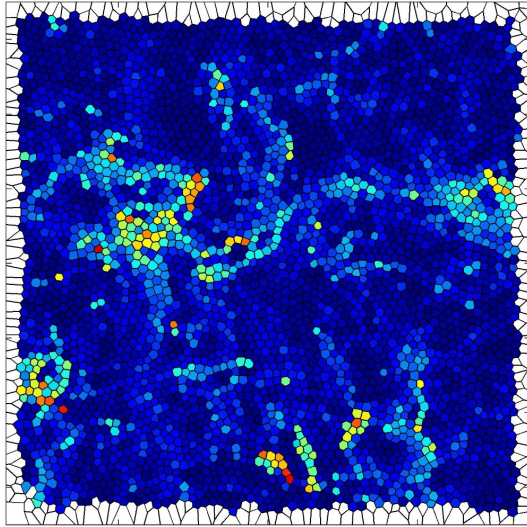
On a timescale  $\tau$  :

$$Q_{p,t} \equiv e^{-\frac{\Delta r^2(\tau)}{2a^2}} \quad a \simeq \frac{d}{5}$$



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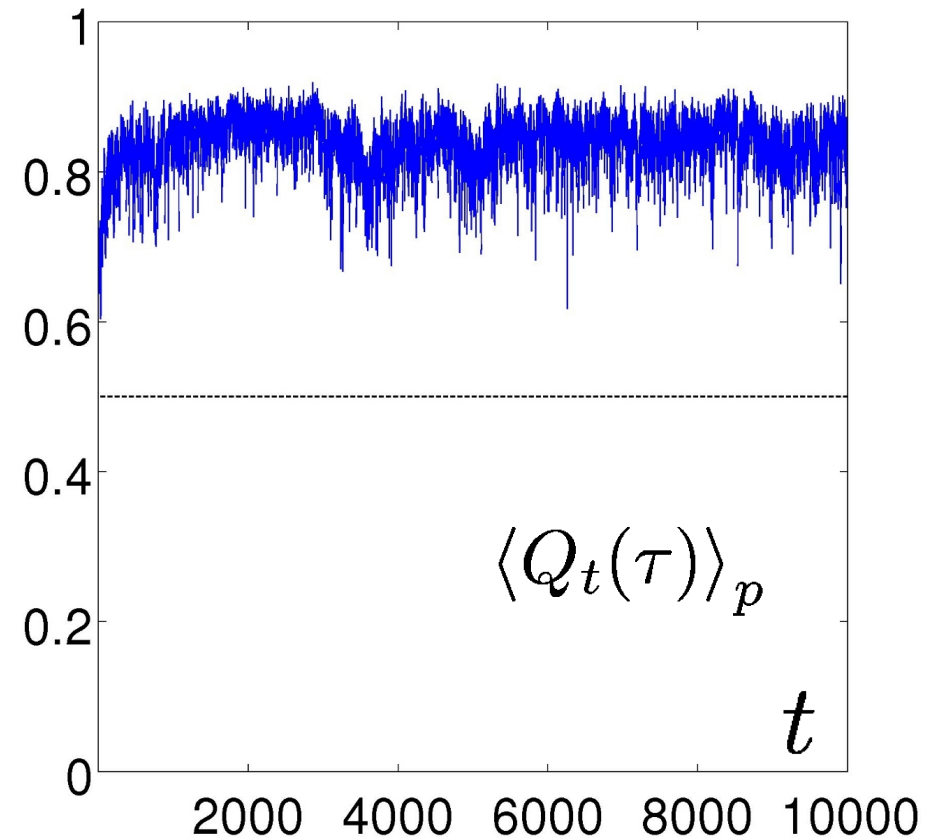
**Correlated**



$$\tau = 10$$

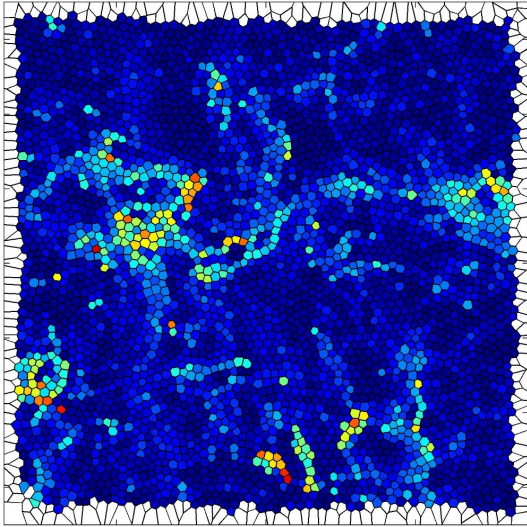
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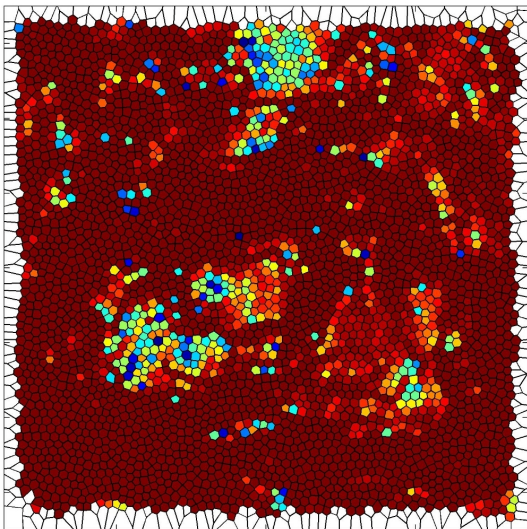


# An heterogeneous relaxation (1)

**Correlated**



$\tau = 10$



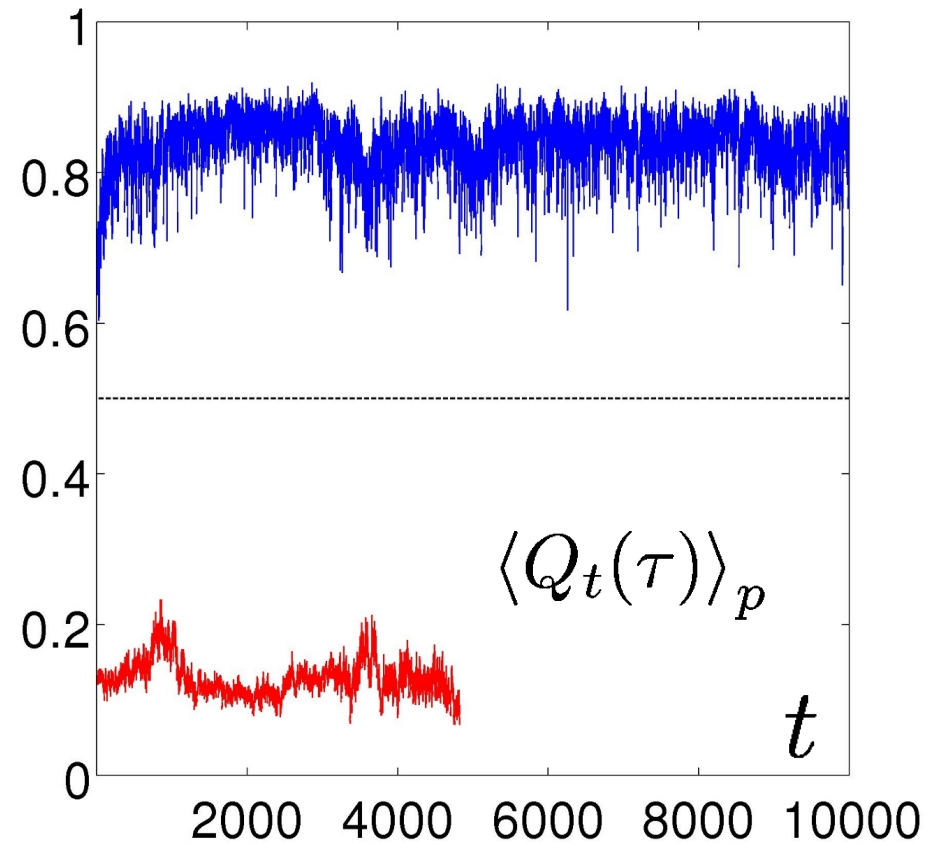
$\tau = 5000$

**Decorrelated**

On a timescale  $\tau$  :

$$Q_{p,t} \equiv e^{-\frac{\Delta r^2(\tau)}{2a^2}}$$

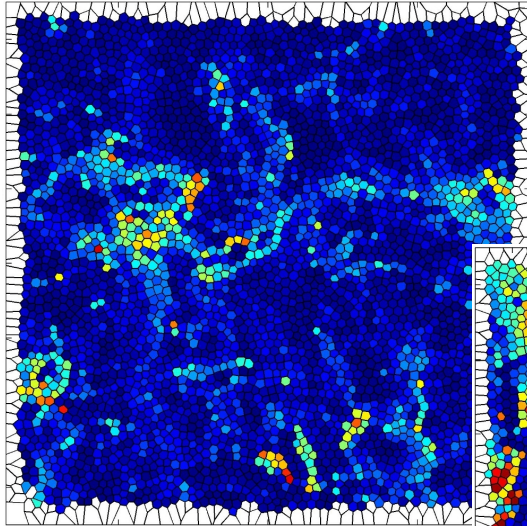
$$a \simeq \frac{d}{5}$$





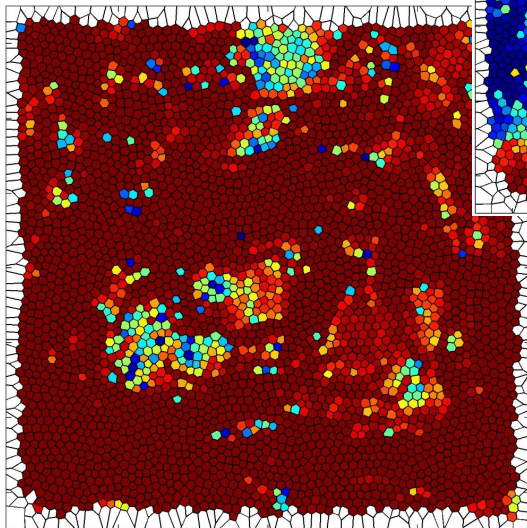
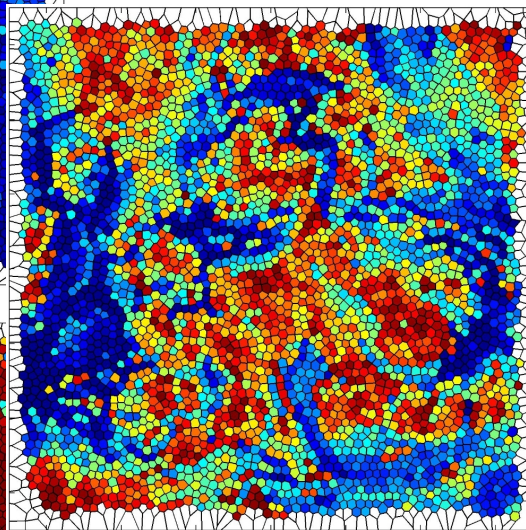
# An heterogeneous relaxation (1)

**Correlated**



$\tau = 10$

$\tau = 720$

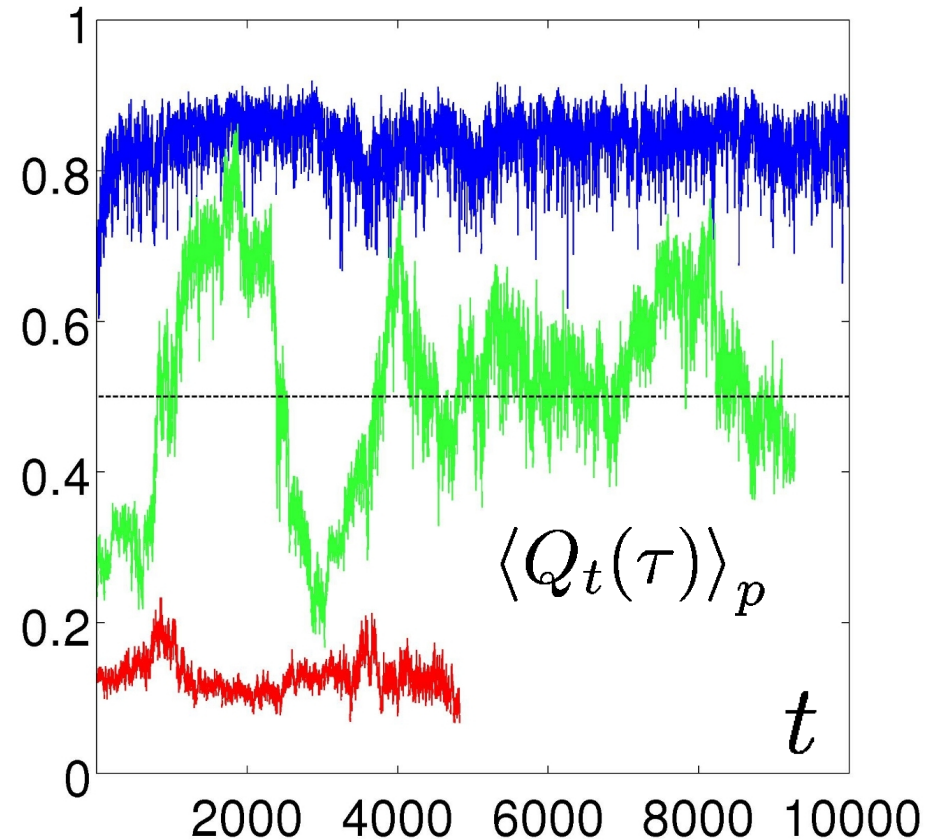


$\tau = 5000$

**Decorrelated**

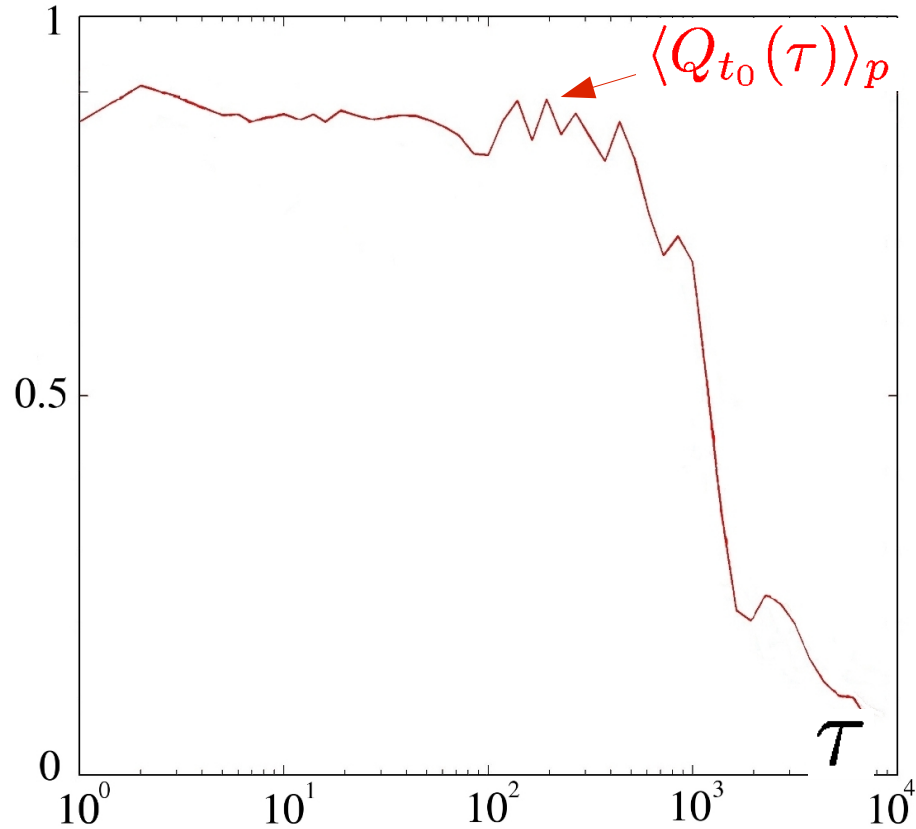
On a timescale  $\tau$  :

$$Q_{p,t} \equiv e^{-\frac{\Delta r^2(\tau)}{2a^2}} \quad a \simeq \frac{d}{5}$$



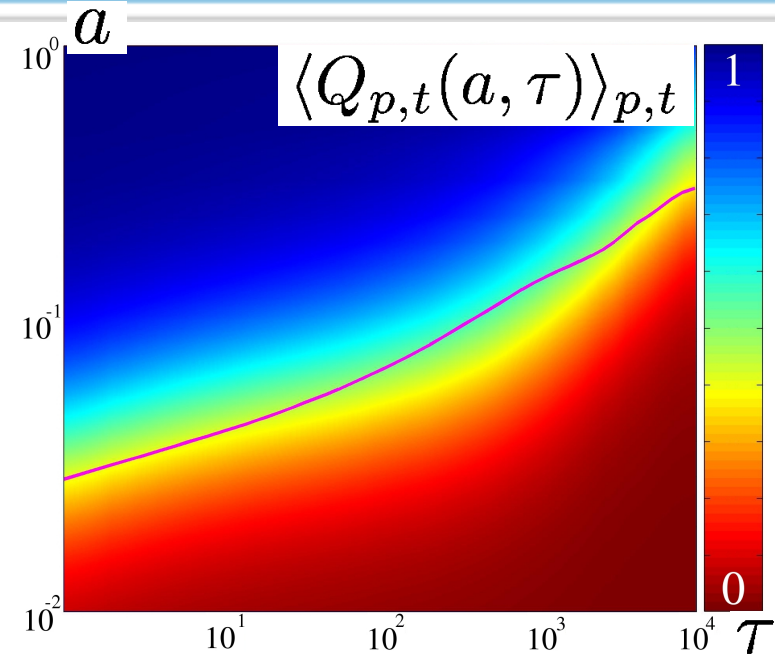
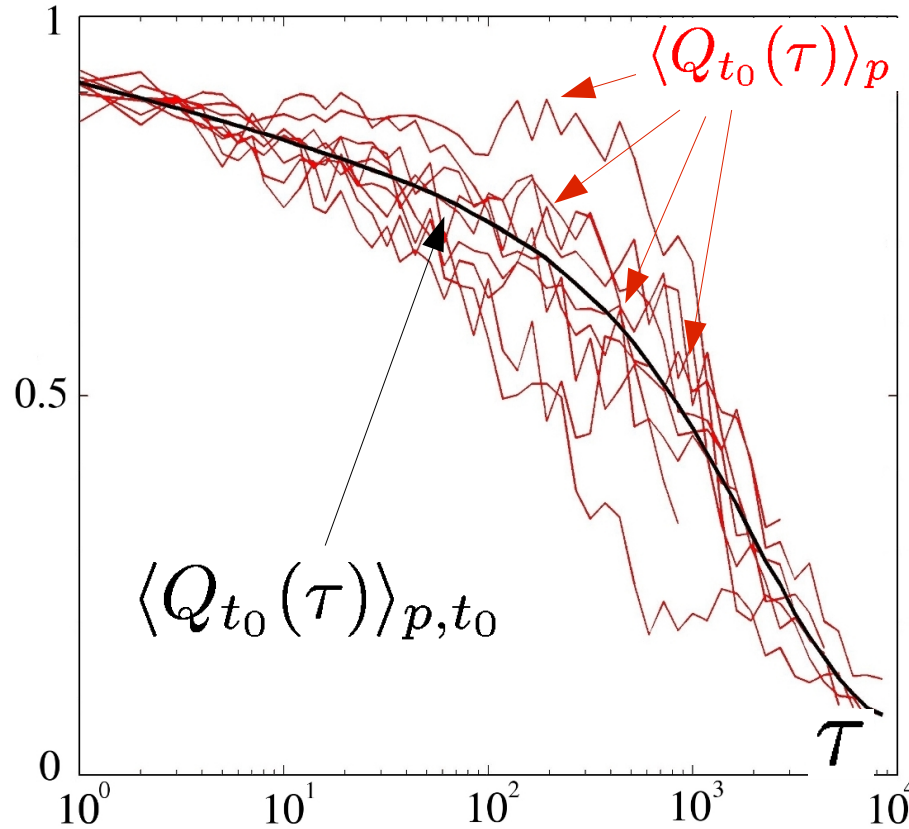
# An heterogeneous relaxation (2)

For  $a=d/5$  :



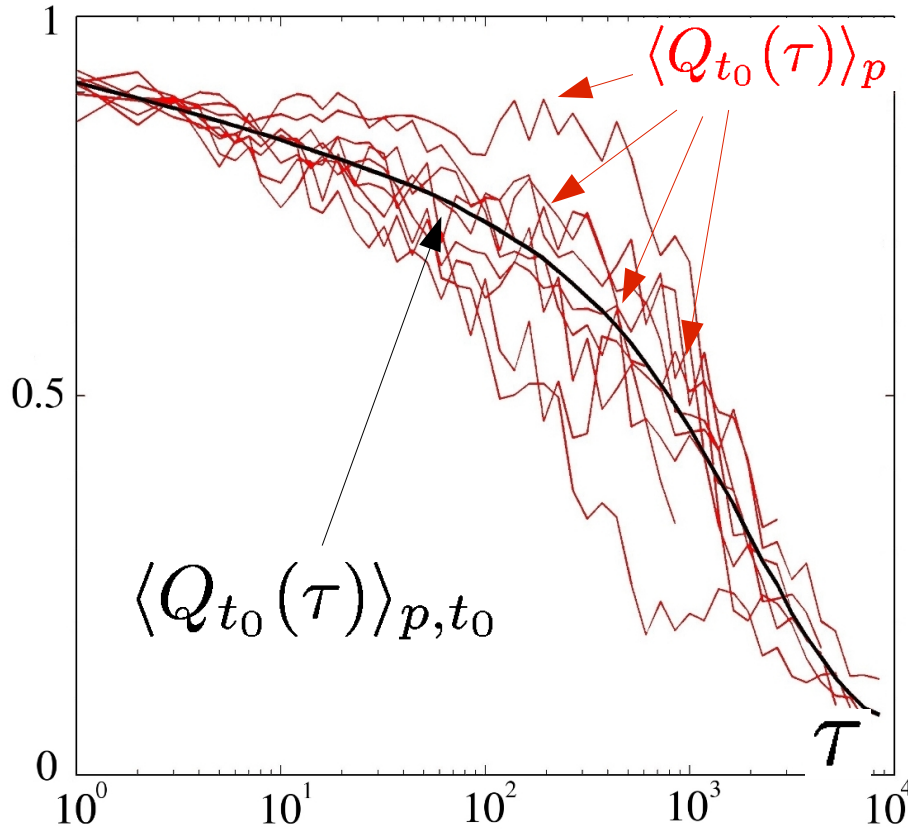
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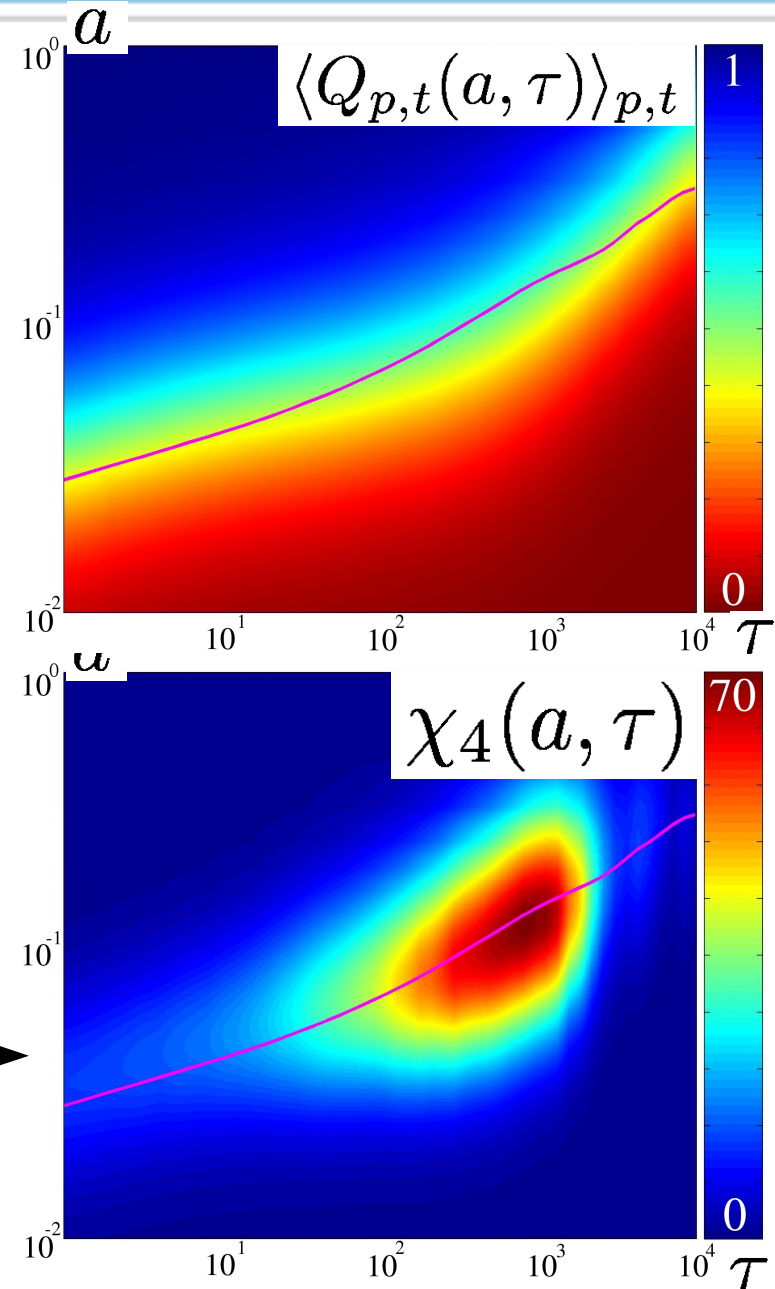


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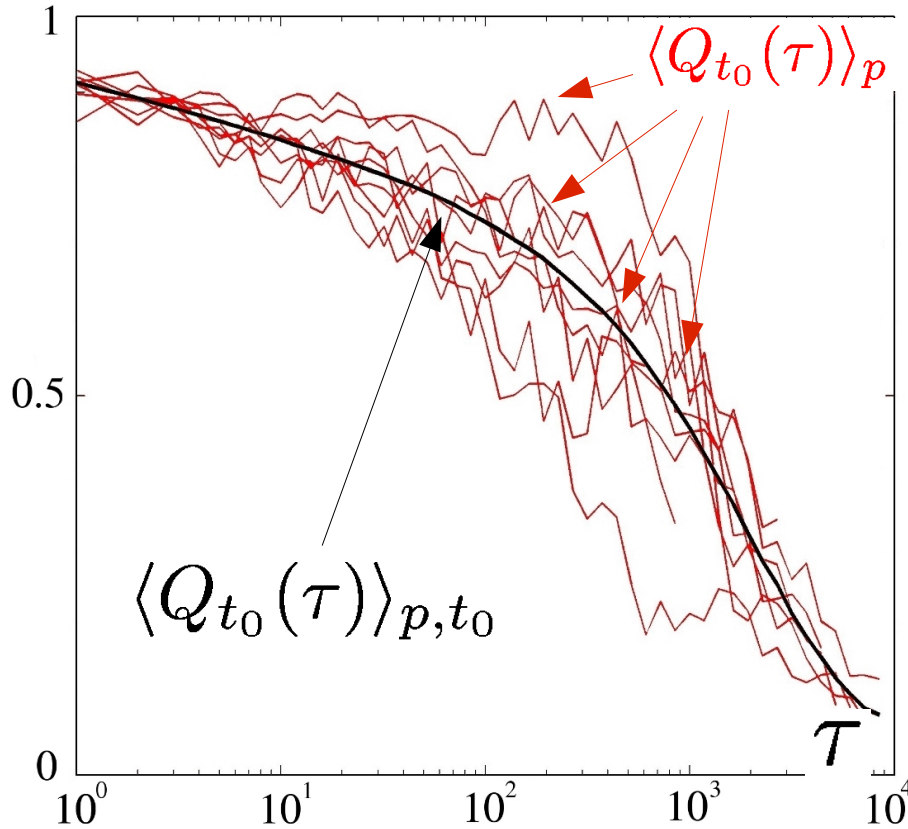


$$\chi_4(a, \tau) \equiv N \text{var}_t (\langle Q_t(a, \tau) \rangle_p) \rightarrow$$



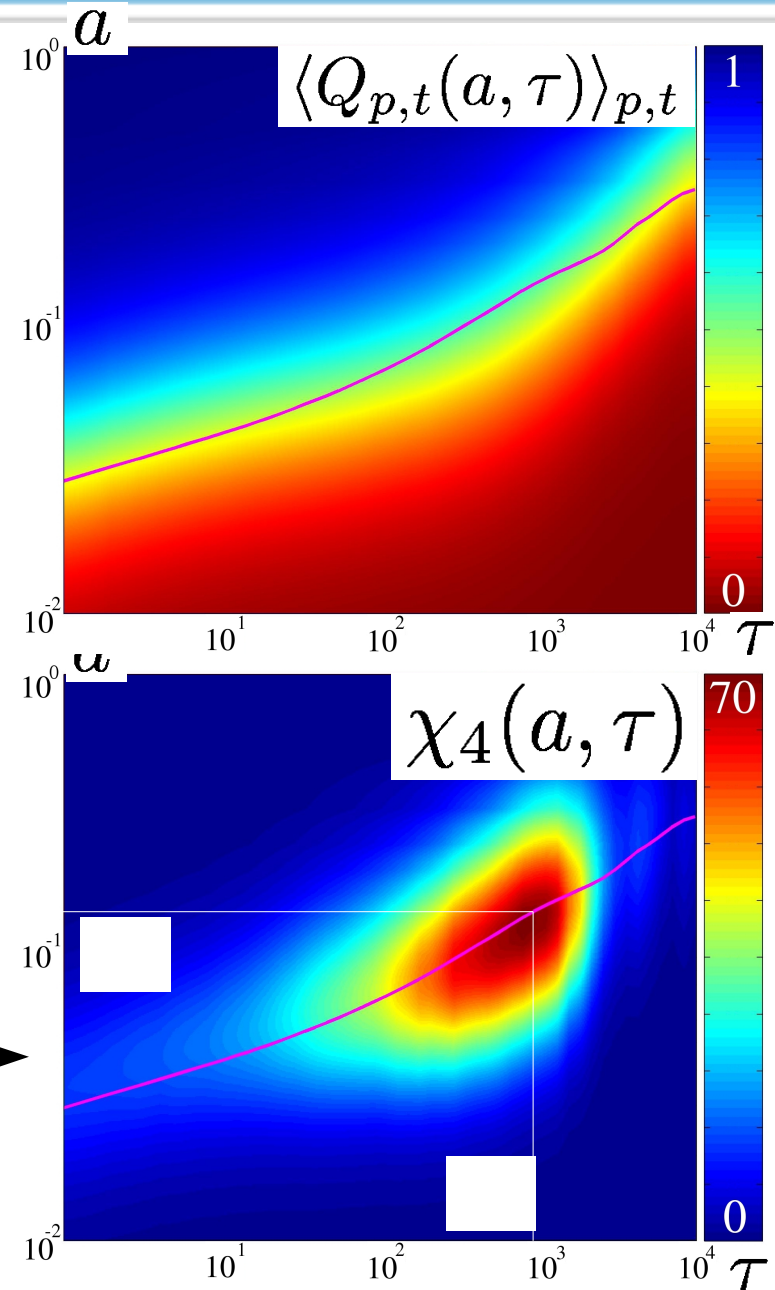
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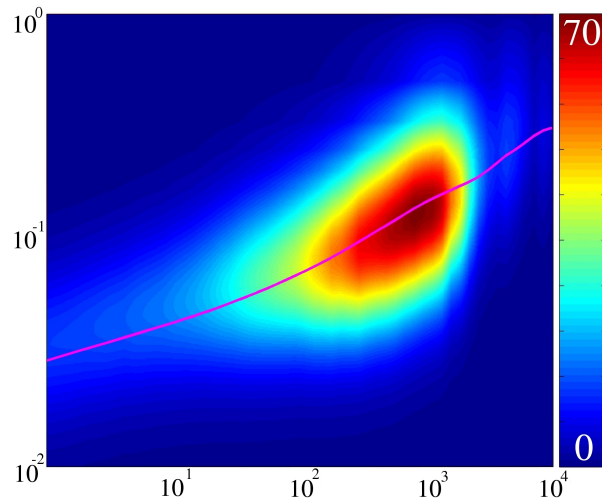
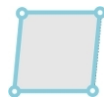
$$\chi_4(a, \tau) \equiv N \text{var}_t (\langle Q_t(a, \tau) \rangle_p) \rightarrow$$

$$a^* \simeq d/5 ; \tau^* = 720 \simeq \tau_\alpha$$

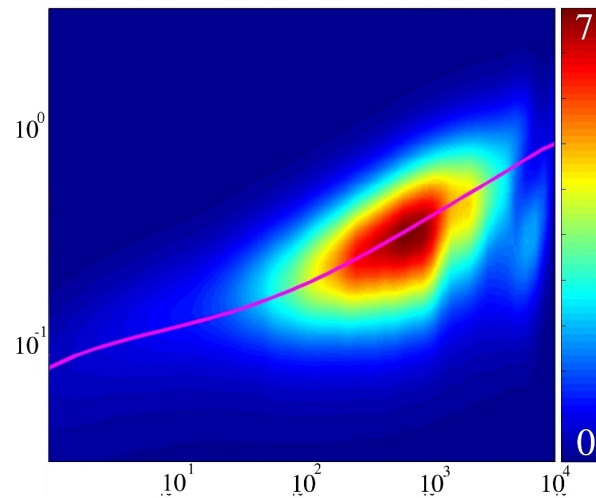


# An heterogeneous relaxation (3)

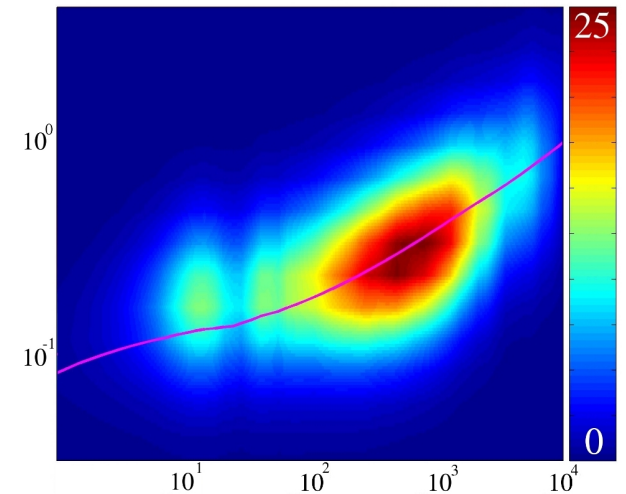
Cyclic shear  
experiment



Fluidized bed  
experiment



Lennard-Jones  
Simulation



- Same statistical properties of the dynamics
- In very different systems : validates the phenomenological analogy and broaden the scope of the description
- We can use each system's specificity to build this description

# Roadmap

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## Part 1 : Heterogeneous relaxations

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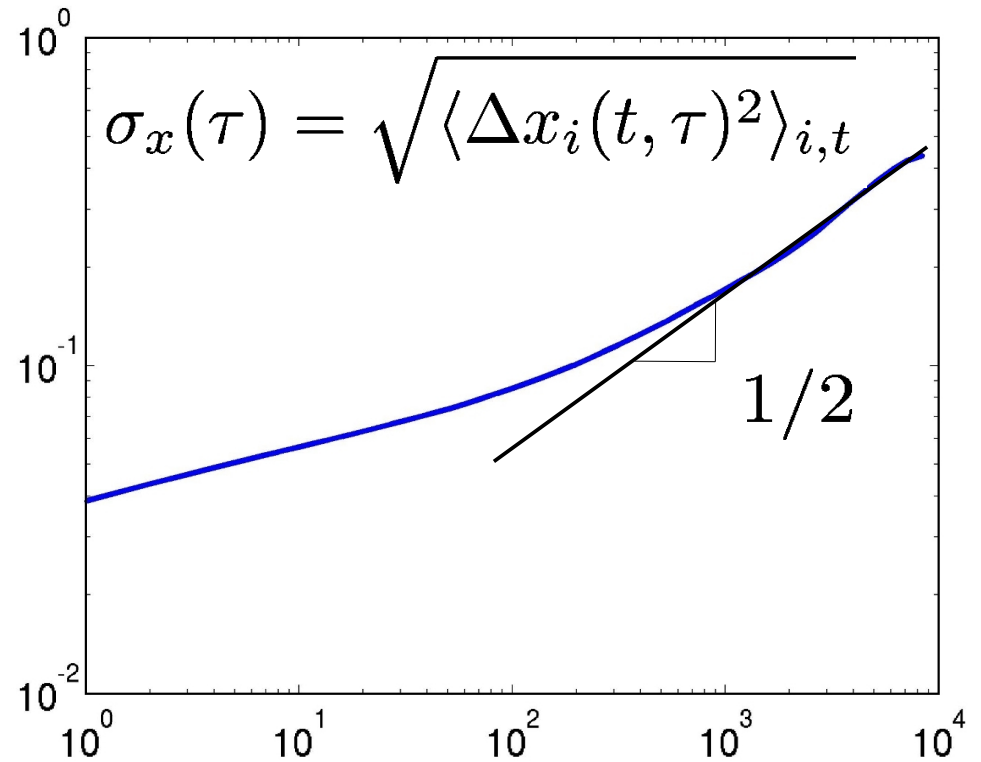
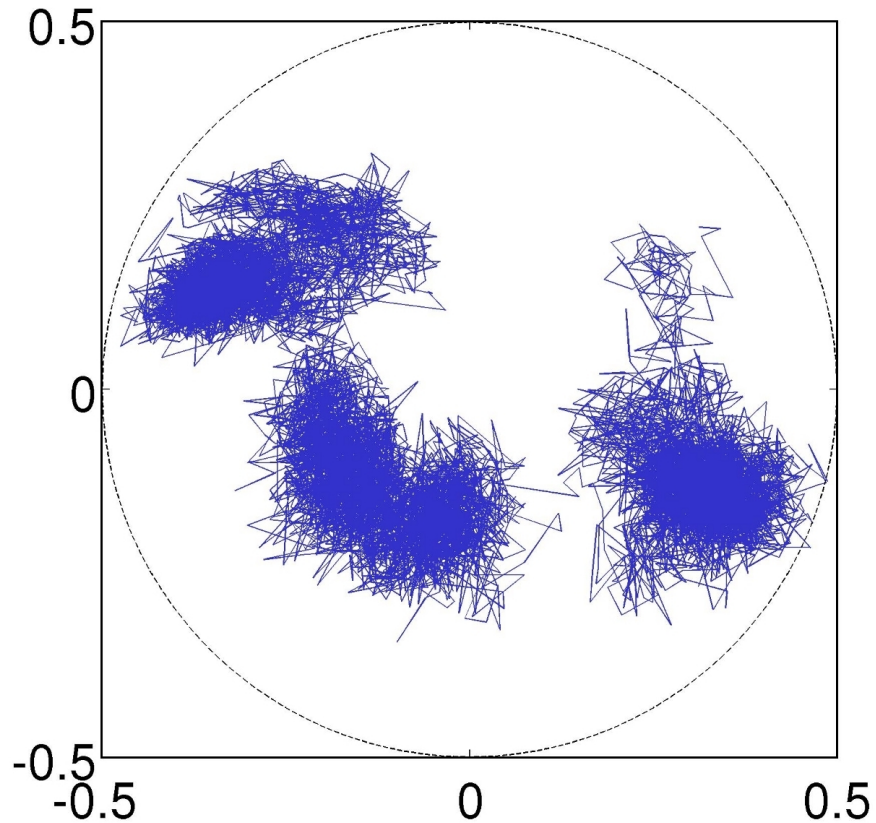
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## Conclusion and perspectives

# A « Cage » dynamics

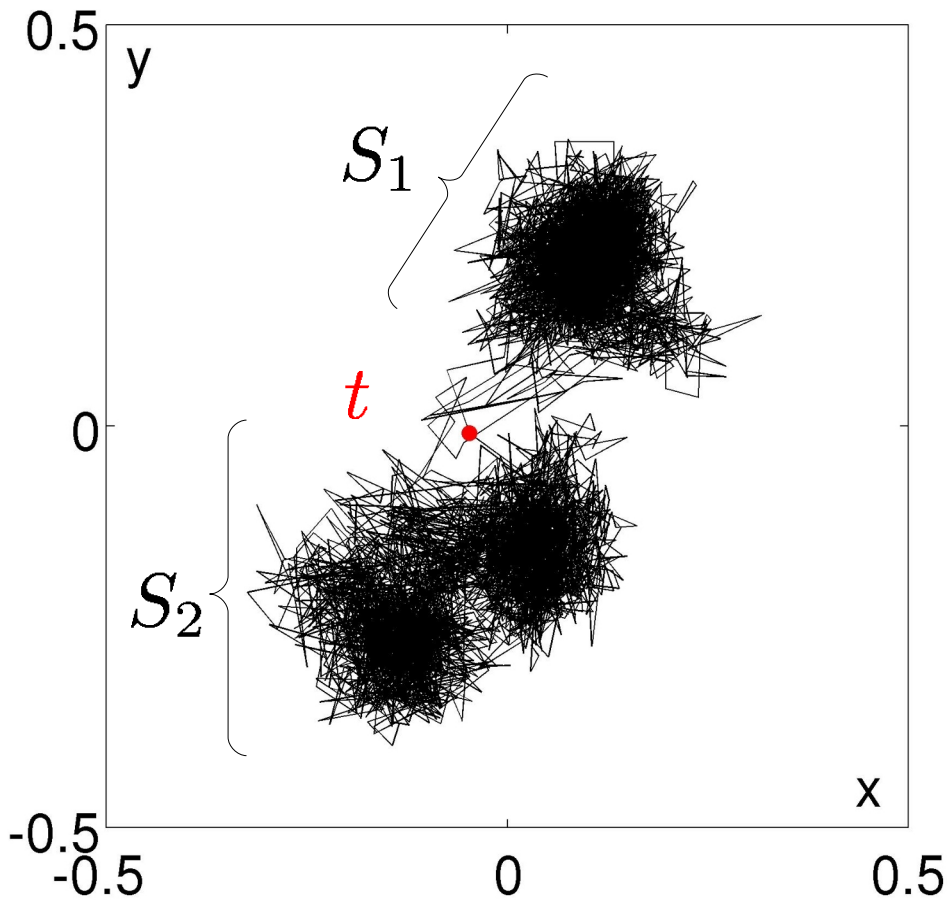




# Finding cage jumps (1)

Take one trajectory and compute :  $p(t) = \xi(t) \cdot \sqrt{\langle d_1(i)^2 \rangle_{i \in S_2} \cdot \langle d_2(j)^2 \rangle_{j \in S_1}}$

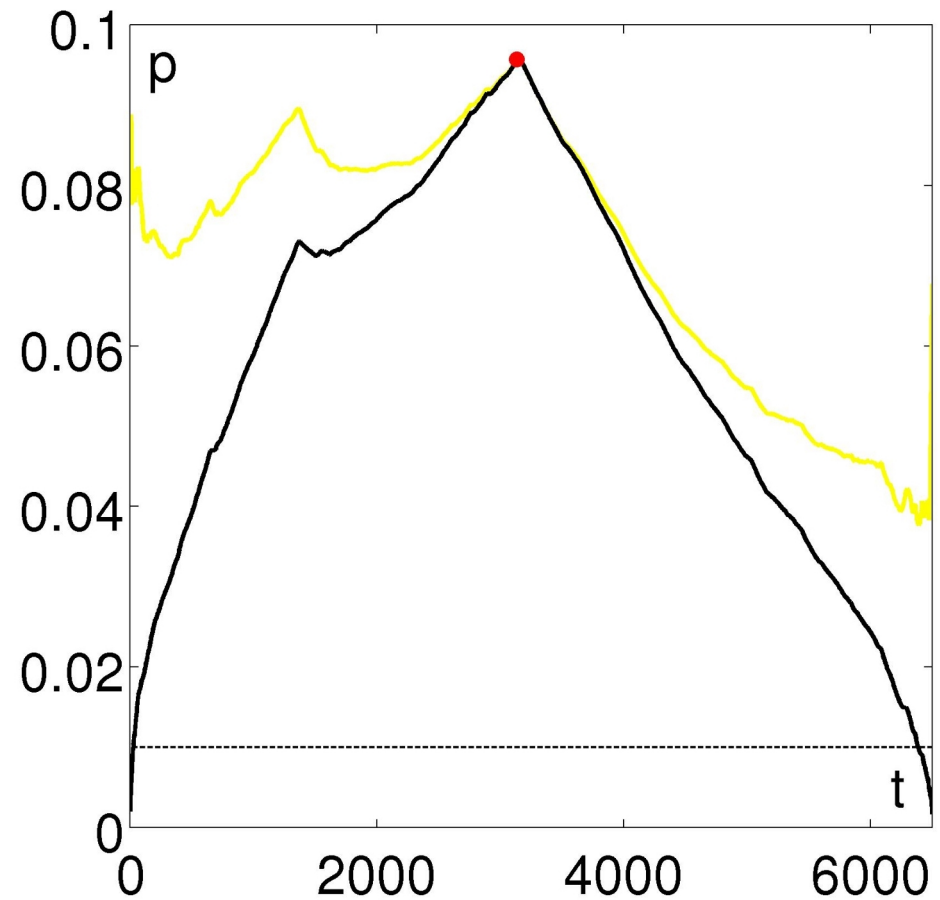
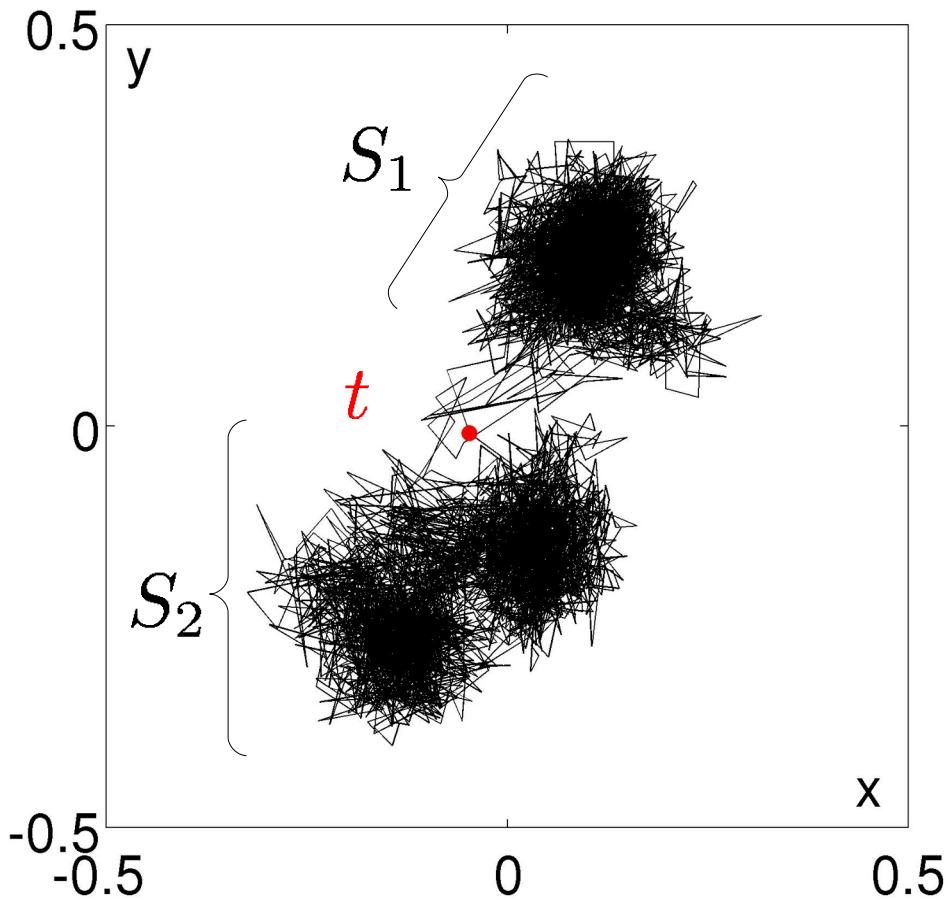
Mean quadratic distance to the center of mass of the **other** set of points



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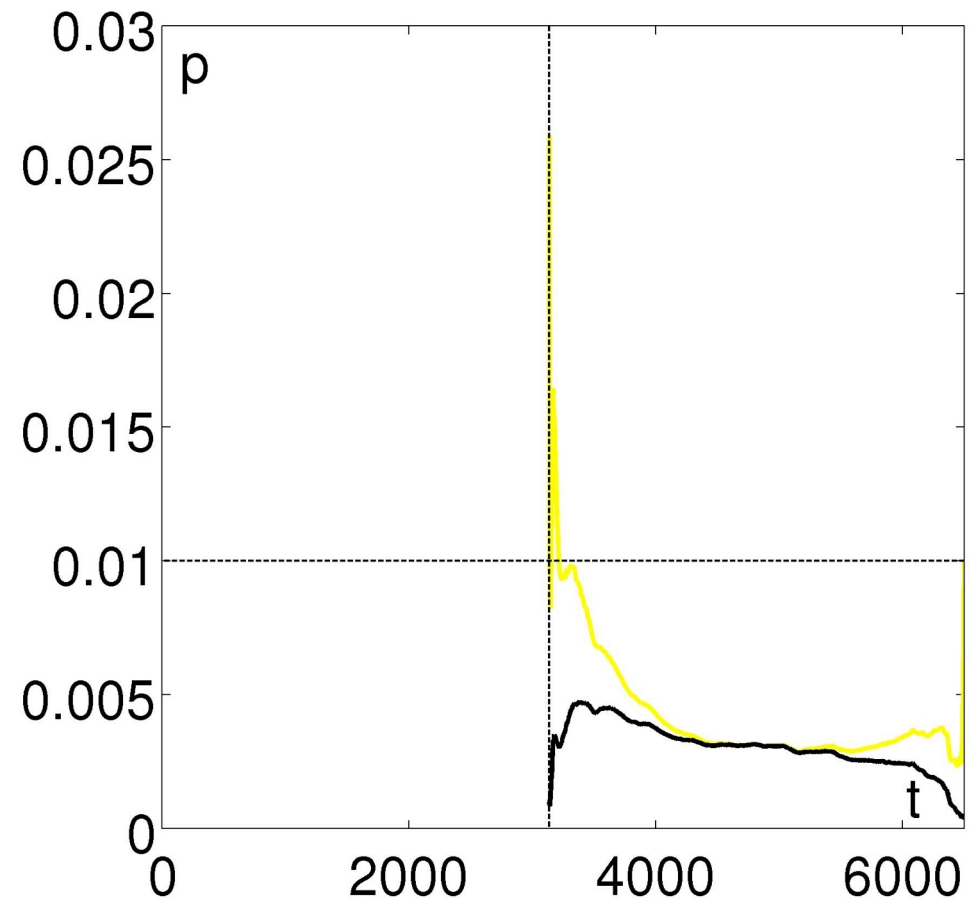
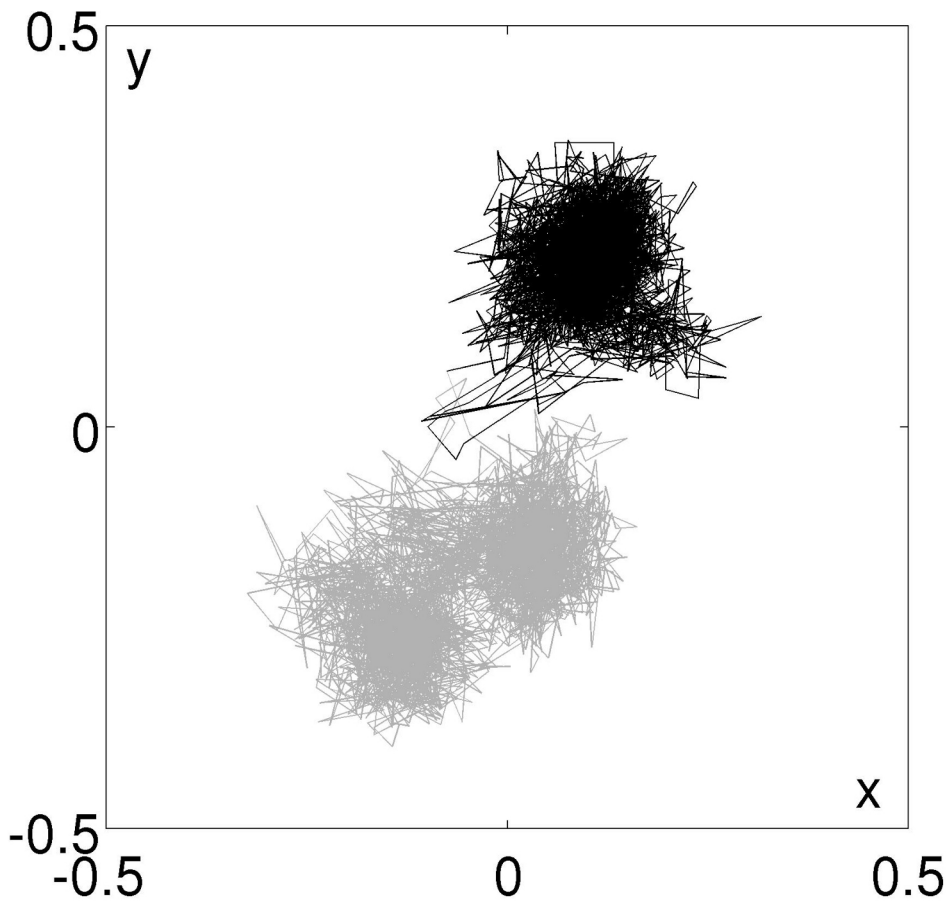
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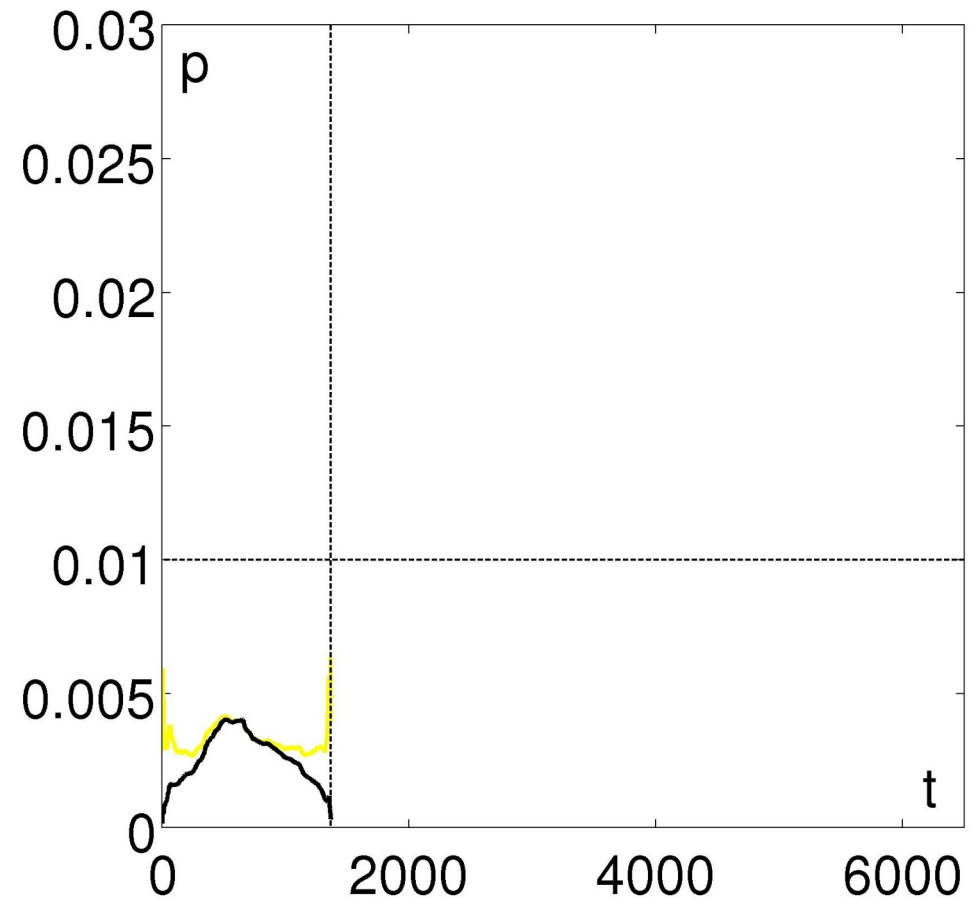
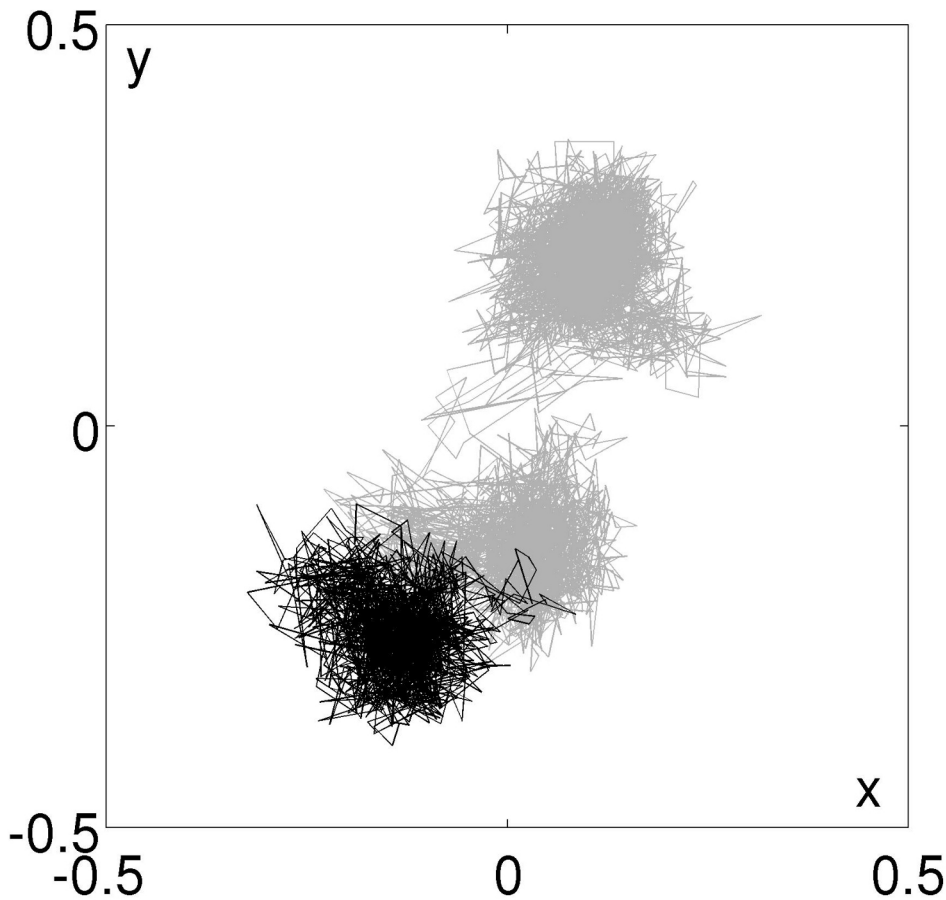
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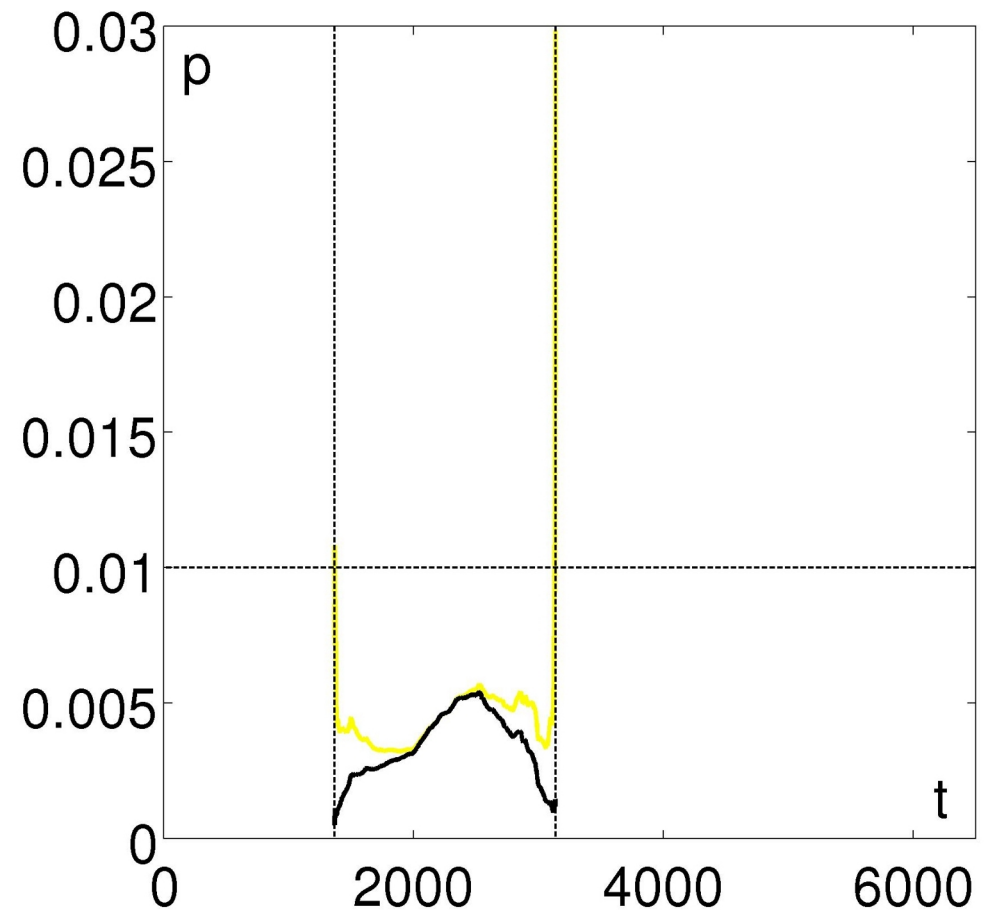
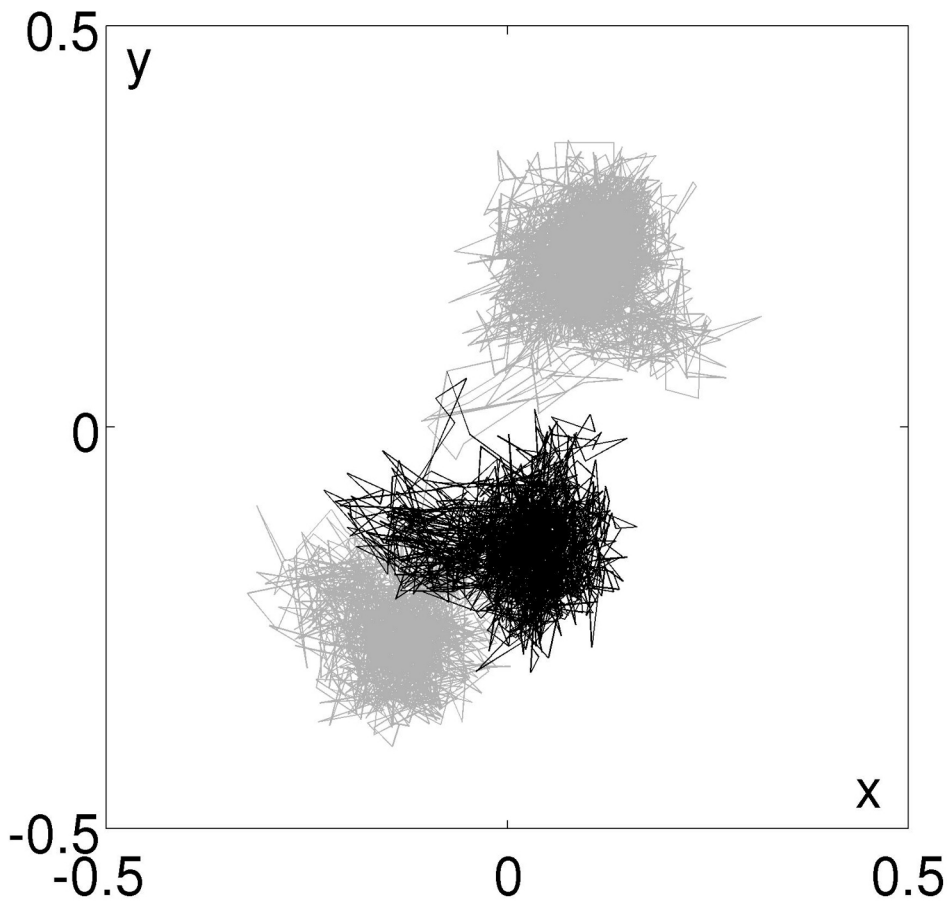
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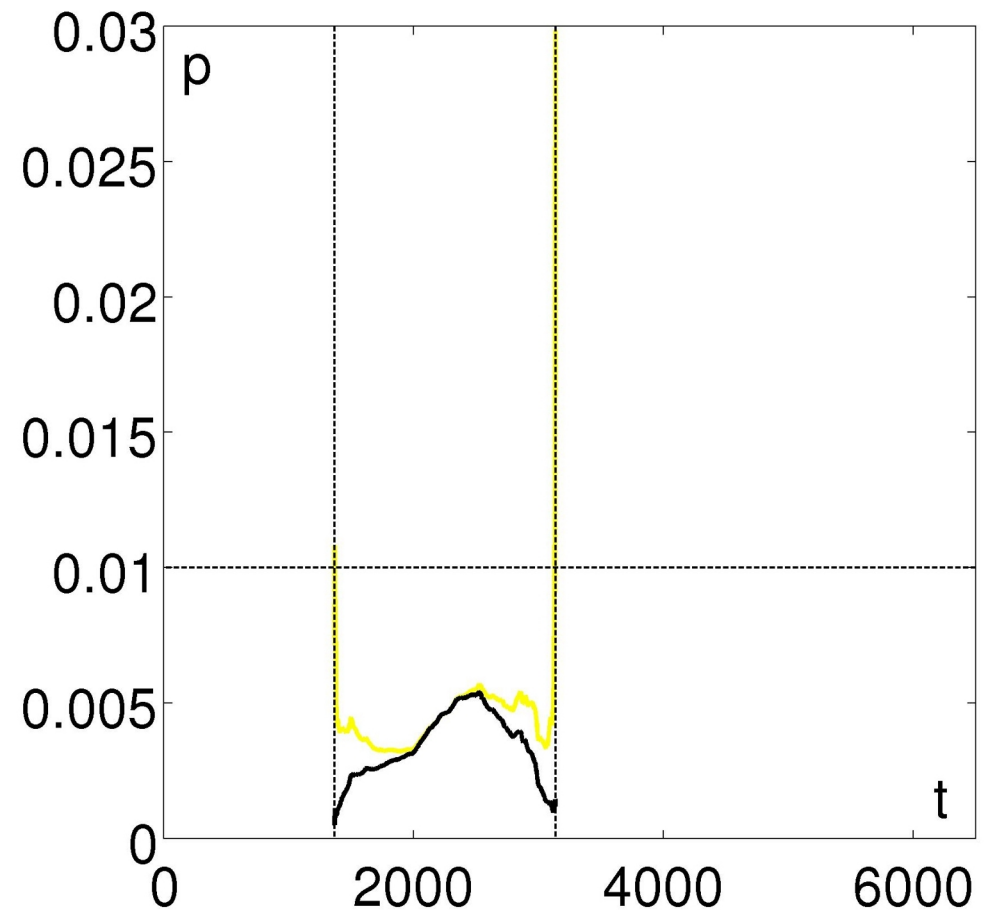
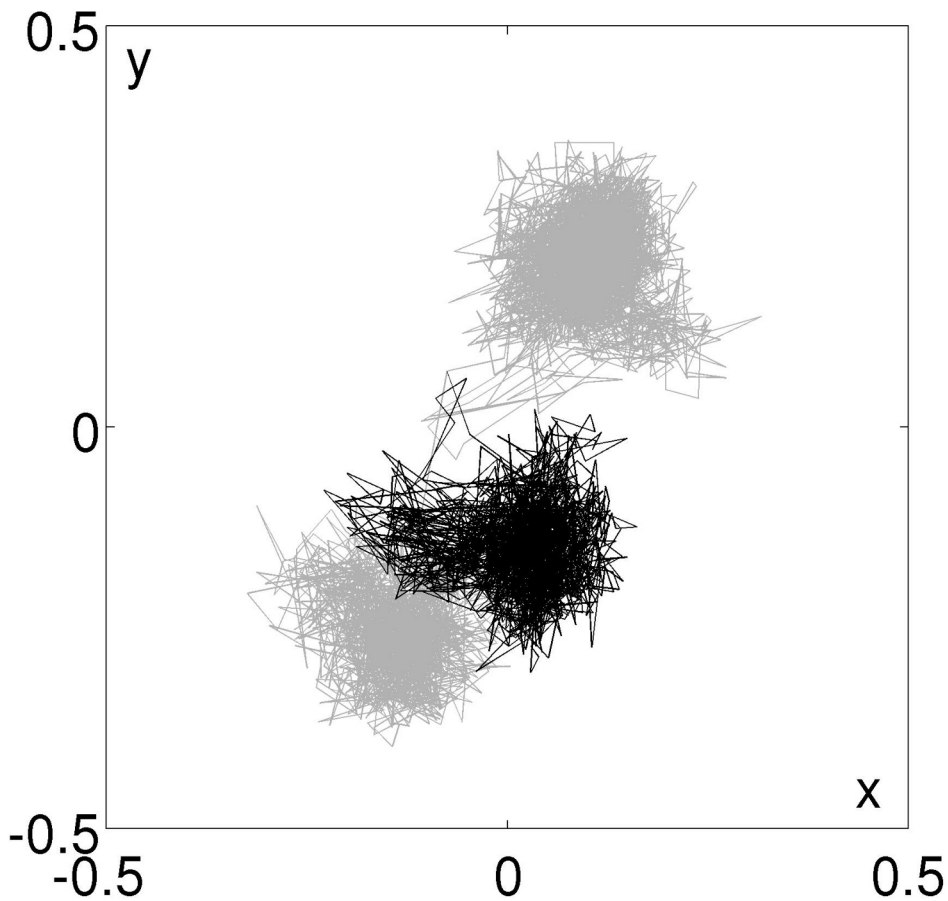
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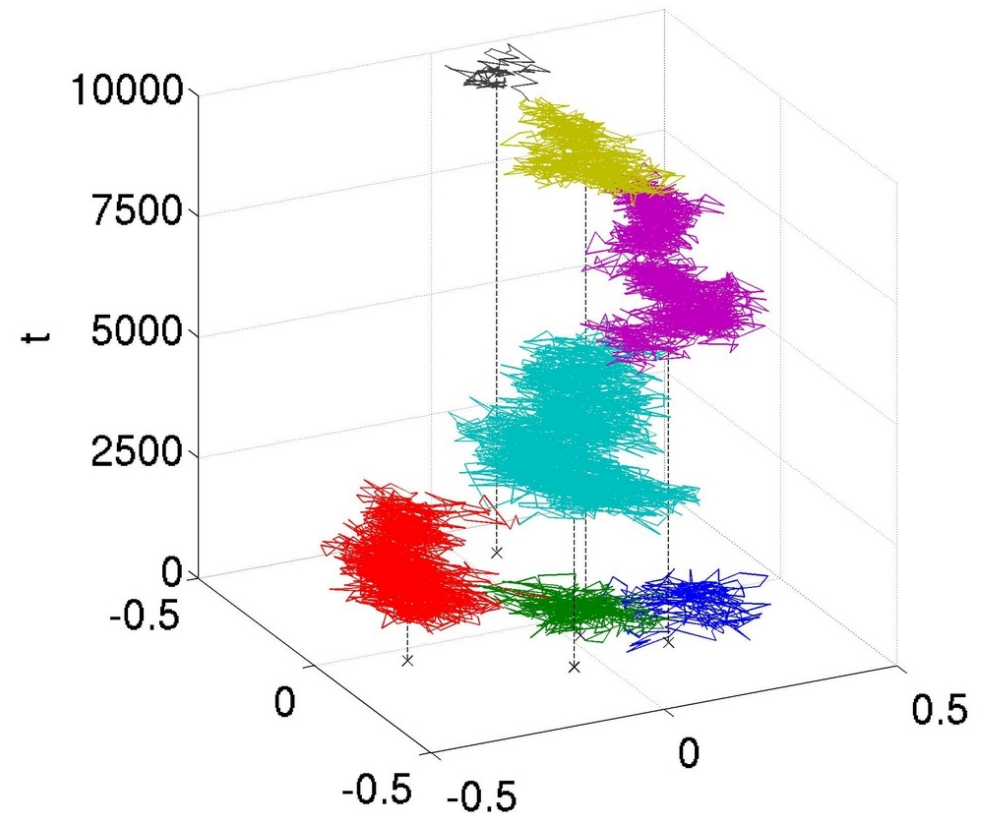
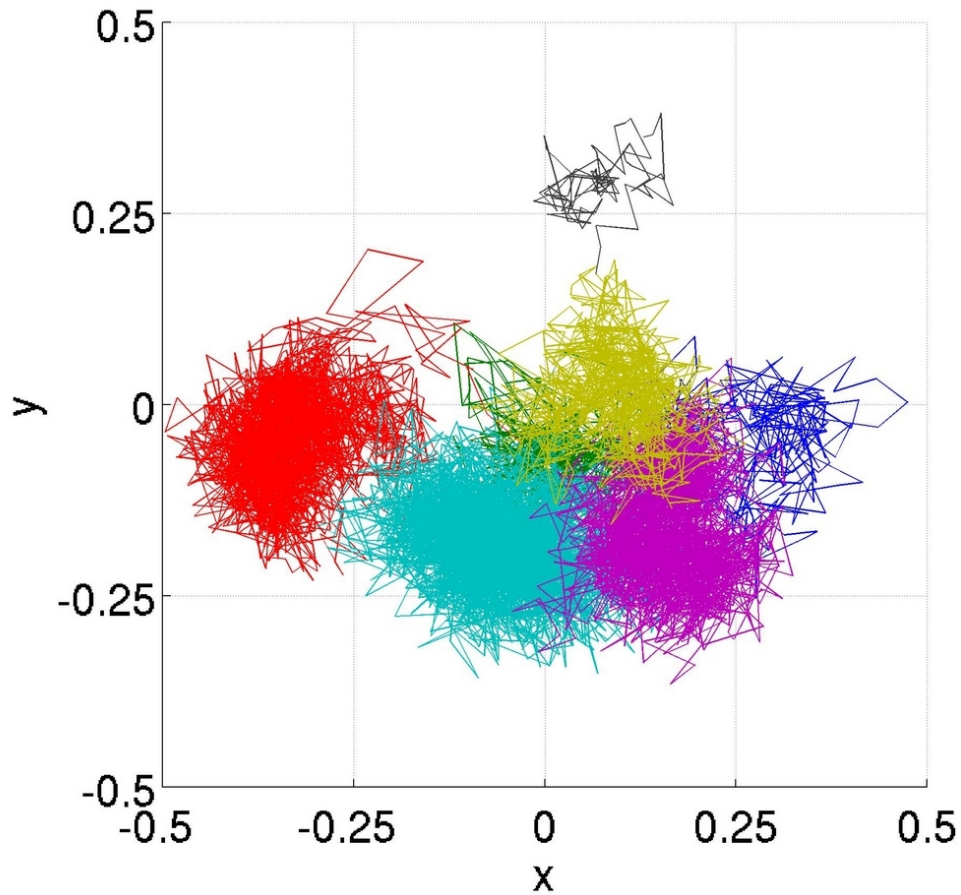
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Mean quadratic distance to the center of mass of the **other** set of points

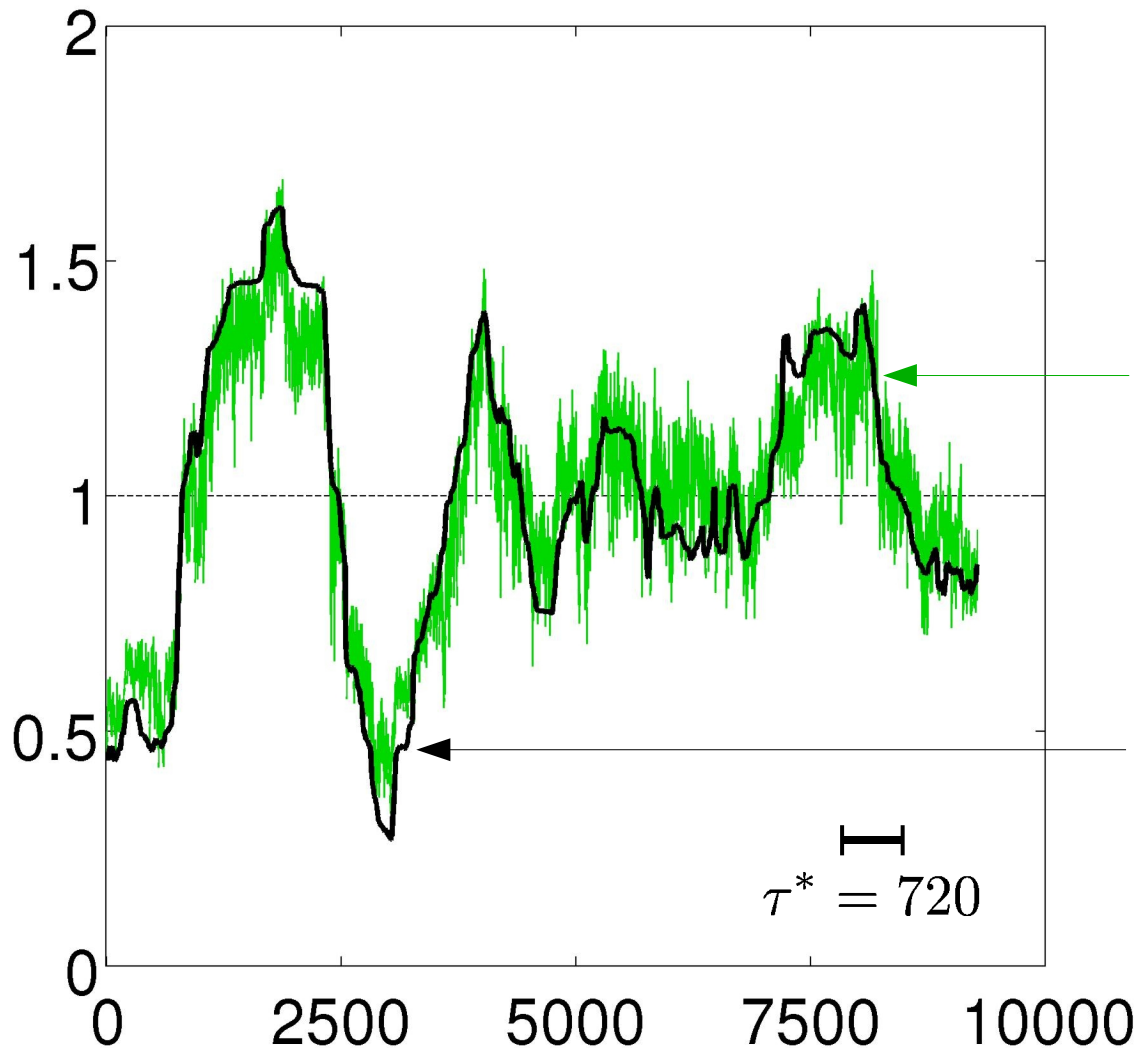


# Finding cage jumps (2)

Recomposition of a trajectory :



# Relaxation is dominated by cage jumps



$$Q_{\tau}(t) = \left\langle e^{-\frac{\Delta r^2(\tau)}{2a^2}} \right\rangle_p$$

Fluctuations of the dynamics

$$\frac{Q_{\tau^*}(t)}{\langle Q_{\tau^*} \rangle_t}$$

Relative number of particles that **haven't jumped** between  $t$  and  $t+\tau^*$

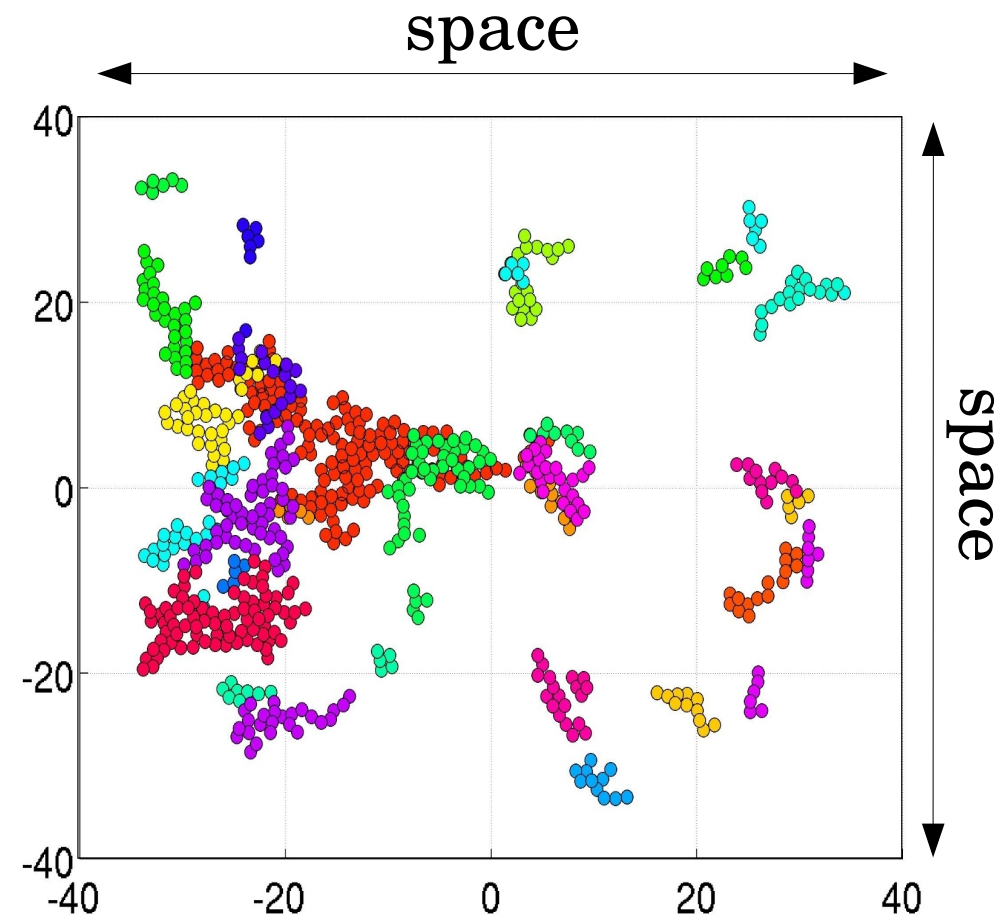
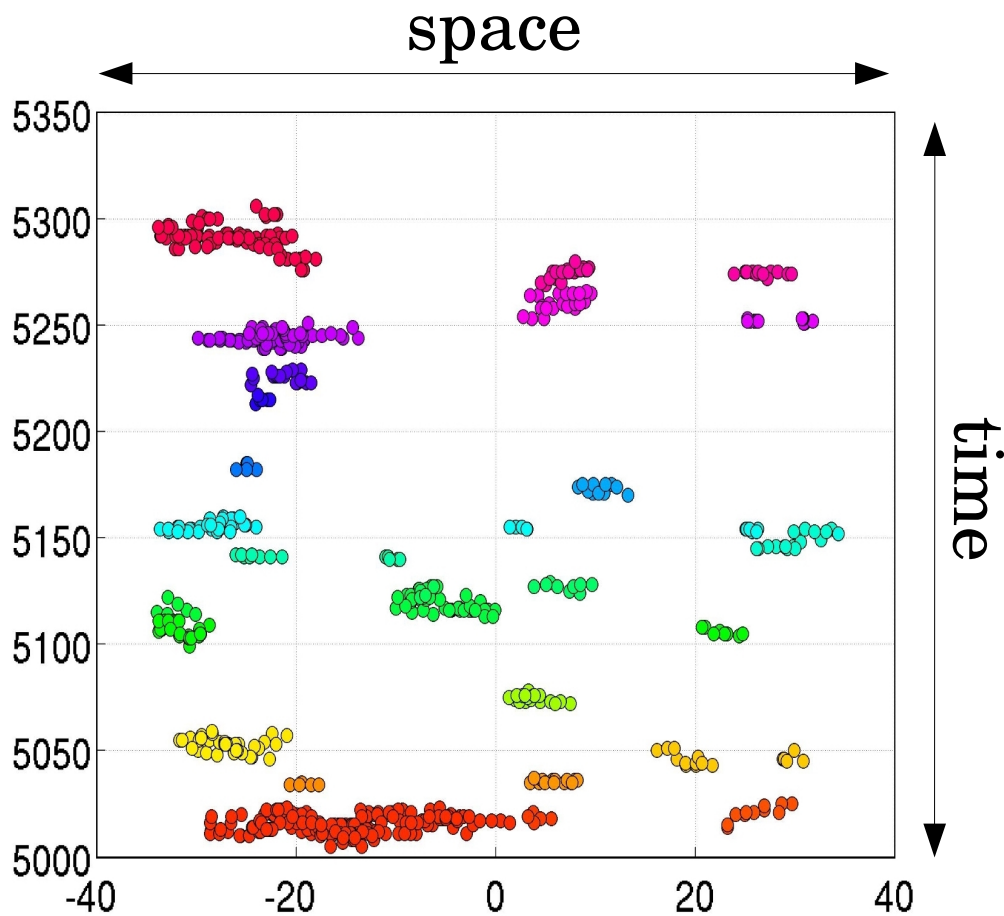
$$\frac{N_{\tau^*}(t)}{\langle N_{\tau^*} \rangle_t}$$

→ Cage jumps are enough to describe the relaxation



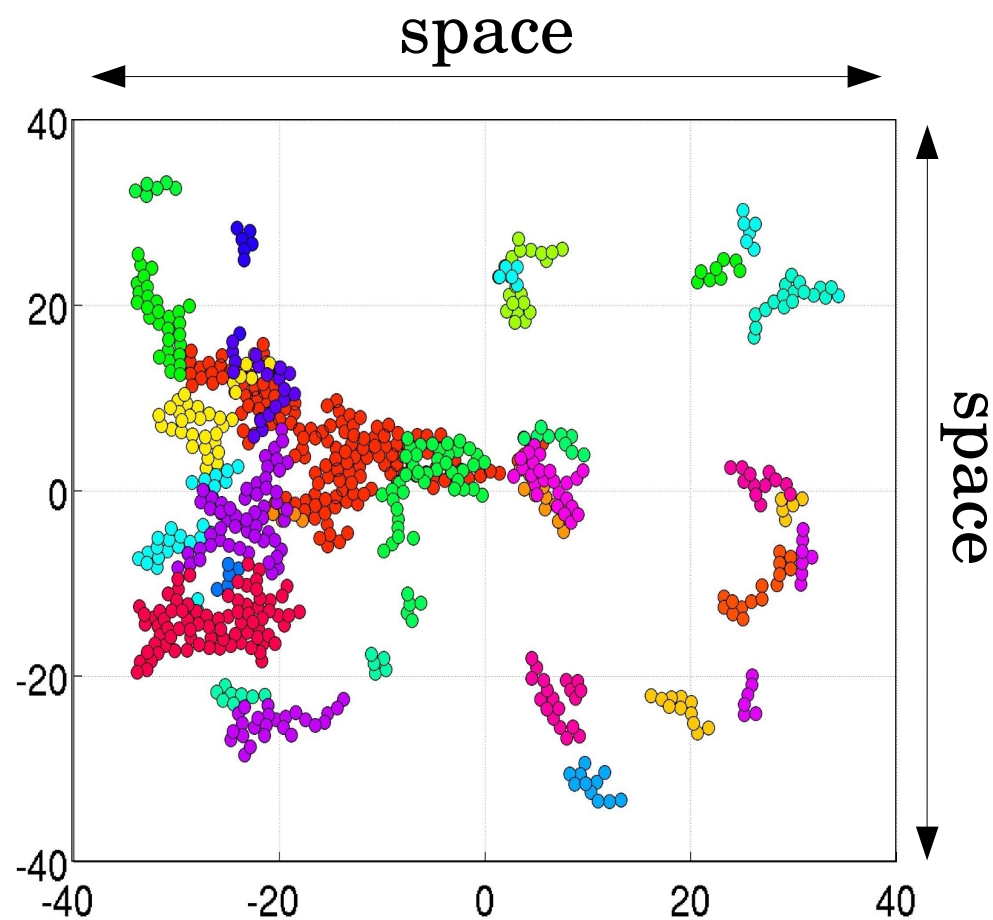
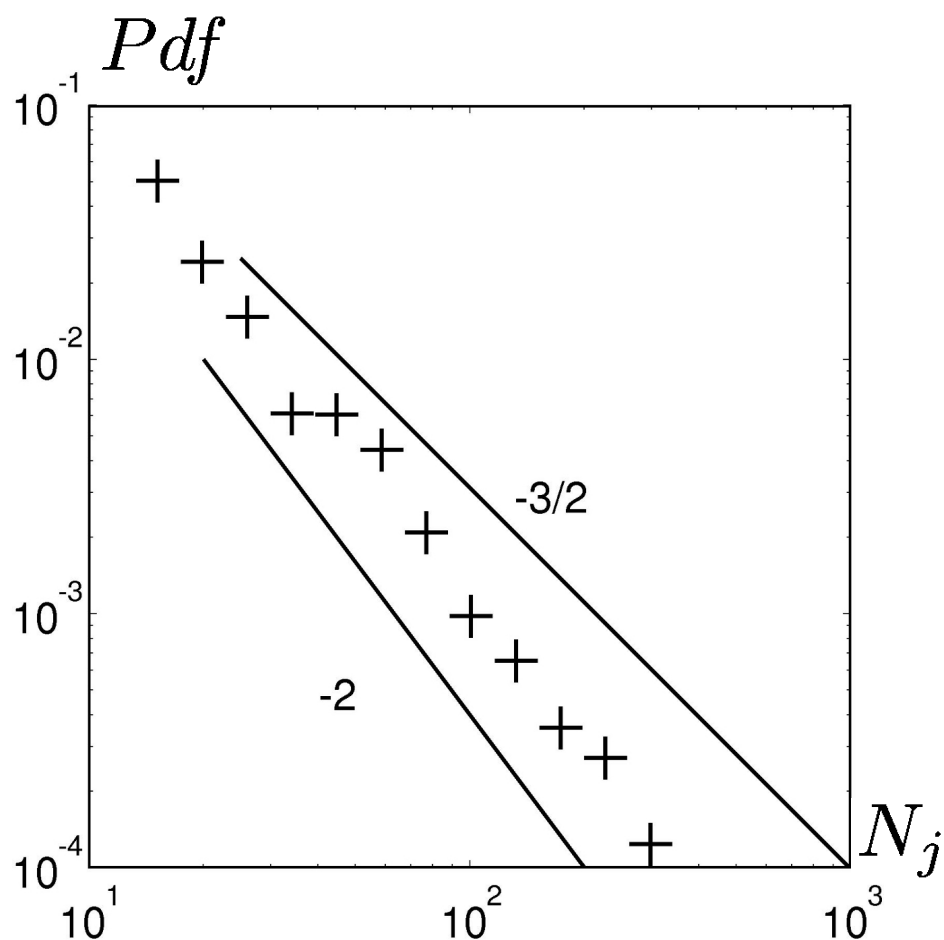
# Clusters of cage jumps

Cage jumps form **clusters** :  
quasi-instantaneous cooperative events



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Cage jumps form **clusters** :  
quasi-instantaneous cooperative events

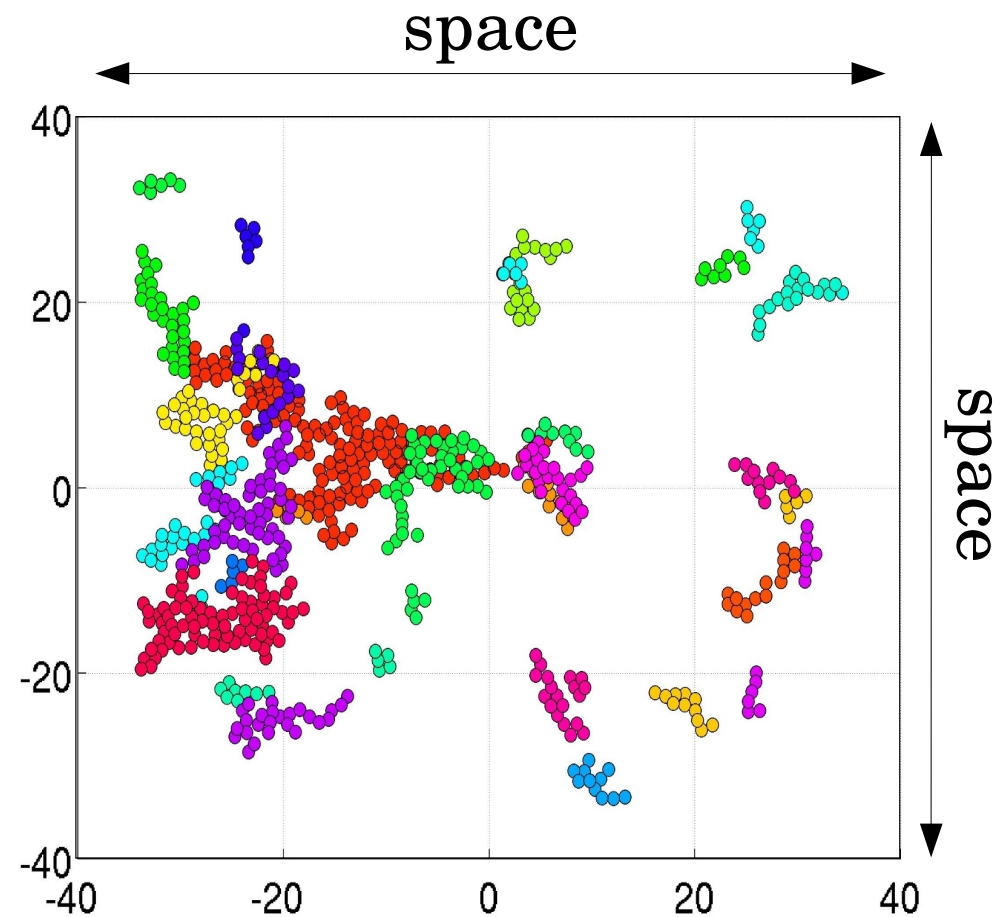
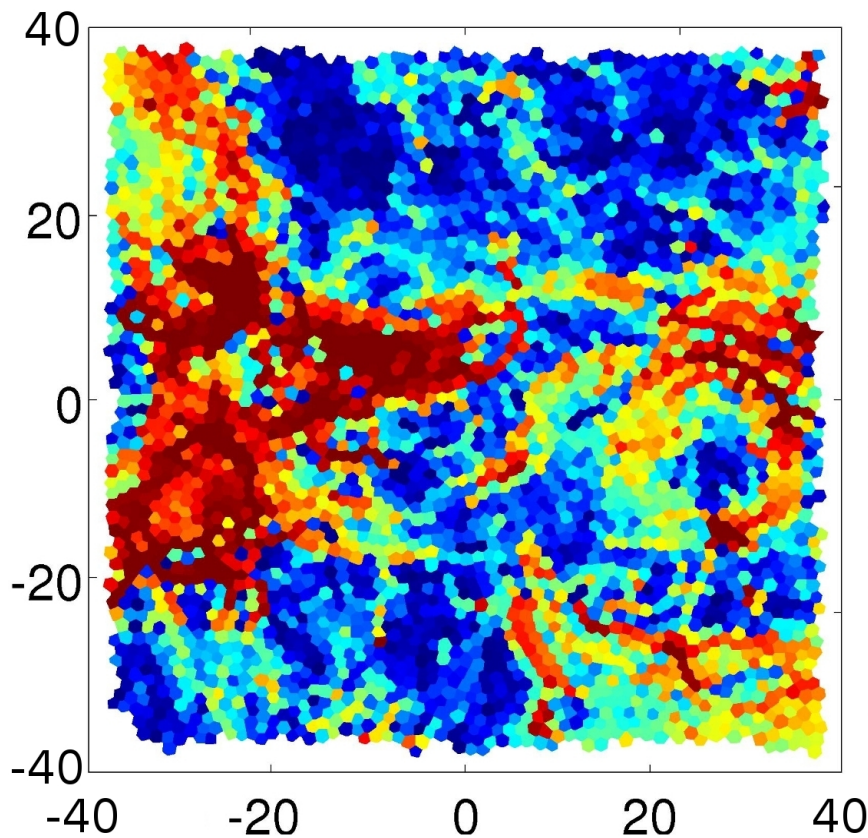


# Clusters of cage jumps

Cage jumps form **clusters** :  
quasi-instantaneous cooperative events



$$Q_{p,t} \equiv e^{-\frac{\Delta r^2(\tau)}{2a^2}}$$

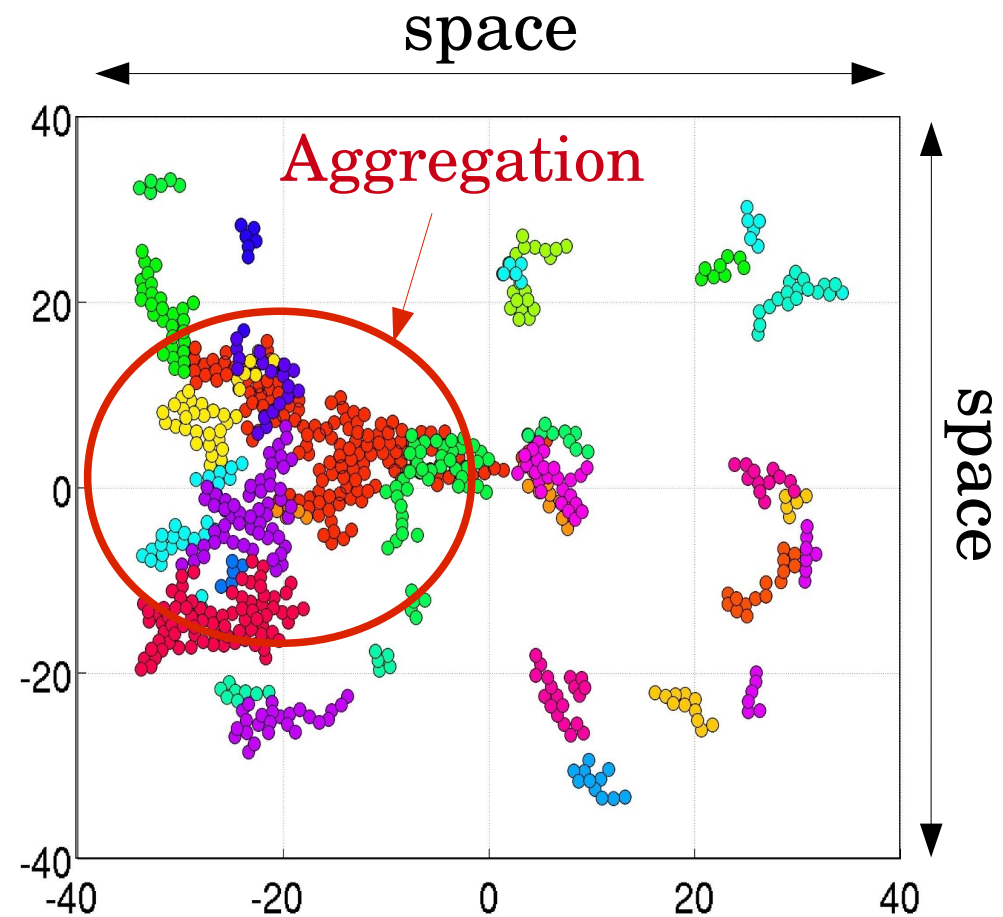
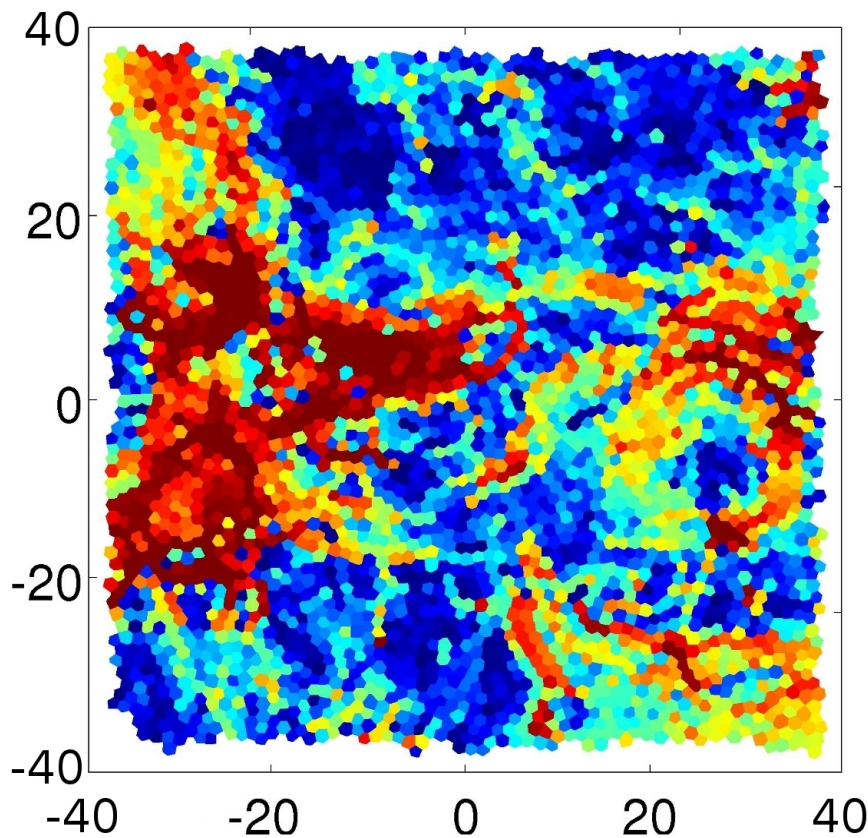


# Clusters of cage jumps

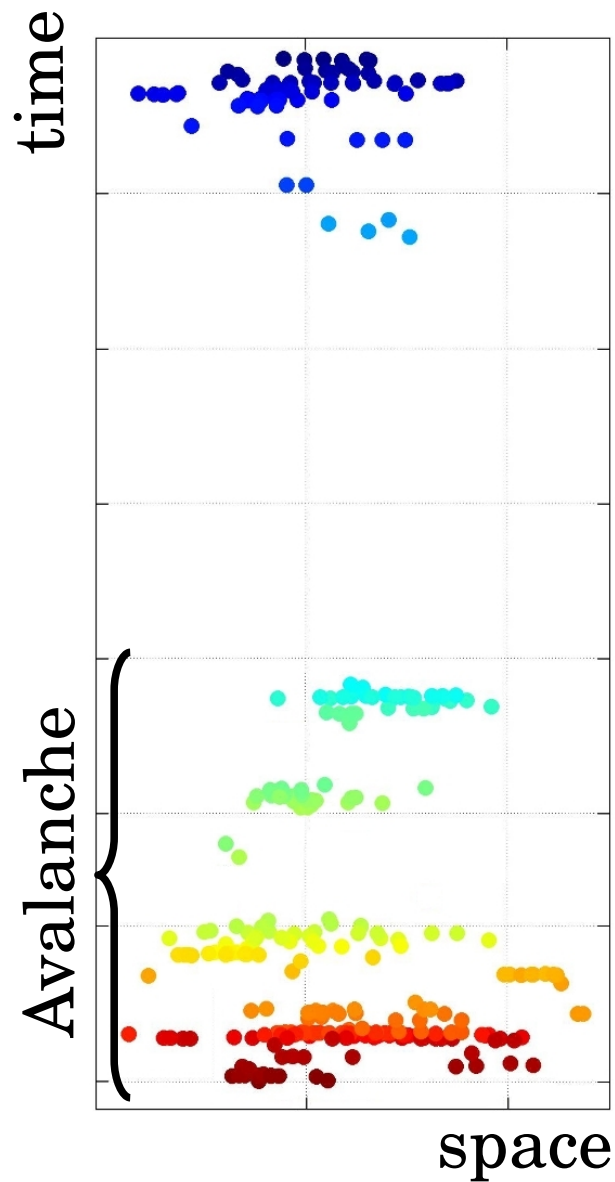
Cage jumps form **clusters** :  
quasi-instantaneous cooperative events



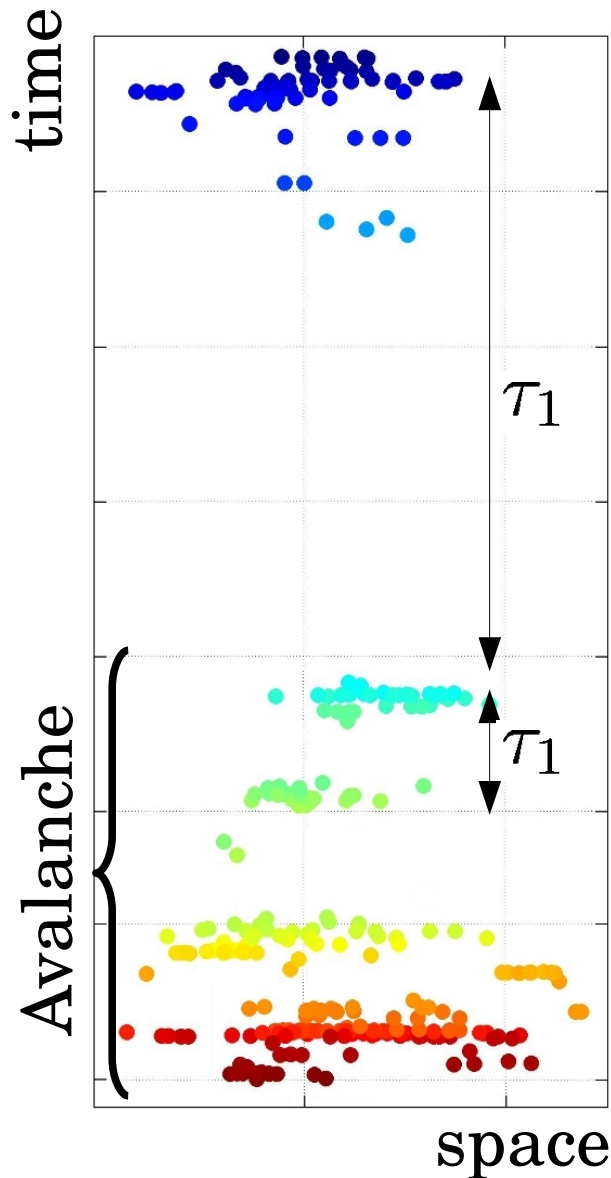
$$Q_{p,t} \equiv e^{-\frac{\Delta r^2(\tau)}{2a^2}}$$



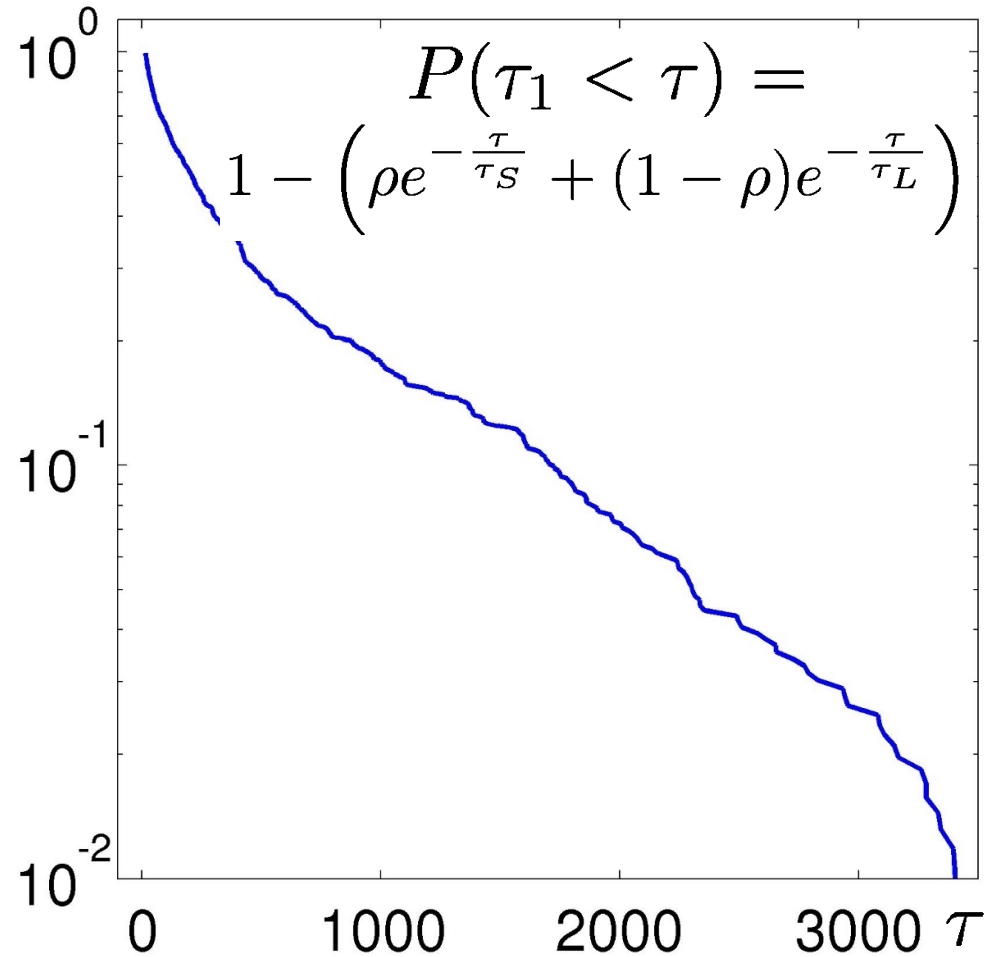
# A two-timescale process



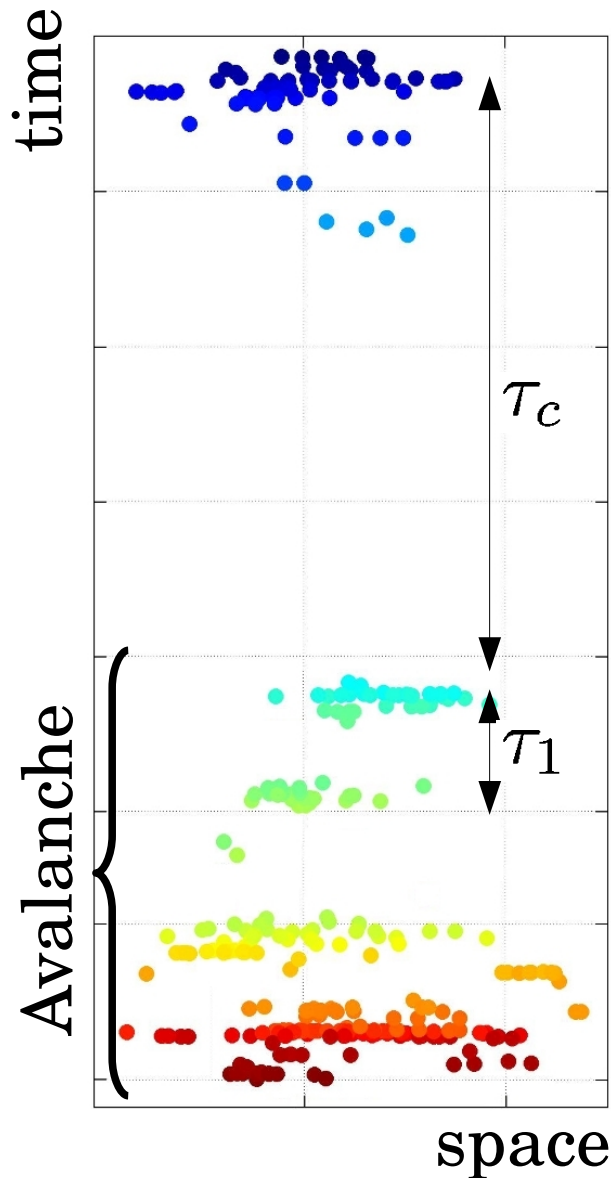
# A two-timescale process



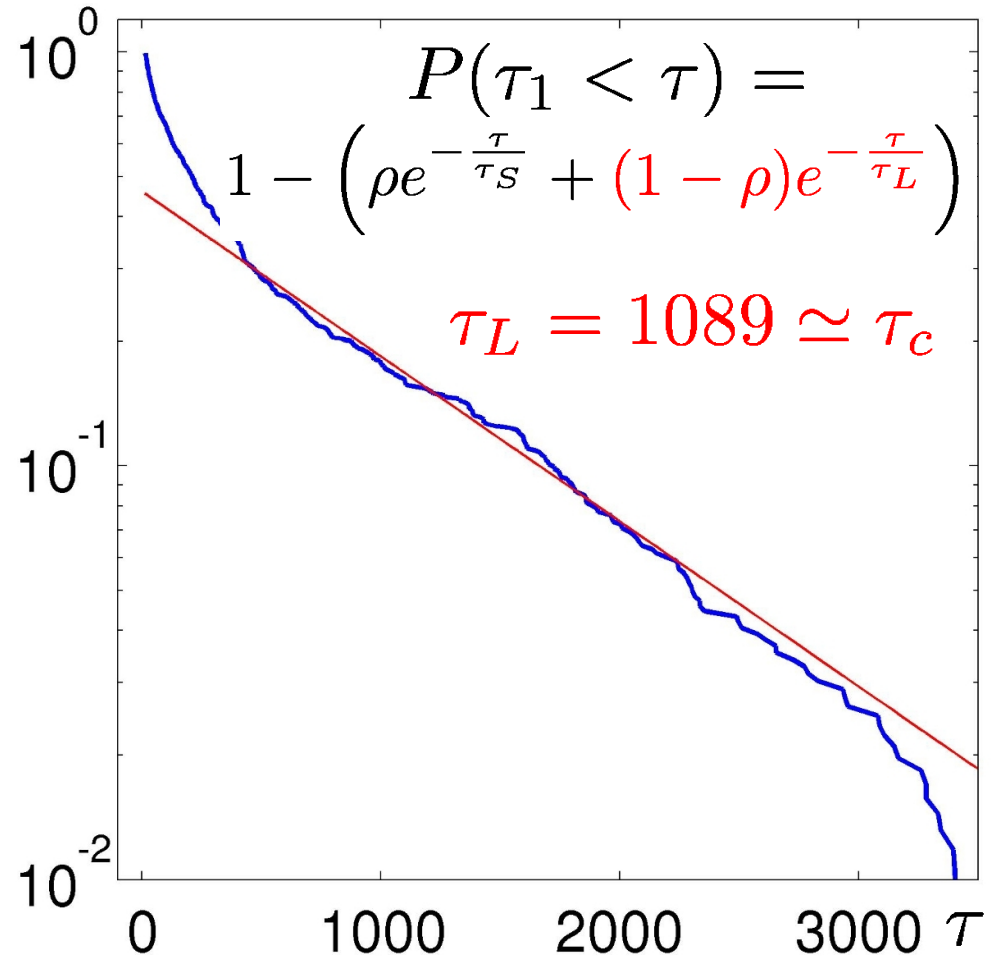
Distribution of the time  $\tau_1$   
to the 1<sup>st</sup> next adjacent cluster



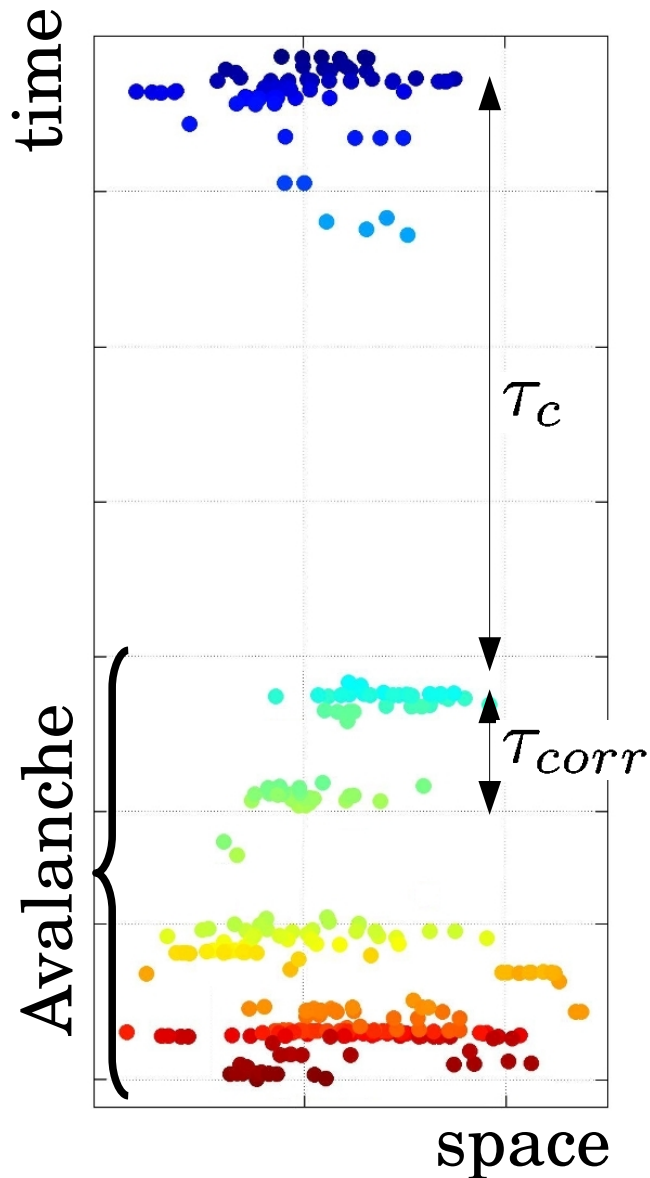
# A two-timescale process



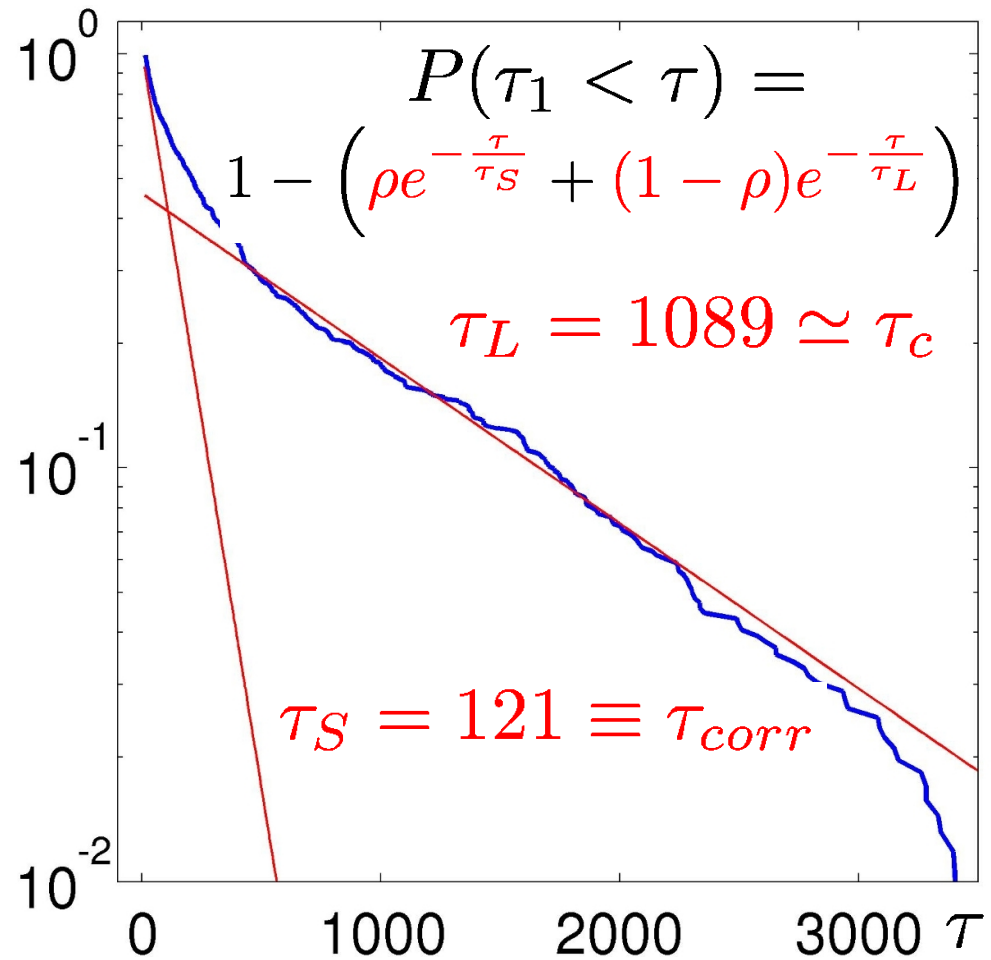
Distribution of the time  $\tau_1$   
to the 1<sup>st</sup> next adjacent cluster



# A two-timescale process

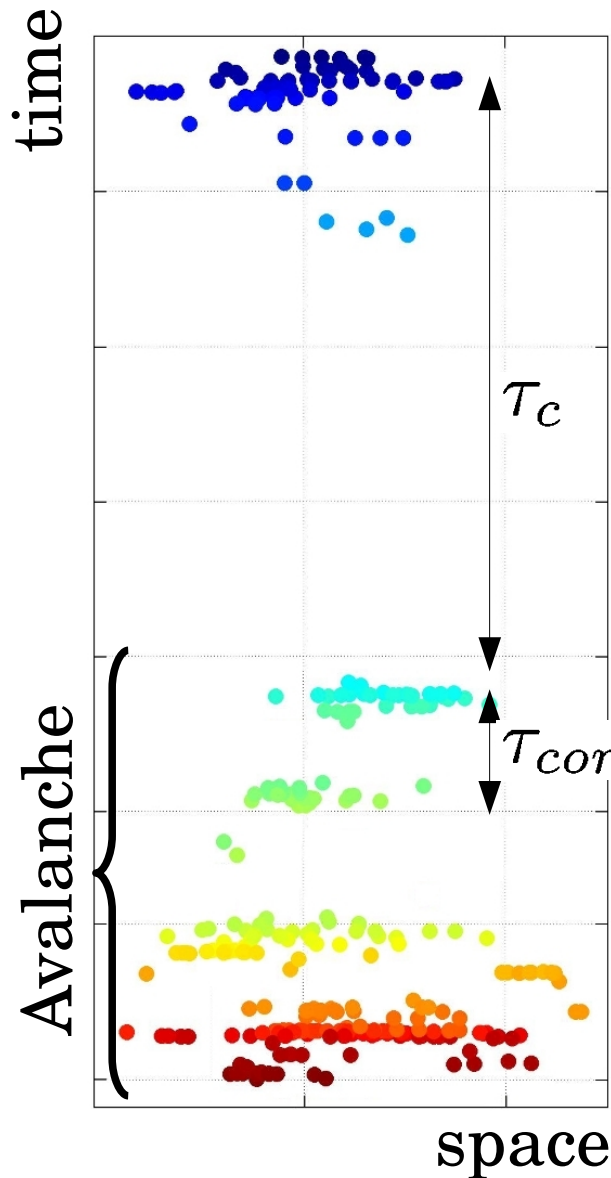


Distribution of the time  $\tau_1$   
to the 1<sup>st</sup> next adjacent cluster



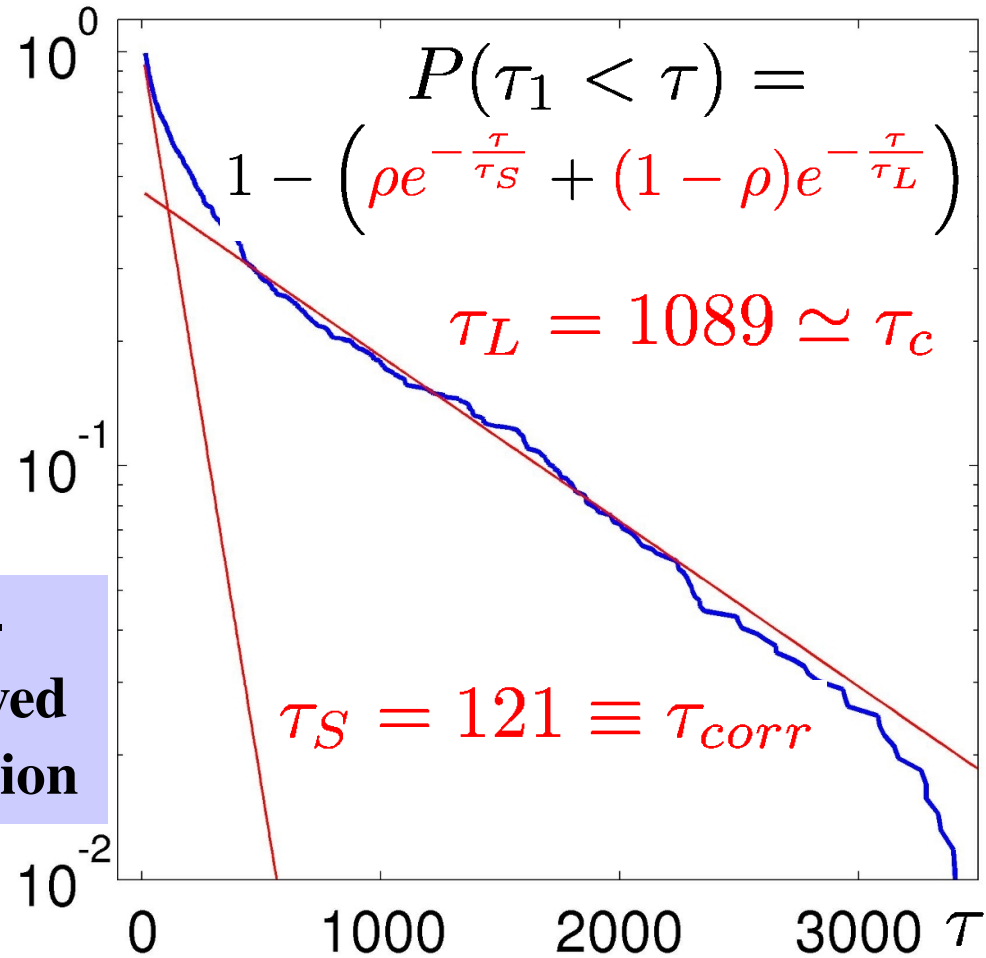


# A two-timescale process

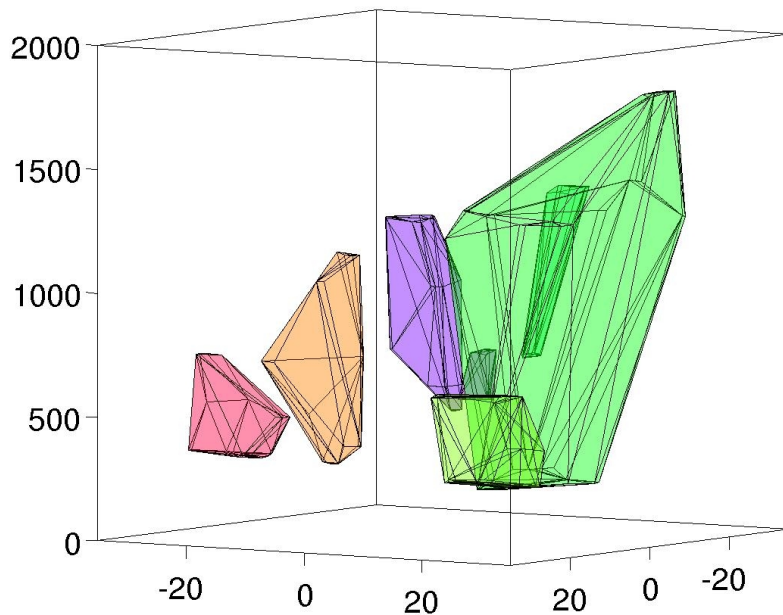
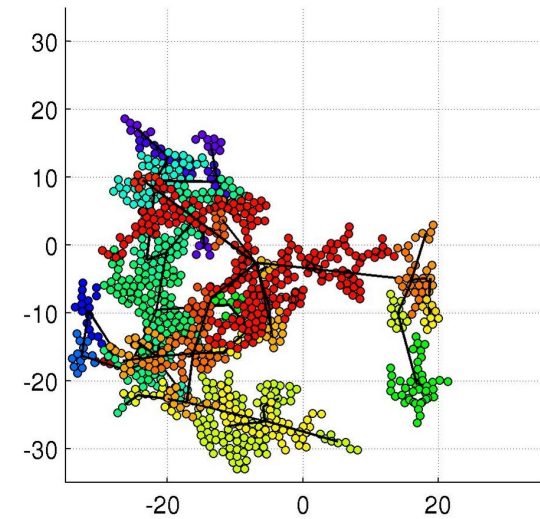
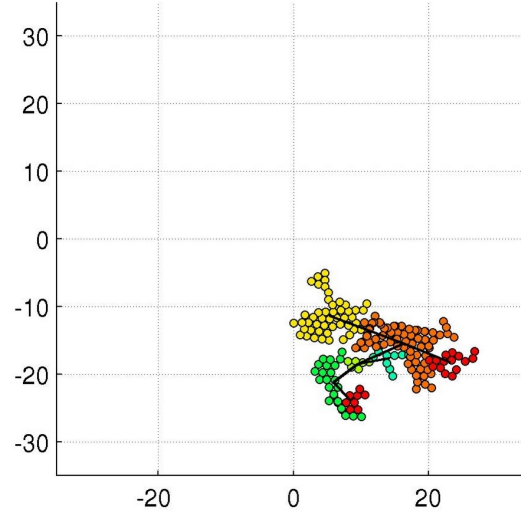
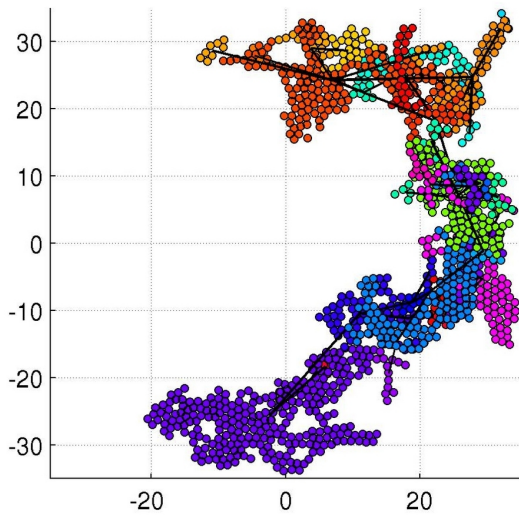


Non-  
conserved  
facilitation

Distribution of the time  $\tau_1$   
to the 1<sup>st</sup> next adjacent cluster

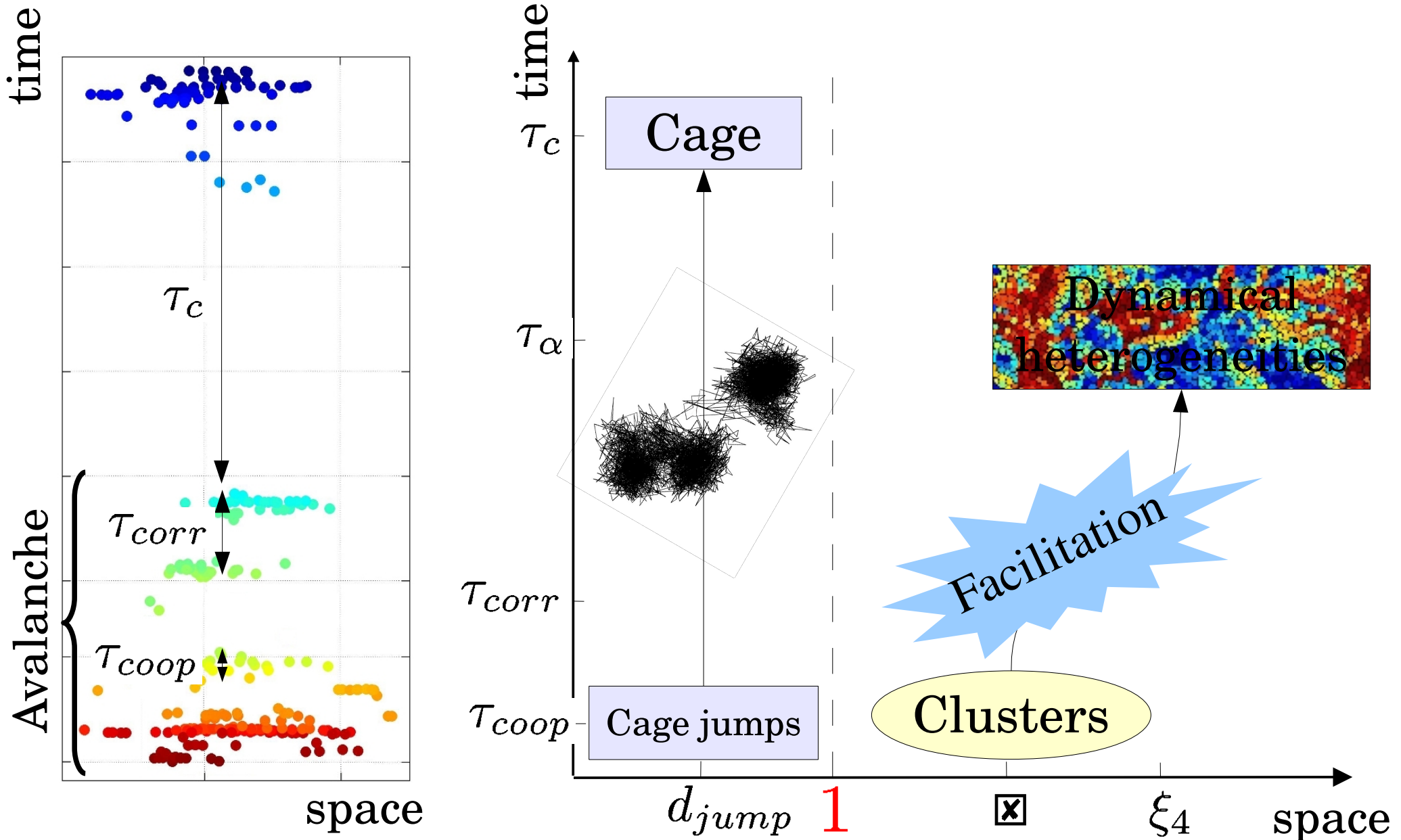


# Avalanches

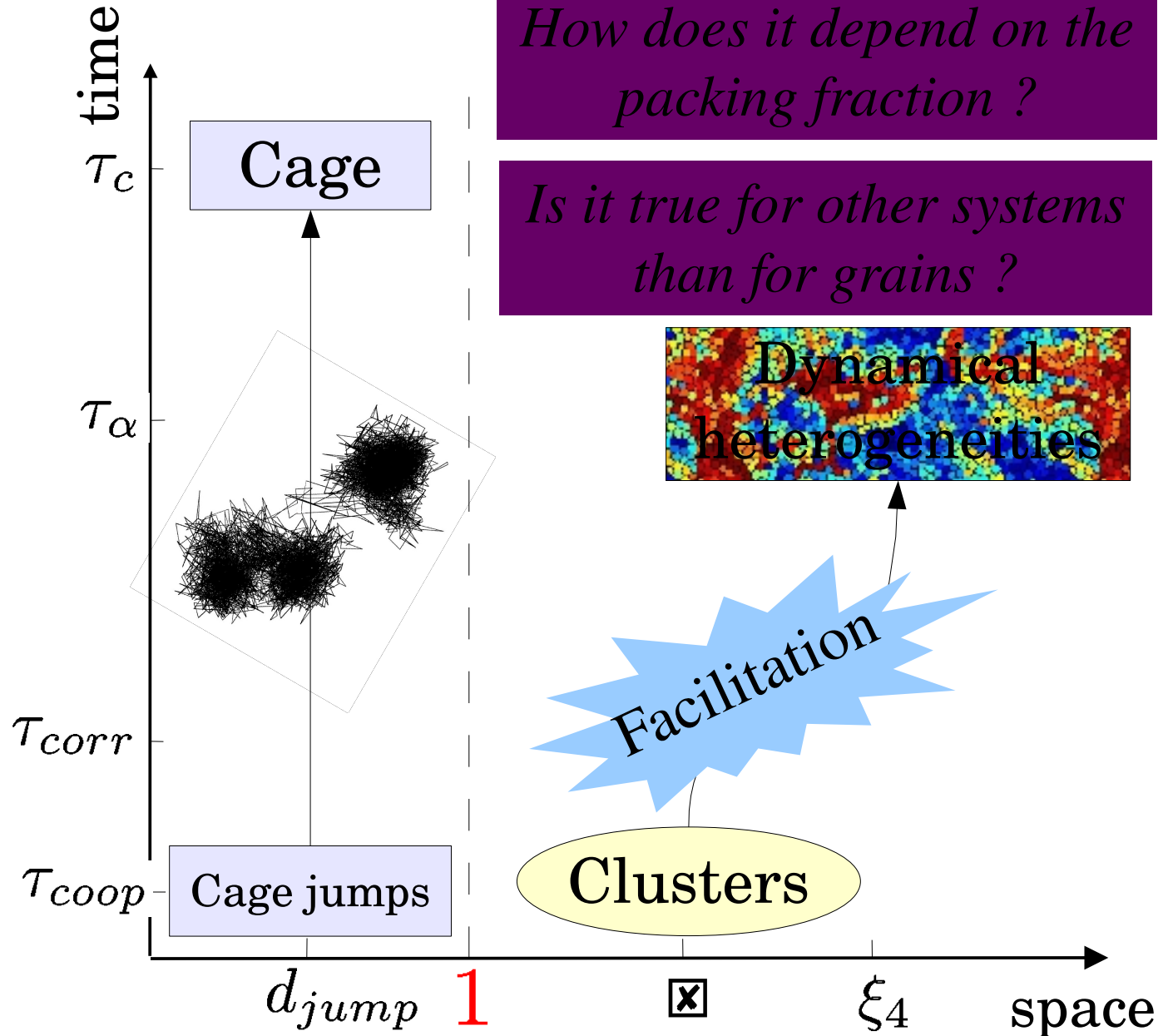
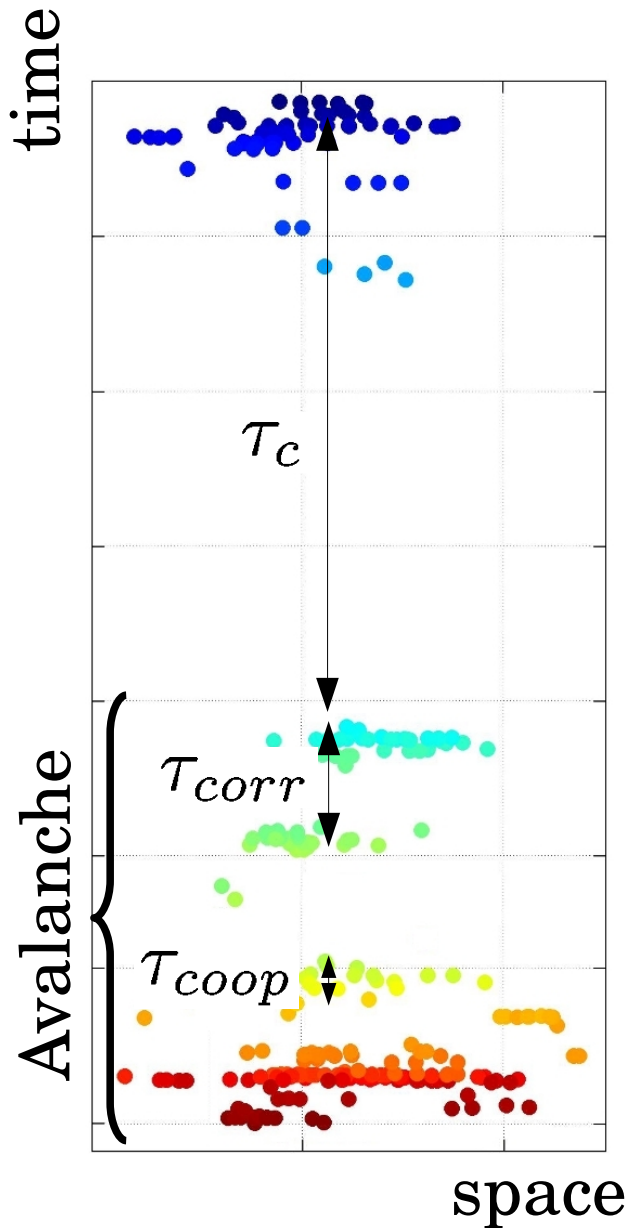


- Many different sizes and shapes
- A finite distance between avalanches
- Avalanches occur in distinct regions of space

# Summary of time and length scales



# Summary of time and length scales

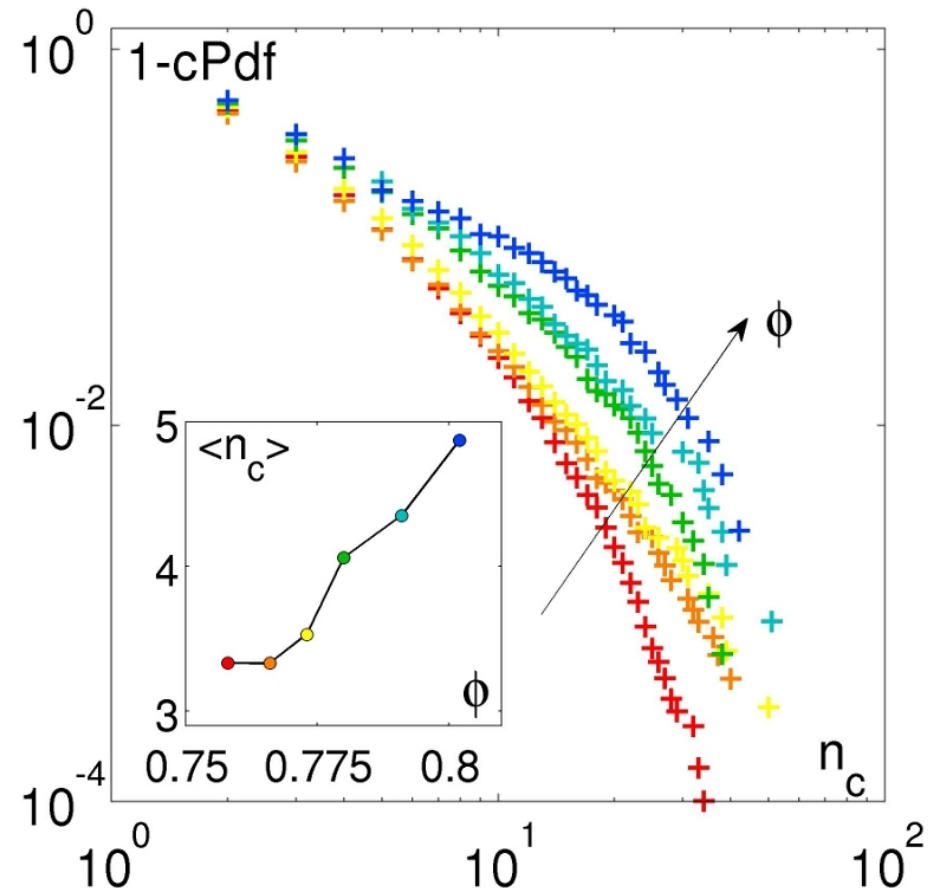
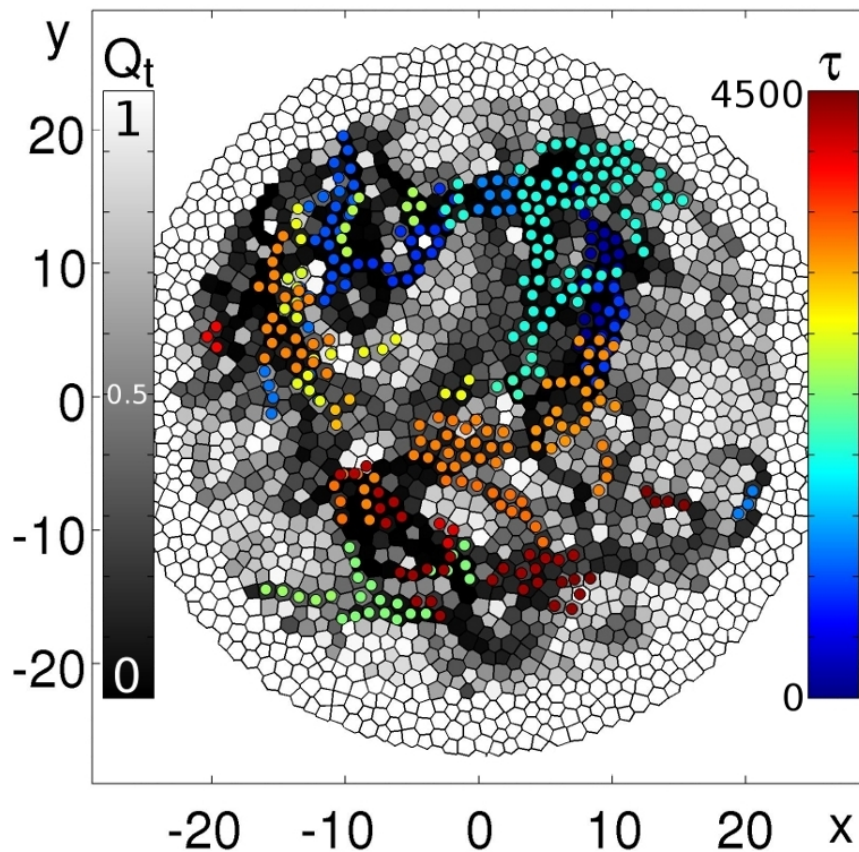


# The fluidized bed experiment

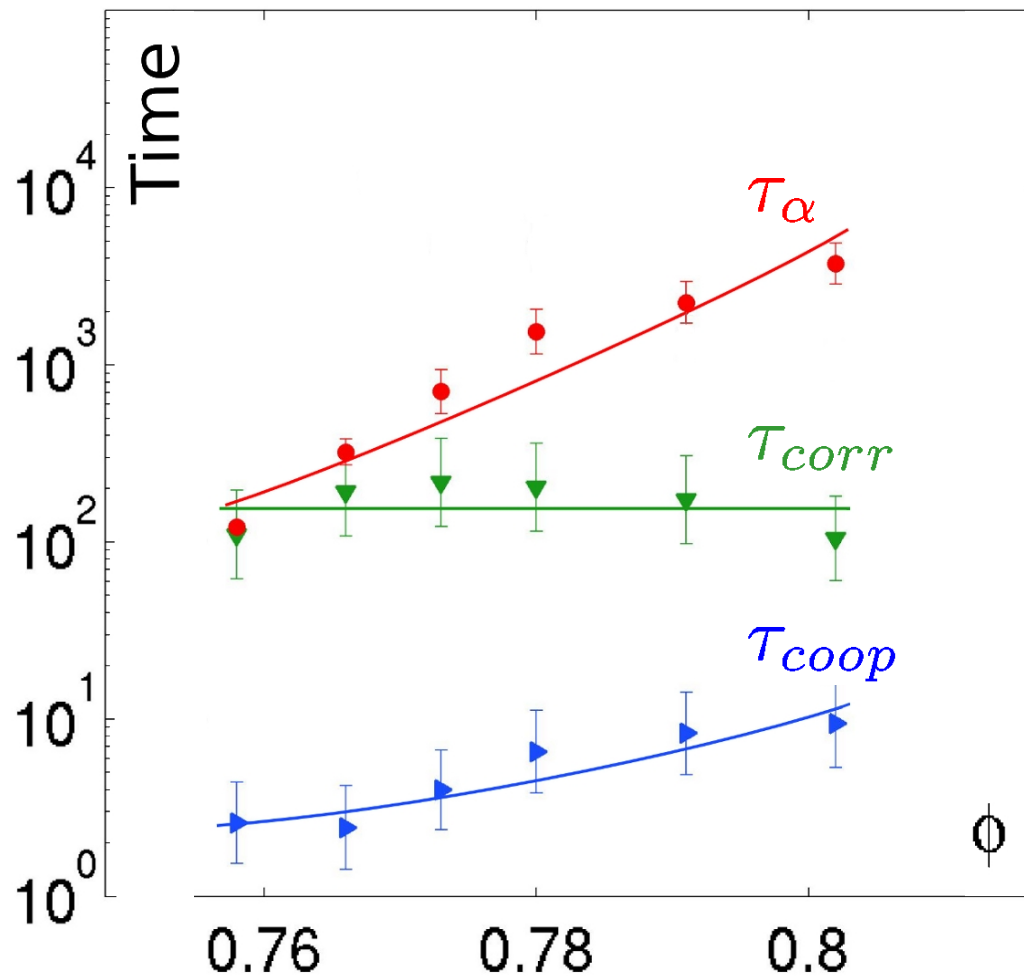
→ *Fluidized bed experiment*



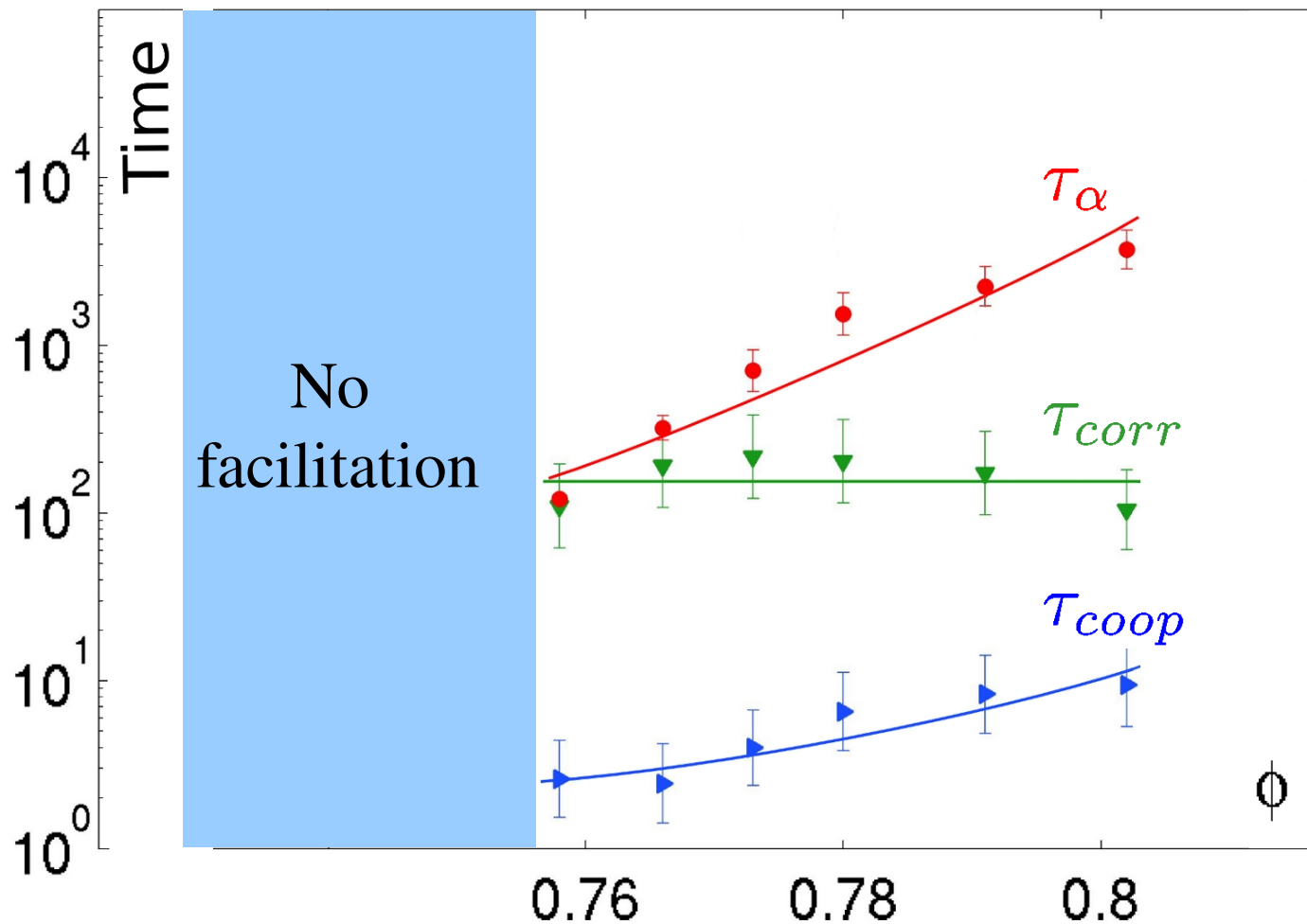
- Same observations !
- Clusters have large tail distributions
- They grow as the packing fraction increase



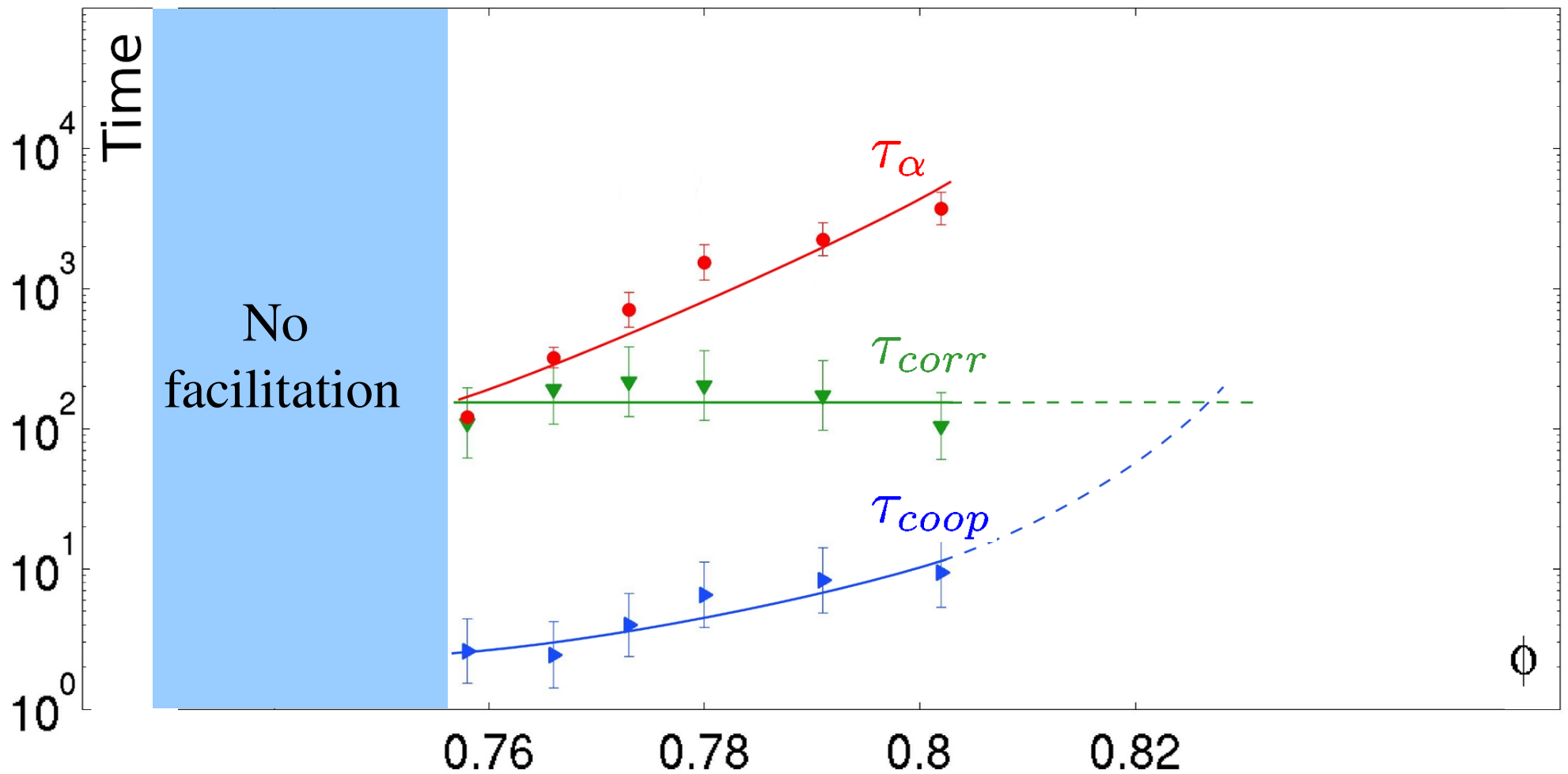
# Evolution of timescales



# Evolution of timescales

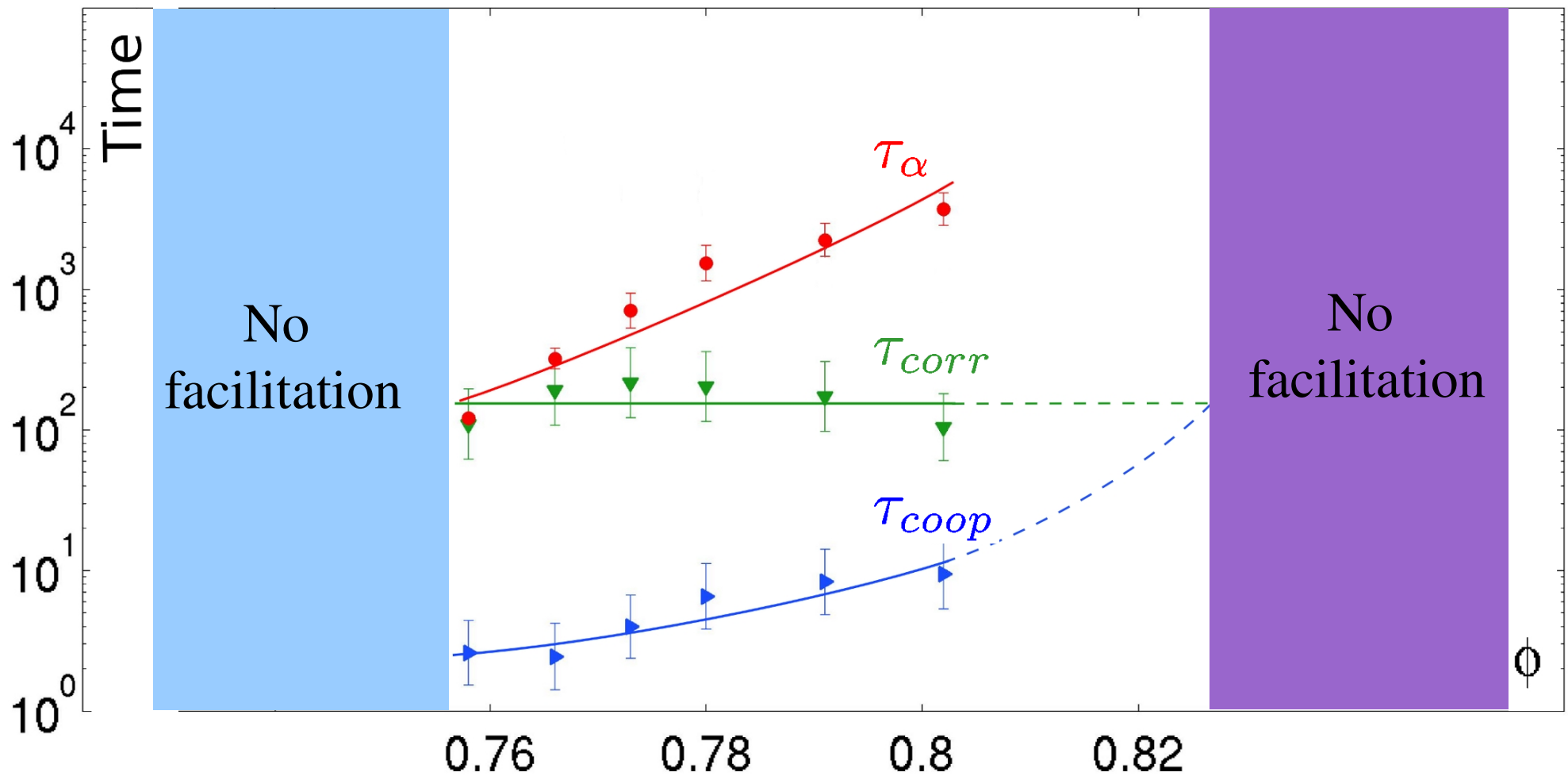


# Evolution of timescales

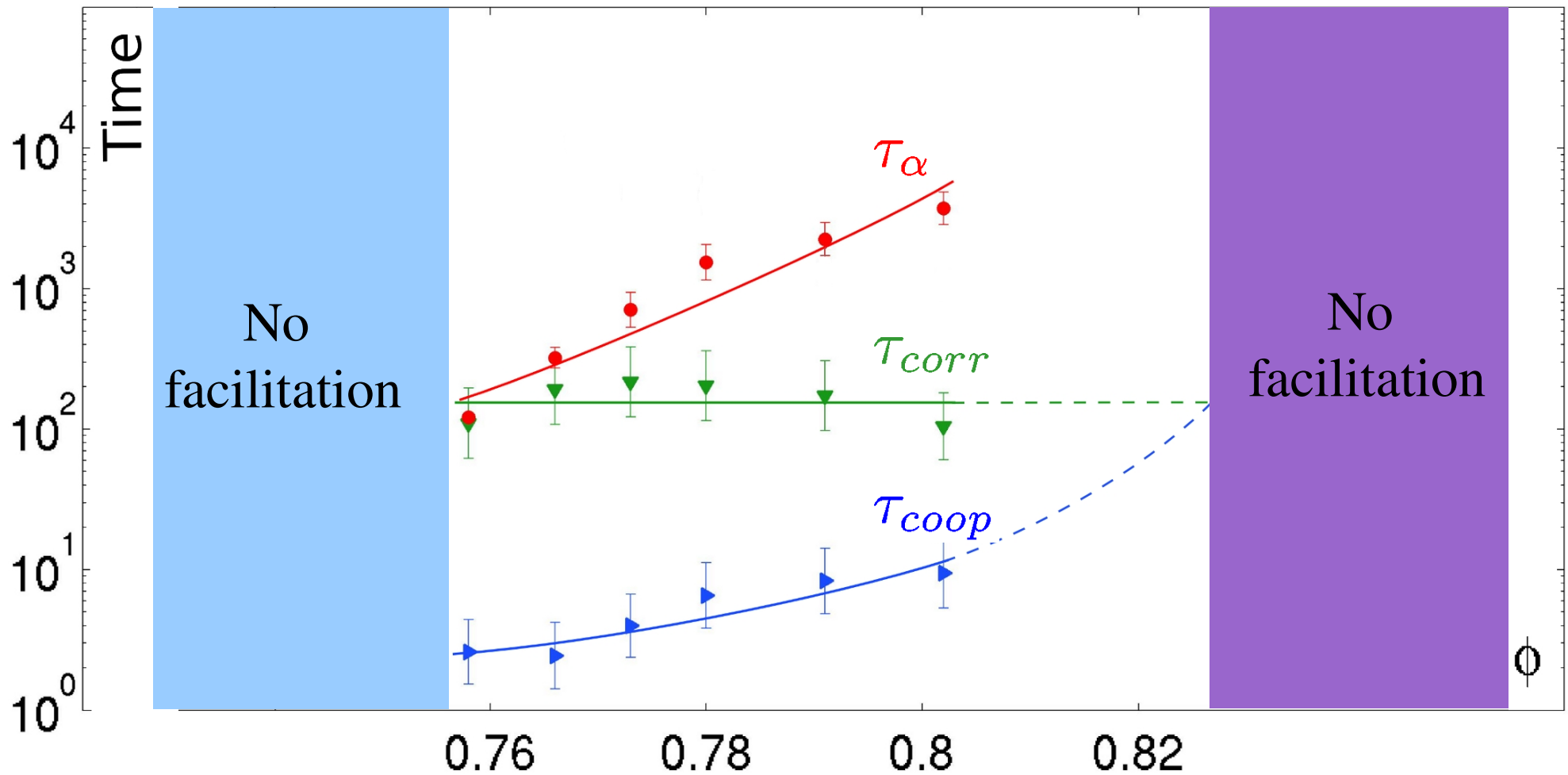




# Evolution of timescales



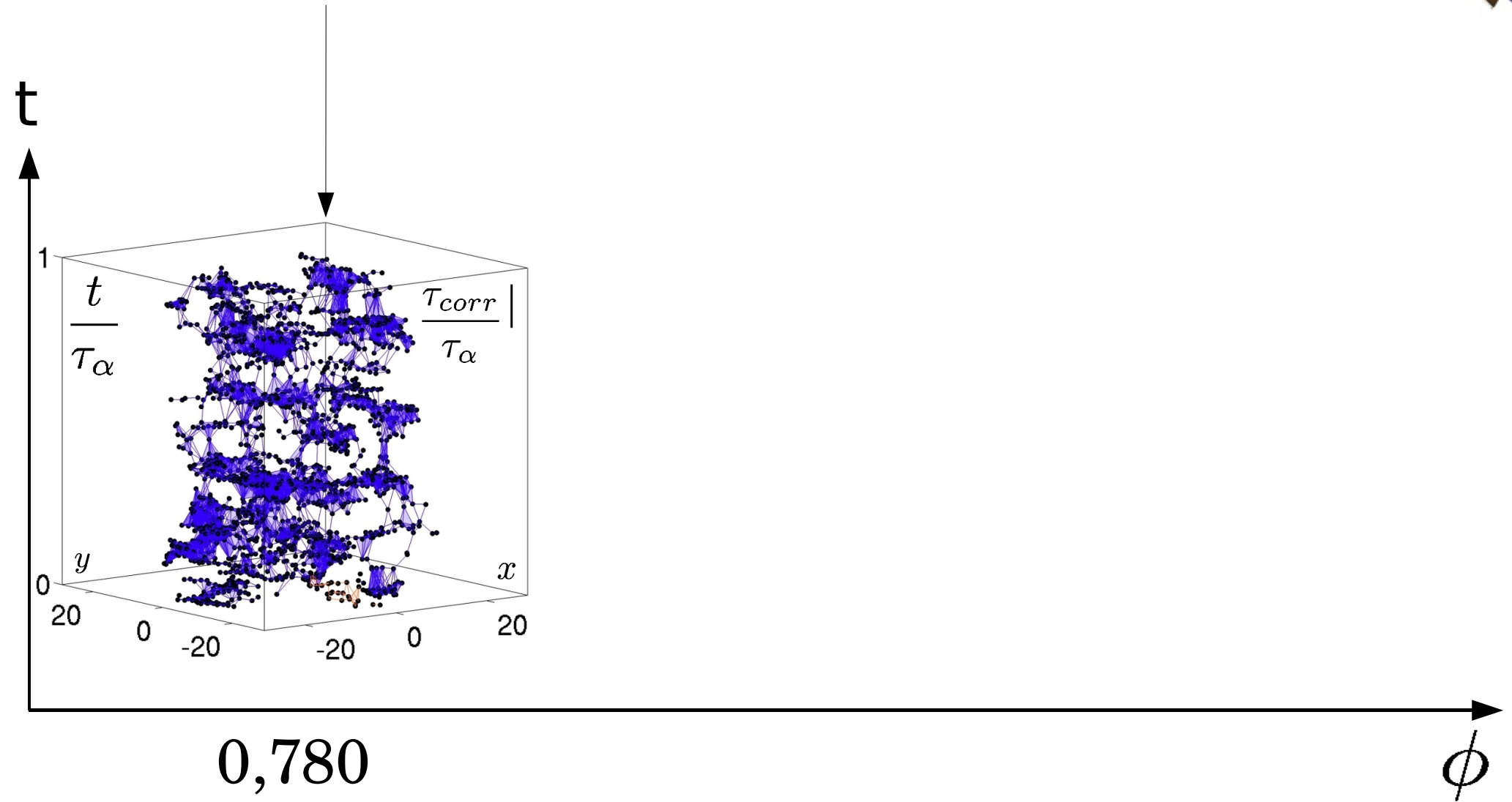
# Evolution of timescales



Is facilitation conserved ?

# Evolution with the packing fraction

An **entangled network** at low  $\phi$ : *conserved facilitation*

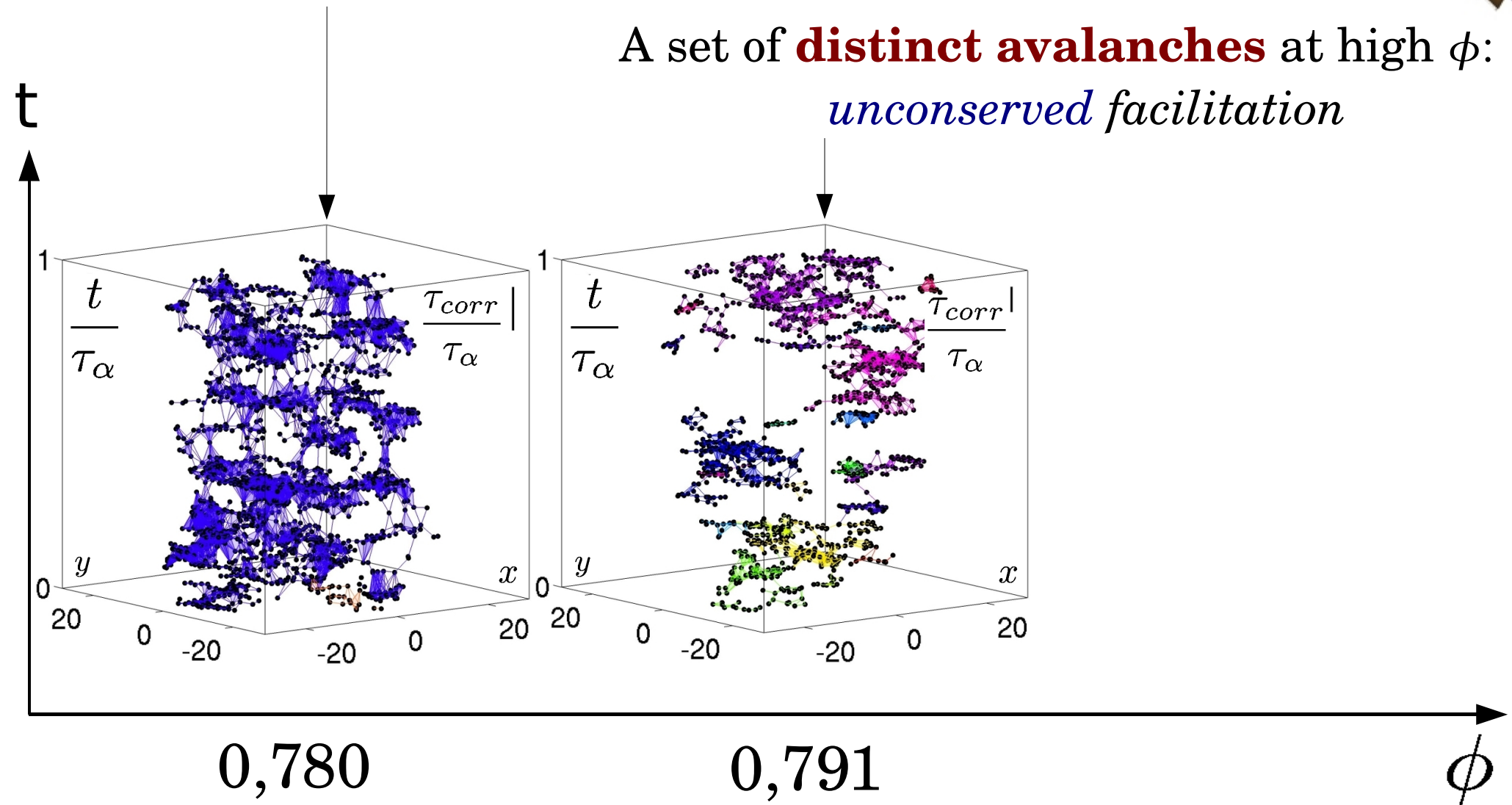


# Evolution with the packing fraction

An **entangled network** at low  $\phi$ : *conserved facilitation*



A set of **distinct avalanches** at high  $\phi$ :  
*unconserved facilitation*

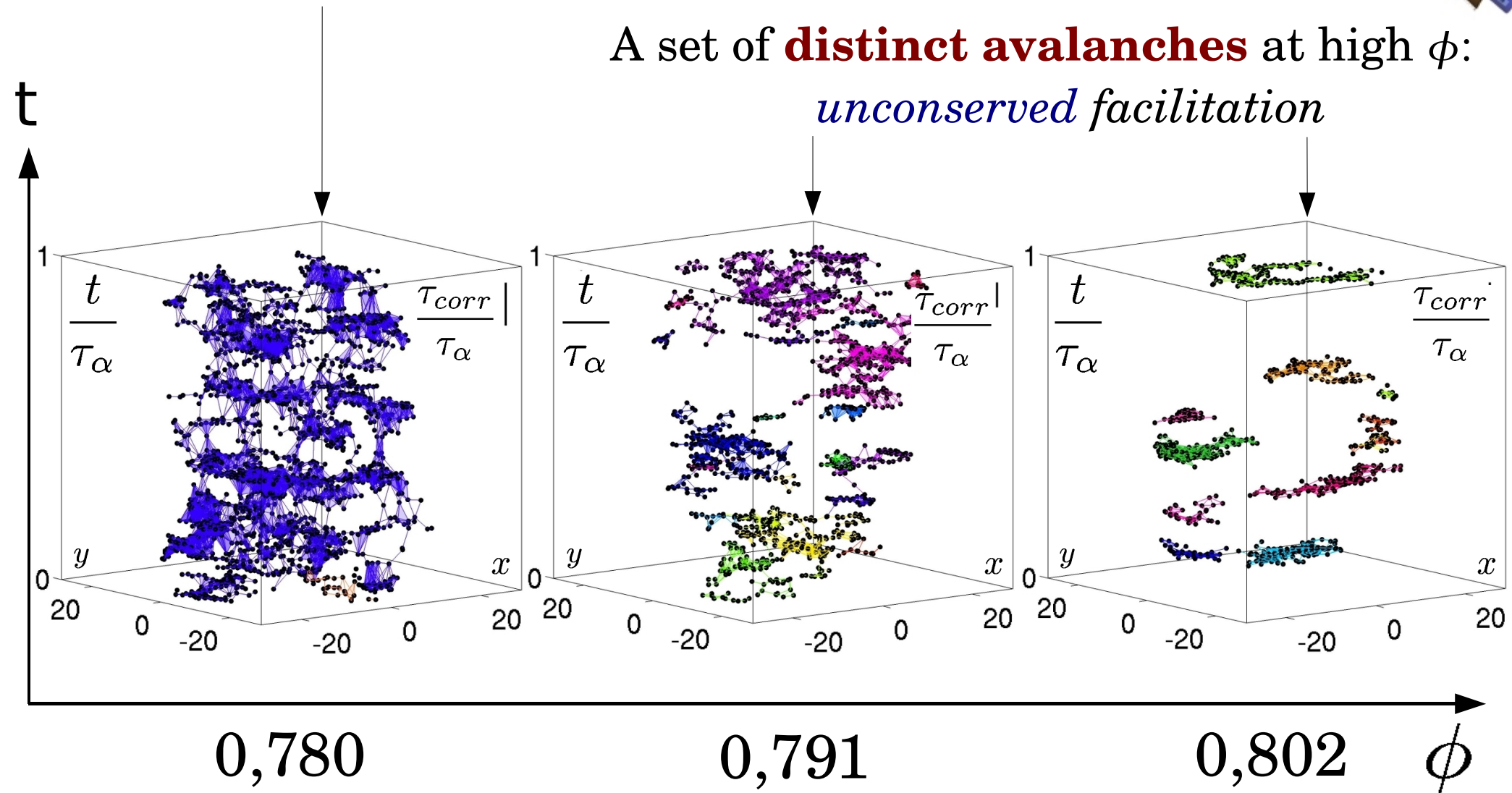


# Evolution with the packing fraction

An **entangled network** at low  $\phi$ : *conserved facilitation*

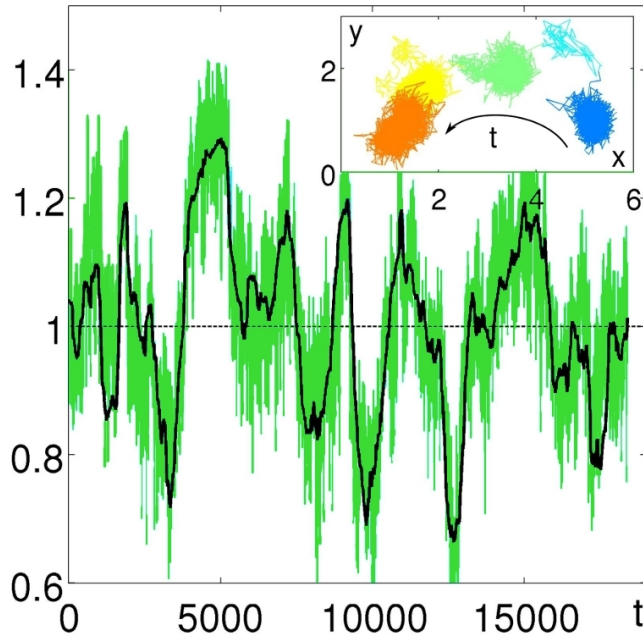


A set of **distinct avalanches** at high  $\phi$ :  
*unconserved facilitation*

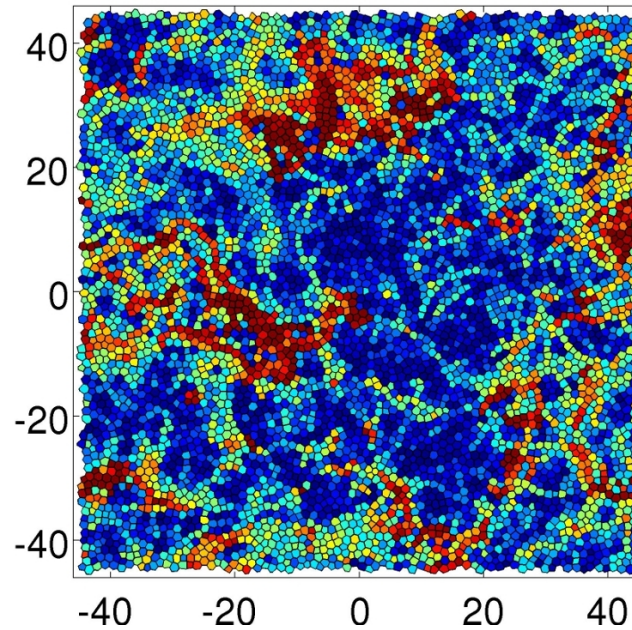


# When « real liquids » jump

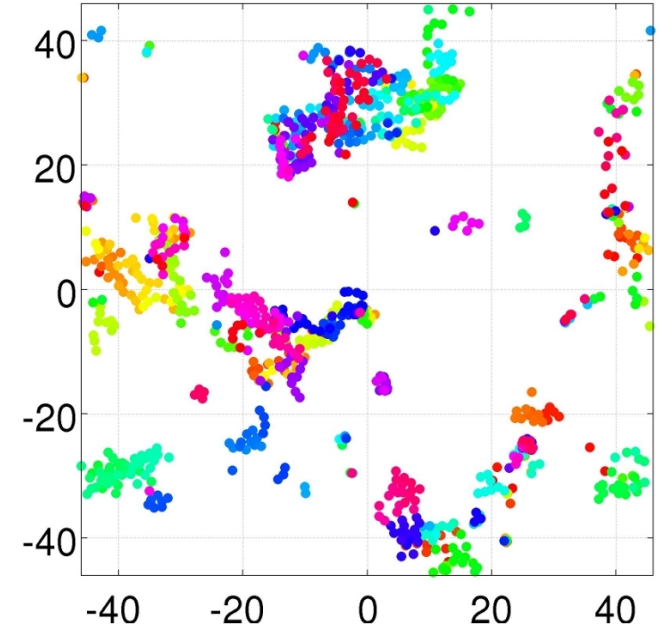
→ *Simulation*



$$\frac{Q_{\tau^*}(t)}{\langle Q_{\tau^*} \rangle_t}$$



$$Q_{p,t}(a^*, \tau^*)$$



*Clusters of cage jumps*

Despite the systems are very different,  
the phenomenology is similar to granular materials

# Roadmap

## Systems

- Why granular media ?
- Systems investigated during my thesis



## Part 1 : Heterogeneous relaxations

- From granular media to Lennard-Jones liquids

## Part 2 : The Building Blocks of Dynamical Heterogeneities

- From cage jumps to avalanches, a non conserved facilitation
- Evolution of dynamical facilitation with  $\phi$
- An identical scenario for Lennard-Jones liquids

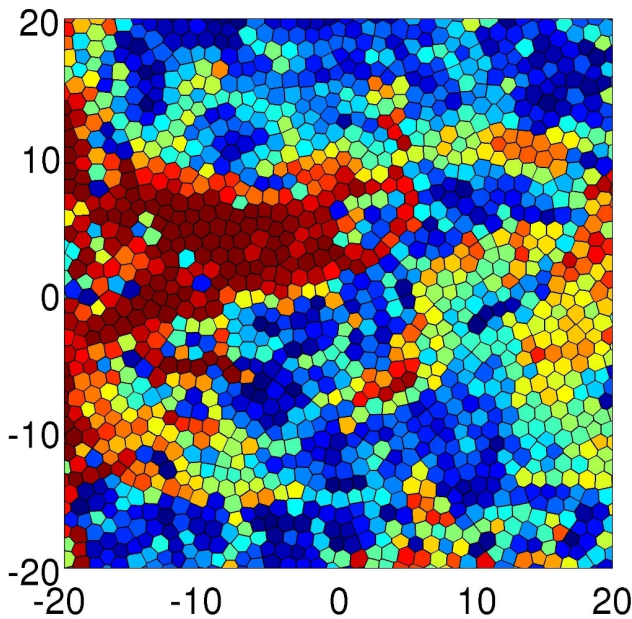
## Part 3 : Relation to Structure ?

- No clear signature of the dynamics in the structure
- More on simulated liquids : the isoconfigurational ensemble

## Conclusion and perspectives

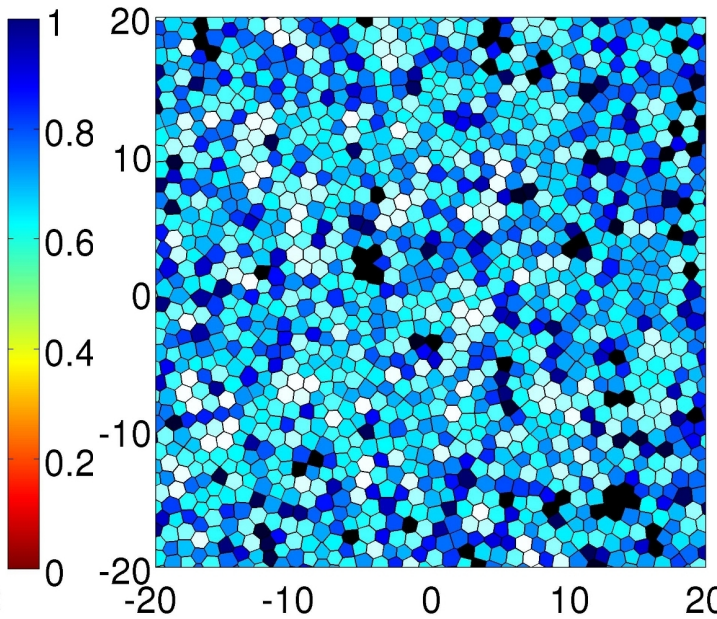
# No obvious relation in our systems

- Some attempts to correlate structure and dynamics :



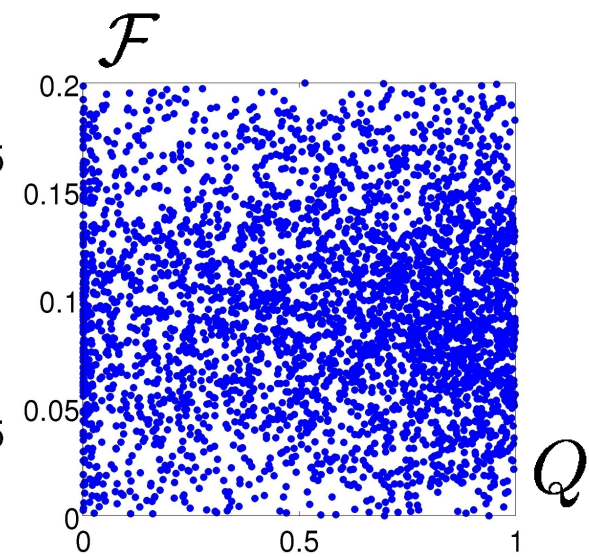
**Local relaxation**

$$Q_{p,t}(a^*, \tau^*)$$



**Free volume**

$$\mathcal{F}(p) = \frac{S_{VC} - S_{hex}}{S_{hex}}$$



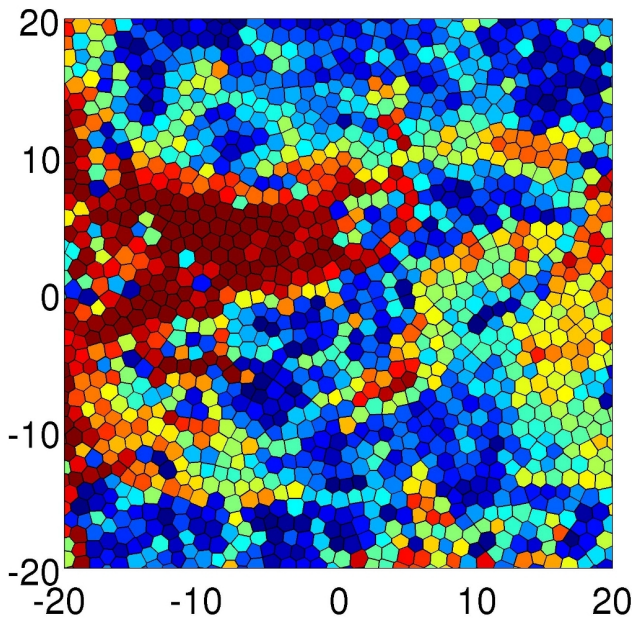
Correlation :

**-0.0189 X**



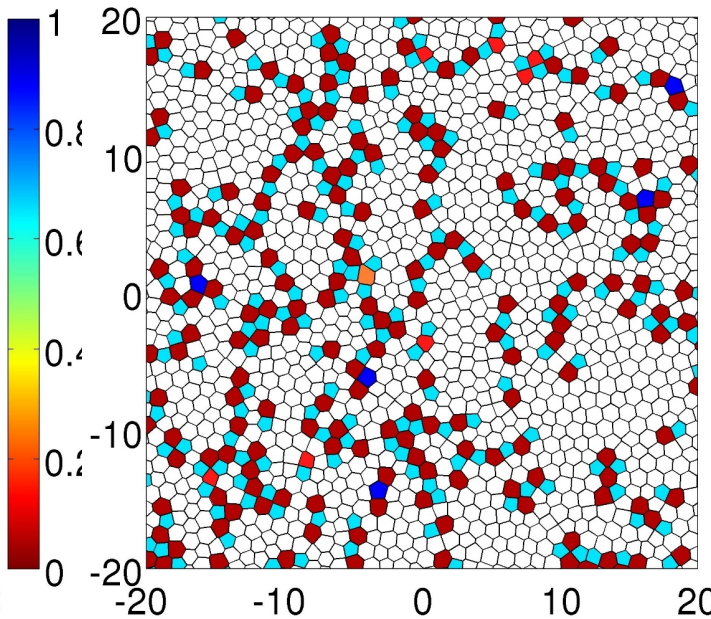
# No obvious relation in our systems

- Some attempts to correlate structure and dynamics :



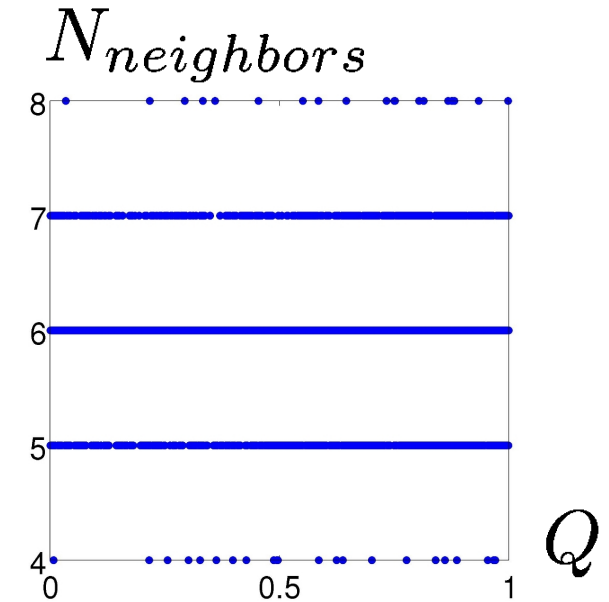
**Local relaxation**

$$Q_{p,t}(a^*, \tau^*)$$



**Topological defects**

particles with  $N \neq 6$   
neighbors

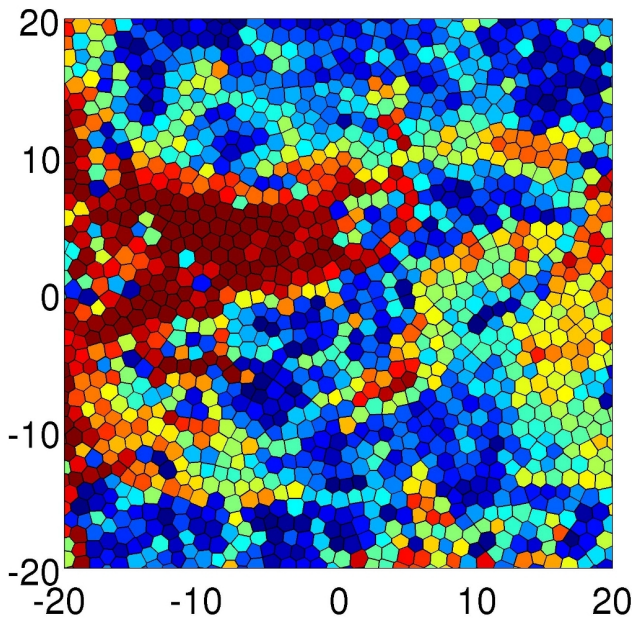


Correlation :

**-0.0069 X**

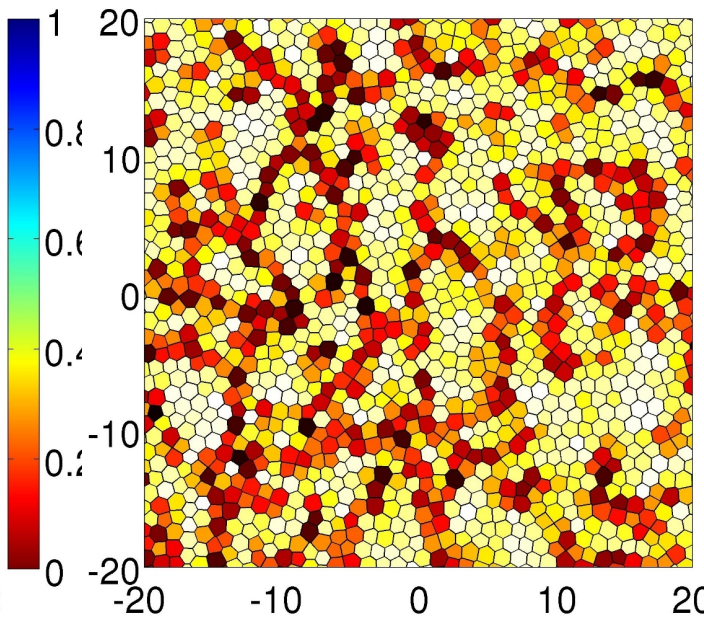
# No obvious relation in our systems

- Some attempts to correlate structure and dynamics :



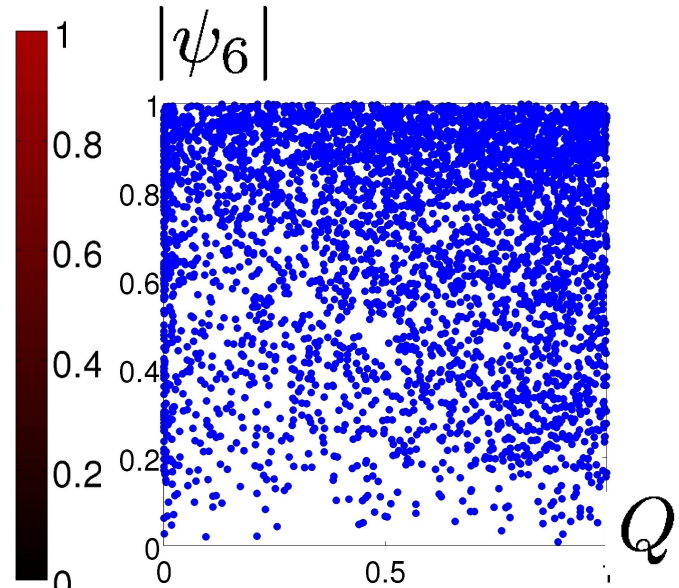
**Local relaxation**

$$Q_{p,t}(a^*, \tau^*)$$



**Orientalional order**

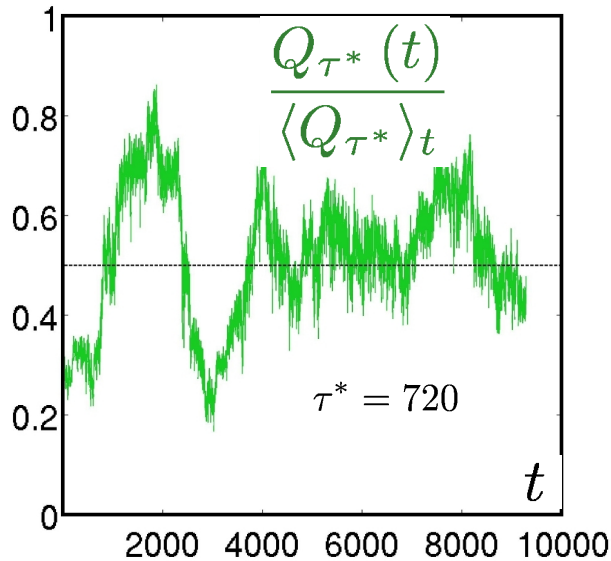
$$\psi_6^j = \frac{1}{n_j} \sum_{k=1}^{n_j} e^{i6\theta_{jk}}$$



Correlation :

**-0.0208 X**

# Inherent difficulties



## Temporal intermittency

The dynamics is intrinsically *intermittent* :  
→ the structural quantity we are searching for should also be !

## Delocalization

No connection between the static and dynamic properties of glass formers at the particle level, but on larger length scales

→ Berthier *et. al.*, Phys. Rev. E. 76, 041509 (2007)


Can one find a structural quantity that meets these two criteria ?

# The isoconfigurational Debye-Waller

- Run 100 evolutions starting from **the same structure** but **random initial momenta** → « isoconfigurations »

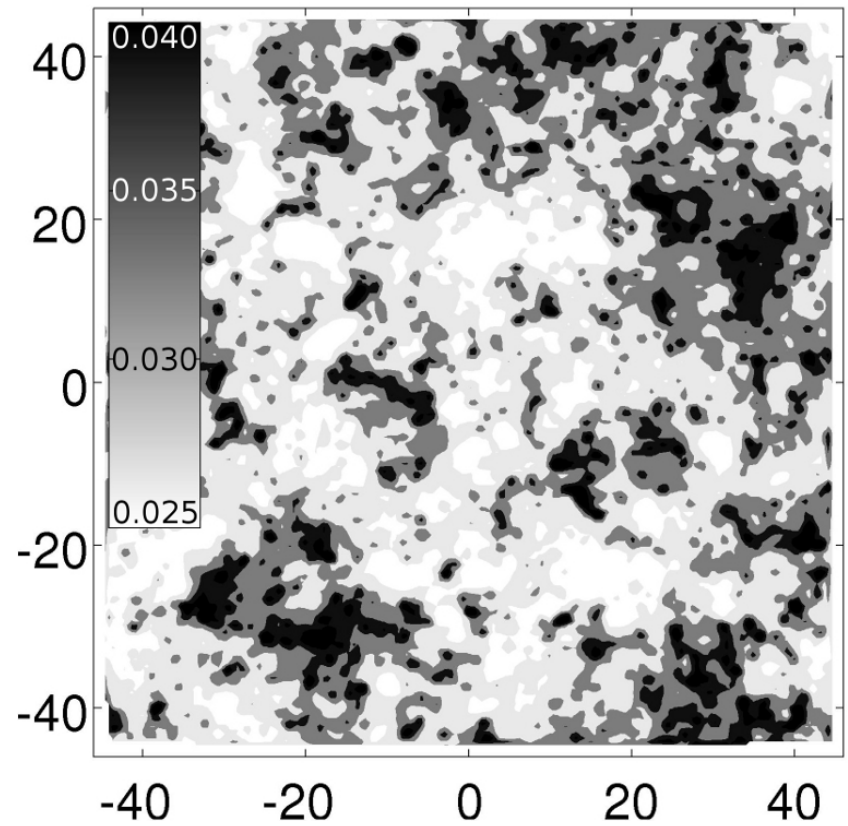
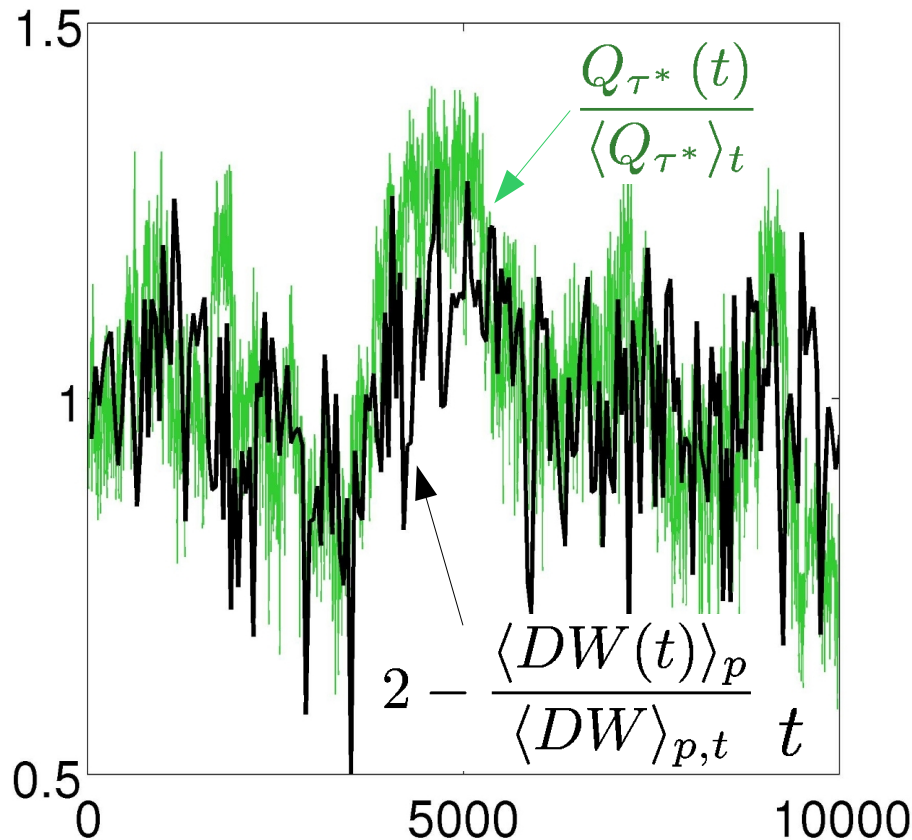


# The isoconfigurational Debye-Waller

- Run 100 evolutions starting from **the same structure** but **random initial momenta** → « isoconfigurations » 
- The Debye-Waller :  $DW_p \equiv \langle \langle [\Delta r(t) - \langle \Delta r \rangle_t]^2 \rangle_{\Delta t} \rangle_c$   
*at short timescale ( $\Delta t=17$ )*

# The isoconfigurational Debye-Waller

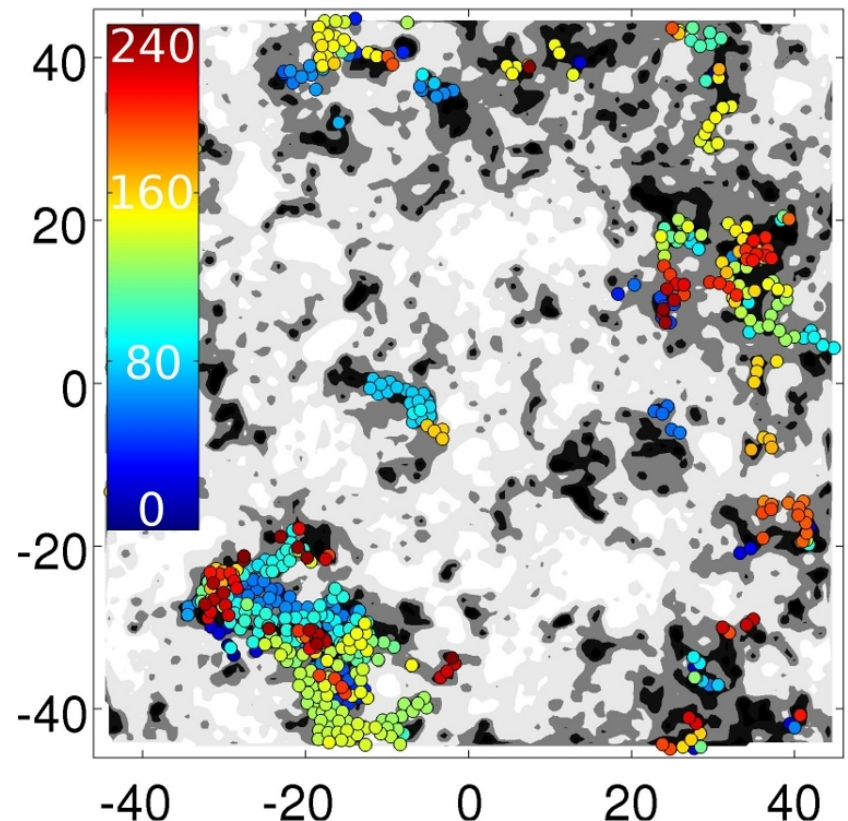
- Run 100 evolutions starting from **the same structure** but **random initial momenta** → « isoconfigurations »
- The Debye-Waller :  $DW_p \equiv \langle \langle [\Delta r(t) - \langle \Delta r \rangle_t]^2 \rangle_{\Delta t} \rangle_c$   
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# The isoconfigurational Debye-Waller

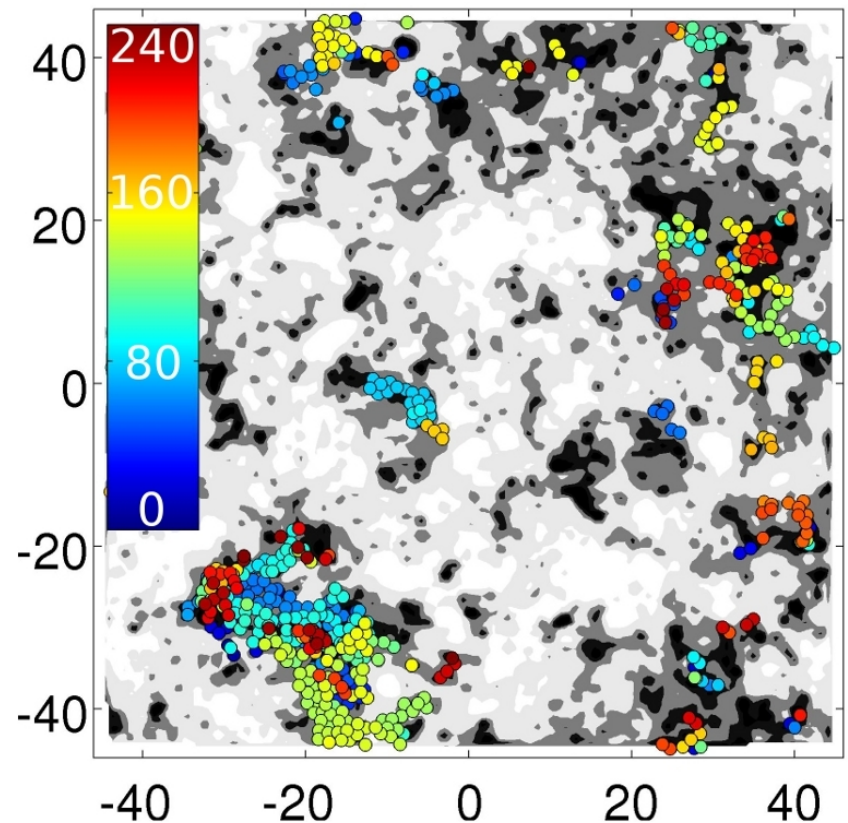
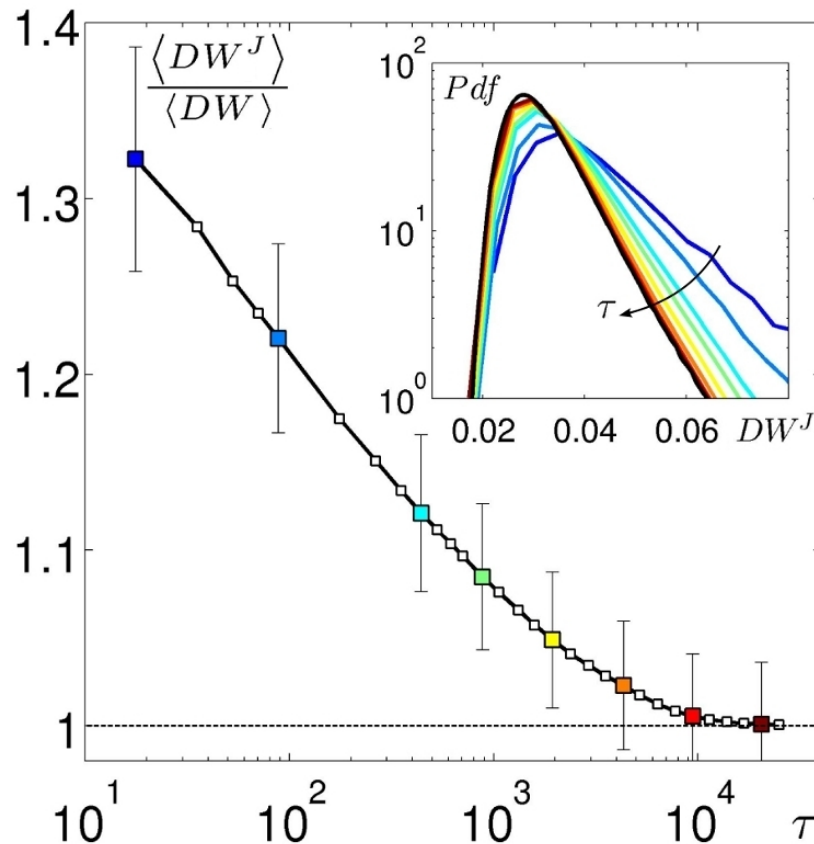
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*at short timescale ( $\Delta t=17$ )*

Predicts the dynamics on  
much longer timescales  
(jumps during  $\sim \tau_\alpha / 3$ )



# The isoconfigurational Debye-Waller

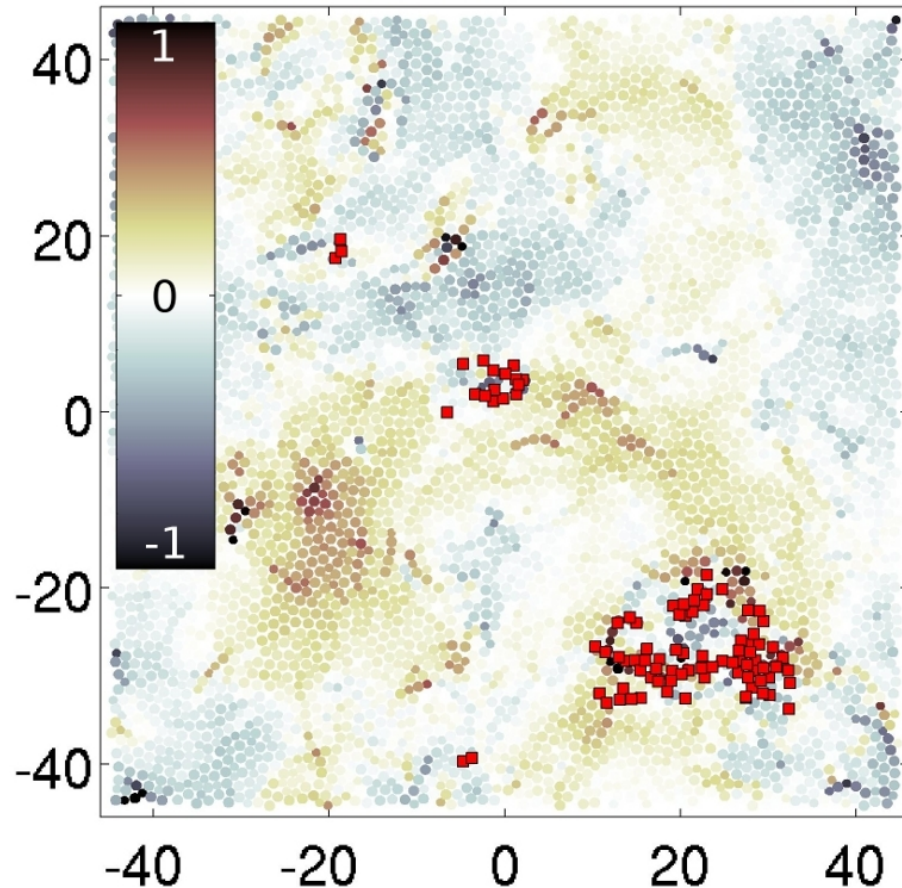
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*at short timescale ( $\Delta t=17$ )*





# Do cage jumps influence Debye-Waller ?

Fluctuations of DW factor  
occur on large length scales ...



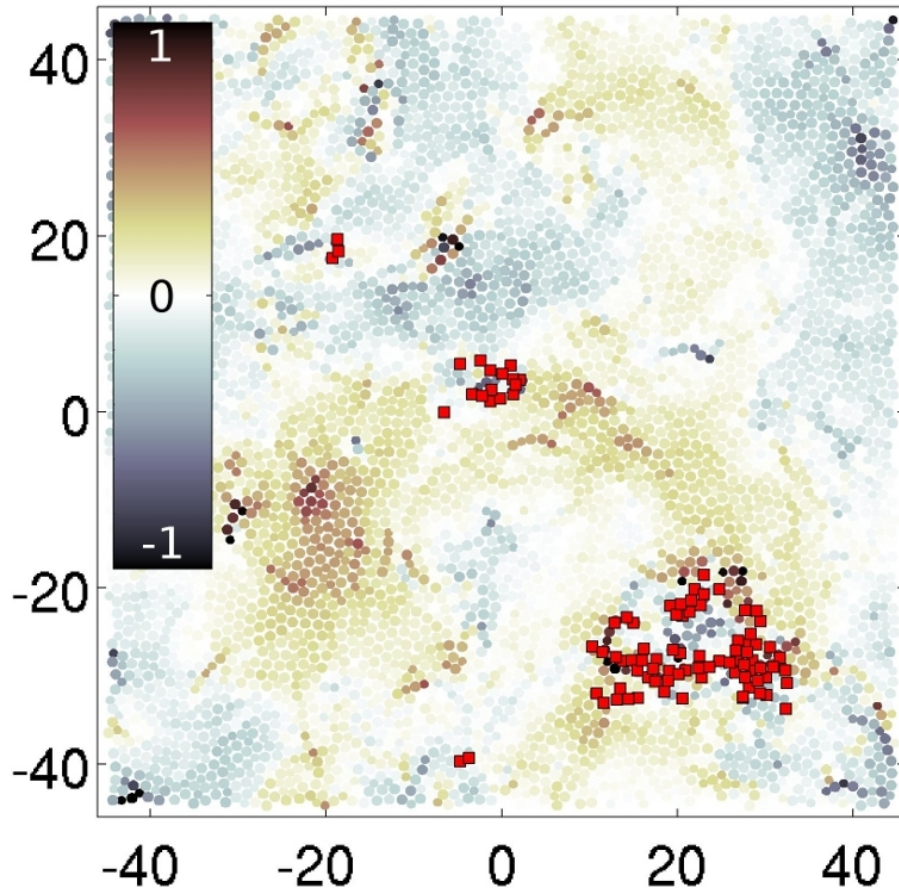
$$\delta DW(\tau) = DW(t + \tau) - DW(t)$$

$(\tau = 17)$

# Do cage jumps influence Debye-Waller ?

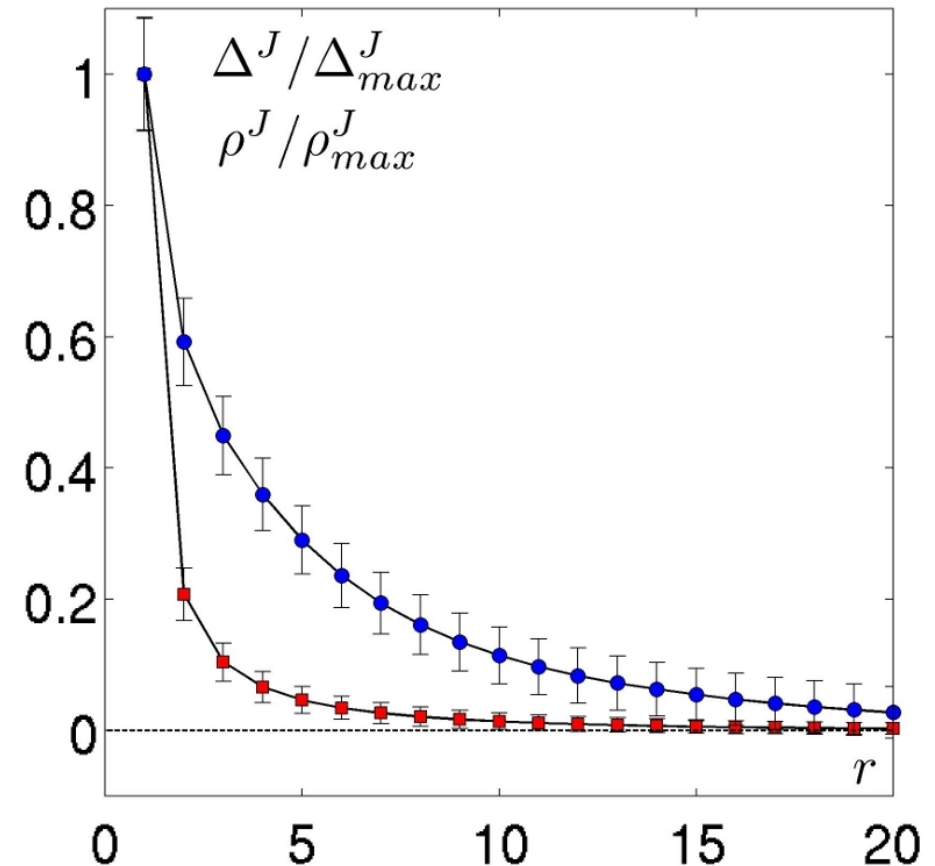
Fluctuations of DW factor  
occur on large length scales ...

... larger than the  
cluster scale



$$\delta DW(\tau) = DW(t + \tau) - DW(t)$$

$(\tau = 17)$



# General conclusion

- Heterogeneous relaxation is well described with cage jumps
- Large decorrelation domains arise from the complex spatio-temporal facilitation patterns :
  - Single entangle network at low  $\phi$
  - Distinct « avalanches » at high  $\phi$
- Isoconfigurational studies reveal that a « static » quantity can predict future evolution of the system
- The same description applies for three different systems

# Perspectives

- **In simulations :**
  - Apply the same methodology for several temperatures
    - *Is the phenomenology similar to the evolution with  $\phi$  ?*
  - Link between the soft modes analyses and the isoconfigurational studies.
- **In grains :** explore higher densities
  - *Is facilitation disappearing for systems closer to  $\phi_G$  ?*
  - Relation to Jamming ?

# Thanks to ...

Olivier Dauchot and Giulio Biroli for their wise advices

Asaph Widmer-Cooper, David Reichman, Peter Harrowell, Jonathan Kummerfeld, Douglas Durian and Jean-Philippe Bouchaud for fruitful scientific collaboration

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*and so many more ... you included !*