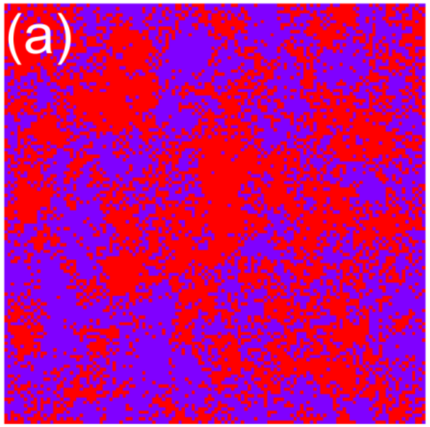
Elasticity, Facilitation, Avalanches of Relaxation, and Dynamic Heterogeneity in Glass-Forming Liquids



Misaki Ozawa

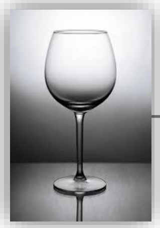
Univ. Grenoble Alpes

Collaboration with Giulio Biroli, Marko Popovic, Ali Tahaei, and Matthieu Wyart

Ozawa and Biroli, PRL, 2023

Tahaei, Biroli, Ozawa, Popovic, and Wyart, PRX, 2023

Supercooled liquids approaching glass transition

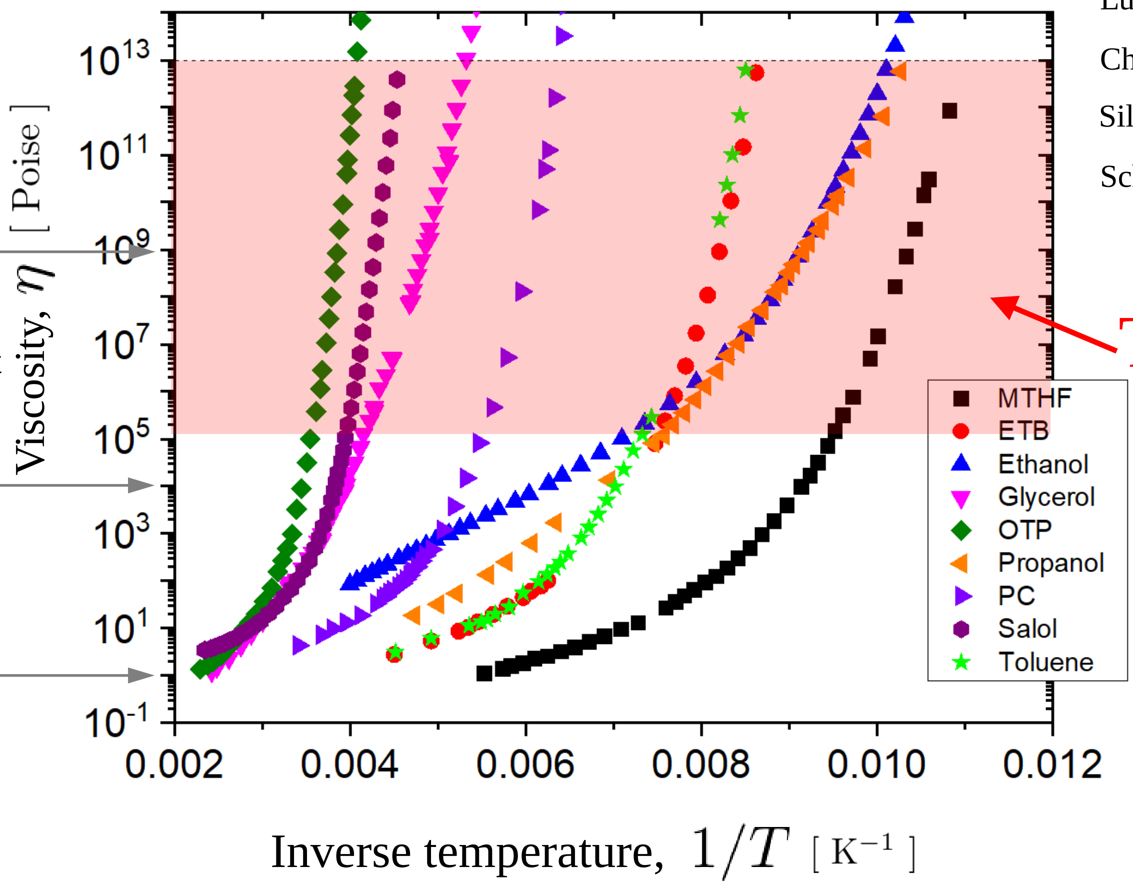


Glass

Pitch drop experiment

Peanut butter

Olive oil



Lunkenheimer et al., PRE 2010

Chen and Richert, JCP 2011

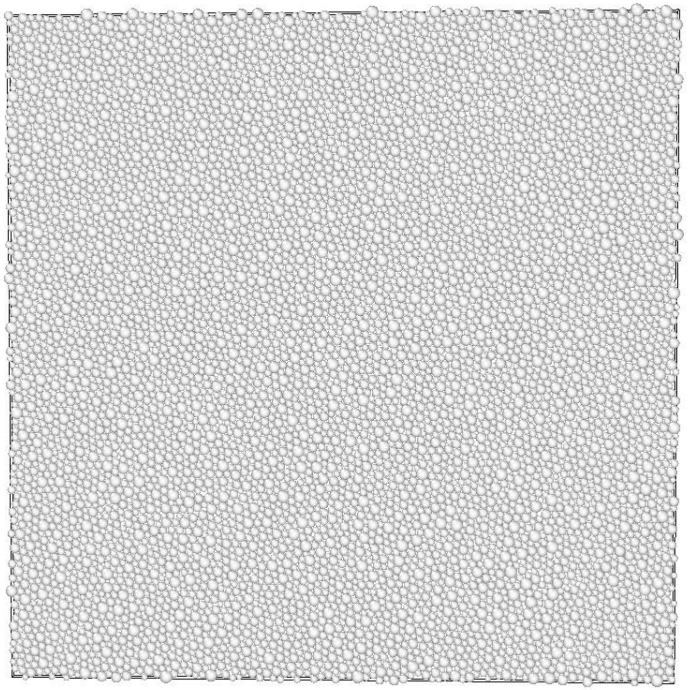
Sillren et al., JCP 2014

Schmidtke et al., PRE 2012

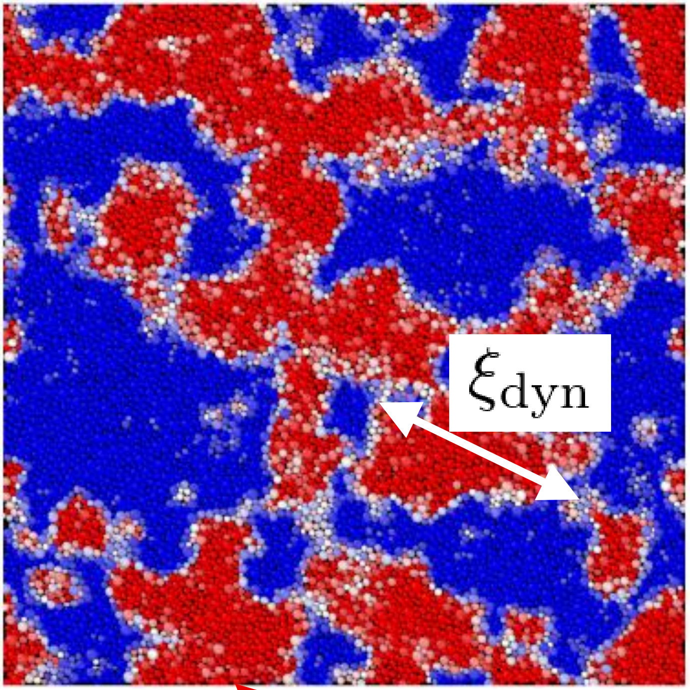
Target regime

Dynamical heterogeneity

Snapshot



Dynamics



Immobile

Mobile

Hurley and Harrowell, PRE 1995

Karmakar, Dasgupta, and Sastry, PNAS 2009

Kob, Donati, Plimpton, Poole, and Glotzer, PRL 1997

Scalliet, Guiselin, and Berthier, PRX 2022

Mechanism?

Many theories and scenarios

- Random first order transition theory

  Kirkpatrick, Thirumalai, and Wolyness, PRA 1989

  Bouchaud and Biroli, JCP 2004

- Dynamic facilitation

  Chandler and Garrahan, Annu. Rev. Phys. Chem. 2010

- Frustration-limited domain theory

  Tarjus, Kivelson, Nussinov, and Viot, J. Phys. Condens. Matter 2005

- Elasticity scenarios

  Dyre, RMP 2006

  Lemaitre, PRL 2014

- Geometrical considerations

  Tanaka, Kawasaki, Shintani, and Watanabe, Nat. Mater 2010

- Etc..

The New York Times

The Nature of Glass Remains Anything but Clear

Many theories and scenarios

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  Kirkpatrick, Thirumalai, and Wolyness, PRA 1989

  Bouchaud and Biroli, JCP 2004

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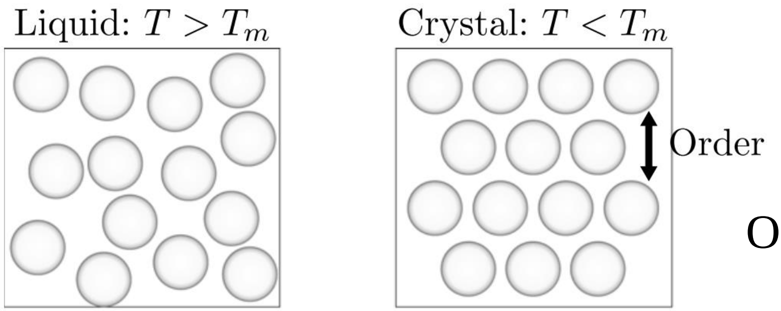
The New York Times

The Nature of Glass Remains Anything but Clear

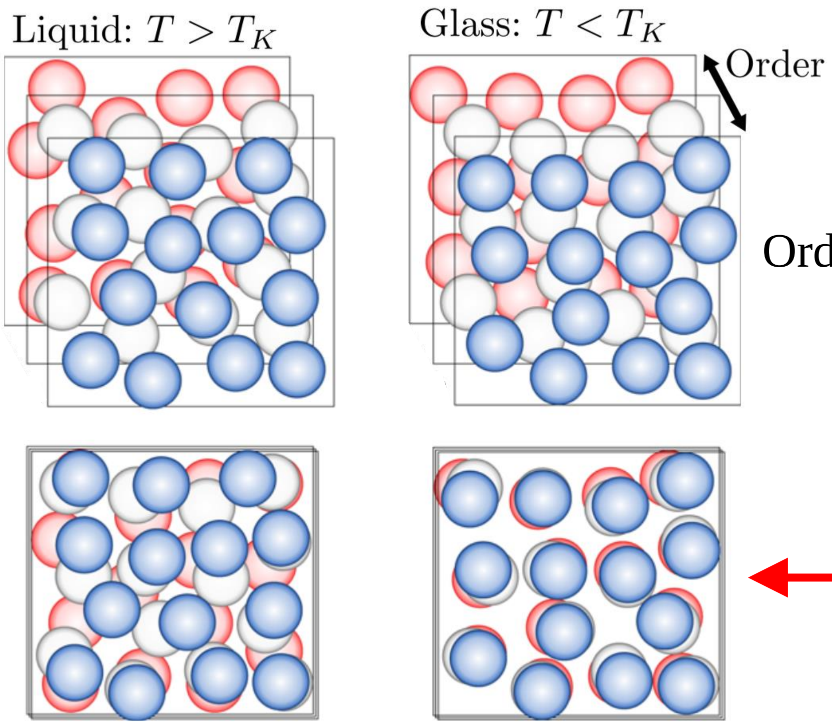
Order parameter for the glass transition in RFOT

Liquid to Crystal transition



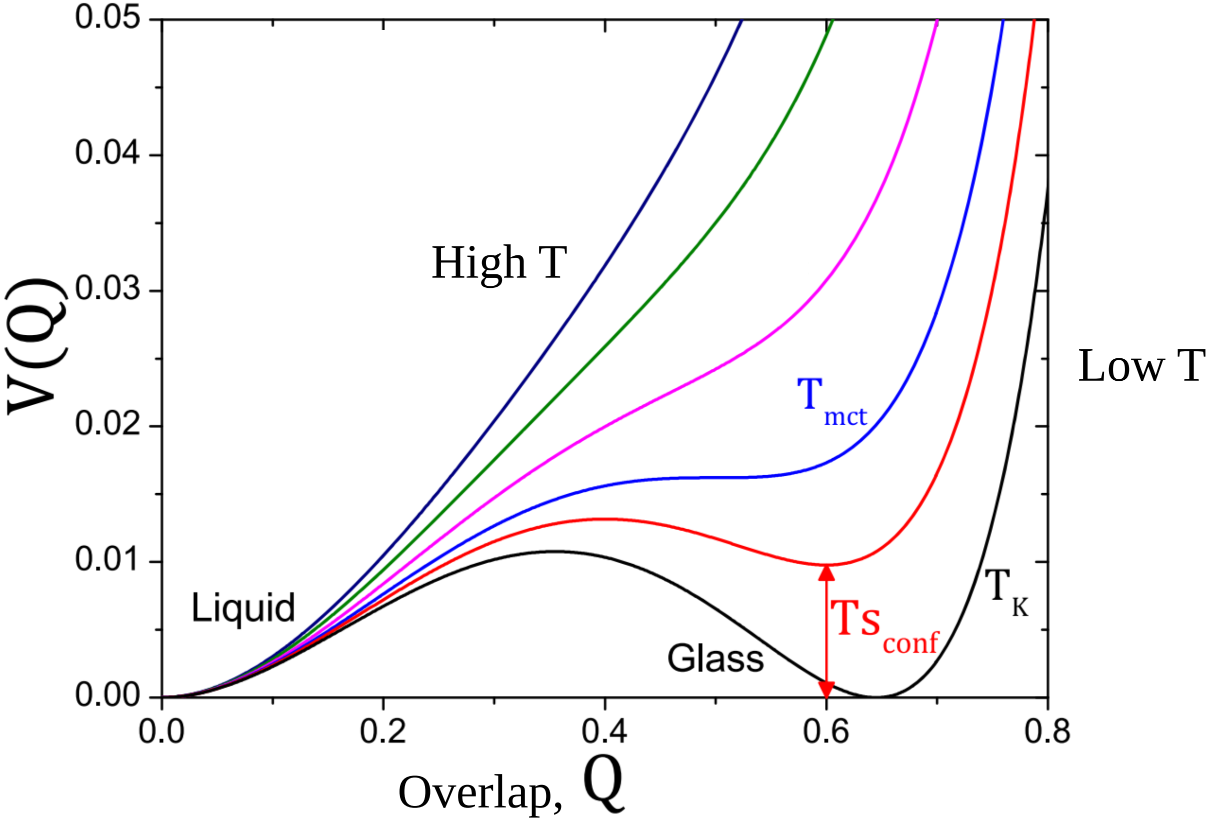
Order within one configuration

Liquid to Glass transition



Order among different configurations

← Overlap!



Landau free energy for the glass transition (Franz-Paris potential)

Franz and Parisi, Journal de Physique 1995

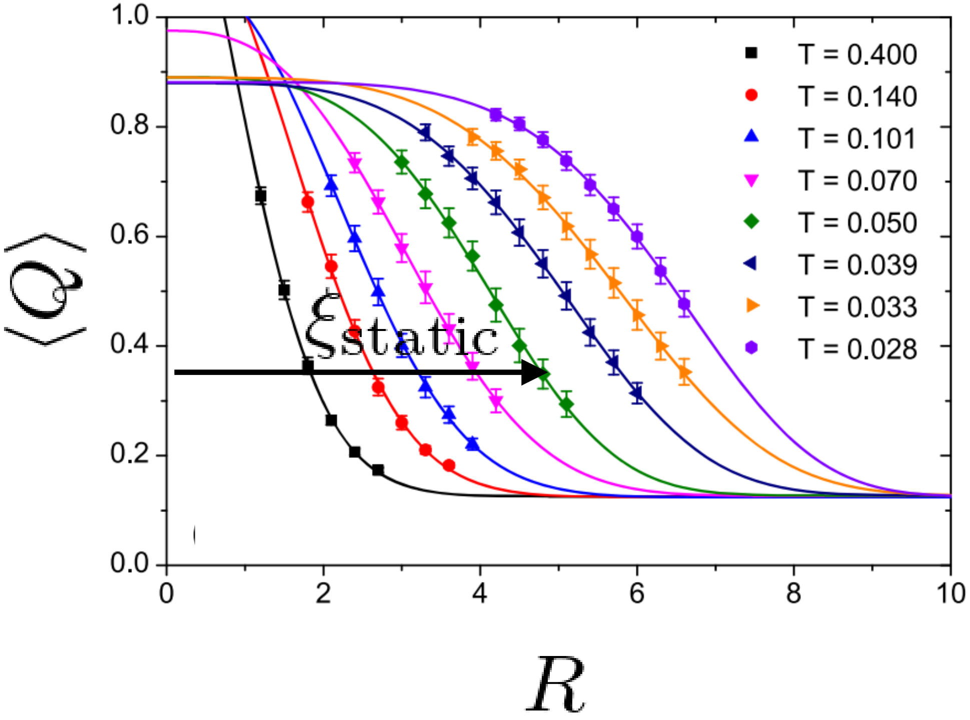
Random “first-order” transition (Kauzmann ideal glass transition) at T_{K}

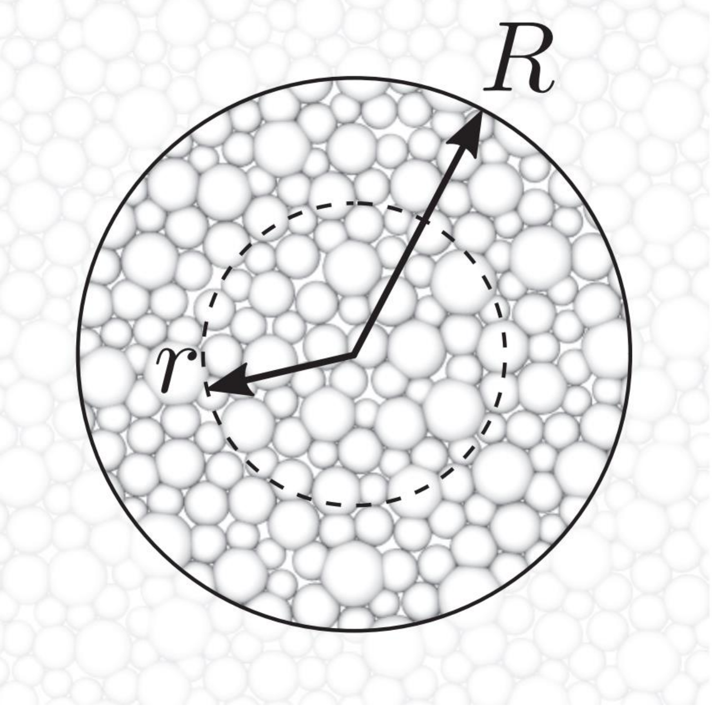
Verified at
$d \to \infty$

Merzard and Parisi, PRL 1999

Charbonneau, Kurchan, Parisi, Urbani, and Zamponi, Annu. Rev. Condens. Matter Phys. 2017

Finite dimensions d





Biroli, Bouchaud, Cavagna, Grigera, Verrocchio, Nat. Phys. 2008

Kurchan and Levine, J. Phys. A 2010

Berthier, Ozawa, and Scalliet, JCP 2019

Extracting “static” correlation length $\xi_{\mathtt{static}}$ by overlap
(Amorphous order)

$\xi_{\mathtt{static}}$ increases with decreasing temperature

RFOT prediction for dynamics

Relaxation time

$$\tau_\alpha \sim \exp\left[\frac{\xi_{\mathtt{static}}^{\psi}}{T}\right] \qquad \psi > 0$$

$$\xi_{\mathtt{static}} \to \infty \quad \text{when} \quad T \to T_{\mathrm{K}}$$

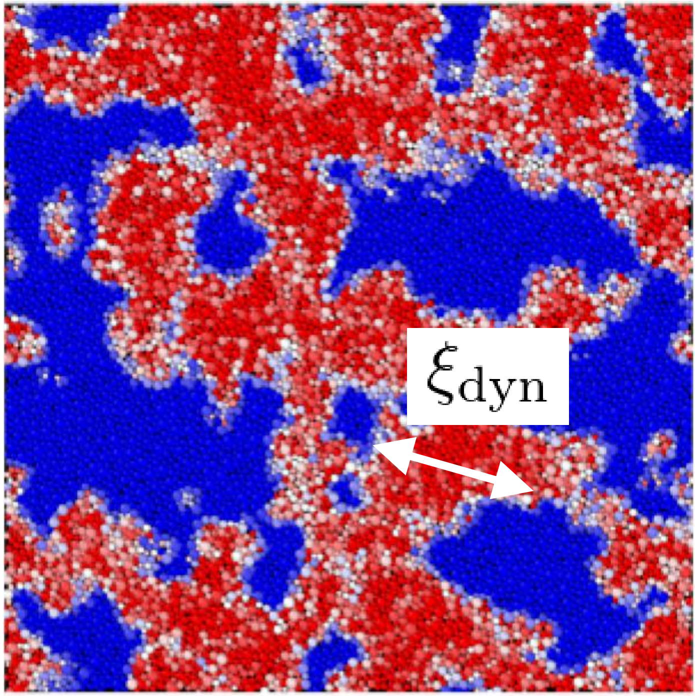
Thus, $\tau_\alpha \to \infty$ when $T \to T_{\mathrm{K}}$

Kirkpatrick, Thirumalai, and Wolyness, PRA 1989

Bouchaud and Biroli, JCP 2004

Relaxation time diverges toward T_{K}

Comparison between static and dynamic lengthscales



Mobile

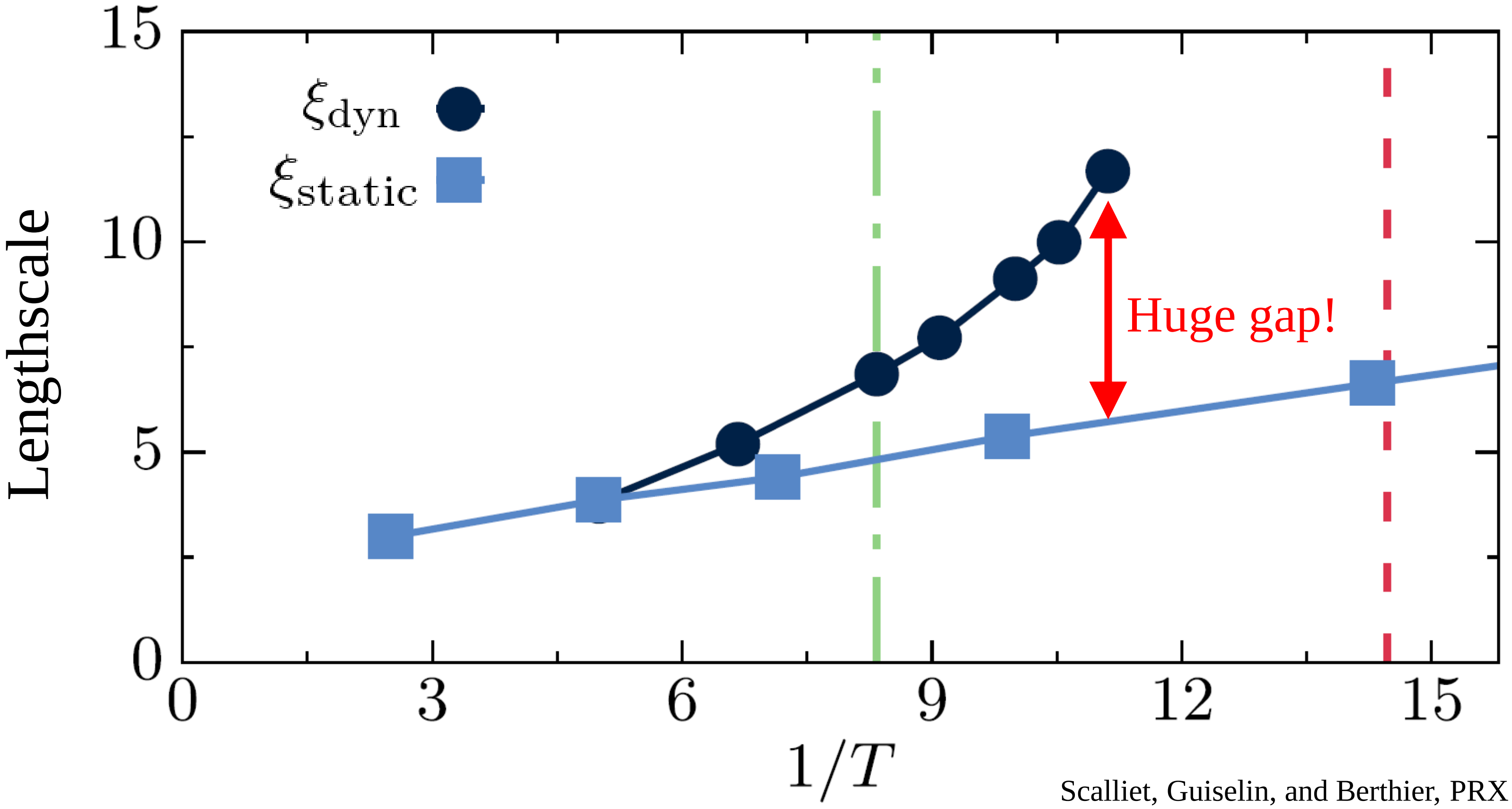
Immobile

ξ_{static}

Scalliet, Guiselin, and Berthier, PRX 2022

ξ_{static} is much smaller than ξ_{dyn}

Comparison between static and dynamic lengthscales



Scalliet, Guiselin, and Berthier, PRX 2022

$\xi_{\rm static}$ is much smaller than $\xi_{\rm dyn}$

ξ_{static} alone cannot explain dynamical heterogeneity ξ_{dyn}

What is missing?


Many theories and scenarios

- Random first order transition theory

  Kirkpatrick, Thirumalai, and Wolyness, PRA 1989

  Bouchaud and Biroli, JCP 2004

- Dynamic facilitation

  Chandler and Garrahan, Annu. Rev. Phys. Chem. 2010

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  Lemaitre, PRL 2014

- Geometrical considerations

  Tanaka, Kawasaki, Shintani, and Watanabe, Nat. Mater 2010

- Etc..

The New York Times

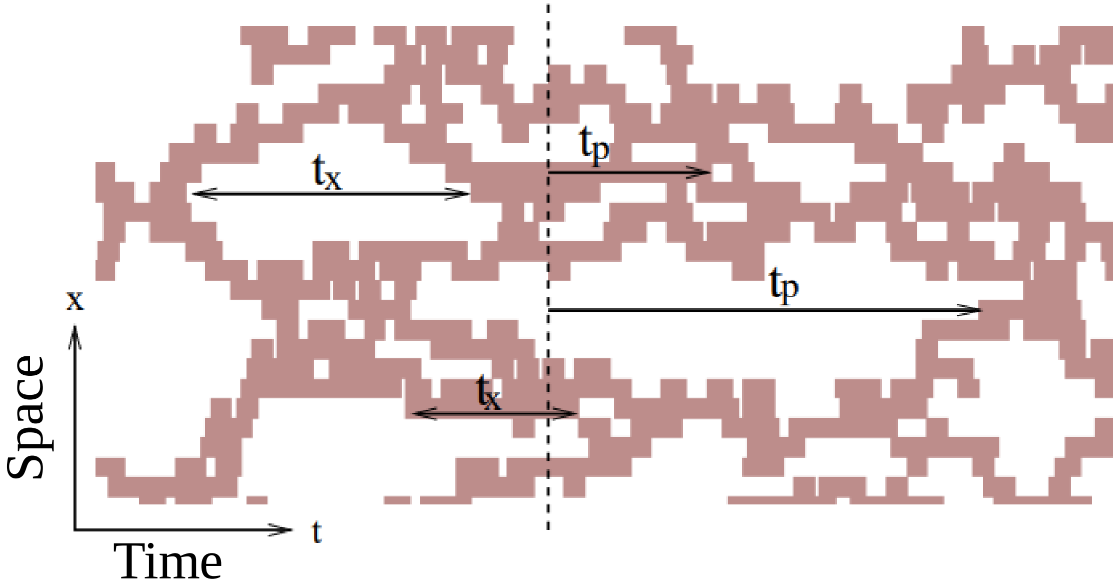
The Nature of Glass Remains Anything but Clear

Kinetically constrained models: ideal gas of mobile defects diffusing on a lattice.

Cancrini, Martinelli, Roberto, Toninelli 2009

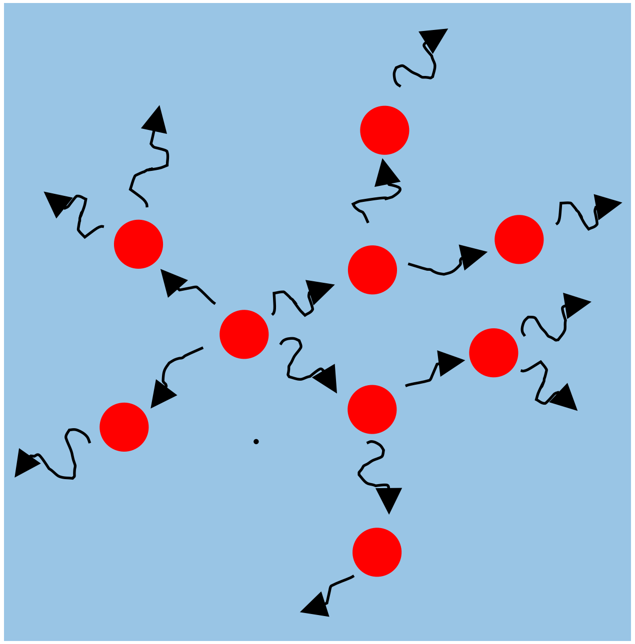
Chandler and Garrahan, Annu. Rev. Phys. Chem. 2010

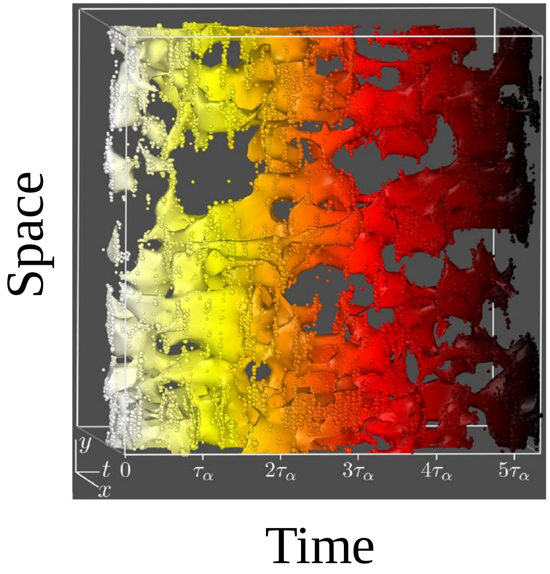
Trivial thermodynamics yet constrained dynamical rules for defect mobility.
(Abstract mathematical rule)



Reproduces super-Arrhenius behavior, growth of dynamic lengthscale.

Dynamical facilitation





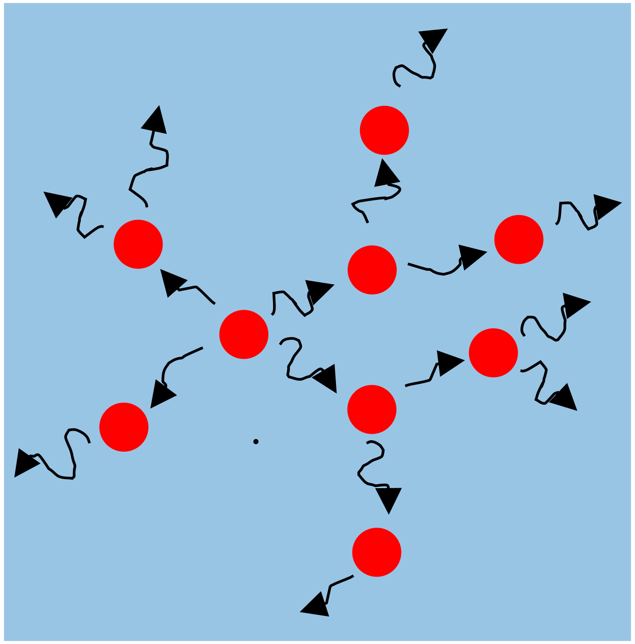
Mobile sites induce mobile sites nearby:
Cascade of mobility

Dynamical facilitation is getting dominant at lower temperatures

Guiselin, Scalliet, and Berthier, Nat Phys. 2021

Scalliet, Guiselin, and Berthier, PRX 2022

Dynamical facilitation is there. But what is physical mechanism responsible for it?



What is ⤳ ?

Many theories and scenarios

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- Etc..

The New York Times

The Nature of Glass Remains Anything but Clear

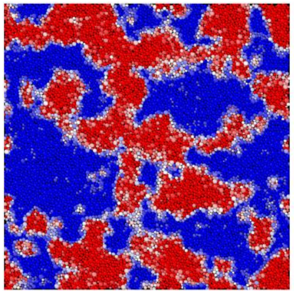
Different approaches

?

Solid that flows

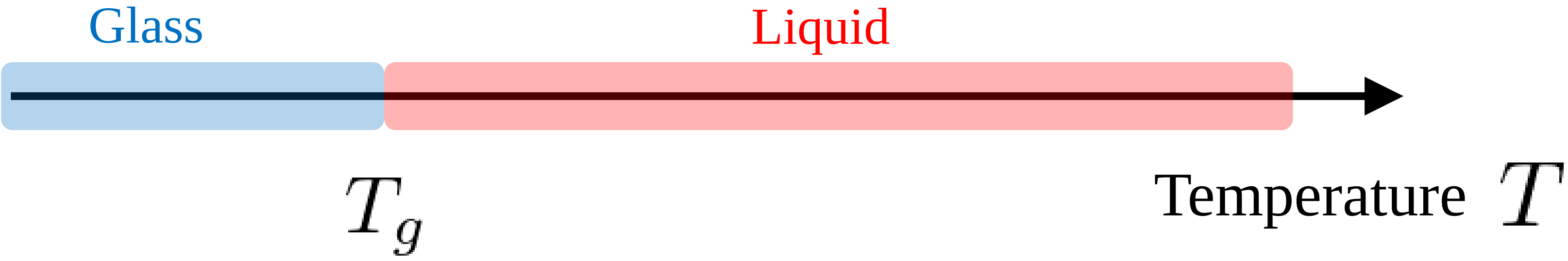
Dyre, RMP 2006

Lemaitre, PRL 2014



Viscous fluid

Majority of theories



Glass

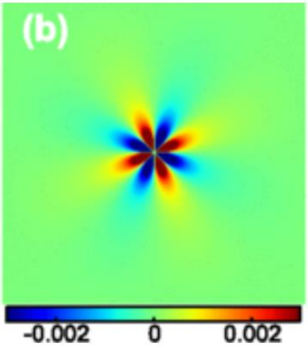
Liquid

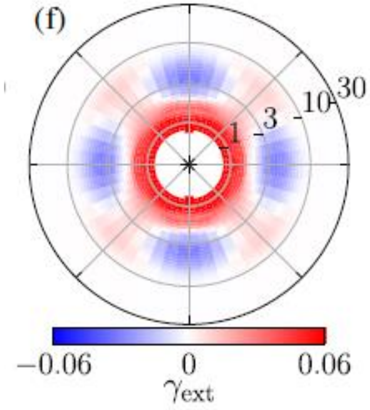
T_g

Temperature T

Elasticity and plasticity in liquids

Elasticity and long-range stress correlations





Lemaitre, PRL 2014

Wu, Iwashita, and Egami, PRE 2015

Flenner and Szamel, PRL 2015

Chowdhury, Abraham, Hudson, and Harrowell, JCP 2016

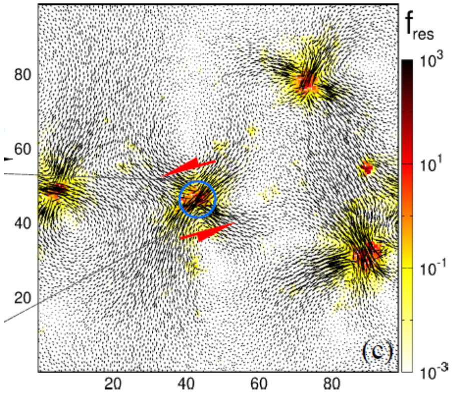
Maier, Zippelius, and Fuchs, PRL 2017

Chacko, Landes, Biroli, Dauchot, Liu, and Reichman, PRL 2021

Steffen, Schneider, Muller, and Rottler, JCP 2022

Ghanekarade, Phan, Schweizer, and Simmons, Nat. Phys. 2023

Plasticity and shear transformations

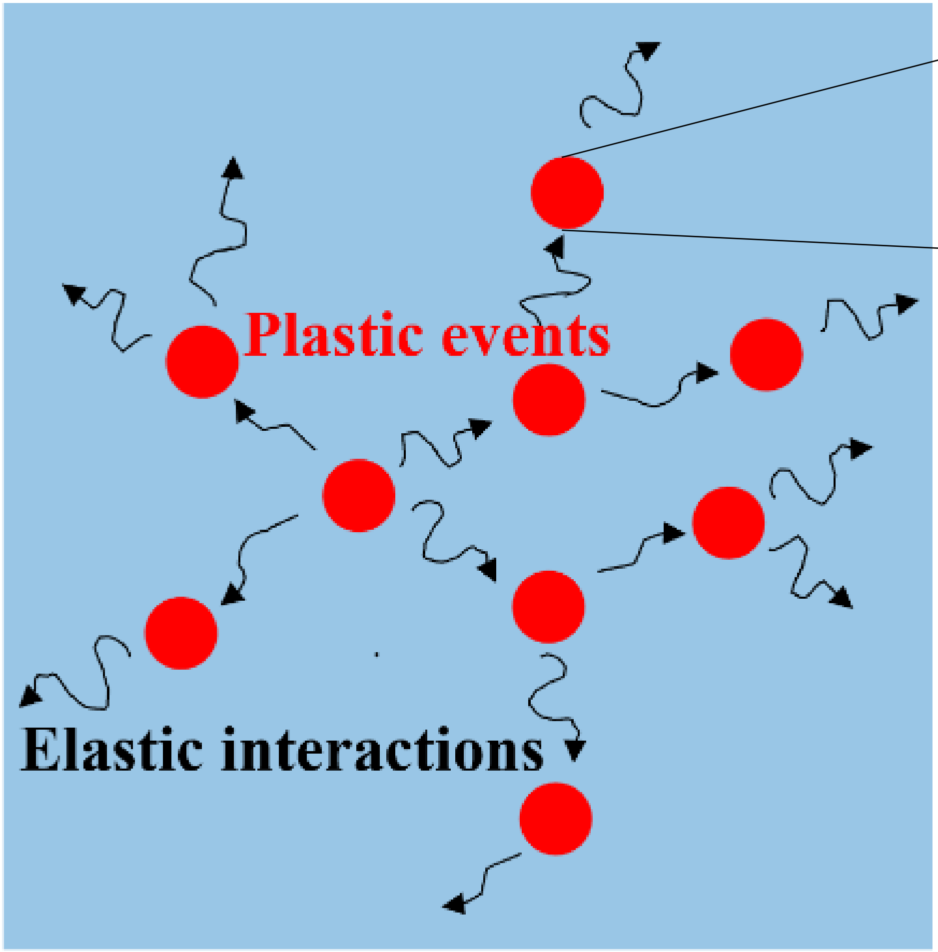


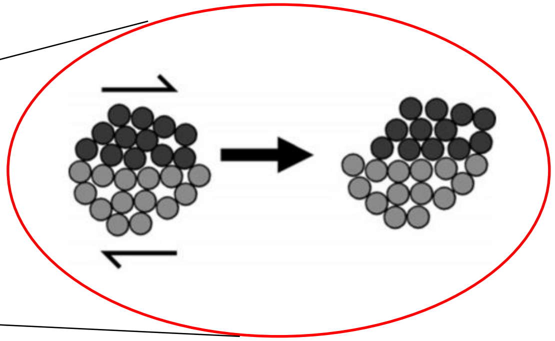
Lerbinger, Barbot, Vandembroucq, and Patinet, PRL 2022

Falk and Langer, PRE 1998

Maloney and Lemaitre, PRE 2006

Mechanism of dynamical heterogeneity





Localized plastic events induced by thermal fluctuations and elasticity

Chacko, Landes, Biroli, Dauchot, Liu, and Reichman PRL 2021

Lerbinger, Barbot, Vandembroucq, and Patinet, PRL 2022

Cascade of events: Facilitation

Chandler and Garrahan, Annu. Rev. Phys. Chem. 2010

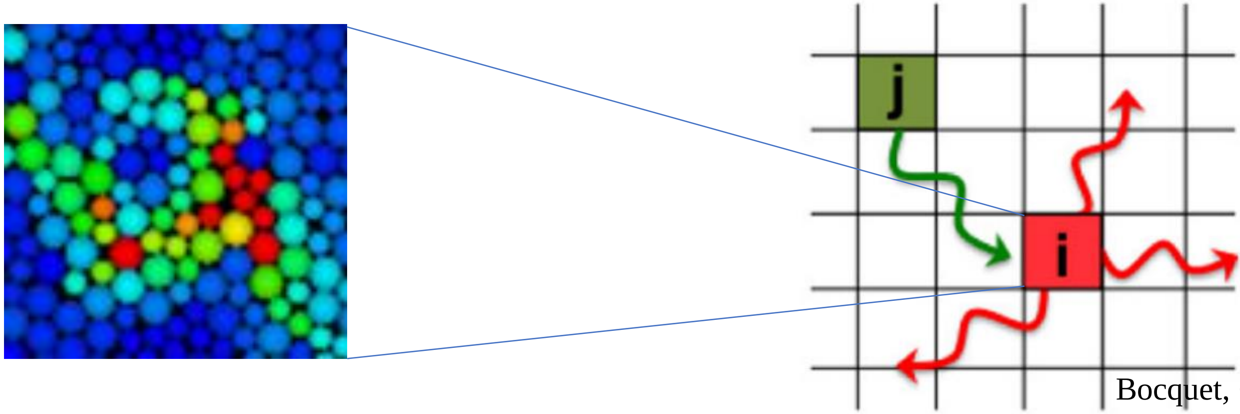
Elasticity induced facilitation

Hasyima and Mandadapu, JCP 2021 Ozawa and Biroli, PRL, 2023

Elastoplastic models

Microscopic scale

Mesoscopic scale



Bocquet, Colin, Ajdari, PRL 2009

Molecular simulations (MD)

Nicolas, Ferrero, Martens, and Barrat, RMP 2018

Bulatov and Argon, Mod. Sim. 1994

Baret, Vandembroucq, and Roux, PRL 2002

Elastoplastic models (EPM)

Onuki, PRE 2003

Jagla, PRE 2007

Lin, Lerner, Rosso, and Wyart PNAS 2014

1) Local dynamics condition for local stress σ_i

i) $\sigma_i \leq \sigma^{\mathrm{th}}$: Elastic (immobile)

ii) $\sigma_i > \sigma^{\mathrm{th}}$: Plastic (mobile) with a stress drop $\sigma_i \to \sigma_i - \delta\sigma_i$

iii) Thermal fluctuation: Elastic → Plastic with probability $e^{-E(\sigma_i)/T}$

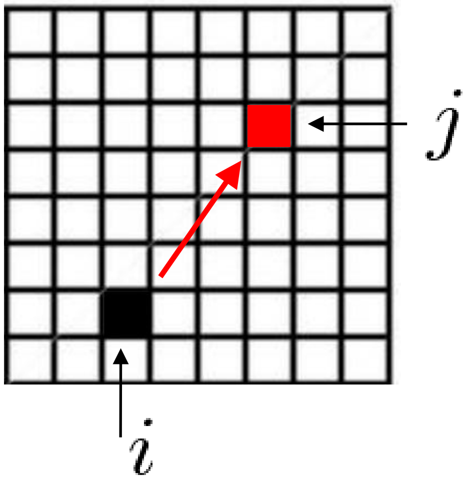
Maloney and Lacks, PRE 2006

Ferrero, Martens, and Barrat, PRL 2014

Lerbinger, Barbot, Vandembroucq, and Patinet, PRL 2022

$$E(\sigma_i) = (\sigma^{\mathrm{th}} - \sigma_i)^{3/2}$$

2) Elastic interaction

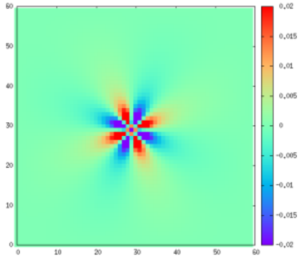


$$\sigma_j \to \sigma_j + g(\mathbf{r}_{ij})\delta\sigma_i$$

$$g(\mathbf{r}_{ij}) = \frac{\cos(4\theta)}{r_{ij}^2}$$

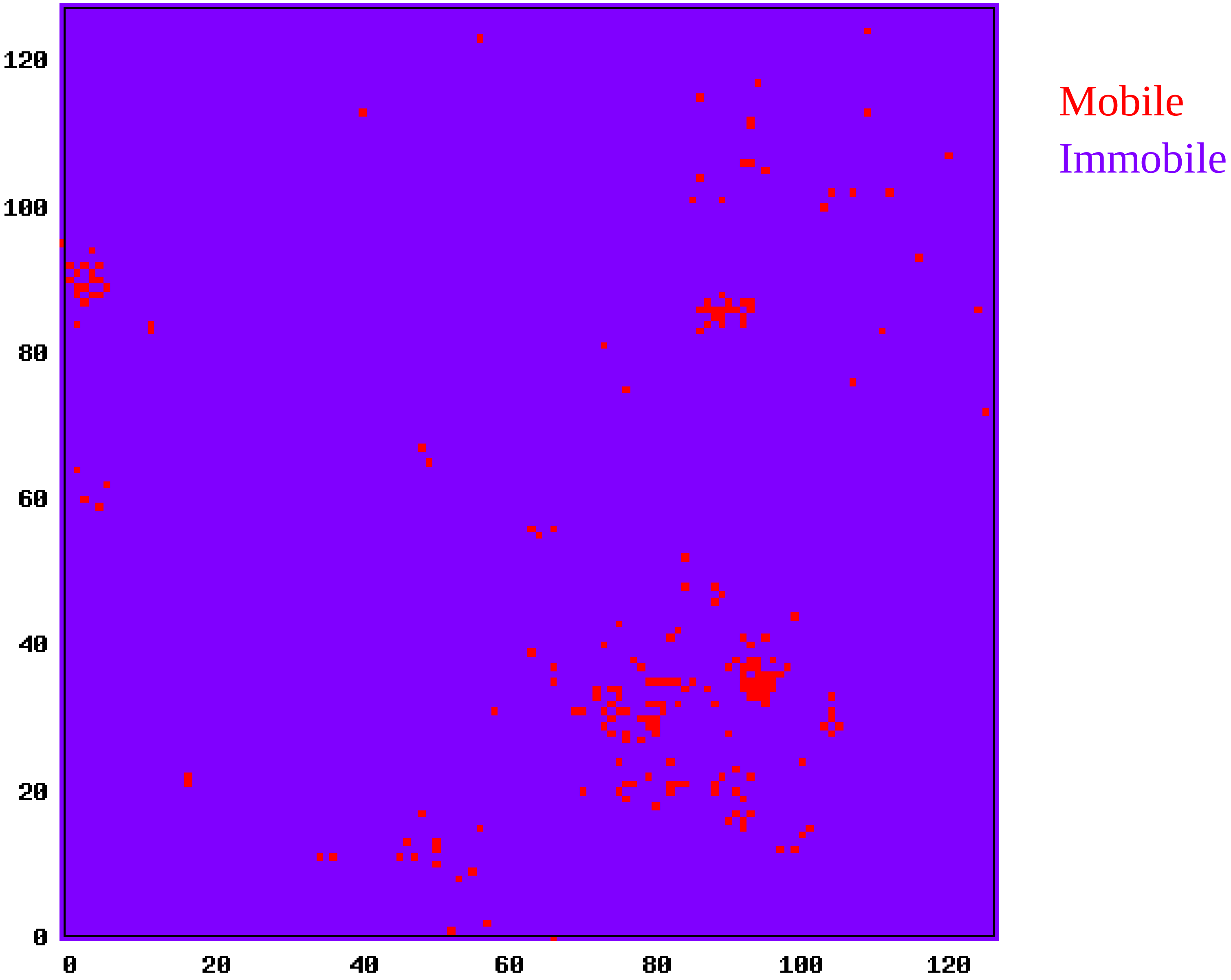
Picard, Ajdari, Lequeux, and Bocquet, EPJE 2004

Random rotation of Eshelby kernel

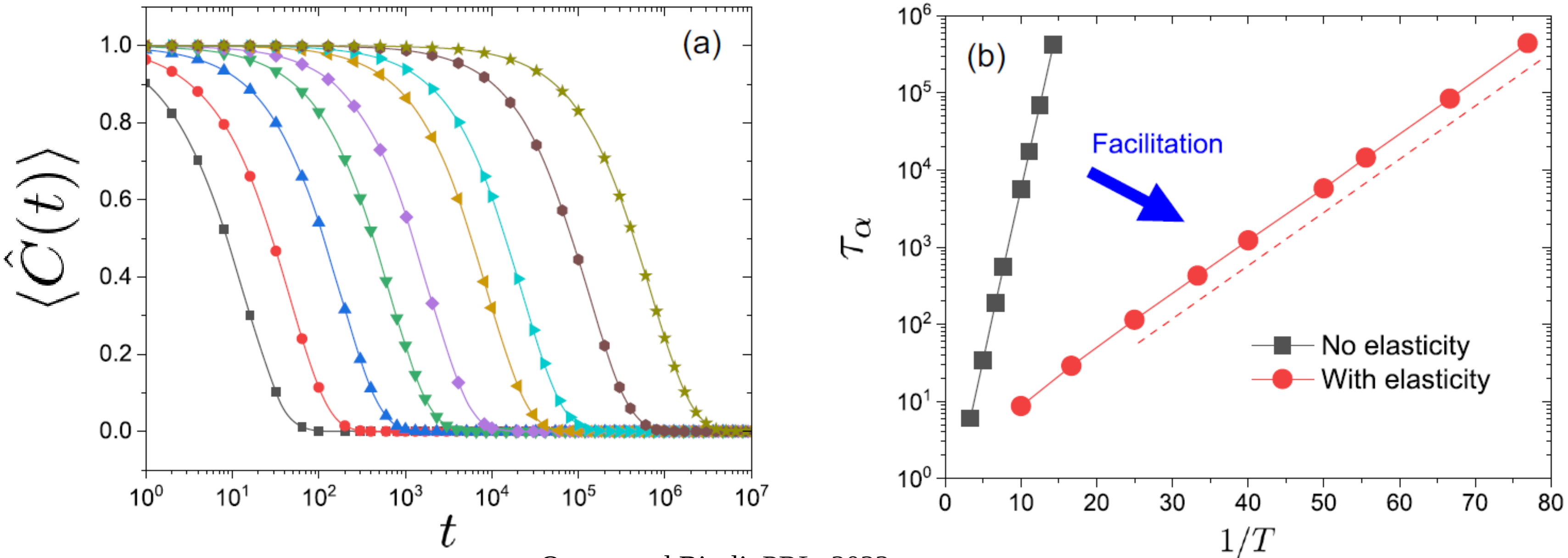


3) Repeat 1) - 2)

Movie



Two-point time correlation function



Ozawa and Biroli, PRL, 2023

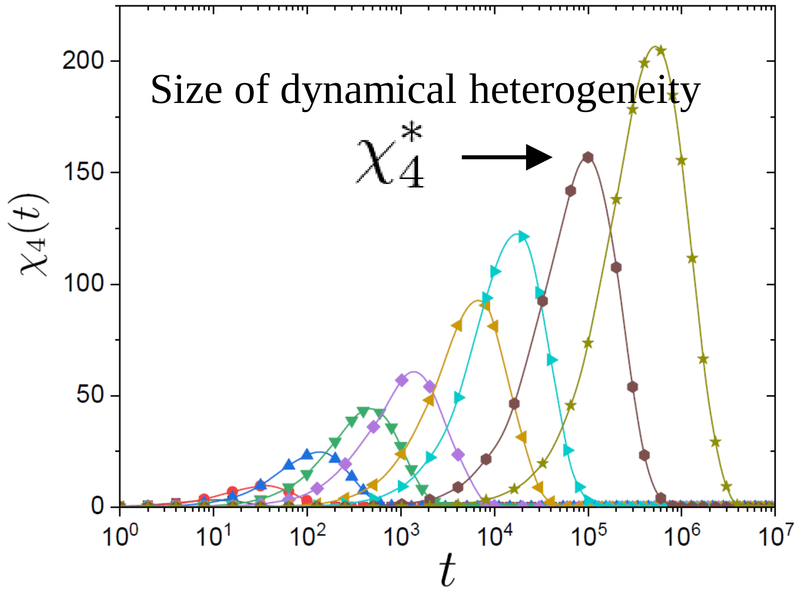
$\langle \hat{C}(t) \rangle$: Persistence two-point time correlation function
(à la Intermediate scattering function)

Berthier and Garrahan, PRE 2003

τ_α : Relaxation time defined by $\langle \hat{C}(\tau_\alpha) \rangle = 1/e$

Four-point correlation function

Elastoplastic model

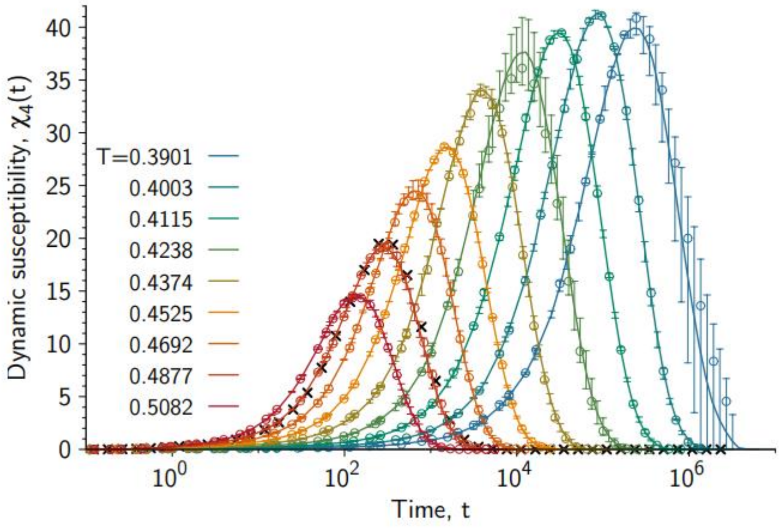


Size of dynamical heterogeneity

χ_4^* →

Ozawa and Biroli, PRL, 2023

Molecular simulation



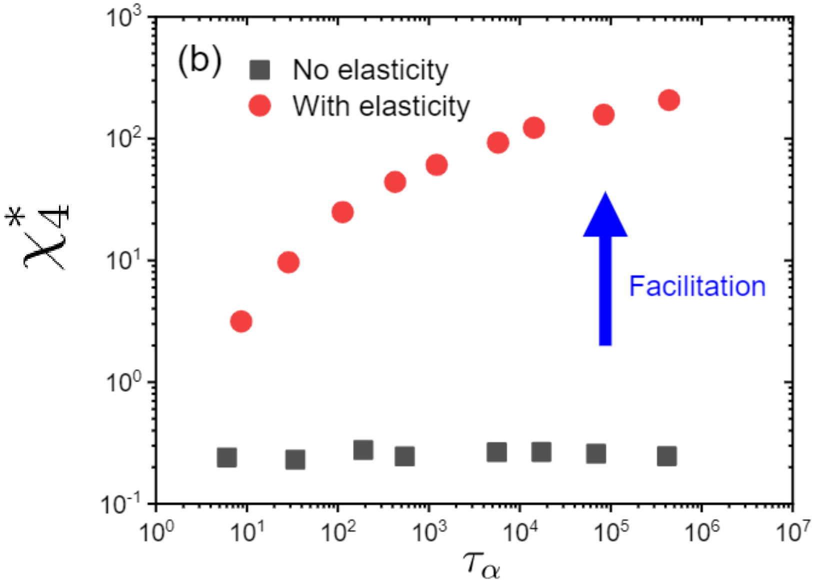
Coslovich, Ozawa, and Kob, EPJE 2018

$$\chi_4(t) = N\mathrm{Var}(\hat{C}(t)) = N\left(\langle \hat{C}^2(t)\rangle - \langle \hat{C}(t)\rangle^2\right)$$

Lacevic, Starr, Schroder, and Glotzer, JCP 2003

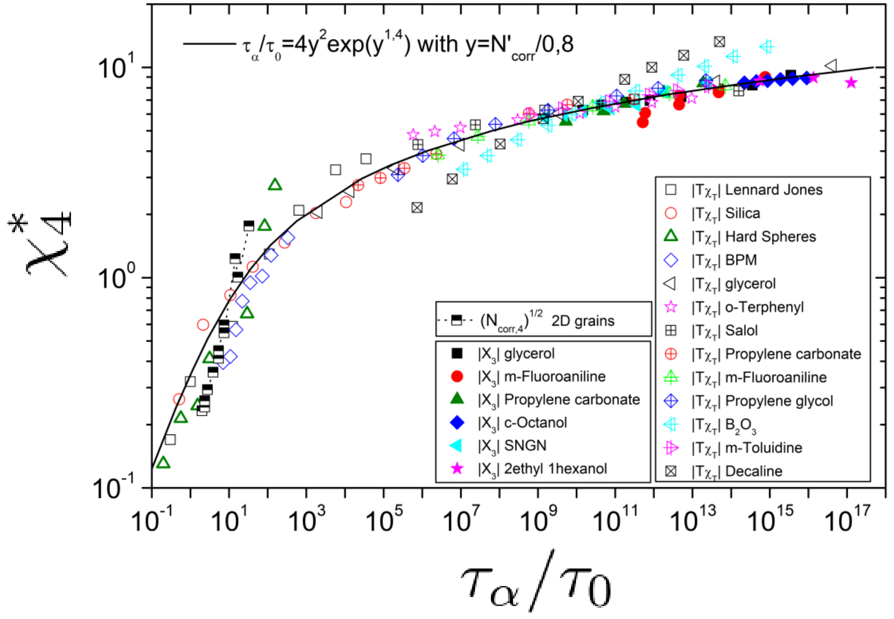
Four-point correlation function

Elastoplastic model



Ozawa and Biroli, PRL, 2023

Experiments



Dalle-Ferrier, Thibierege, Alba-Simionesco, Berthier, Biroli, Bouchaud, Ladieu, L'Hote, and Tarjus, PRE 2007

Dauchot, Ladieu, and Royall, Comptes Rendus. Physique 2023

Mean-field theory

$$\frac{\partial P(\sigma,t)}{\partial t} = \alpha\Gamma(t)\frac{\partial^2 P(\sigma,t)}{\partial \sigma^2} - \nu(\sigma,\sigma_c)P(\sigma,t) + \Gamma(t)y(\sigma)$$

Hébraud and Lequeux, PRL 1998

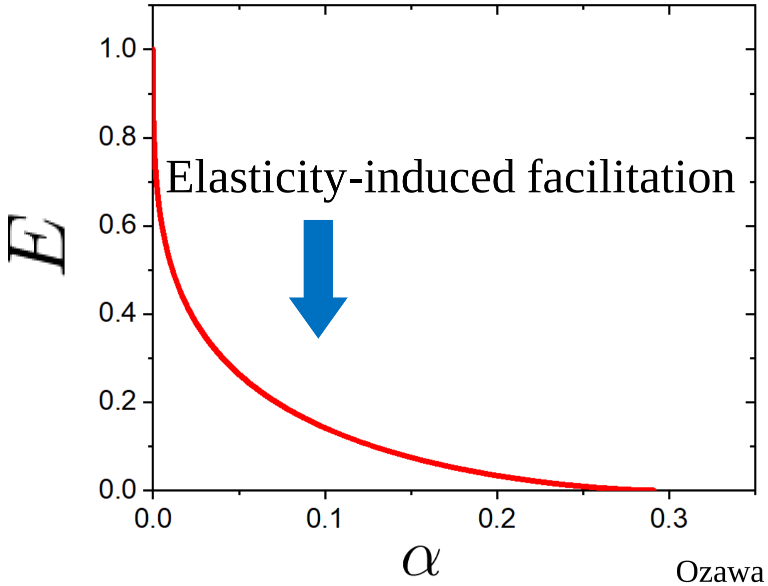
Agoritsas, Bertin, Martens, and Barrat, EPJE 2015

Magnitude of elastic interaction

$$\alpha = \frac{1}{2}\sum_{j\neq i}\left(g(\mathbf{r}_{ij})\delta\sigma_j\right)^2$$

Bocquet, Colin, Ajdari, PRL 2009

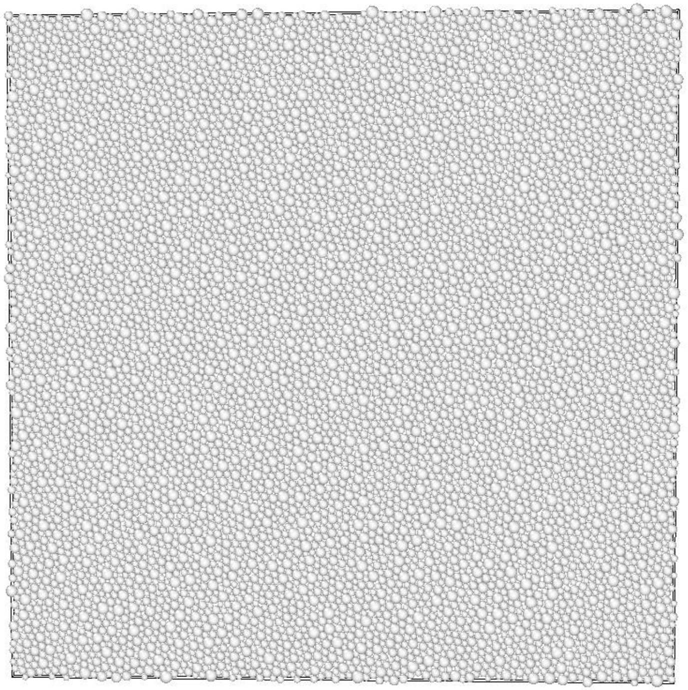
$$\tau_\alpha \simeq \Gamma^{-1} \simeq \tau_0 e^{\frac{E}{T}}$$



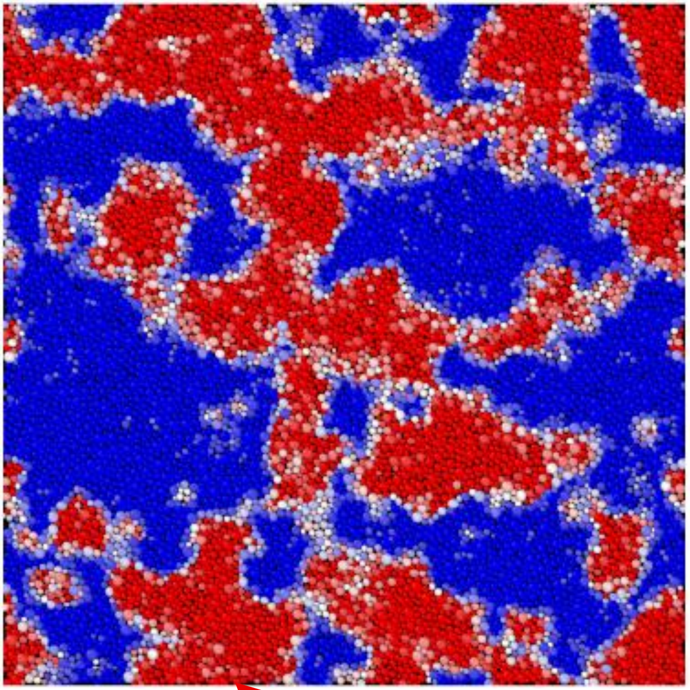
Ozawa and Biroli, PRL, 2023

Dynamical heterogeneity

Snapshot



Dynamics



Immobile

Mobile

Hurley and Harrowell, PRE 1995

Karmakar, Dasgupta, and Sastry, PNAS 2009

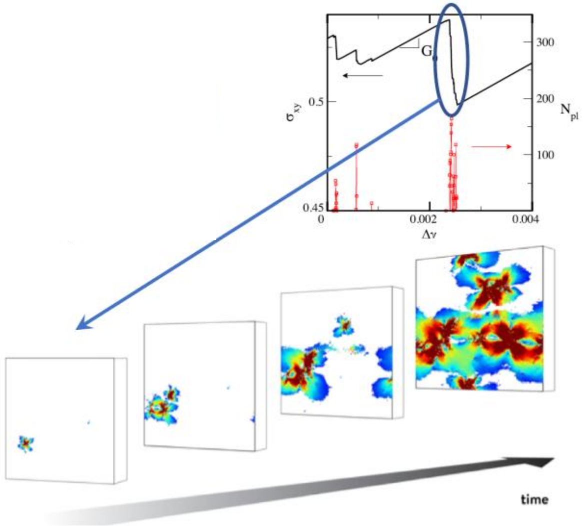
Kob, Donati, Plimpton, Poole, and Glotzer, PRL 1997

Scalliet, Guiselin, and Berthier, PRX 2022

Mechanism? Scaling theory?

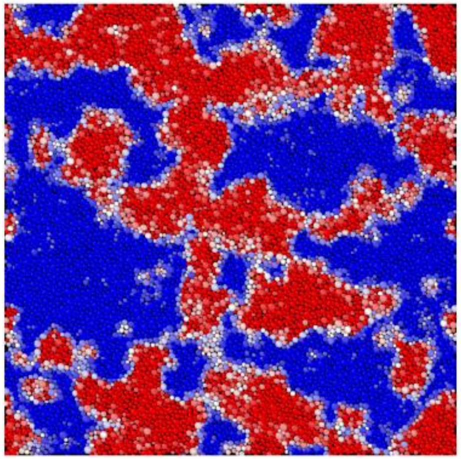
Motivation

Amorphous solids under shear



Barrat, Physica A 2018

Supercooled liquids in equilibrium



Scalliet, Guiselin, and Berthier, PRX 2022

Amorphous solids under shear are well understood: Scaling theory etc.

Nicolas, Ferrero, Martens, and Barrat, RMP 2018

Lin, Lerner, Rosso, and Wyart, PNAS 2014

Korchinski and Rottler, PRE 2022

Can we understand supercooled liquids (without shear) in a similar way?

Candelier, Widmer-Cooper, Kummerfeld, Dauchot, Biroli, Harrowell, and Reichman, PRL 2010

Yanagishima, Russo, and Tanaka, Nat. Commun. 2017

Scaling theory (summary)

$T > 0$

Four point correlation

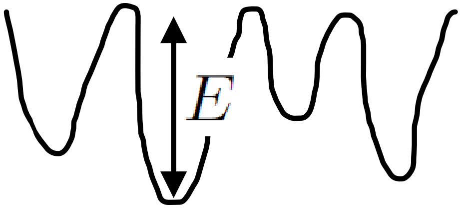
$$\chi_4^* \sim T^{-\gamma}$$

Dynamic correlation length

$$\xi \sim T^{-\nu}$$

$T = 0$ (Critical point)

Local energy barrier E



Statistics of energy barriers

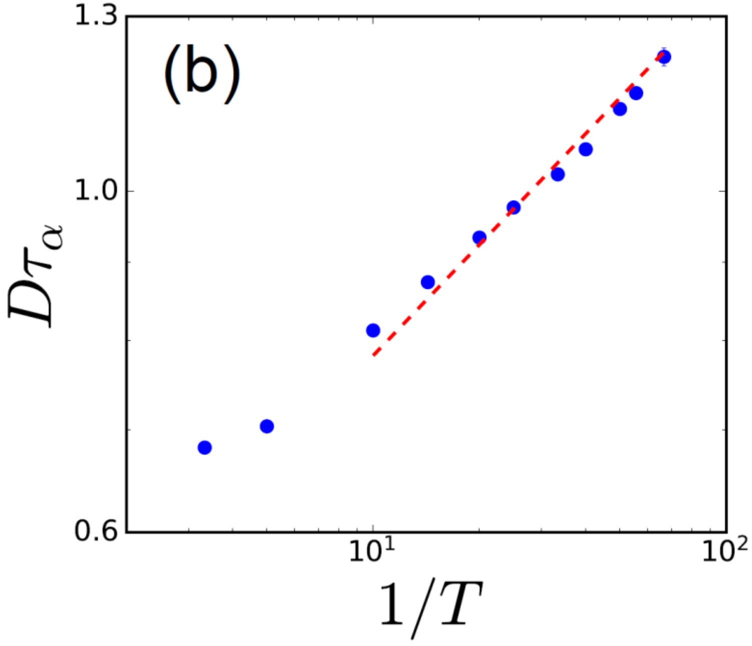
$$\langle E_{\text{second}} - E_{\text{min}} \rangle \sim N^{-\delta} \qquad N = L^d$$

Scaling relations

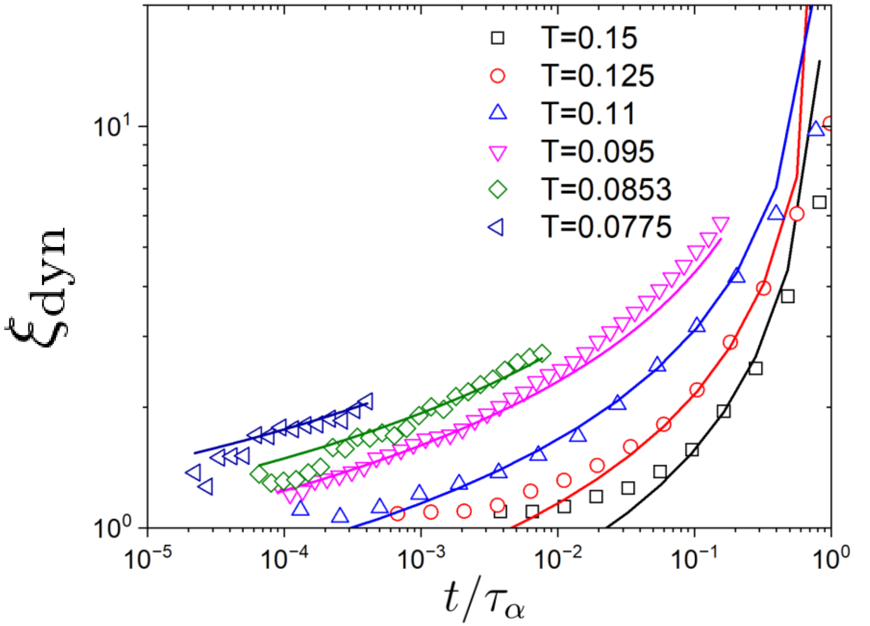
$$\gamma = \frac{\tilde{d}_f}{\delta d} \qquad \nu = \frac{1}{\delta d}$$

Other predictions

Stokes-Einstein violation



Time-evolution of $\xi_{\rm dyn}$



Accumulation of relaxation events

$$D\tau_\alpha \sim T^{-h}$$

$$h = \nu(d_f - \tilde{d}_f)$$

Logarithmic time dependence

$$\xi_{\rm dyn} = A(T)\left[T\ln(\tau_\alpha/t)\right]^{-1/(\tilde{\sigma}\tilde{d}_f)}$$

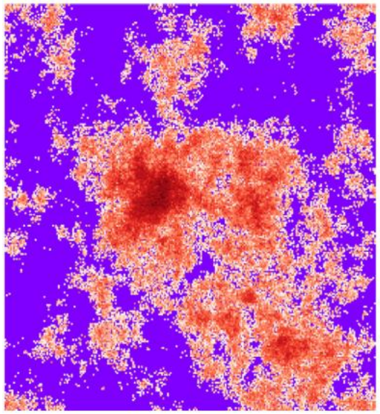
Jung, Garrahan, and Chandler, PRE 2004

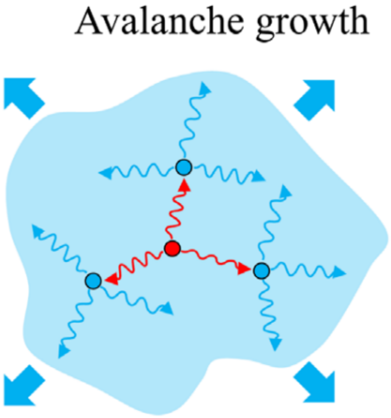
Pastore, Kikutsuji, Rusciano, Matsubayashi, Kim, and Greco, JCP 2021

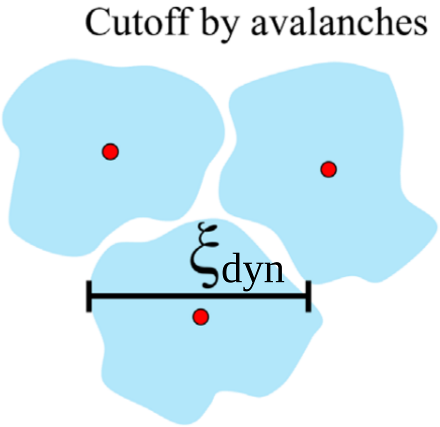
Scalliet, Guiselin, and Berthier, PRX. 2022

Tahaei, Biroli, Ozawa, Popovic, and Wyart, PRX, 2023

Avalanche of relaxation




Avalanche growth


Cutoff by avalanches

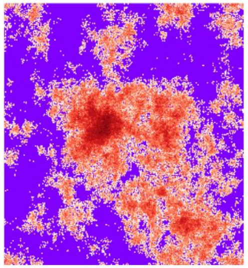
Dynamical heterogeneity is an avalanche induced by thermal fluctuations

Tahaei, Biroli, Ozawa, Popovic, and Wyart, PRX 2023

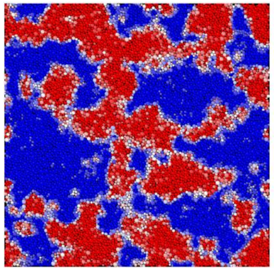
Open discussions

Avalanche-like

Coarsening-like



?



Tahaei, Biroli, Ozawa, Popovic, and Wyart, PRX 2023

Scalliet, Guiselin, and Berthier, PRX. 2022

Detailed balance, time reversal symmetry is important?

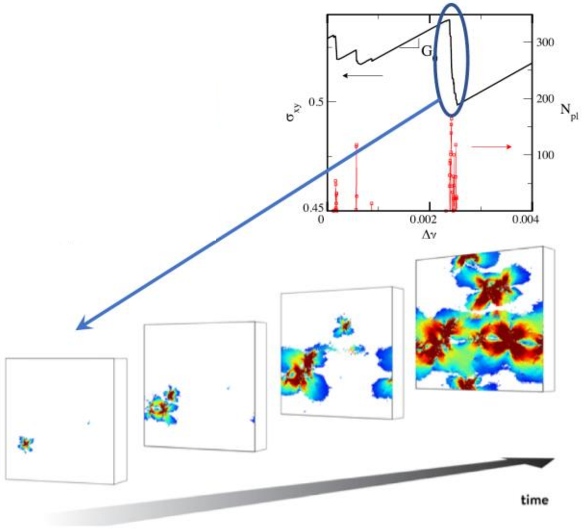
Ridout and Liu, arXiv 2024

Are elastic interactions screened at finite temperature?

Lemaître, Mondal, Moshe, Procaccia, Roy, Screiber-Re’em, PRE 2021

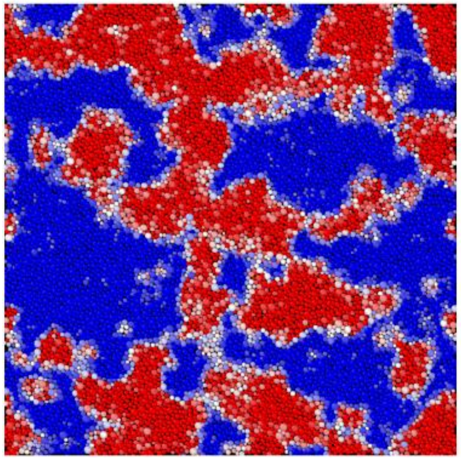
Motivation

Amorphous solids under shear



Barrat, Physica A 2018

Supercooled liquids in equilibrium



Scalliet, Guiselin, and Berthier, PRX 2022

Amorphous solids under shear are well understood: Scaling theory etc.

Nicolas, Ferrero, Martens, and Barrat, RMP 2018

Lin, Lerner, Rosso, and Wyart, PNAS 2014

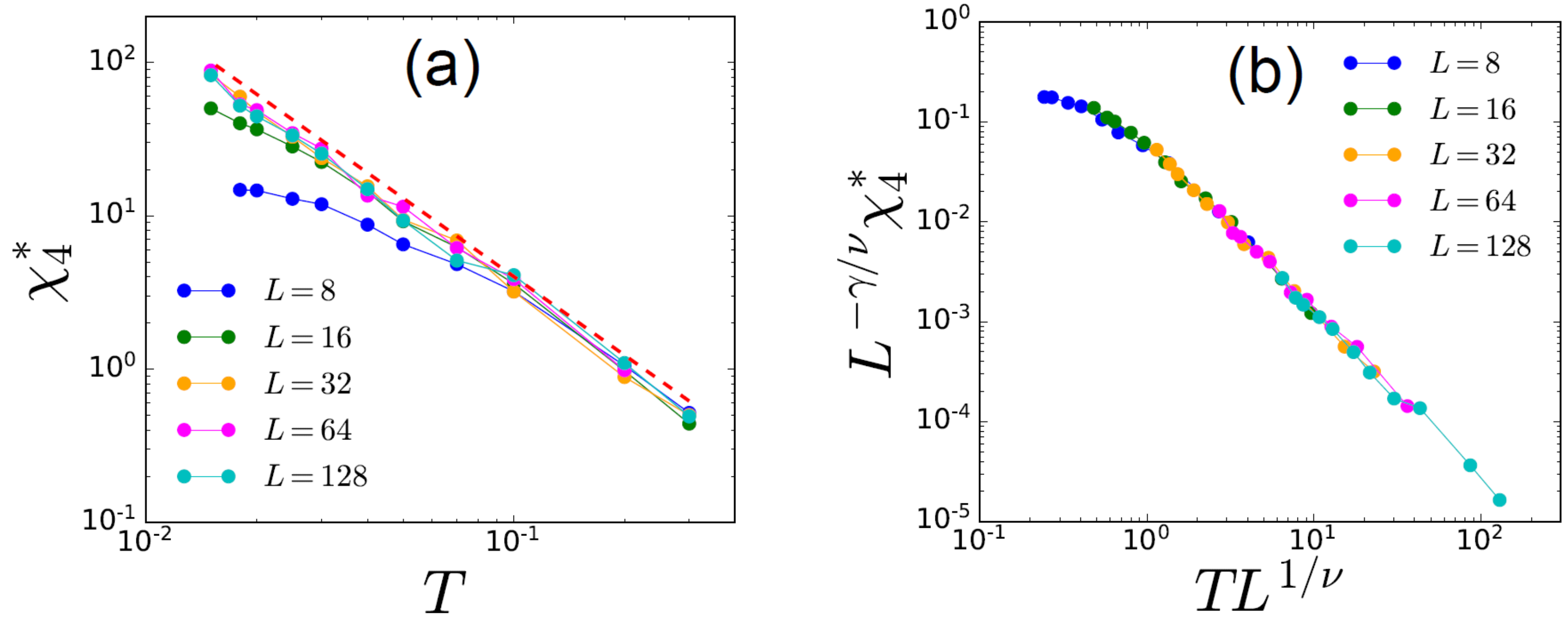
Korchinski and Rottler, PRE 2022

Can we understand supercooled liquids (without shear) in a similar way?

Candelier, Widmer-Cooper, Kummerfeld, Dauchot, Biroli, Harrowell, and Reichman, PRL 2010

Yanagishima, Russo, and Tanaka, Nat. Commun. 2017

Finite size scaling at $T > 0$



We confirmed scalings $\chi_4^* \sim T^{-\gamma}$, $\xi \sim T^{-\nu}$,

and determined critical exponents γ, ν

Tahaei, Biroli, Ozawa, Popovic, and Wyart, PRX, 2023

Extremal dynamics at $T = 0^+$

$T > 0$ simulation

iii) Thermal fluctuation: Elastic → Plastic with probability $e^{-E(\sigma_i)/T}$

$T = 0^+$ simulation

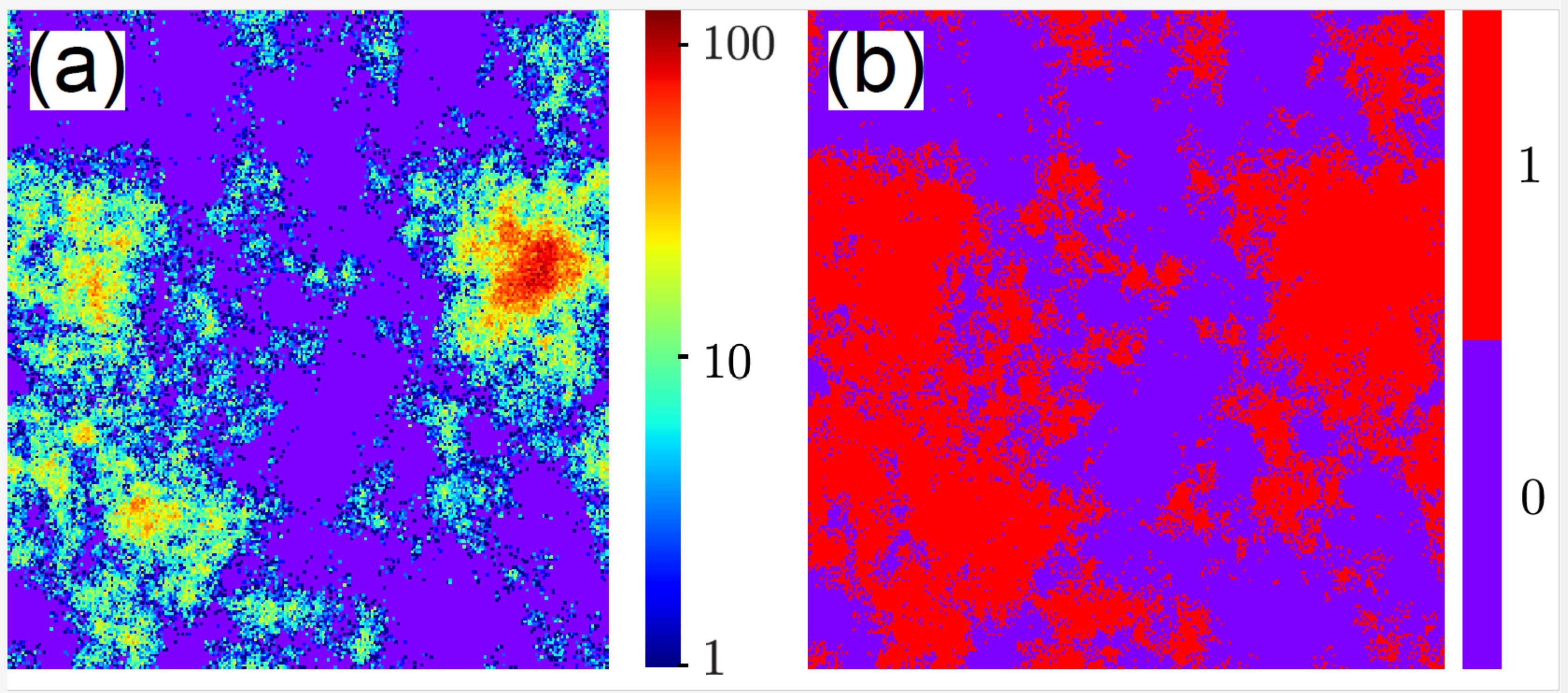
iii) Elastic → Plastic for the site with the lowest energy barrier E_{min}

Always choosing the weakest site: Extremal dynamics

Paczuski, Maslov, and Bak, PRE 1996

Kumar, Patinet, Maloney, Regev, Vandembroucq, and Mungan, JCP 2022

Avalanches in extremal dynamics



Event based avalanche size : S
(Total number of relaxation events)

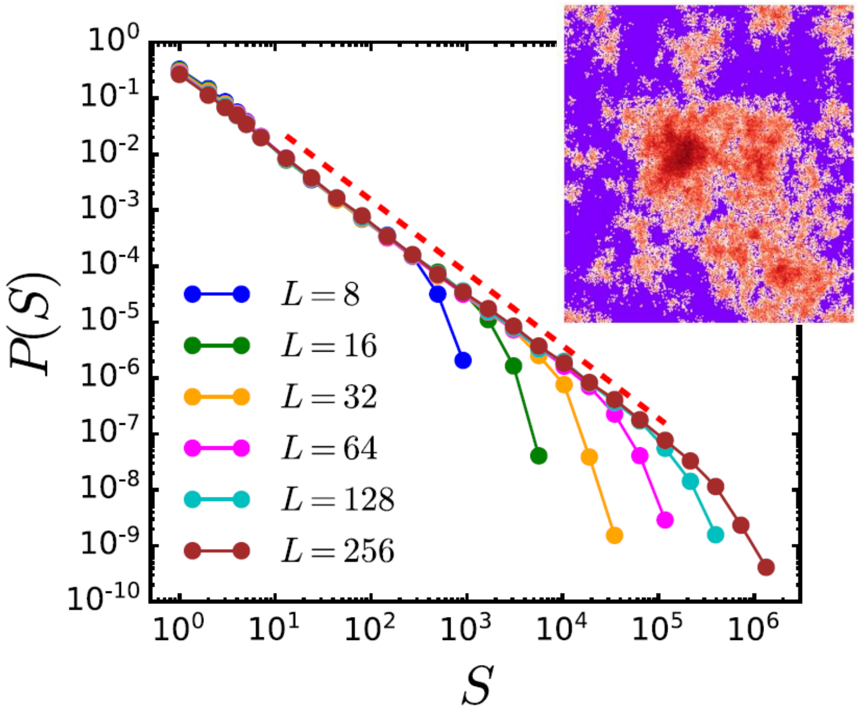
Site based avalanche size: $\tilde{S}$
(Total number of sites relaxed at least once)

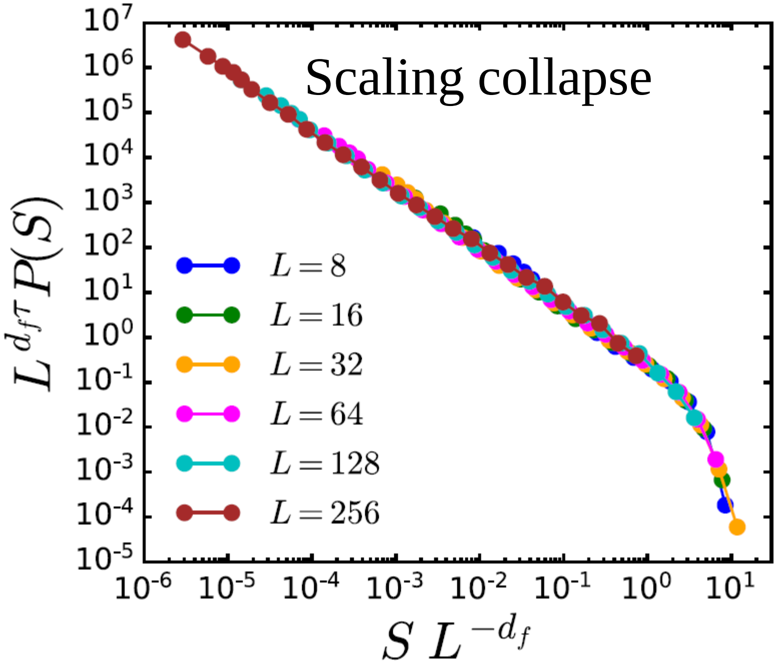
Accumulation of events

Related to χ_4

Tahaei, Biroli, Ozawa, Popovic, and Wyart, PRX, 2023

Avalanches statistics



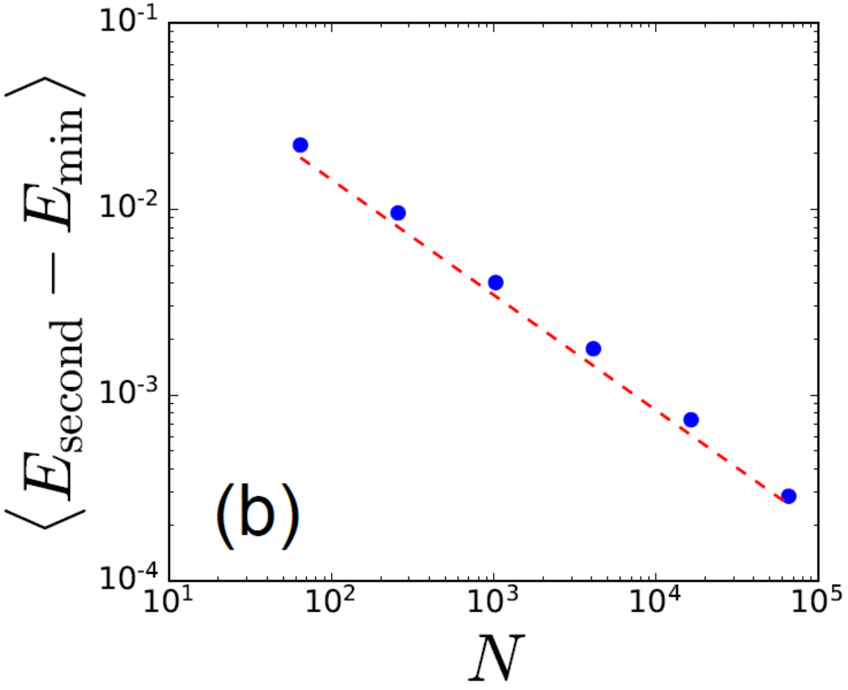


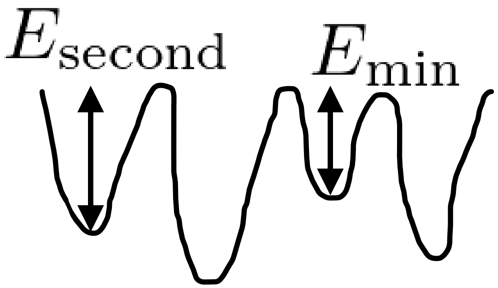
S : Size of an avalanche

$P(S) \sim S^{-\tau} f(S/L^{d_f})$: Its distribution

Dynamics at $T = 0^{+}$ follows avalanche statistics

Statistics of local energy barriers at $T = 0^+$





$E_{\rm min}$: The smallest energy barrier

$E_{\rm second}$: The second smallest energy barrier

We confirmed $\langle E_{\rm second} - E_{\rm min} \rangle \sim N^{-\delta}$

Tahaei, Biroli, Ozawa, Popovic, and Wyart, PRX, 2023

Scaling argument

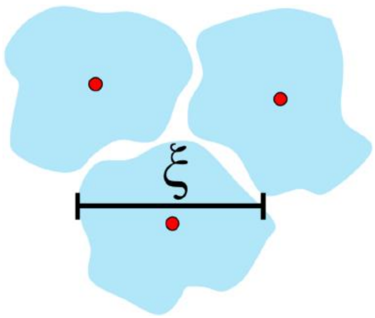
Tahaei, Biroli, Ozawa, Popovic, and Wyart, PRX, 2023

See also Korchinski and Rottler, PRE 2022

i) $k_\mathrm{B}T \ll \langle E_\mathrm{second} - E_\mathrm{min} \rangle$ Extremal dynamics holds

ii) $k_\mathrm{B}T \gg \langle E_\mathrm{second} - E_\mathrm{min} \rangle$ Extremal dynamics breaks down

Crossover lengthscale ξ



$$k_\mathrm{B}T \sim \langle E_\mathrm{second} - E_\mathrm{min} \rangle \sim N^{-\delta} \sim (\xi^d)^{-\delta}$$

$$\Rightarrow \quad \xi \sim T^{-\nu} \text{ with } \nu = \frac{1}{\delta d}$$

$$\chi_4^* \sim \xi^{\tilde{d}_f}$$

$$\Rightarrow \quad \chi_4^* \sim T^{-\gamma} \text{ with } \gamma = \frac{\tilde{d}_f}{\delta d}$$