Structural glasses, from Flatland to infinite dimensions

Elisabeth Agoritsas
(DQMP, University of Geneva, Switzerland)

Dense amorphous materials which exhibits a solid-like behaviour, or 'structural glasses', are ubiquitous around us. They span a very wide range of experimental systems in soft matter, condensed matter, or biophysics, such as emulsions, foams, biological tissues, metallic glasses, etc. From a material sciences perspective, **their structural disorder can be tuned depending on their initial preparation and the subsequent driving history.** This has key implications for the mechanical and transport properties of such materials, quite distinct compared to crystalline materials, a**nd paves the way to engineer specific disorder-induced properties**.

Theoretical descriptions of dense amorphous materials remain challenging, though crucially needed for rationalising the wide range of observed dynamical features (and assessing their potential universality). One successful approach is based on coarsed-grained descriptions, with ad hoc effective ingredients. Another approach, which might seem more abstract at first, consists in playing with the spatial dimension. In this talk I will discuss **which (exact) insights can be obtained from the infinite-dimensional limit, and why/how they can be informative for the low-dimensional space (2D, 3D)** we actually live in.

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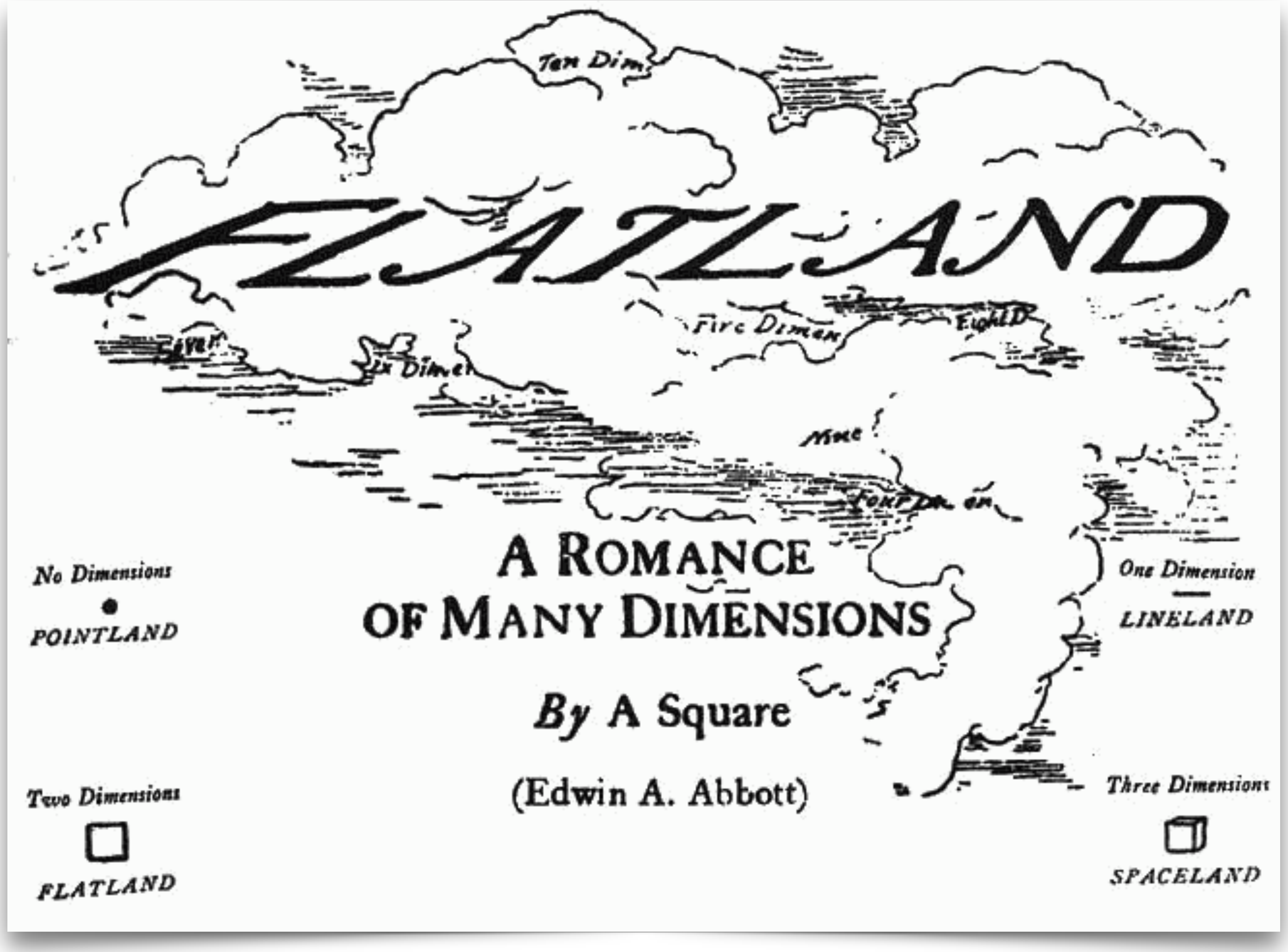


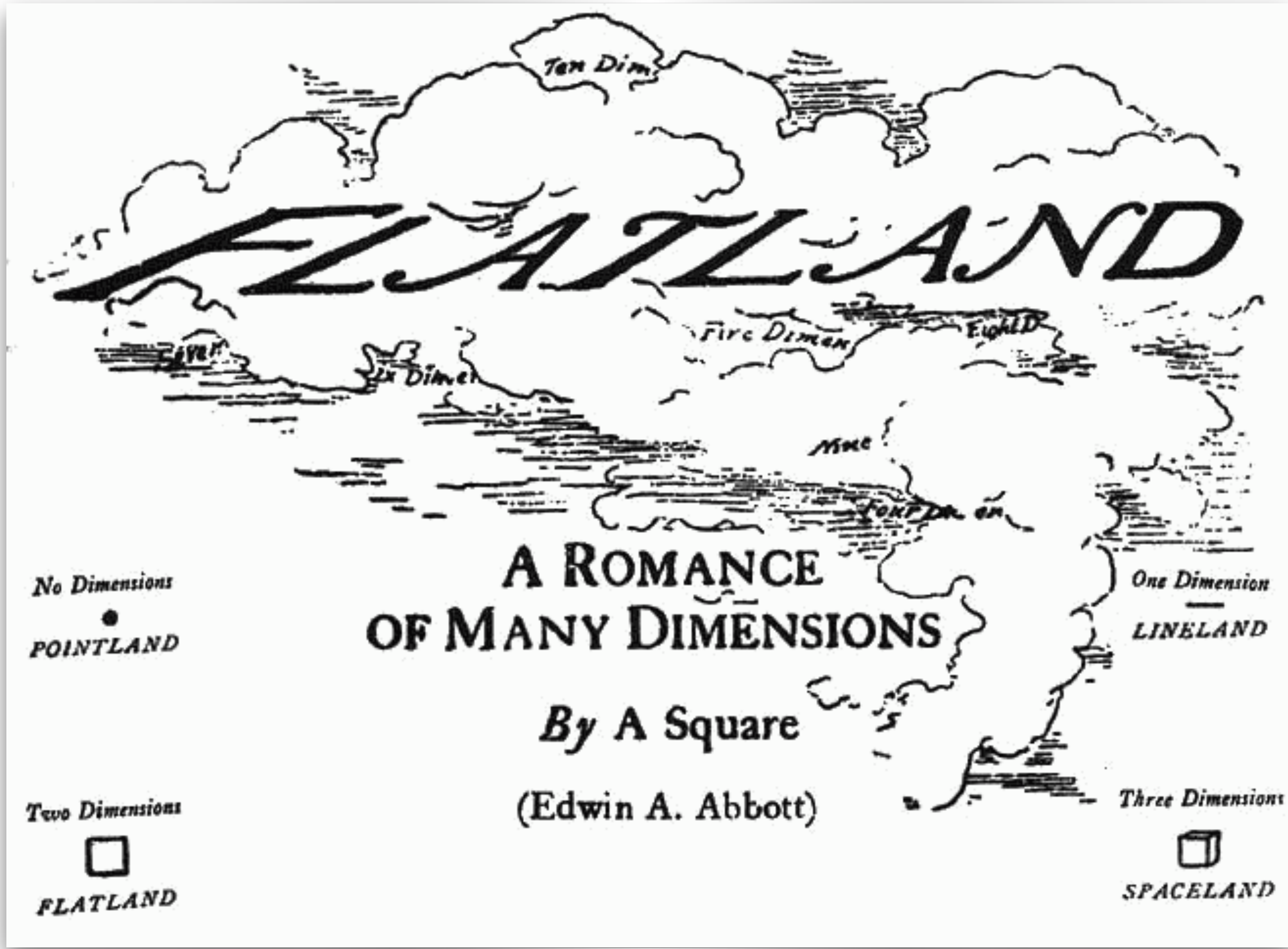
SIMONS FOUNDATION
Advancing Research in Basic Science and Mathematics

"Crackling the Glass Problem" (2015-23)

[https://scglass.uchicago.edu]

ENS École Normale Supérieure | PSL Research University Paris | erc European Research Council Established by the European Commission | cnrs | Swiss National Science Foundation | EPFL | Schola Genevensis

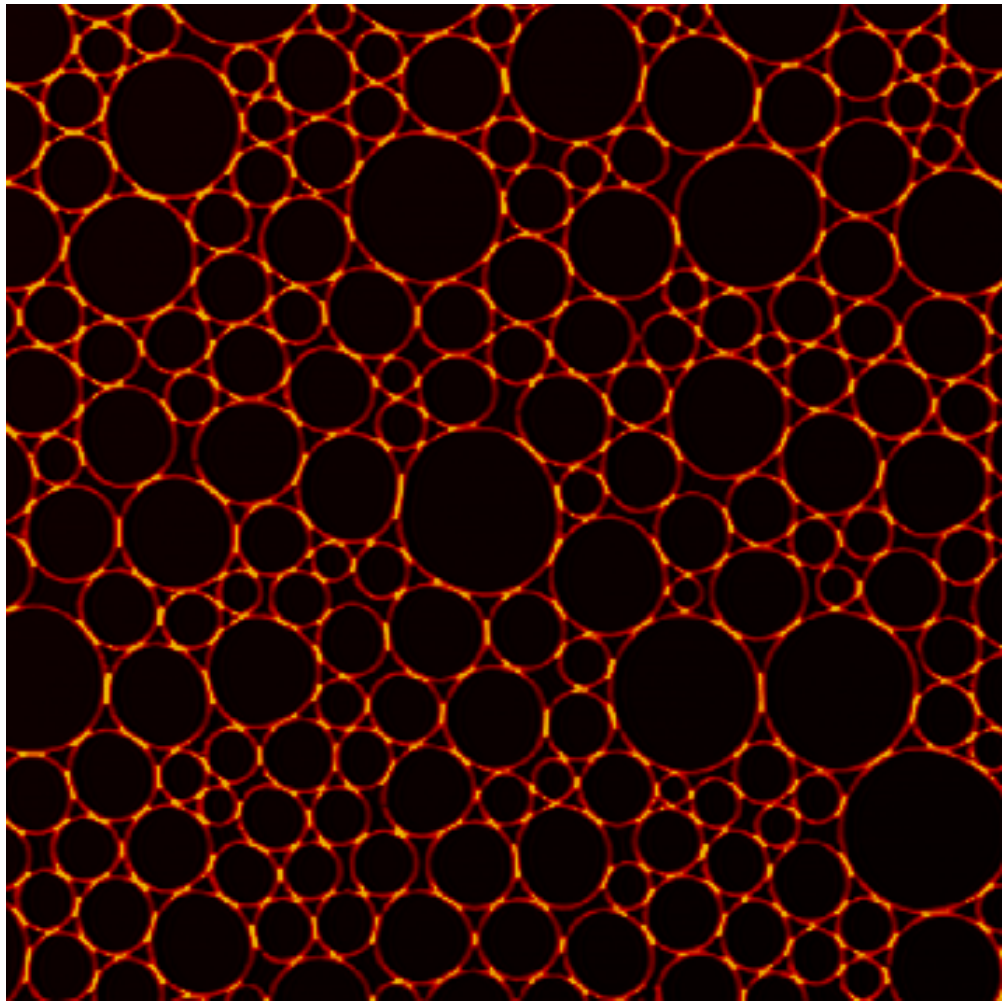
Structural glasses, from Flatland to infinite dimensions



...

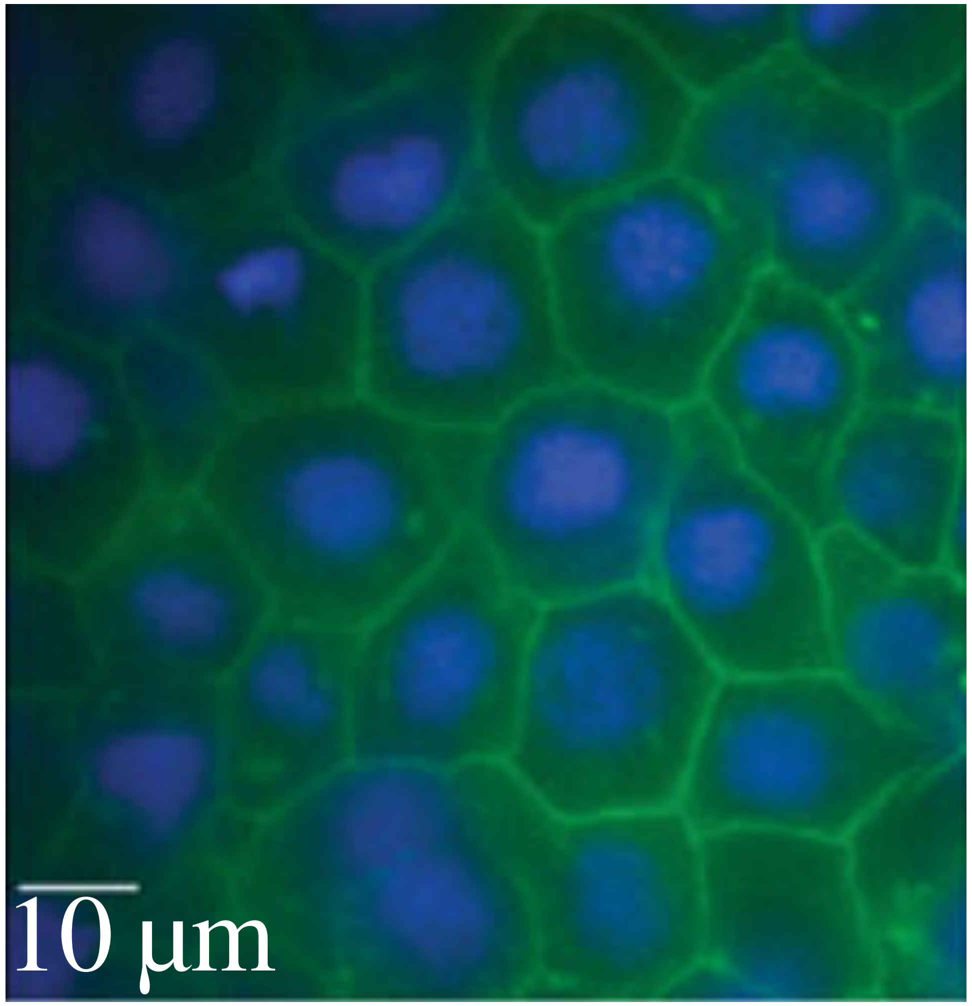
Infinite Dimensions
???

Dense structurally disordered systems



Emulsion droplets, sandwiched between two glass plates.
Picture from Ken Desmond (Emory University).

Dense amorphous materials



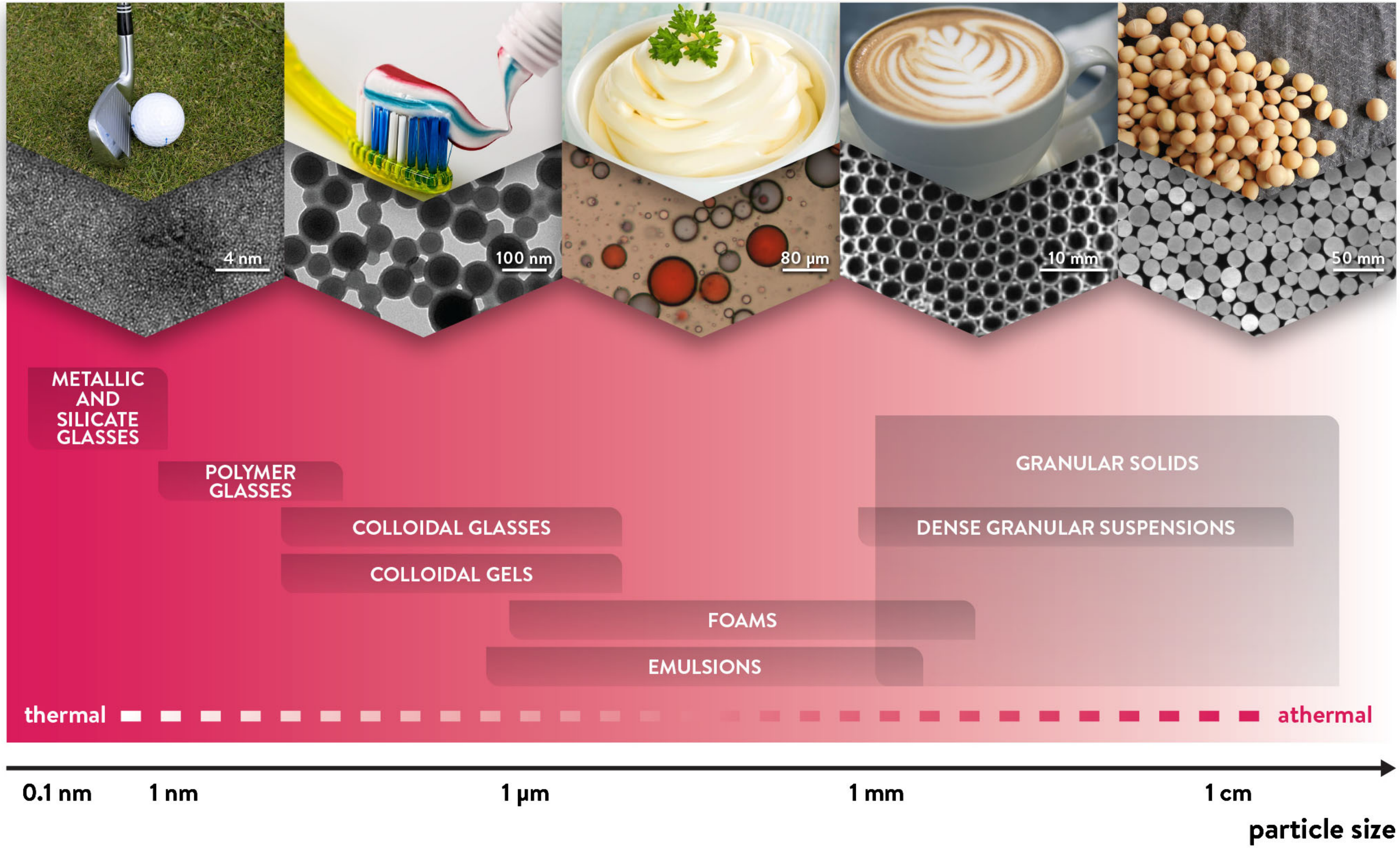
2D cross-section of zebrafish embryonic tissue
E.-M. Schötz, et al. *J. R. Soc. Interface* 10, 20130726 (2013).

Dense active matter

Structural disorder (dynamical) ⇄ Mechanical/rheological/transport properties

Broad range of characteristic scales

A. Nicolas, E. E. Ferrero, K. Martens, J.-L. Barrat, *Rev. Mod. Phys.* 90, 045006 (2018).

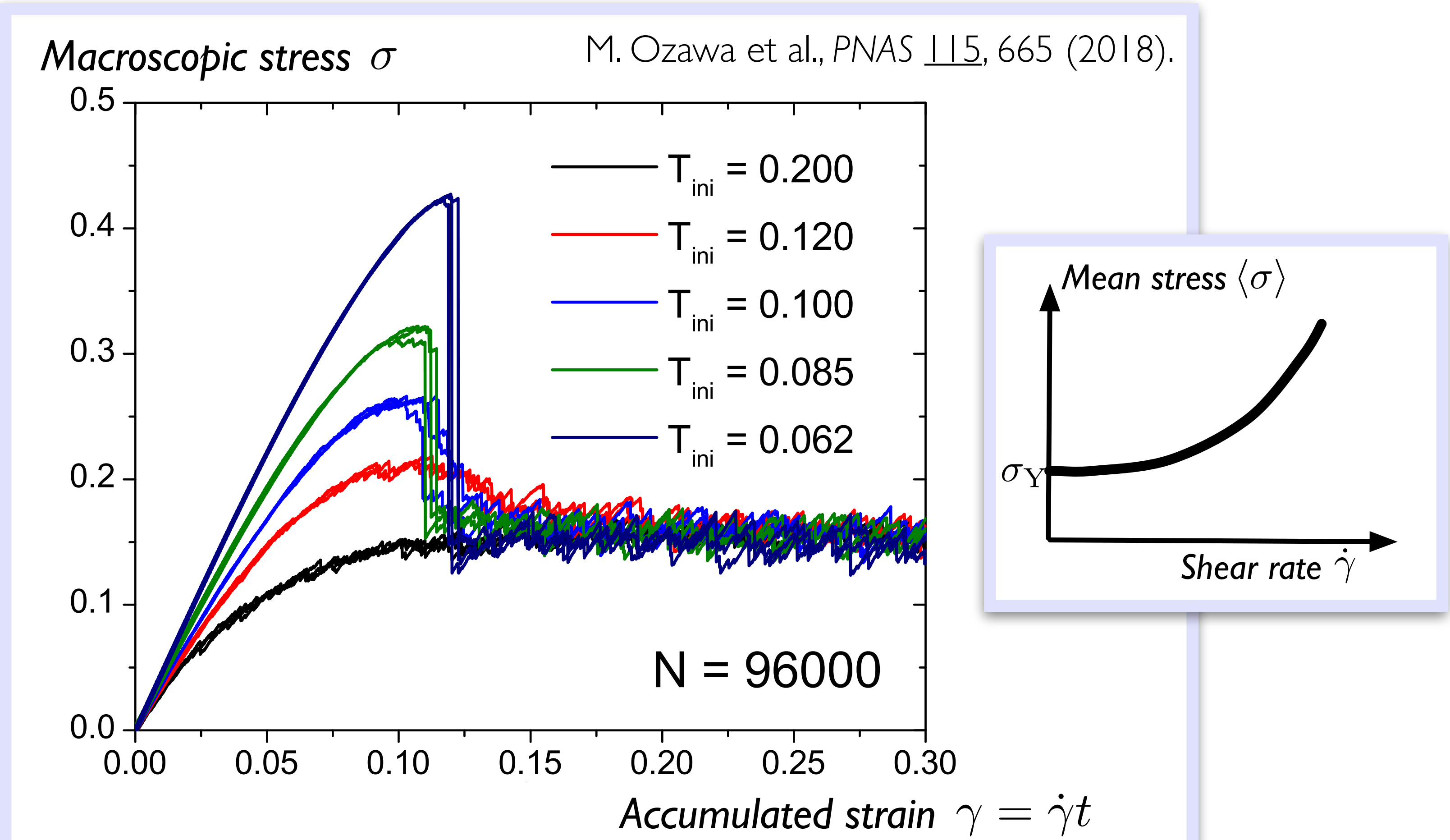


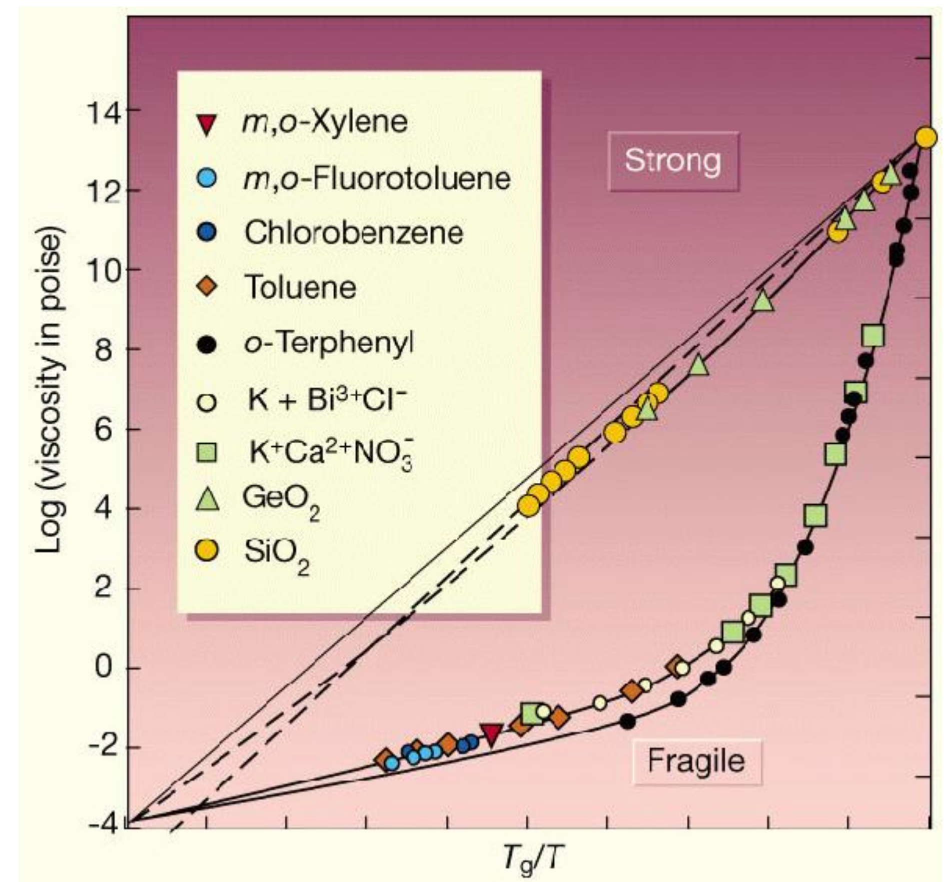
Solid-like behaviours: **Mechanics**?

Liquid-like behaviours: **Rheology**?

Disorder-induced properties

- **Mechanical** properties: e.g. stress-strain curves
- **Rheological** properties: e.g. flow curves, viscosity





P. G. Debenedetti & F. H. Stillinger, *Nature* 410, 259 (2001).

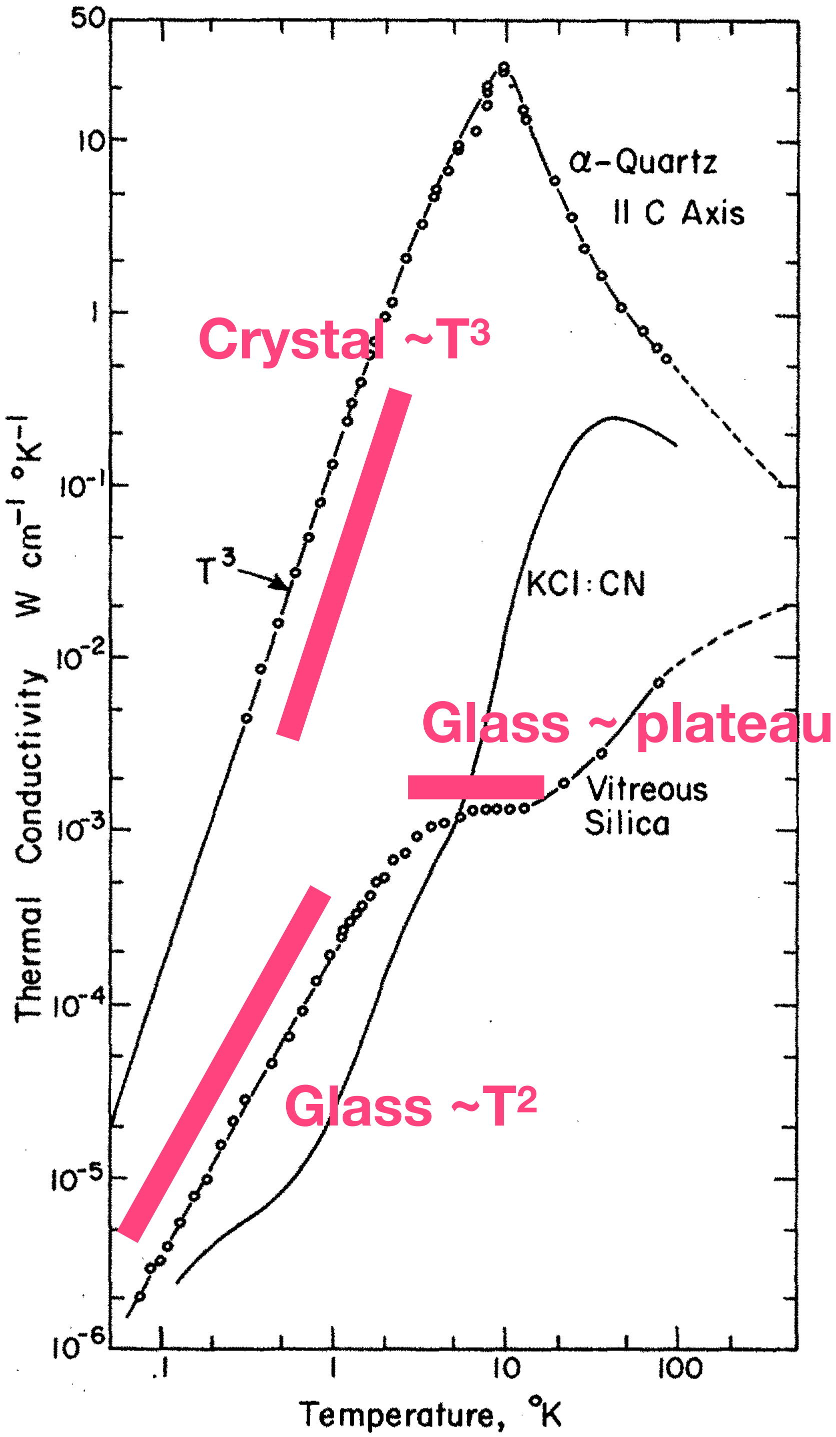
Brittle-to-ductile

Yielding transition

Glass transition

Disorder-induced properties

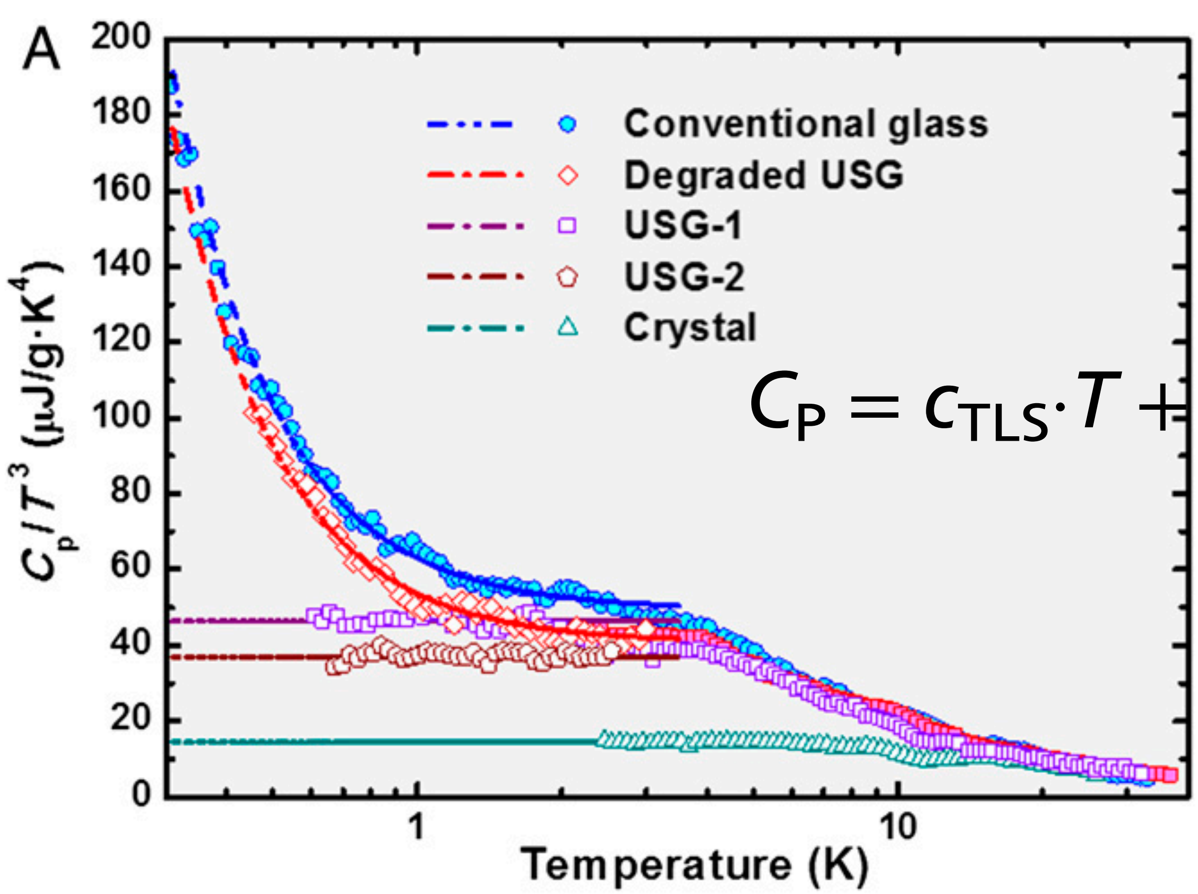
- **Transport** properties: e.g. density of two-level systems (TLSs)



R. C. Zeller & R. O. Pohl, *Phys. Rev. B* 4, 2029 (1971), "Thermal Conductivity and Specific Heat of Noncrystalline Solids"

Low-temperature anomalies in structural glasses

Tunable density of two-level systems (TLSs)?



$$C_P = c_{TLS} \cdot T + c_D \cdot T^3$$

T. Perez-Castaneda et al., *PNAS* 111, 11275 (2014), "Suppression of tunneling two-level systems in ultrastable glasses of indomethacin" [vapor-deposited glasses]

D. Khomenko *et al.*, *Phys. Rev. Lett.* 124, 225901 (2020)., "**Depletion of Two-Level Systems in Ultrastable Computer-Generated Glasses**"

Disorder-induced properties

- **Optical** properties: e.g. structural colors

__Brand new review__: K. Vynck et al., "Light in correlated disordered media", *Rev. Mod. Phys.* __95__, 045003 (2023).

[⇒ Prof. F. Scheffold @UniFribourg]

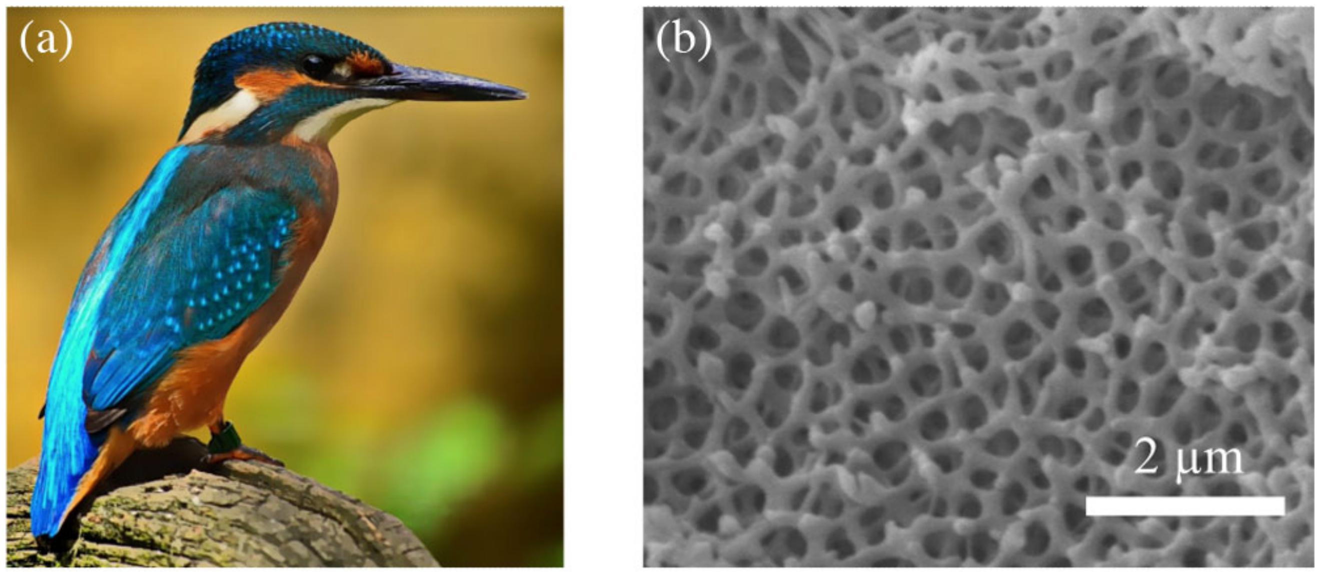
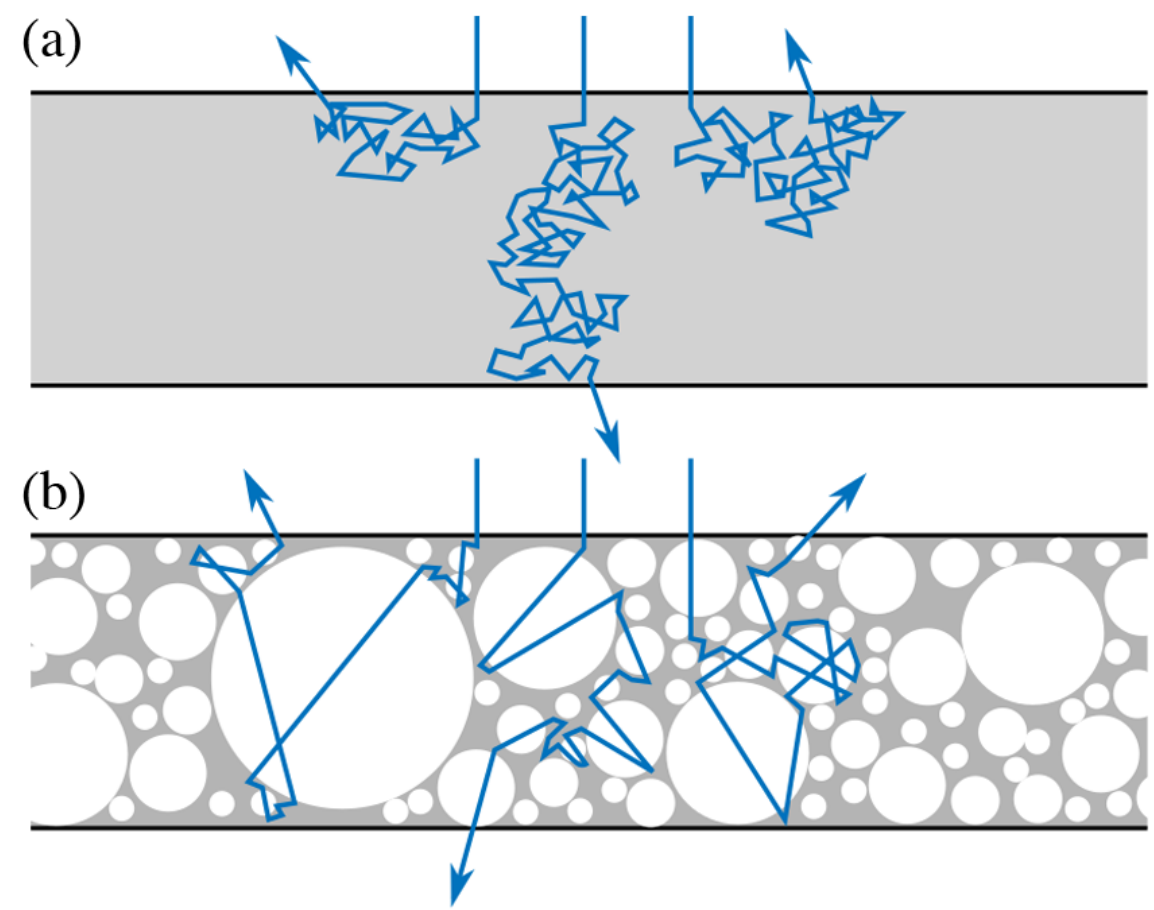


FIG. 17. (a) Kingfisher. The angular-independent blue (light gray) coloration of the bird feather is the result of a correlated 3D structure, which is shown in the SEM image in (b). See Stavenga *et al.* (2011) for more information. (a) Courtesy of Pixabay.

Stavenga, D. G., J. Tinbergen, H. L. Leertouwer, and B. D. Wilts, 2011, "Kingfisher feathers—Colouration by pigments, spongy nanostructures and thin films," J. Exp. Biol. **214**, 3960–3967.

FIG. 8. Impact of large-scale heterogeneity on transport



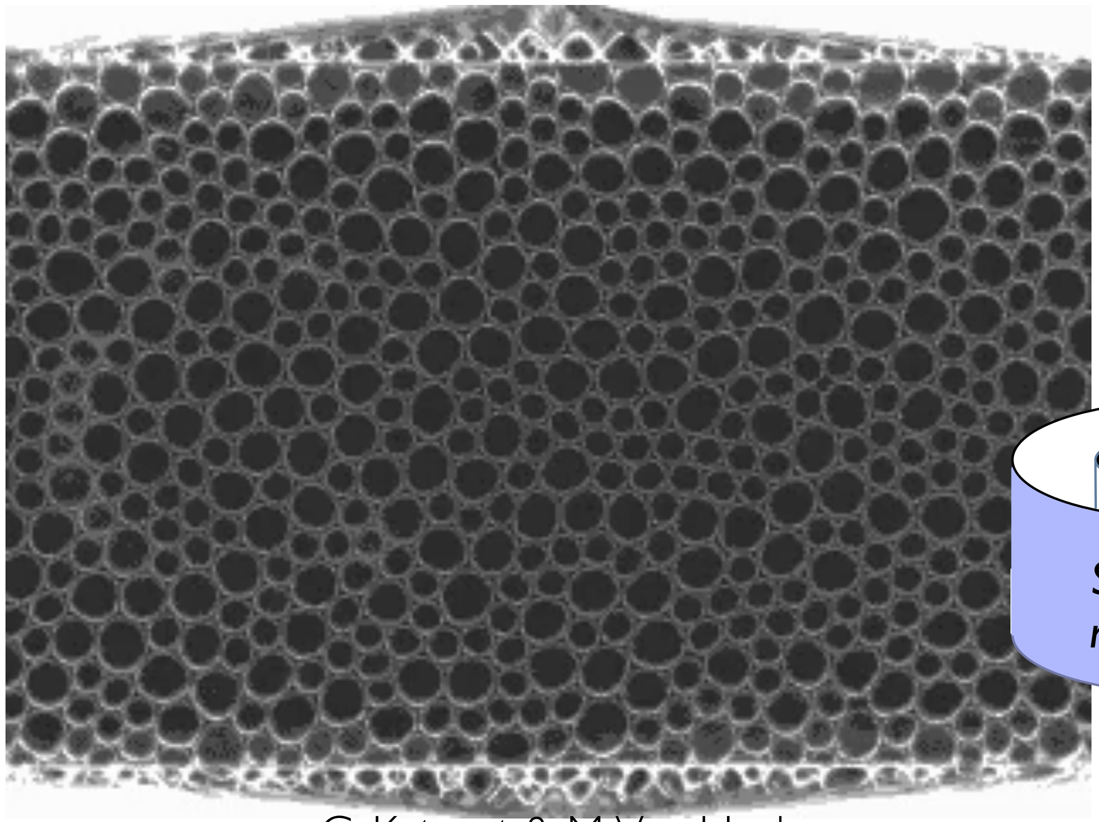
- Upscaled experimental models for metals: monolayers of bubbles

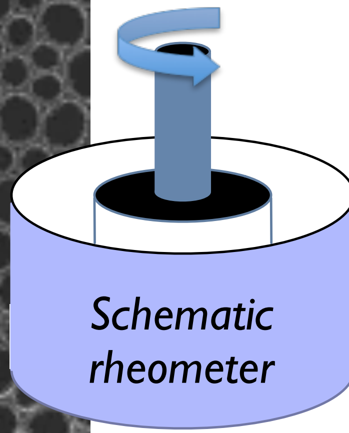
Monodisperse: crystalline

L. Bragg & J.F. Nye, *Proc. R. Soc. A* 190, 474 (1947)

Bidisperse/polydisperse: amorphous

A.S. Argon & H.Y. Kuo, *Mater. Sci. Eng.* 39, 101 (1979)

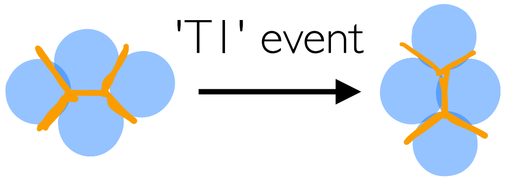




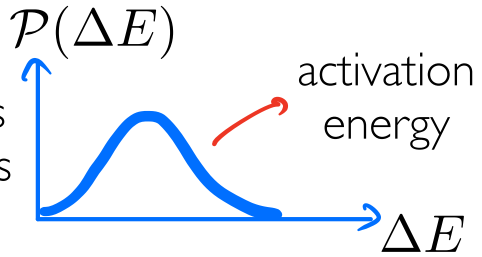
G. Katgert & M. Van Hecke,
http://www.physics.leidenuniv.nl/vanhecke

Dislocation dynamics

Key excitations: rapid **irreversible/plastic local rearrangements**



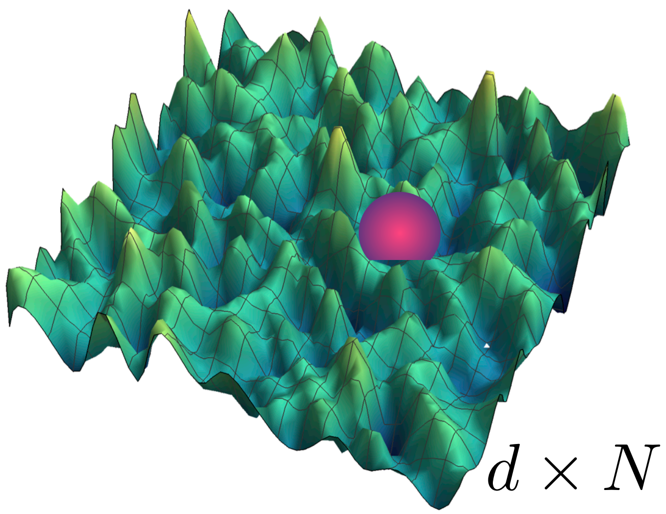
in metallic glasses
~20-700 particles
involved



Landscape picture & local excitations/defects

Structural disorder (dynamical)

Mechanical/rheological/transport properties

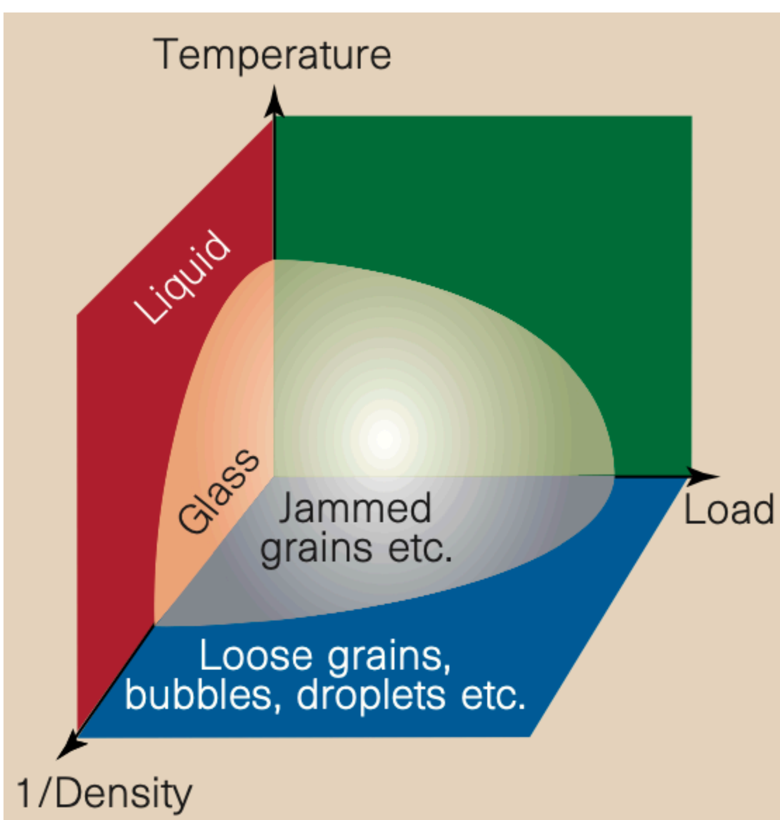


$\approx$ Statistical features of high-dim. rough landscape

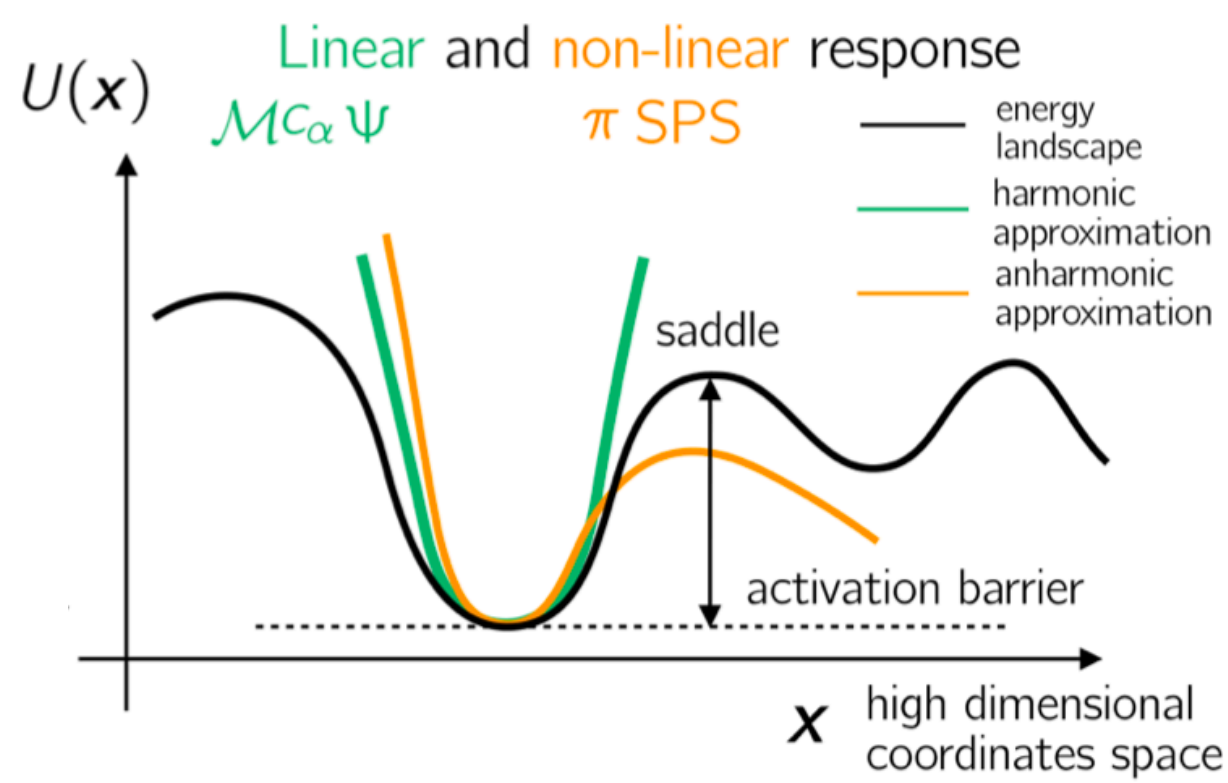
& 3 elementary excitations

Curvature of minima: density of states
Distribution of barriers, saddles, flat directions
Connectivity between minima

Shear Transformation Zones (STZs)
Thermal activation
Quantum tunneling in TLSs



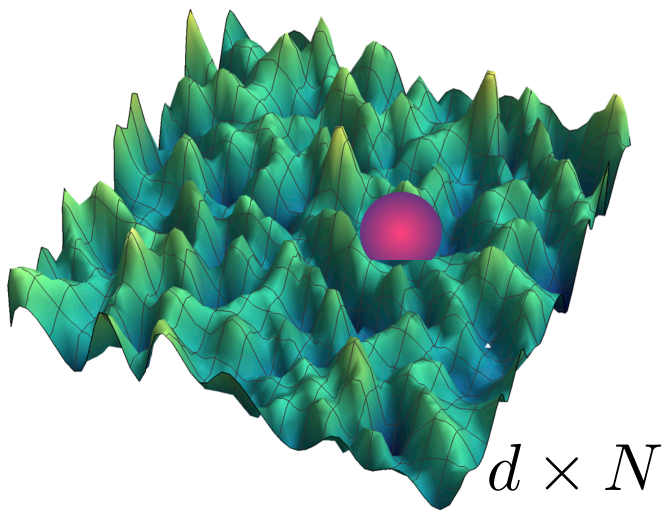
A.J. Liu & S.R. Nagel, *Nature* 396, 6706 (1998).
A.J. Liu & S.R. Nagel, *Annu. Rev. Condens. Matter Phys.* 1, 347 (2010).



D. Richard *et al.*, *Phys. Rev. Materials* 4, 113609 (2020)

Landscape picture & local excitations/defects

Structural disorder (dynamical) ⇄ Mechanical/rheological/transport properties

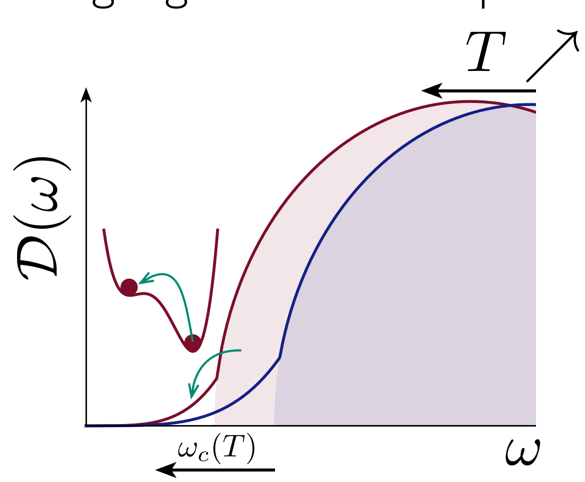


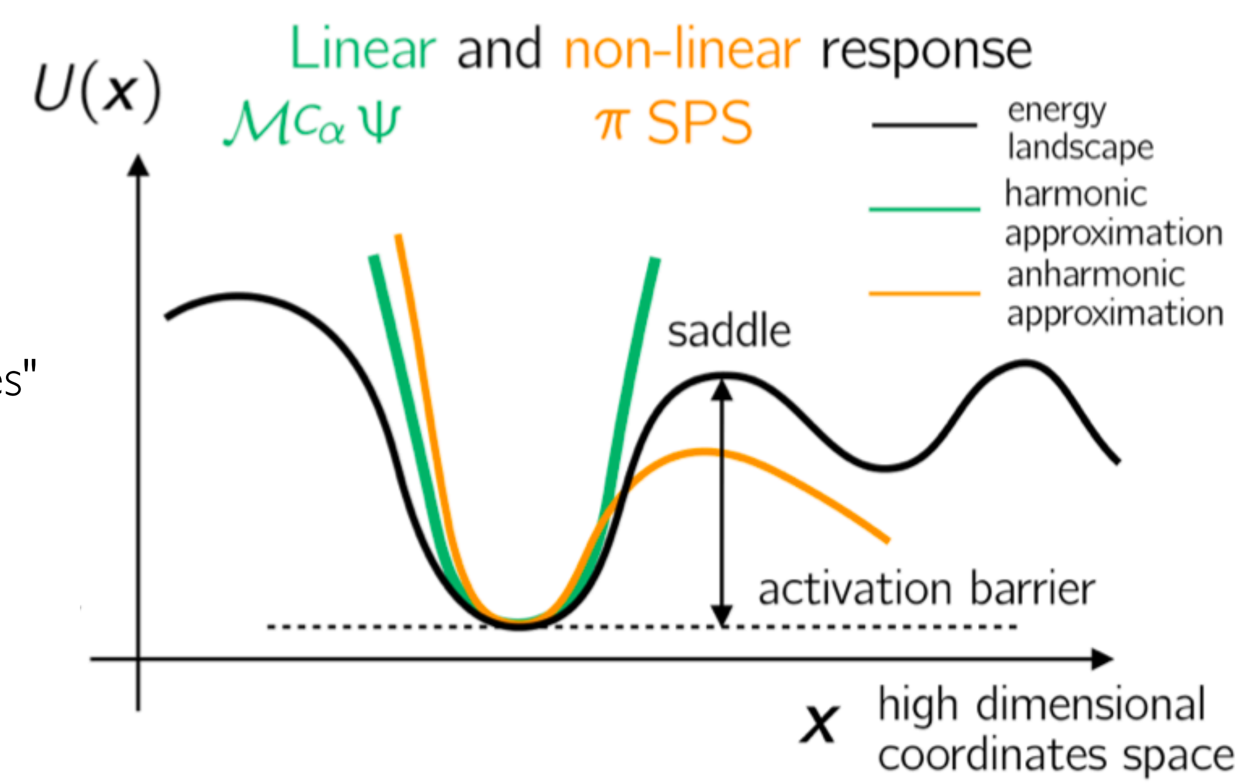
$\approx$ Statistical features of high-dim. rough landscape & 3 elementary excitations

Curvature of minima: density of states
Distribution of barriers, saddles, flat directions
Connectivity between minima

Shear Transformation Zones (STZs)
Thermal activation
Quantum tunneling in TLSs

- Hessian of the energy functional
  ⇒ **Spectrum of vibrational (harmonic) modes**
  ⇒ spectrum of vibrational (harmonic) modes
  $$\mathcal{D}(\omega) \sim \omega^4$$
  ⇒ corresponding eigenmodes = "quasi-localised modes"

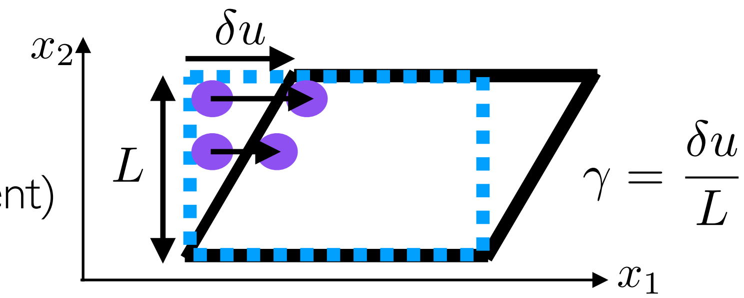




D. Richard *et al.*, *Phys. Rev. Materials* 4, 113609 (2020)

In practice: numerics/analytics

- **Microscopic descriptions**: Langevin-like dynamics



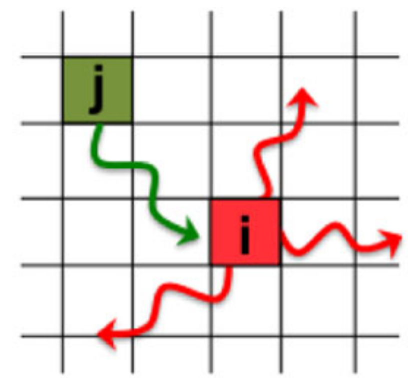
$$\overbrace{\zeta\left[\dot{\mathbf{x}}_i(t) - \dot{\gamma}(t) x_2(t)\, \hat{\mathbf{x}}_1\right]}^{\textit{Local friction}} = \overbrace{\boldsymbol{F}_i(t)}^{\textit{Interaction}} + \overbrace{\boldsymbol{\xi}_i(t)}^{\textit{Noise}}$$

$$\boldsymbol{F}_i(t) = -\sum_{j(\neq i)} \nabla v_{ij}\left(|\mathbf{x}_i(t) - \mathbf{x}_j(t)|\right)$$

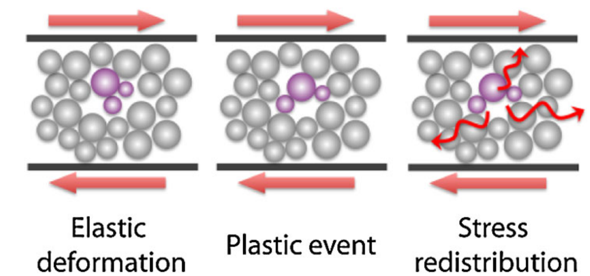
- **Coarse-grained descriptions**: elasto-plastic models, 'trap' models

  = Effective models based on local excitations, with *ad hoc* ingredients

  Review: A. Nicolas et al., *Rev. Mod. Phys.* 90, 045006 (2018).



L. Bocquet et al., *PRL* 103, 036001 (2009).



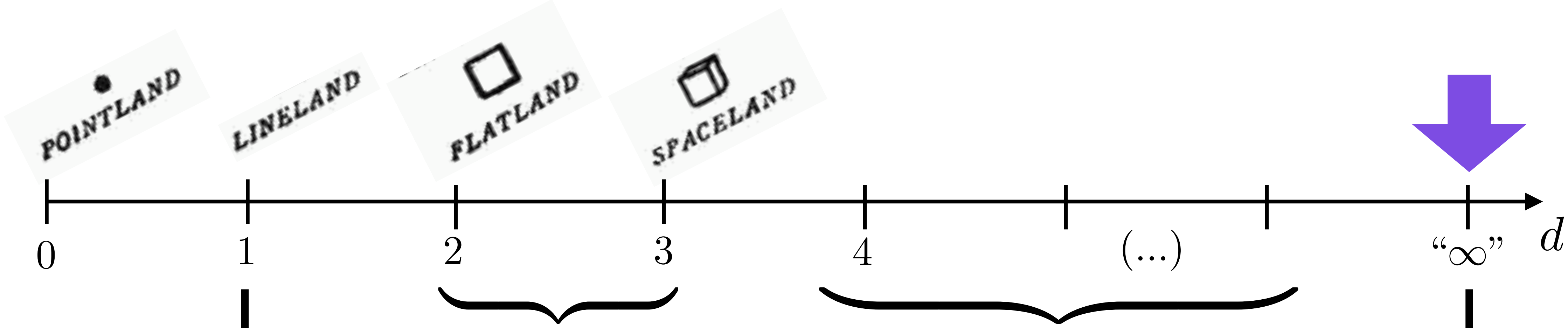
- **Numerical limitations**:
  - Bounded exploration of phase space
  - Finite-size & finite-statistics artefacts / Need to average at least on initial condition & noise
  - Out-of-equilibrium dense systems: very slow to equilibrate ($\Rightarrow$ need SWAP algorithm!)

- **Analytical approaches needed**:

  for supporting/rationalizing numerical findings

  complement numerics beyond their inherent limitations

Playing with spatial dimension



Our physical intuition

Our physical space

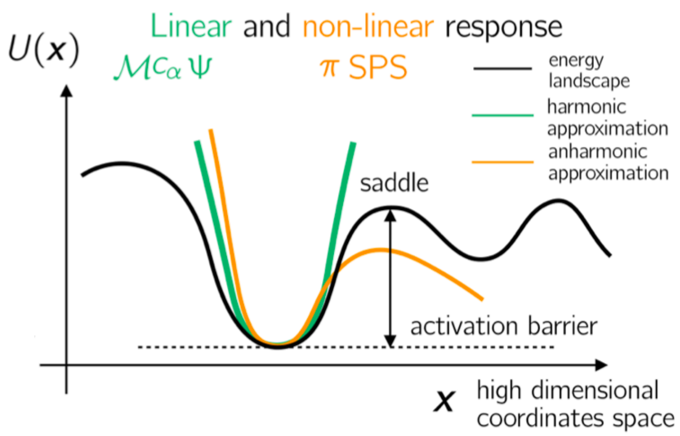
Numerical studies

$$4 \leq d \leq 13$$

e.g. P. Charbonneau et al., *J.Chem.Phys.* 156, 134502 (2022)

Exact analytical benchmark

'Mean-Field'



- **Some freedom in how to generalize the interactions & the dynamics with dimension:**

T. Maimbourg & J. Kurchan, *EPL* 114, 60002 (2016).
J. Kurchan, T. Maimbourg, F. Zamponi, *J. Stat. Mech.* 2016, 033210 (2016),

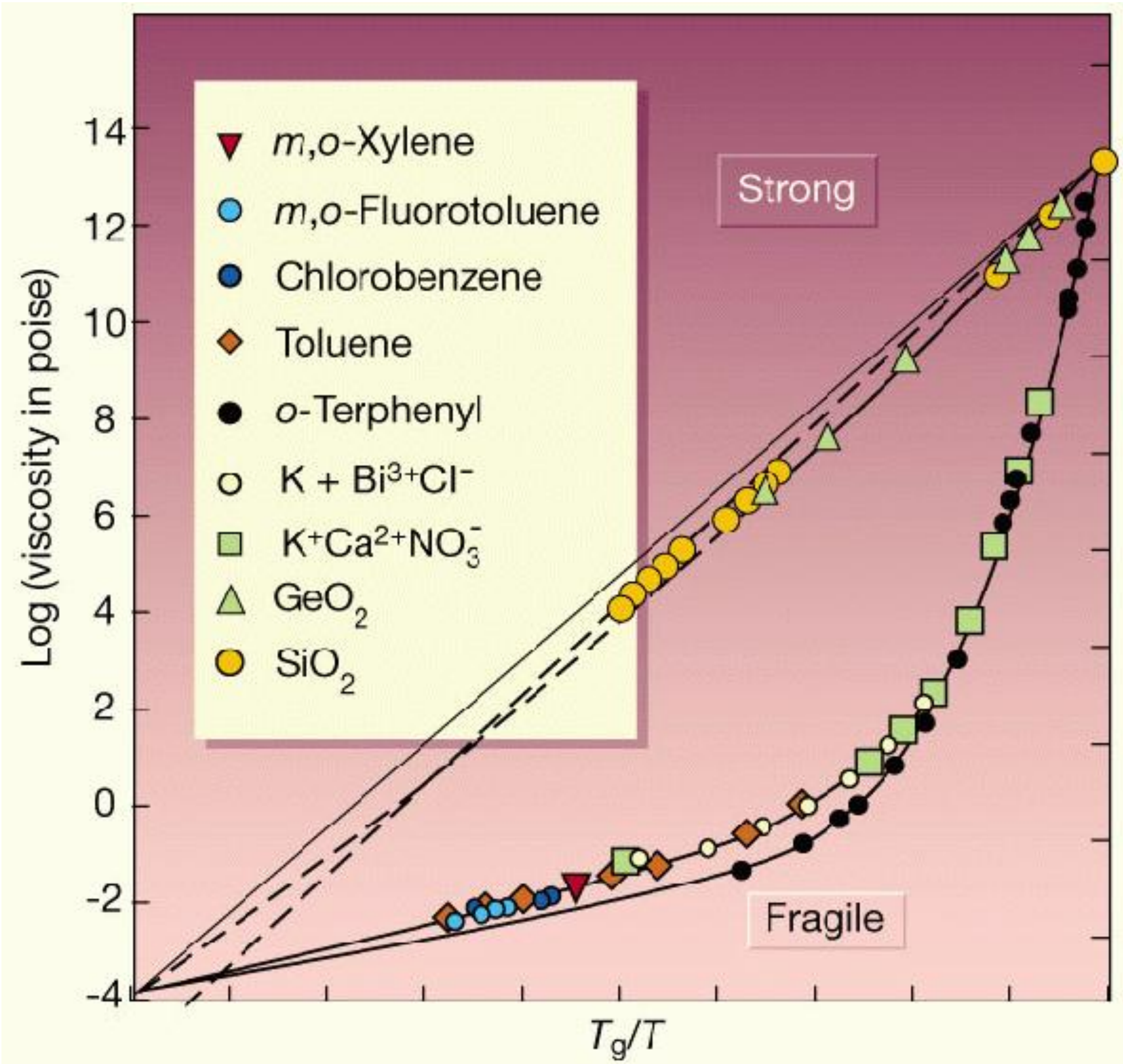
E.g. Soft harmonic spheres: $v(r) = \epsilon d^2 (r/\ell - 1)^2 \theta(\ell - r)$

E.g. Lennard-Jones: $v(r) = \epsilon \left[(\ell/r)^{4d} - (\ell/r)^{2d} \right])$

E.g. Hard spheres: $e^{-v(r)/T} = \theta(r - \ell)$

Infinite-dimensional 'mean-field' successes

Input 1: Glass transition



Dramatic increase in the viscosity with inverse temperature

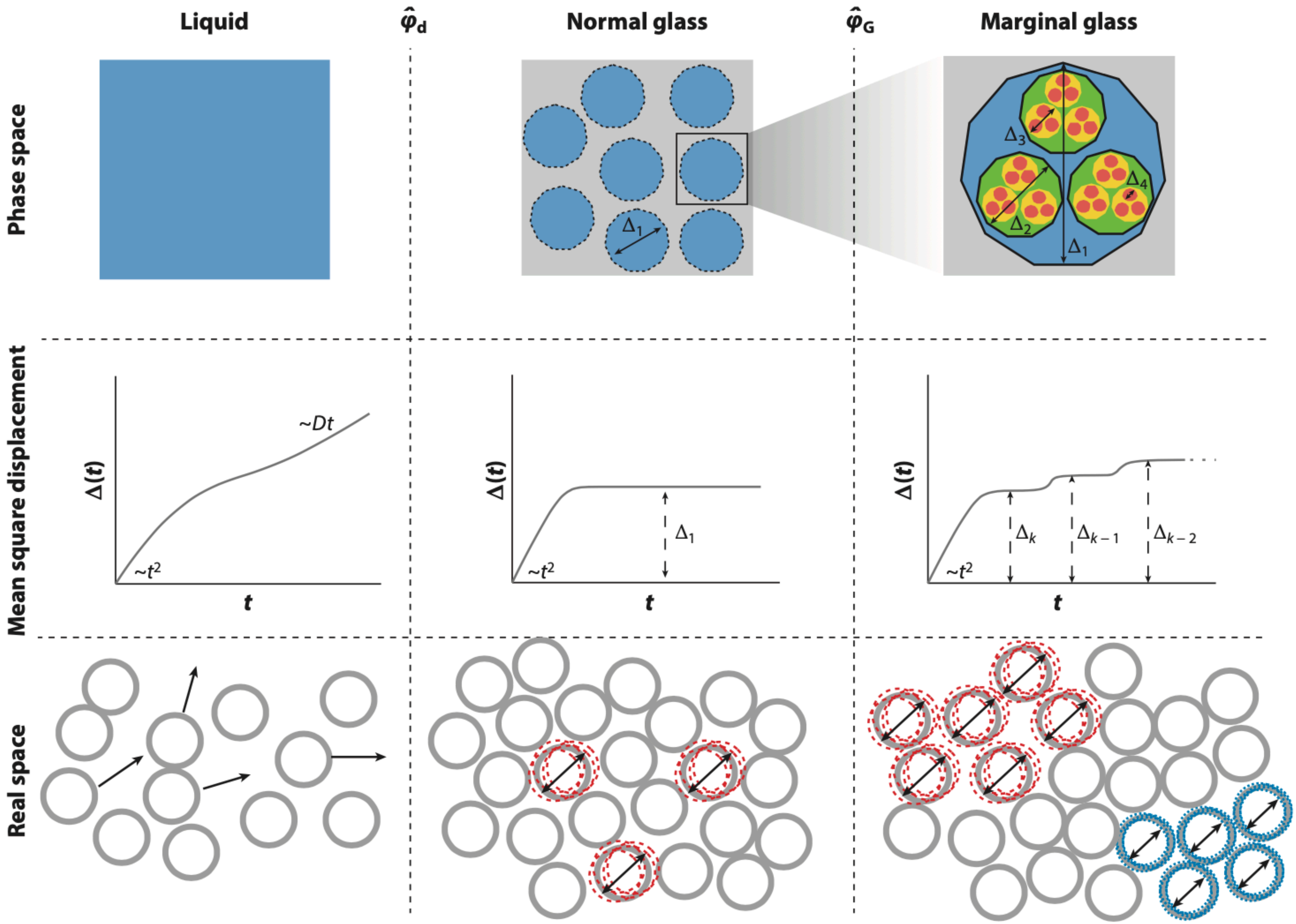
No significant change in the amorphous structure

P. G. Debenedetti & F. H. Stillinger, "Supercooled liquids and the glass transition", *Nature* 410, 259 (2001).

Review: F. Landes et al., "Glasses and aging: A Statistical Mechanics Perspective", *Encyclopedia of Complexity and Systems Science (2022)* / arXiv:2006.09725 [cond-mat.stat-mech].

Input 1: Glass transition — Fractal free energy landscapes in structural glasses

<u>*Review*</u>: P. Charbonneau et al., *Annual Review of Condensed Matter Physics* <u>8</u>, 265 (2017).



Input 2: Glass transition — Fractal free energy landscapes in structural glasses

P. Charbonneau et al., *Nature Communications* 5, 3725 (2014).

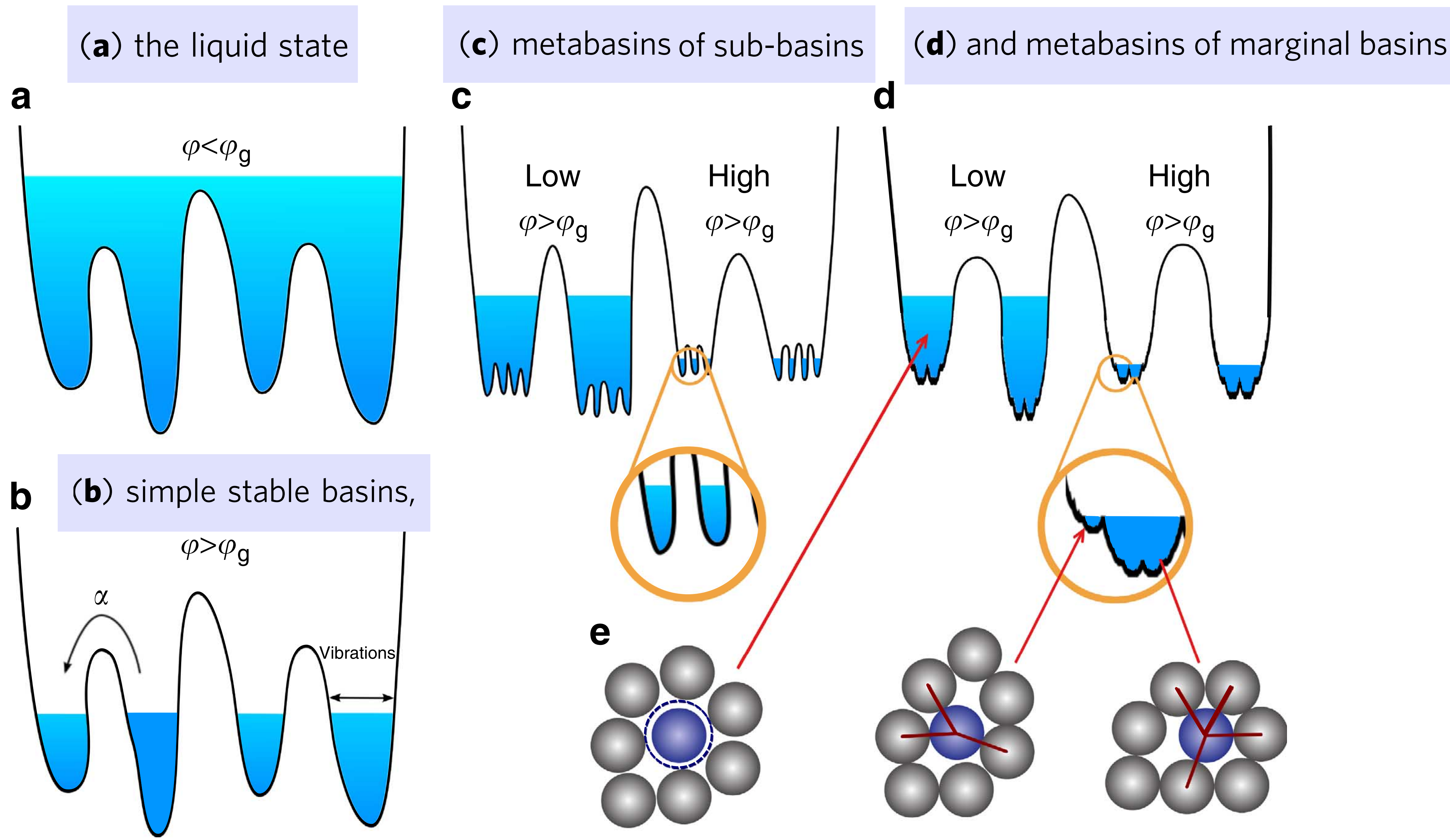
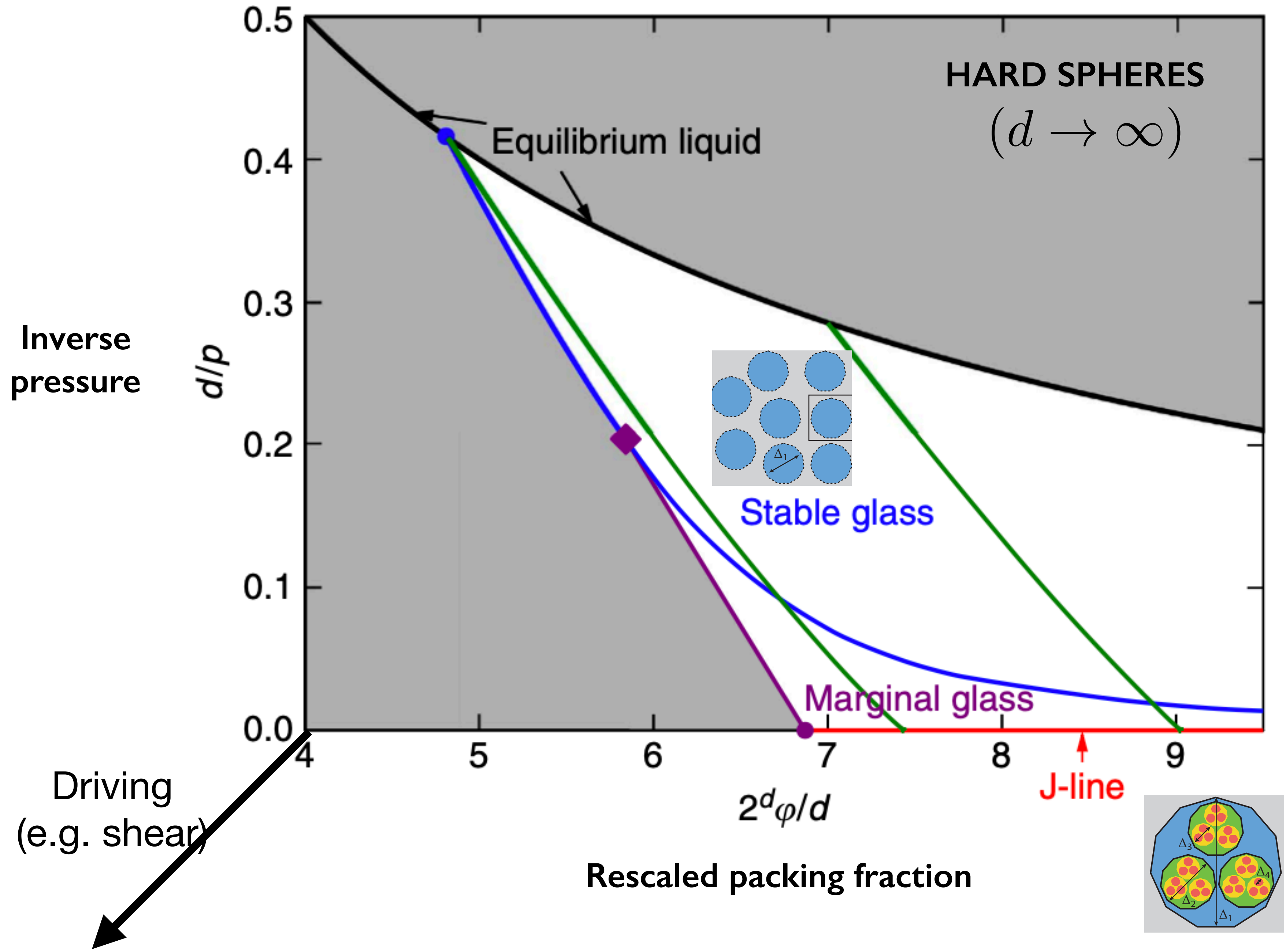
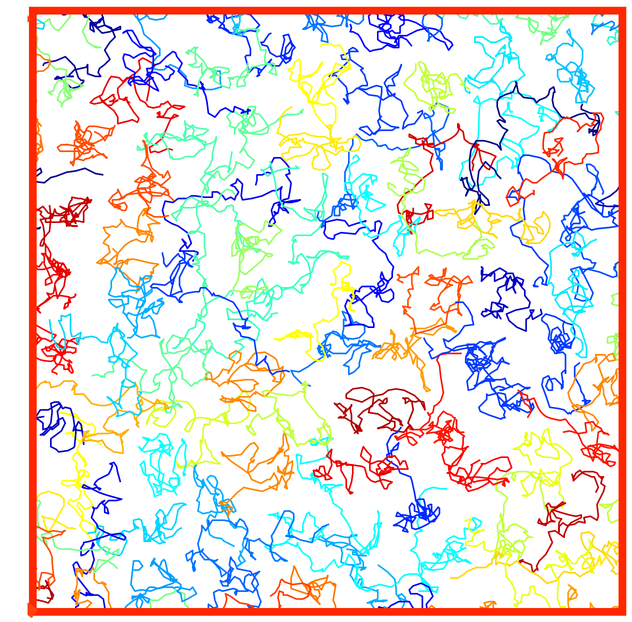


Figure 1 | Free energy landscape with simple basins, metabasins and fractal basins.

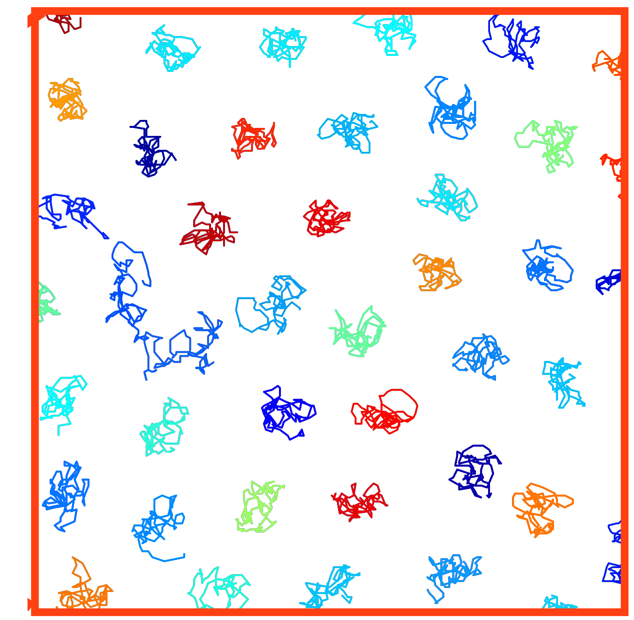
Input 1: Glass transition — Equilibrium phase diagramme of dense packings

P. Charbonneau et al., *Nature Communications* 5, 3725 (2014).





Diffusion vs caging



D. Bi *et al.*, *Phys. Rev. X* 6, 021011 (2016).

Equilibrium dynamics

T. Maimbourg, J. Kurchan & F. Zamponi, *Phys. Rev. Lett.* 116, 015902 & *J. Stat. Mech.* 2016, 033210 (2016).

G. Szamel, *Phys. Rev. Lett.* 119, 155502 (2017).

A. Manacorda, G. Schehr & F. Zamponi, *J. Chem. Phys.* 152, 164506 (2020).

Input 2: Quasistatic shear

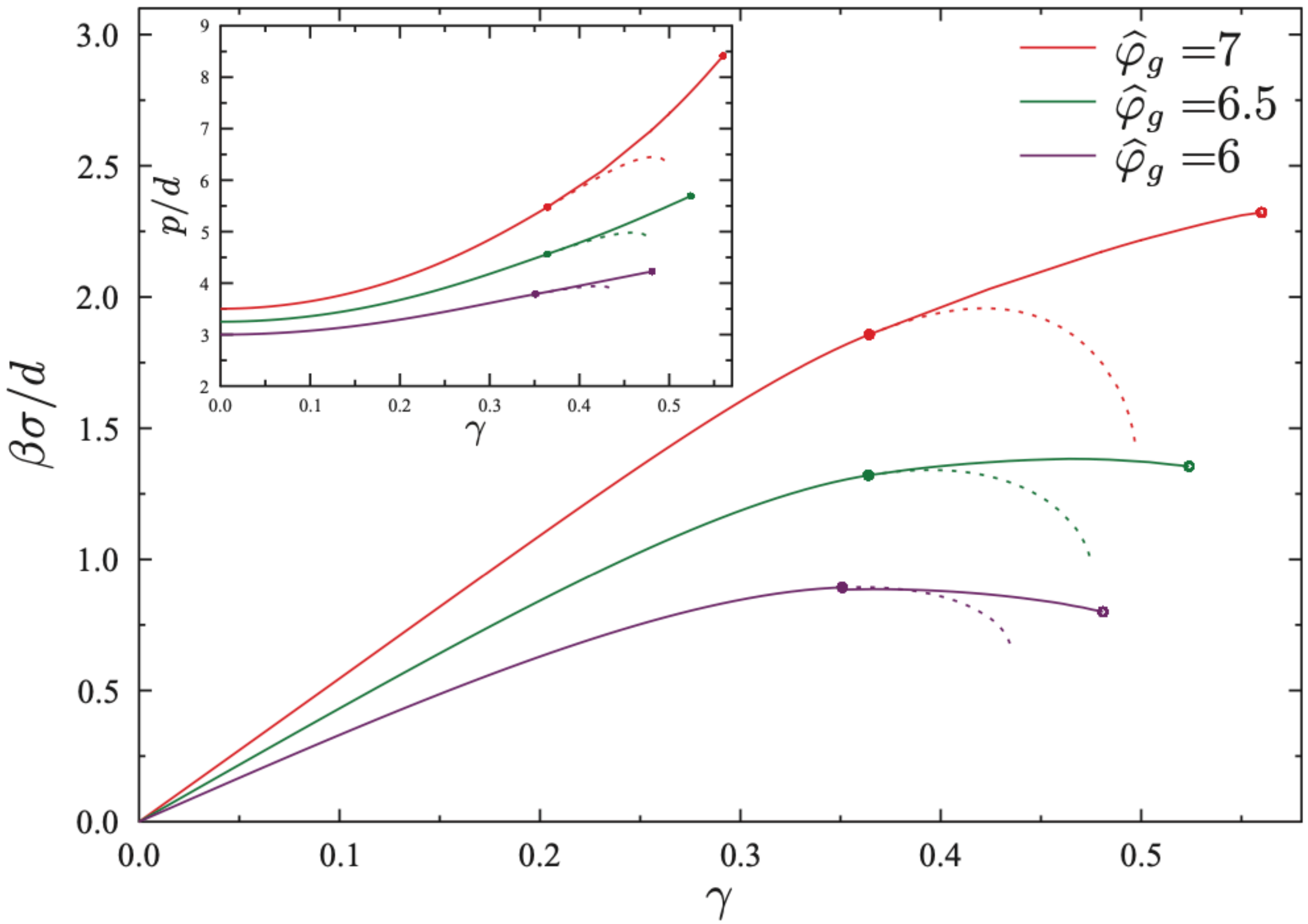
Equilibrium dynamics

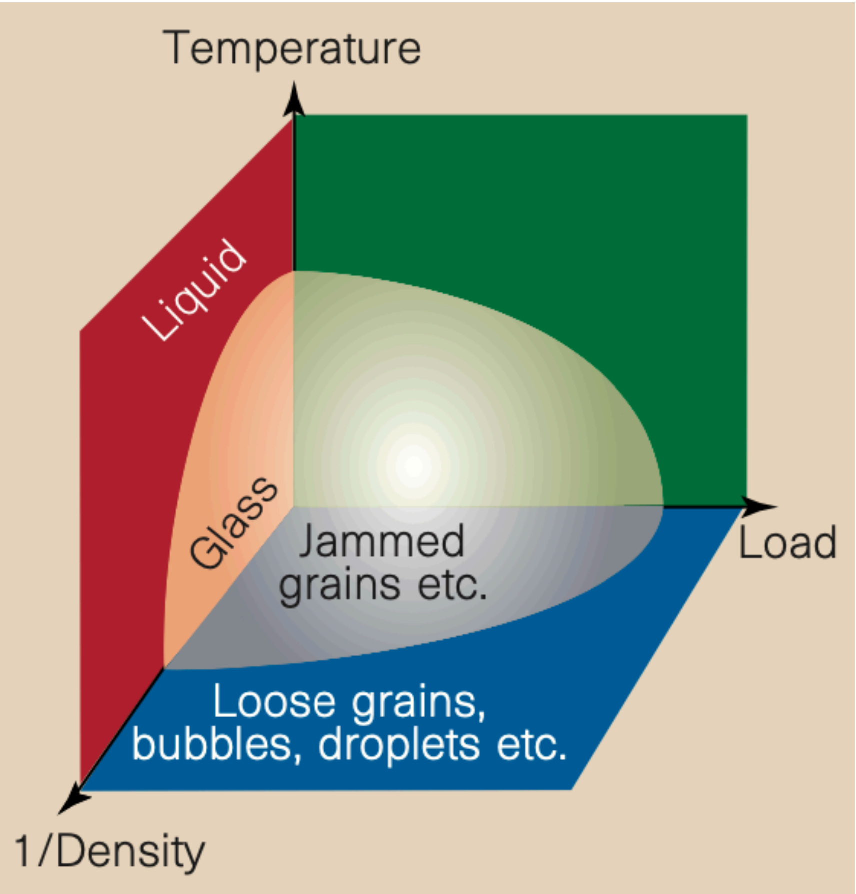
T. Maimbourg, J. Kurchan & F. Zamponi, *Phys. Rev. Lett.* 116, 015902 & *J. Stat. Mech.* 2016, 033210 (2016).

G. Szamel, *Phys. Rev. Lett.* 119, 155502 (2017).

A. Manacorda, G. Schehr & F. Zamponi, *J. Chem. Phys.* 152, 164506 (2020).

C. Liu, G. Biroli, D. R. Reichman, & G. Szamel, *Phys. Rev. E* 104, 054606 (2021).





A.J. Liu & S.R. Nagel, *Nature* 396, 6706 (1998).

RS solution: C. Rainone, P. Urbani, H. Yoshino, F. Zamponi, *Phys. Rev. Lett.* 114, 015701 (2015).

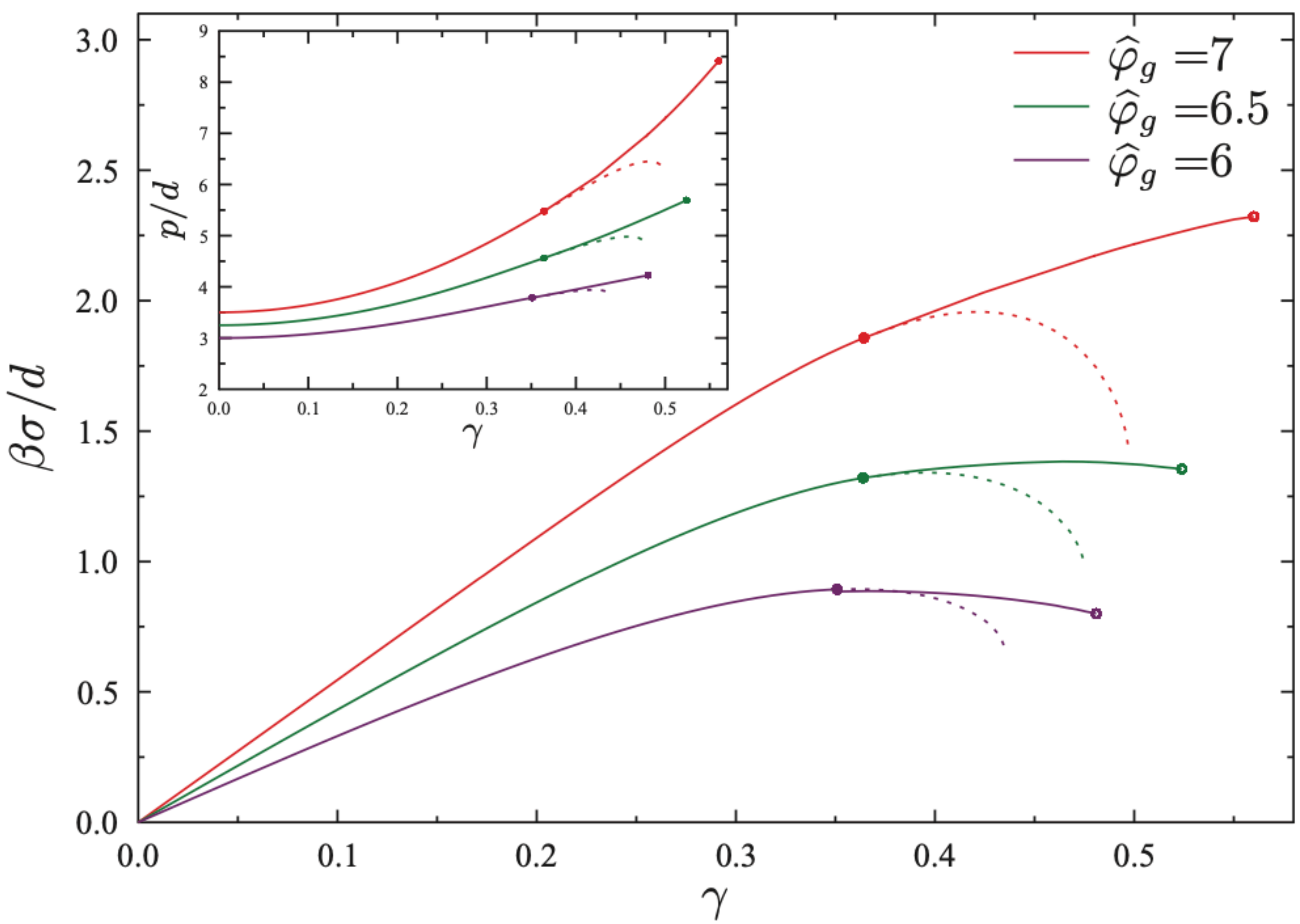
Full-RSB solution: C. Rainone & P. Urbani, *J. Stat. Mech.* 2016, 053302 (2016).

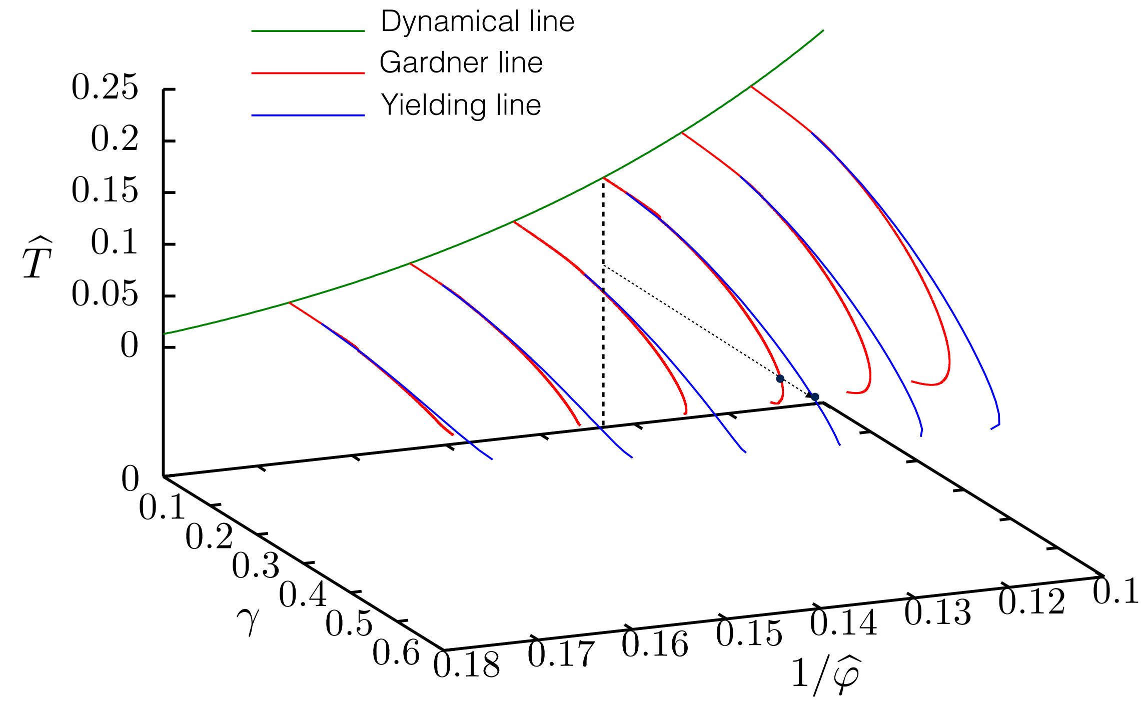
Liu-Nagel diagram in infinite dimension: G. Biroli & P. Urbani, *SciPost Phys.*. 4, 020 (2018).

Input 2: Quasistatic shear

Equilibrium dynamics

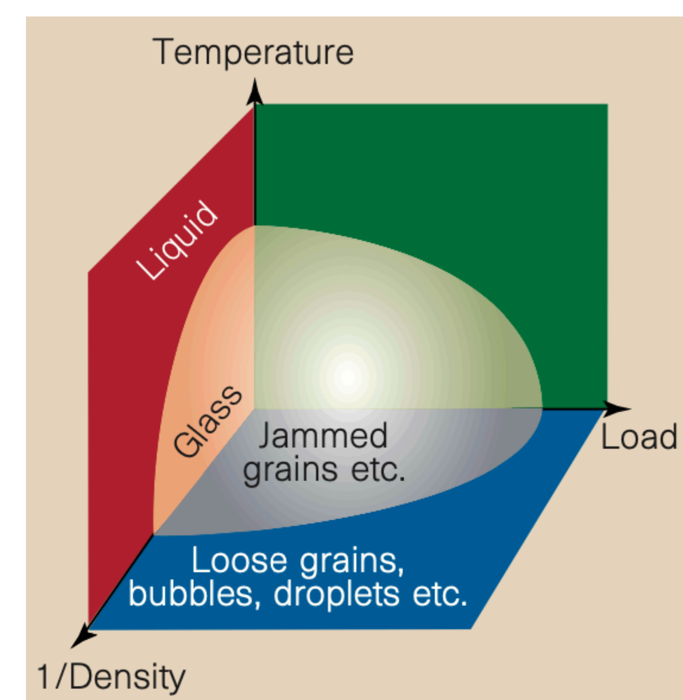
T. Maimbourg, J. Kurchan & F. Zamponi, *Phys. Rev. Lett.* 116, 015902 & *J. Stat. Mech.* 2016, 033210 (2016).
G. Szamel, *Phys. Rev. Lett.* 119, 155502 (2017).
A. Manacorda, G. Schehr & F. Zamponi, *J. Chem. Phys.* 152, 164506 (2020).
C. Liu, G. Biroli, D. R. Reichman, & G. Szamel, *Phys. Rev. E* 104, 054606 (2021).





RS solution: C. Rainone, P. Urbani, H. Yoshino, F. Zamponi, *Phys. Rev. Lett.* 114, 015701 (2015).
Full-RSB solution: C. Rainone & P. Urbani, *J. Stat. Mech.* 2016, 053302 (2016).

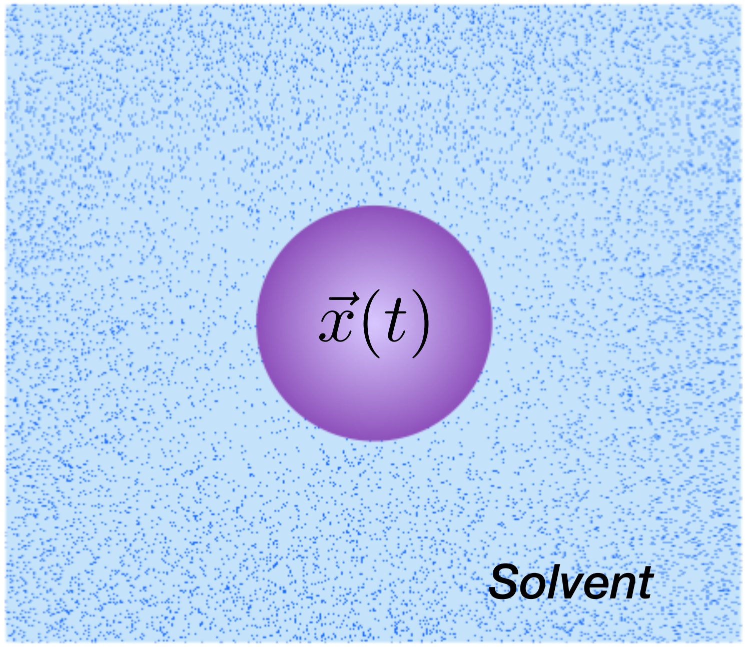
Liu-Nagel diagram in infinite dimension: G. Biroli & P. Urbani, *SciPost Phys.*. 4, 020 (2018).



Out-of-equilibrium
Dynamical Mean-Field Theory (DMFT)

DMFT — Starting point: Langevin dynamics

Brownian motion: thermal bath



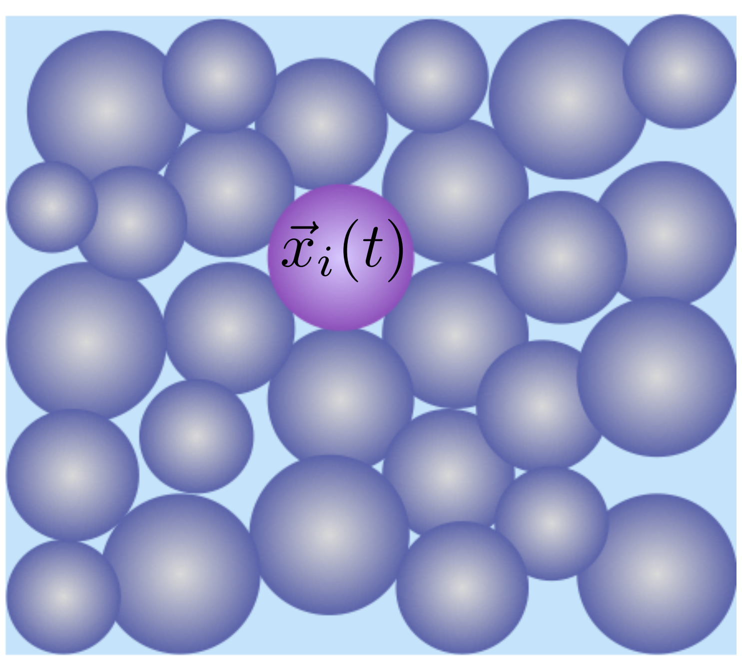
Acceleration	Friction	Noise	Driving

$$m\ddot{\vec{x}}(t) + \zeta\dot{\vec{x}}(t) = \vec{\xi}(t) + \vec{F}(t)$$

Equilibrium-like white Gaussian noise

$$\begin{cases} \langle \vec{\xi}(t) \rangle = 0 \\ \langle \xi_\mu(t)\, \xi_\nu(t') \rangle = 2\zeta T\, \delta(t-t') \end{cases}$$

Dense assemblies of particles
of similar sizes



Acceleration	Interactions	Driving

$$m_i\ddot{\vec{x}}_i(t) = \sum_{j(\neq i)} \nabla v_{ij}(\vec{x}_i(t) - \vec{x}_j(t)) + \vec{F}_i(t)$$

$$m_i\ddot{\vec{x}}_i(t) + \underbrace{\boxed{\text{Effective "friction"}} = \boxed{\text{Effective "noise"}} + \boxed{\text{Local "disorder"}}}_{\text{Self-generated by the interactions}} + \boxed{\text{Driving}}$$

DMFT — Limit of infinite spatial dimension

Review book for $d \to \infty$: G. Parisi, P. Urbani & F. Zamponi, "Theory of simple glasses", Cambridge University Press (2020)

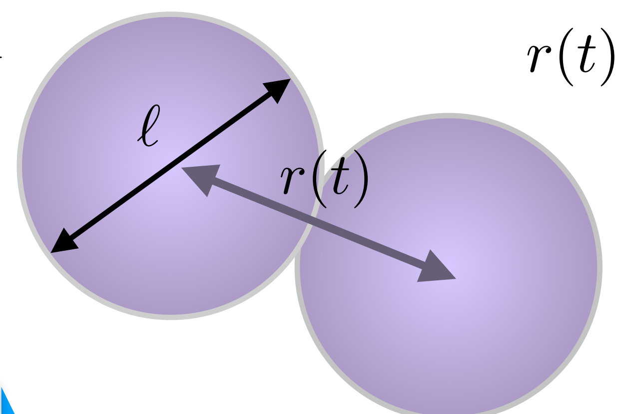
Many-body Langevin

Spatial fluctuations are suppressed with $d \to \infty$

Effective scalar Langevin

$\Rightarrow$ ***mean-field description exact*** in this limit

$$\{\mathbf{r}_{ij}(t) = \mathbf{x}_i(t) - \mathbf{x}_j(t)\}$$



$r(t) = \ell\left(1 + \frac{h(t)}{d}\right)$ with the gap $h(t) \sim \mathcal{O}(d^0)$

Gap fluctuations: $\mathcal{P}(h(t))$

Energy $\sim \langle \bar{v}(h(t)) \rangle_h$

Pressure $\sim \langle \bar{v}'(h(t)) \rangle_h$

E.g. Soft harmonic spheres:

$$v(r) = \epsilon d^2 (r/\ell - 1)^2 \theta(\ell - r)$$

$$\bar{v}(h) = \epsilon h^2 \theta(-h)$$

$$\lim_{d \to \infty} v(r) = \bar{v}(h)$$

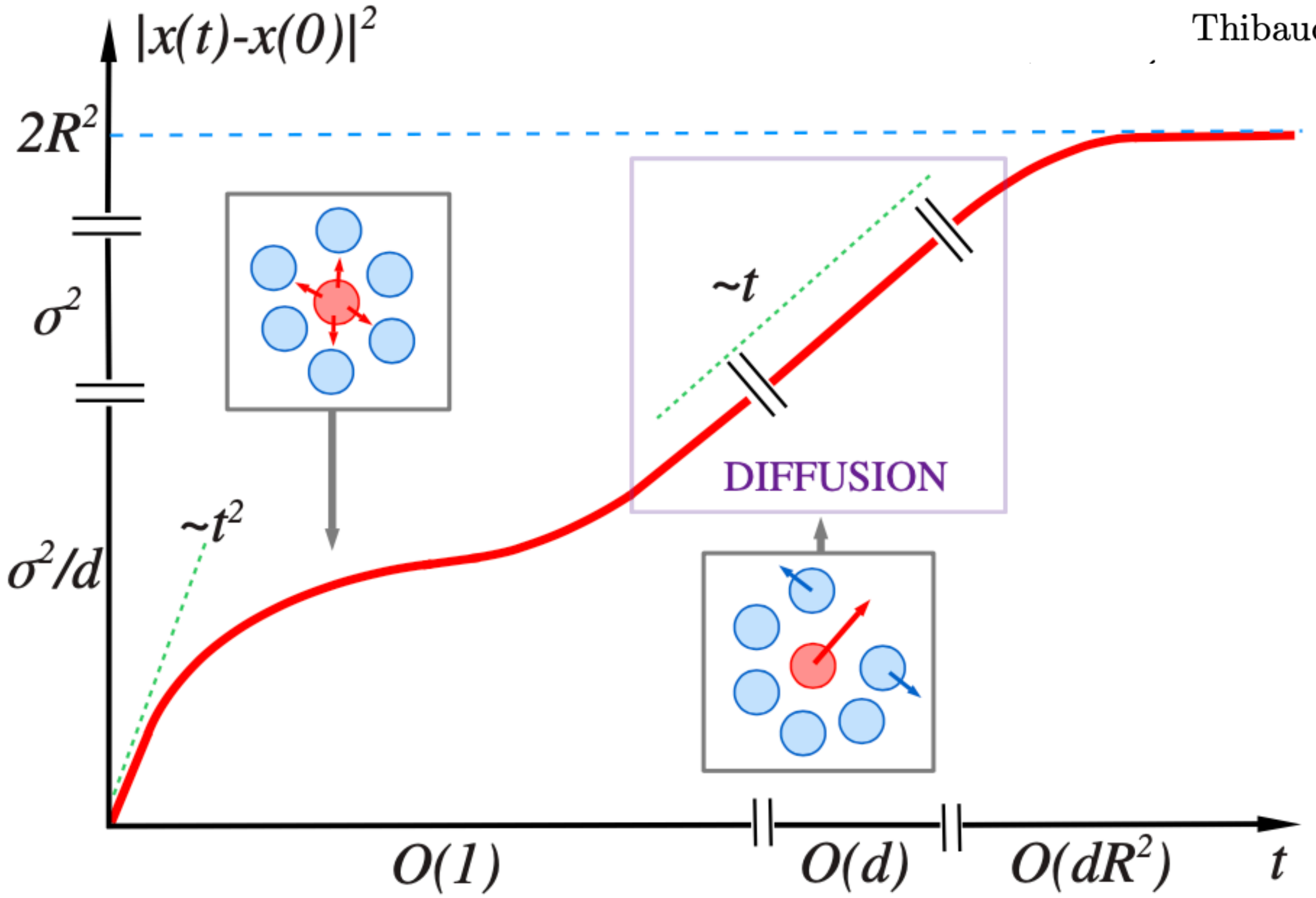
T. Maimbourg & J. Kurchan, *EPL* 114, 60002 (2016).

Initial condition $\mathcal{P}[\{\mathbf{r}_{ij}(0)\}] \Rightarrow \mathcal{P}(h_0)$

Sampling trajectories: $h(t)|h_0$

Rescaled MSD: $\Delta(t,t') = \dfrac{d^2}{\ell^2} D(t,t')$

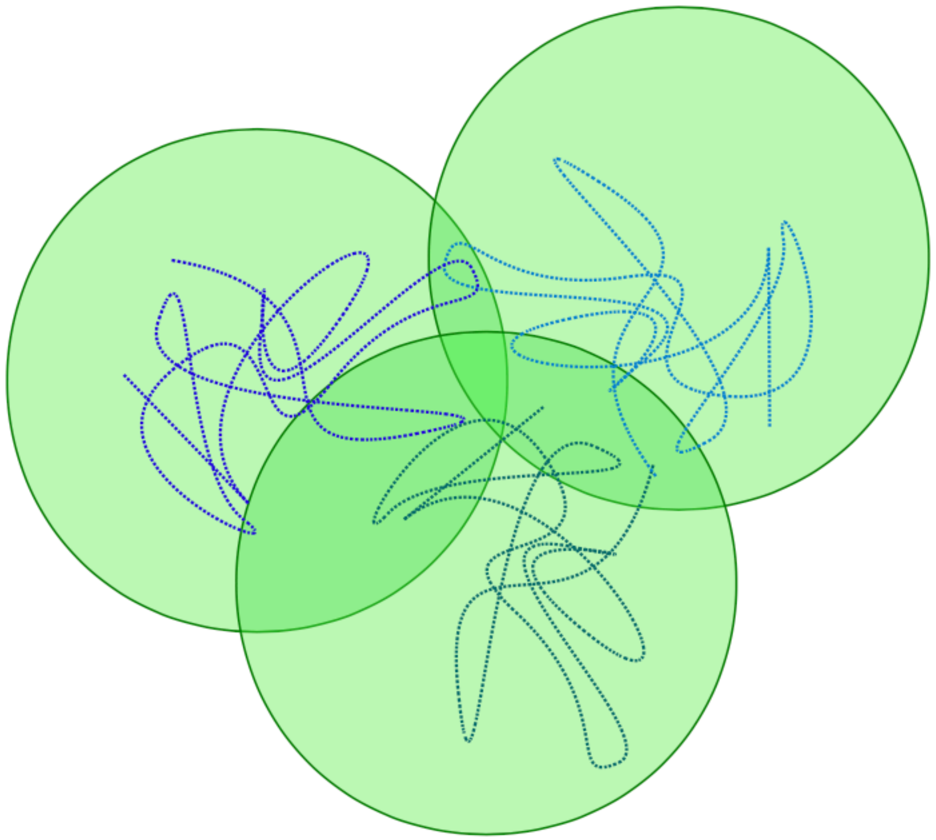
DMFT — Limit of infinite spatial dimension



Supplementary information for
"Solution of the dynamics of liquids in the large-dimensional limit"

Thibaud Maimbourg,[1] Jorge Kurchan,[2] and Francesco Zamponi[1]

J. Stat. Mech. 2016, 033210 (2016).



E.g. Soft harmonic spheres:

$$v(r) = \epsilon d^2 (r/\ell - 1)^2 \theta(\ell - r)$$

$$\bar{v}(h) = \epsilon h^2 \theta(-h)$$

$$\lim_{d\to\infty} v(r) = \bar{v}(h)$$

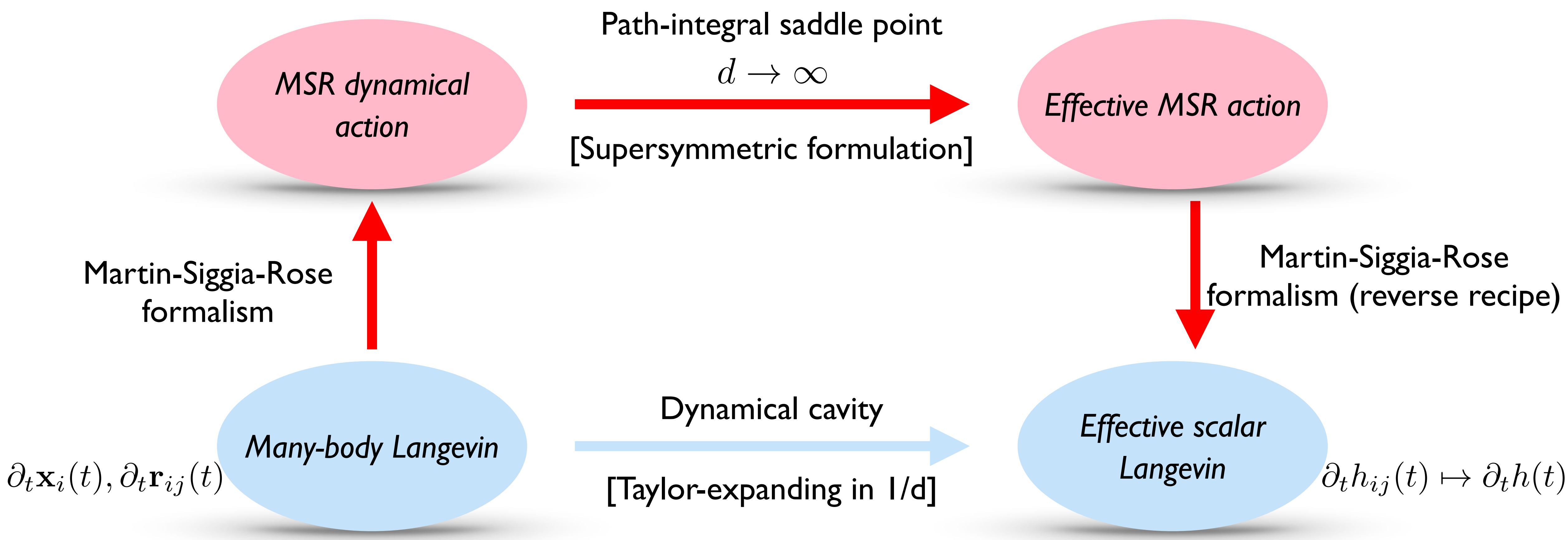
Initial condition $\mathcal{P}[\{\mathbf{r}_{ij}(0)\}] \Rightarrow \mathcal{P}(h_0)$

Sampling trajectories: $h(t)|h_0$

Rescaled MSD: $\Delta(t,t') = \dfrac{d^2}{\ell^2} D(t,t')$

T. Maimbourg & J. Kurchan, *EPL* 114, 60002 (2016).

DMFT — Two complementary derivations



Equilibrium dynamics

T. Maimbourg, J. Kurchan & F. Zamponi, *Phys. Rev. Lett.* 116, 015902 & *J. Stat. Mech.* 2016, 033210 (2016).
G. Szamel, *Phys. Rev. Lett.* 119, 155502 (2017).
A. Manacorda, G. Schehr & F. Zamponi, *J. Chem. Phys.* 152, 164506 (2020).
C. Liu, G. Biroli, D. R. Reichman, & G. Szamel, *Phys. Rev. E* 104, 054606 (2021).

$$\boxed{\langle \xi_{i\mu}(t)\xi_{j\nu}(t')\rangle = 2\zeta T\, \delta_{ij}\delta_{\mu\nu}\delta(t-t')}$$

Out-of-equilibrium

Isotropic case: E. Agoritsas, T. Maimbourg & F. Zamponi, ***J. Phys. A* 52, 144002 (2019).**
Under global shear: E. Agoritsas, T. Maimbourg & F. Zamponi, ***J. Phys. A* 52, 334001 (2019).**
Under local forcing: E. Agoritsas, ***J. Stat. Mech.* 2021, 033501(2021).**

Generic approach!

Continuous random perceptron (similar derivations)

E. Agoritsas, G. Biroli, P. Urbani & F. Zamponi, *J. Phys. A* 51, 085002 (2018).

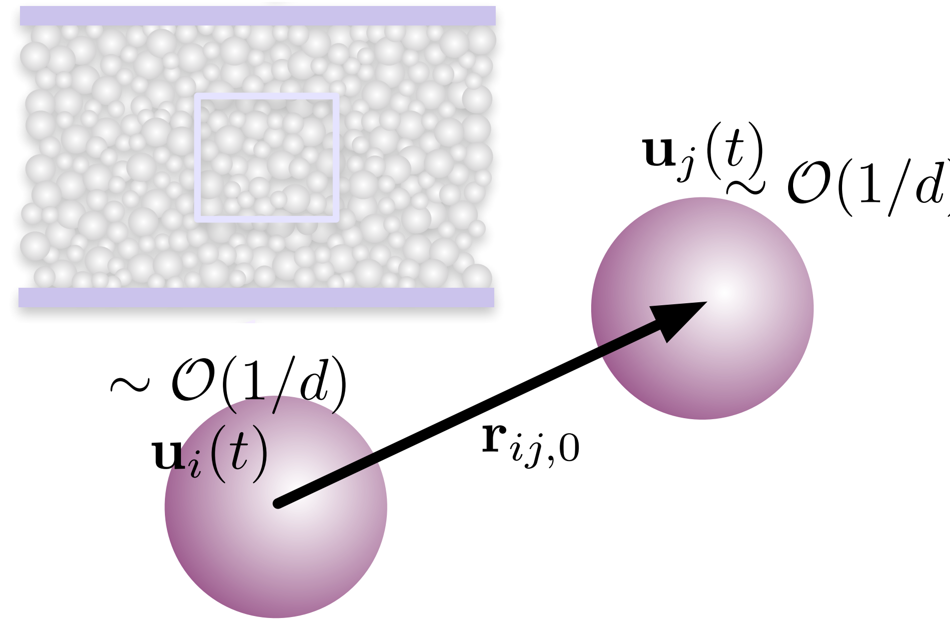
- A few key physical assumptions specific to high dimensions:

  - Particles stay 'close' to their initial position:

$$\mathbf{u}_i(t) = \mathbf{x}_i(t) - \mathbf{x}_i(0) \sim \mathcal{O}(1/d)$$
$$\mathbf{w}_{ij}(t) = \mathbf{u}_i(t) - \mathbf{u}_j(t) \sim \mathcal{O}(1/d)$$

  - Each particle has numerous uncorrelated neighbours.

  - Statistical isotropy of the system (in absence of shear).



- Interactions treated perturbatively for 'small' displacements:

$$\zeta \dot{\mathbf{u}}_i(t) = \boxed{\text{Interaction term}} + \xi_i(t)$$

DMFT — Dynamical cavity: Effective stochastic processes WITHOUT SHEAR

- A few key physical assumptions specific to high dimensions:
  - Particles stay 'close' to their initial position:

$$\mathbf{u}_i(t) = \mathbf{x}_i(t) - \mathbf{x}_i(0) \sim \mathcal{O}(1/d)$$
$$\mathbf{w}_{ij}(t) = \mathbf{u}_i(t) - \mathbf{u}_j(t) \sim \mathcal{O}(1/d)$$

  - Each particle has numerous uncorrelated neighbours.
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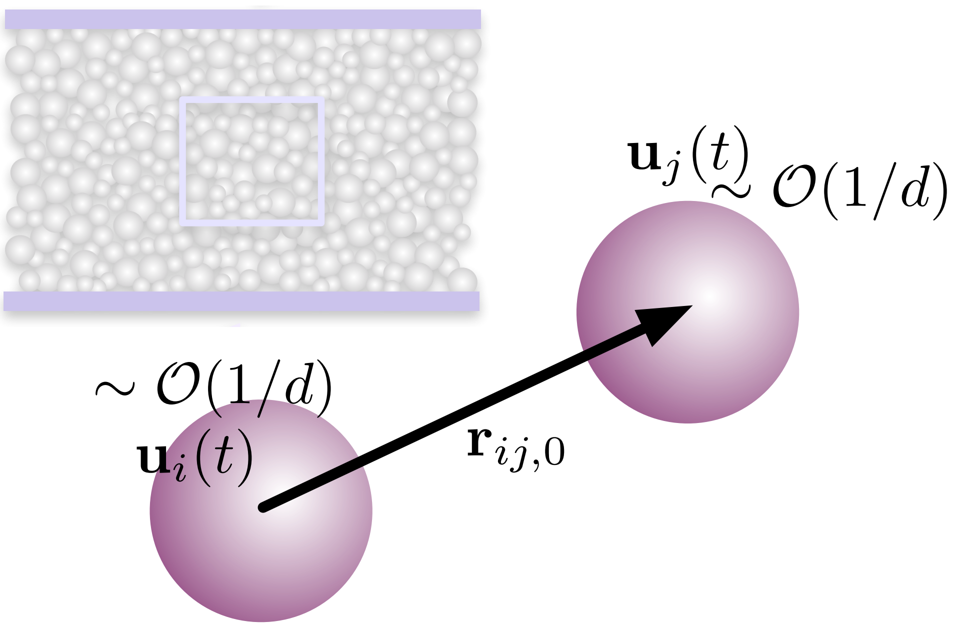


- Interactions treated perturbatively for 'small' displacements:

$$\zeta \dot{\mathbf{u}}_i(t) = \boxed{-k(t)\,\mathbf{u}_i(t) + \int_0^t ds\, M_R(t,s)\,\mathbf{u}_i(s) + \tilde{\mathbf{F}}_i^f(t)} + \xi_i(t)$$

with $\left\langle \tilde{\mathbf{F}}^f_{i\mu}(t) \right\rangle = 0\,, \quad \left\langle \tilde{\mathbf{F}}^f_{i\mu}(t) \tilde{\mathbf{F}}^f_{j\nu}(s) \right\rangle = \delta_{ij}\, M_C^{\mu\nu}(t,s)\,.$

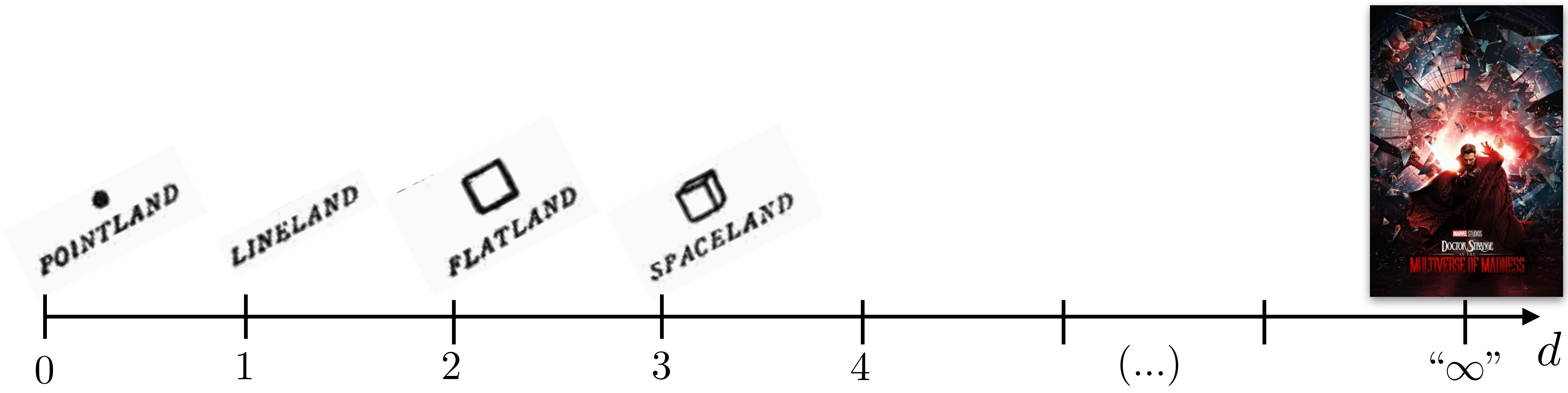
Mean-field kernels

$$k(t) \sim \langle \nabla^2 v \rangle\,,$$
$$M_C(t,s) \sim \langle \nabla v \cdot \nabla v \rangle$$
$$M_R(t,s) \sim \delta \langle \nabla v \rangle / \delta P$$

Same procedure for the interparticle distance $\mathbf{w}_{ij}(t) = \mathbf{u}_i(t) - \mathbf{u}_j(t)$

$\Rightarrow$ Same for the gap $h_{ij}(t) \sim \mathcal{O}(d^0)$

Take-home messages



- **Structural glasses ↔ landscape picture & its statistical features**

  ⇒ Direct translation to specific properties, therefore **tunable by well-devised preparation/driving protocols**

- **High-dimensional landscape** with extrema/saddle-points/flat directions ⇒ *Lineland* intuition might be misleading

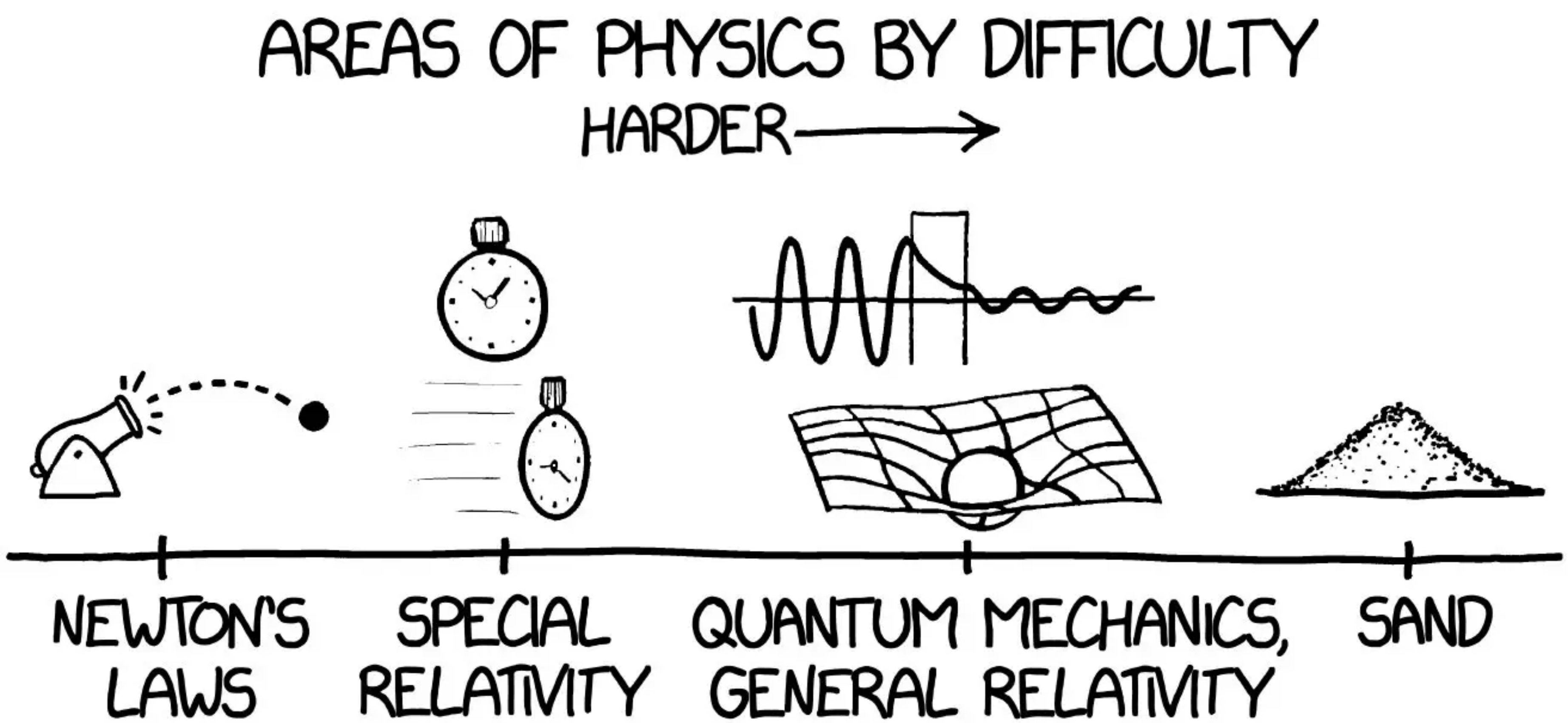
- Infinite-dim. limit provides **exact analytical benchmarks**, useful to capture low-dim. physics related to metastable states

- Great 'mean-field' successes: glass transition, mechanical heterogeneities, jamming criticality (based on <u>statics</u>)

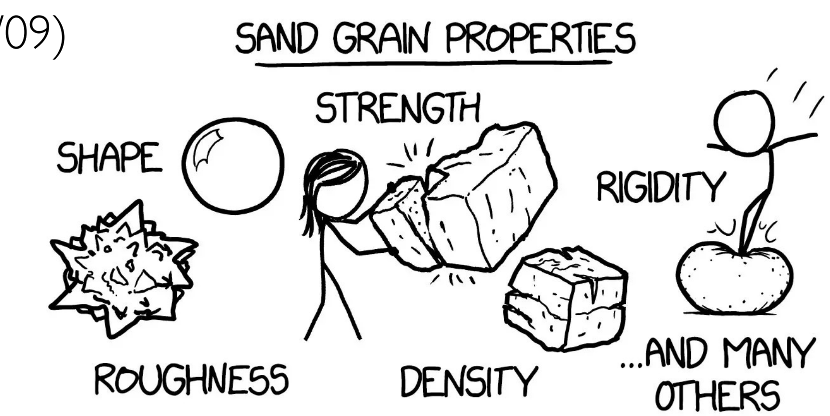
- Next step: <u>dynamics</u> ⇒ **Out-of-equilibrium Dynamical Mean-Field Theory (DMFT)**, e.g. link to dense active matter

- Questions generic for driven disordered/complex systems, e.g. in **machine-learning training algorithms**

"More is different"



XKCD for the New York Times, "What Makes Sand Soft?" (2020/11/09)



I do not mean to give the impression that all is settled. For instance, I think there are still fascinating questions of principle about glasses and other amorphous phases, which may reveal even more complex types of behavior.

It seems to me that the next stage is to consider the system which is regular but contains information. *[⇒ Memory formation in matter]*

P.W. Anderson, *Science* (1972) "**More is different**"