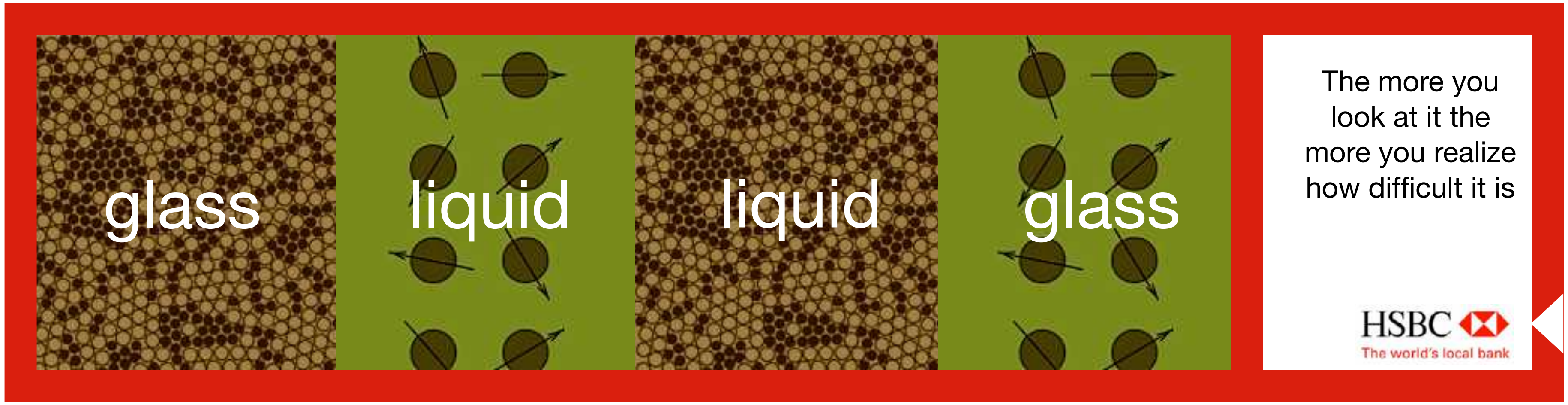
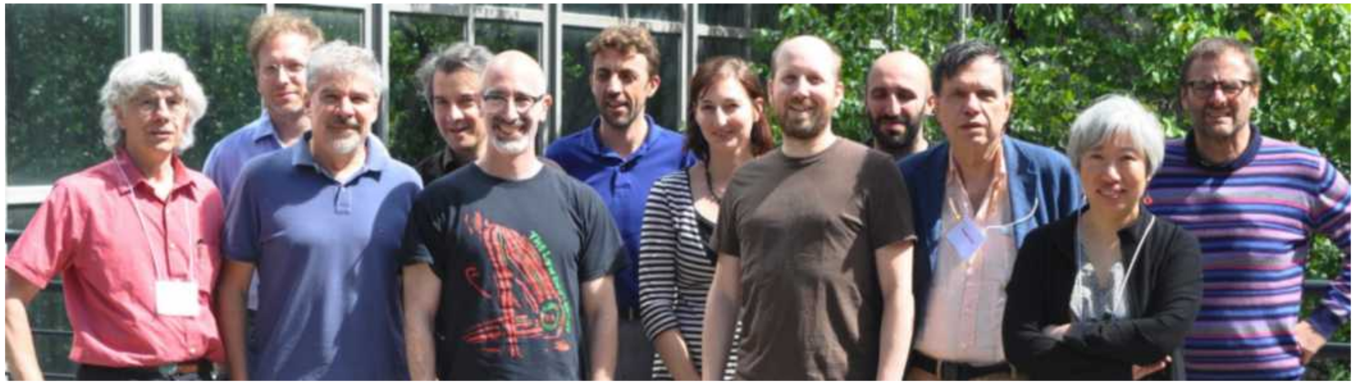
Cracking the Glass Problem: Dynamics



Patrick Charbonneau and Grzegorz Szamel

Collaborative Glassiness

Simons Collaboration on Cracking the Glass Problem @ hardsphere.glass



Yi Hu (Duke)
Yuliang Jin (now CAS-Physics)
Joyjit Kundu (Duke)
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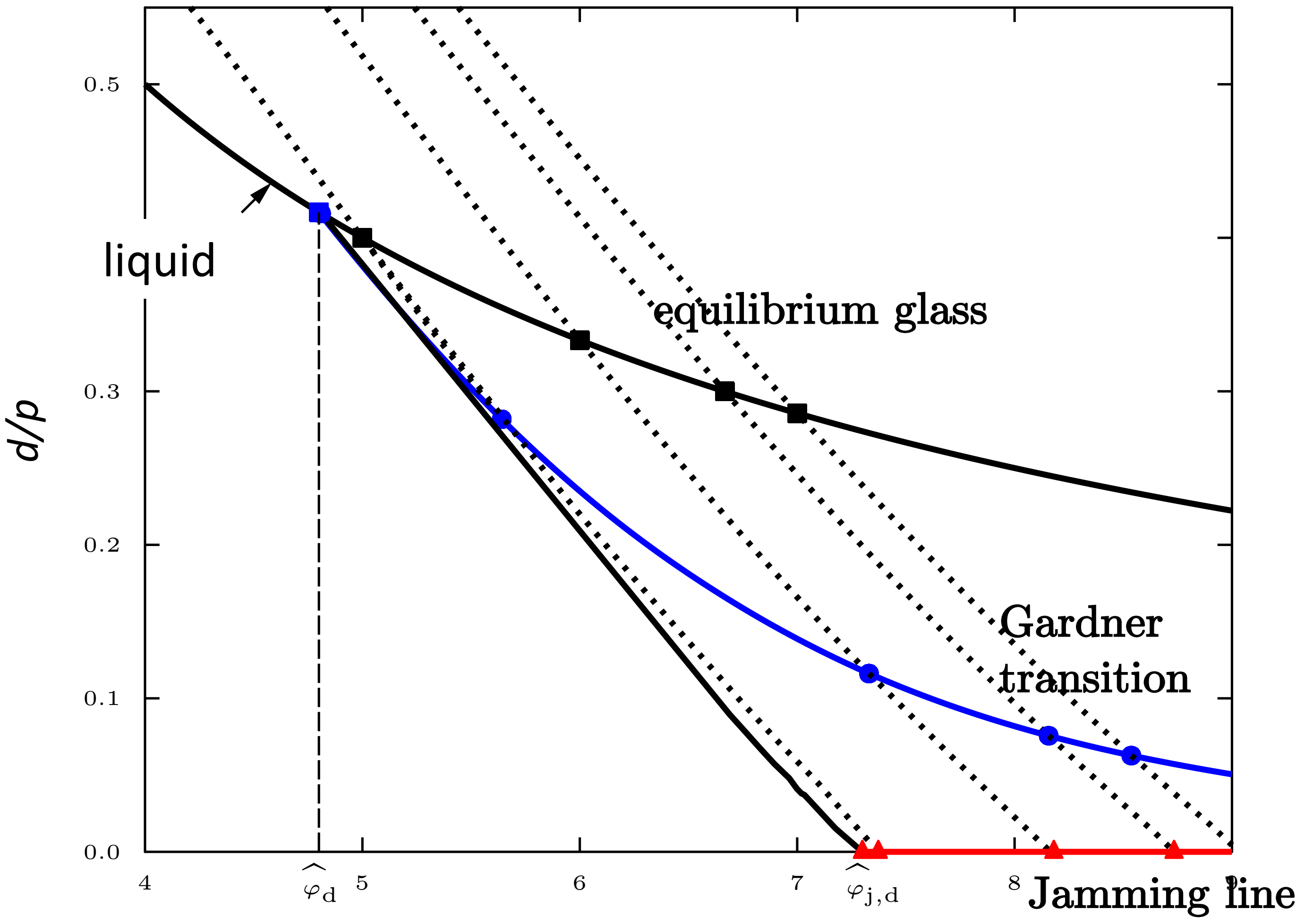
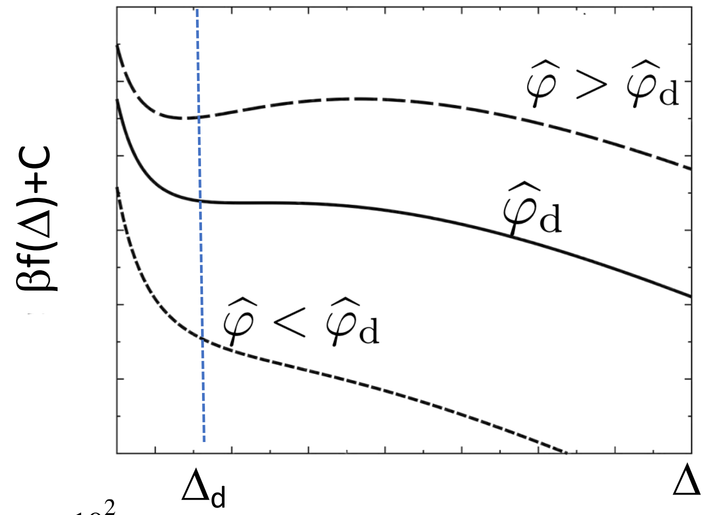
Benoit Charbonneau (Waterloo)
Harukuni Ikeda (ENS-Paris)

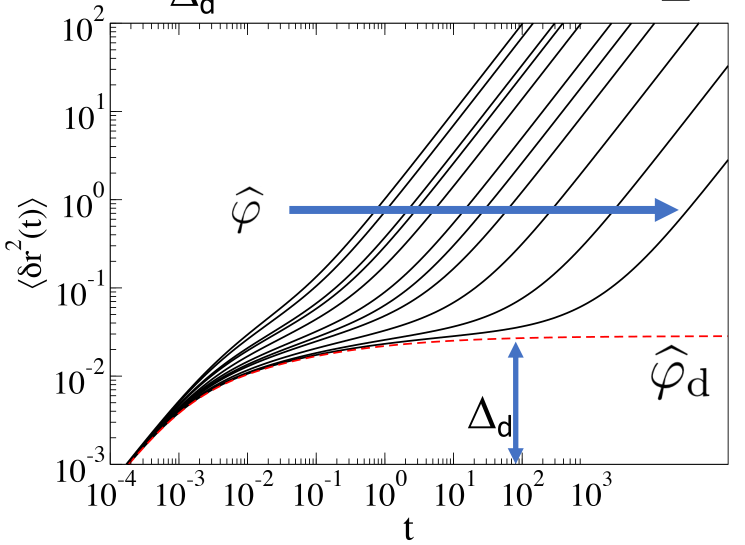
SIMONS FOUNDATION

Exact HS PD in $d \to \infty$



$$\widehat{\varphi} = 2^d \varphi / d$$



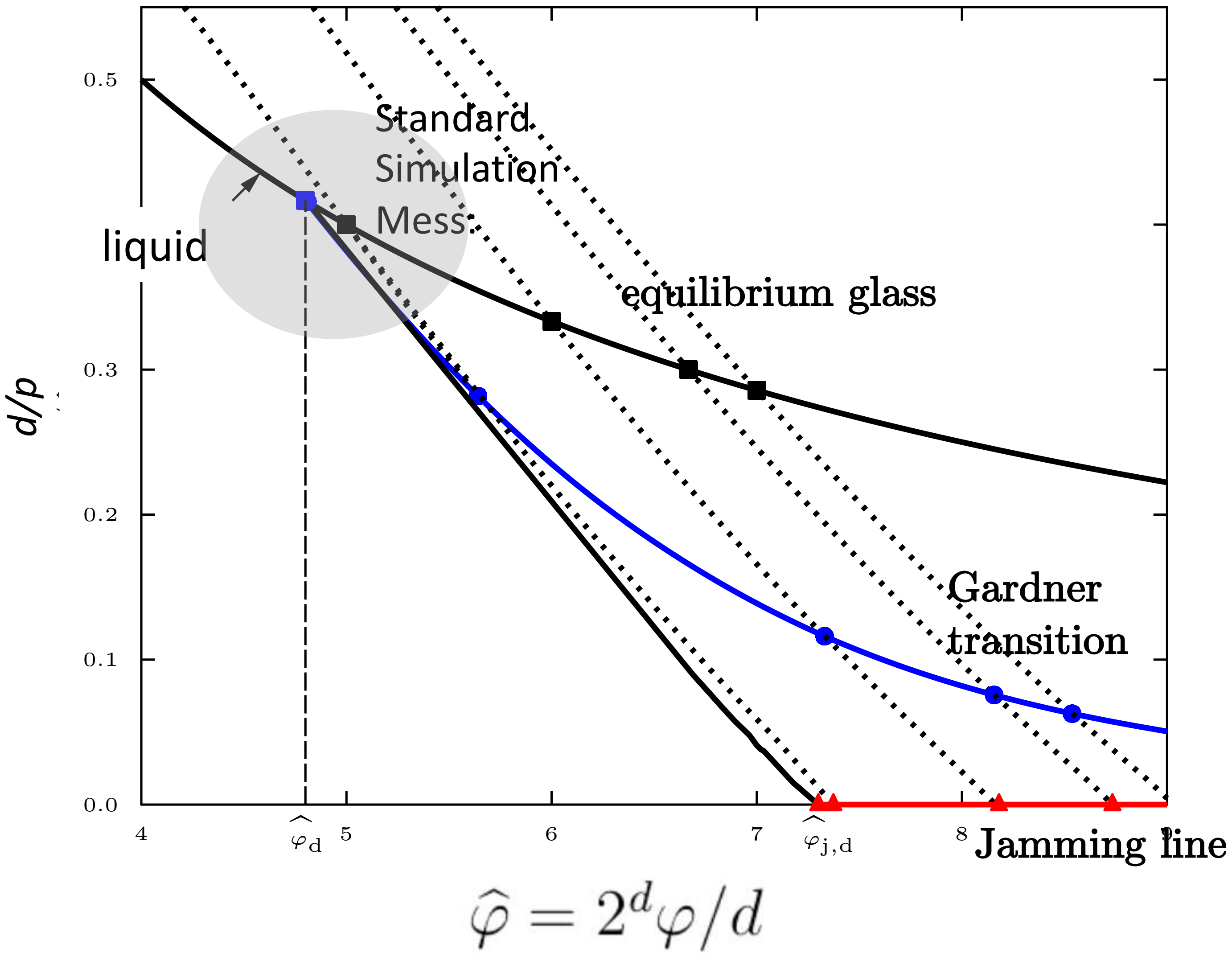


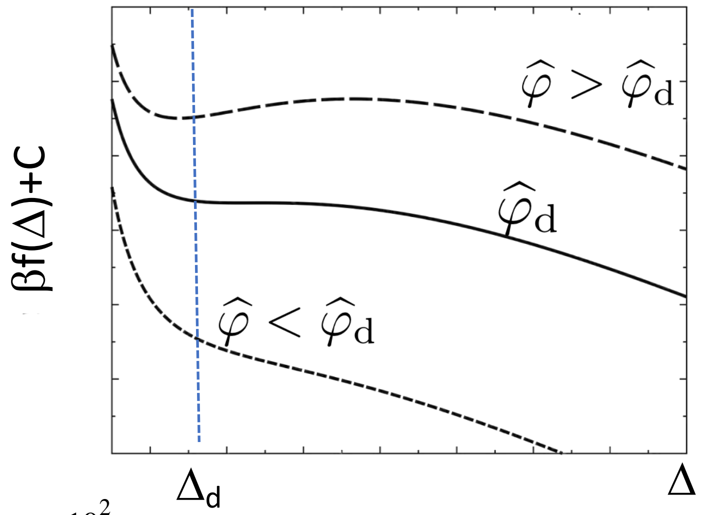
Localization by caging

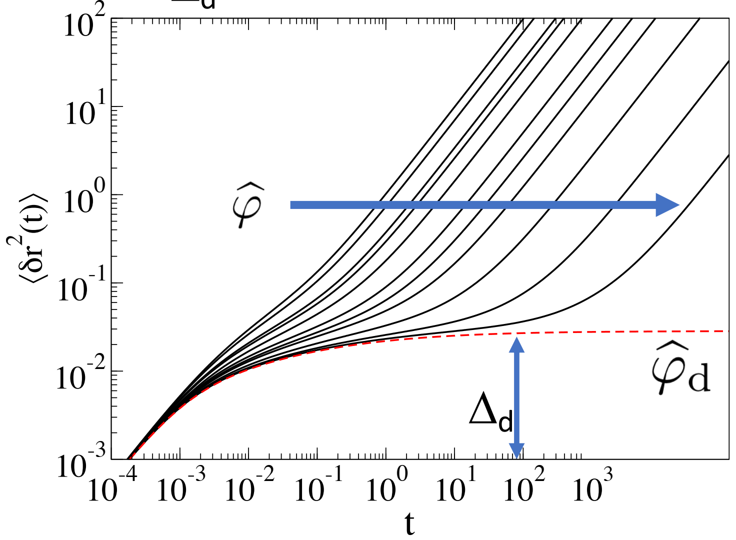
First formulation and analytical attempt: Kirkpatrick and Wolynes, 1987; PZ, KPZ, KPUZ, CKPUZ, RU, RUYZ (2005-2016)

Exact Realization of the RFOT scenario without activation from Kirkpatrick, Thirumalai and Wolynes (1989)

Dynamics is Only Simple in $d \to \infty$







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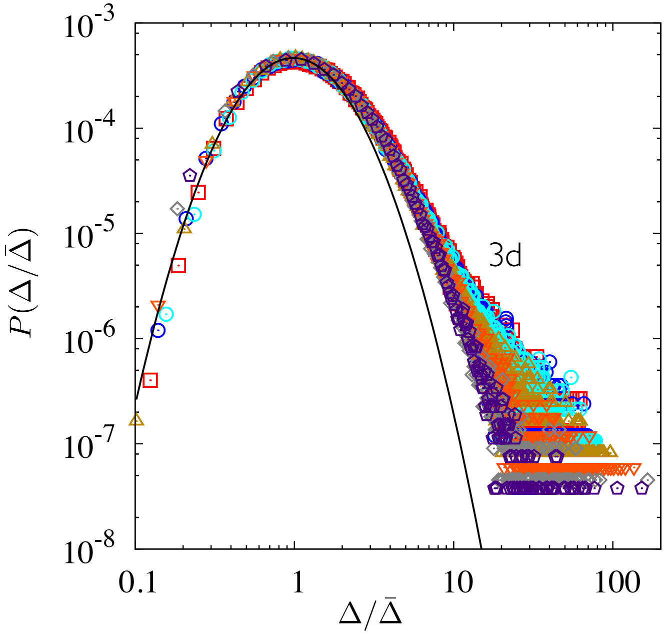
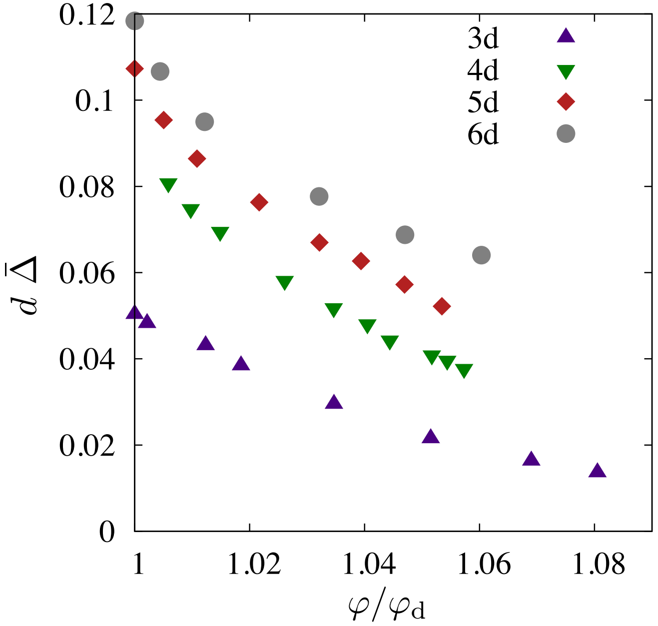
Challenges of Relating $d\to\infty$ and "Real" Glasses around the dynamical transition.

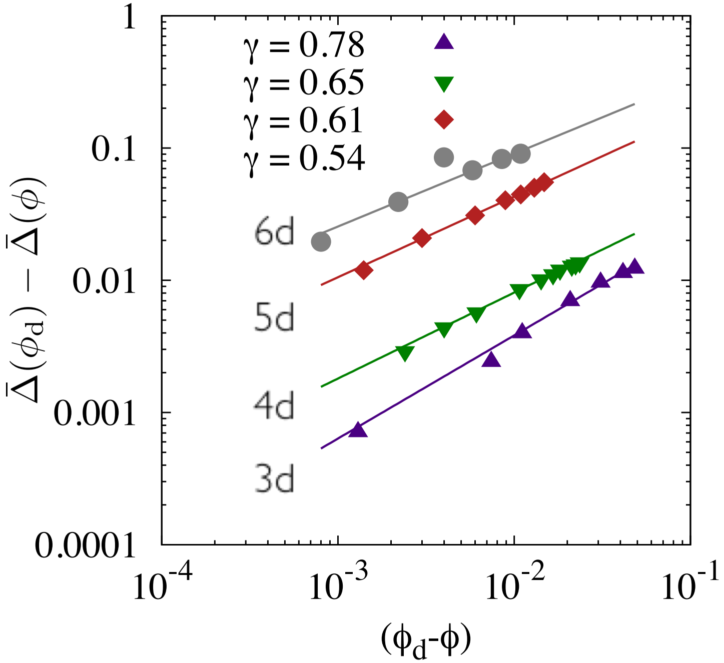
System	Glass Nucl.	Comp. Struct.	$d < d_{\mathrm{u}}$
HS in $d = 2$ and 3	✓	✓	✓
HS in $4 \leq d < 6$	small	small	✓
HS in $6 \leq d < 8$	small	small	✓
HS in $d \geq 8$	small	small	✗
HS/MK in $d \to \infty$	✗	✗	✗

In addition, some of the critical exponents are not universal even in MFT.

Ex: Typical HS Cages Display non-MF Criticality?







Log-normal cage size distribution

Upon increasing *d*, θ approaches MF prediction, θ =1/2

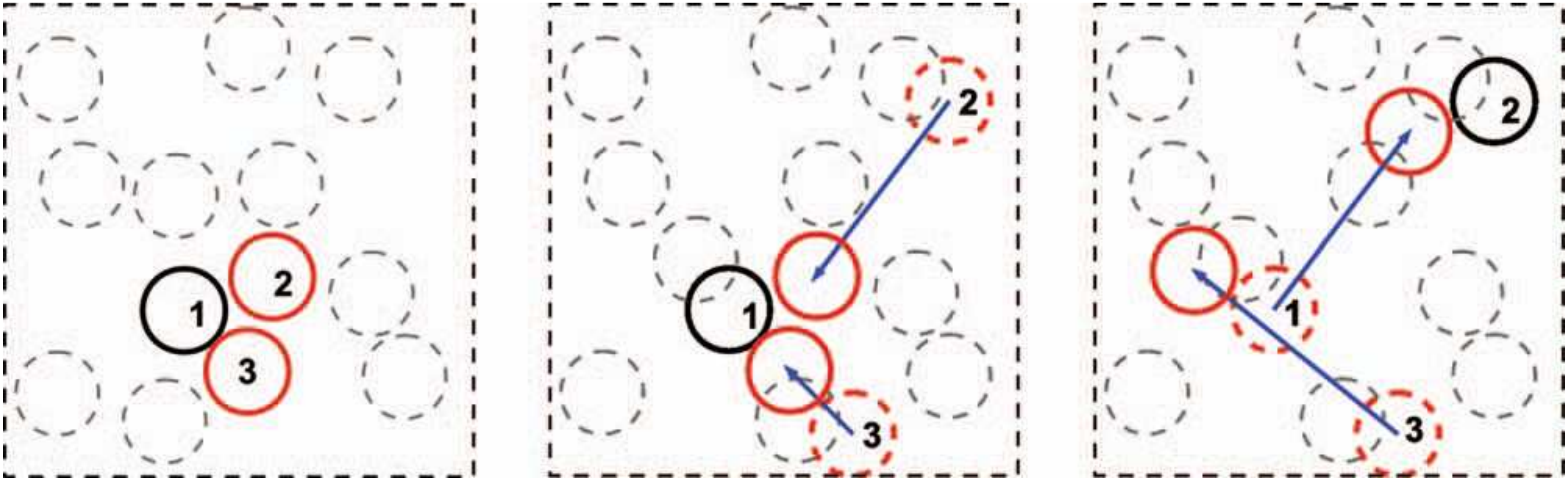
$$\bar{\Delta}(\phi_{\rm d}) - \bar{\Delta}(\phi) = A(\phi - \phi_{\rm d})^{\theta}$$

In progress: extend density and *d* range and compute dynamical susceptibility.

Kundu *et al*. in preparation (2019)

MK(K): structureless and interfaceless

$$\mathcal{H}_{\{\vec{\Lambda}\}} = \sum_{i}^{N} \frac{|\vec{p}_i|^2}{2m} + \sum_{i<j}^{N} u(|\vec{r}_{ij} + \vec{\Lambda}_{ij}|) \qquad \vec{\Lambda}_{ij} \in [0, L]^d$$



Mari, Kurchan, J. Chem. Phys. (2011)

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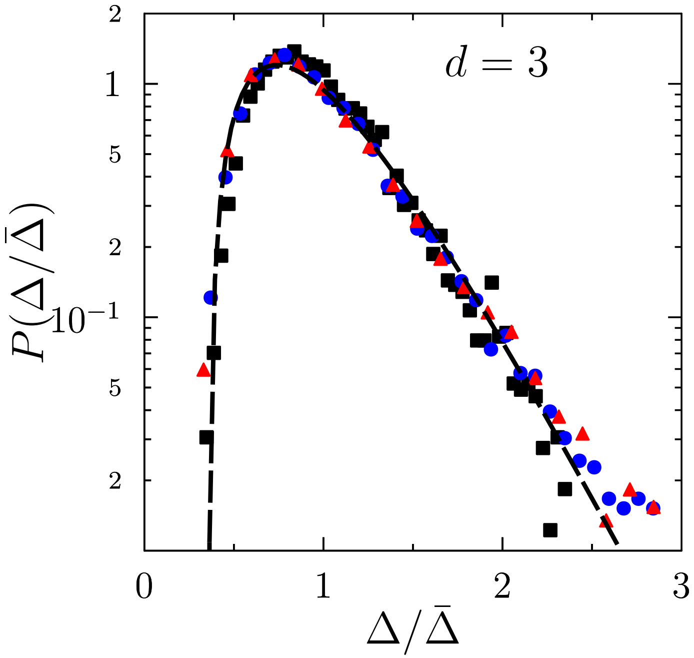
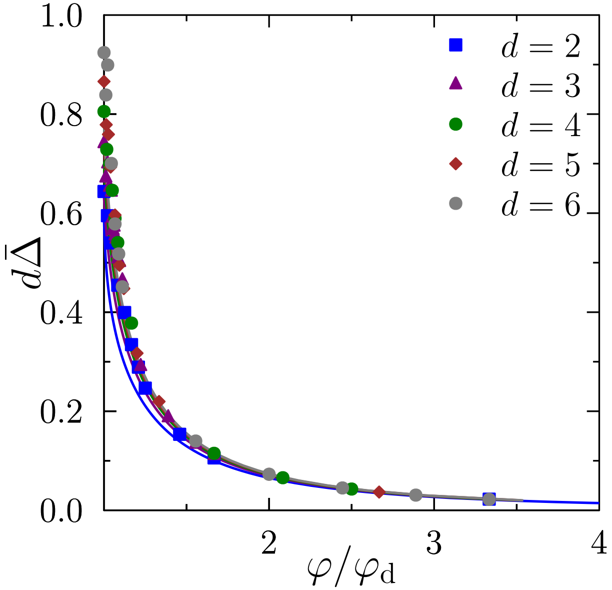
HS/MK in $d \to \infty$	✗	✗	✗
MK in $d = 3$	✗	✗	✗

Typical MK Cages Display MF Criticality



Log-normal cage size distribution

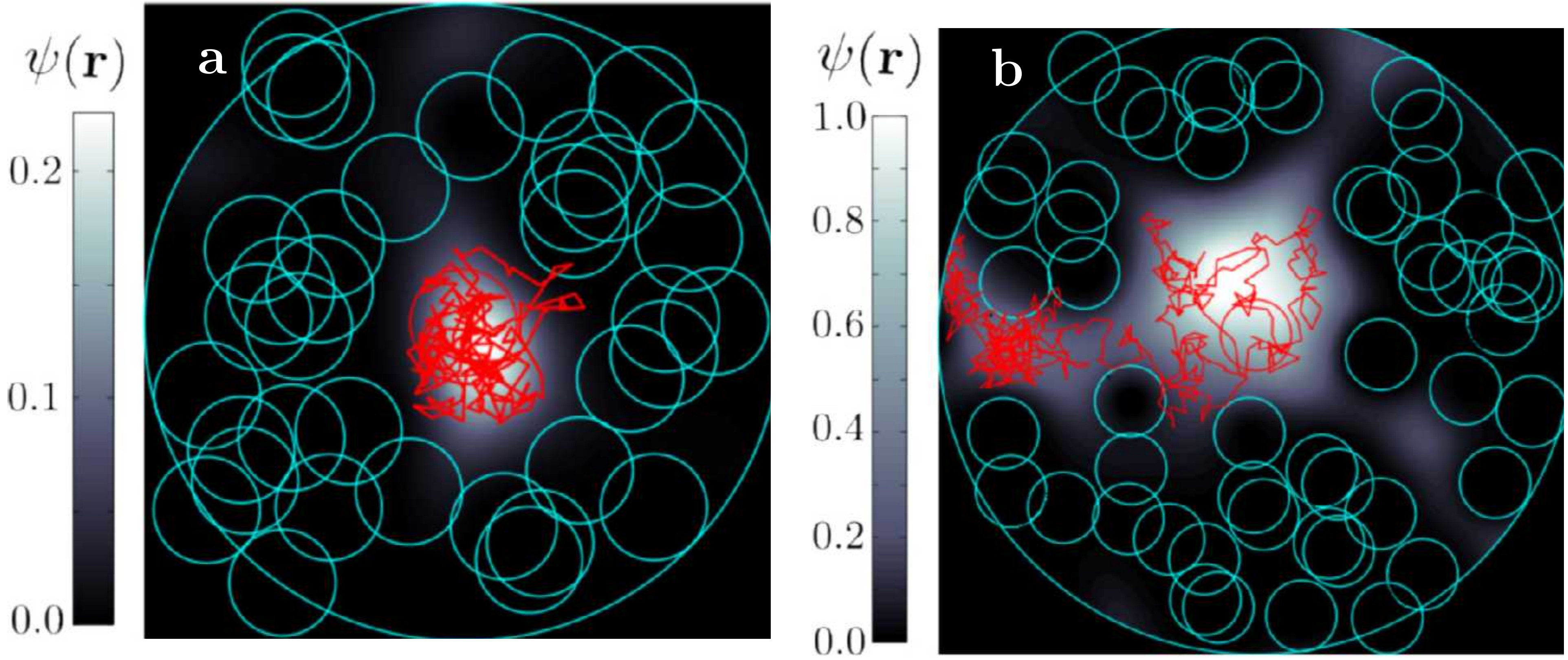


MF critical scaling

$$\bar{\Delta}(\phi_d) - \bar{\Delta}(\phi) = A(\phi - \phi_d)^{0.5}$$

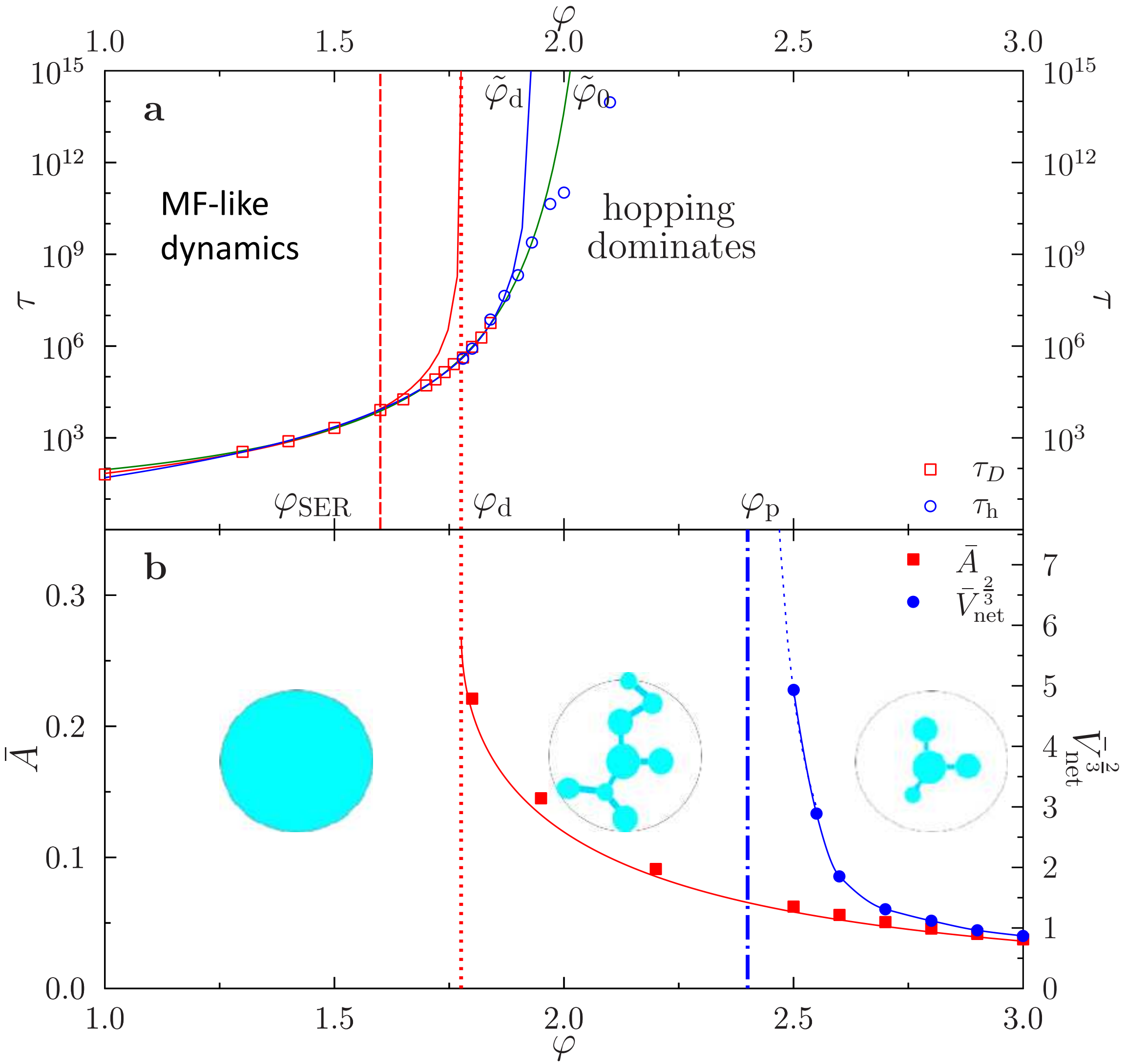
Charbonneau *et al*. PNAS (2015)

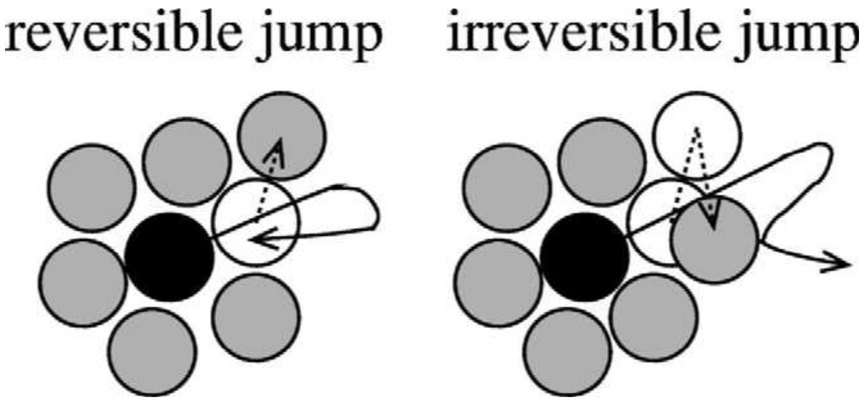
But Not Every Particle is Caged!



Mézard *et al.* (2011); Charbonneau *et al*. PNAS (2015)

Hopping Is the Unifying Hypothesis



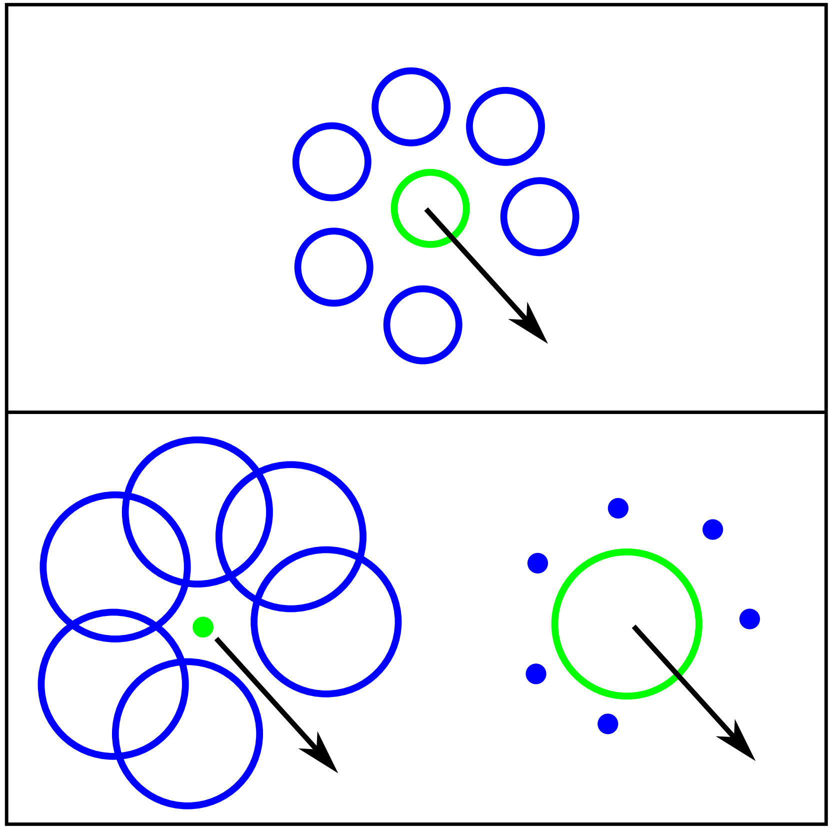


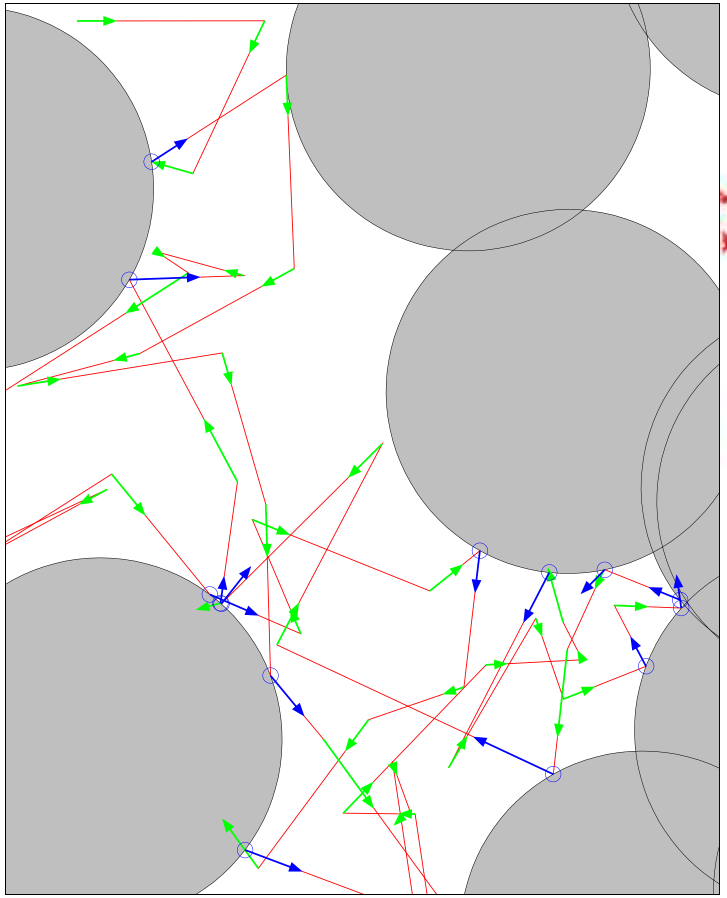
But hopping is completely absent from HS description in $d \to \infty$

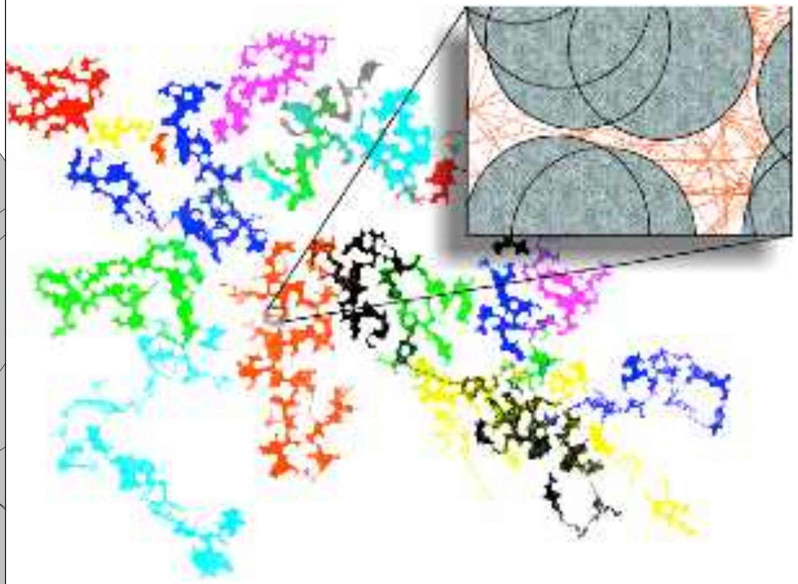
Charbonneau *et al*. (2014); Vollmayr-Lee (2004)

Minimal Model for Caging vs Hopping?

Freezing the obstacles in MK makes the problem equivalent to RLG and Void Percolation

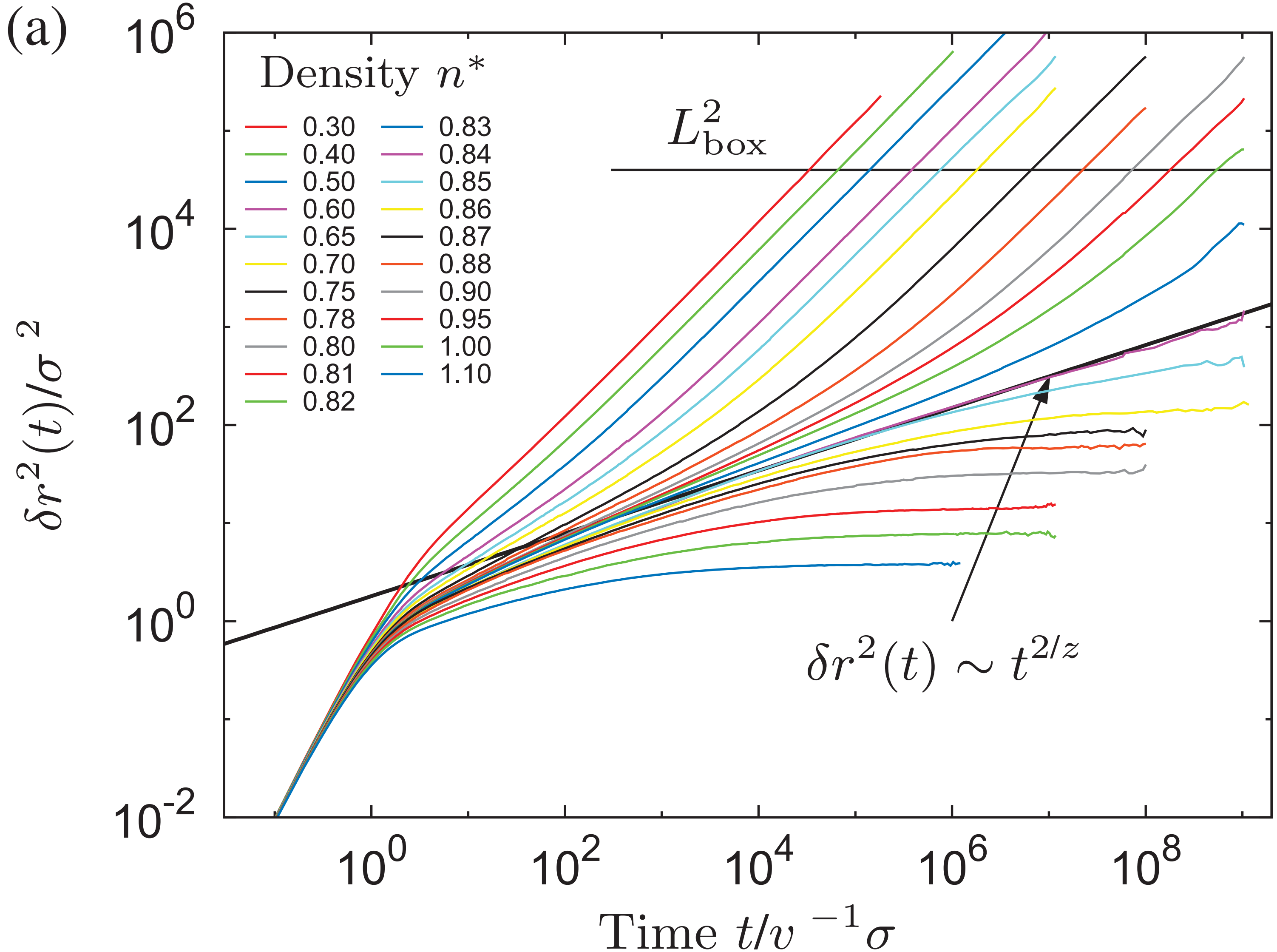




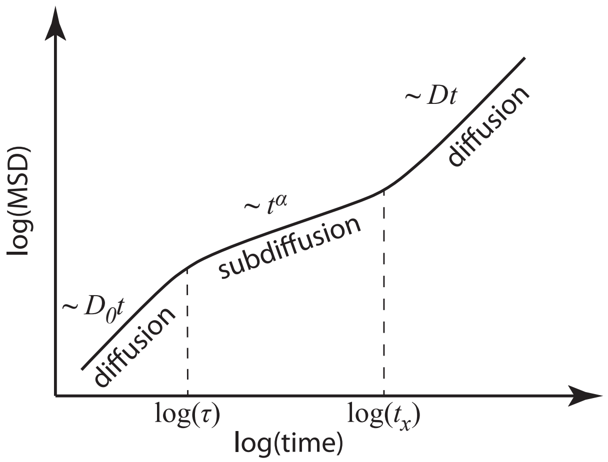


Jin and Charbonneau (2015); Franosch *et al*. (2008)

Finite-*d* RLG Localizes But Doesn't Cage.



Localization transition is continuous…



Franosch *et al*. (2008)

Lorentz gas in high d

Three assumptions:

1. Dynamics of the particle can be described by a generalized Langevin equation with Gaussian noise:

$$\gamma \dot{\mathbf{r}}(t) = -\int_0^t M^{\text{irr}}(t-t')\dot{\mathbf{r}}(t')dt' + \boldsymbol{\eta}(t) + \boldsymbol{\xi}(t)$$

γ - single-particle friction coefficient;

$M^{\text{irr}}(t)$ - irreducible memory function (internal friction kernel);

$\boldsymbol{\eta}_i(t)$ - Gaussian colored noise describing the fluctuating force due to the obstacles, $\langle \boldsymbol{\eta}_i(t)\boldsymbol{\eta}_j(t')\rangle = T\delta_{ij}\boldsymbol{I}M^{\text{irr}}(t-t')$;

$\boldsymbol{\xi}_i(t)$ - Gaussian white noise, $\langle \boldsymbol{\xi}_i(t)\boldsymbol{\xi}_j(t')\rangle = 2\gamma T\delta_{ij}\boldsymbol{I}\delta(t-t')$.

Three assumptions (cont.)

2. Approximate expression for the irreducible memory function:

$$M^{\text{irr}}(t) = \beta \left\langle \sum_i \hat{\mathbf{k}} \cdot \mathbf{F}_i(t^{\text{irr}}) \sum_j \hat{\mathbf{k}} \cdot \mathbf{F}_j(0) \right\rangle$$
$$\approx \beta \sum_i \left\langle \hat{\mathbf{k}} \cdot \mathbf{F}_i(t) \hat{\mathbf{k}} \cdot \mathbf{F}_i(0) \right\rangle$$

$\hat{\mathbf{k}}$ - unit vector

$\mathbf{F}_i(t^{\text{irr}})$ - force acting on the moving particle due to obstacle i, at time t, evolving with irreducible dynamics

$\mathbf{F}_i(t)$ - force acting on the moving particle due to obstacle i, at time t, evolving with normal dynamics.

Three assumptions (cont.)

3. The last assumption was based on the absence of the correlations between forces due to different obstacles; this is consistent with the following equation of motion of the particle with respect to the obstacle:

$$\gamma\dot{\mathbf{r}}_i(t) = \mathbf{F}(\mathbf{r}_i(t)) - \int_0^t M^{\text{irr}}(t-t')\dot{\mathbf{r}}_i(t')dt' + \boldsymbol{\eta}(t) + \boldsymbol{\xi}(t)$$

$\mathbf{r}_i \equiv \mathbf{r} - \mathbf{R}_i$, where $\mathbf{R}_i$ is the position of obstacle i

Noises $\boldsymbol{\eta}(t)$ and $\boldsymbol{\eta}(t)$ are independent realizations of the noises in the equation of motion for the particle.

Self-consistent equation for the Lorentz gas memory function

- The distance between the particle and the obstacle, $\mathbf{r}_1(t) \equiv \mathbf{r}(t) - \mathbf{R}_1$, evolves according to:

$$\gamma \dot{\mathbf{r}}_1(t) = \mathbf{F}(\mathbf{r}_1(t)) - \int_0^t M^{\mathrm{irr}}(t-t')\dot{\mathbf{r}}_1(t')dt' + \boldsymbol{\eta}(t) + \boldsymbol{\xi}(t)$$

$$\langle \boldsymbol{\eta}(t)\boldsymbol{\eta}(t')\rangle = T\boldsymbol{I}M^{\mathrm{irr}}(t-t') \quad \& \quad \langle \boldsymbol{\xi}(t)\boldsymbol{\xi}(t')\rangle = 2\gamma T\boldsymbol{I}\delta(t-t')$$

  where the memory function is determined by the process itself

$$M^{\mathrm{irr}}(t) = n\beta \int d\mathbf{r}_1 \left\langle \hat{\mathbf{k}} \cdot \mathbf{F}(\mathbf{r}_1(t))\, \hat{\mathbf{k}} \cdot \mathbf{F}(\mathbf{r}_1) \right\rangle g(r_1).$$

- Having found the memory function, we can study the motion of the particle,

$$\gamma \dot{\mathbf{r}}(t) = -\int_0^t M^{\mathrm{irr}}(t-t')\dot{\mathbf{r}}(t')dt' + \boldsymbol{\eta}(t) + \boldsymbol{\xi}(t).$$

Localization transition

- The theory predicts a localization transition; at the transition the memory function develops a non-decaying component, $\lim_{t\to\infty} M^{\mathrm{irr}}(t) = M_{\mathrm{EA}}$,

$$M_{\mathrm{EA}} = n\beta \int d\mathbf{s}\, P_{\mathrm{slow}}(\mathbf{s}) \left\langle \hat{\mathbf{k}} \cdot \mathbf{F}(\mathbf{r}) \right\rangle_{\mathbf{s}}^2$$

$$\langle \cdots \rangle_{\mathbf{s}} = \frac{\int d\mathbf{r} e^{-\beta\left(V(r)+M_{\mathrm{EA}}\mathbf{r}^2/2-\mathbf{s}\cdot\mathbf{r}\right)} \cdots}{\int d\mathbf{r} e^{-\beta\left(V(r)+M_{\mathrm{EA}}\mathbf{r}^2/2-\mathbf{s}\cdot\mathbf{r}\right)}} \qquad P_{\mathrm{slow}}(\mathbf{s}) = \int d\mathbf{r} \frac{e^{-\beta V(r) - \frac{(\mathbf{s}-M_{\mathrm{EA}}\mathbf{r})^2}{2TM_{\mathrm{EA}}}}}{(2\pi T M_{\mathrm{EA}})^{d/2}}$$

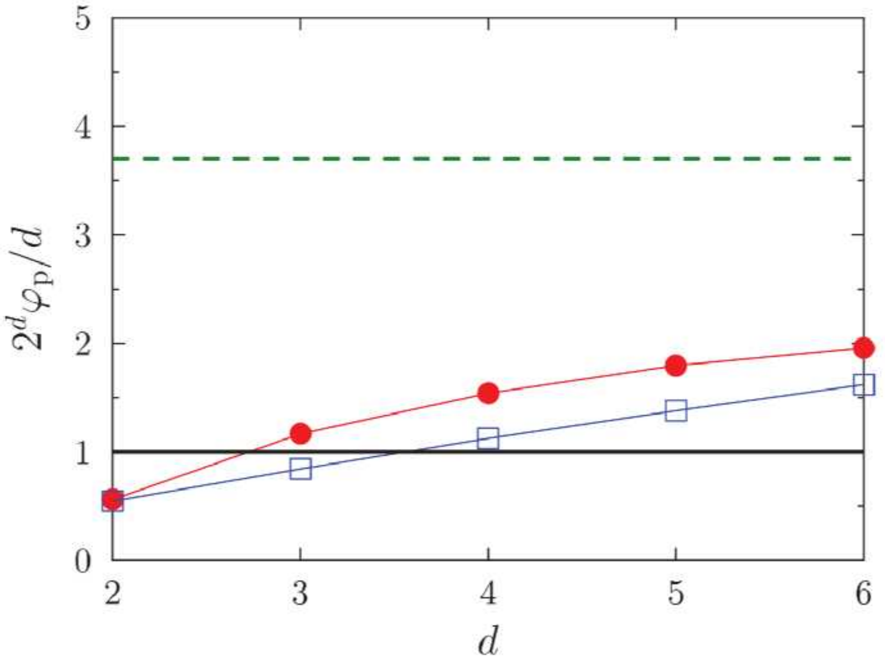
- The self-consistent equation for the localization transition is equivalent to the equation derived from the replica approach:

$$1 = -2\frac{nA}{d} \int d\mathbf{r} \frac{\partial q_{A/2}(r)}{\partial A} \ln q_{A/2}(r)$$ Ikeda & Zamponi, unpublished

$$q_A(r) = \int d\mathbf{r}' f_{2A}^{G}(\mathbf{r}') e^{-\beta V(|\mathbf{r}-\mathbf{r}'|)} \qquad f_{2A}^{G}(\mathbf{r}) = \frac{e^{-\mathbf{r}^2/4A}}{(4\pi A)^{d/2}} \qquad A = T/M_{\mathrm{EA}}$$

Equation for the localization transition is almost identical to the equation for the dynamic HS transition

- Both the dynamic theory and the replica theory predict a localization transition at the (rescaled) volume fraction equal to one half of the volume fraction at the dynamic transition of the hard sphere system: at the localization transition $2^d\phi/d \approx 2.403$.



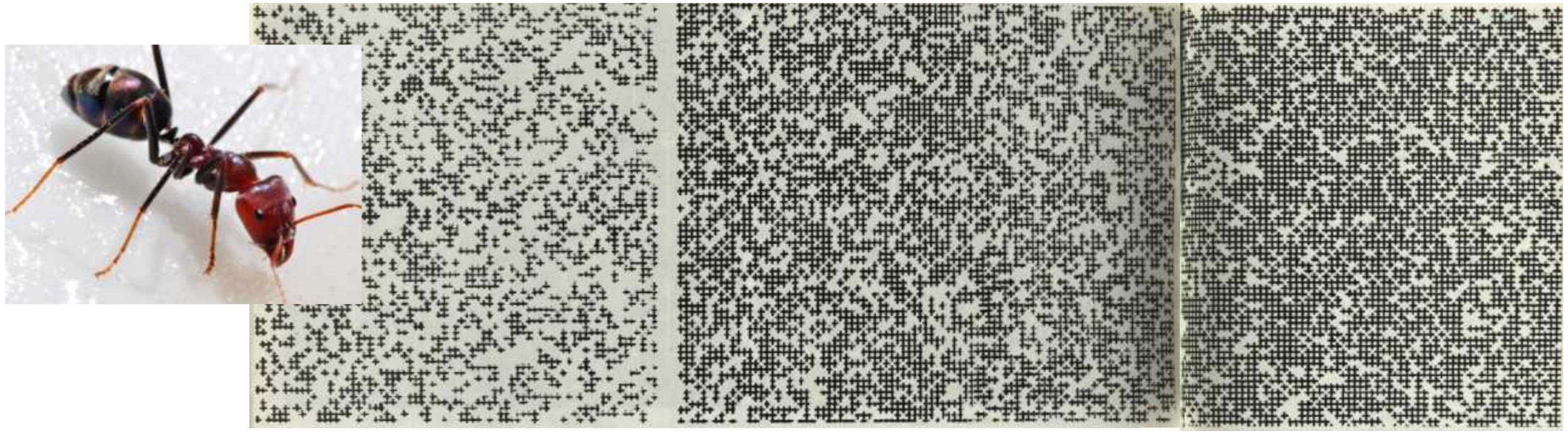
Jin & Charbonneau, PRE 91, 042313 (2015)

- This result is not inconsistent with numerical results (red circles).

RLG in finite *d* vs in the limit $d \to \infty$

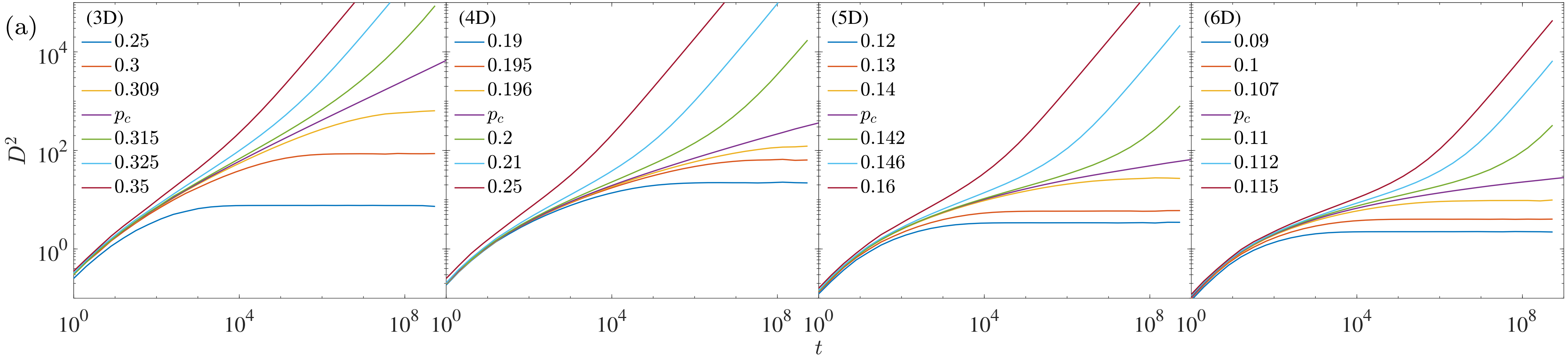
- In finite *d*, localization is continuous and therefore distinct from caging.
- In the limit $d \to \infty$, caging is perfect and appears discontinuously (Charbonneau, Hu, Ikeda, Szamel, and Zamponi, unpublished).
- Two possibilities:
  - Discontinuity is a general property of high-*d* percolation
  - High *d* discontinuity is specific to off-lattice models.

Simpler Analysis: Ant-in-a-labyrinth



The ant-in-a-labyrinth localization is not cage-like in finite *d*.

But for $d > d_u = 6$, (sub)subdiffusion is logarithmic.



Wikipedia; De Gennes (1976); Biroli, Charbonneau, Hu (2018)

Ant-in-a-labyrinth Analysis in $d \to \infty$

- Bethe Lattice for connectivity z:

$$\Delta^2 \approx -\frac{z}{z-2}\ln|p-p_c|$$

- The (sub)subdiffusive prefactor does not vanish in the $d \to \infty$ limit; localization and caging never coexist on a lattice.
- Distinction between caging and hopping might be a feature of off-lattice models!

- How to reconcile RLG results?
- To be continued…

Biroli, Charbonneau, Hu, arXiv:1810.7826 (2018)