

Tracer-bath correlations in d -dimensional interacting particle systems

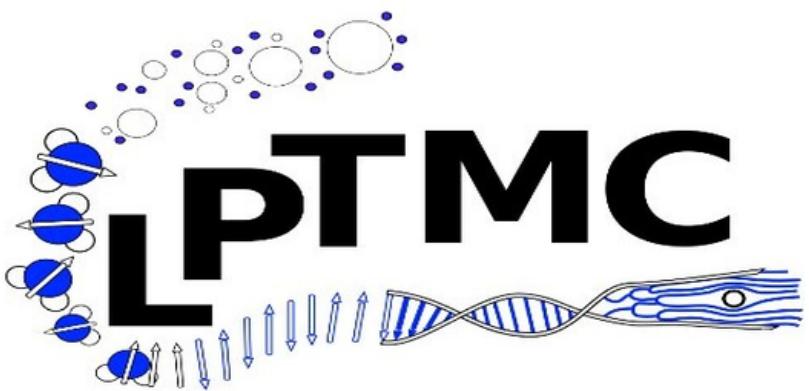
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DAMTP Statistical Physics and Soft Matter Seminar

University of Cambridge, 21 January 2025

Work in collaboration with P. Illien, T. Berlitz, A. Grabsch, O. Bénichou



Tracer particle in a thermal bath



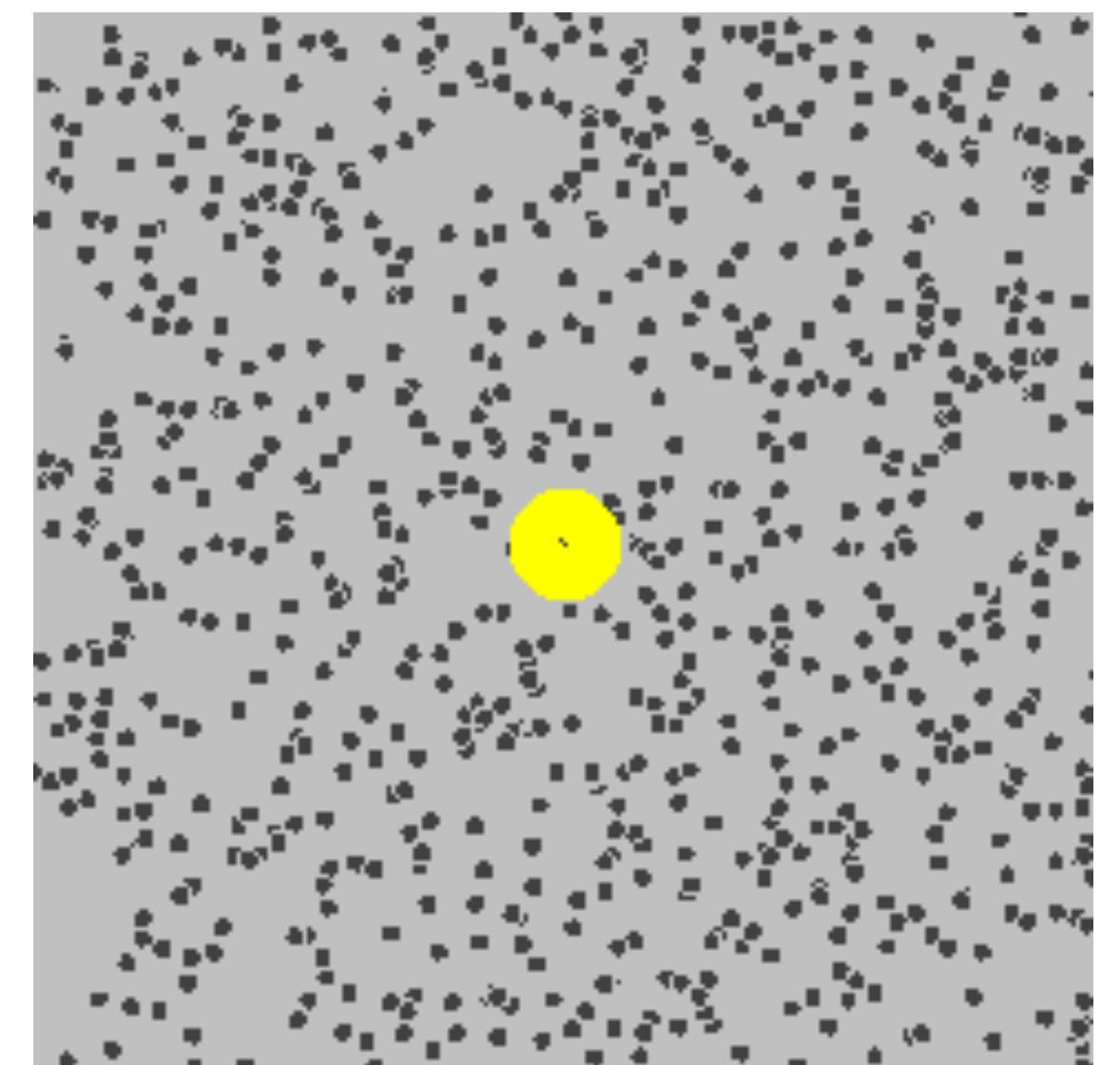
$$m \ddot{X}(t) = -\gamma \dot{X}(t) + \zeta(t)$$

$$\langle \zeta(t)\zeta(t') \rangle = 2\gamma k_B T \delta(t - t')$$

- ✿ **Brownian motion:**
bath in **equilibrium, structureless,**
no **tracer-bath** correlations, **diffusive** behaviour

$$\langle X^n(t) \zeta(t) \rangle = 0, \quad \langle X^2(t) \rangle \propto t$$

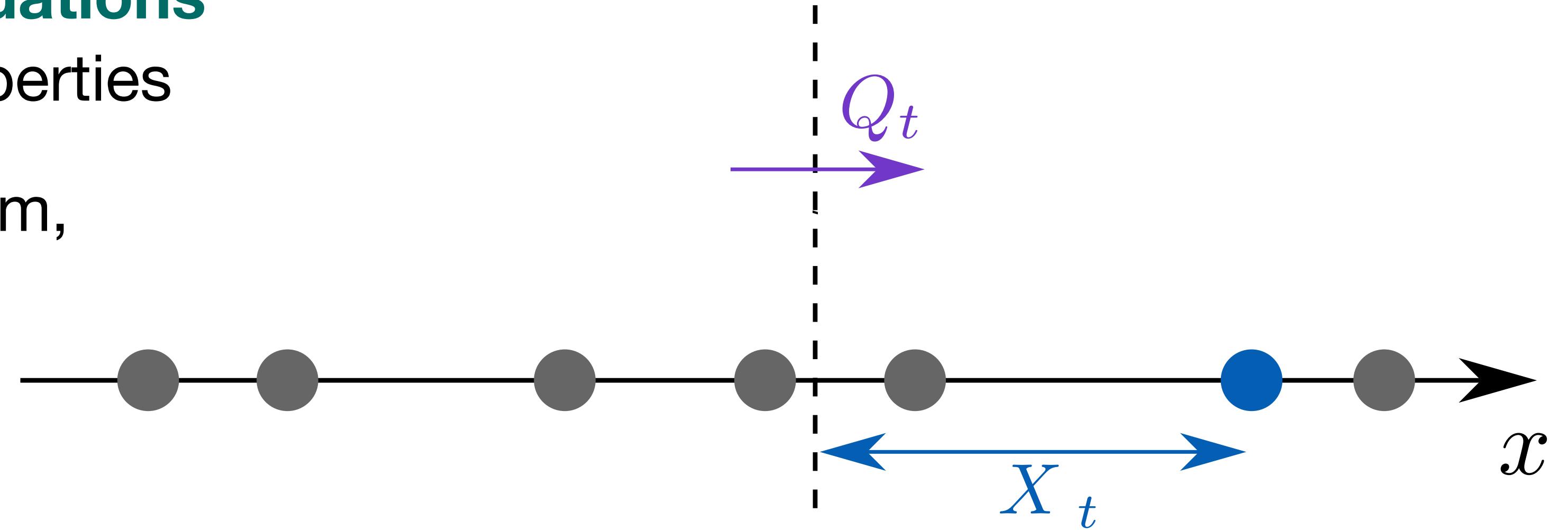
- ✿ What if particles have similar sizes?



Classical interacting particle systems

and why we still talk about them

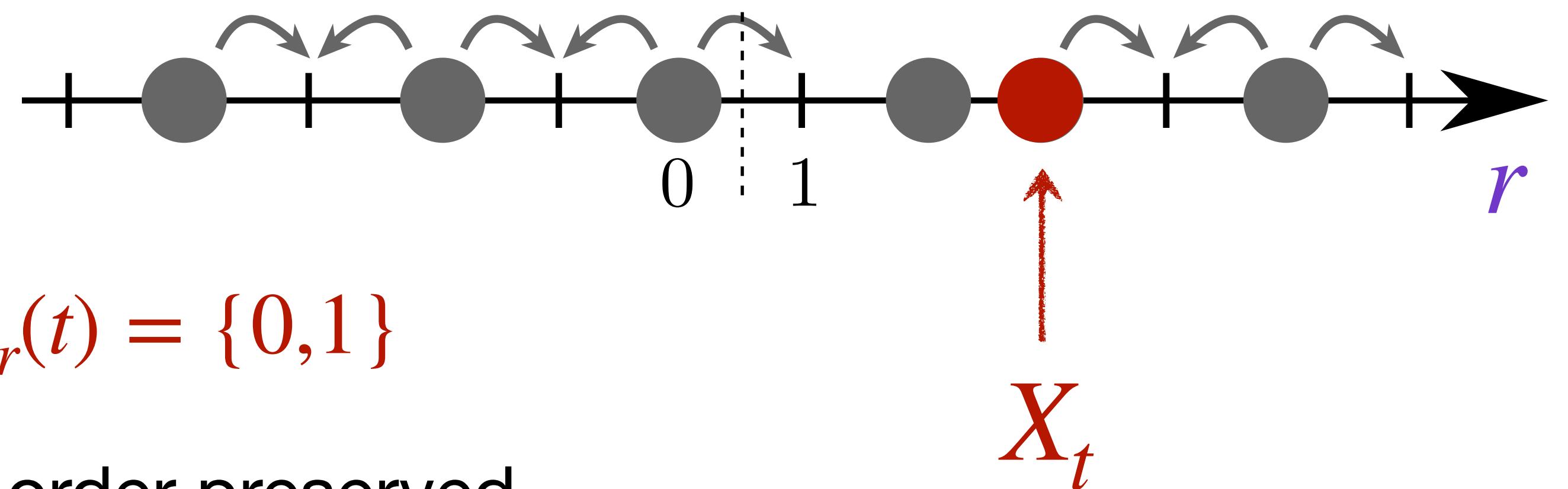
- Lattice gases, interacting Brownian particles, simple liquids...
- Dynamics, transport properties
- **Local observables:** integrated current Q_t , position X_t of tagged particle
- Random due to thermal **fluctuations**
 - determine statistical properties
- Interacting many-body problem,
out of equilibrium



Symmetric Exclusion Process

as paradigmatic diffusive system

- Particles on a lattice
+ random hoppings (equal rates),
only if target site is empty



- State of the system:** occupations $\rho_r(t) = \{0,1\}$
- In 1d, **single-file** geometry \rightarrow initial order preserved
- Subdiffusive behavior of tracer

$$\langle X_t^2 \rangle \propto \sqrt{t}$$

(zeolites, confined colloids, dipolar spheres...)

Lin, Meron, Cui, Rice, Diamant, Phys. Rev. Lett. **94** (21), 216001 (2005)

Wei, Bechinger, Leiderer, Science **287** (5453), 625-7 (2000)

Hahn, Kärger, Kukla, Phys. Rev. Lett. **76** (15), 2762-2765 (1996)

H. Spohn, *Large scale dynamics of interacting particles* (1991)

Role of correlations with surrounding bath

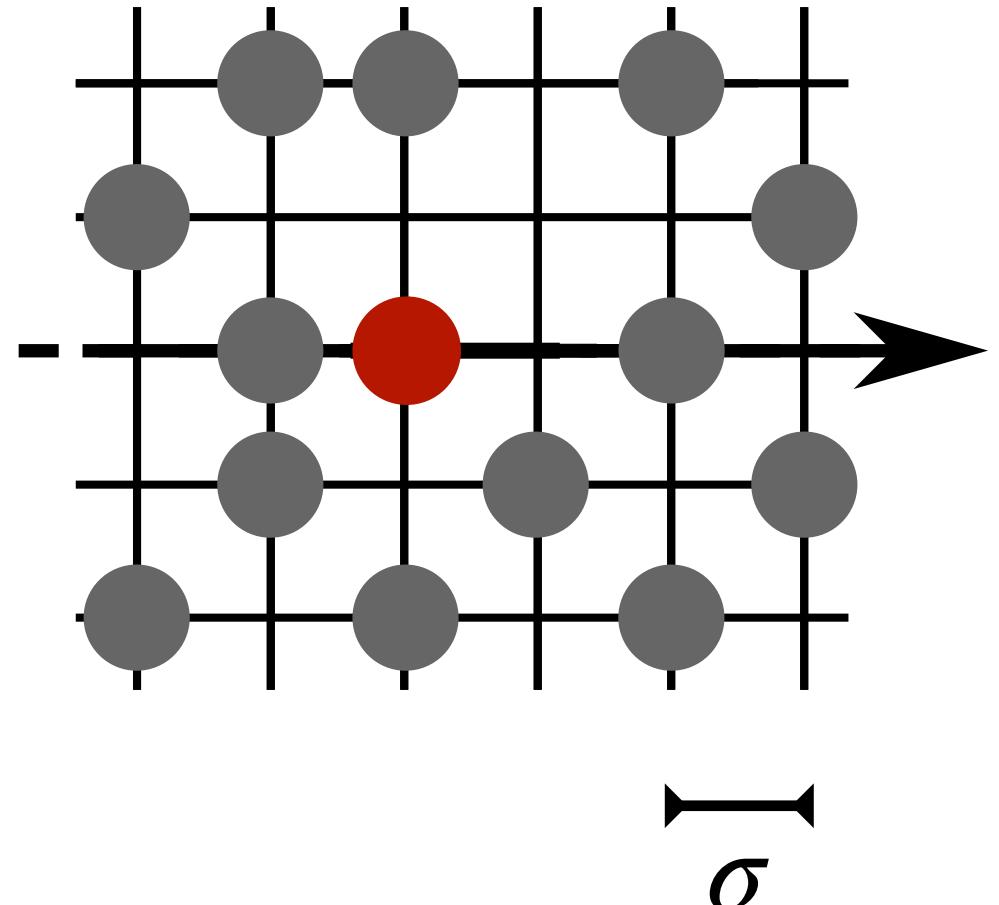
- Master equation $\partial_t P(X, \underline{\rho}, t) = [\mathcal{L}_{\text{tracer}} + \mathcal{L}_{\text{bath}}] P(X, \underline{\rho}, t)$

- Multiply by $e^{\lambda \cdot X}$ and average,

$$\partial_t \Psi(\lambda, t) = \frac{1}{2d\tau} \sum_{\mu=-d}^d \left(e^{\sigma \lambda \cdot \hat{\mathbf{e}}_\mu} - 1 \right) [1 - w_{\mathbf{e}_\mu}(\lambda, t)]$$

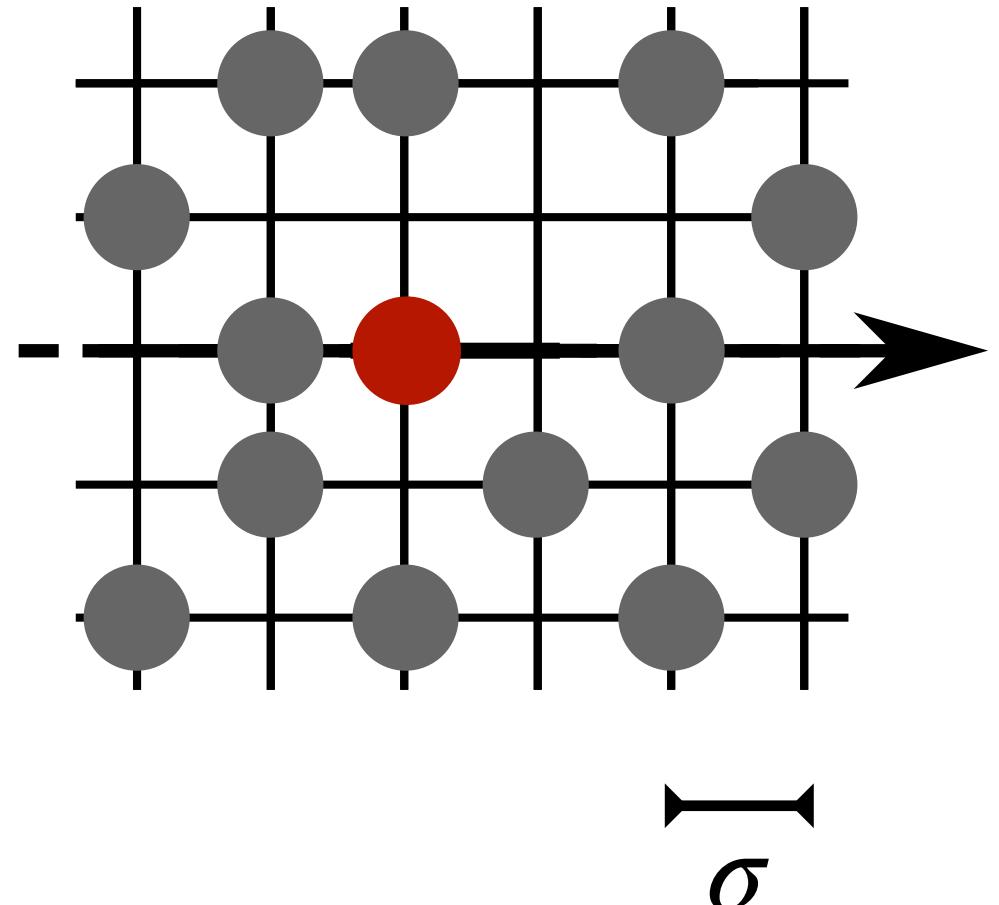
$$\Psi(\lambda, t) = \ln \langle e^{\lambda \cdot X} \rangle = \sum_{n=1}^{\infty} \frac{\lambda^n}{n!} \langle X^n \rangle_c,$$

$$w_r(\lambda, t) = \frac{\langle \rho_{X+r} e^{\lambda \cdot X} \rangle}{\langle e^{\lambda \cdot X} \rangle} = \sum_{n=0}^{\infty} \frac{\lambda^n}{n!} \langle \rho_{X+r} X^n \rangle_c$$

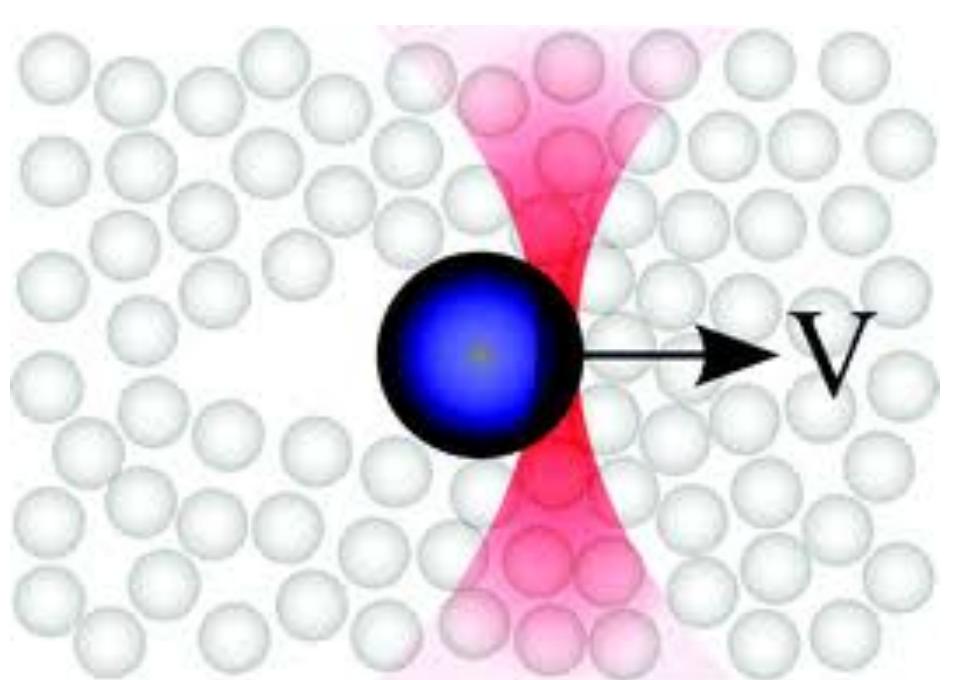


Role of correlations with surrounding bath

$$\partial_t \Psi(\lambda, t) = \frac{1}{2d\tau} \sum_{\mu=-d}^d \left(e^{\sigma \lambda \cdot \hat{\mathbf{e}}_\mu} - 1 \right) \left[1 - w_{\mathbf{e}_\mu}(\lambda, t) \right]$$

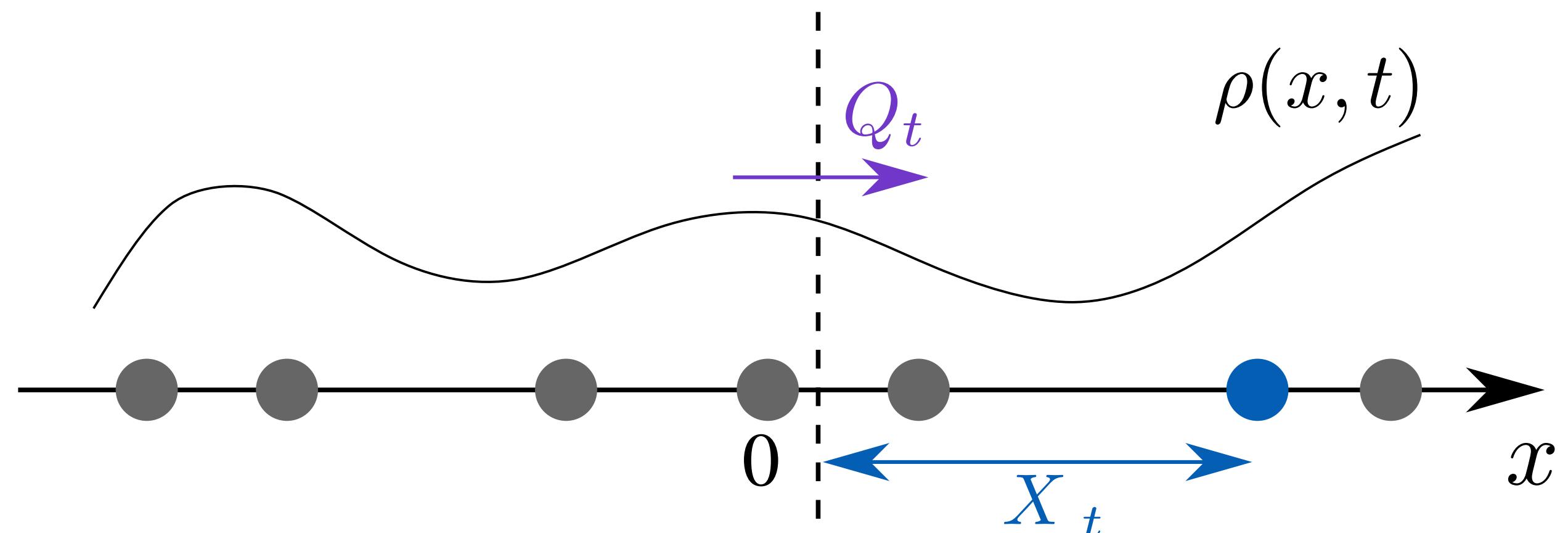


- Knowing $w_{\mathbf{e}_\mu}(\lambda, t)$ on neighbouring sites is enough to deduce $\Psi(\lambda, t)$
- $\partial_t w_{\mathbf{e}_\mu}(\lambda, t) = \dots [w_{\mathbf{r}}(\lambda, t)] \dots$ generically depends on $w_{\mathbf{r}}(\lambda, t)$ even from far away \mathbf{r}
→ **strongly correlated**
- $w_{\mathbf{r}}(\lambda, t)$ encodes the **response** of the bath



Integrated current Q_t

- Q_t = net # of particles crossing (0–1) \rightarrow in $d = 1$, $\langle Q_t^2 \rangle \propto \sqrt{t}$
- A positive fluctuation of Q_t is correlated with an increase of $\rho_r(t)$ on its r.h.s.
 \rightarrow **correlations dictate the subdiffusive behavior of Q_t**
- $\langle \rho_r(t) e^{\lambda Q_t} \rangle$ encodes the **response** of the bath
- Fully understood in 1d SEP
- Open problem in $d > 1$



Grabsch, Poncet, Rizkallah, Illien, Bénichou, Sci. Adv. 8, eabm5043 (2022)

1. Current fluctuations

- As **first step**, focus on

$$c_r(t) \equiv \langle Q_t \rho_r(t) \rangle$$

- Fact 1:** infinite lattice $d = 1$ (no reservoirs, no PBC)

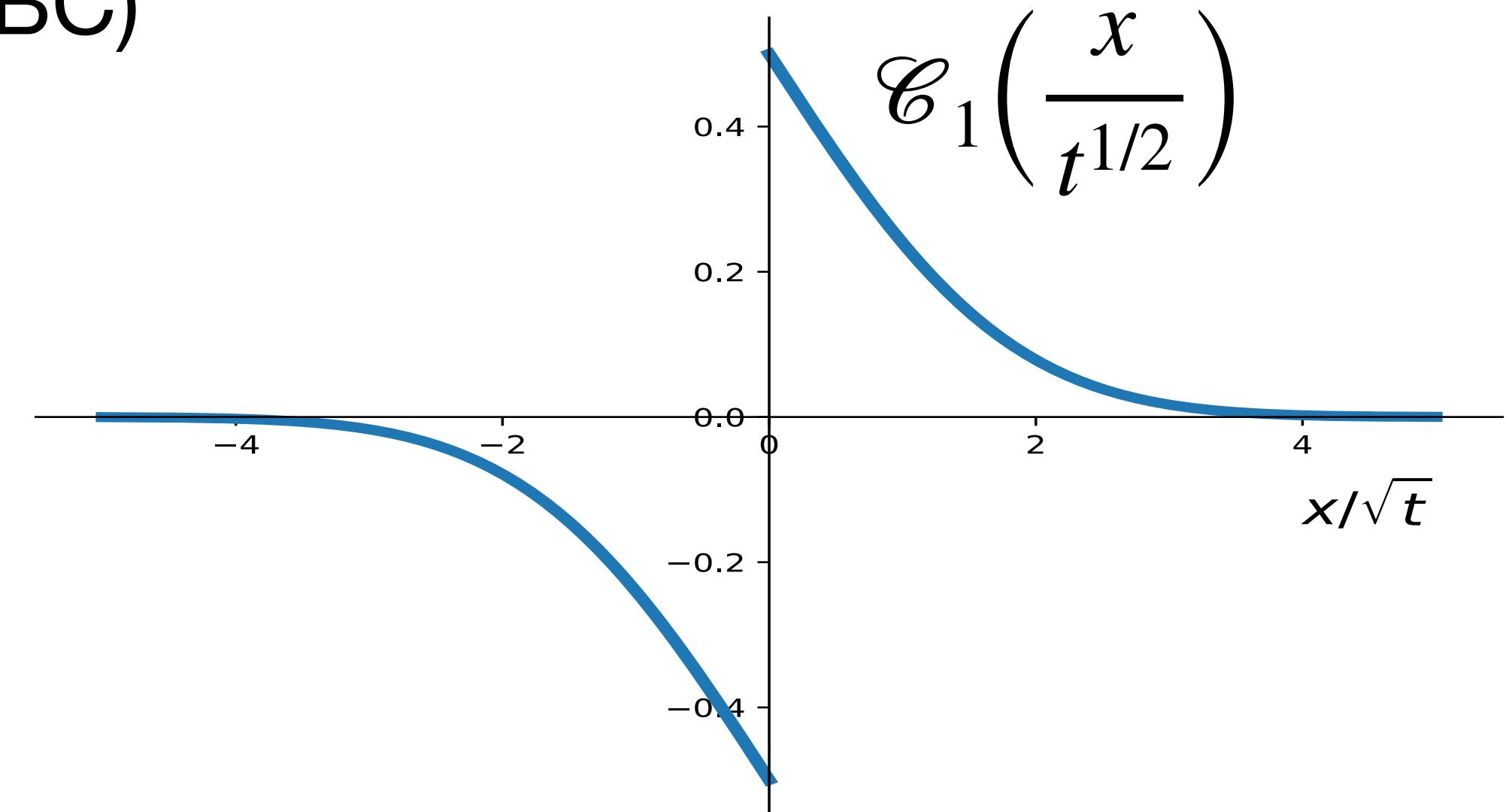
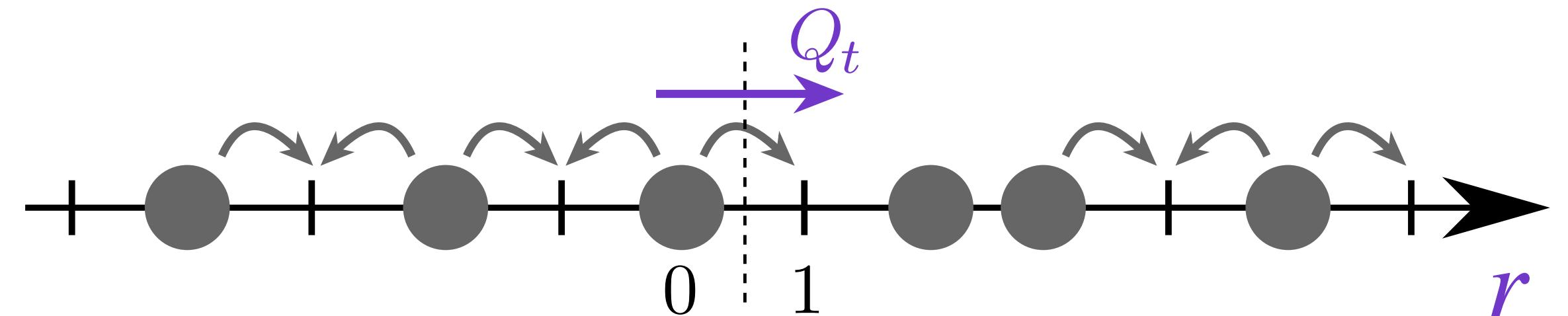
$$c_r(t) \xrightarrow[t \rightarrow \infty]{} \bar{\rho}(1 - \bar{\rho}) \mathcal{C}_1\left(\frac{r}{t^{1/2}}\right),$$

$$\langle Q_t^2 \rangle \propto \bar{\rho}(1 - \bar{\rho}) t^{1/2}$$

- Fact 2:** finite systems (any spatial dimension d)

$$c_{\vec{r}}(t) \xrightarrow[t \rightarrow \infty]{} c_{\vec{r}}, \quad \langle Q_t^2 \rangle \propto t$$

Recall $\partial_t \langle Q_t^2 \rangle = \dots [c_{\vec{r}}(t)] \dots$



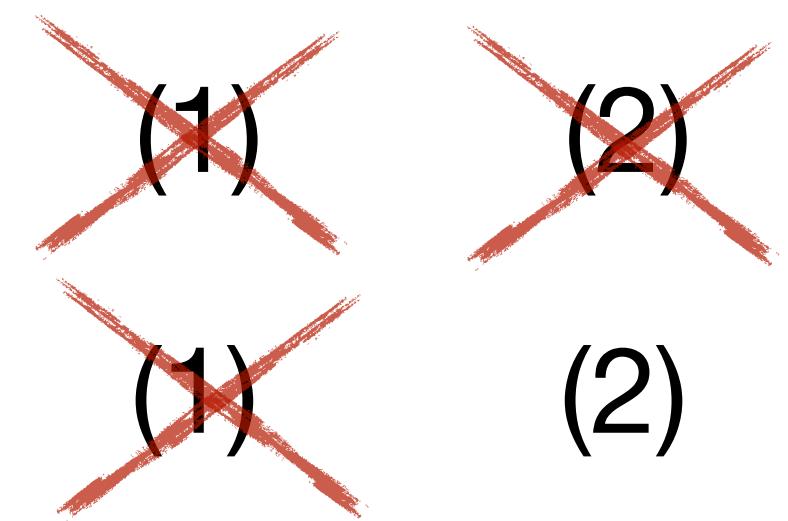
infinite lattices in $d > 1$?

From $d = 1$ to higher d passing through the comb

1. Order-preserving (single-file)
2. Tree-like (no loops!)

Do we need both, to make $c_r(t)$ time dependent?

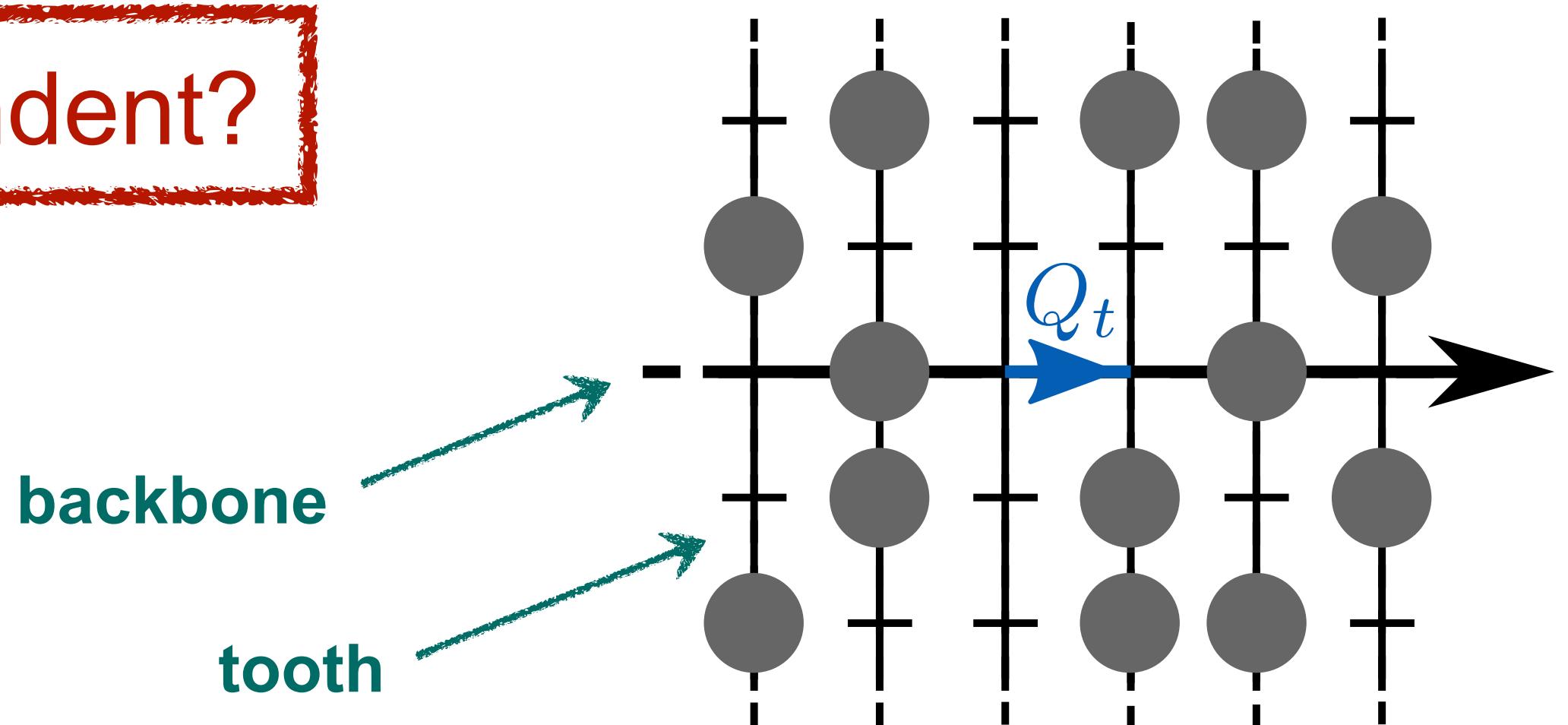
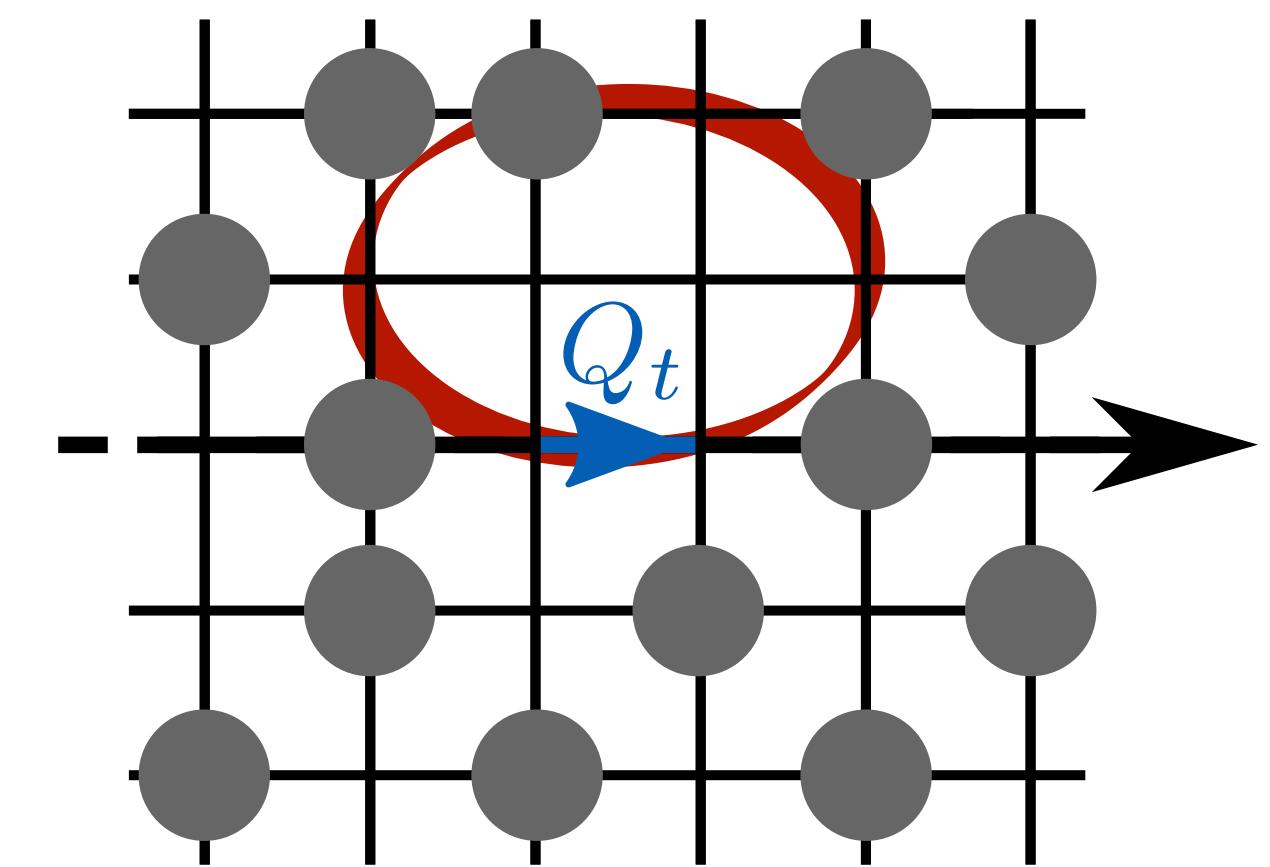
Higher d :



Comb lattice :



Transport in porous media,
subdiffusion (even single particle!)



Bénichou, Illien, Oshanin, Sarracino, Voituriez, Phys. Rev. Lett. 115, 220601 (2015)
Ben-Avraham, Havlin, *Diffusion and reactions in fractals and disordered systems* (2000)

Microscopic calculation

- Master equation

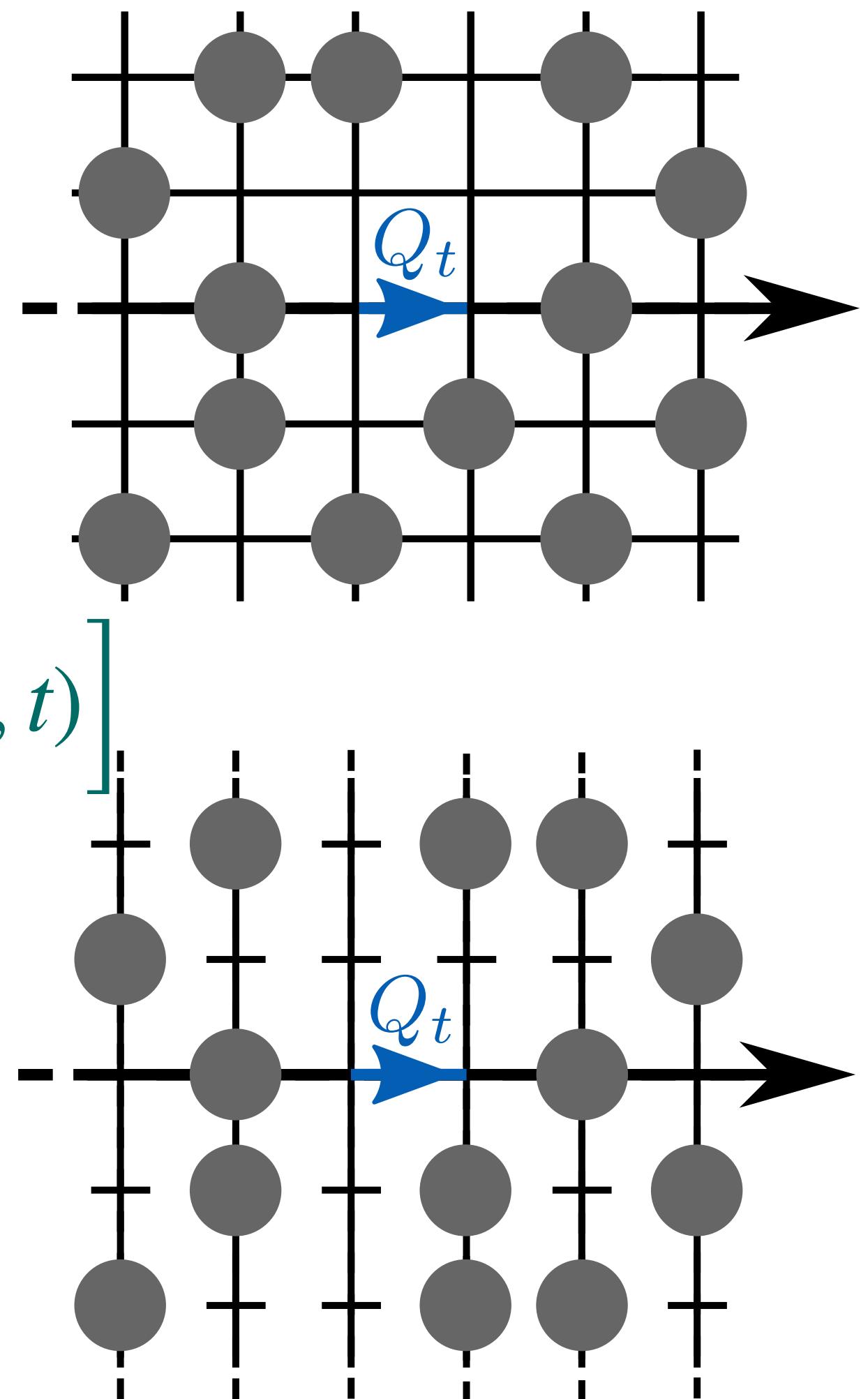


$$\partial_t P_t(\underline{\rho}) = \sum_{x,y} \left[P(\underline{\rho}^{x,y+}, t) - P(\underline{\rho}, t) \right] + \sum_{x,y} \left[P(\underline{\rho}^{x+,y}, t) - P(\underline{\rho}, t) \right]$$

- Look for moments, e.g. $\partial_t \langle Q_t^2 \rangle = \dots [c_{\vec{r}}(t)] \dots$

- Find exact closed equations $\partial_t c_{\vec{r}}(t)$ for $c_{\vec{r}}(t) = \langle Q_t \rho_{\vec{r}}(t) \rangle$

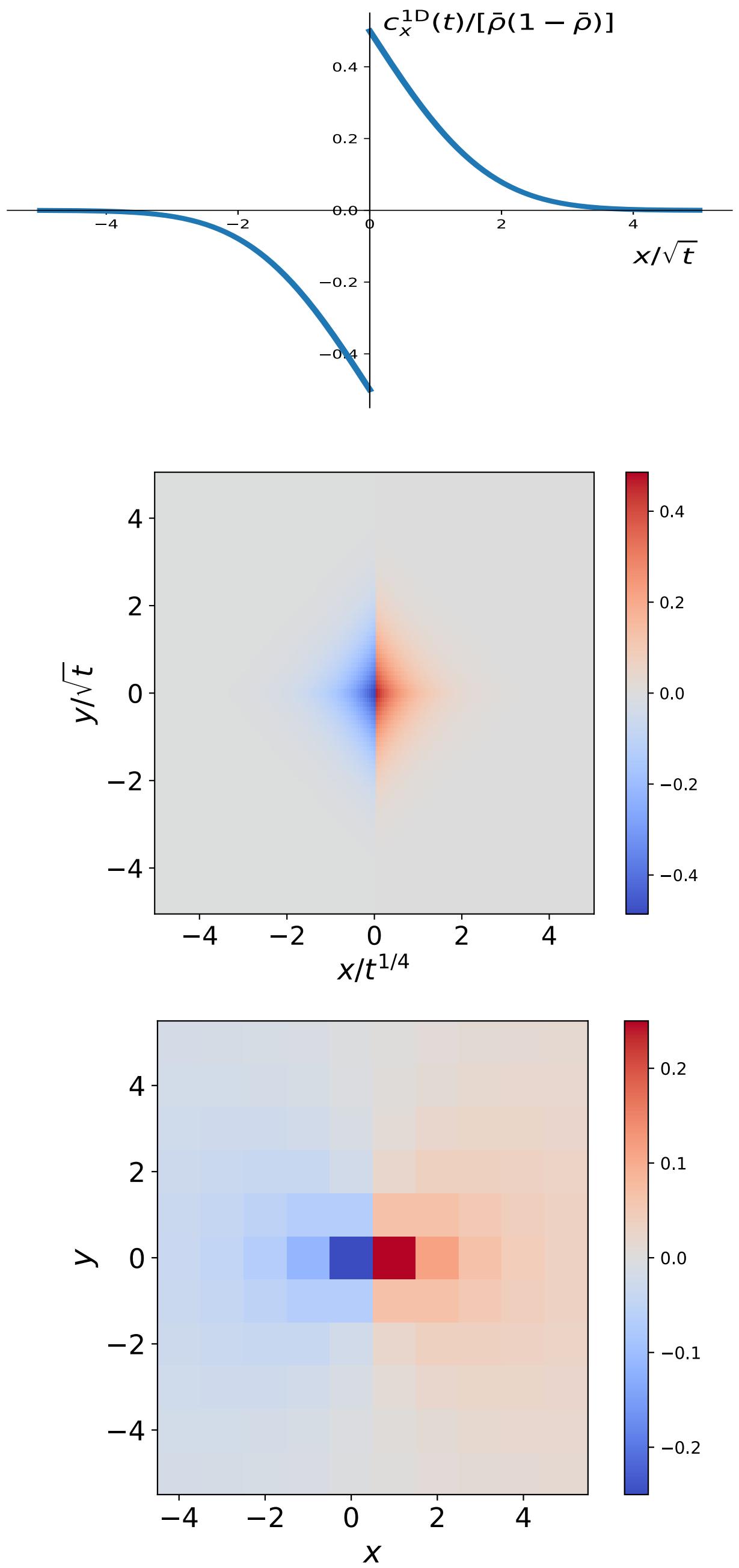
- Solve them in Fourier-Laplace (self-consistently)



Results

- $d = 1$ $c_r(t) \xrightarrow[t \rightarrow \infty]{} \bar{\rho}(1 - \bar{\rho}) \mathcal{C}_1\left(\frac{r}{t^{1/2}}\right)$
 $\langle Q_t^2 \rangle = n_1 \bar{\rho}(1 - \bar{\rho}) t^{1/2}$
- Comb $c_{\vec{r}}(t) \xrightarrow[t \rightarrow \infty]{} \bar{\rho}(1 - \bar{\rho}) \mathcal{C}_c\left(\frac{x}{t^{1/4}}, \frac{y}{t^{1/2}}\right)$
 $\langle Q_t^2 \rangle = n_c \bar{\rho}(1 - \bar{\rho}) t^{3/4}$
- $d = 2$ $c_{\vec{r}}(t) \xrightarrow[t \rightarrow \infty]{} \bar{\rho}(1 - \bar{\rho}) \mathcal{C}_2(\vec{r})$
 $\langle Q_t^2 \rangle = n_2 \bar{\rho}(1 - \bar{\rho}) t$

loops!



Macroscopic Fluctuation Theory

Hydrodynamic description for the occupations,

$$\partial_t \langle \rho_{\vec{r}}(t) \rangle = \delta_{y,0} \Delta_x \langle \rho_{\vec{r}}(t) \rangle + \Delta_y \langle \rho_{\vec{r}}(t) \rangle$$

$$\langle \rho_{\vec{r}}(t) \rangle \simeq \rho \left(\frac{x}{T^{1/4}}, \frac{y}{T^{1/2}}, \frac{t}{T} \right)$$



$$\partial_t \rho + \vec{\nabla} \cdot \vec{j} = 0$$

$$\vec{j} = -\mathbf{D} \vec{\nabla} \rho + \vec{\nu}$$

$$\mathbf{D} = \begin{pmatrix} \delta(y) & 0 \\ 0 & 1 \end{pmatrix}$$

Add noise:

$$\langle \nu_i(x, y, t) \nu_j(x', y', t') \rangle = \Sigma_{i,j}(\rho(x, y, t)) \delta(x - x') \delta(y - y') \delta(t - t')$$

$$\Sigma(\rho) = 2\rho(1 - \rho) \begin{pmatrix} \delta(y) & 0 \\ 0 & 1 \end{pmatrix}$$

T. Berlitz, D. Venturelli, A. Grabsch, O. Bénichou, J. Stat. Mech. (2024) 113208

Macroscopic Fluctuation Theory

on the comb

$$\partial_t \rho(\vec{r}, t) = \vec{\nabla} \cdot [\mathbf{D} \vec{\nabla} \rho + \vec{\nu}] \longrightarrow P[\rho] = \int \mathcal{D}H e^{-T^{3/4} S[\rho, H]}$$

- Integrated current fluctuations

$$Q_T \simeq T^{3/4} \int_0^\infty dx \int_{-\infty}^\infty dy [\rho(x, y, 1) - \rho(x, y, 0)]$$

$$\langle e^{\lambda Q_T} \rangle = \int \mathcal{D}\rho \mathcal{D}H e^{-T^{3/4} S[\rho, H] + \lambda Q_T}$$

- Saddle point

$$\begin{aligned} \rho &= \rho^{(0)} + \lambda \rho^{(1)} + \dots \\ H &= H^{(0)} + \lambda H^{(1)} + \dots \end{aligned}$$

correlation profile

$$\frac{\langle \rho_{\vec{r}=(x,y)}(T) e^{\lambda Q_T} \rangle}{\langle e^{\lambda Q_T} \rangle} \simeq \rho^*(x, y, 1)$$

Macroscopic Fluctuation Theory

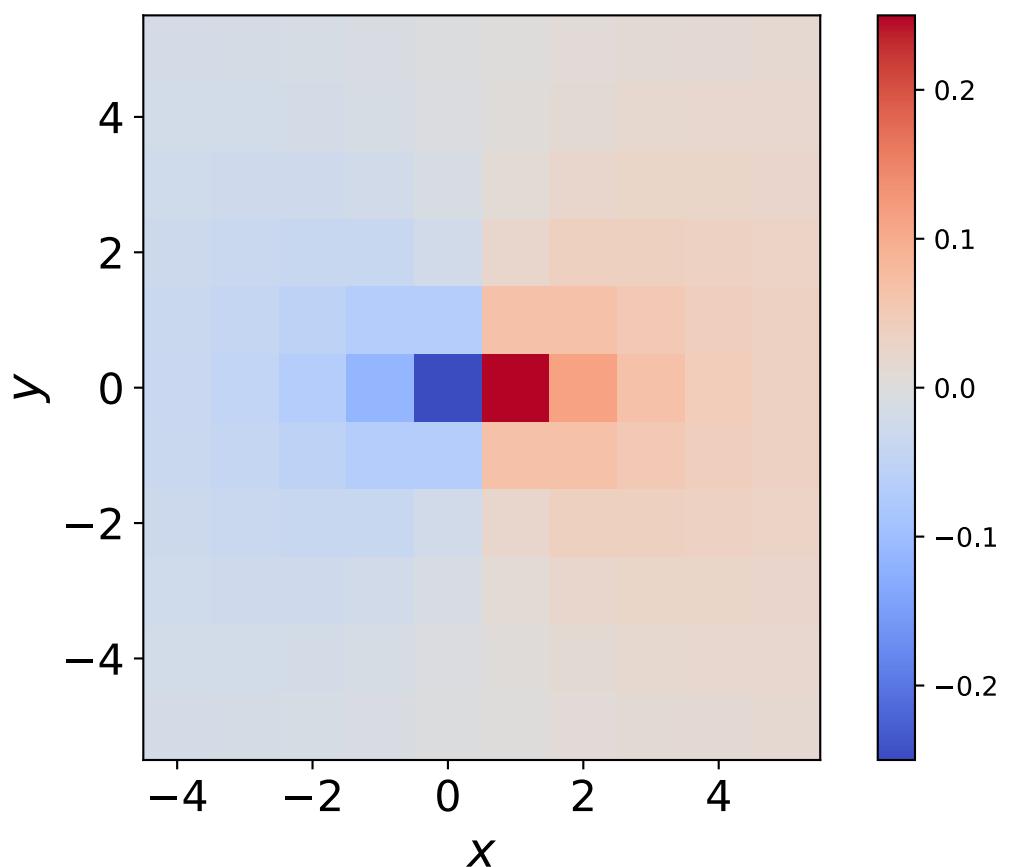
towards higher moments and general diffusive processes

- Recover $\langle \rho_r(T) Q_T \rangle$, $\langle Q_T^2 \rangle$ + in principle higher moments/correlations
- Can be extended to other **models**
- First application of MFT to an **inhomogeneous** system (**comb**)

T. Berlioz, D. Venturelli, A. Grabsch, O. Bénichou, J. Stat. Mech. (2024) 113208

What about higher d ?

- In $1d$ & comb, correlations vary slowly at the lattice scale
- In higher d correlations become stationary, no scaling limit!



Microscopic path-integral representation

$$\rho_{\vec{r}}(t + dt) - \rho_{\vec{r}}(t) = dt \sum_{\vec{\nu}} \left(\vec{j}_{\vec{r}-\vec{\nu}}(t) - \vec{j}_{\vec{r}}(t) \right) \cdot \vec{\nu},$$

$$\vec{j}_{\vec{r}}(t) dt = \sum_{\vec{\nu}} [\rho_{\vec{r}}(1 - \rho_{\vec{r}+\vec{\nu}}) \xi_{\vec{r},\vec{\nu}}(t) - \rho_{\vec{r}+\vec{\nu}}(1 - \rho_{\vec{r}}) \xi_{\vec{r}+\vec{\nu},-\vec{\nu}}(t)] \vec{\nu},$$

equivalent to the M.E. if Poissonian noise is $\xi_{\vec{r},\vec{\mu}}(t) = \begin{cases} 1 & \text{prob. } \gamma dt, \\ 0 & \text{prob. } 1 - \gamma dt. \end{cases}$

Usual **MSR** machinery gives

$$\langle e^{\lambda Q_T} \rangle = \int \mathcal{D}\theta_{\vec{r}} \mathcal{D}\vec{\varphi}_{\vec{r}} e^{-S[\rho_{\vec{r}}, \vec{j}_{\vec{r}}, \theta_{\vec{r}}, \vec{\varphi}_{\vec{r}}] + \lambda Q_T} \longrightarrow$$

$$Q_T = \int_0^T dt \left(\vec{j}_{\vec{r}=\vec{0}}(t) \right)_1$$

A. Lefèvre, G. Biroli, J. Stat. Mech. (2007) P07024

Microscopic path-integral representation

- Saddle-point eqs are **difference equations** for $\rho_{\vec{r}}, \vec{j}_{\vec{r}}, \theta_{\vec{r}}, \vec{\varphi}_{\vec{r}}$
- Turn out to relax to a stationary limit,

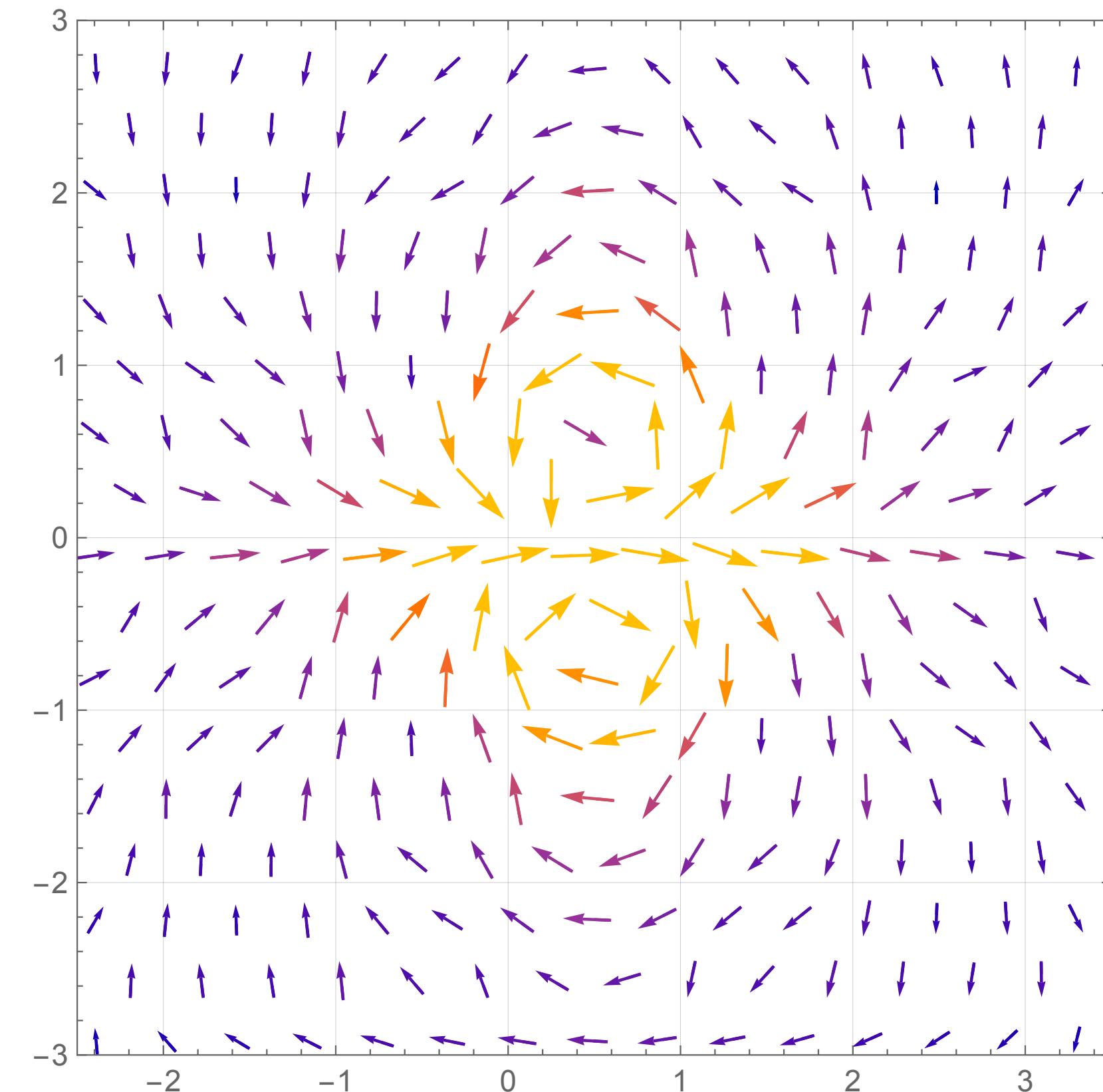
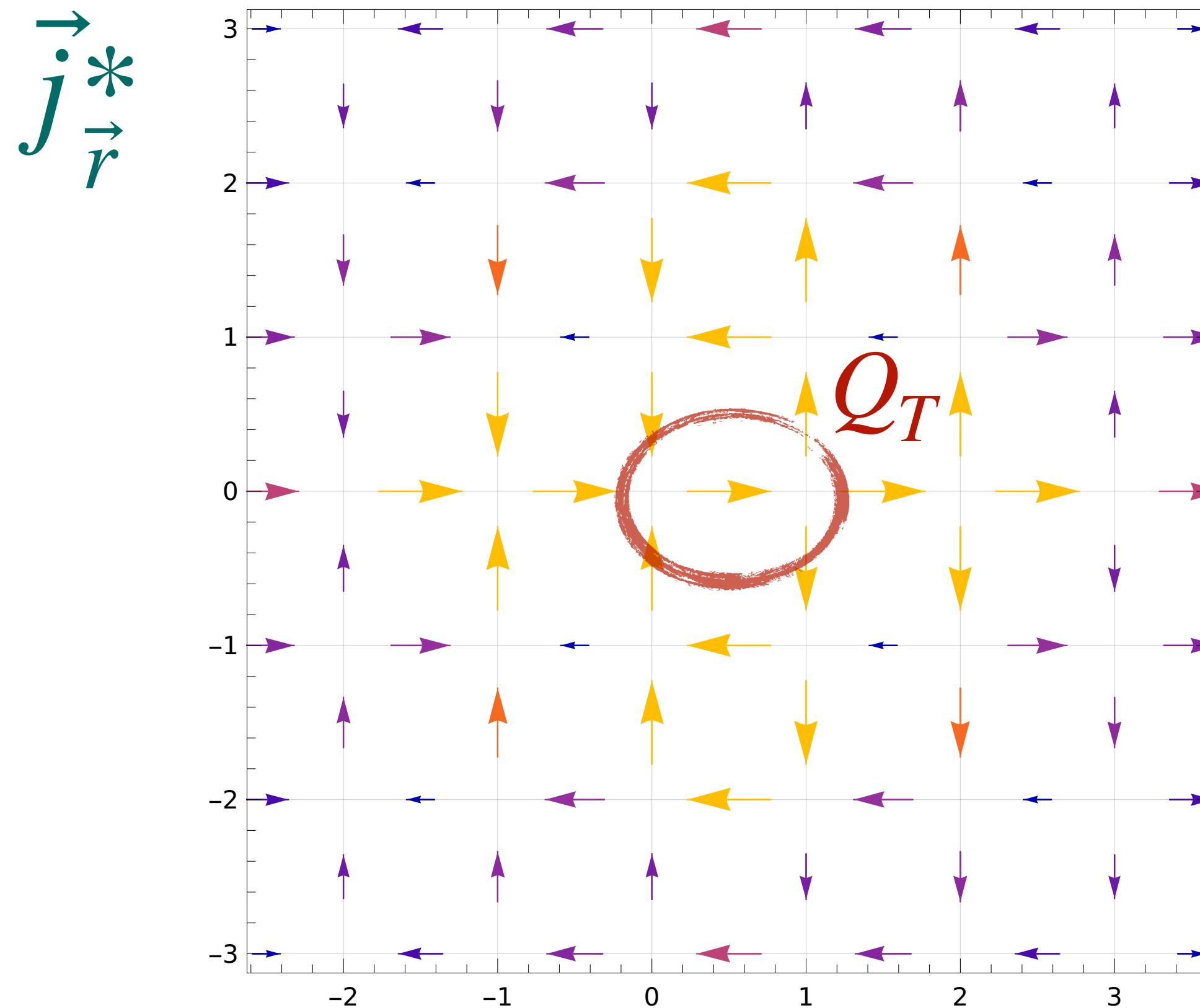
$$\mathcal{S} = \int_0^T dt \mathcal{L}[\{\rho, \vec{j}, \theta, \vec{\varphi}\}] \simeq T \mathcal{L}^*[\{\rho^*, \vec{j}^*, \theta^*, \vec{\varphi}^*\}]$$

- Can be used to recover

$$\langle e^{\lambda Q_T} \rangle \simeq \exp\{-T[\mathcal{L}^* - \lambda(\vec{j}_{\vec{r}=0}^*)_1]\} \rightarrow \langle Q_t^2 \rangle = 2\gamma \left(1 - \frac{1}{d}\right) \bar{\rho}(1 - \bar{\rho}) t$$

Role of loops

$$\frac{\langle \vec{j}_{\vec{r}}(t) e^{\lambda Q_T} \rangle}{\langle e^{\lambda Q_T} \rangle} \simeq \vec{j}_{\vec{r}}^*(t) \rightarrow \langle \vec{j}_{\vec{r}}(t) Q_T \rangle$$



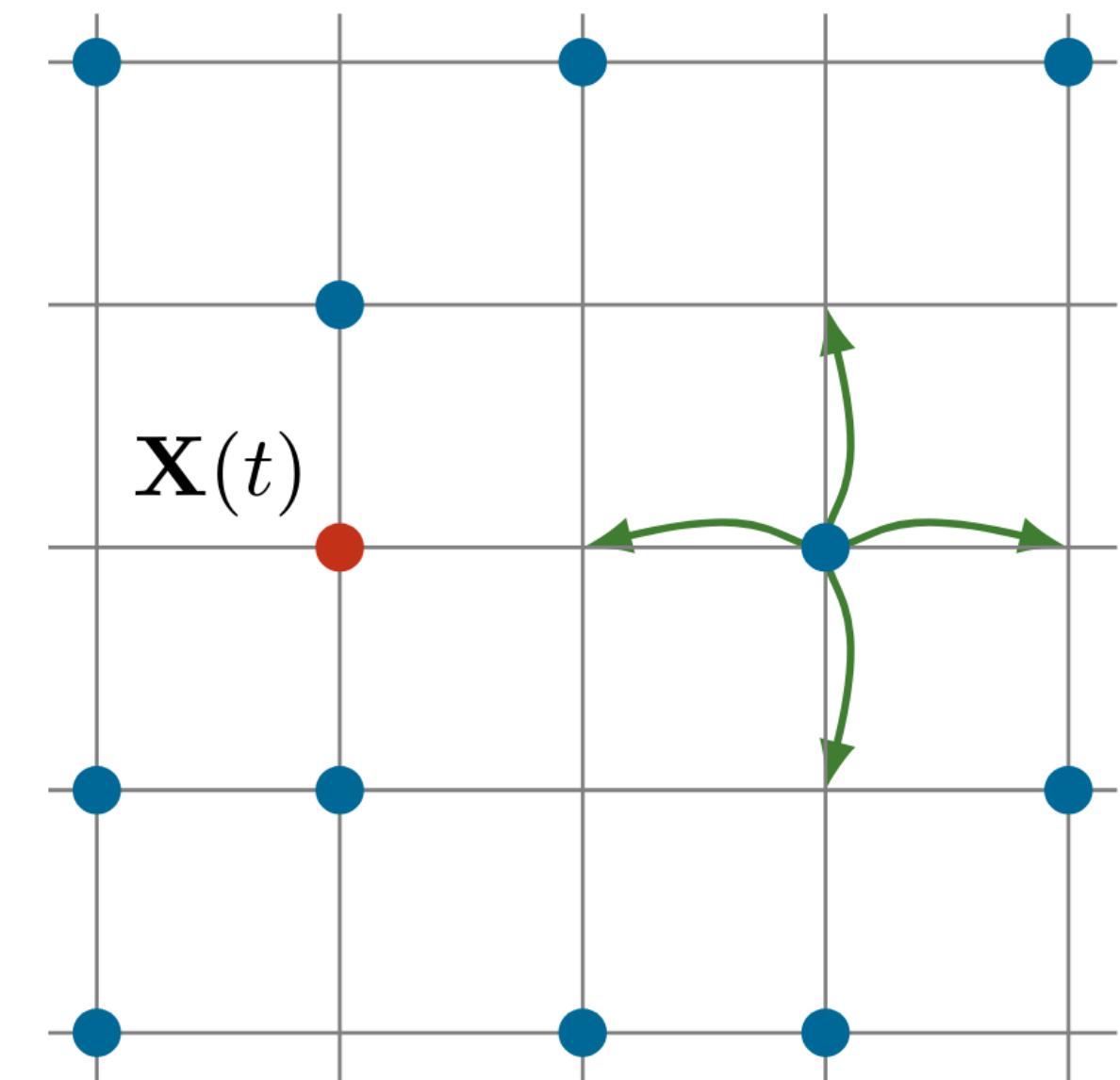
looped structure of the lattice allows for **vortex** configurations, and thus for stationary $c_{\vec{r}}$

2. Tracer-bath correlations

Hard-core lattice gas in d dimensions

- Tagged tracer at X_t , bath occupations $\rho_r(t) = \{0,1\}$
- Using the ME, get an equation for $g_r(t) = \langle X_t \rho_{X+r}(t) \rangle \rightarrow \text{not closed!}$
- $\partial_t g_r(t) = (\dots)$ can be closed upon **decoupling**
$$\langle \rho_{X+r} \rho_{X+r'} \rangle \simeq \langle \rho_{X+r} \rangle \langle \rho_{X+r'} \rangle$$
$$\langle X_t \rho_{X+r} \rho_{X+r'} \rangle \simeq \langle X_t \rho_{X+r} \rangle \langle \rho_{X+r'} \rangle + \langle X_t \rho_{X+r'} \rangle \langle \rho_{X+r} \rangle$$
(exact for large/small $\bar{\rho}$)

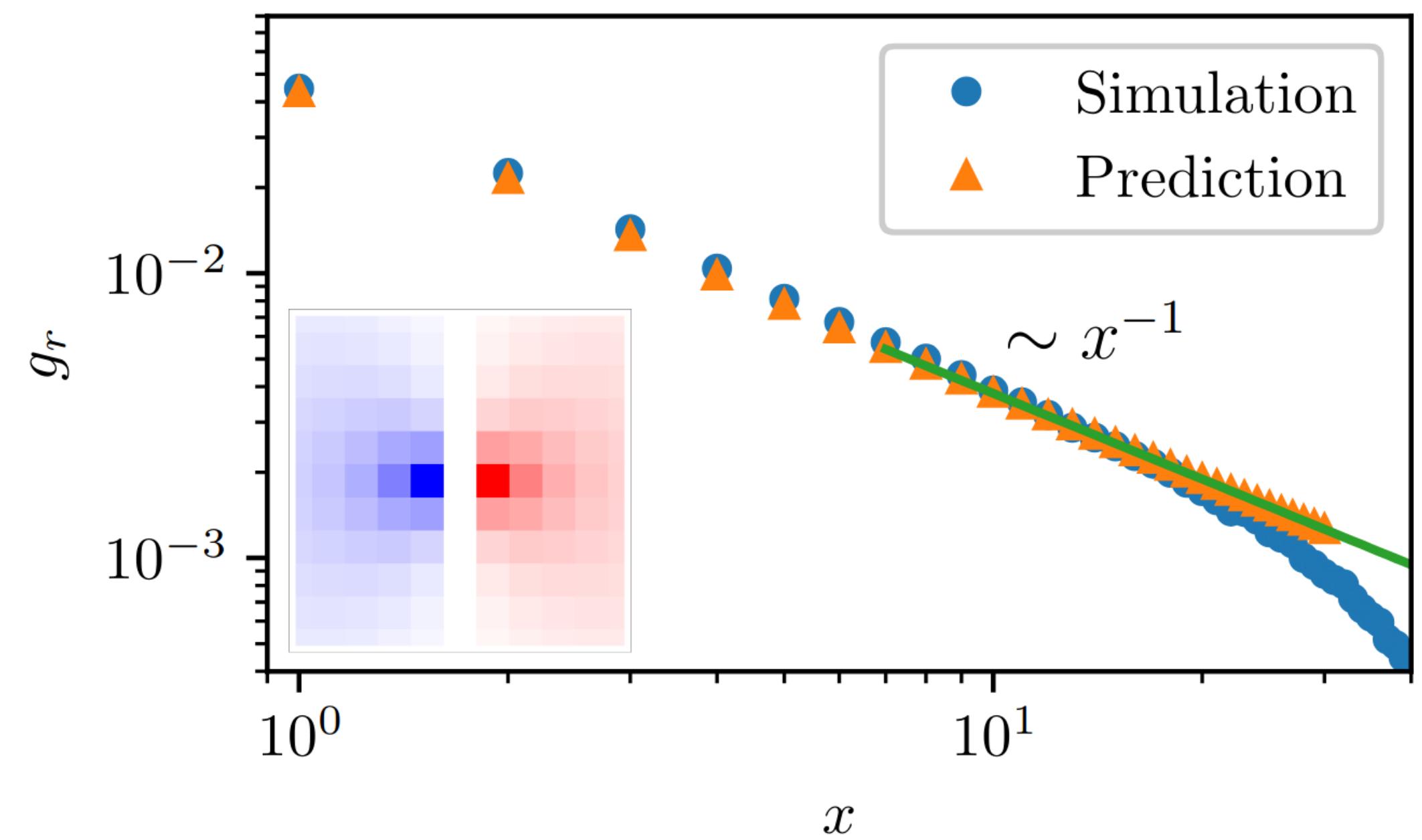
Bénichou, Illien, Oshanin, Sarracino, Voituriez, Phys. Rev. Lett. 115, 220601 (2015)



Hard-core lattice gas in d dimensions

- Self-consistent *difference* eq. for $g_r(t) = \langle X_t \rho_r(t) \rangle$
- Assume $g_r(t) \rightarrow g_r$ at long t
- For large $x = \mathbf{r} \cdot \hat{\mathbf{e}}_1$,

$$g_x \sim x^{1-d}$$



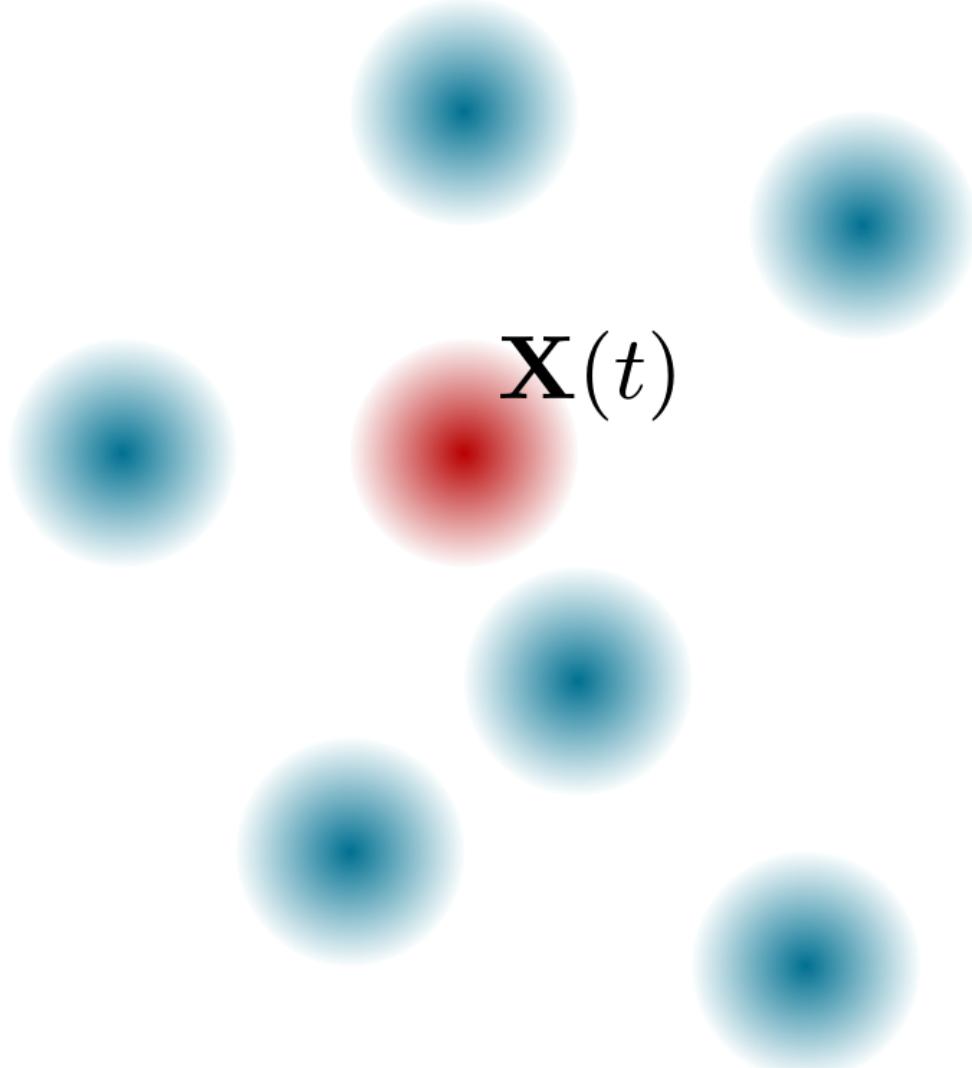
Soft interacting particles in d dimensions

- Overdamped Brownian particles, $i = 0, \dots, N$

$$\dot{\mathbf{X}}_i(t) = -\mu \sum_{j \neq i} \nabla_i U(\mathbf{X}_i(t) - \mathbf{X}_j(t)) + \boldsymbol{\eta}_i(t),$$

$$\langle \boldsymbol{\eta}_i(t)^T \boldsymbol{\eta}_j(t') \rangle = 2\mu T \delta_{ij} \delta(t - t') I_d$$

- “soft” potentials $U(\mathbf{x})$, e.g. Gaussian core
- **Tracer** $i = 0$ (omitted)
- Correlation profiles?



Coarse-grained dynamics with a tracer

- Dean-Kawasaki equation for $\rho(x, t) = \sum_{i=1}^N \delta(x - X_i(t))$,

$$\partial_t \mathbf{X}(t) = -\mu \nabla_{\mathbf{X}} \mathcal{F}[\rho, \mathbf{X}] + \boldsymbol{\eta}_0(t),$$

$$\partial_t \rho(\mathbf{x}, t) = \mu \nabla \cdot \left[\rho(\mathbf{x}, t) \nabla \frac{\delta \mathcal{F}}{\delta \rho(\mathbf{x}, t)} \right] + \nabla \cdot \left[\rho^{\frac{1}{2}}(\mathbf{x}, t) \boldsymbol{\xi}(\mathbf{x}, t) \right],$$

$$\mathcal{F}[\rho, \mathbf{X}] = T \int d\mathbf{x} \rho(\mathbf{x}) \log \left(\frac{\rho(\mathbf{x})}{\rho_0} \right) + \frac{1}{2} \int d\mathbf{x} d\mathbf{y} \rho(\mathbf{x}) U(\mathbf{x} - \mathbf{y}) \rho(\mathbf{y}) + \int d\mathbf{y} \rho(\mathbf{y}) U(\mathbf{y} - \mathbf{X})$$

- Linearize about $\rho(\mathbf{x}, t) = \rho_0 + \sqrt{\rho_0} \phi(\mathbf{x}, t)$, assuming $\phi/\sqrt{\rho_0} \ll 1$.



FIG. 1. David Dean and Kyozi Kawasaki, who are very happy that their equation is being used for the zillionth time.

Tracer statistics from correlation profiles

$$\partial_t \Psi(\lambda, t) = \frac{1}{2d\tau} \sum_{\mu=-d}^d \left(e^{\sigma\lambda \cdot \hat{\mathbf{e}}_\mu} - 1 \right) \left[1 - w_{\mathbf{e}_\mu}(\lambda, t) \right]$$

- $\rho(\mathbf{x}, t) = \rho_0 + \sqrt{\rho_0} \phi(\mathbf{x}, t) \rightarrow$ coupled eqs for $\mathbf{X}(t), \phi(\mathbf{x}, t)$ (linear in ϕ)
- How does $\Psi(\lambda, t) = \ln \langle e^{\lambda \cdot \mathbf{X}(t)} \rangle$ evolve?

$$\partial_t \Psi(\lambda, t) = \lambda^2 \mu T + \sqrt{\rho_0} \mu \lambda \cdot \int d^d x U(\mathbf{x}) \nabla_{\mathbf{x}} w(\mathbf{x}, \lambda, t),$$

with the profile

$$w(\mathbf{x}, \lambda, t) = \frac{\langle \phi(\mathbf{x} + \mathbf{X}(t), t) e^{\lambda \cdot \mathbf{X}(t)} \rangle}{\langle e^{\lambda \cdot \mathbf{X}(t)} \rangle} = \langle \phi(\mathbf{x} + \mathbf{X}(t), t) \rangle + \lambda \cdot \langle \mathbf{X}(t) \phi(\mathbf{x} + \mathbf{X}(t), t) \rangle + \mathcal{O}(\lambda^2)$$

average density profile

correlation profile $\mathbf{g}(\mathbf{x}, t)$

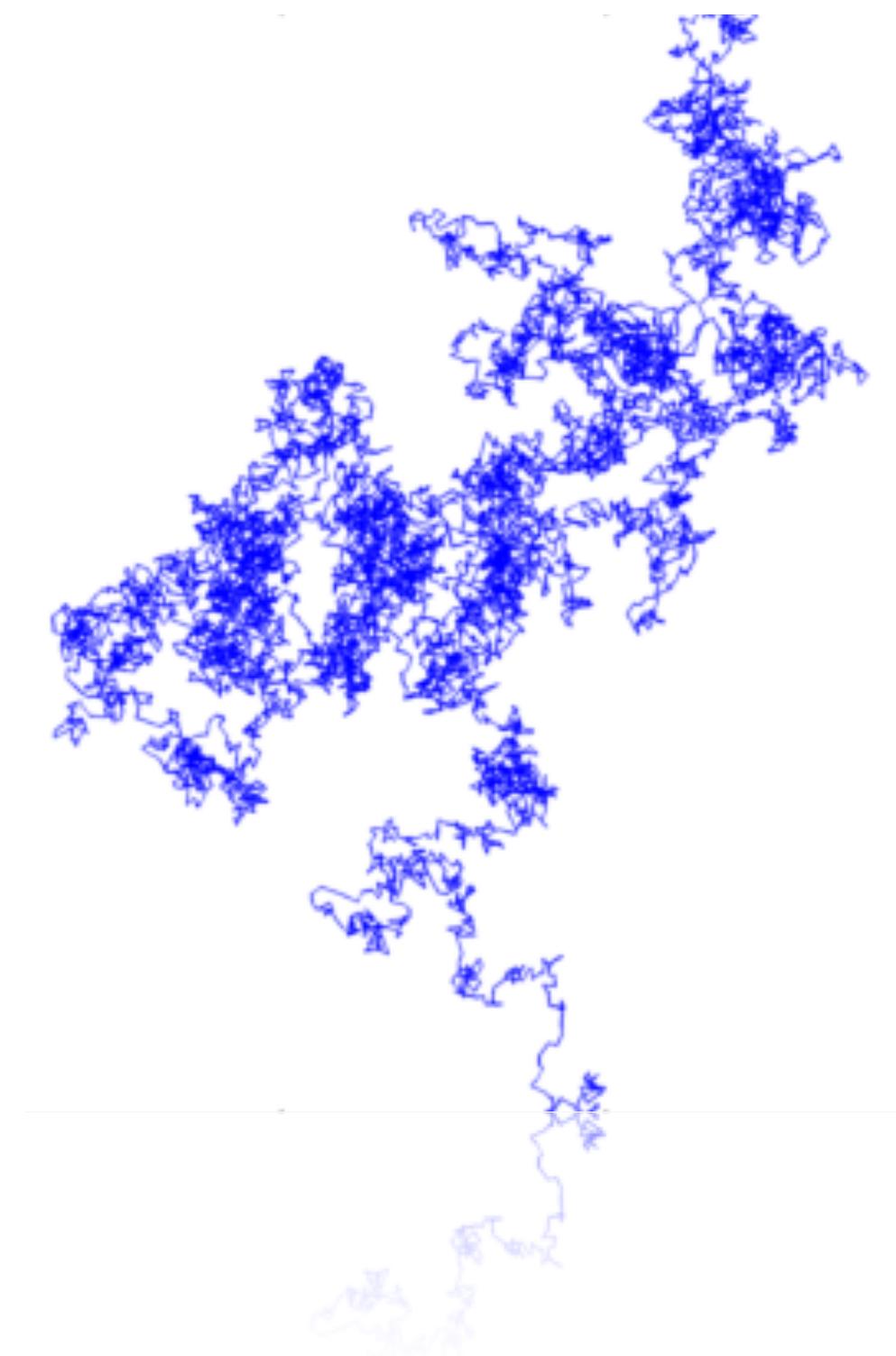
Beware of zero modes

- In the theory of fluids, stationary quantities are computed as

$$\langle \mathbf{X} \rho(\mathbf{x} + \mathbf{X}) \rangle \propto \int \mathcal{D}\rho \int d\mathbf{X} e^{-\frac{1}{T}\mathcal{F}[\rho, \mathbf{X}]} [\mathbf{X} \rho(\mathbf{x} + \mathbf{X})] = 0$$

upon defining $\rho'(\mathbf{x}) = \rho(\mathbf{x} + \mathbf{X})$.

- But thermodynamic quantities only depend on $|\mathbf{X}_i - \mathbf{X}_j|$, whereas $[\mathbf{X} \rho(\mathbf{x} + \mathbf{X})]$ depends also on **COM**
- Trivial zero! → Need to use EOM to predict stationary profiles.



Stationary correlation profile

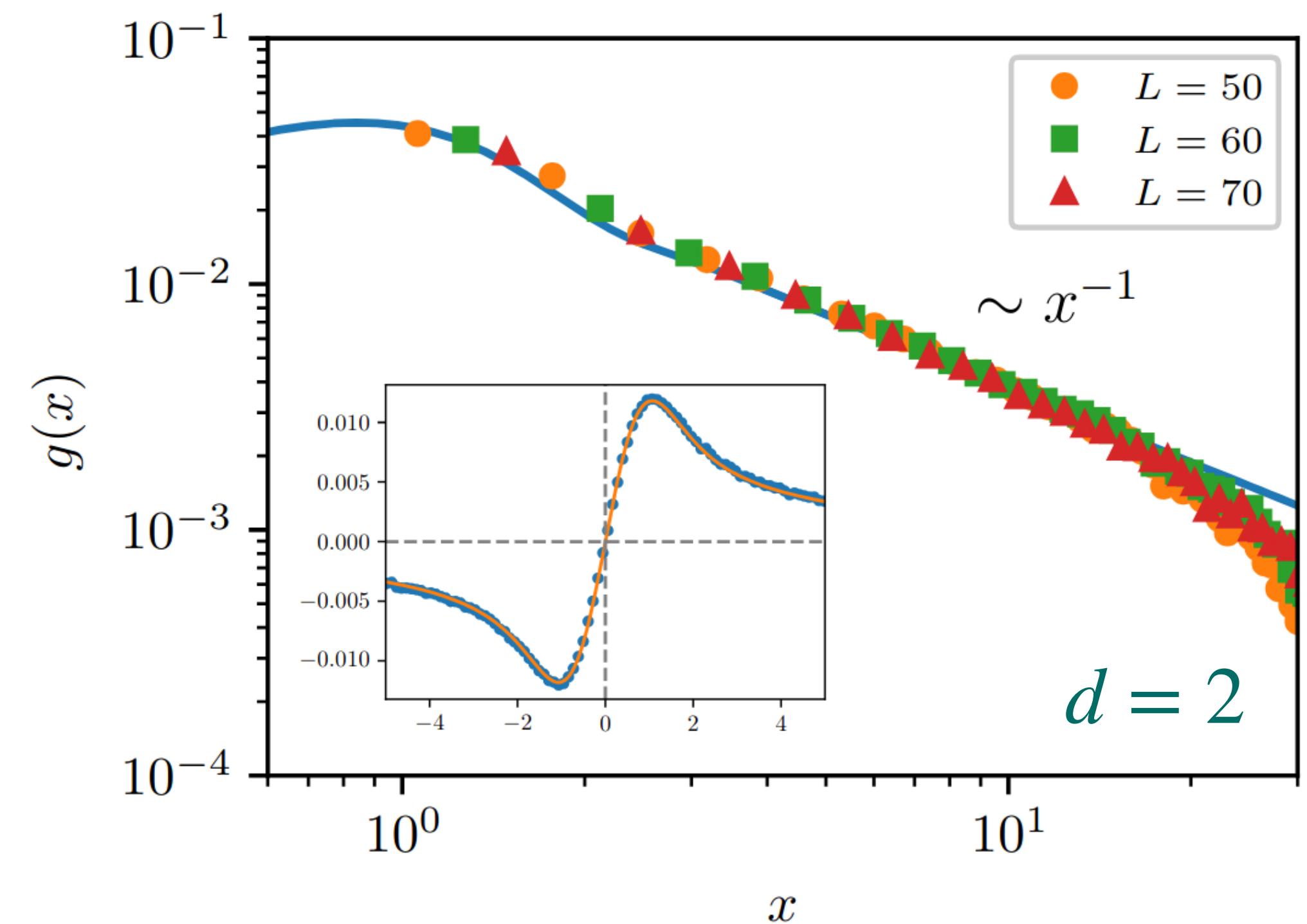
large-distance behavior

- From coupled EOM for $\partial_t \mathbf{X}(t)$ and $\partial_t \phi(\mathbf{x}, t)$, write one for

$$g(\mathbf{x}, t) = \hat{\mathbf{e}}_1 \cdot \langle \mathbf{X}(t) \phi(\mathbf{x} + \mathbf{X}(t), t) \rangle$$

- Compute $g(\mathbf{x})$ perturbatively
- At large distance $x = \mathbf{x} \cdot \hat{\mathbf{e}}_1$,

$$g(x) \sim x^{1-d}$$

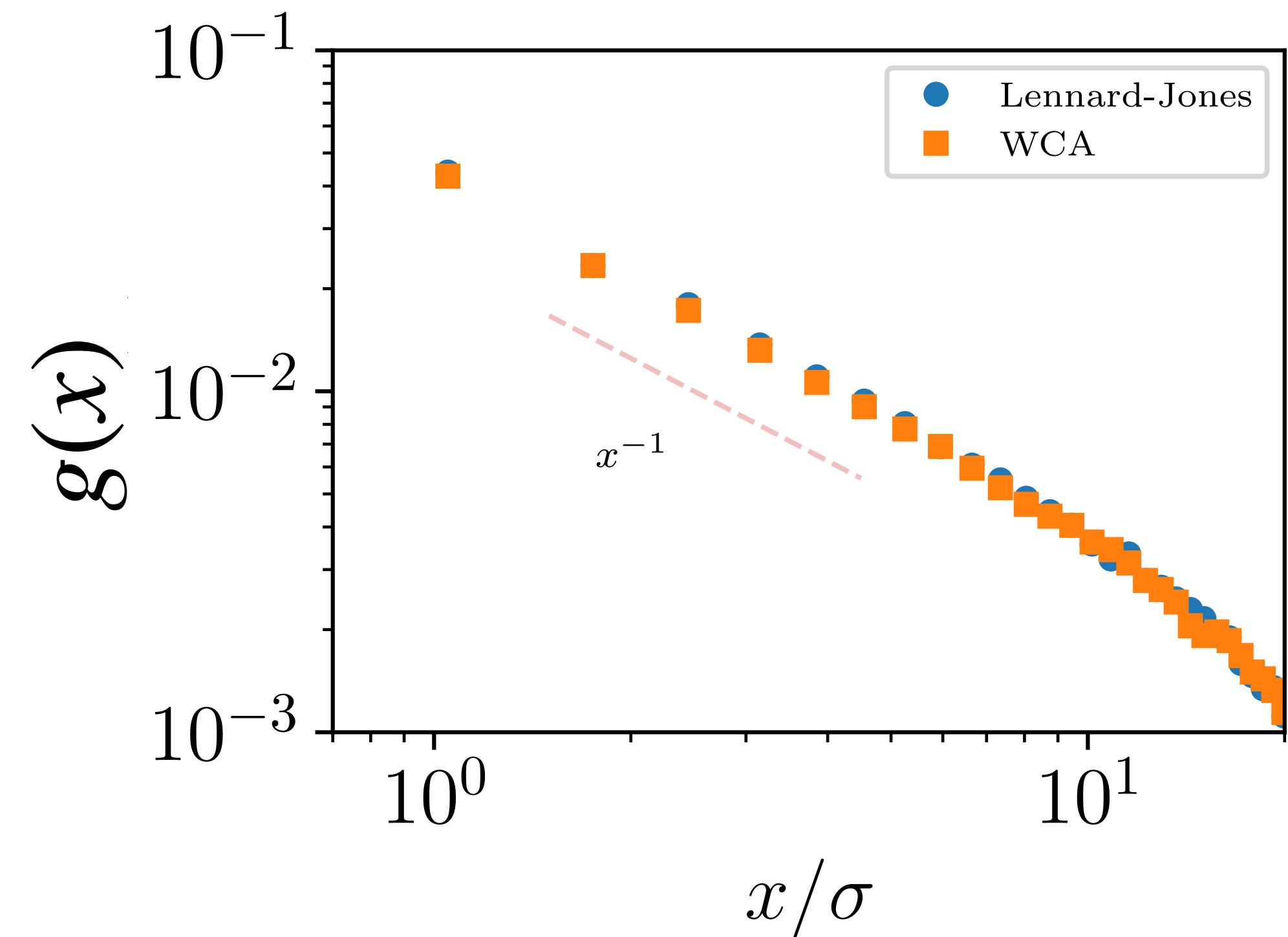
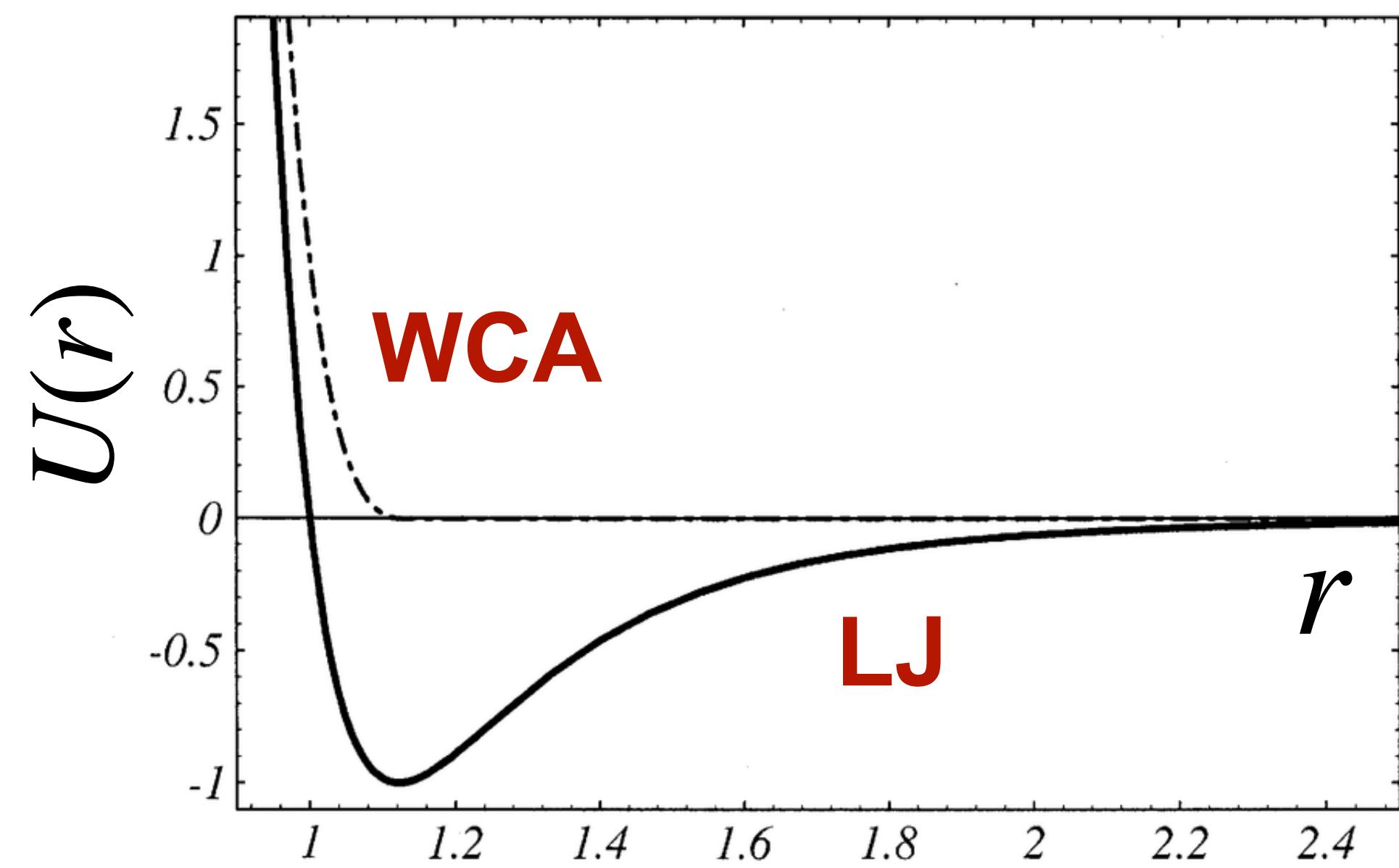


Lennard-Jones fluids

stationary correlation profile

Are short-distance details of interactions irrelevant for large-distance behaviour of the bath response?

- Strong repulsion beyond linearised D-K theory
- Simulate LJ and WCA suspensions



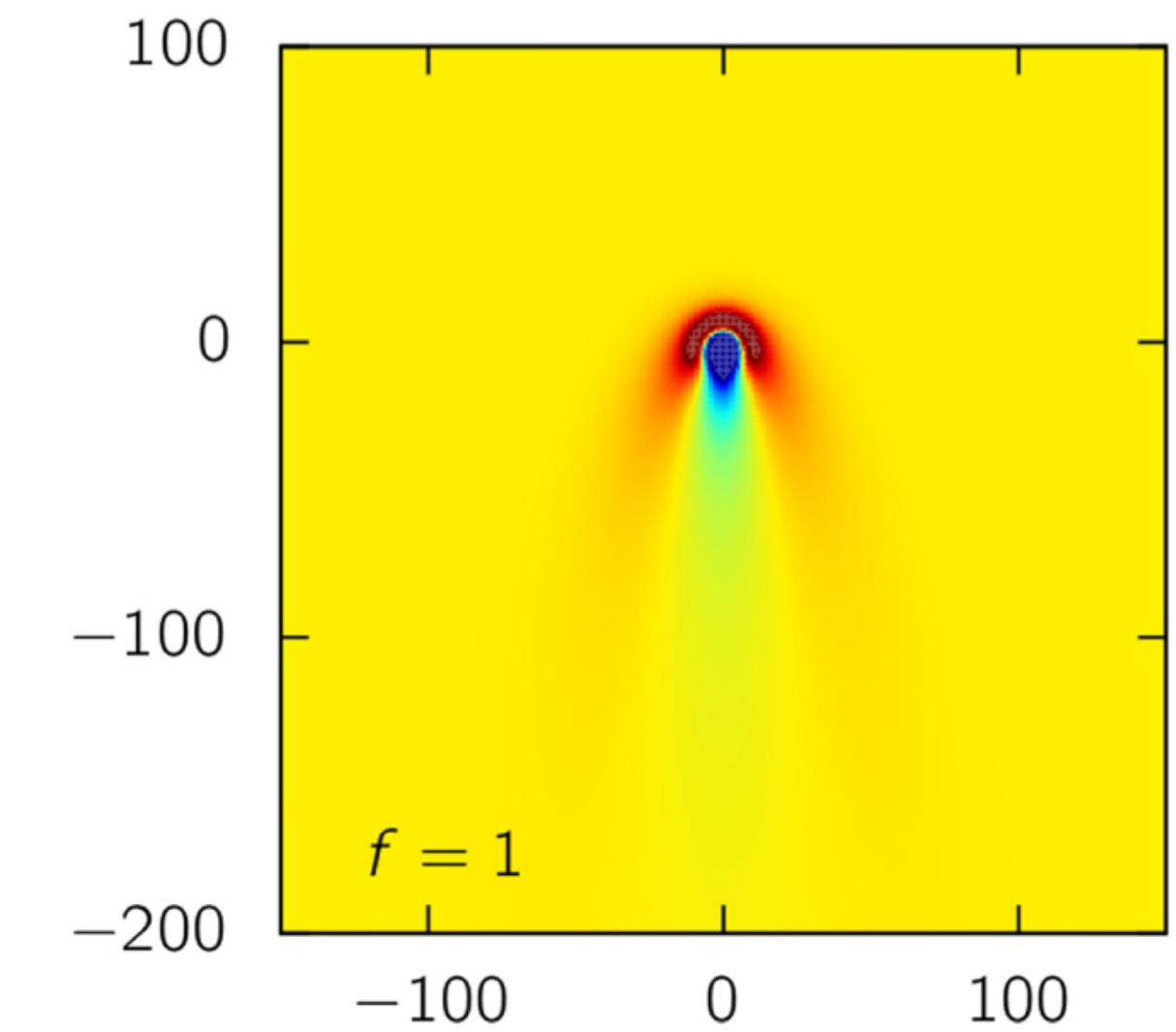
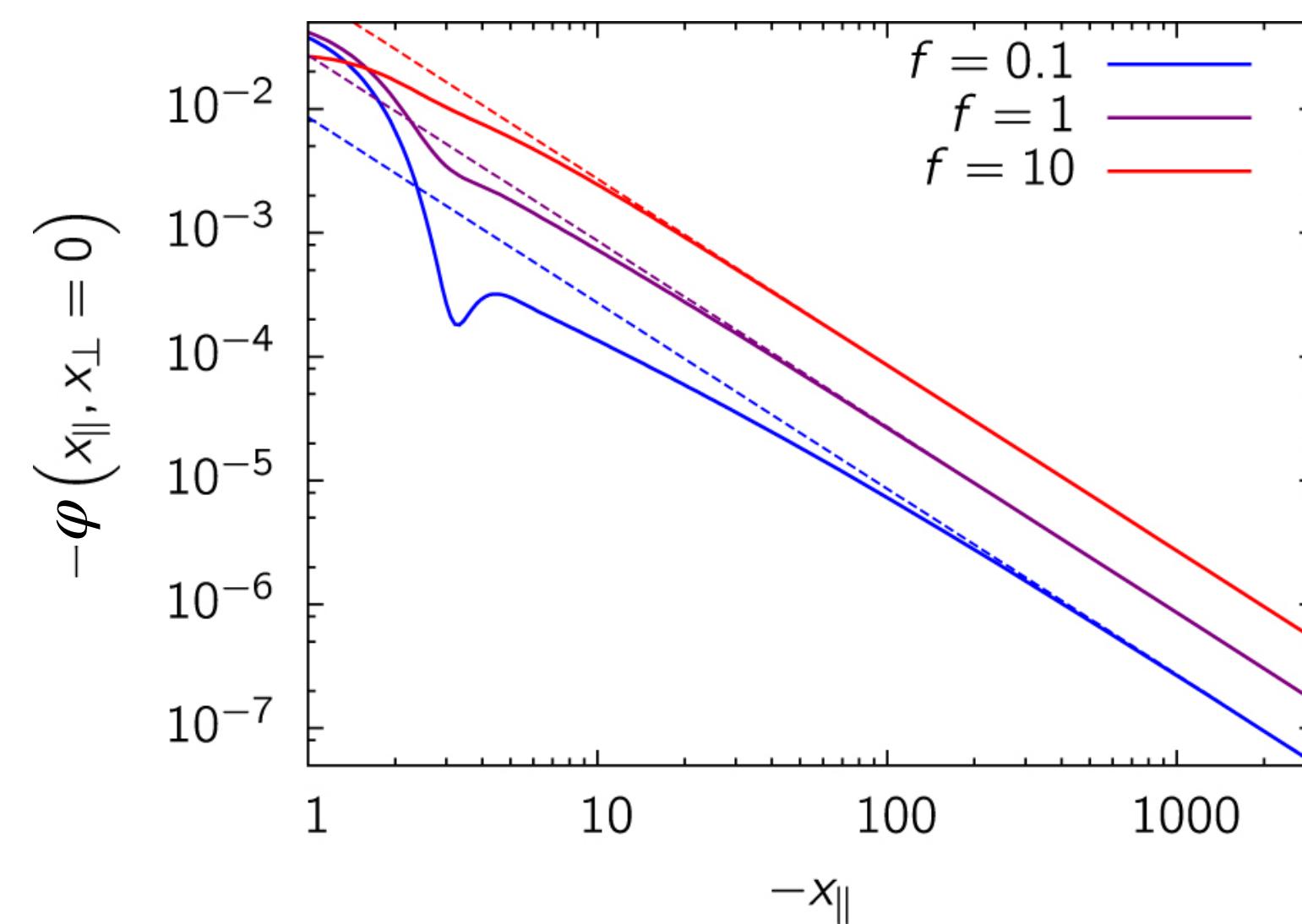
Average density profile under steady driving

- Stationary density profile in the frame of the tracer,

$$\varphi(\mathbf{x}, t) = \langle \phi(\mathbf{x} + \mathbf{X}(t), t) \rangle$$

- For $x_{||} \rightarrow -\infty$,

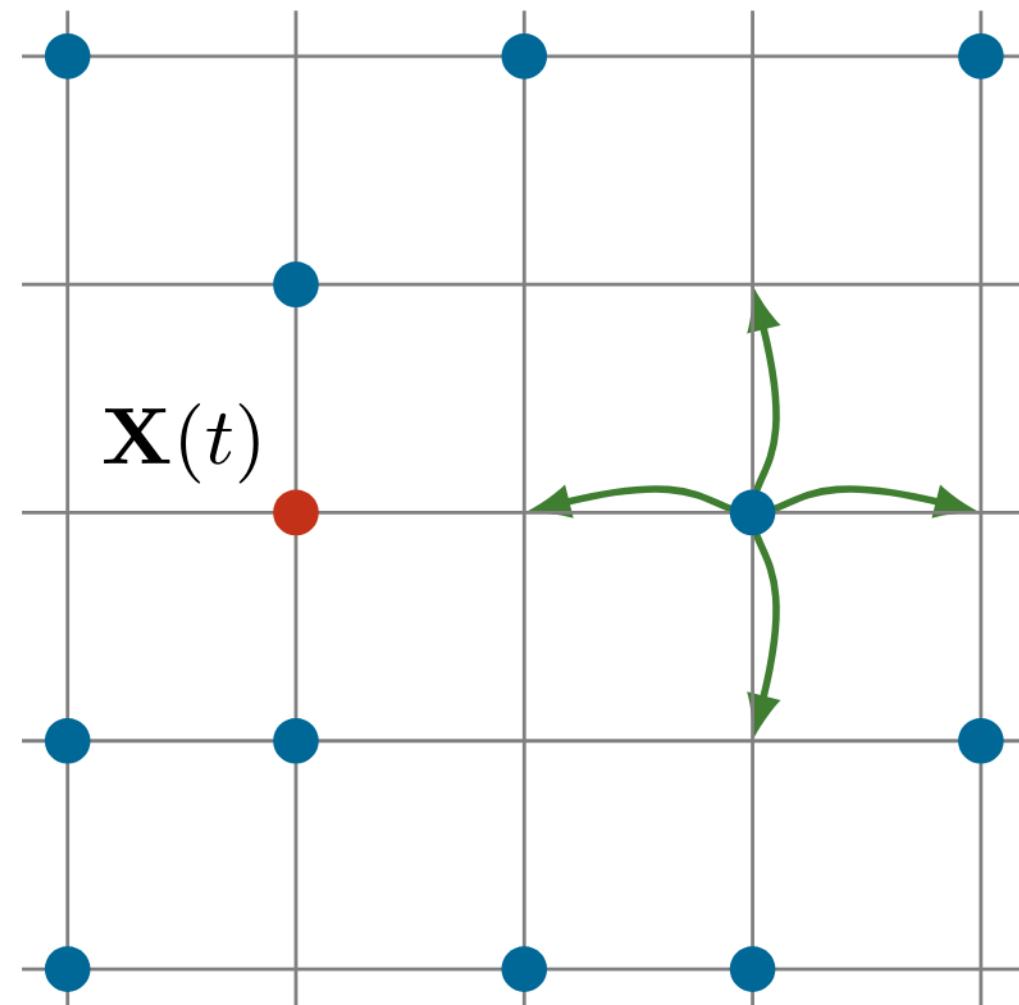
$$\varphi(x_{||}, \mathbf{x}_{\perp} = \mathbf{0}) \sim -|x_{||}|^{-\frac{1+d}{2}}$$



V. Démery et al., New J. Phys. **16** (2014) 053032

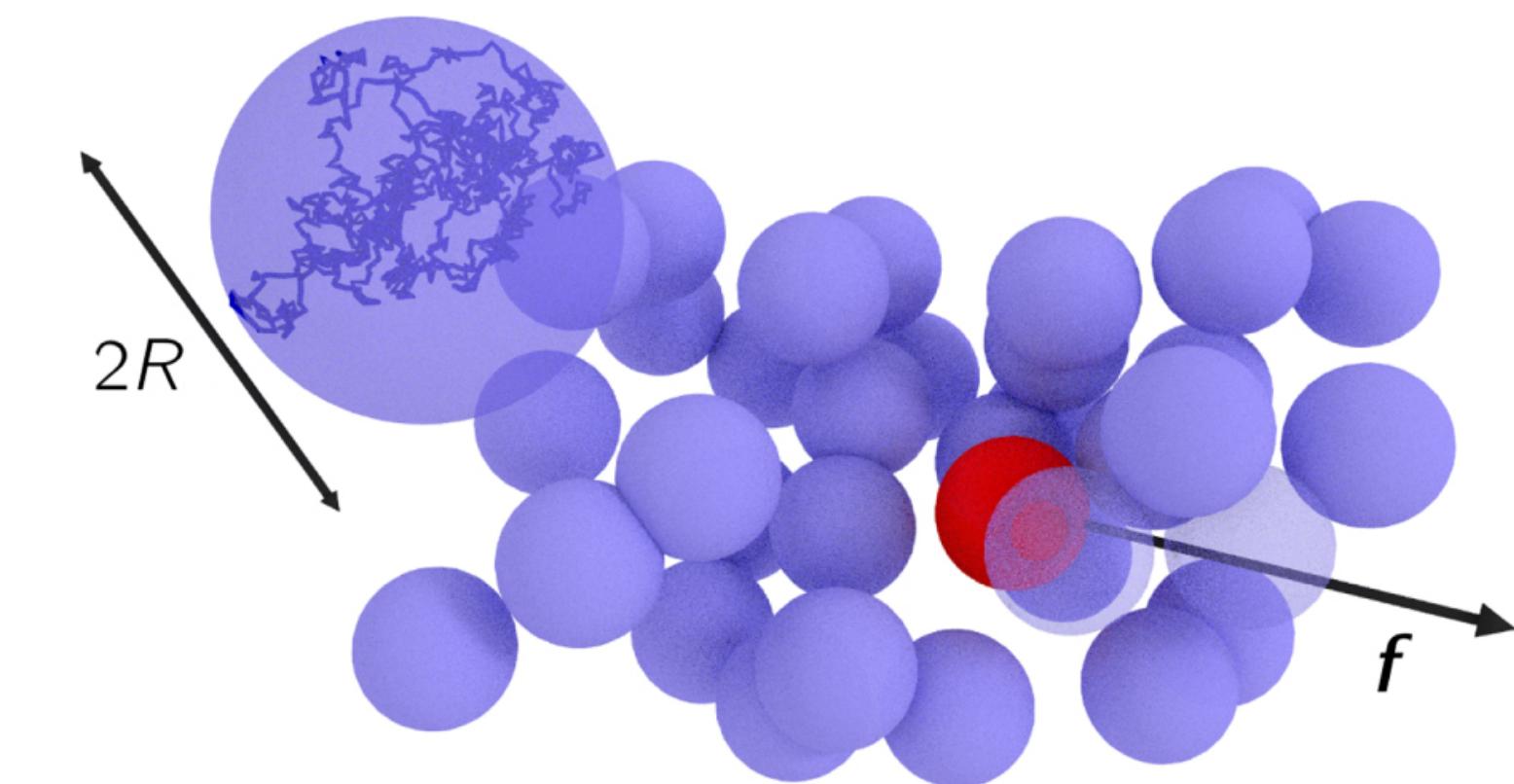
O. Bénichou et al., Phys. Rev. Lett. **84**, 511 (2000)

Universality in diffusive systems



Fluctuating hydrodynamics,
macroscopic fluctuation theory

large-distance behaviour
of tracer-bath correlations



??

Dean-Kawasaki theory

Summing up

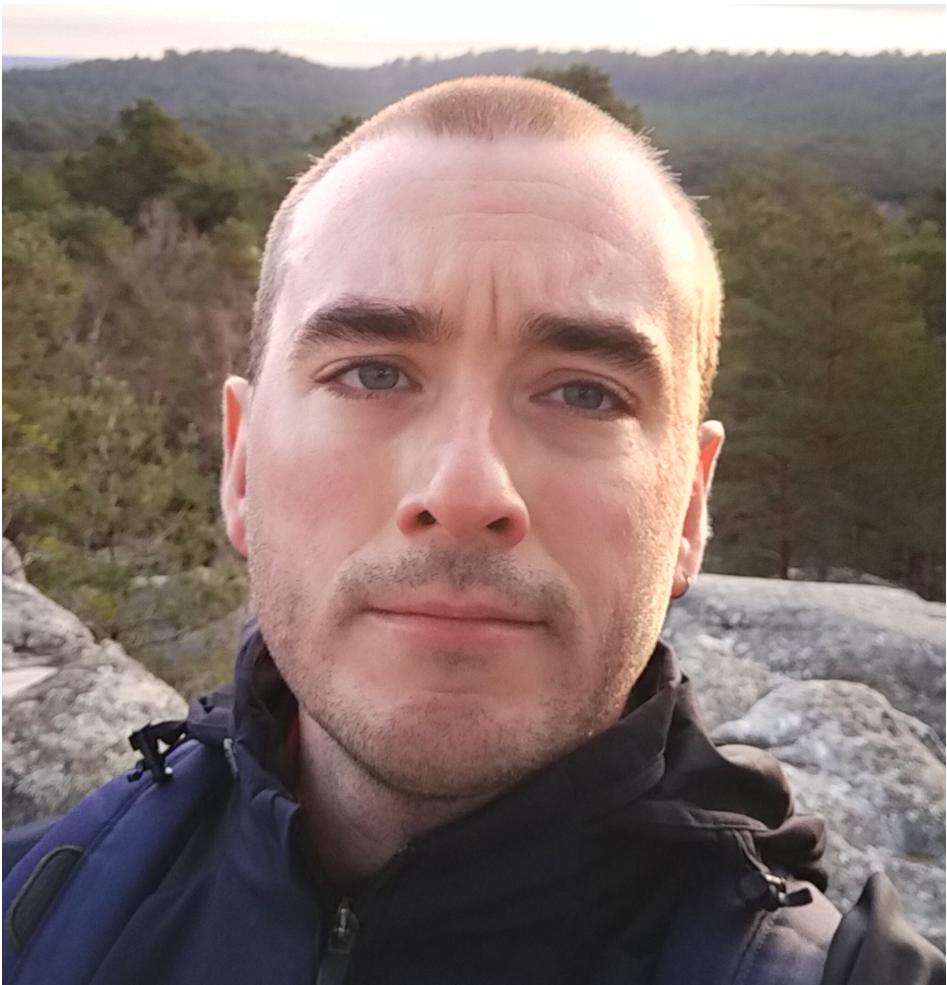
Spatial correlation profiles are worth checking out!

- $\langle Q_t \rho_r(t) \rangle$ gives info on **response** of the bath (e.g. role of **loops**),
- and gives access to **moments**, e.g. $\langle Q_t^2 \rangle$ on infinite lattices in $d > 1$.
- $\langle X_t \rho_r(t) \rangle \sim r^{1-d}$ for large r in a hard-core lattice gas,
- and for Brownian suspensions with both **weak/strong** repulsion.
- Why?

T. Berlitz, D. Venturelli, A. Grabsch, O. Bénichou, J. Stat. Mech. (2024) 113208

D. Venturelli, P. Illien, A. Grabsch, O. Bénichou, arXiv:2411.09326 (2024)

Thanks!



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