# Geometry-Induced Superdiffusion in Driven Crowded Systems

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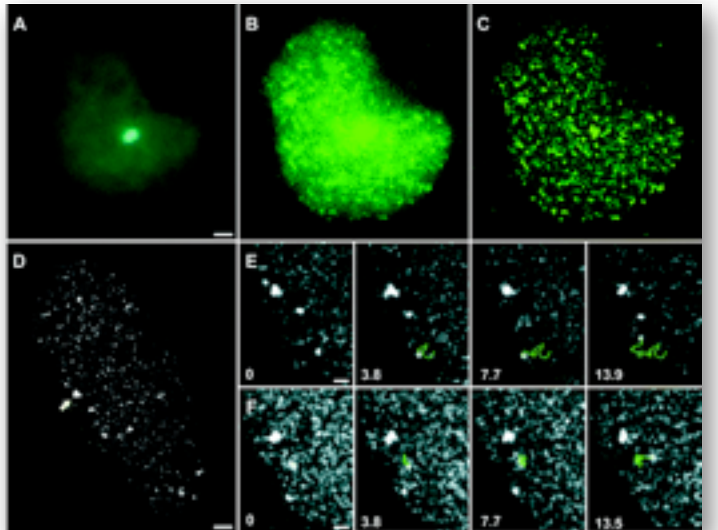
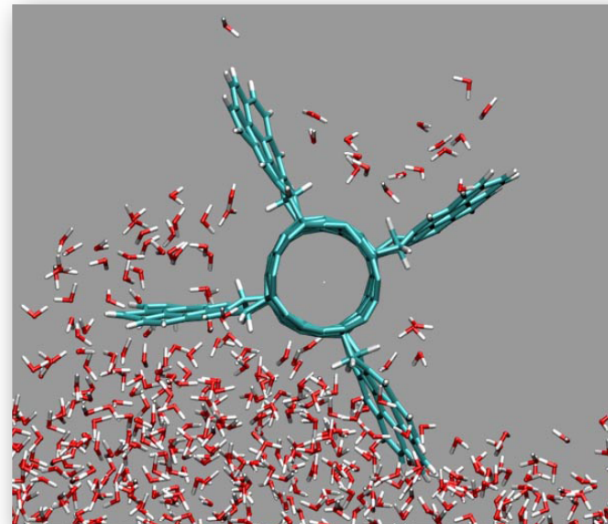
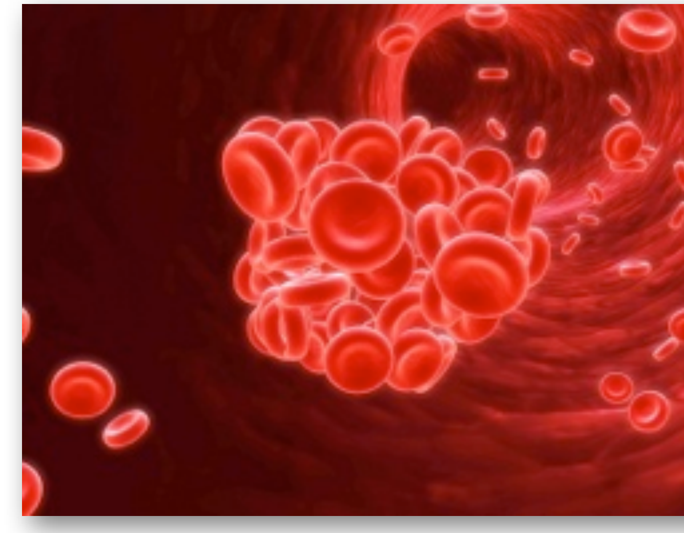
# Active Microrheology

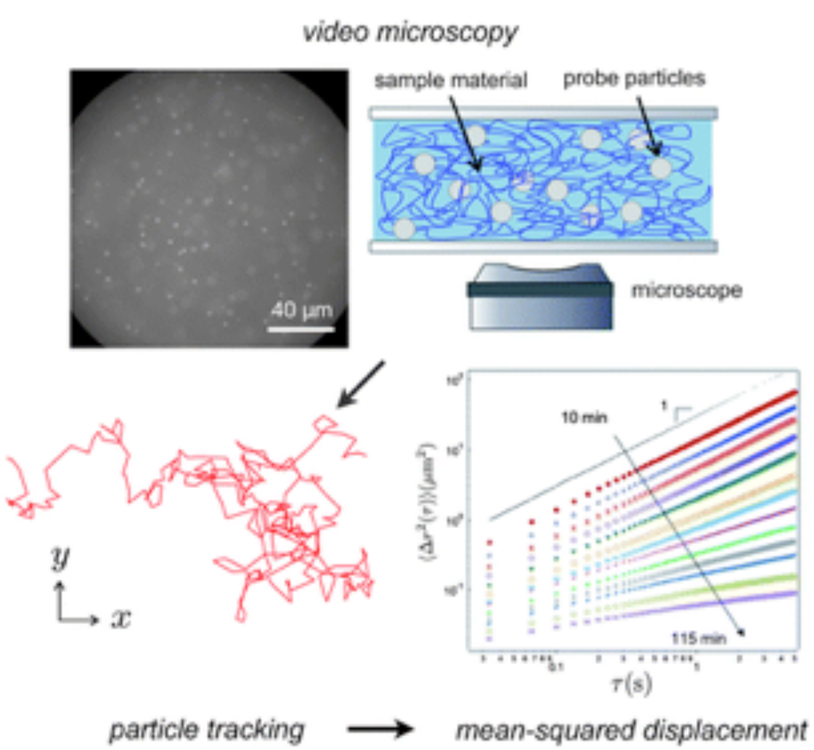


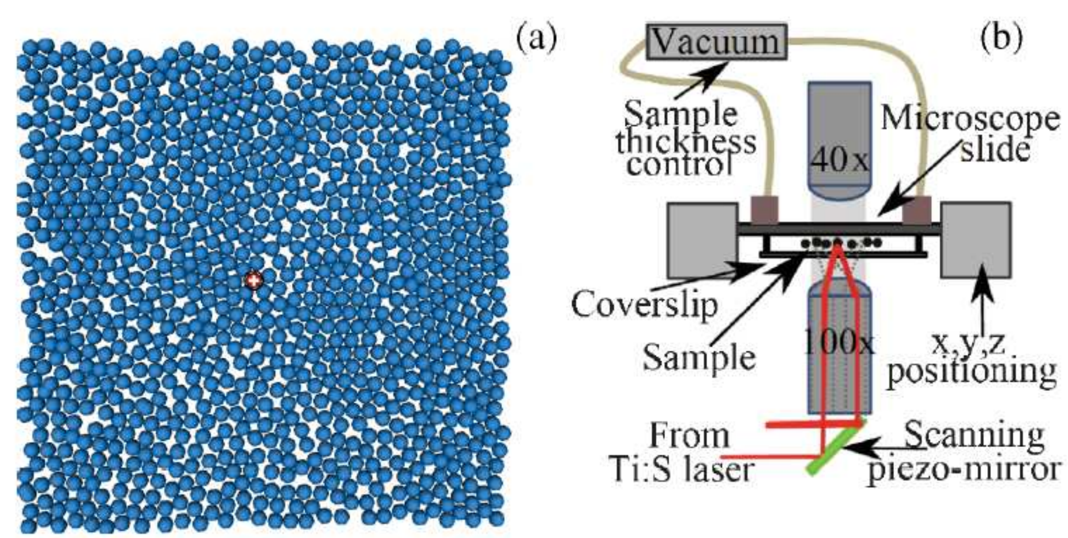


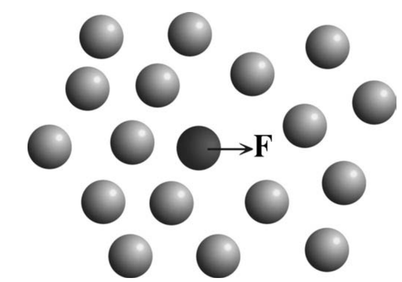


To probe the material response on micrometer length scales with microliter sample volumes.

*viscocity,*
*viscoelasticity,*
*structural changes*

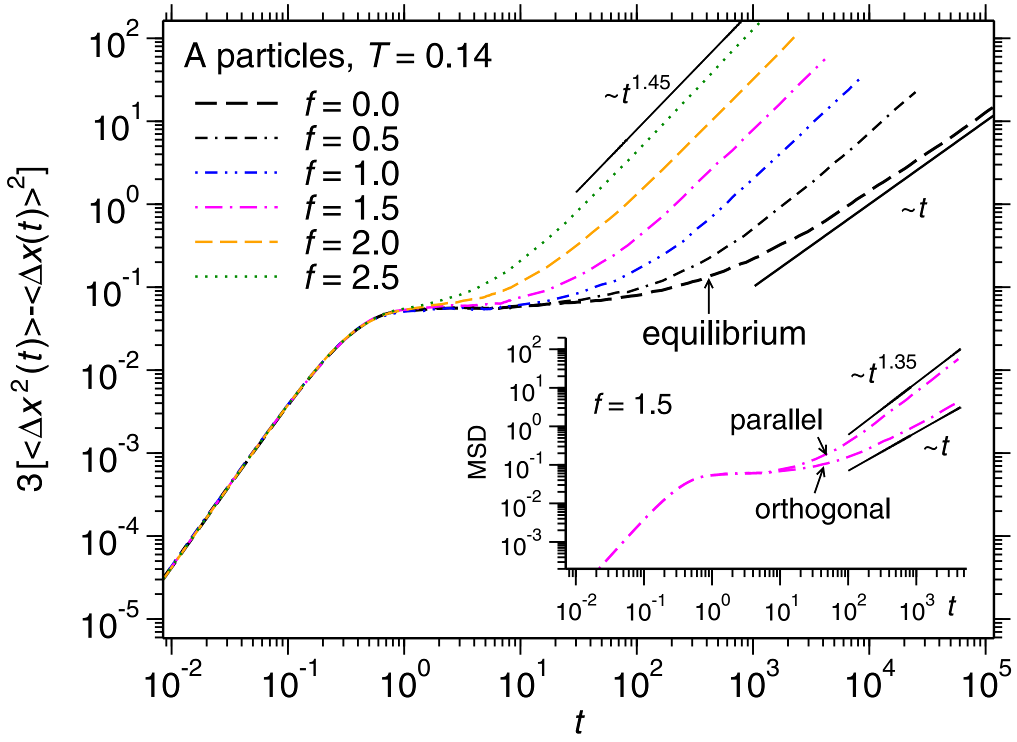


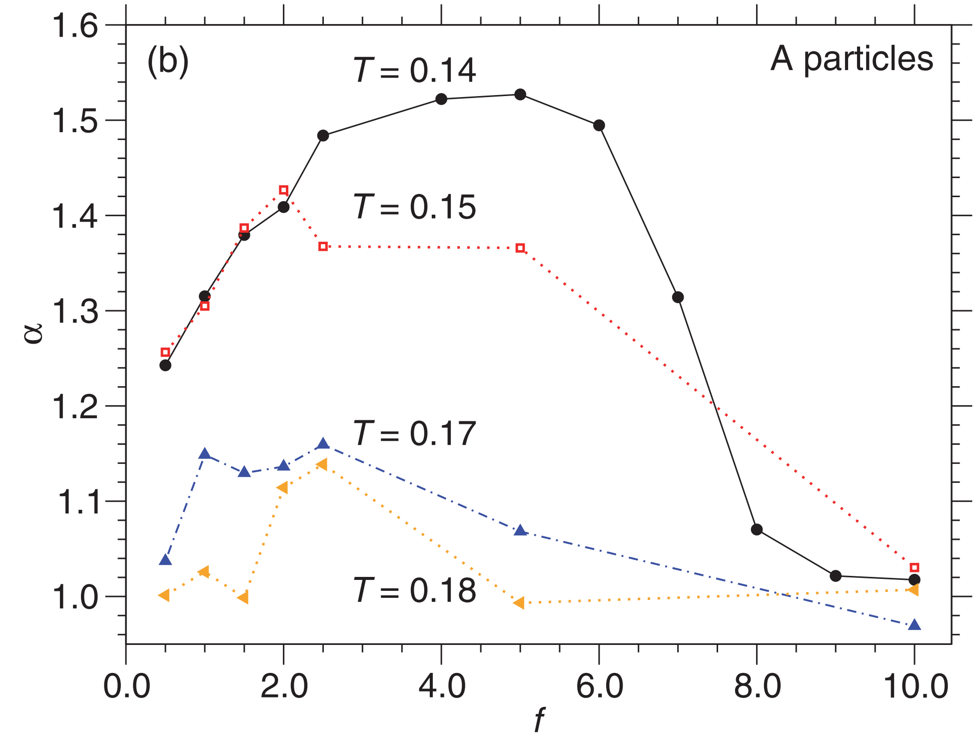




Is the standard setting for studying the Einstein relation and other LR fluctuation-dissipation relations.

# A glass forming binary Yukawa fluid




$$\sigma_{\parallel}^2 \equiv \langle X_t^2 \rangle - \langle X_t \rangle^2 \sim t^{\alpha}$$

$$\alpha \approx 1.3 - 1.6$$

Superdiffusive fluctuations

but only in the vicinity of the glass transition

Winter et al PRL **108** (2012) 028303

## Theoretical descriptions

Mode-coupling theory: unable to describe superdiffusion — Harrer et al J.Phys. Cond Mat 2012

Effective CTRW approach: parameters inferred only numerically — Schroer, Heuer et al PRL 2013

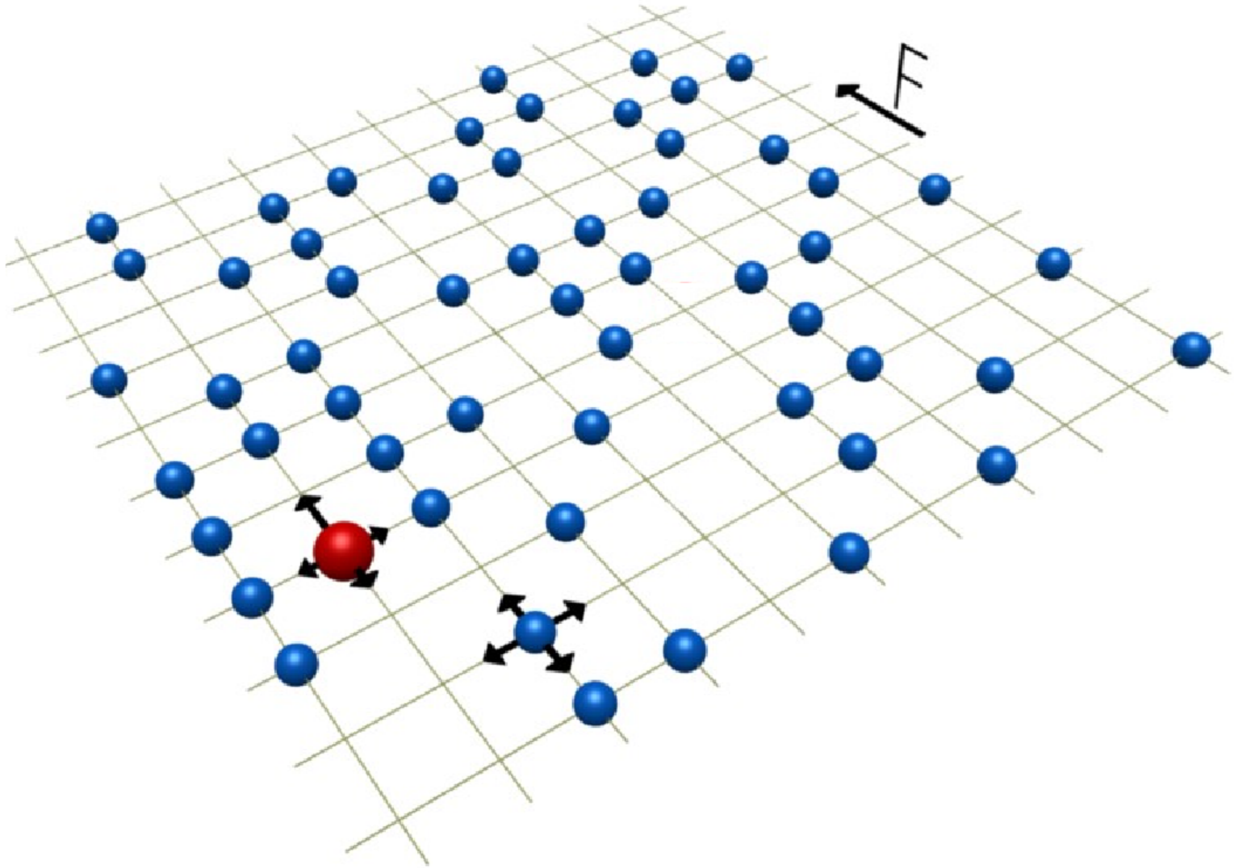
There is no clear understanding of superdiffusion

- is it related to glass transition?
- is it transient or the ultimate regime?

# Our model

Biased intruder (TP) in a gas of unbiased hard-core particles on a $d$-dimensional hypercubic lattice

- We consider a square lattice of $L_x \times L_y$ sites, of unit spacing, with P.B.C and populated with hard-core particles.
- Each site can be either empty or occupied by at most one particle.
- The system evolves in discrete time $n$ and particles move randomly.
- One particle, *the intruder*, is subject to a constant force $\mathbf{F}$




Simple Exclusion Process:
an ASEP in a sea of SEP's

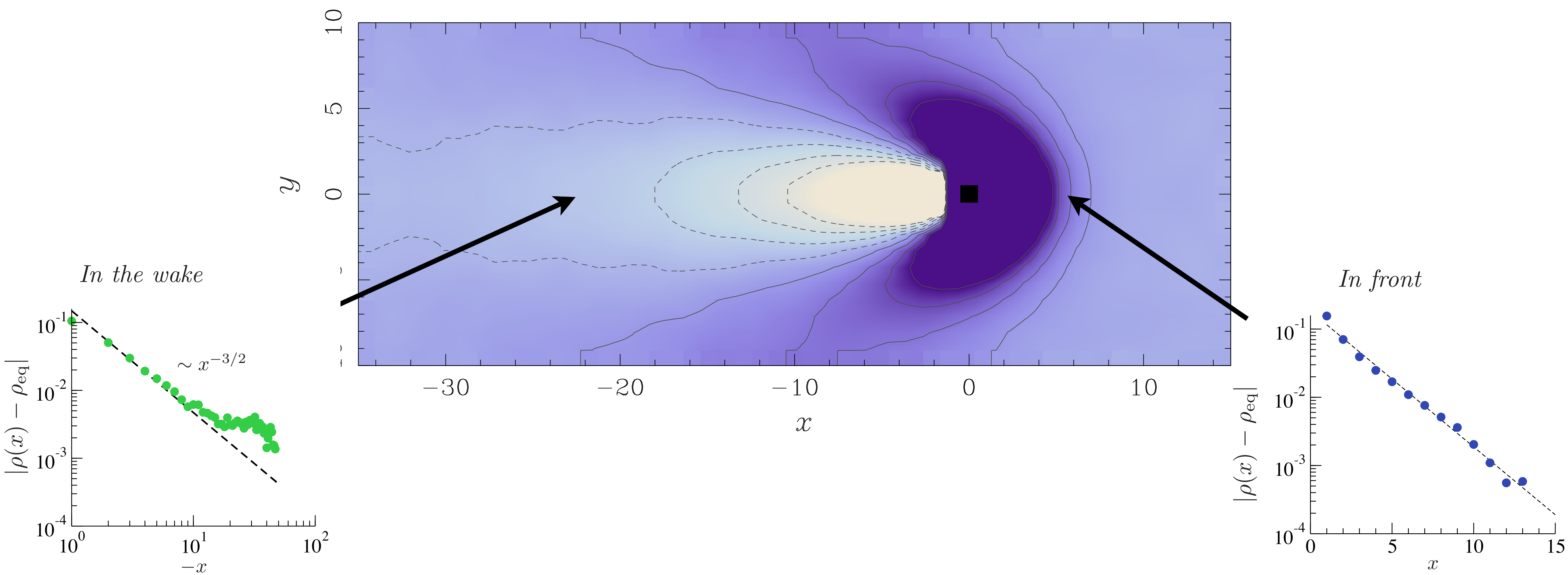
• Bath particles move in either direction with equal jump probability $1/4$.
• The intruder moves in direction $\mathbf{e}_\nu$ with probability

$$p_\nu = Z^{-1} e^{\frac{\beta}{2}\mathbf{F}\cdot\mathbf{e}_\nu} \; ,$$

where $Z = 2(1 + \cosh(\beta\sigma F/2))$ and $\beta$ is the inverse temperature.

# Nonequilibrium inhomogeneity

As the force increases structural changes around the TP are observed

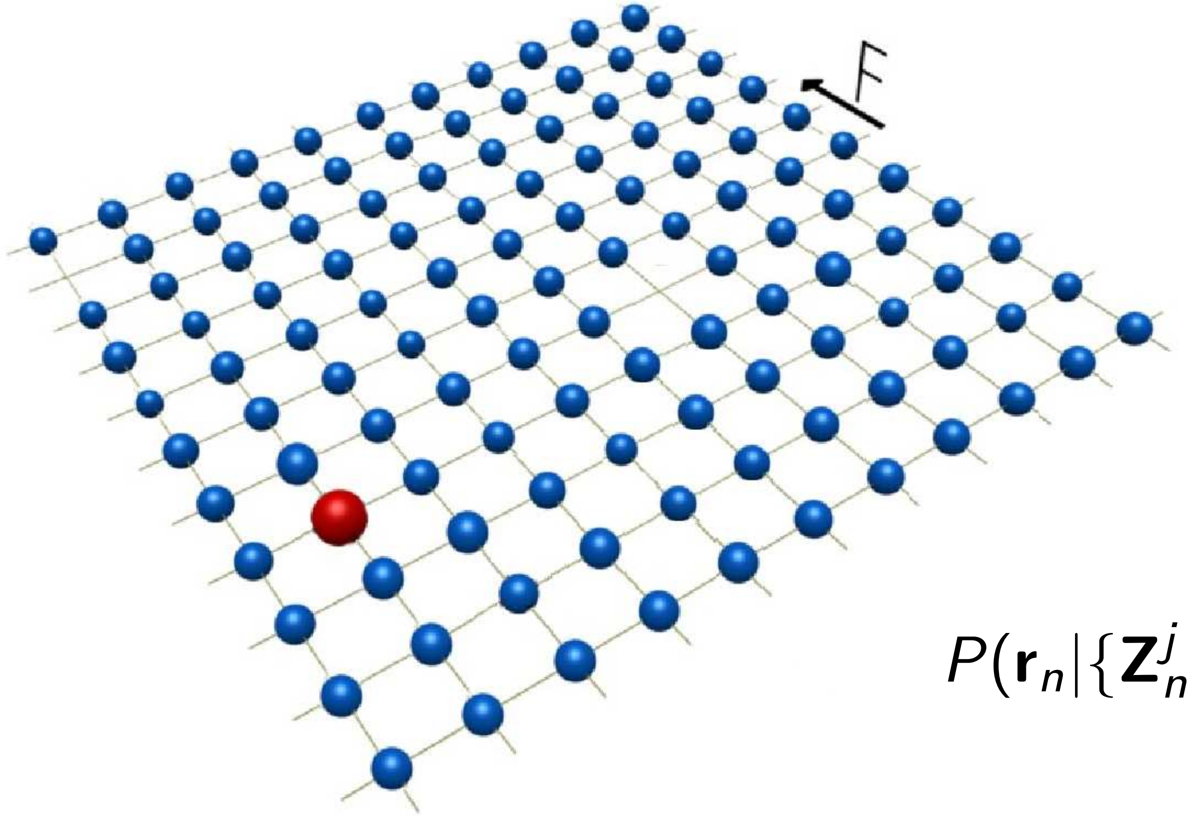



The medium remembers the passage of the intruder on very large spatial and temporal scales

observed in colloidal suspensions, monolayers of vibrated grains and in glass systems.

Mejia-Monasterio & Oshanin Soft matter **7** (2011) 993

# Overcrowding. Theoretical approach




For a given configuration of the vacancies $\{Z^j_n\}$, the probability of finding the TP at position $r_n$ at discrete time $n$ is

$$P(\mathbf{r}_n|\{\mathbf{Z}^j_n\}) = \sum_{\mathbf{r}^1_n}\cdots\sum_{\mathbf{r}^M_n}\delta(\mathbf{r}_n, \mathbf{r}^1_n + \cdots + \mathbf{r}^M_n)P(\mathbf{r}^1_n, \ldots, \mathbf{r}^M_n|\{\mathbf{Z}^j_n\})$$

In the lowest order in the vacancy density $\rho_0$ the interactions with different vacancies can be considered independent and

$$P(\mathbf{r}^1_n, \ldots, \mathbf{r}^M_n|\{\mathbf{Z}^j_n\}) \simeq \prod_{j=1}^{M} P(\mathbf{r}_n|Z^j_n)$$

The problem reduces to that of $M$ single vacancies with corrections smaller than $\mathcal{O}(\rho_0)$.

The interactions with a single vacancy is written in terms of first return probabilities by summing over all jump moments and jump directions.

Brummelhuis & Hilhorst, JSP (1988)
Bénichou & Oshanin PRE (2002)

# Theoretical approach

In the limit of large $n$ and low vacancy density $\rho_0$

$$P(\mathbf{R}_n) \simeq \frac{1}{4\pi^2} \int_{-\pi}^{\pi} \mathrm{d}\mathbf{k} \, \exp\left(-\mathrm{i}\left(\mathbf{k} \cdot \mathbf{R}_n\right) - \rho_0 \, \Omega_n(\mathbf{k})\right)$$

$$\Omega_z(\mathbf{k}) = \sum_{n=0}^{\infty} \Omega_n(\mathbf{k}) \, z^n$$

$$\sim \frac{1}{(1-z)} \frac{\Phi(\mathbf{k})}{1 - z + \Phi(\mathbf{k})/\chi_z}$$

$$\chi_z \sim -\frac{\pi}{(1-z)\ln(1-z)}$$

$$\Phi(\mathbf{k}) = -ia_0 k_x + a_1 k_x^2/2 + a_2 k_y^2/2$$

$$a_0 = \frac{\sinh(\beta F/2)}{(2\pi - 3)\cosh(\beta F/2) + 1},$$

$$a_1 = \frac{\cosh(\beta F/2)}{(2\pi - 3)\cosh(\beta F/2) + 1},$$

$$a_2 = \frac{1}{\cosh(\beta F/2) + 2\pi - 3}.$$

is the leading asymptotic term of the generating function of the mean number of new sites visited on the $n$-th time step.

Bénichou, Mejia-Monasterio, Oshanin PRE **87** (2013) 020103
Bénichou et al JSTAT P05008 (2013)

# Convergence to a gaussian distribution

Non-trivial behavior because the system is not in equilibrium and density profiles of the host medium particles around the TP are highly asymmetric!

In the limit $\rho_0 \to 0$

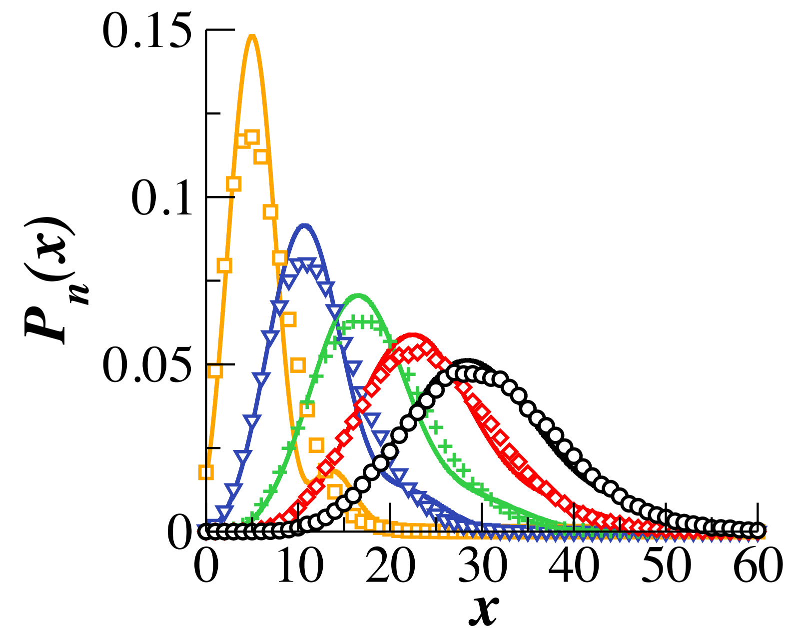
$$P_n(x) = (2\pi\sigma_x^2)^{-1/2} \mathrm{e}^{-\frac{(x-vn)^2}{2\sigma_x^2}} \left(1 + A/n + \ldots\right) \ ,$$

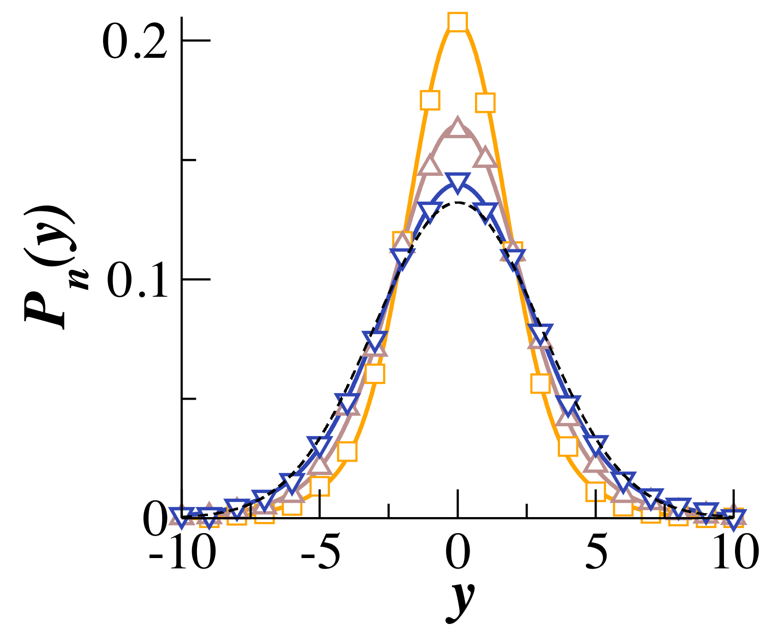
$$P_n(y) = (2\pi\sigma_y^2)^{-1/2} \mathrm{e}^{-\frac{y^2}{2\sigma_y^2}} \left(1 + B \ln n/n + \ldots\right) \ ,$$

$$v \sim \rho_0 \, a_0 \ ,$$

$$\sigma_x^2 \sim \rho_0 \left( a_1 + \frac{2a_0^2}{\pi} (\gamma - 1) + \frac{2a_0^2}{\pi} \ln(n) \right) n \ ,$$

$$\sigma_y^2 \sim \rho_0 \, a_2 n \ , \qquad a_i \equiv a_i(\beta F)$$



# Drift velocity of the TP

The ultimate velocity of the TP is $v = \rho_0 \cdot A_0(F)$

$$_0(\quad) = \frac{\sinh(\beta \quad 2)}{\left(1 + \frac{2d\alpha}{2d-\alpha}\right)\cosh(\beta \quad 2) + d + 1}$$

$$\alpha = \lim_{\xi \to 1^-} \left(P(0|0;\xi) - P(2\hat{e}_1|0;\xi)\right)$$

a nonlinear force-velocity relation and for

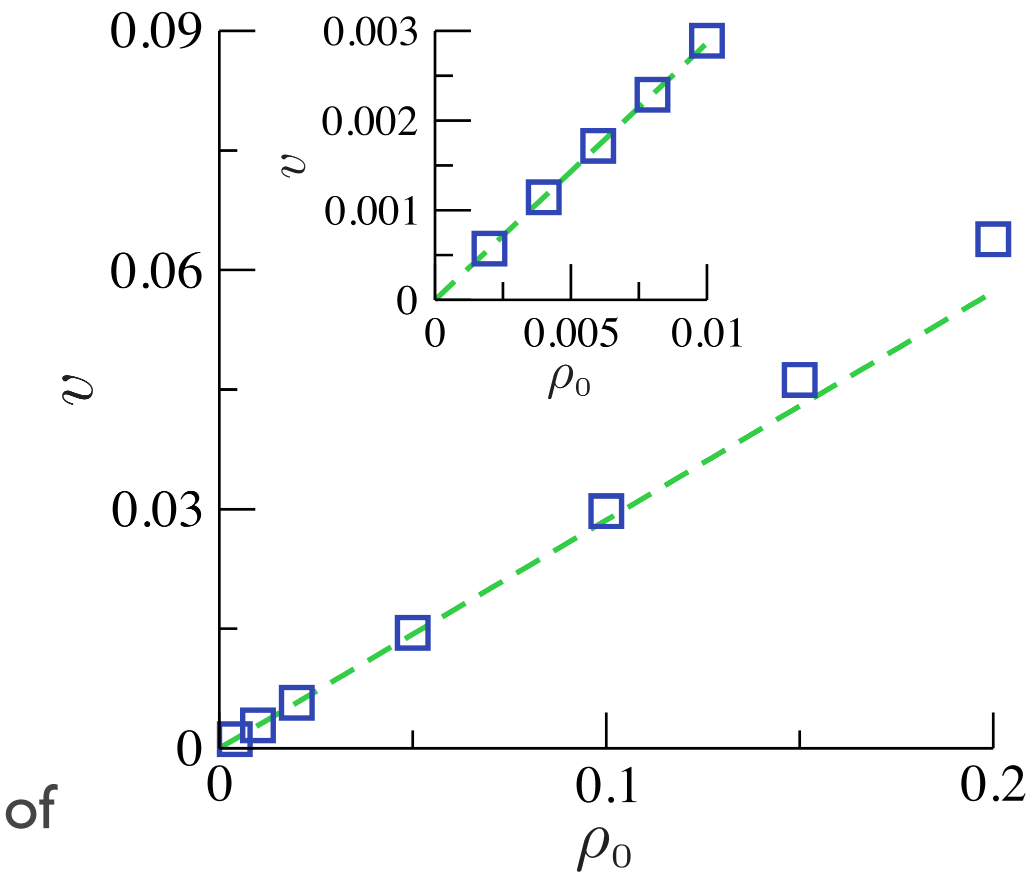
$$\beta F \ll 1\,, \quad A_0(F) \approx F$$

$P(\mathbf{r}, (r)_0; \xi)$ is the generating function of the propagator of a symmetric RW on a *d*-dimensional hypercubic lattice

Note that the velocity vanishes for single-files as it should (since $\alpha = 2$).

At equilibrium ($F = 0$), the TP's diffusion coefficient is

$$D = \frac{1}{2d}\left(\frac{2d-\alpha}{2d+\alpha}\right)\rho_0$$




Exact results to $\mathcal{O}(\rho_0)$.

Bénichou, Illien, Bodrova, Chakraborty, Law, M-M, Oshanin, Voituriez PRL **111** (2013) 260601

# Superdiffusive broadening of the fluctuations

Exact results for the variance of the TP's position at intermediate times ( $\rho_0^2 t \ll 1$ ).

Along the direction of the bias the variance is

$$\sigma_x^2 \approx \rho_0 A_0(F) t + 2\rho_0 A_0^2(F)\theta(t)$$

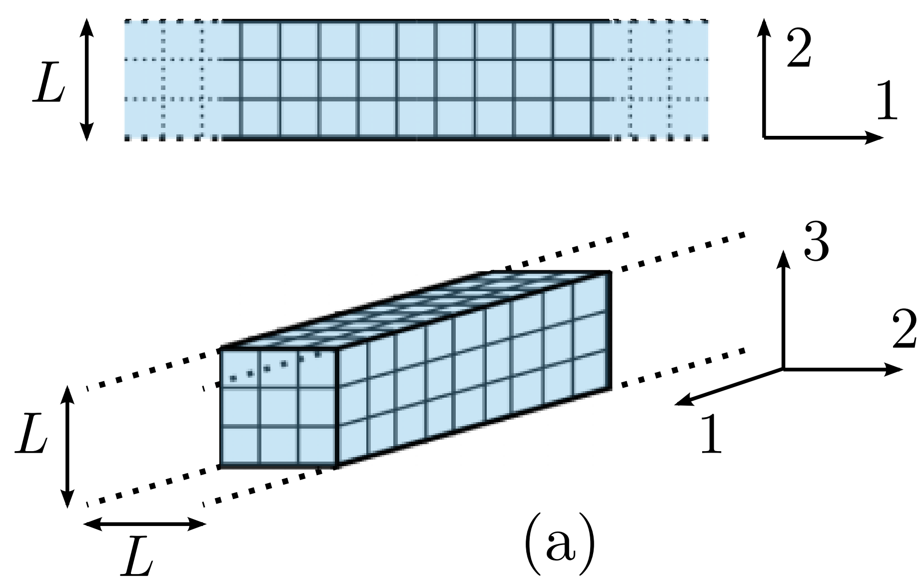
- First term is always "diffusive" and is, in fact, responsible for the validity of the Einstein relation ($A_0(F) \approx F$ for small $F$).
- Second term is "superdiffusive" in confined geometries, which signifies that is not the real spatial dimension $d$ that matters, but the effective one.
- The coefficient in the second term is proportional to $F^2$, which signifies that the superdiffive behavior emerges "beyond the linear response".
- Note that $A_0(F) = 0$ for single file case. It is a special singular.

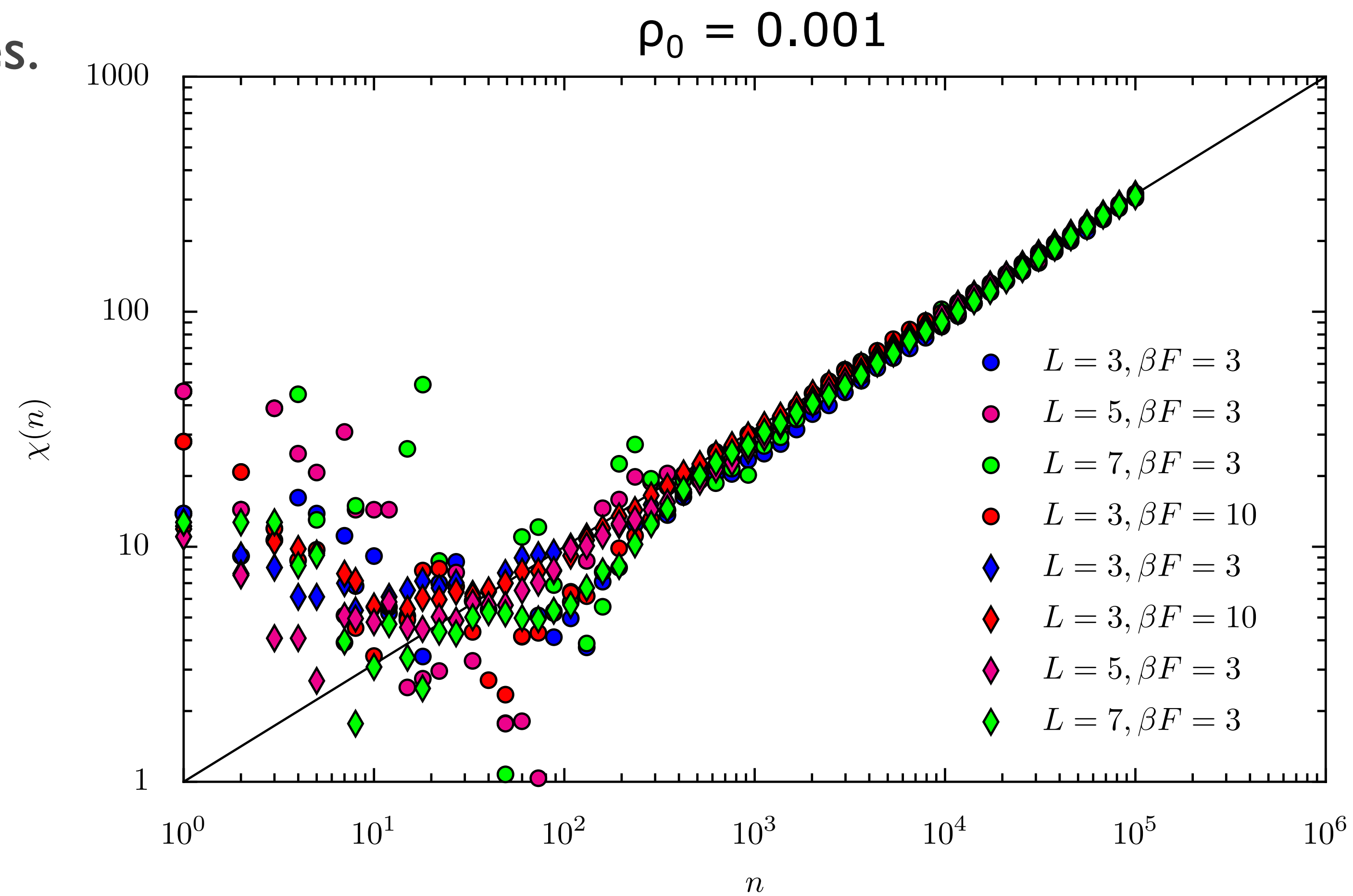
| *geometry* | $\theta(t)$ | |
|---|---|---|
| infinite 3D | $t$ | diffusive |
| 3D capillary | $t^{3/2}/L^2$ | strong superdiffusion |
| 3D slit pore | $t\ln(t)/L$ | weak superdiffusion |
| infinite 2D | $t\ln(t)$ | weak superdiffusion |
| 2D stripe | $t^{3/2}/L$ | strong superdiffusion |

The variance in the perpendicular direction to the bias is always diffusive.

# Superdiffusive broadening of the fluctuations

Biased intruders in dense hard-core lattice gases, confined in 2D stripes and 3D capillaries.




For 3D capillaries of cross-section *L* x *L*

$$\chi(t) = \sigma^2 L^2 / (t A_0^2(F))$$

For *2D* stripes of width *L*

$$\chi(t) = \sigma^2 L/(t A_0^2(F))$$

Diamonds correspond to stripes and circles to capillaries.
The solid line stands for $\sim t^{3/2}$

*data collapses*

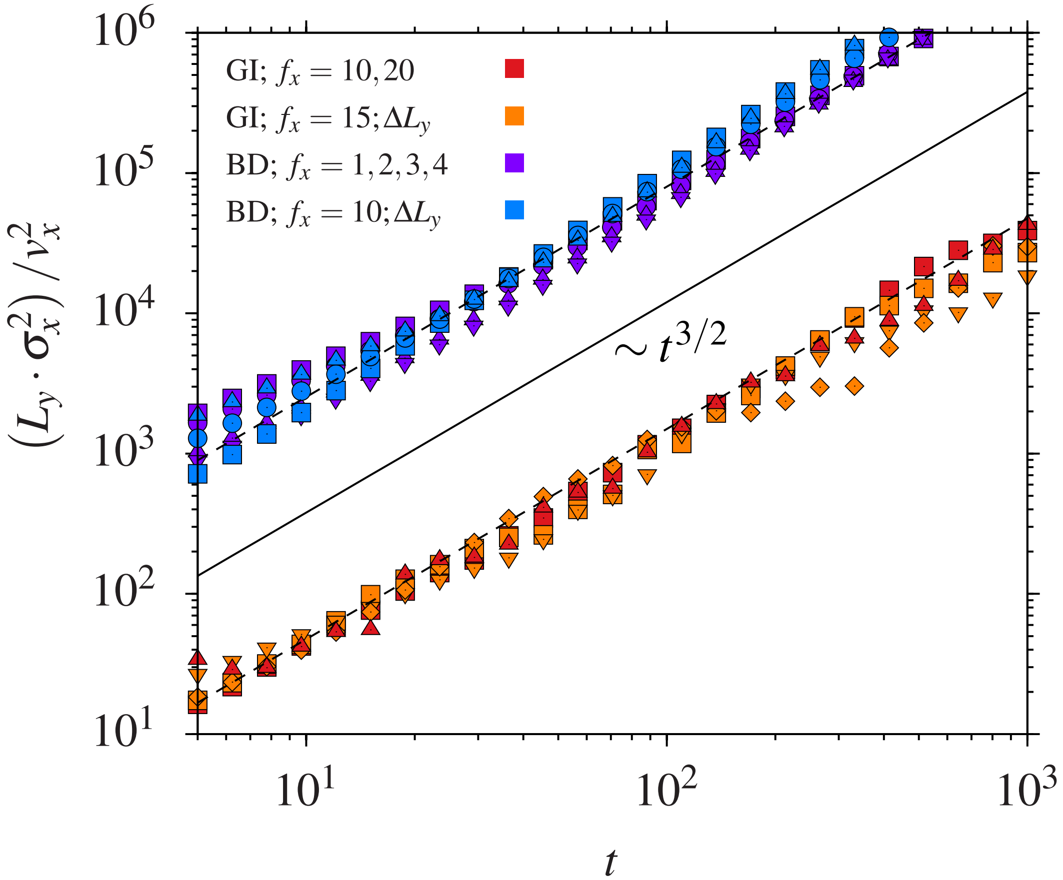
# Superdiffusive broadening of the fluctuations

Simulations of non-glass-formers (monodisperse Yukawa and granular fluids) in 2D stripes

For both systems the variance grows superdiffusively, $\approx t^{3/2}$

Note the data collapse for the variance divided by $v^2$ (and multiplied by L)

This signifies that superdiffusive broadening of fluctuations occurs beyond the linear response




Yukawa liquid Granular fluid
volume fraction *0.5*

$\sigma^2$ changes by three orders of magnitude (spatially resolved regime) time $t$ by more than two orders of magnitude (time resolved regime).
Superdiffusive growth starts from the earliest times when the TP makes just few steps.

# Ultimate behaviour in the large time limit

The long time behaviour is always diffusive

$$\lim_{t\to\infty}\frac{\sigma_x^2}{t}\underset{\rho_0\to 0}{\sim}\begin{cases} B & \text{quasi-1D,} \\ 4a_0^2\pi^{-1}\rho_0\ln(\rho_0^{-1}) & \text{2D lattice,} \\ 2a_0^2[A+\coth(f/2)/(2a_0)]\rho_0 & \text{3D lattice,} \end{cases}$$

In quasi-1D the longitudinal diffusivity is enhanced

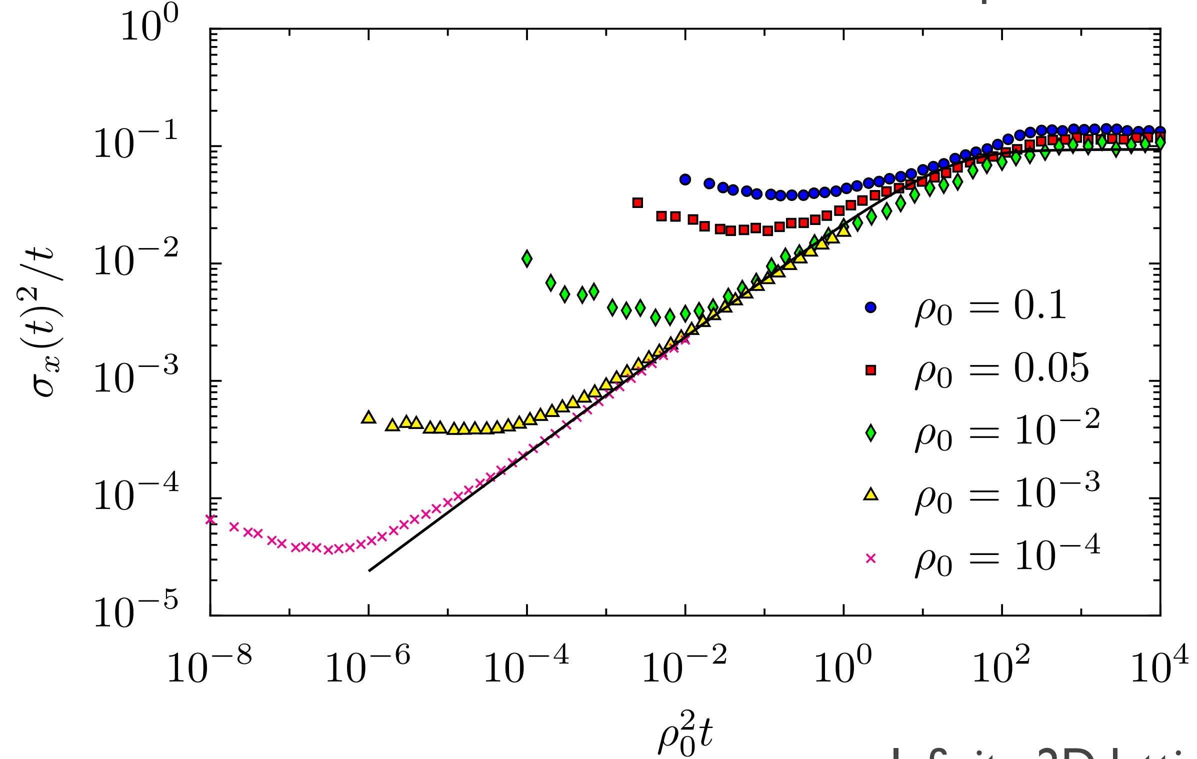
$$\frac{D_\parallel}{D_\perp}\sim\frac{1}{\rho_0}$$

In 2D

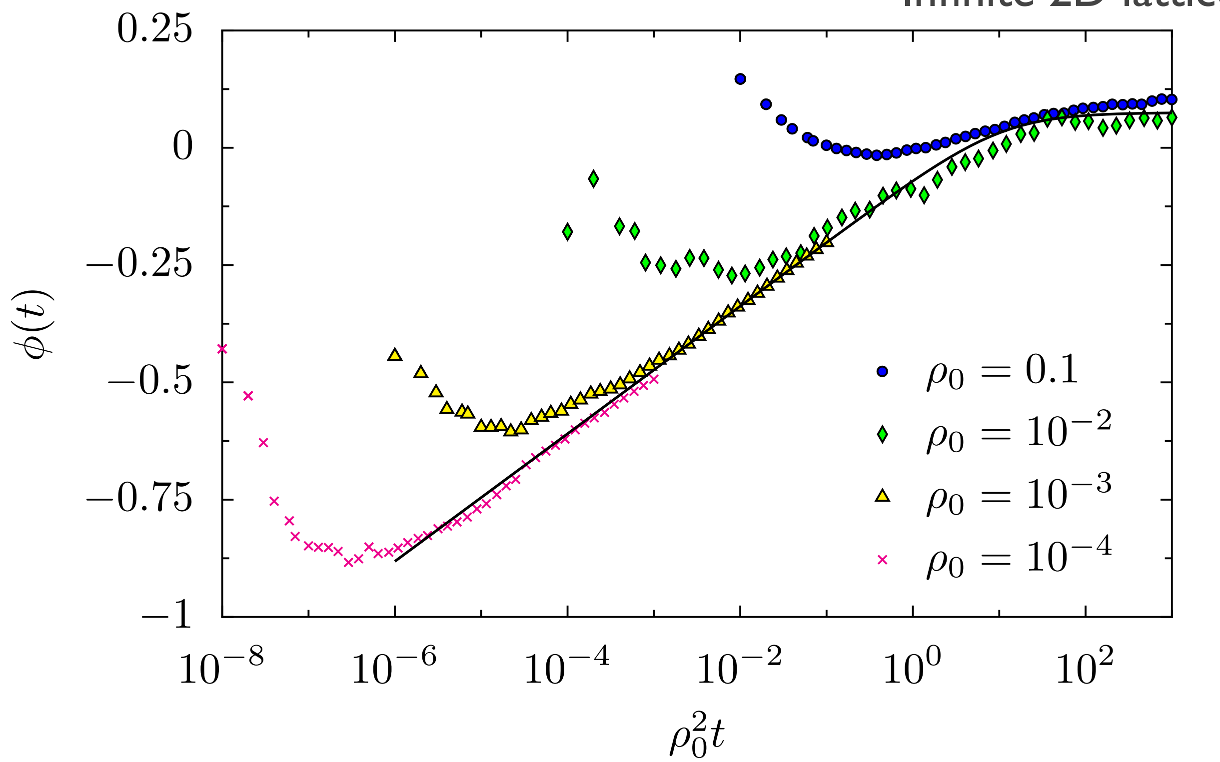
$$\frac{D_\parallel}{D_\perp}\sim\ln(\rho_0^{-1})$$

No enhancement is observed in 3D

2D stripe with L =3




Infinite 2D lattice



# Conclusions

- New phenomena: field-induced broadening of fluctuations in crowded environments.

- Biased TP dynamics in different geometries, infinite 3D, 2D, and in confined geometries, 3D slit pores, 2D stripes, 3D capillaries

- Exact results for the TP velocity (F > 0) and the TP diffusion coefficient (F = 0).

- Variance of the TP displacement :
  - Intermediate time behavior. Qualitative explanation.
  - Simulations for lattices and off-lattice systems.
  - Long-time behavior. Qualitative CTRW-picture
  - Convergence of the distribution to the Gaussian function

- New results for single files

Mejia-Monasterio & Oshanin Soft matter **7** (2011) 993
Bénichou, Mejia-Monasterio, Oshanin PRE **87** (2013) 020103
Bénichou et al JSTAT P05008 (2013)
Illien et al PRL **111** (2013) 038102
Bénichou et al PRL **111** (2013) 260601
Illien et al PRL **113** (2014) 030603

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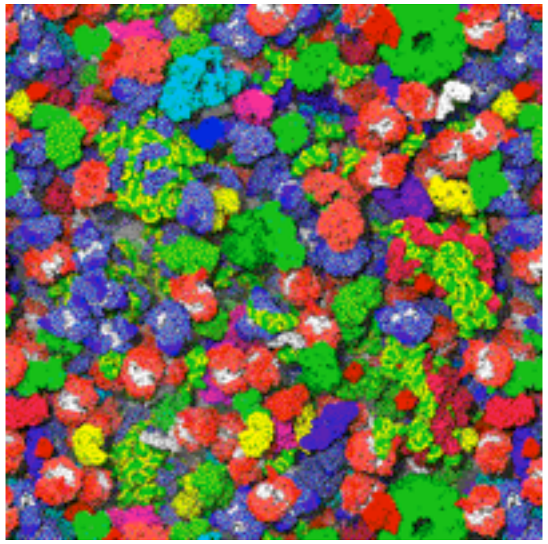
McGuffee & Elcock (2010)



*In confined geometries, transport is passively subdiffusive but actively superdiffusive*