Insights from single-file diffusion into glassy dynamics

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Question:

What is single-file diffusion (SFD)?

Why is it interesting?

A short answer:
Single-file diffusion is a 1D model of caged dynamics.

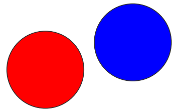
Further questions:

- What does SFD tell us about cages?
- How can we detect collective motions in SFD and determine its correlation length?
- Is the method of analysis extensible to 2D colloidal liquids?
- How can we introduce cage-breaking events into SFD?
- Does SFD help us to choose "good" statistical quantities for proper characterization of glassy dynamics?

— All right, I will answer these questions one by one.

Kinetic theories of particle systems

- Rarefied: 一期一会 one life, one meeting

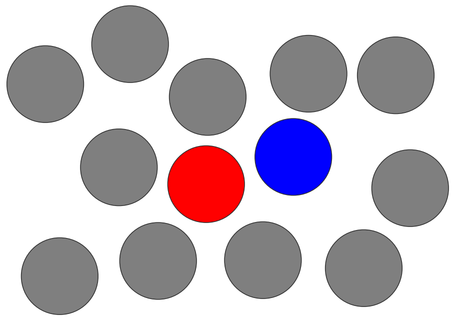


  - always encounter a new partner

$$\frac{\partial f}{\partial t} + \mathbf{v} \cdot \frac{\partial f}{\partial \mathbf{r}} = \int \Phi f f \qquad \text{Boltzmann eq.}$$

  - reunions or ring collisions as exceptional events

- Dense: 一蓮托生 to be reborn on the same lotus
  - always the same neighbors:
    motion blocked by "cage effect"
  - solidification without crystallization



Single-File Diffusion (SFD)

1D system of particles (typically Brownian)

+ "no-passing" repulsive interaction

$$m\ddot{X}_i = -\mu \dot{X}_i - \frac{\partial}{\partial X_i} \sum_{j<k} V(X_k - X_j) + \mu f_i(t)$$

interaction random force

- thermodynamics:

  ideal gas or Tonks–Takahashi gas

  Takahashi (1942)*

- dynamics:

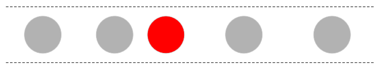
  a problem of 1965-vintage

  Harris (1965), Jepsen (1965)*, ...

  still studied actively

  as a model of ideal cage effect

$*$ reprinted in: "Mathematical Physics in One Dimension", Lieb & Mattis (1966)



demo
in Java

- free Brownian particles ($V = 0$):

  $\langle R^2 \rangle = 2Dt$

  Einstein (1905)

- "no passing" interaction:

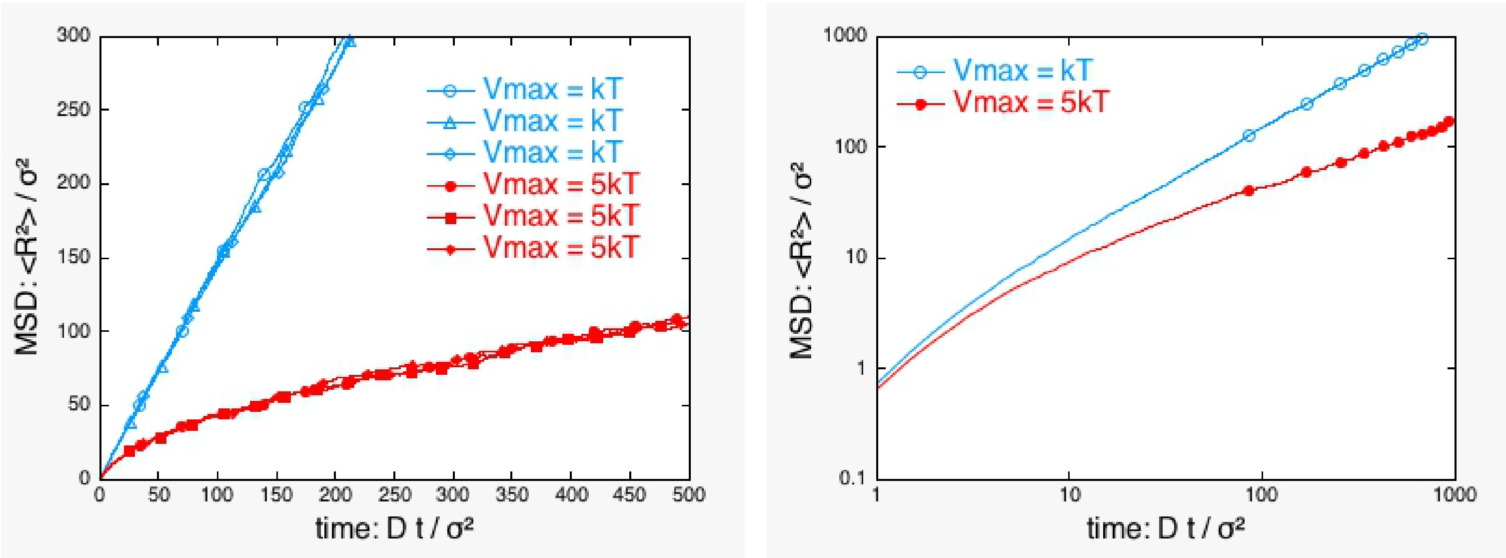
  $\langle R^2 \rangle = ??$

SFD is slow

$\rho_0^{-2} \ll Dt \ll L^2 \to \infty$

$R_j \stackrel{\text{def}}{=} X_j(t) - X_j(0)$; study long-time behavior of MSD

- free Brownian particles ($V = 0$): $\left\langle R^2 \right\rangle \propto t$
- "no passing" ($V_{\max} = \infty$): $\left\langle R^2 \right\rangle = \dfrac{2S}{\rho_0}\sqrt{\dfrac{D^{\mathrm{c}} t}{\pi}} \propto t^{1/2}$
  Kollmann, PRL **90** (2003)



Basic setup

1D Langevin eq. with repulsive interaction

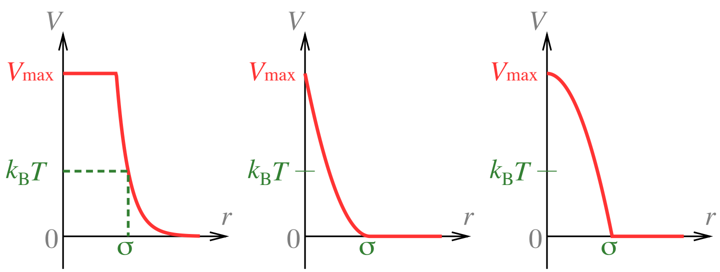
$$m\ddot{X}_i = -\mu\dot{X}_i - \frac{\partial}{\partial X_i}\sum_{j<k} V(X_k - X_j) + \mu f_i(t)$$

interaction   random force

- periodic BC: $X_{i+N} = X_i + L$
- equilibrium IC, statistically homogeneous & steady
- large system ($N \to \infty$) with $\rho_0 = N/L$ kept finite
- $V_{\text{max}} \gg k_B T$: no overtaking

Variants (mainly in the latter half of the talk)

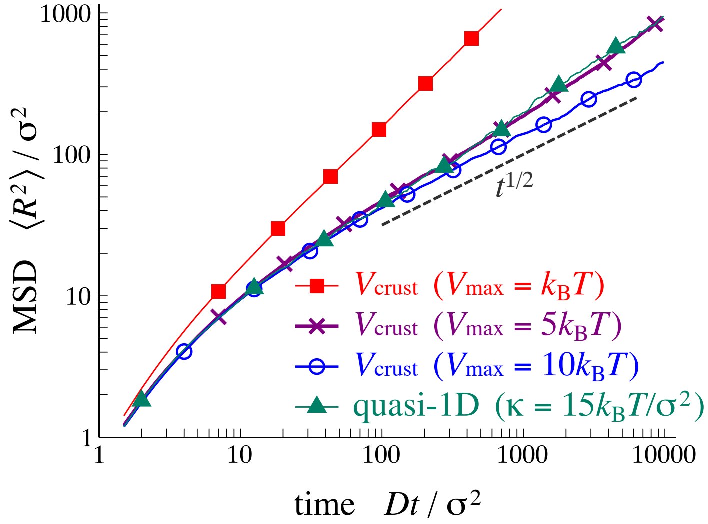
- finite system size L, N
- finite $V_{\text{max}}/k_B T$ & long time: overtaking



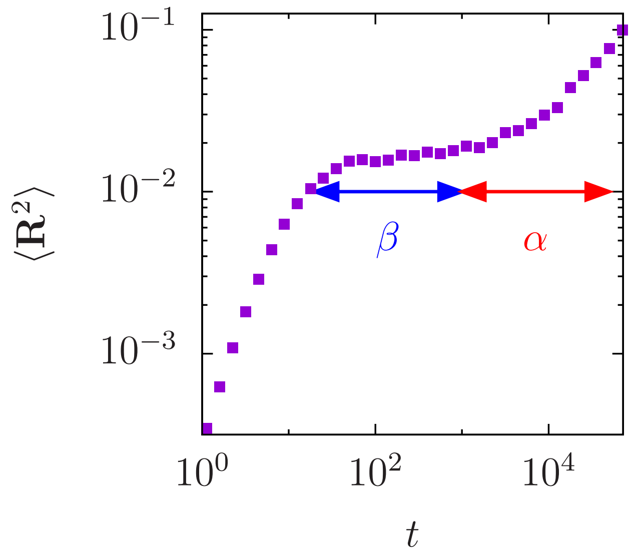
Slowdown of diffusion (1/2): MSD

$$\left\langle \mathbf{R}^2 \right\rangle = \left\langle [\mathbf{r}_j(t) - \mathbf{r}_j(0)]^2 \right\rangle$$

mean square displacement



SFD (1D & quasi-1D)



typical glassy MD in 3D

Ooshida *et al.*, MPLB **29** (2015)

the $\sqrt{t}$-behavior in SFD corresponds to the plateau in 3D case

Slowdown of diffusion (2/2): velocity autocorrelation

SFD

$$\left\langle \dot{X}_i(t)\dot{X}_i(0)\right\rangle \simeq -\sqrt{\frac{k_{\mathrm{B}}T}{\gamma\pi}}(4\rho)^{-1}t^{-3/2}$$

Taloni & Lomholt, PRE **78** (2008)

3D Newtonian MD

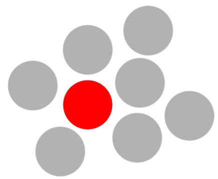
$$Z(t) = \frac{1}{3}\langle \mathbf{v}_i(t)\cdot\mathbf{v}_i(t)\rangle \sim \begin{cases} +t^{-3/2} & (\text{low } \phi) \\ -t^{-5/2} & (\text{high } \phi) \end{cases}$$

Williams *et al.*, PRL **96** (2006)

negative velocity autocorrelation:

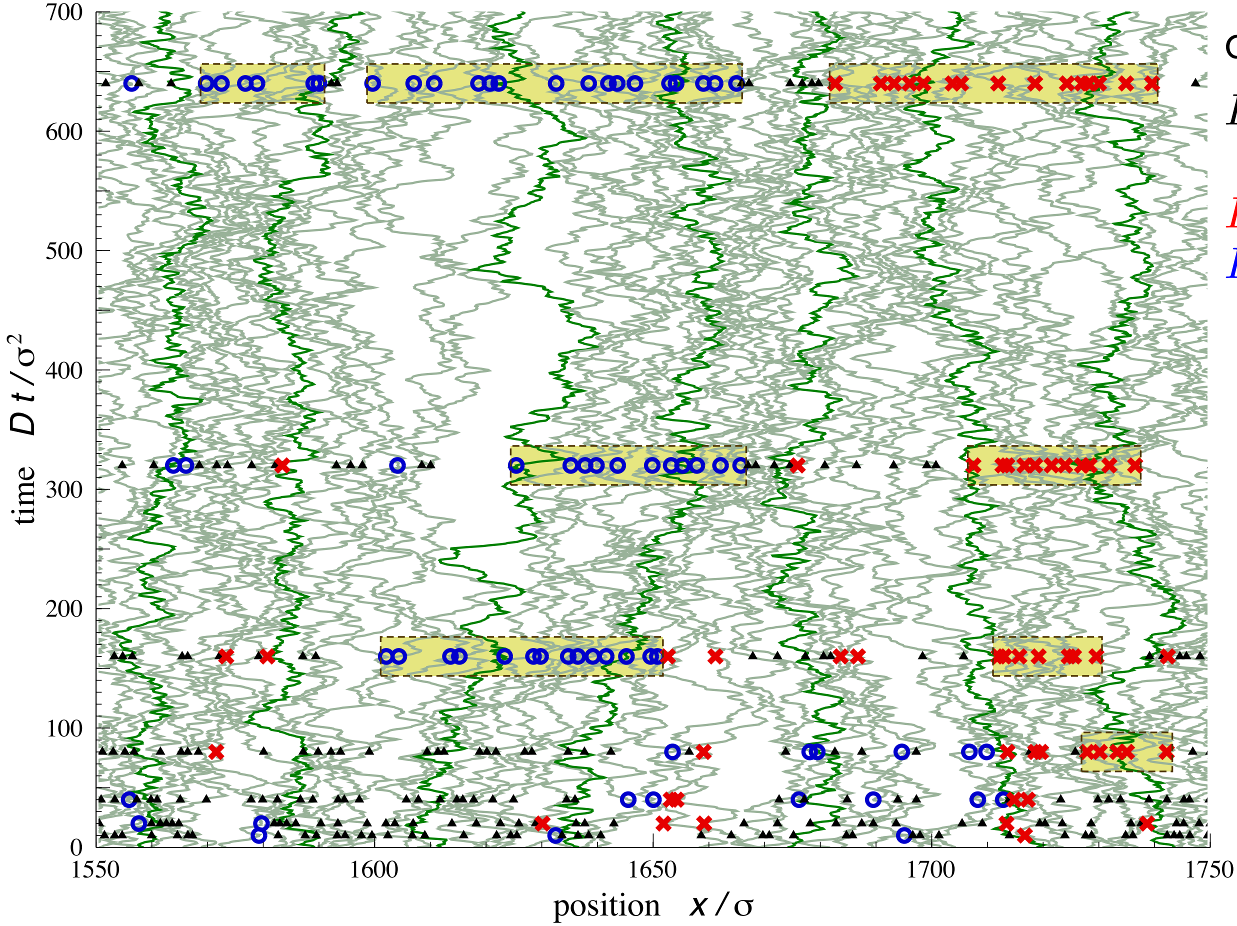
the particle is pushed back by the **neighbors**

what do they do?



Collective motion in space–time diagram

particles moved leftwards (×) and rightwards (◯)



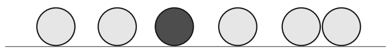
displacement

$$R_j \overset{\text{def}}{=} X_j(t) - X_j(0)$$

$R_j < 0$: leftwards

$R_j > 0$: rightwards

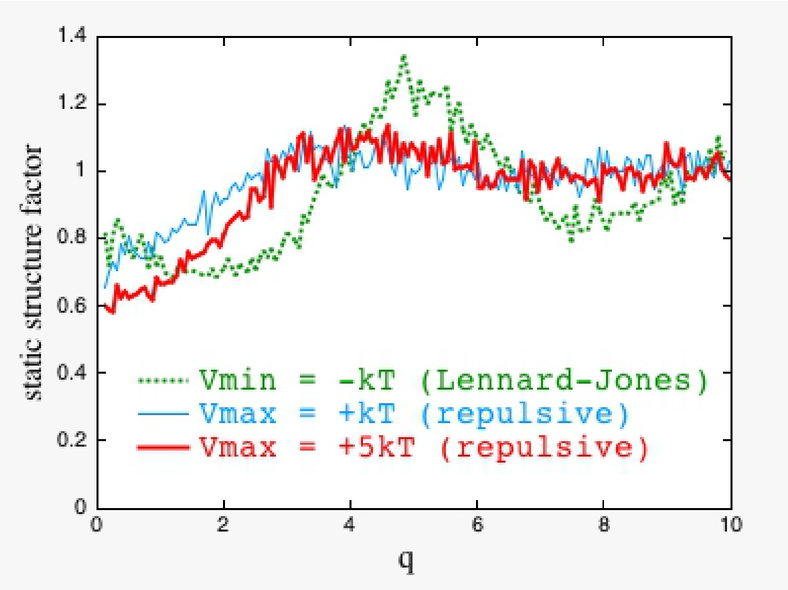
neighbors must cooperate



SFD is "glassy": structure behind the slow dynamics

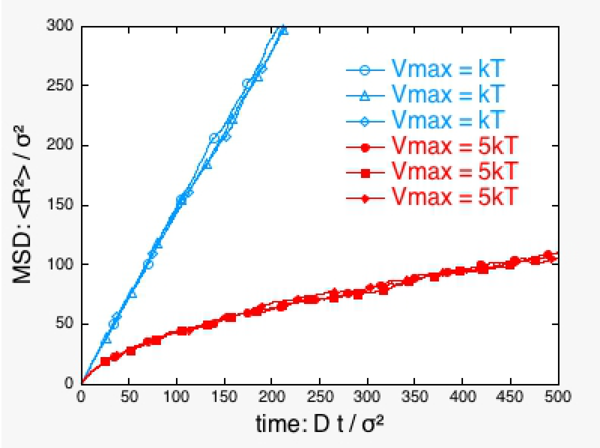
static structure factor

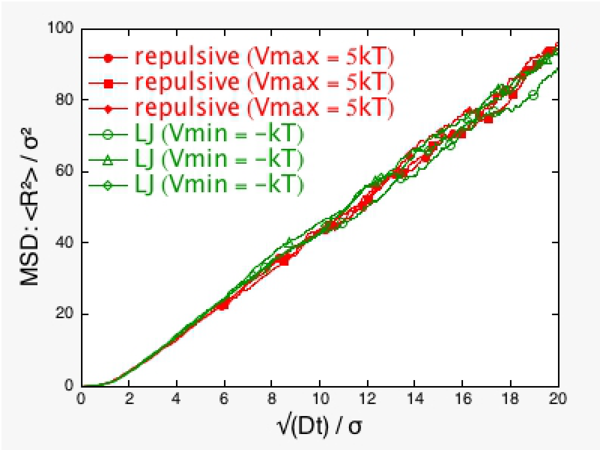
$$S(q) = \frac{1}{N} \sum_{i,j} \left\langle \exp\left[\mathrm{i} q \left(X_j - X_i \right) \right] \right\rangle$$



something beyond $S(q)$:
glassy dynamical structure?

$\langle R^2 \rangle$ vs t

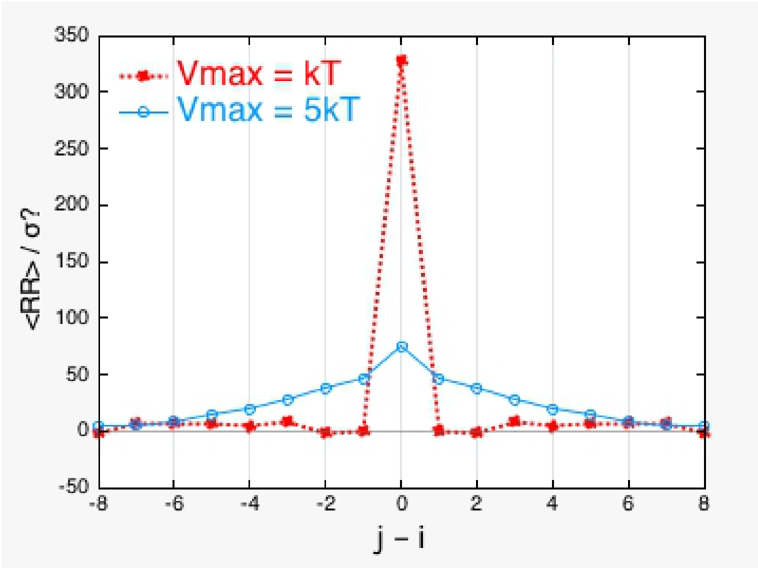




Displacement correlation (DC) in SFD distinguishes anomalous diffusions from normal diffusion

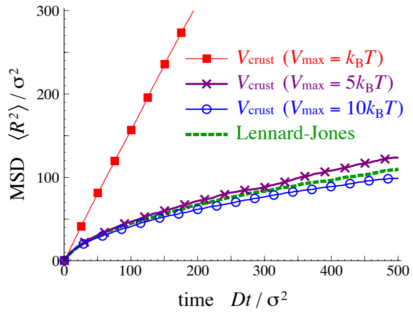
$\langle R_i R_j \rangle$ $(t = 200\,\sigma^2/D)$

displ. corr.



presence/absence of correlation

MSD $\langle R^2 \rangle$



Problem: calculate $\langle RR \rangle$ in the absence of overtaking

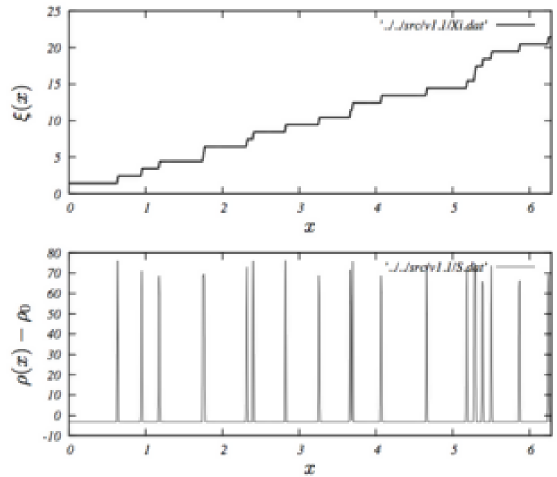
Idea for analytical calculation of DC: label variable

- Continuum description in standard (Eulerian) variable:
  Dean–Kawasaki eq.

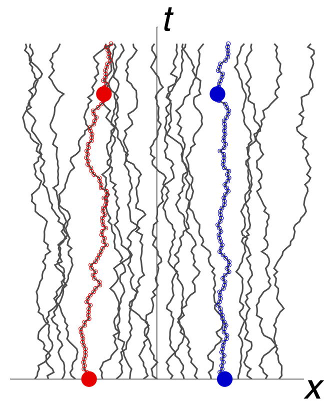
$$\rho = \sum_j \rho_j = \sum_j \delta(x - X_j(t))$$

$$Q = -D\left(\partial_x \rho + \frac{\rho}{k_{\mathrm{B}}T}\partial_x U\right) + \sum_j \rho_j(x,t) f_j(t)$$

$$\partial_t \rho + \partial_x Q = 0$$



- Introduce label variables ξ as
  a solution to $(\partial_x \xi, -\partial_t \xi) = (\rho, Q)$
  and define the mapping $\xi \mapsto x(\xi, t)$
  (Lagrangian description)
  - by construction, ξ satisfies
    the convective eq. $(\rho\partial_t + Q\partial_x)\xi = 0$
  - in the absence of overtaking,
    ξ labels the worldline
  - displacement: $R = x(\xi, t) - x(\xi, 0)$

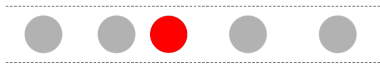


Ideal SFD = elastic chain = roughening surface

- Eulerian-Lagrangian map: $(\xi, t) \mapsto x = x(\xi, t)$
- introduce $\psi = \psi(\xi, t)$ to express $\dfrac{\partial x}{\partial \xi} = \ell_0(1+\psi)$

  deformation grad.

  interpretation:
  - "vacancy" in (lattice) SFD
  - "stretch" in elastic chains

  - slope of $h = x - \ell_0 \xi$ regarded as a roughening surface
- displacement $R(\xi, t) = x(\xi, t) - x(\xi, 0)$

  $$\Rightarrow \quad \partial_\xi R = \ell_0 \left[\psi(\xi, t) - \psi(\xi, 0)\right]$$

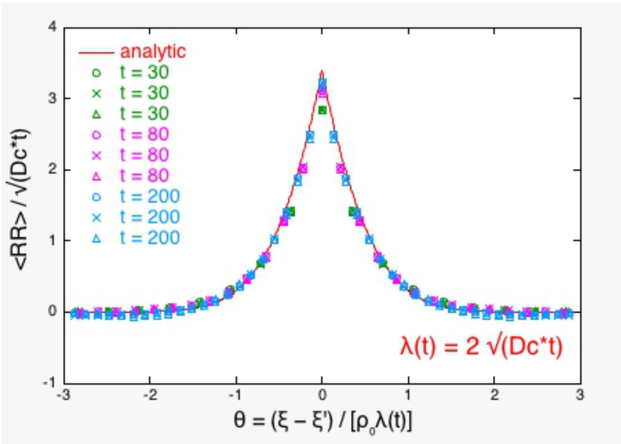
Alexander–Pincus formula relates $\langle RR \rangle$ to $C \propto \langle \psi\psi \rangle$:

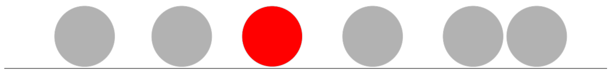
$$\left\langle R^2 \right\rangle \propto \int_{-\infty}^{\infty} \frac{C(k,0) - C(k,t)}{k^2} \mathrm{d}k$$

reduced to Edwards–Wilkinson integral if $C \sim e^{-Dk^2 t}$

more generally: use DK eq.

Analytical result: DC in SFD without overtaking





$$\frac{\langle R(\xi,t)R(\xi',t)\rangle}{2S\ell_0^2\sqrt{D_*^{\mathrm{C}}t}} = \frac{e^{-\theta^2}}{\sqrt{\pi}} - |\theta|\,\mathrm{erfc}\,|\theta|$$

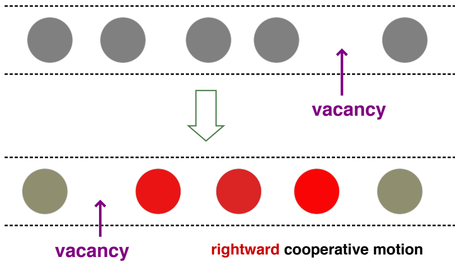
$$\theta \stackrel{\mathrm{def}}{=} \frac{\xi-\xi'}{\rho_0\lambda(t)} = \frac{\xi-\xi'}{2\sqrt{D_*^{\mathrm{C}}t}}, \qquad \ell_0 = \frac{1}{\rho_0}$$

dynamical correlation length

- key ingredient: elongation or "vacancy" field ψ

  displacement gradient $\dfrac{\partial x}{\partial \xi} = \ell_0(1+\psi)$

- correlated motion with diffusive dynamical length $\lambda(t) \propto \sqrt{t}$



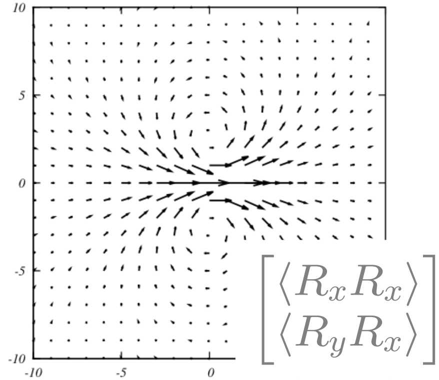
Question:

How can we extend the calculation of displacement correlation to 2D?

Definitions in 2D

label variables $\boldsymbol{\xi} = (\xi, \eta)$: curvilinear coordinate system sticking to the particles, $\mathbf{r}(\boldsymbol{\xi}, t + \Delta t) - \mathbf{r}(\boldsymbol{\xi}, t) = \mathbf{u}(\boldsymbol{\xi}, t)\Delta t$

time integr. $\mathbf{R} = \mathbf{R}(\boldsymbol{\xi}, t - s) = \mathbf{r}(\boldsymbol{\xi}, t) - \mathbf{r}(\boldsymbol{\xi}, s)$
displacement



displacement corr. (DC) tensor

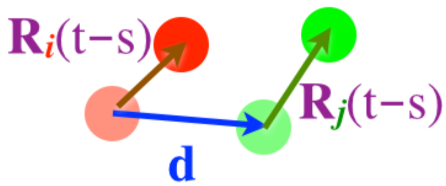
$$\langle \mathbf{R} \otimes \mathbf{R} \rangle_{\tilde{\mathbf{d}}} = \begin{bmatrix}\langle R_x R_x \rangle & \langle R_x R_y \rangle \\ \langle R_y R_x \rangle & \langle R_y R_y \rangle\end{bmatrix}$$

$$= X_{\parallel}(\tilde{d}/\ell_0, t - s)\frac{\tilde{\mathbf{d}} \otimes \tilde{\mathbf{d}}}{\tilde{\mathbf{d}}^2} + X_{\perp}(\tilde{d}/\ell_0, t - s)\left(\mathbb{1} - \frac{\tilde{\mathbf{d}} \otimes \tilde{\mathbf{d}}}{\tilde{\mathbf{d}}^2}\right)$$

longitudinal transverse

as a function of $\tilde{\mathbf{d}} =$ "initial" relative position

$$\tilde{\mathbf{d}} \simeq \ell_0(\boldsymbol{\xi} - \boldsymbol{\xi}'),\ \ell_0 = \frac{1}{\sqrt{\rho_0}}$$



Strategy for analytical calculation of DC in 2D

- deformation grad. $\partial(x,y)/\partial(\xi,\eta)$ taken as field var.:

$$(\partial_\xi \mathbf{r}, \partial_\eta \mathbf{r}) = \ell_0 \begin{bmatrix} 1+\Psi_1 & * \\ * & 1+\Psi_2 \end{bmatrix}, \quad \ell_0 = \frac{1}{\sqrt{\rho_0}}$$

- rewrite Dean–Kawasaki eq. in terms of Ψ_a and $\mathbf{u}$:

$$\ell_0 \partial_t \begin{bmatrix} \Psi_1(\xi,t) \\ \Psi_2(\xi,t) \end{bmatrix} = \begin{bmatrix} \partial_\xi u_x(\xi,t) \\ \partial_\eta u_y(\xi,t) \end{bmatrix}$$

$$\mathbf{u} = -D\left(\frac{\nabla\rho}{\rho} + \frac{\nabla U}{k_\mathrm{B}T}\right) + \sum_j \delta^2(\boldsymbol{\xi} - \boldsymbol{\Xi}_j)\mathbf{f}_j(t)$$

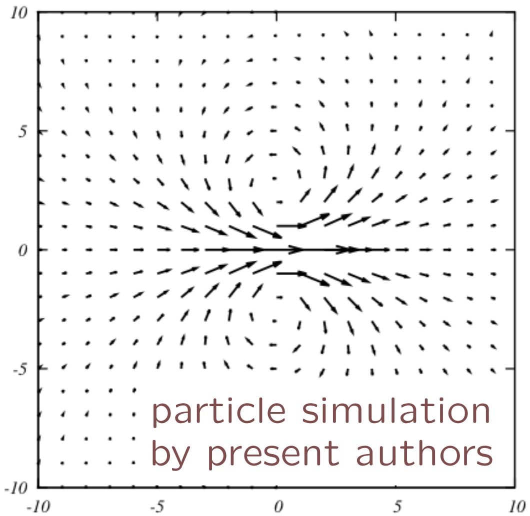
where $\nabla = (\nabla\xi)\partial_\xi + (\nabla\eta)\partial_\eta$ $\rightarrow$ expressible with Ψ

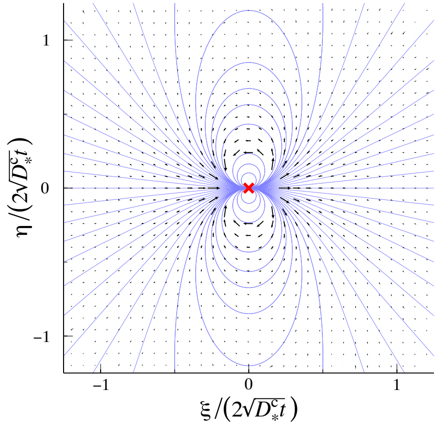
- Calculate $C_{\alpha\beta} \propto \left\langle \check{\Psi}_\alpha \check{\Psi}_\beta \right\rangle$ in Fourier representation

- Alexander–Pincus formula:
inv. Fourier trf. of $\left\langle \check{\Psi}_\alpha \check{\Psi}_\beta \right\rangle / (k_\alpha k_\beta)$ yields $\langle \mathbf{R} \otimes \mathbf{R} \rangle$

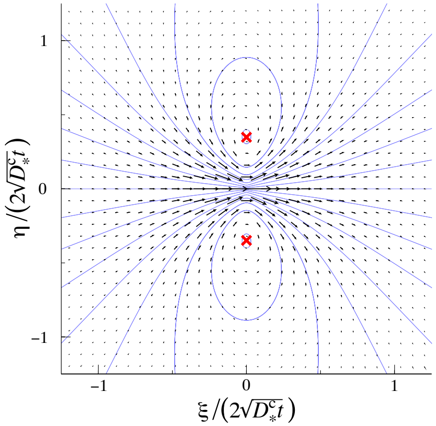
DC in 2D colloidal liquid

label variable theory: DK eq. with $\mathbf{r} = \mathbf{r}(\boldsymbol{\xi}, t)$





linear analysis



linear analysis + correction

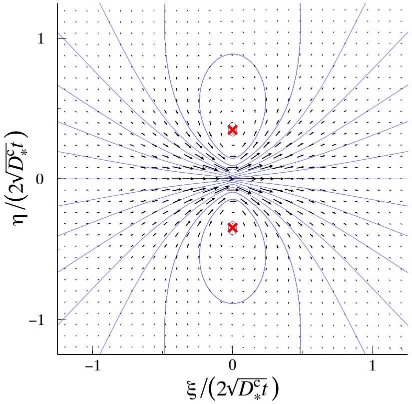
compare:

Doliwa & Heuer, PRE **61** (2000)

- linear analysis (with Helmholtz decomposition in $\boldsymbol{\xi}$-space) suffices to explain DC at *larger* scales
- need of correction for *rotational* modes at *smaller* scales: elasticity

Ooshida *et al.*, PRE **94** (2016)

Remark: difference from Alder–Wainwright backflow

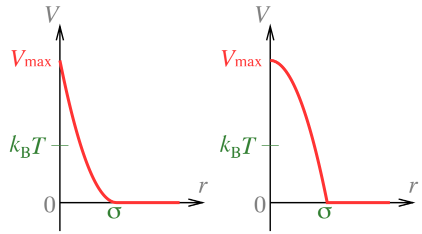
	Alder & Wainwright	present
spatially:	vortex pair (with "back flow") PRA **1** (1970)	
temporally: $\langle \mathbf{u}(t) \cdot \mathbf{u}(s) \rangle$	**positive** tail	**negative** tail (i.e. cage effect)

N.B. velocity autocorrelation in SFD is also negative

Question:

How can we introduce a 1D analogue of rotational modes into SFD?

Overtaking: a cage-breaking event allowed by finite barrier



The same 1D Langevin eq.

$$m\ddot{X}_i = -\mu \dot{X}_i - \frac{\partial}{\partial X_i} \sum_{j<k} V(X_k - X_j) + \mu f_i(t)$$

interaction random force

but now with finite $V_{\max}/k_B T$ & long time

$\Rightarrow$ overtaking rate $\nu_\alpha > 0$

finite $V_{\max}$ can be interpreted as modeling quasi-1D system of hardcore particles, with $\mathbf{r}_i = (X_i, Y_i)$:

$$m\ddot{\mathbf{r}}_i = -\mu \dot{\mathbf{r}}_i - \frac{\partial}{\partial \mathbf{r}_i} \left[\sum_{j<k} V_{\text{hardcore}}(r_{jk}) + \sum_j \frac{\kappa}{2} Y_j^2 \right] + \mu \mathbf{f}_i(t)$$

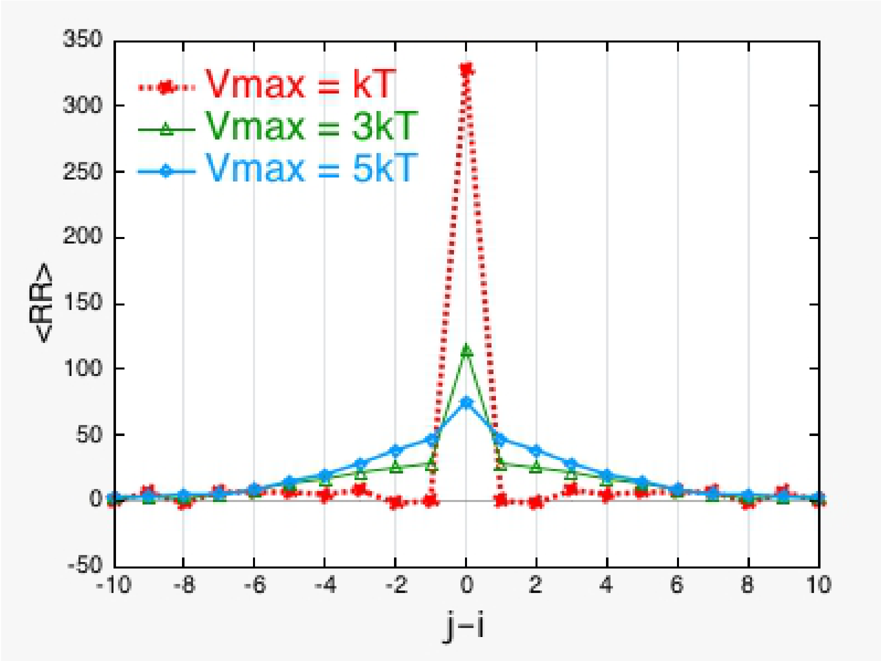
DC from simulation with different barrier heights

V_{max}	overtaking
$+\infty$	never
high	seldom
medium	sometimes
low	often
0	always

Problem 2: how does the presence of *infrequent* overtaking affect the displacement correlation?

Numerical solution: decrease in *short-distance* correlations (except for $j - i = 0$)

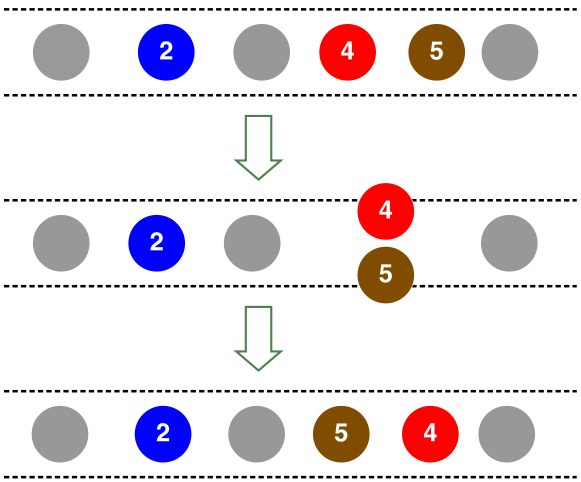
$\rightarrow$ analytical clarification?



$\langle R_i R_j \rangle$ plotted against $j - i$ at $t = 200$

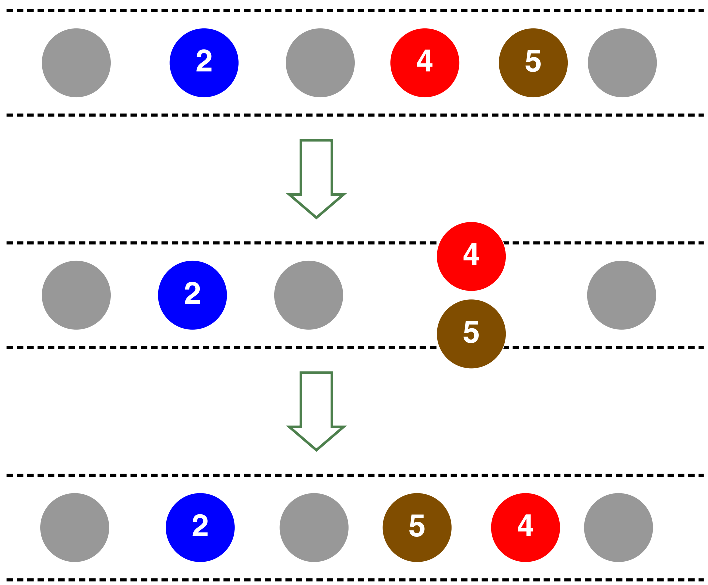
Theoretical treatment of overtaking: formulation

- Correspondence among three variables:
  numbering j, label variable ξ, and particle position x
- label variable $\xi = \xi(x, t)$, by construction, obeys the convective eq. $(\rho\partial_t + Q\partial_x)\xi = 0$
- define $\Xi_j(t) \stackrel{\text{def}}{=} \xi(X_j(t), t)$
  Ooshida *et al.*, JPSJ **80** (2011)
  - no passing: $\Xi_j(t) = \Xi_j(0)$
    time-independent
  - overtaking: $\Xi_j(t) \neq \Xi_j(0)$



$$\frac{\mathrm{d}\Xi_j}{\mathrm{d}t} = \sum_i \int \left(\rho_i Q_j - \rho_j Q_i\right) \mathrm{d}x$$

Theoretical treatment of overtaking: example



before it

$$\Xi_2 = 2, \quad \Xi_4 = 4, \quad \Xi_5 = 5$$

during it

$$\frac{\mathrm{d}\Xi_4}{\mathrm{d}t} = \int (\rho_5 Q_4 - \rho_4 Q_5)\mathrm{d}x = -\frac{\mathrm{d}\Xi_5}{\mathrm{d}t}$$

after it

$$\Xi_2 = 2, \quad \Xi_4 = 5, \quad \Xi_5 = 4$$

$$\Xi_j(t) \stackrel{\text{def}}{=} \xi(X_j(t), t),$$

$$\rho_j = \delta(x - X_j(t)), \quad \partial_t \rho_j + \partial_x Q_j = 0$$

Calculation of DC in SFD with overtaking

- express $R_j(t) = X_j(t) - X_j(0)$ in terms of $\check{\psi}(k,t)$ and $\delta\Xi_j(t) \overset{\text{def}}{=} \Xi_j(t) - \Xi_j(0)$
- $\check{\psi}(k,t)$, Fourier modes of vacancy field, is still governed by Dean–Kawasaki eq.
- dynamics of $\delta\Xi$ (i.e. overtaking) modeled by random exchange with frequency ν_α

  cf. *Amidakuji* (Amitabha's Lottery)

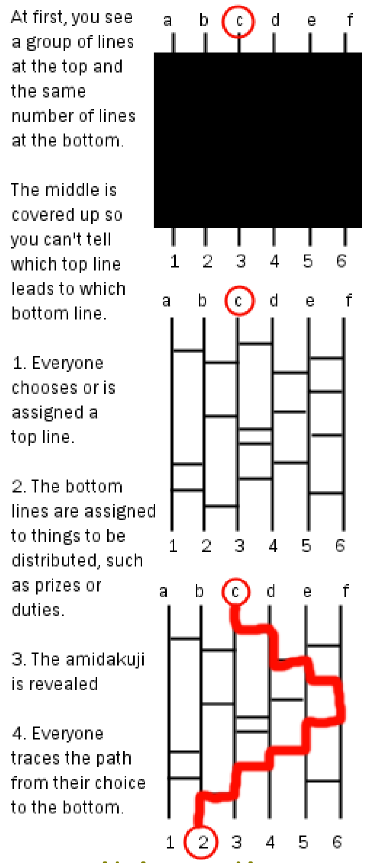
  $\to$ PDF for $(\delta\Xi_i, \delta\Xi_j)$
- calculate DC within linear approx.:

$$\left\langle R_i R_j \right\rangle = \ell_0^2 \sum_{k\neq 0} \frac{\left\langle e^{\mathrm{i}k(\Xi_j - \Xi_i)} \left|\check{\psi}(k,t) - \check{\psi}(k,0)\right|^2 \right\rangle}{k^2} + \ell^2 \left\langle \delta\Xi_i \delta\Xi_j \right\rangle$$

$$= \cdots$$

assumption:
no correlation between ψ and Ξ

Amidakuji



wikimedia

Analytical result: DC in SFD with overtaking

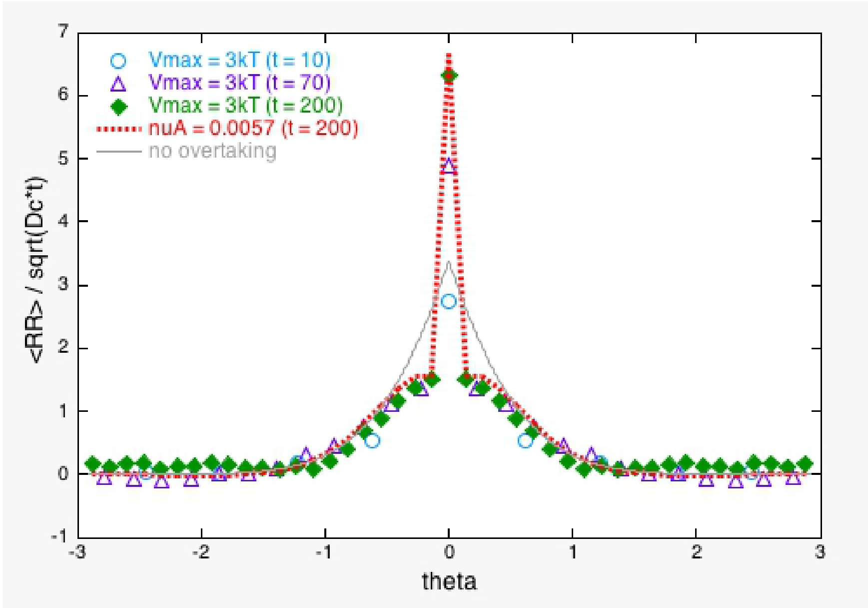
$$\frac{\langle R_i R_j \rangle}{\ell_0^2} = \begin{cases} 2S\sqrt{D'_* t} + 2\nu_\alpha t & (i=j) \\ S\left[2\sqrt{D'_* t}\,\varphi\left(\frac{|j-i|}{\sqrt{4D'_* t}}\right) - \sqrt{2\nu_\alpha t}\,\varphi\left(\frac{|j-i|}{\sqrt{8\nu_\alpha t}}\right)\right] + \langle \delta\Xi_i \delta\Xi_j \rangle & (i \neq j) \end{cases}$$

where $\ell_0 = L/N$, $D'_* = D_*^{\mathrm{c}} + \nu_\alpha$,

$$\varphi(\theta) = \frac{1}{\sqrt{\pi}} e^{-\theta^2} - |\theta| \, \mathrm{erfc}\, |\theta| \,,$$

$\langle \delta\Xi_i \delta\Xi_{i+\Delta} \rangle$ ↓ modified Bessel func.

$$= -2\nu_\alpha t\, e^{-4\nu_\alpha t} \left[I_{\Delta-1}(4\nu_\alpha t) + I_\Delta(4\nu_\alpha t) \right] + \left(\Delta - \frac{1}{2}\right) e^{-4\nu_\alpha t} \sum_{n=\Delta}^{\infty} I_n(4\nu_\alpha t)$$



Comparison w particle simulation

$\frac{\langle R_i R_j \rangle}{\sqrt{D^{\mathrm{c}} t}}$ plotted against $\vartheta = \frac{|j-i|}{2\sqrt{D_*^{\mathrm{c}} t}}$

- - - - : theory with overtaking ($\nu_\alpha = 0.0057\, D/\sigma^2$)

—— : theory without overtaking ($\nu_\alpha = 0$)

What determines the overtaking rate?

- 1D interaction potential: harmonic repulsion

$$V(r) = \begin{cases} V_{\mathrm{max}} \left(1 - |r|/\sigma\right)^2 & (|r| < \sigma) \\ 0 & (|r| > \sigma) \end{cases}$$

- density $\rho_0 = N/L$

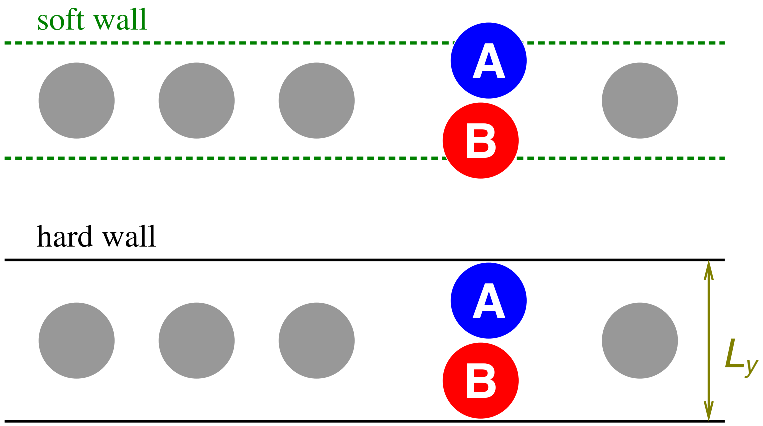
count overtaking events numerically

$\Rightarrow$ compute ν_α $\Rightarrow$ fitting: $a_0 \approx 1/2$ and $a_1 \approx 1/6$

$$\nu_\alpha = D \left(a_0 \frac{\rho_0}{\sigma} + a_1 \rho_0^2 \, \beta V_{\mathrm{max}} \right) e^{-\beta V_{\mathrm{max}}} \qquad \left(\beta = \frac{1}{k_{\mathrm{B}} T} \right)$$

Arrhenius-like, with a prefactor

1D potential reflects particle-to-channel size ratio



- soft wall:

$$V_{\mathsf{max}} \sim \frac{\kappa\sigma^2}{k_{\mathsf{B}}T}$$

no singularity

- hard wall:

$$V_{\mathsf{max}} \sim k_{\mathsf{B}}T \ln \frac{\sigma}{L_y - 2\sigma}$$

singular for $\sigma \to L/2$

Numerical values of ν_α
$\Rightarrow$ plot it against confinement strength such as κ
Lucena *et al.*, PRE **85** (2012)

cf. Angel plot

Last question:

Is the MSD a “good variable”

to observe overtaking?

1D elastic fluctuation eclipses overtaking

cf. Shiba *et al.*, PRL **117** (2016): "eclipse" by 2D fluctuation

MSD in SFD with $L \to \infty$ and finite V_{max}:

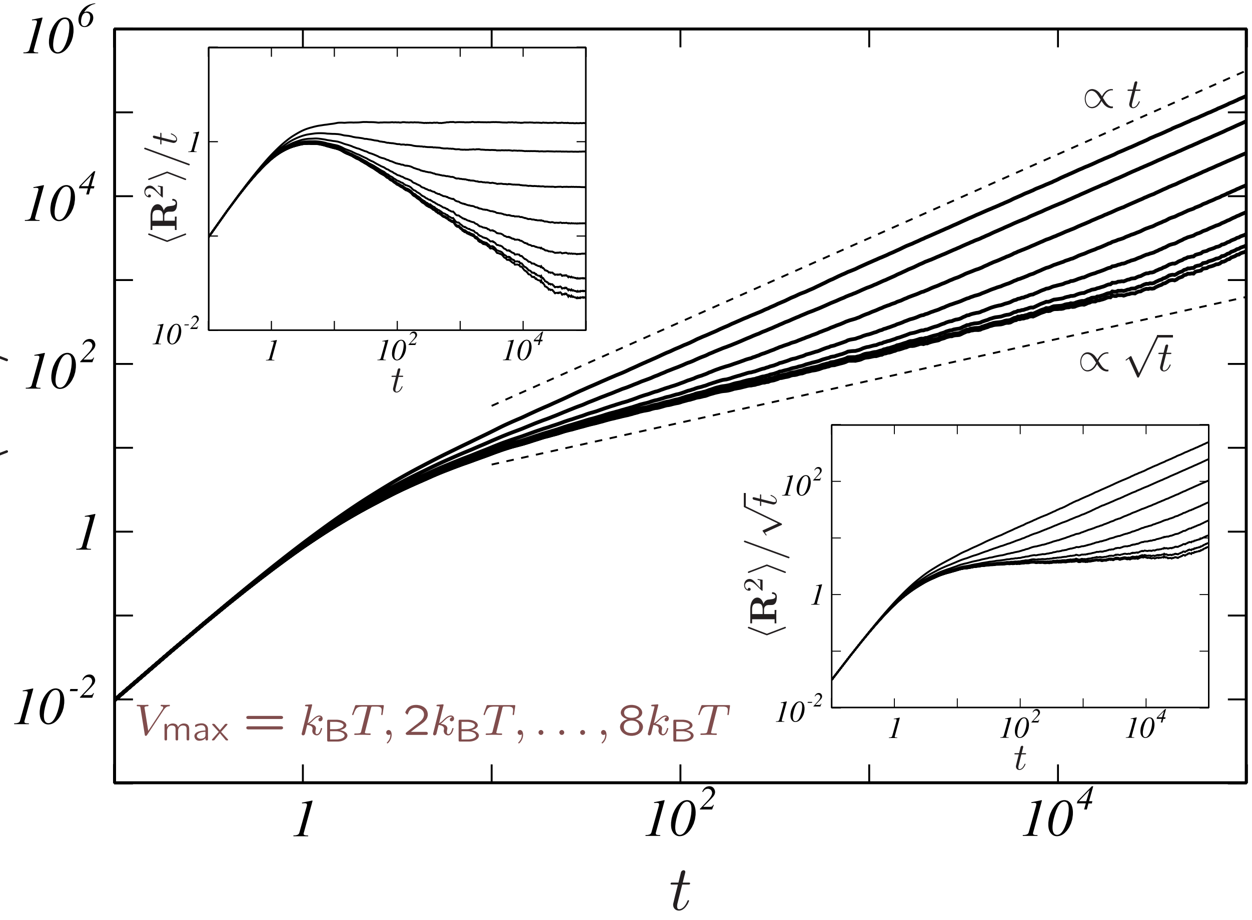
$$\left\langle R^2 \right\rangle = \Bigl(K\sqrt{t} + [\text{correction}] \Bigr) + 2D_\alpha t$$

elastic modes   overtaking

remedies:

- *ultra-longtime* simulation so that $2D_\alpha t$ dominates
- *smaller* system size L to suppress elastic fluctuation
- better statistical quantities (less sensitive to drift)

Remedy 1: longtime simulation



Remedy 2: smaller system size

MSD in SFD with finite (N, L) and finite V_{max}:

$$\left\langle R^2 \right\rangle \simeq I_{\mathrm{EW}}(t) + \frac{2Dt}{N} + 2D_\alpha t$$

c-o-mass overtaking

elastic modes

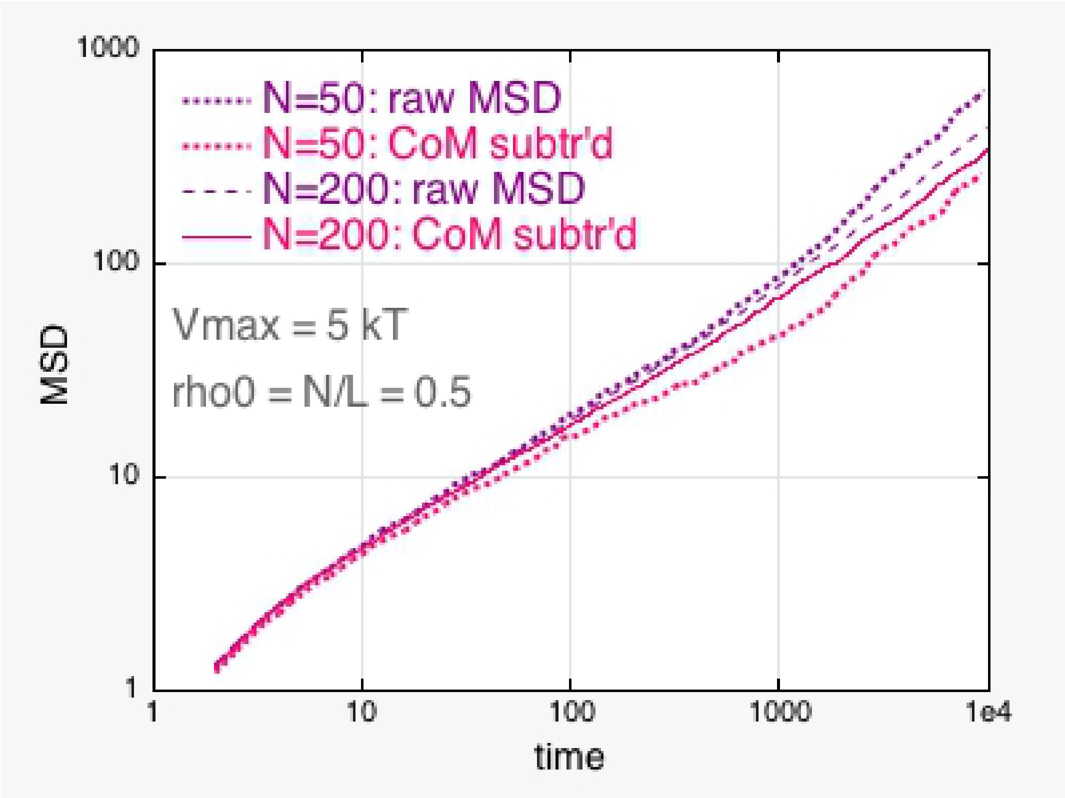
$$I_{\mathrm{EW}}(t) = \frac{2S\ell_0^2}{\pi} \int_{\pi/N}^{\infty} \frac{1 - e^{-D_*^{\mathrm{c}} k^2 t}}{k^2} \mathrm{d}k \simeq \begin{cases} K\sqrt{t} & (\lambda \ll L) \\ \frac{2S}{\pi^2} \ell_0 L & (\lambda \gg L) \end{cases}$$

$$\lambda = \lambda(t) = 2\sqrt{D^{\mathrm{c}} t}$$

- elastic fluctuation, given by I_{EW}, saturates when λ reaches the system size L
- contribution from the center-of-mass motion is not negligible anymore

subtract center-of-mass motion:

$$\left\langle R^2 \right\rangle - \frac{2Dt}{N} \simeq I_{\mathrm{EW}}(t) + 2D_\alpha t, \qquad I_{\mathrm{EW}}(t) \simeq \begin{cases} K\sqrt{t} & (\lambda \ll L) \\ \frac{2S}{\pi^2}\ell_0 L & (\lambda \gg L) \end{cases}$$



be careful!
the result still depends on L

Remedy 3: statistical quantity less sensitive to drift

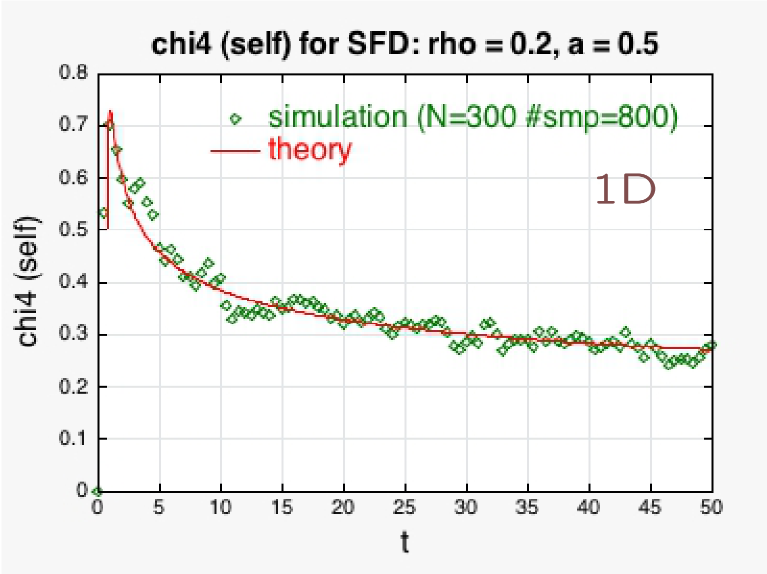
a "bad example":

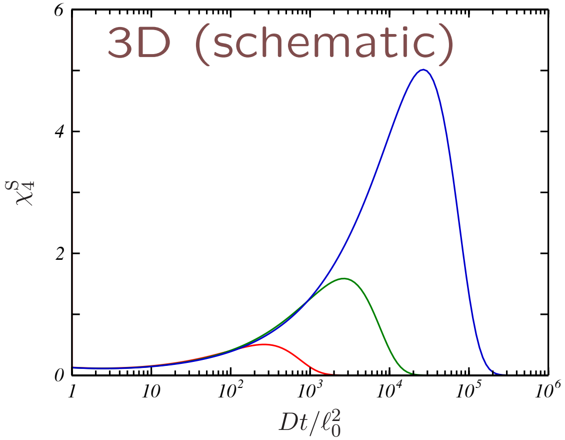
$\mathcal{Q}$-based $\chi_4 \propto \langle \mathcal{Q}^2 \rangle - \langle \mathcal{Q} \rangle^2$

overlap density

$$\mathcal{Q} = \sum_{i,j} \bar{\delta}_a(\mathbf{r}_j(t) - \mathbf{r}_i(0))$$

Lačević *et al.*, JCP **119** (2003)



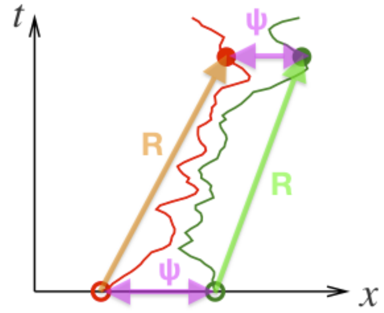


χ_4^{S} does *not* grow in SFD with $\nu_\alpha = 0$ because it is too sensitive to drift!

Ooshida *et al.*, PRE **88** (2013)

A better bridge between theory & simulation?

- $\mathcal{Q}$-based χ_4 is too sensitive to drift
- $\langle RR \rangle$ is also subject to drift
- Since ψ is frame-independent, its particle-based counterpart should have the same property



proposal

elongation correlation:

analytical tractable variant of bond breakage correlation

Ooshida & Otsuki, JPSJ **86** (2017)

Definitions: elongation correlation

Elongation for the pair of particles (i, j): $\ell_0 = L/N$

$$\varepsilon_{i,j}(t) \stackrel{\text{def}}{=} \frac{X_j(t) - X_i(t)}{X_j^\natural - X_i^\natural} - 1 = \frac{X_j(t) - X_i(t) - (j-i)\ell_0}{(j-i)\ell_0}$$

natural distance

As a function of two times s, t (such that $0 \le s \le t$) and the label distance $\Delta = \tilde{d}/\ell_0 \in \mathbf{Z}_+$, we define

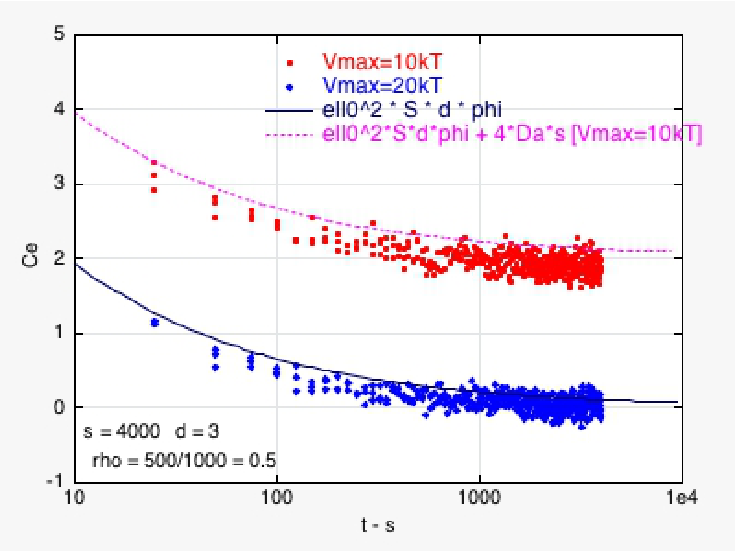
$$C_\varepsilon(\Delta, t, s) \stackrel{\text{def}}{=} \Delta^2 \ell_0^2 \left\langle \varepsilon_{i,j}(t)\varepsilon_{i,j}(s) \right\rangle_{j-i=\Delta}$$
$$= \frac{1}{N} \sum_i \left\langle \left[X_{i+\Delta}(t) - X_i(t) - \tilde{d}\right]\left[X_{i+\Delta}(s) - X_i(s) - \tilde{d}\right] \right\rangle$$

Equal-time correlation:

$$C_\varepsilon^0(\Delta, s) = C_\varepsilon(\Delta, s, s) = \frac{1}{N} \sum_i \left\langle \left[X_{i+\Delta}(s) - X_i(s) - \tilde{d}\right]^2 \right\rangle$$

Effect of overtaking on elongation correlation

$$C_\varepsilon(\Delta, t, s) = \frac{L^4}{\pi N^2} \int_{-\infty}^{\infty} \frac{1 - \cos k\Delta}{k^2} \overset{\downarrow \text{ corr. of } \psi}{C(k,t,s)} \mathrm{d}k + \underset{\text{overtaking}}{4 D_\alpha s}$$



$C_\varepsilon(k,t,s)$ vs $t-s$

- $C = \langle\psi\psi\rangle$ is *insensitive* to overtaking; ψ is designed so as to measure the distance from the "inherent structure"
- $C_\varepsilon = \langle\varepsilon\varepsilon\rangle$ is subject to the disordered numbering due to the overtaking events before s

Concluding remarks

- A cage is not made of the first neighbors only; rather, it is like a "space-time matryoshka"
- While the inner layers of the cage is broken by overtaking, the outer layers are still governed by elastic fluctuations alone
- SFD with overtaking is useful as a test bed of new ideas of statistical quantity for 2D colloidal glasses

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