Calculation of displacement correlation tensor indicating vortical cooperative motion in two-dimensional colloidal liquids

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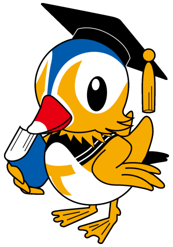


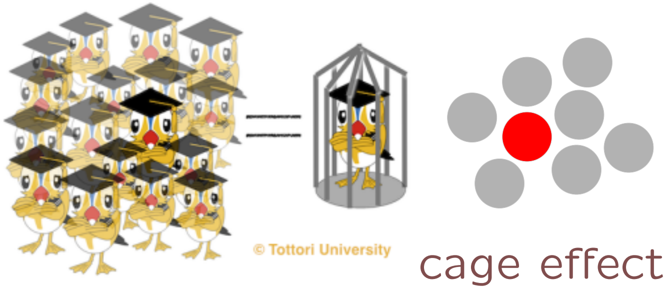
in collaboration with
Otsuki M., Goto S., & Matsumoto T.

YITP Workshop (Kyoto) 2016-Mar-09

Motivation & Target

- Rheology of granular/colloidal pastes (plasticity)
  $\leftarrow$ strategy: solid-based approach to jammed systems
  via **tensorial** MCT with deformation tensor

- Present target:
  colloidal liquid (Langevin eq.)



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cage effect

$$\mu\dot{\mathbf{r}}_i = -\frac{\partial}{\partial \mathbf{r}_i}\sum_{j<k} V(r_{jk}) + \mu\mathbf{f}_i(t)$$

$$\left\langle \mathbf{f}_i(t)\otimes\mathbf{f}_j(t')\right\rangle = \frac{2k_{\mathrm{B}}T}{\mu}\delta_{ij}\delta(t-t')\mathbb{1}$$

  continuum description: Dean–Kawasaki eq.

- Problem:
  calculate displacement correlation tensor analytically
  (quantitative indicator of *vortical* cooperative motion)

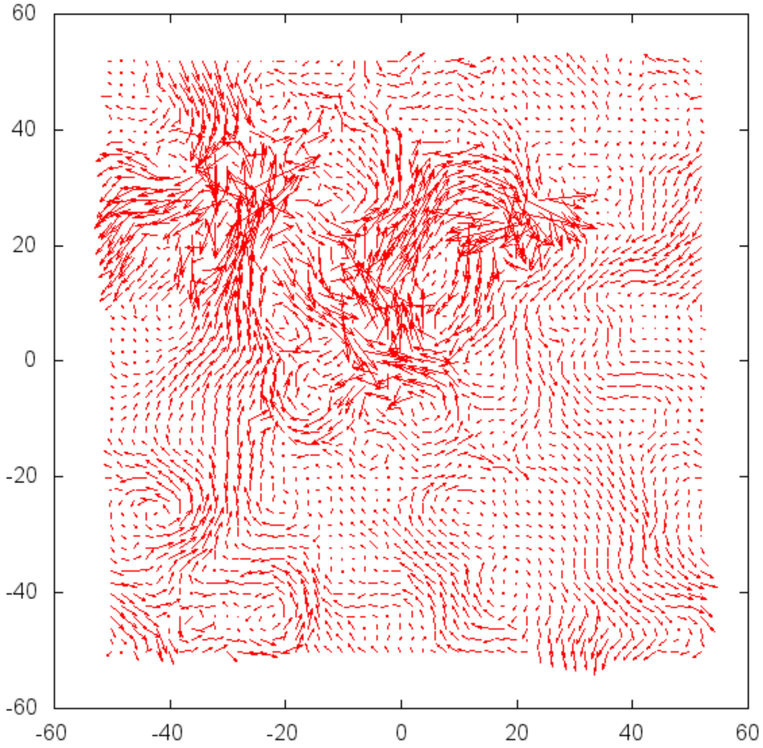
Crowded particles can move only cooperatively

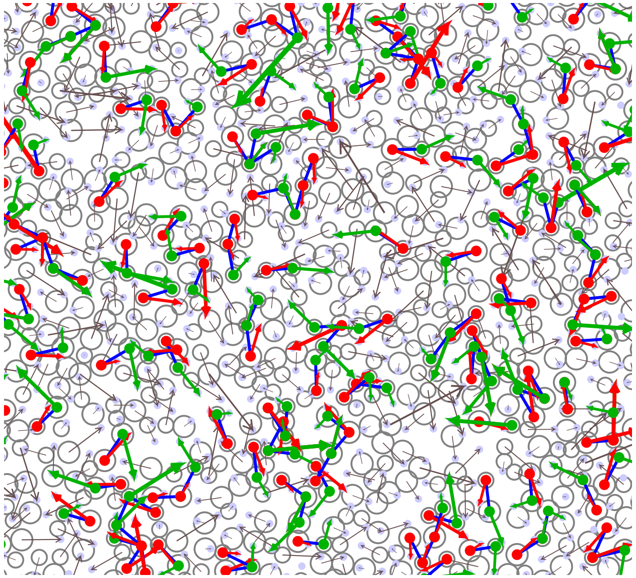
granular (left) or colloidal (right), the particles tend to move together

$$\tilde{\mathbf{d}}_{ij} = \boldsymbol{r}_j(s) - \boldsymbol{r}_i(s)$$

$$\mathbf{R}_i = \boldsymbol{r}_i(t) - \boldsymbol{r}_i(s)$$

$$\mathbf{R}_j = \boldsymbol{r}_j(t) - \boldsymbol{r}_j(s)$$

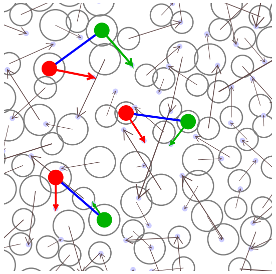


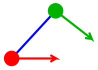


$\cos \angle(\mathbf{R}_i, \tilde{\mathbf{d}}_{ij}) > 0.9$

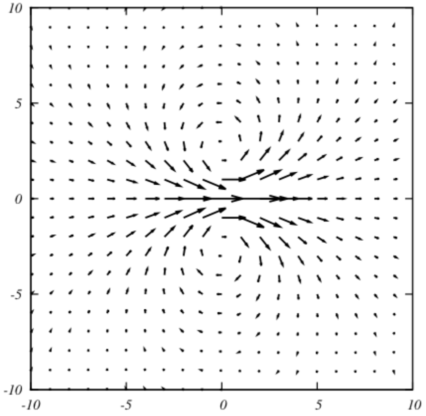
$\Rightarrow$ often $\mathbf{R}_j \parallel \tilde{\mathbf{d}}_{ij}$ as well

Displacement correlation: definition (1/2)

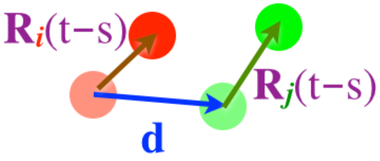


average  etc.

$$\Rightarrow \begin{cases} \left\langle (R_{xj}, R_{yj}) \right\rangle_{\tilde{\mathbf{d}}, \mathbf{R}_i} \\ \left\langle (R_{xi} R_{xj}, R_{xi} R_{yj}) \right\rangle_{\tilde{\mathbf{d}}} \end{cases}$$



$\mathbf{e}_1 \cdot \langle \mathbf{R} \otimes \mathbf{R} \rangle_{\tilde{\mathbf{d}}}$ (present)



$$\tilde{\mathbf{d}}_{ij} = \boldsymbol{r}_j(s) - \boldsymbol{r}_i(s)$$
$$\mathbf{R}_i = \boldsymbol{r}_i(t) - \boldsymbol{r}_i(s)$$
$$\mathbf{R}_j = \boldsymbol{r}_j(t) - \boldsymbol{r}_j(s)$$

cf.

Fig. 8,

Doliwa & Heuer,

PRE **61** (2000)

Displacement correlation: definition (2/2)

Tensorial indicator of cooperatively in colloidal systems:

displacement correlation tensor

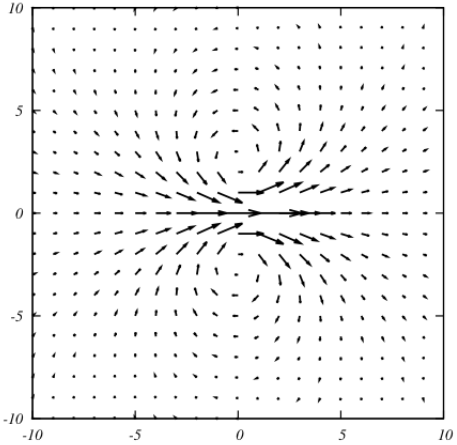
$$\langle \mathbf{R} \otimes \mathbf{R} \rangle_{\tilde{\mathbf{d}}} = \begin{bmatrix} \langle R_x R_x \rangle_{\tilde{\mathbf{d}}} & \langle R_x R_y \rangle_{\tilde{\mathbf{d}}} \\ \langle R_y R_x \rangle_{\tilde{\mathbf{d}}} & \langle R_y R_y \rangle_{\tilde{\mathbf{d}}} \end{bmatrix}$$

where

$$\mathbf{R} = (R_x(t,s), R_y(t,s)) = \mathbf{r}(t) - \mathbf{r}(s)$$

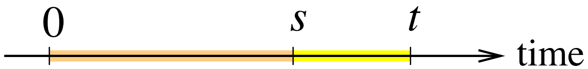
$(s < t)$

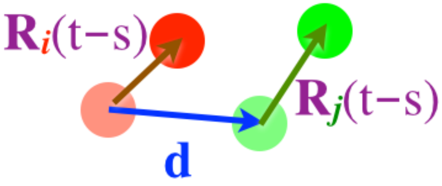
$\langle \quad \rangle_{\tilde{\mathbf{d}}}$ = average over pairs (i, j)
such that $\mathbf{r}_j(s) - \mathbf{r}_i(s) = \tilde{\mathbf{d}}$



from DNS

How can we reproduce the vortex pair (semi)analytically?





Outline

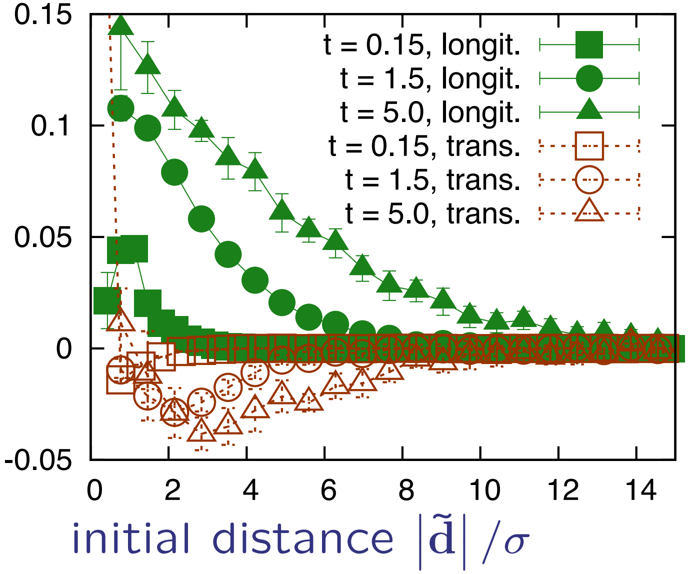
- Introduction
- Calculation of displacement corr. (DC) $\langle \mathbf{R} \otimes \mathbf{R} \rangle$
  - Direct numerical simulation (DNS) of 2D Brownian particles
  - 1D theory (single-file diffusion = SFD)
    linear & nonlinear
  - **2D linear theory [main result]**
    → analytical expression of $\langle \mathbf{R} \otimes \mathbf{R} \rangle$
- Discussion & perspectives

Longitudinal & transverse components of DC tensor

Assume isotropy (rotational & reflectional symmetries):

$$\langle \mathbf{R}\otimes\mathbf{R}\rangle_{\tilde{\mathbf{d}}} = \begin{bmatrix} \langle R_xR_x\rangle_{\tilde{\mathbf{d}}} & \langle R_xR_y\rangle_{\tilde{\mathbf{d}}} \\ \langle R_yR_x\rangle_{\tilde{\mathbf{d}}} & \langle R_yR_y\rangle_{\tilde{\mathbf{d}}} \end{bmatrix}$$

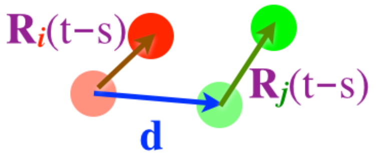
$$= \underbrace{X_{\parallel}(\tilde{d}/\ell_0, t-s)\frac{\tilde{\mathbf{d}}\otimes\tilde{\mathbf{d}}}{\tilde{\mathbf{d}}^2}}_{\text{longitudinal}} + \underbrace{X_{\perp}(\tilde{d}/\ell_0, t-s)\left(\mathbb{1} - \frac{\tilde{\mathbf{d}}\otimes\tilde{\mathbf{d}}}{\tilde{\mathbf{d}}^2}\right)}_{\text{transverse}}$$



DNS of Brownian particles

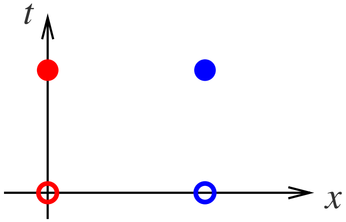
$N = 4000$, area fraction $\phi = 0.5$

$V_{\text{max}} = 50\, k_{\text{B}}T$

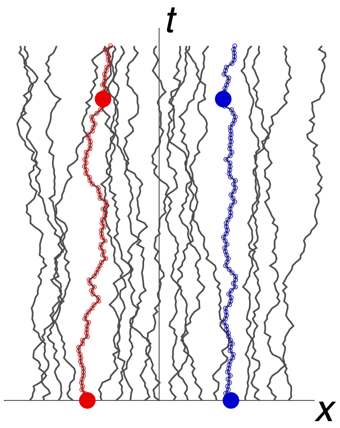


Problem:
calculate $X_{\parallel}$ and $X_{\perp}$ analytically

Idea for analytical calculation of DC: label variable



- DC is a space-time 4-point correlation
- Standard (Eulerian) field description:
  $\langle\rho(\mathbf{r}_1,0)\rho(\mathbf{r}_1,t)\rho(\mathbf{r}_2,0)\rho(\mathbf{r}_2,t)\rangle \leftarrow$ 4-body correlation
- Introduce label variables $\boldsymbol{\xi}=(\xi,\eta)$:
  curvilinear coordinate system sticking to the particles
  $\mathbf{r}(\boldsymbol{\xi},t+\Delta t)-\mathbf{r}(\boldsymbol{\xi},t)=\mathbf{u}(\boldsymbol{\xi},t)\Delta t$
  (Lagrangian description)
  time integr. $\mathbf{R}=\mathbf{R}(\boldsymbol{\xi},t)=\mathbf{r}(\boldsymbol{\xi},t)-\mathbf{r}(\boldsymbol{\xi},0)$
  displacement



- DC as a space-time 4-point correlation
  $\rightarrow$ 2-body Lagrangian correlation

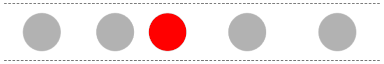
$*$ Eulerian/Lagrangian: terminology of fluid mechanics
in regard to choice of the independent variables, $(\mathbf{r},t)$ or $(\boldsymbol{\xi},t)$

Single-File Diffusion (SFD)

1D system of Brownian particles

+ "no-passing" repulsive interaction

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



Harris (1965), Jepsen (1965), Levitt (1973), ...

Slow diffusion: asymptotic behavior of MSD

$$\boxed{\left\langle R^2 \right\rangle = \frac{2S}{\rho_0}\sqrt{\frac{D^{\mathrm{c}}t}{\pi}}}$$

Hahn & Kärger, JPA **28** (1995)
Kollmann, PRL **90** (2003)

Ooshida *et al.*, JPSJ **80** (2011)

Continuum theory of SFD

Dean–Kawasaki eq.

$$\partial_t \rho + \partial_x Q = 0 \qquad \rho = \sum_j \rho_j, \quad \rho_j = \delta(x - X_j(t))$$

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

$$\Downarrow \ \text{change of variable}: \ \rho = \frac{\partial \xi}{\partial x} \text{ etc.}$$

$$\ell_0 \partial_t \psi(\xi, t) = \partial_\xi u(\xi, t) \qquad \psi = \frac{\rho_0}{\rho} - 1$$

$$u = -D \rho \partial_\xi \left(\log \frac{\rho}{\rho_0} + \frac{U}{k_{\mathrm{B}} T} \right) + \sum_j \delta(\xi - \Xi_j) f_j(t)$$

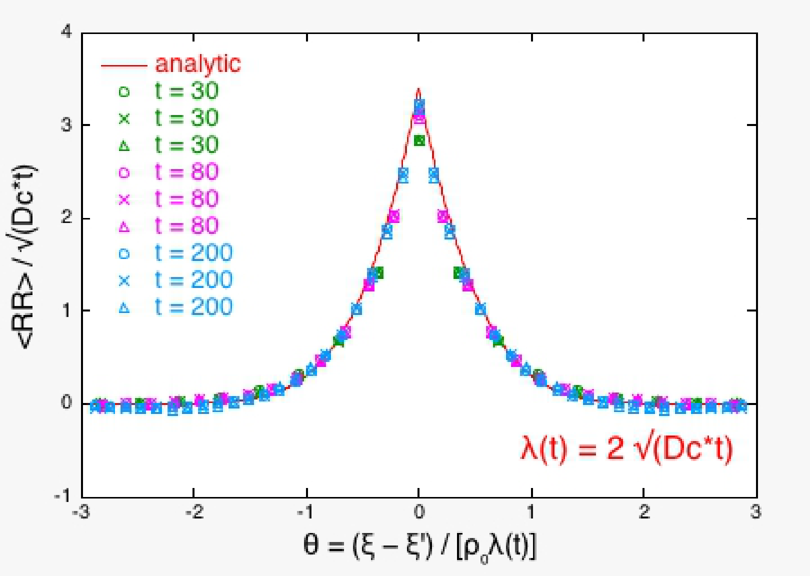
$$\Downarrow$$

correlations: $\langle \psi \psi \rangle = \cdots, \quad \langle R R \rangle = \cdots$

DC from Lagrangian continuum theory of SFD

$$\left\langle R(\xi,t)R(\xi',t)\right\rangle = \frac{2S}{\rho_0}\sqrt{\frac{D^{\mathrm{c}}t}{\pi}}\,\exp\left[-\frac{(\xi-\xi')^2}{4\rho_0^2 D^{\mathrm{c}}t}\right] - \frac{S}{\rho_0^2}|\xi-\xi'|\,\mathrm{erfc}\,\frac{|\xi-\xi'|}{2\rho_0\sqrt{D^{\mathrm{c}}t}} + [\mathrm{correction}]$$

dynamical corr. length: $\lambda(t) = 2\sqrt{D^{\mathrm{c}}t}$, grows in time diffusively



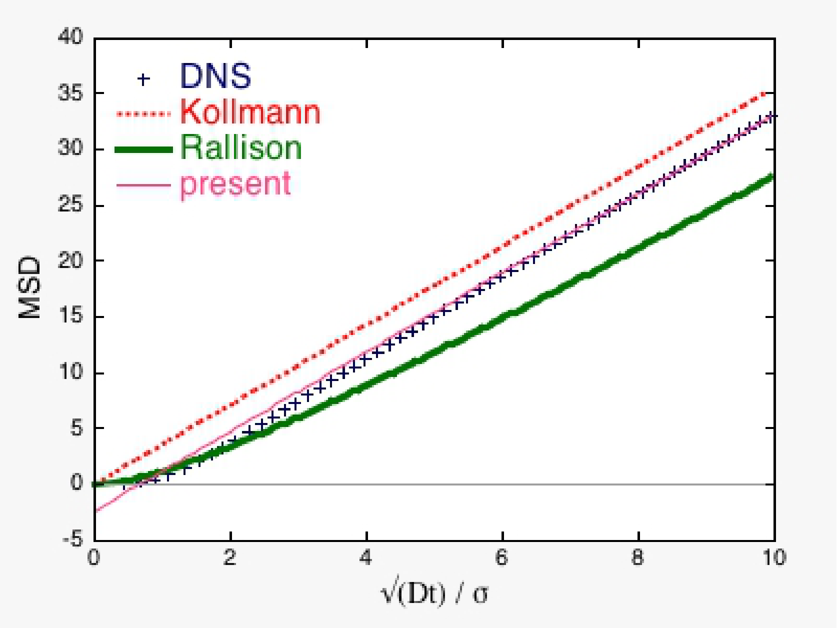
$$\theta \overset{\mathrm{def}}{=} \frac{\xi-\xi'}{\rho_0\lambda(t)} = \frac{\xi-\xi'}{2\rho_0\sqrt{D^{\mathrm{c}}t}}$$

$$\frac{\langle R(\xi,t)R(\xi',t)\rangle}{\sigma\sqrt{D^{\mathrm{c}}t}} \simeq \varphi(\theta)$$

$$= \frac{2S}{\rho_0\sigma}\left(\frac{e^{-\theta^2}}{\sqrt{\pi}} - |\theta|\,\mathrm{erfc}\,|\theta|\right)$$

← DNS (Brownian particles)
$N = 3000$
$\rho_0 = N/L = 0.2\,\sigma^{-1}$

MCT correction to asymptotic behavior of SFD



$\rho_0 \sigma = 0.25, \ S = 0.624$

Hahn & Kärger (1995);
Kollmann (2003)

$$\langle R^2 \rangle \simeq \frac{2S}{\rho_0} \sqrt{\frac{D^c t}{\pi}}$$

Rallison, JFM **186** (1988)

$$\langle R^2 \rangle = \frac{2S}{\rho_0} \sqrt{\frac{D^c t}{\pi}} - \frac{S}{\pi \rho_0^2} \log \left(1 + \rho_0 \sqrt{4\pi D^c t} \right)$$

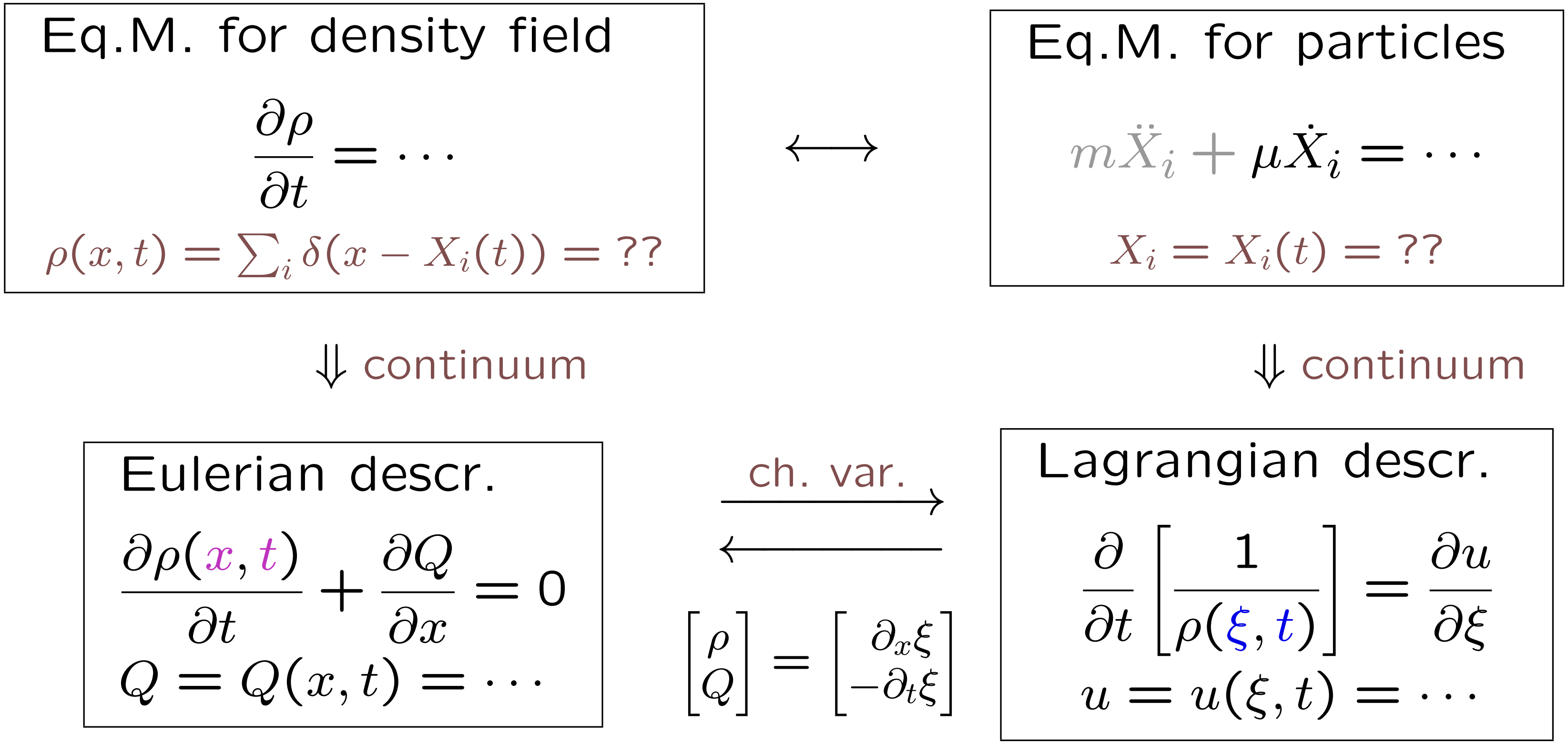
present (Lagrangian MCT)

$$\langle R^2 \rangle = \frac{2S}{\rho_0} \sqrt{\frac{D^c t}{\pi}} - \frac{\sqrt{2}}{3\pi} \rho_0^{-2}$$

How to extend the theory of SFD to 2D?

- How to construct Lagrangian description
- Strategy for calculation of DC
  - choice of field variable
  - conversion from the field variable to displacement

To switch from Eulerian to Lagrangian (1D)



Eq.M. for density field

$$\frac{\partial\rho}{\partial t} = \cdots$$

$\rho(x,t) = \sum_i \delta(x - X_i(t)) = ??$

$\longleftrightarrow$

Eq.M. for particles

$$m\ddot{X}_i + \mu\dot{X}_i = \cdots$$

$X_i = X_i(t) = ??$

$\Downarrow$ continuum

$\Downarrow$ continuum

Eulerian descr.

$$\frac{\partial\rho(x,t)}{\partial t} + \frac{\partial Q}{\partial x} = 0$$

$$Q = Q(x,t) = \cdots$$

ch. var.

$\longrightarrow$

$\longleftarrow$

$$\begin{bmatrix}\rho\\ Q\end{bmatrix} = \begin{bmatrix}\partial_x\xi\\ -\partial_t\xi\end{bmatrix}$$

Lagrangian descr.

$$\frac{\partial}{\partial t}\left[\frac{1}{\rho(\xi,t)}\right] = \frac{\partial u}{\partial\xi}$$

$$u = u(\xi,t) = \cdots$$

Ooshida *et al.*, JPSJ **80**, 074007 (2011); PRE **88**, 062108 (2013)
Spohn, J. Stat. Phys. **154**, 1191 (2014)

To switch from Eulerian to Lagrangian (2D)

Eq.M. for density field

$$\frac{\partial\rho}{\partial t} = \cdots$$

$$\rho(\mathbf{r},t) = \textstyle\sum_i \delta(\mathbf{r}-\mathbf{r}_i(t)) = ??$$

$$\longleftrightarrow$$

Eq.M. for particles

$$m\ddot{\mathbf{r}}_i + \mu\dot{\mathbf{r}}_i = \cdots$$

$$\mathbf{r}_i = \mathbf{r}_i(t) = ??$$

$\Downarrow$ continuum

$$\cancel{\Downarrow}$$

Eulerian descr.

$$\frac{\partial\rho(\mathbf{r},t)}{\partial t} + \nabla\cdot\mathbf{Q} = 0$$

$$\mathbf{Q} = \mathbf{Q}(\mathbf{r},t) = \cdots$$

$$\xrightarrow{\text{ch. var.}}$$

$$\longleftarrow$$

$$\begin{bmatrix}\rho\\ \mathbf{Q}\end{bmatrix} = \begin{vmatrix} \mathbf{e}_0 & \partial_t\xi & \partial_t\eta \\ \mathbf{e}_1 & \partial_x\xi & \partial_x\eta \\ \mathbf{e}_2 & \partial_y\xi & \partial_y\eta \end{vmatrix}$$

Lagrangian descr.

$$\ell_0\frac{\partial\Psi_a(\boldsymbol{\xi},t)}{\partial t} = \frac{\partial u_a}{\partial\xi_a}$$

$$\mathbf{u} = \mathbf{u}(\boldsymbol{\xi},t) = \cdots$$

N.B.: regular numbering (crystalline order) is not required

Ooshida *et al.*, arXiv:1507.05714 (to appear in Biophys. Rev. Lett.)

Strategy for calculation of DC in 2D

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

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

- 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 $\nabla = (\nabla \xi)\partial_\xi + (\nabla \eta)\partial_\eta$ → expressible with Ψ

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

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

Alexander & Pincus, PRB **18** (1978); Oo. *et al.*, arXiv:1507.05714

Linear analysis in 2D

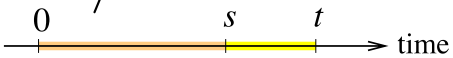
Langevin noise
↓

Fourier repr.
+ linearization

$$\boxed{\partial_t \begin{bmatrix}\check{\Psi}_1\\ \check{\Psi}_2\end{bmatrix} = -D_*^{\mathsf{c}} \begin{bmatrix}k_1^2 & k_1^2\\ k_2^2 & k_2^2\end{bmatrix} \begin{bmatrix}\check{\Psi}_1\\ \check{\Psi}_2\end{bmatrix} + \begin{bmatrix}\check{f}_1\\ \check{f}_2\end{bmatrix}}$$

eigenvalues $\nu_{\mathsf{d}} = D_*^{\mathsf{c}}\mathbf{k}^2$ and $\nu_{\mathsf{r}} = 0$

calculate $C_{\alpha\beta}(\mathbf{k},t,s) \overset{\mathsf{def}}{=} \frac{N}{L^4}\left\langle \check{\Psi}_\alpha(\mathbf{k},t)\check{\Psi}_\beta(-\mathbf{k},s)\right\rangle$



$$C_{11}(\mathbf{k},t,s) = \left(\frac{k_1^2}{\mathbf{k}^2}\right)^2 \frac{S}{L^4}\,\mathsf{exp}\left(-D_*^{\mathsf{c}}\mathbf{k}^2(t-s)\right) + \frac{k_1^2k_2^2}{\mathbf{k}^2}\frac{2D_*}{L^4}s$$

from eigenvalue $\mathbf{k}^2$ from eigenvalue 0

$$C_{22}(\mathbf{k},t,s) = \cdots$$
$$C_{12}(\mathbf{k},t,s) = C_{21}(\mathbf{k},t,s) = \cdots$$

"d"-mode $C_{\mathsf{d}}(t,s) \propto e^{-\nu_{\mathsf{d}}(t-s)}$; "r"-mode $C_{\mathsf{r}}(t,s) \propto s$

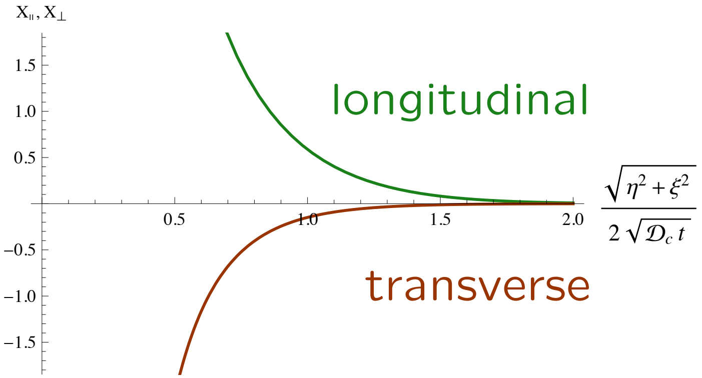
$$C_{\alpha\beta} \propto \left\langle \check{\Psi}_\alpha\check{\Psi}_\beta\right\rangle \xrightarrow{\mathsf{AP\ formula}} \langle \mathbf{R}\otimes\mathbf{R}\rangle = \begin{bmatrix}\langle R_xR_x\rangle & \langle R_xR_y\rangle\\ \langle R_yR_x\rangle & \langle R_yR_y\rangle\end{bmatrix}$$

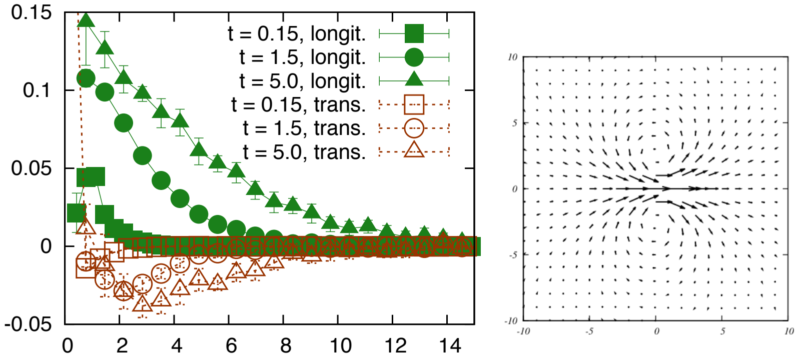
DC from 2D linear solution

$$X_{\|}(\xi, t-s) = \frac{S}{4\pi\rho_0}\left\{\vartheta^{-2}\exp\left(-\vartheta^{2}\right) + E_1\left(\vartheta^{2}\right)\right\}$$

$$X_{\perp}(\xi, t-s) = \frac{S}{4\pi\rho_0}\left\{-\vartheta^{-2}\exp\left(-\vartheta^{2}\right) + E_1\left(\vartheta^{2}\right)\right\}$$

$$\vartheta = \frac{|\xi-\xi'|}{2\sqrt{D_*^{\mathrm{C}}(t-s)}}, \qquad E_1(z) = \int_z^{\infty} \frac{\exp(-\zeta)}{\zeta}\mathrm{d}\zeta$$





DNS of Brownian particles

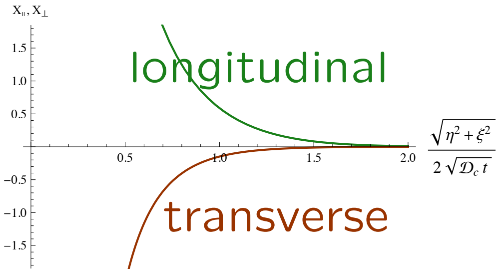
$X_{\|} > 0$ and $X_{\perp} < 0$ everywhere

$X_{\perp} \to -\infty$ for $\vartheta \to +0$ limitation of linear analysis

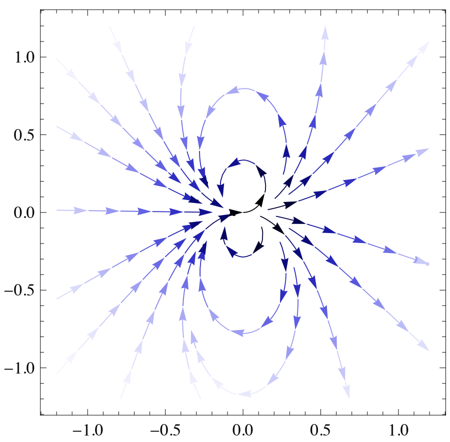
DC from 2D linear theory captures vortical motion

$$X_\parallel(\xi, t-s) = \frac{S}{4\pi\rho_0}\left\{\vartheta^{-2}\exp\left(-\vartheta^2\right)+E_1\left(\vartheta^2\right)\right\}$$

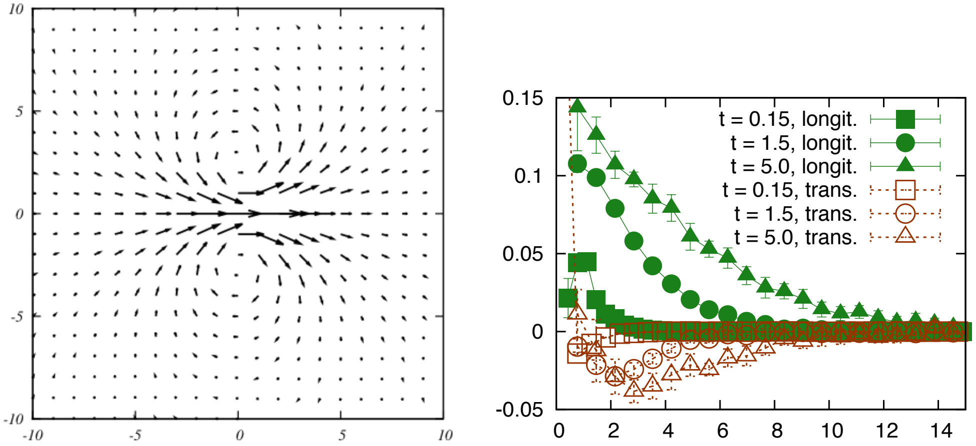
$$X_\perp(\xi, t-s) = \frac{S}{4\pi\rho_0}\left\{-\vartheta^{-2}\exp\left(-\vartheta^2\right)+E_1\left(\vartheta^2\right)\right\}$$



$$\tilde{\mathrm{d}} \simeq \ell_0(\boldsymbol{\xi}-\boldsymbol{\xi}'), \quad \vartheta = \frac{|\boldsymbol{\xi}-\boldsymbol{\xi}'|}{2\sqrt{D_*^c(t-s)}}, \quad E_1(z) = \int_z^\infty \frac{e^{-\zeta}}{\zeta}\mathrm{d}\zeta$$



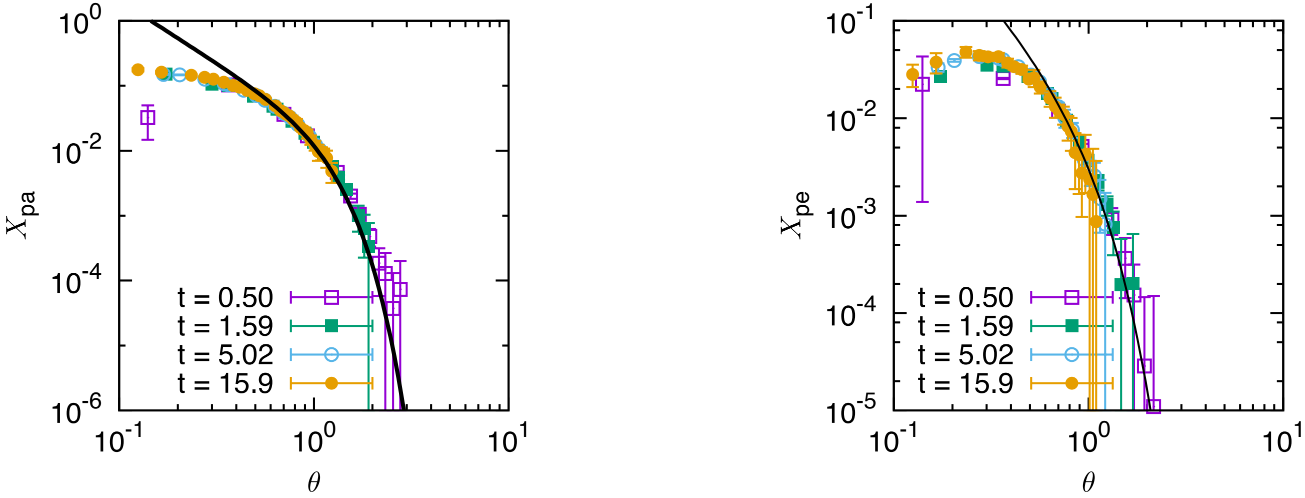
analytical



DNS for Brownian particles

Displacement correlation vs similarity variable

longitudinal corr. $X_\parallel$ vs ϑ transverse corr. $X_\perp$ vs ϑ



$$\vartheta = \frac{|\boldsymbol{\xi} - \boldsymbol{\xi}'|}{2\sqrt{D_*^{\mathrm{c}} t}} \simeq \frac{|\tilde{\mathrm{d}}|}{2\sqrt{D^{\mathrm{c}} t}}$$

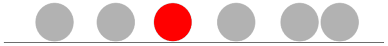
- DCs for different intervals collapse onto a single curve
- The master curve agrees with the linear theory on the long-wave side (but not on the short-wave side)

Discussion & perspectives

- Displacement correlation $\langle \mathbf{R} \otimes \mathbf{R} \rangle$

  expressible with $\vartheta = \frac{|\boldsymbol{\xi} - \boldsymbol{\xi}'|}{2\sqrt{D^{\mathsf{C}}_{*} t}} \simeq \frac{|\tilde{\mathbf{d}}|}{2\sqrt{D^{\mathsf{C}} t}}$

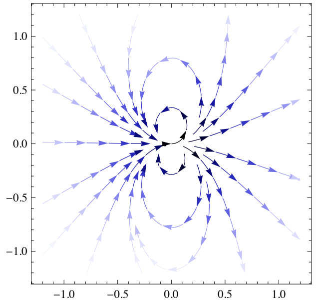
  cages are nested



- Limitation of the linear theory:

  disagreement on the short-wave side

  need of a nonlinear theory, such as MCT



- Possible advantage over the standard MCT:
  - DC is tensorial and can capture the vortex pair
  - The field variable Ψ is also tensorial

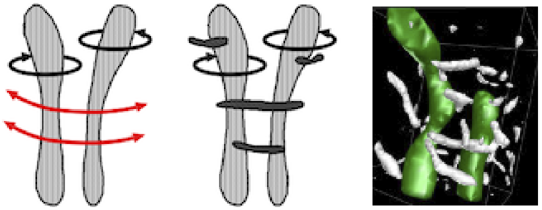
Nonlinear theory: mode-coupling theory (MCT)

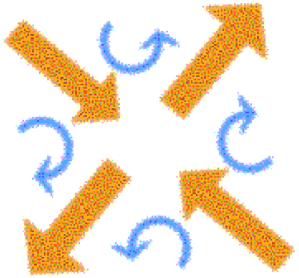
$$(\partial_t + D^{\mathrm{c}} k^2) C(t,s) + \int M(t,u)\, \partial_u C(u,s) \mathrm{d}u = 0, \qquad M \sim \sum CC$$

- standard MCT: $M \sim \langle \rho\rho \rangle \langle \rho\rho \rangle$ insensitive to vortex quads
- tensorial MCT: $M \sim \langle \Psi\Psi \rangle \langle \Psi\Psi \rangle$ can detect them

vortices & shear
in turbulence

Goto, PTPS **195** (2012)





plastic event

in sheared

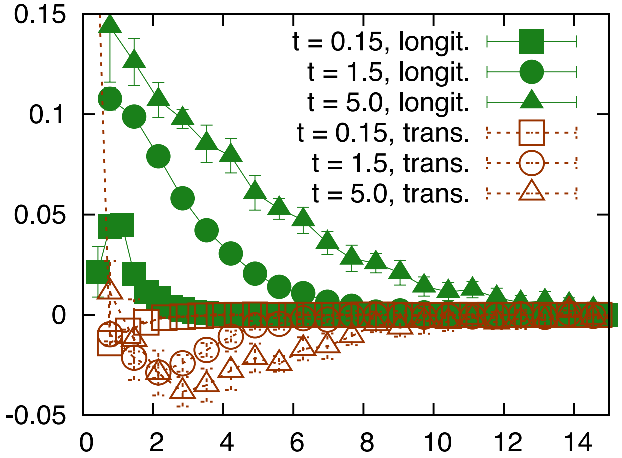
amorphous solid

cf. Dasgupta *et al.*, PRE **87** (2013)

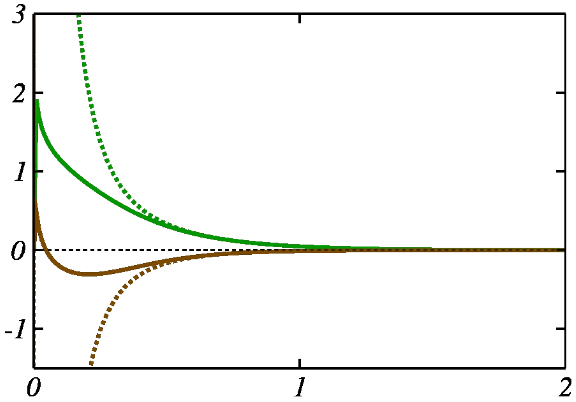
DC suggested by preliminary nonlinear analysis

$$X_{\parallel}(\xi,t) \propto \frac{e^{-\vartheta^2} - e^{-\vartheta^2/\mu_r}}{\vartheta^2} + E_1\left(\vartheta^2\right) + E_1\left(\vartheta^2/\mu_r\right)$$

$$X_{\perp}(\xi,t) \propto -\frac{e^{-\vartheta^2} - e^{-\vartheta^2/\mu_r}}{\vartheta^2} + E_1\left(\vartheta^2\right) + E_1\left(\vartheta^2/\mu_r\right)$$

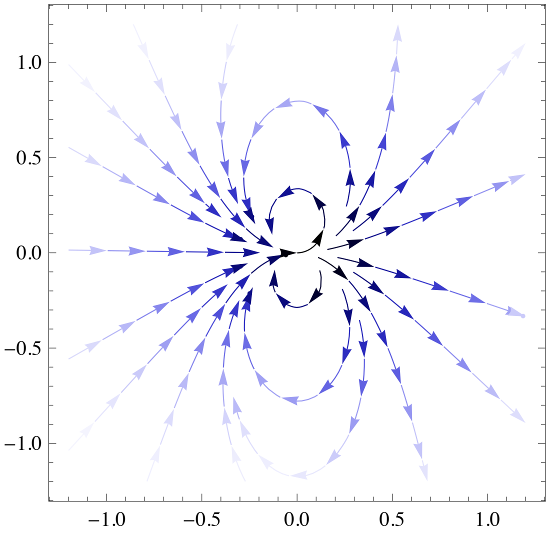


initial distance $\left|\tilde{\mathrm{d}}\right|/\sigma$

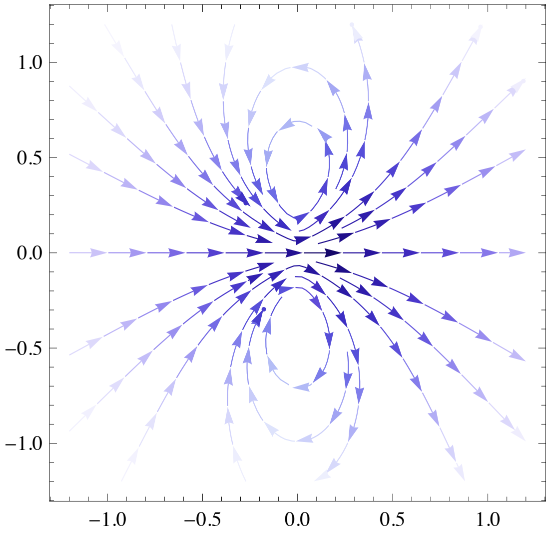


$$\vartheta = \frac{|\xi-\xi'|}{2\sqrt{D^{\mathrm{c}}_{*}t}} \simeq \frac{\left|\tilde{\mathrm{d}}\right|}{2\sqrt{D^{\mathrm{c}}t}}$$

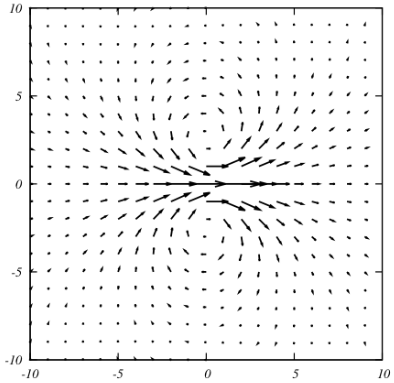
Implications of nonlinear correction for vortex pair



linear analysis



+ correction



Concluding remarks

- DC obtained analytically from 2D linear theory

$$X_{\parallel}(\xi, t-s) = \frac{S}{4\pi\rho_0}\left\{\vartheta^{-2}\exp\left(-\vartheta^2\right) + E_1\left(\vartheta^2\right)\right\}$$

$$X_{\perp}(\xi, t-s) = \frac{S}{4\pi\rho_0}\left\{-\vartheta^{-2}\exp\left(-\vartheta^2\right) + E_1\left(\vartheta^2\right)\right\}$$

where $\vartheta = \frac{|\xi-\xi'|}{2\sqrt{D_*^{\mathrm{C}}(t-s)}}$

- Nonlinear theory: tensorial MCT
  - memory kernel $M \sim \langle\Psi\Psi\rangle\langle\Psi\Psi\rangle$ sensitive to vortex quads (plastic events)
  - preliminary analysis: modification to linear theory, finite vortex–vortex distance