Brownian dynamics of confined suspensions of driven active colloids

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Non-Spherical Colloids near Boundaries

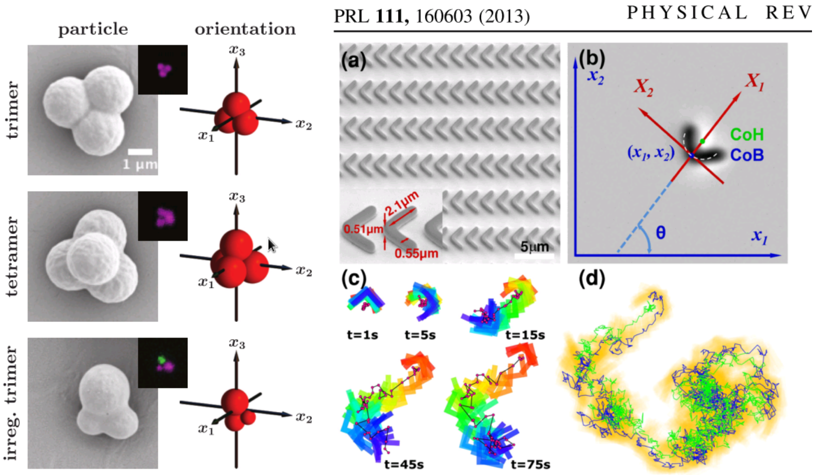


Figure: (Left) Cross-linked spheres; Kraft et al. (PRE 2013). (Right) Lithographed boomerangs; Chakrabarty et al. (PRL 2013).

Light-Activated Diffusion/Osmophoresis

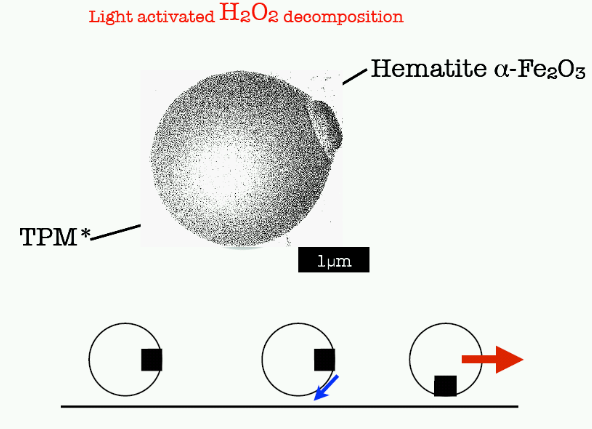
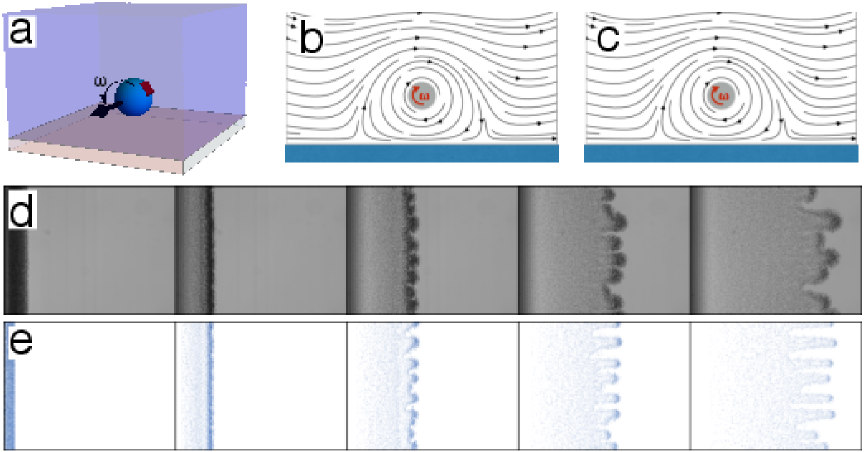


Figure: From Jeremie Palacci (now UCSD), was at Paul Chaikin lab (NYU Physics) (Science 2013).

Light-Activated Colloidal Surfers

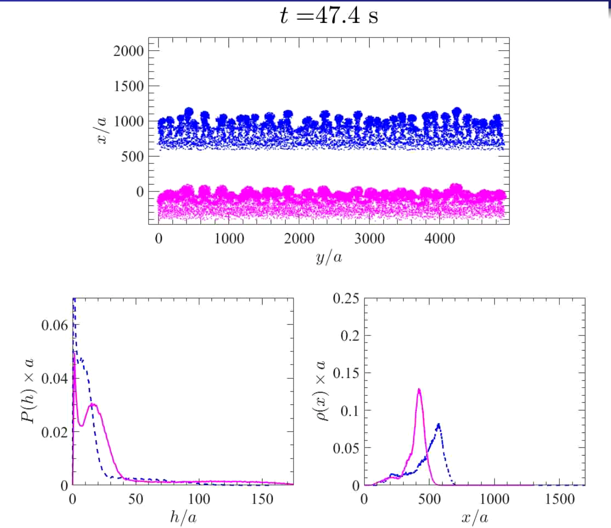
Movie from Jeremie Palacci

Microrollers: Fingering Instability



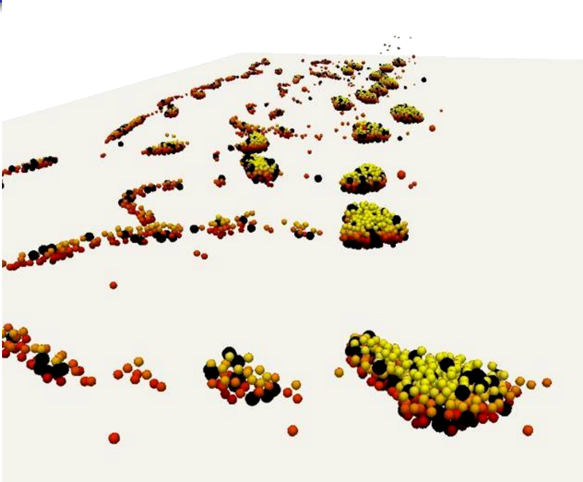
Experiments by Michelle Driscoll (was in the Chaikin lab at NYU Physics, now at Northwestern Physics), simulations by **Blaise Delmotte** (was at Courant, now at LadHyX Paris) [1, 2].

Role of Brownian Motion



Simulations show that thermal fluctuation are quantitatively important because they set the **gravitational height** [2].

Critters

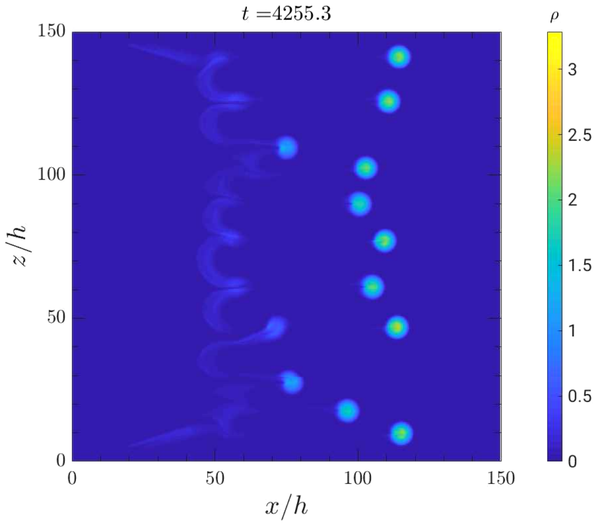


Simulations by **Blaise Delmotte** revealed that stable motile clusters termed **critters can form purely by hydrodynamic interactions** [1]. Still trying to create critters that don't shed particles in the lab...

Continuum models of rollers: 2D

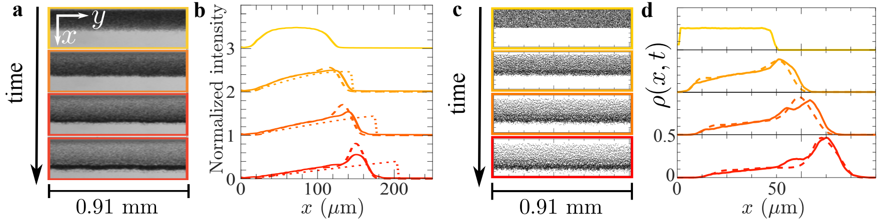
- Consider an infinite sheet of rotlets with **planar density** $\rho(x, y, t)$, which is fixed at a height $z = h$ given by the gravitational height
$$h = a + k_B T/mg.$$
- A **point torque** $T\hat{\mathbf{y}}$ at $(x', y';\ h)$ induces a fluid velocity in the (x, y) plane given by the kernel (**Green's function**), with $S = 3T/(4\pi\eta)$,
$$K_x\left(x - x', y - y';\ h\right) = Sh\frac{(x-x')^2}{\left[(x-x')^2+(y-y')^2+4h^2\right]^{5/2}},$$
$$K_y\left(x - x', y - y';\ h\right) = Sh\frac{(x-x')(y-y')}{\left[(x-x')^2+(y-y')^2+4h^2\right]^{5/2}}.$$
- The conservation law for the rotlet density in the sheet is given by the **nonlocal conservation law PDE**
$$\frac{\partial \rho(x, y, t)}{\partial t} = -\frac{\partial\left(\rho\left(K_x * \rho\right)\right)}{\partial x} - \frac{\partial\left(\rho\left(K_y * \rho\right)\right)}{\partial y}, \qquad (1)$$
where $*$ denotes convolution, $\left(K * \rho\right)(\mathbf{r}) = \int K\left(\mathbf{r} - \mathbf{r}'\right)\rho\left(\mathbf{r}'\right)\, d\mathbf{r}'$.

Deterministic 2D non-local conservation law



Quasi-2D simulations show that continuum deterministic models can reproduce the fingering instability.

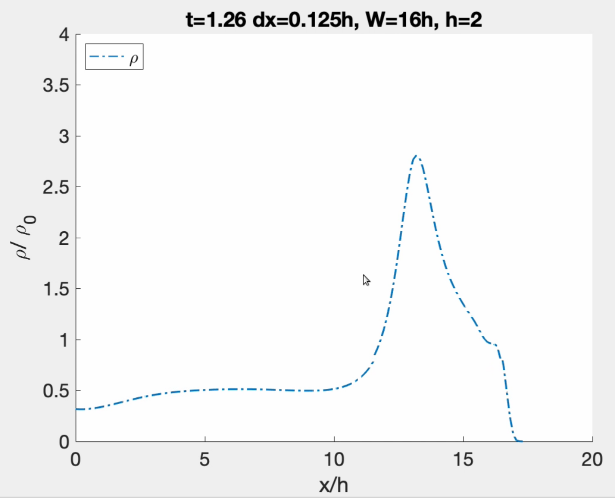
Continuum models of rollers: 1D



$$\frac{\partial \rho(x,t)}{\partial t} = -\frac{\partial\left[\rho\left(K*\rho\right)\right]}{\partial x}, \tag{2}$$

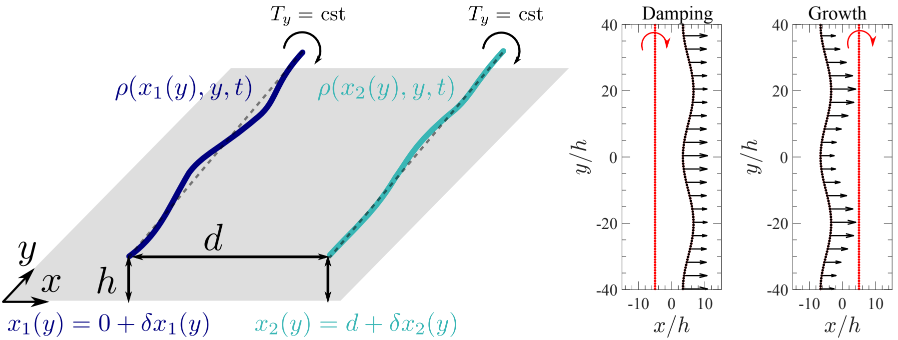
$$K = \int_{-\infty}^{+\infty} K_x(x-x',y;\ h)dy = \frac{4Sh}{3}\frac{(x-x')^2}{\left[(x-x')^2+4h^2\right]^2}.$$

Deterministic 1D non-local conservation law



Quasi-1D simulations show the formation of a **front of finite width** $\sim (5-10)h$ (made by Zhe Chen using aperiodic FFT-based convolutions + BDS advection by Wenjun Zhao).

Continuum models of fingering instability



Linear stability analysis of fingering: Set $d = 10h$ based on 1D model. The stability analysis shows that the line in the back is **linearly unstable** with dominant wavelength $\sim 10h$.

Minimally-Resolved Simulations

- Represent each spherical particle by a **single blob**, and solve the Ito equations of **Brownian HydroDynamics** for the (correlated) positions of the N spherical microrollers $\mathbf{Q}(t) = \{\mathbf{q}_1(t), \ldots, \mathbf{q}_N(t)\}$,

$$d\mathbf{Q} = \boldsymbol{\mathcal{M}}\mathbf{F}dt + \boldsymbol{\mathcal{M}}_c\mathbf{T}dt + (2k_B T\,\boldsymbol{\mathcal{M}})^{\frac{1}{2}}\,d\boldsymbol{\mathcal{B}} + k_B T\,(\partial_{\mathbf{Q}} \cdot \boldsymbol{\mathcal{M}})\,dt, \quad (3)$$

  where $\boldsymbol{\mathcal{B}}(t)$ is a vector of Brownian motions, and $\mathbf{F}(\mathbf{Q})$ are applied forces, and $\mathbf{T}$ the external magnetic torques.
- How to compute **deterministic velocities** $\boldsymbol{\mathcal{M}}\mathbf{F}$ efficiently?
- How to generate **Brownian increments** $(2k_B T\,\boldsymbol{\mathcal{M}})^{\frac{1}{2}}\,\Delta\boldsymbol{\mathcal{B}}$ efficiently?
- How to generate **stochastic drift** $k_B T\,(\partial_{\mathbf{Q}} \cdot \boldsymbol{\mathcal{M}})$ efficiently by only solving mobility problems?

Blobs in Stokes Flow

- The symmetric positive semidefinite (SPD) **blob-blob mobility matrix** $\boldsymbol{\mathcal{M}}$ encodes the hydrodynamics:
  3×3 block $\mathbf{M}_{ij}$ maps a force on blob j to a velocity of blob i.
- The mobility is approximated to have a far-field **pairwise approximation**

$$\mathbf{M}_{ij}(\mathbf{Q}) \equiv \mathbf{M}_{ij}\left(\mathbf{q}_i, \mathbf{q}_j\right) = \boldsymbol{\mathcal{R}}\left(\mathbf{q}_i, \mathbf{q}_j\right),$$

where the **hydrodynamic kernel** $\boldsymbol{\mathcal{R}}$ for spheres of radius a is

$$\boldsymbol{\mathcal{R}}\left(\mathbf{q}_i, \mathbf{q}_j\right) \approx \eta^{-1}\left(\mathbf{I} + \frac{a^2}{6}\boldsymbol{\nabla}^2_{\mathbf{r}'}\right)\left(\mathbf{I} + \frac{a^2}{6}\boldsymbol{\nabla}^2_{\mathbf{r}''}\right)\mathbb{G}(\mathbf{r}', \mathbf{r}'')\Big|_{\mathbf{r}''=\mathbf{q}_i}^{\mathbf{r}'=\mathbf{q}_j} \quad (4)$$

where $\mathbb{G}$ is the **Green's function** for steady Stokes flow, *given* the appropriate boundary conditions.

Confined Geometries

- The Green's function is only known explicitly in some very special circumstances, e.g., for a **single no-slip boundary** $\mathbb{G}$ is the **Oseen-Blake** tensor.
- For blobs next to a wall the **Rotne-Prager-Blake** tensor has been computed by Swan (MIT) and Brady (Caltech) and we will use it here. It is still missing corrections when the blobs overlap the wall so we have made a heuristic fix [2].
- We compute $\boldsymbol{\mathcal{M}\lambda}$ using **GPU-accelerated** $O(N_b^2)$ sum. Often faster than Fast Multipole Methods for up to 10^5 blobs.
- For slit channels we can use a grid-based **fluid Stokes solver** to compute the (action of the) **Green's functions on the fly** [3] In the triply periodic case [4] or explicit Stokes solver [3] approach adding thermal fluctuations (Brownian motion) can be done using **fluctuating hydrodynamics**.

Generating Brownian increments

- We want to sample random **Brownian "velocities"** with covariance $\boldsymbol{\mathcal{M}}$:

$$\mathbf{U}_b = \sqrt{\frac{2k_B T}{\Delta t}}\, \boldsymbol{\mathcal{M}}^{\frac{1}{2}} \mathbf{W} \quad \Rightarrow \quad \langle \mathbf{U}_b \mathbf{U}_b^T \rangle = \left(\frac{2k_B T}{\Delta t} \right) \boldsymbol{\mathcal{M}}$$

- The product $\boldsymbol{\mathcal{M}}^{\frac{1}{2}} \mathbf{W}$ can be computed iteratively by **repeated multiplication** of a vector by $\boldsymbol{\mathcal{M}}$ using (preconditioned) Krylov subspace **Lanczos methods**.
- Close to a bottom wall pairwise hydrodynamic interactions **decay rapidly like** $1/r^3$, so the Krylov method converges in a **small constant number of iterations**, without any preconditioning.
- One can use **fluctuating hydrodynamics** to generate $\boldsymbol{\mathcal{M}}^{\frac{1}{2}} \mathbf{W}$ with only a few **FFTs** in near **linear time** for periodic suspensions (also works with **multigrid**) [4, 3].

Stochastic drift term

$$\frac{d\mathbf{Q}(t)}{dt} = \mathcal{M}\mathbf{F} + (2k_BT\,\mathcal{M})^{\frac{1}{2}}\,\mathcal{W}(t) + (k_BT)\,\partial_{\mathbf{Q}}\cdot\mathcal{M}$$

- Key idea to get $(\partial_{\mathbf{Q}}\cdot\mathcal{M})_i = \partial\mathcal{M}_{ij}/\partial Q_j$ is to use **random finite differences (RFD)** [2]: If $\langle\Delta\mathbf{P}\Delta\mathbf{Q}^T = \mathbf{I}\rangle$,

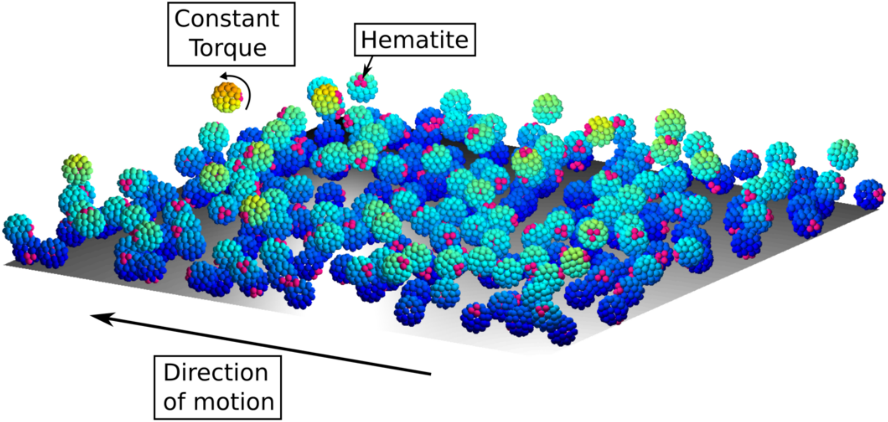
$$\lim_{\delta\to 0}\frac{1}{\delta}\langle\left\{\mathcal{M}\left(\mathbf{Q}+\frac{\delta}{2}\Delta\mathbf{Q}\right)-\mathcal{M}\left(\mathbf{Q}-\frac{\delta}{2}\Delta\mathbf{Q}\right)\right\}\Delta\mathbf{P}\rangle = \qquad (5)$$

$$\{\partial_{\mathbf{Q}}\mathcal{M}(\mathbf{Q})\} : \langle\Delta\mathbf{P}\Delta\mathbf{Q}^T\rangle = k_BT\,\partial_{\mathbf{Q}}\cdot\mathcal{M}(\mathbf{Q})\,. \qquad (6)$$

- This leads to a **stochastic Adams-Bashforth** temporal integrator [2],

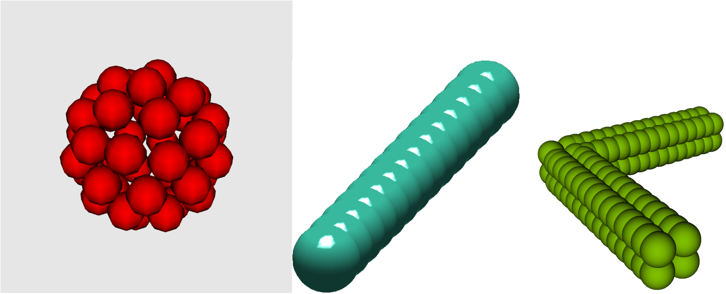
$$\frac{\mathbf{Q}^{n+1}-\mathbf{Q}^n}{\Delta t} = \left(\frac{3}{2}\mathcal{M}^n\mathbf{F}^n - \frac{1}{2}\mathcal{M}^{n-1}\mathbf{F}^{n-1}\right) + \sqrt{\frac{2k_BT}{\Delta t}}\,(\mathcal{M}^n)^{\frac{1}{2}}\,\mathbf{W}^n$$
$$+\frac{k_BT}{\delta}\left(\mathcal{M}\left(\mathbf{Q}+\frac{\delta}{2}\widetilde{\mathbf{W}}^n\right)-\mathcal{M}\left(\mathbf{Q}-\frac{\delta}{2}\widetilde{\mathbf{W}}^n\right)\right)\widetilde{\mathbf{W}}^n.$$

Microrollers: Uniform Suspension



Simulations by **Brennan Sprinkle**+Blaise Delmotte [3] of a uniform suspension of microrollers at packing fraction $\phi = 0.4$ (GIF). Compare to experiments (AVI) by **Michelle Driscoll**.

Rigid MultiBlob Models



- The rigid body is discretized through a number of "**beads**" or "**blobs**" with hydrodynamic radius *a*.
- Standard is **stiff springs** but we want **rigid multiblobs**.
- Equivalent to a **(smartly!) regularized first-kind boundary integral formulation**.
- **We can efficiently simulate the driven and Brownian motion of the rigid multiblobs.**

Rigid MultiBlobs

- We add **rigidity forces** as Lagrange multipliers $\boldsymbol{\lambda} = \{\boldsymbol{\lambda}_1, \dots, \boldsymbol{\lambda}_n\}$ to constrain a group of blobs forming body p to move rigidly,

$$\begin{aligned}\sum_j \mathcal{M}_{ij}\boldsymbol{\lambda}_j =& \mathbf{u}_p + \boldsymbol{\omega}_p \times \left(\mathbf{r}_i - \mathbf{q}_p\right) \qquad (7)\\ \sum_{i \in \mathcal{B}_p} \boldsymbol{\lambda}_i =& \mathbf{f}_p \\ \sum_{i \in \mathcal{B}_p} (\mathbf{r}_i - \mathbf{q}_p) \times \boldsymbol{\lambda}_i =& \boldsymbol{\tau}_p.\end{aligned}$$

  where $\mathbf{u}$ is the velocity of the tracking point $\mathbf{q}$, $\boldsymbol{\omega}$ is the angular velocity of the body around $\mathbf{q}$, $\mathbf{f}$ is the total force applied on the body, $\boldsymbol{\tau}$ is the total torque applied to the body about point $\mathbf{q}$, and $\mathbf{r}_i$ is the position of blob i.

- This can be a **very large linear system** for suspensions of many bodies discretized with many blobs:
  Use **iterative solvers** with a **good preconditioner**.

Suspensions of Rigid Bodies

- In matrix notation we have a **saddle-point** linear system of equations for the rigidity forces $\boldsymbol{\lambda}$ and unknown motion $\mathbf{U}$,

$$\begin{bmatrix} \mathcal{M} & -\mathcal{K} \\ \mathcal{K}^T & \mathbf{0} \end{bmatrix} \begin{bmatrix} \boldsymbol{\lambda} \\ \mathbf{U} \end{bmatrix} = \begin{bmatrix} \breve{\mathbf{u}} \\ \mathbf{F} \end{bmatrix}. \tag{8}$$

Same as **first-kind boundary integral methods**!

- The **surface velocity** $\breve{\mathbf{u}}$ can be used to model **active slip** or to generate **Brownian velocities** [3].
- Solution gives the **mobility matrix**

$$\mathcal{N} = \left(\mathcal{K}^T \mathcal{M}^{-1} \mathcal{K}\right)^{-1} \tag{9}$$

$$\mathbf{U} = \mathcal{N}\mathbf{F} - \left(\mathcal{N}\mathcal{K}^T \mathcal{M}^{-1}\right) \breve{\mathbf{u}}$$

Lubrication for **spherical colloids**

- Use **Stokesian Dynamics** approach introduced by Brady, but with more accurate rigid multiblob "far-field" mobility:

$$\begin{pmatrix} \mathcal{M} & -\mathcal{K} \\ \mathcal{K}^T & \boldsymbol{\Delta}_{MB} \end{pmatrix} \begin{pmatrix} \boldsymbol{\lambda} \\ \mathbf{U} \end{pmatrix} = \begin{pmatrix} -\breve{\mathbf{u}} \\ \mathbf{F} \end{pmatrix}, \tag{10}$$

- $\boldsymbol{\Delta}_{MB}$ is a **lubrication correction to the resistance matrix** formed by adding **pairwise** contributions for each pair of nearby surfaces (either particle-particle or particle-wall).
- The pairwise terms in $\boldsymbol{\Delta}_{MB}$ can be computed analytically using asymptotic expansion (for very close particles) or tabulated by using a more accurate reference method (e.g., boundary integral).
- Lubrication-corrected mobility matrix

$$\overline{\mathcal{N}} = \left[\mathcal{N}^{-1} + \boldsymbol{\Delta}_{MB}\right]^{-1} = \mathcal{N} \cdot \left[\mathbf{I} + \boldsymbol{\Delta}_{MB} \cdot \mathcal{N}\right]^{-1}.$$

Linear Algebra

- Without lubrication corrections, we have had great success with the indefinite **block-diagonal preconditioner**

$$\mathcal{P} = \begin{bmatrix} \mathcal{M}_{\text{diag}} & -\mathcal{K} \\ \mathcal{K}^T & \mathbf{0} \end{bmatrix} \tag{11}$$

  where we neglect all hydrodynamic interactions between blobs on distinct bodies in the preconditioner.
- For the **mobility problem**, we find a **small constant number of GMRES iterations** independent of the number of rigid multiblobs.
- For minimally-resolved single blob models we get the saddle-point system

$$\begin{pmatrix} \mathcal{N}_{\text{min}} & -I \\ I & \boldsymbol{\Delta}_{\text{min}} \end{pmatrix} \begin{pmatrix} \boldsymbol{\lambda} \\ \mathbf{U} \end{pmatrix} = \begin{pmatrix} -\breve{\mathbf{u}} \\ \mathbf{F} \end{pmatrix},$$

  where $\mathcal{N}_{\text{min}}$ is the generalized RPY mobility **including rotation**. Brennan Sprinkle is working on preconditioners.

Generating Brownian Displacements $\sim \mathcal{N}^{\frac{1}{2}}\mathbf{W}$

- Assume that we knew how to efficiently generate Brownian blob velocities $\mathcal{M}^{\frac{1}{2}}\mathbf{W}$ (PSE for periodic, Lanczoz for sedimented suspensions, fluctuating Stokes solver for slit channels).
  For rigid multiblobs use the **block-diagonal preconditioner** in the Lancsoz iteration.
- *Key idea*: Solve the mobility problem with random slip $\breve{\mathbf{u}}$,

$$\begin{bmatrix} \mathcal{M} & -\mathcal{K} \\ -\mathcal{K}^T & \mathbf{0} \end{bmatrix} \begin{bmatrix} \boldsymbol{\lambda} \\ \mathbf{U} \end{bmatrix} = - \begin{bmatrix} \breve{\mathbf{u}} = (2k_B T)^{1/2} \mathcal{M}^{\frac{1}{2}}\mathbf{W} \\ \mathbf{F} \end{bmatrix}, \qquad (12)$$

$$\mathbf{U} = \mathcal{N}\mathbf{F} + (2k_B T)^{\frac{1}{2}} \mathcal{N}\mathcal{K}^T\mathcal{M}^{-1}\mathcal{M}^{\frac{1}{2}}\,\mathbf{W} = \mathcal{N}\mathbf{F} + (2k_B T)^{\frac{1}{2}} \mathcal{N}^{\frac{1}{2}}\mathbf{W}.$$

which defines a $\mathcal{N}^{\frac{1}{2}} = \mathcal{N}\mathcal{K}^T\mathcal{M}^{-1}\mathcal{M}^{\frac{1}{2}}$:

$$\mathcal{N}^{\frac{1}{2}}\left(\mathcal{N}^{\frac{1}{2}}\right)^{\dagger} = \mathcal{N}\left(\mathcal{K}^T\mathcal{M}^{-1}\mathcal{K}\right)\mathcal{N} = \mathcal{N}\mathcal{N}^{-1}\mathcal{N} = \mathcal{N}.$$

Random Traction Euler-Maruyuama

- One can use the RFD idea to make more efficient temporal integrators for Brownian rigid multiblobs [3], such as the following **Euler scheme**:

1. Solve a mobility problem with a **random force+torque**:

$$\begin{bmatrix} \mathcal{M} & -\mathcal{K} \\ -\mathcal{K}^T & \mathbf{0} \end{bmatrix}^n \begin{bmatrix} \boldsymbol{\lambda}^{RFD} \\ \mathbf{U}^{RFD} \end{bmatrix} = \begin{bmatrix} \mathbf{0} \\ -\widetilde{\mathbf{W}} \end{bmatrix}. \tag{13}$$

2. Compute **random finite differences**:

$$\mathbf{F}^{RFD} = \frac{k_B T}{\delta} \left(\mathcal{K}^T \left(\mathbf{Q}^n + \delta \widetilde{\mathbf{W}} \right) - \left(\mathcal{K}^n \right)^T \right) \boldsymbol{\lambda}^{RFD}$$

$$\breve{\mathbf{u}}^{RFD} = \frac{k_B T}{\delta} \left(\mathcal{M} \left(\mathbf{Q}^n + \delta \widetilde{\mathbf{W}} \right) - \mathcal{M}^n \right) \boldsymbol{\lambda}^{RFD} + \\ - \frac{k_B T}{\delta} \left(\mathcal{K} \left(\mathbf{Q}^n + \delta \widetilde{\mathbf{W}} \right) - \mathcal{K}^n \right) \mathbf{U}^{RFD}.$$

Random Traction EM contd.

1. Compute **correlated random slip**:

$$\breve{\mathbf{u}}^n = \left(\frac{2k_B T}{\Delta t}\right)^{1/2} (\mathcal{M}^n)^{\frac{1}{2}} \mathbf{W}^n$$

2. Solve the saddle-point system:

$$\begin{bmatrix} \mathcal{M} & -\mathcal{K} \\ -\mathcal{K}^T & \mathbf{0} \end{bmatrix}^n \begin{bmatrix} \boldsymbol{\lambda}^n \\ \mathbf{U}^n \end{bmatrix} = -\begin{bmatrix} \breve{\mathbf{u}}^n + \breve{\mathbf{u}}^{RFD} \\ \mathbf{F}^n - \mathbf{F}^{RFD} \end{bmatrix}. \tag{14}$$

3. Move the particles (rotate for orientation)

$$\mathbf{Q}^{n+1} = \mathbf{Q}^n + \Delta t\, \mathbf{U}^n.$$

Conclusions

- It is possible to construct **efficient algorithms** for Brownian HydroDynamics of **nonspherical colloids in the presence of boundaries**.
- *Collective dynamics of active colloidal suspensions above a wall is strongly affected by the bottom wall!*
- Specialized temporal integrators employing **random finite differences** are required to obtain the correct stochastic drift terms.
- **Fast methods** for convolving with the (regularized) Green's function for Stokes flow in partially-confined geometries with mixed periodicity are still under active development in my group.
- Higher accuracy can be reached by using our recently-developed **fluctuating boundary integral method (FBIM)** [5], which uses the same ideas I described here for rigid multiblobs but replaces the RPY tensor with a **high-order singular quadrature**.

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