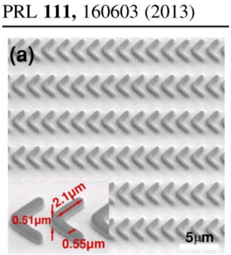
Brownian Dynamics of Confined Colloidal Suspensions

Aleksandar Donev, CIMS
& Blaise Delmotte (CIMS), Florencio Balboa (Flatiron), Brennan Sprinkle (Northwestern)

Courant Institute, *New York University*

Widely Applied Math Seminar, Harvard University
October 12th 2017

Confined Boomerang Colloids



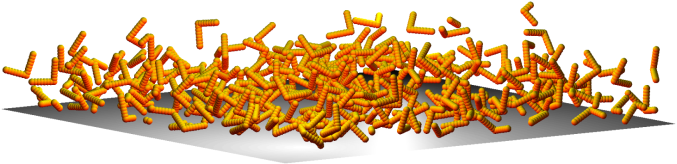
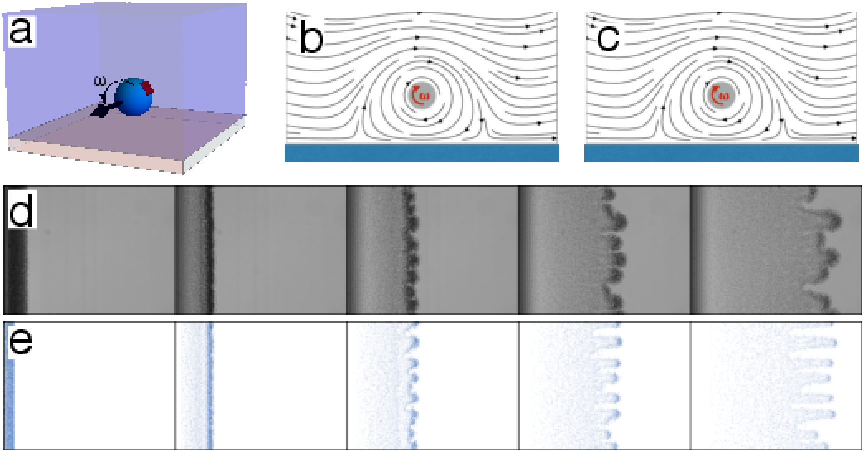


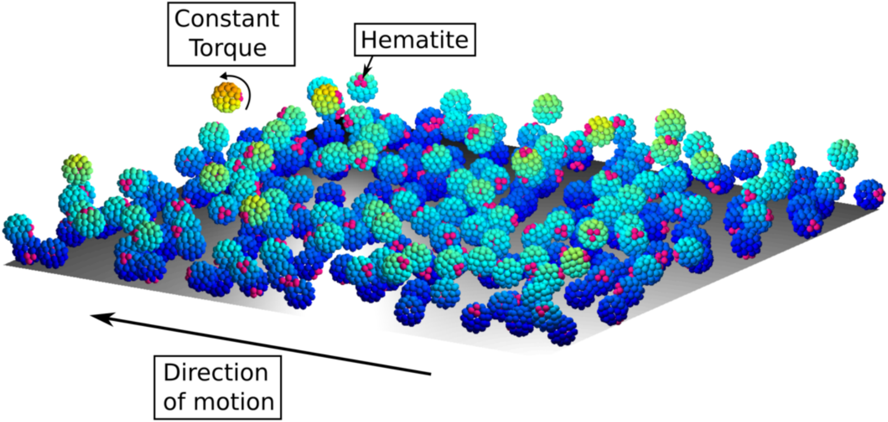
Figure: (Left) Lithographed boomerangs in a slit channel from Chakrabarty et al. (Right) Brownian dynamics of boomerangs (**Brennan Sprinkle**+Florencio Balboa) [1].

Microrollers: Fingering Instability



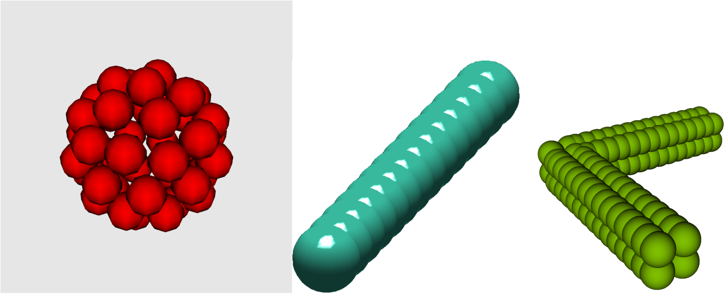
Experiments by Michelle Driscoll (lab of Paul Chaikin, NYU Physics, now at Northwestern). Simulations by **Blaise Delmotte [2]** show that **Brownian motion affects the fingering instability**(AVI).

Microrollers: Uniform Suspension



Simulations by **Brennan Sprinkle**+Blaise Delmotte [1] of a uniform suspension of microrollers at packing fraction $\phi = 0.4$ (MP4). 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** [3].
- Equivalent to a **(smartly!) regularized first-kind boundary integral formulation** [3].
- **How to efficiently simulate the active and Brownian motion of the rigid particles?**

Fluctuating Hydrodynamics

We consider a rigid body Ω immersed in a fluctuating fluid. In the fluid domain, we have the **fluctuating Stokes equation**

$$\rho\partial_t \mathbf{v} + \boldsymbol{\nabla}\pi = \eta\boldsymbol{\nabla}^2\mathbf{v} + (2k_B T\eta)^{\frac{1}{2}}\,\boldsymbol{\nabla}\cdot\boldsymbol{\mathcal{Z}}$$
$$\boldsymbol{\nabla}\cdot\mathbf{v} = 0,$$

with **no-slip BCs** on the bottom wall, and the fluid stress tensor

$$\boldsymbol{\sigma} = -\pi\mathbf{I} + \eta\left(\boldsymbol{\nabla}\mathbf{v} + \boldsymbol{\nabla}^T\mathbf{v}\right) + (2k_B T\eta)^{\frac{1}{2}}\,\boldsymbol{\mathcal{Z}} \tag{1}$$

consists of the usual **viscous stress** as well as a **stochastic stress** modeled by a symmetric **white-noise** tensor $\boldsymbol{\mathcal{Z}}(\mathbf{r},t)$, i.e., a Gaussian random field with mean zero and covariance

$$\langle \mathcal{Z}_{ij}(\mathbf{r},t)\mathcal{Z}_{kl}(\mathbf{r}',t')\rangle = (\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})\,\delta(t-t')\delta(\mathbf{r}-\mathbf{r}').$$

Fluid-Body Coupling

At the fluid-body interface the **no-slip boundary condition** is assumed to apply,

$$\mathbf{v}(\mathbf{q}) = \mathbf{u} + \boldsymbol{\omega} \times \mathbf{q} - \breve{\mathbf{u}}(\mathbf{q}) \text{ for all } \mathbf{q} \in \partial\Omega, \quad (2)$$

with the **inertial body dynamics**

$$m\frac{d\mathbf{u}}{dt} = \mathbf{F} - \int_{\partial\Omega} \boldsymbol{\lambda}(\mathbf{q})\, d\mathbf{q}, \quad (3)$$

$$\mathbf{I}\frac{d\boldsymbol{\omega}}{dt} = \boldsymbol{\tau} - \int_{\partial\Omega} [\mathbf{q} \times \boldsymbol{\lambda}(\mathbf{q})]\, d\mathbf{q} \quad (4)$$

where $\boldsymbol{\lambda}(\mathbf{q})$ is the normal component of the stress on the outside of the surface of the body, i.e., the **traction**

$$\boldsymbol{\lambda}(\mathbf{q}) = \boldsymbol{\sigma} \cdot \mathbf{n}(\mathbf{q}).$$

To model activity we can add **active slip** $\breve{\mathbf{u}}$ due to active boundary layers, or consider external forces/torques.

Mobility Problem

From linearity, the rigid-body motion is defined by a linear mapping $\mathbf{U} = \mathcal{N}\mathbf{F}$ via the deterministic **mobility problem**:

$$\boldsymbol{\nabla}\pi = \eta\boldsymbol{\nabla}^2\mathbf{v} \quad \text{and} \quad \boldsymbol{\nabla}\cdot\mathbf{v}=0 \quad +\text{BCs}$$

$$\mathbf{v}(\mathbf{q}) = \mathbf{u} + \boldsymbol{\omega}\times\mathbf{q} - \breve{\mathbf{u}}(\mathbf{q}) \;\; \text{for all } \mathbf{q}\in\partial\Omega, \tag{5}$$

With **force and torque balance**

$$\int_{\partial\Omega}\boldsymbol{\lambda}(\mathbf{q})\,d\mathbf{q} = \mathbf{F} \quad \text{and} \quad \int_{\partial\Omega}[\mathbf{q}\times\boldsymbol{\lambda}(\mathbf{q})]\,d\mathbf{q} = \boldsymbol{\tau}, \tag{6}$$

where $\boldsymbol{\lambda}(\mathbf{q}) = \boldsymbol{\sigma}\cdot\mathbf{n}(\mathbf{q})$ with

$$\boldsymbol{\sigma} = -\pi\mathbf{I} + \eta\left(\boldsymbol{\nabla}\mathbf{v} + \boldsymbol{\nabla}^T\mathbf{v}\right). \tag{7}$$

Overdamped Brownian Dynamics

- Consider a suspension of N_b rigid bodies with **configuration** $\mathbf{Q} = \{\mathbf{q}, \theta\}$ consisting of **positions and orientations** (described using **quaternions**) immersed in a Stokes fluid.
- By eliminating the fluid from the equations in the **overdamped limit** (infinite Schmidt number) we get the equations of **Brownian Dynamics**

$$\frac{d\mathbf{Q}(t)}{dt} = \mathbf{U} = \mathcal{N}\mathbf{F} + (2k_B T \mathcal{N})^{\frac{1}{2}} \mathcal{W}(t) + (k_B T)\, \partial_{\mathbf{Q}} \cdot \mathcal{N},$$

where $\mathcal{N}(\mathbf{Q})$ is the **body mobility matrix**, with "square root" given by **fluctuation-dissipation balance**

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

$\mathbf{U} = \{\mathbf{u}, \omega\}$ collects the **linear and angular velocities**
$\mathbf{F}(\mathbf{Q}) = \{\mathbf{f}, \tau\}$ collects the **applied forces and torques**.

Difficulties/Goals

Complex shapes We want to stay away from analytical approximations that only work for spherical particles.

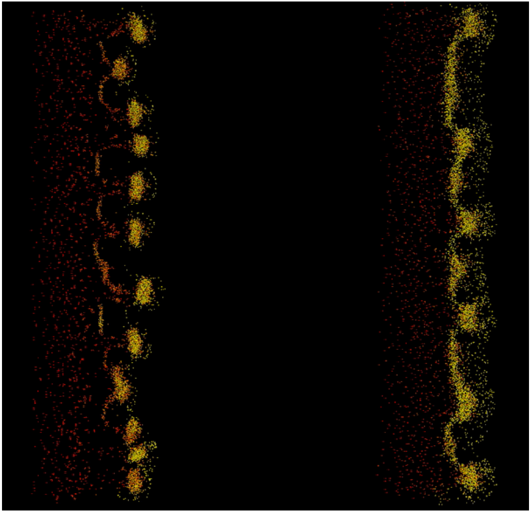
Boundary conditions Whenever observed experimentally there are microscope slips (glass plates) that modify the hydrodynamics strongly. Because of **gravity** the particles sediment **close to the bottom wall** ($\sim$ 100nm).

Many-body hydrodynamics Want to be able to scale the algorithms to suspensions of **many particles**.

Brownian increments How to generate $\mathcal{N}^{\frac{1}{2}}\mathbf{W}$, i.e., Gaussian random variables with covariance $\mathcal{N}$.

Stochastic drift How to include the $(k_B T)\, \partial_{\mathbf{Q}} \cdot \mathcal{N}$ term in **temporal integrators**.

Magnetic Rollers with Brownian Diffusion



Left: Without + Right: With **Brownian motion [2]**

Minimally-Resolved Simulations

- Represent each spherical particle by a **single blob**, and solve the Ito equations of **Brownian Dynamics with Hydrodynamic Interactions** (BD-HI) for the (correlated) positions of the N spherical microrollers $\mathbf{Q}(t) = \{\mathbf{q}_1(t), \ldots, \mathbf{q}_N(t)\}$ subjected to external magnetic torques $\mathbf{T}$,

$$d\mathbf{Q} = \mathcal{M}\mathbf{F}dt + \mathcal{M}_c\mathbf{T} + (2k_B T\,\mathcal{M})^{\frac{1}{2}}\,d\mathcal{B} + k_B T\,(\partial_{\mathbf{Q}} \cdot \mathcal{M})\,dt, \quad (8)$$

  where $\mathcal{B}(t)$ is a vector of Brownian motions, and $\mathbf{F}(\mathbf{Q})$ are applied forces.

- How to compute **deterministic velocities** $\mathcal{M}\mathbf{F}$ efficiently?
- How to generate **Brownian increments** $(2k_B T\,\mathcal{M})^{\frac{1}{2}}\,\Delta\mathcal{B}$ efficiently?
- How to generate **stochastic drift** $k_B T\,(\partial_{\mathbf{Q}} \cdot \mathcal{M})$ efficiently by only solving mobility problems?

Blobs in Stokes Flow

- The **blob-blob mobility matrix** $\mathcal{M}$ describes the hydrodynamic relations between the blobs, accounting for the influence of the boundaries:

$$\mathbf{v}\left(\mathbf{r}\right)\approx\mathbf{w}=\mathcal{M}\boldsymbol{\lambda}. \tag{9}$$

- The 3×3 block $\mathbf{M}_{ij}$ maps a force on blob j to a velocity of blob i.
- For well-separated spheres of radius a we have the **Faxen expressions**

$$\mathcal{M}_{ij}\approx\eta^{-1}\left(\mathbf{I}+\frac{a^2}{6}\boldsymbol{\nabla}^2_{\mathbf{r}'}\right)\left(\mathbf{I}+\frac{a^2}{6}\boldsymbol{\nabla}^2_{\mathbf{r}''}\right)\mathbb{G}(\mathbf{r}',\mathbf{r}'')\Big|^{\mathbf{r}'=\mathbf{r}_j}_{\mathbf{r}''=\mathbf{r}_i} \tag{10}$$

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

Rotne-Prager-Yamakawa tensor

- For homogeneous and isotropic systems (no boundaries!),

$$\boldsymbol{\mathcal{M}}_{ij} = f\left(r_{ij}\right)\boldsymbol{\mathcal{I}} + g\left(r_{ij}\right)\hat{\mathbf{r}}_{ij}\otimes\hat{\mathbf{r}}_{ij}, \tag{11}$$

- For a three dimensional unbounded domain, the Green's function is the **Oseen tensor**,

$$\mathbb{G}(\mathbf{r},\mathbf{r}') \equiv \mathbb{O}(\mathbf{r}-\mathbf{r}') = \frac{1}{8\pi r}\left(\mathbf{I} + \frac{\mathbf{r}\otimes\mathbf{r}}{r^2}\right). \tag{12}$$

- This gives the well-known **Rotne-Prager-Yamakawa (RPY) tensor** for the mobility of pairs of blobs,

$$f(r) = \frac{1}{6\pi\eta a}\begin{cases}\frac{3a}{4r}+\frac{a^3}{2r^3}, & r_{ij} > 2a\\ 1-\frac{9r}{32a}, & r_{ij} \leq 2a\end{cases}$$

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].
- General requirements for a proper RPY tensor:
  - Asymptotically **converge to the Faxen expression** for large distances from particles and walls.
  - Be **non-singular and continuous** for all configurations including overlaps of blobs and blobs with walls.
  - Mobility must **vanish** identically when a blob is exactly **on the boundary** (no motion next to wall).
  - Mobility must be symmetric positive semidefinite (**SPD**) for all configurations.

How to Approximate the Mobility

- In order to make this method work we need a way to compute the (action of the) blob-blob mobility $\boldsymbol{\mathcal{M}}$.
- It all depends on **boundary conditions**:
  - In unbounded domains we can just use the **RPY tensor** (always SPD!) with a Fast Multipole Method (FMM).
  - For single wall we use the **Rotne-Prager-Blake** tensor of Swan/Brady with **GPU-accelerated** $O(N_b^2)$ matrix-vector product.
  - For periodic domains we can use the **spectral Ewald method** [4] with FFTs.
  - In more general cases we can use a **FD/FE/FV fluid Stokes solver** [3] To compute the (action of the) **Green's functions on the fly** [5] In the spectral Ewald [4] or Stokes solver [5] approach adding thermal fluctuations (Brownian motion) can be done using **fluctuating hydrodynamics**.

Generating Brownian increments

- We need a fast way to compute the **Brownian velocities**

$$\mathbf{U}_b = \sqrt{\frac{2k_B T}{\Delta t}}\, \mathcal{M}^{\frac{1}{2}} \mathbf{W}$$

  where **W** is a vector of Gaussian random variables.
- The product $\mathcal{M}^{\frac{1}{2}} \mathbf{W}$ can be computed iteratively by **repeated multiplication** of a vector by $\mathcal{M}$ using (preconditioned) Krylov subspace **Lanczos methods**.
- When particles are sedimented close to a bottom wall, pairwise hydrodynamic interactions decay rapidly like $1/r^3$, which appears to be enough to make the Krylov method converge in a **small constant number of iterations**, without any preconditioning.

Conditioning

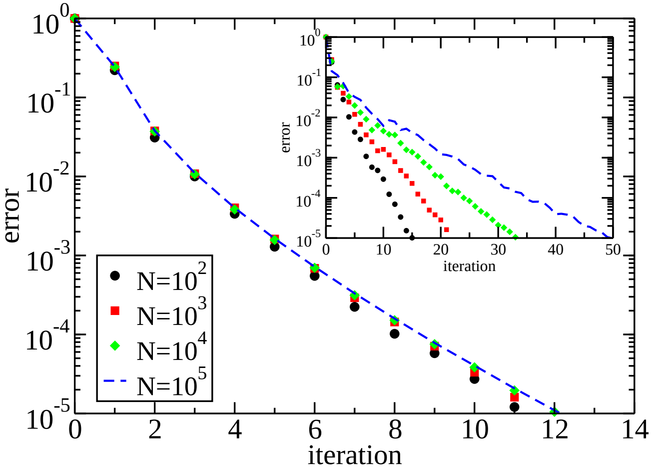


Figure: Convergence of Lancsoz iteration for $\mathcal{M}^{\frac{1}{2}}\mathbf{W}$ (inset without wall).

Periodic suspensions

- Because of the long-ranged $1/r$ nature of the Oseen kernel in free space, the number of iterations is found to grow with the number of particles, leading to an overall complexity of at least $O\left(N^{4/3}\right)$.
- More precisely, we want to sample Gaussian random variables with mean zero and covariance $\boldsymbol{\mathcal{M}}$:

$$\langle \mathbf{U}_b \mathbf{U}_b^T \rangle = \boldsymbol{\mathcal{M}}$$

- This is **easier** than computing some specific square roots, since there is a **lot of freedom**! For example, if $\boldsymbol{\mathcal{M}} = \boldsymbol{\mathcal{M}}_1 + \boldsymbol{\mathcal{M}}_2$, where $\boldsymbol{\mathcal{M}}_{1/2}$ are **both SPD**, then in law

$$\boldsymbol{\mathcal{M}}^{\frac{1}{2}}\mathbf{W} \equiv \boldsymbol{\mathcal{M}}_1^{\frac{1}{2}}\mathbf{W}_1 + \boldsymbol{\mathcal{M}}_2^{\frac{1}{2}}\mathbf{W}_2.$$

- With the group of James Swan (MIT ChemE), we have combined this with fluctuating hydrodynamics in our **Positively Split Ewald** (PSE) method [4]: $\boldsymbol{\mathcal{M}}^{\frac{1}{2}}\mathbf{W}$ with only a few **FFTs** in **linear time** for periodic suspensions (also works with **multigrid** [5]).

Stochastic drift term

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

- 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 $\langle\Delta\mathbf{P}\Delta\mathbf{Q}^T = \mathbf{I}\rangle$,

$$\lim_{\delta\to 0}\frac{1}{\delta}\left\langle\left\{\mathcal{M}\left(\mathbf{Q}+\frac{\delta}{2}\Delta\mathbf{Q}\right)-\mathcal{M}\left(\mathbf{Q}-\frac{\delta}{2}\Delta\mathbf{Q}\right)\right\}\Delta\mathbf{P}\right\rangle = \tag{13}$$

$$\left\{\partial_{\mathbf{Q}}\mathcal{M}(\mathbf{Q})\right\} : \left\langle\Delta\mathbf{P}\Delta\mathbf{Q}^T\right\rangle = k_B T\,\partial_{\mathbf{Q}}\cdot\mathcal{M}(\mathbf{Q}). \tag{14}$$

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

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

Nonspherical Rigid Multiblobs

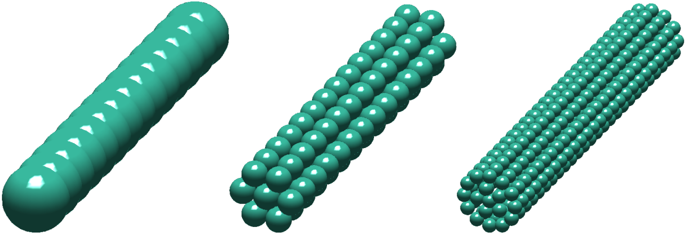


Figure: Rigid multiblob models of a rigid cylinder (rod) going from **minimally resolved** (left) to **well-resolved** (right).

Rigidly-Constrained Blobs

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

$$\sum_j \mathcal{M}_{ij}\boldsymbol{\lambda}_j = \mathbf{u}_p + \boldsymbol{\omega}_p \times \left(\mathbf{r}_i - \mathbf{q}_p\right) + \breve{\mathbf{u}}_i \tag{15}$$

$$\sum_{i \in \mathcal{B}_p} \boldsymbol{\lambda}_i = \mathbf{f}_p$$

$$\sum_{i \in \mathcal{B}_p} (\mathbf{r}_i - \mathbf{q}_p) \times \boldsymbol{\lambda}_i = \boldsymbol{\tau}_p.$$

  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 $\mathbf{U}$,

$$\begin{bmatrix} \mathcal{M} & -\mathcal{K} \\ -\mathcal{K}^T & \mathbf{0} \end{bmatrix} \begin{bmatrix} \boldsymbol{\lambda} \\ \mathbf{U} \end{bmatrix} = \begin{bmatrix} \breve{\mathbf{u}} \\ -\mathbf{F} \end{bmatrix}. \tag{16}$$

- Solve formally using Schur complements

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

- The **many-body mobility matrix** $\mathcal{N}$ takes into account **rigidity** and higher-order **hydrodynamic interactions**,

$$\mathcal{N} = \left(\mathcal{K}^T\mathcal{M}^{-1}\mathcal{K}\right)^{-1} \tag{17}$$

- For much improved accuracy one can use a **first-kind boundary integral formulation** to define a somewhat modified $\mathcal{M}$, but almost everything I say here can be generalized [6].

Active Nanorod Clusters

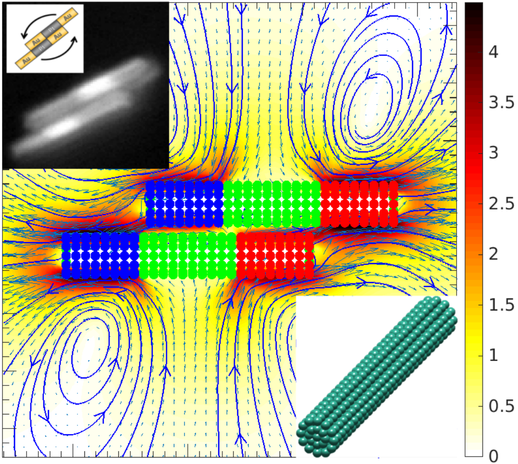


Figure: Active flow around a pair of three-segment nanorods (Au-Pt-Au) sedimented on top of a no-slip boundary and viewed from above, rotating at $\approx$ 0.7Hz in the **counter-clockwise** direction, consistent with experiments by Megan Davies Wykes (Mike Shelley lab).

Preconditioned Iterative Solver

- So far everything I wrote is well-known and used by others as well. But **dense linear algebra does not scale!**
- To get a fast and scalable method we need an **iterative method**:
  1. A fast method for performing the **matrix-vector product**, i.e., computing $\boldsymbol{\mathcal{M}}\boldsymbol{\lambda}$.
  2. A suitable **preconditioner**, which is an approximate solver for (16), to bound the number of GMRES iterations.
- How to do the fast $\boldsymbol{\mathcal{M}}\boldsymbol{\lambda}$ depends on the geometry (boundary conditions) and number of blobs N_b:
  - **fast-multipole method** (FMM), **spectral Ewald** (FFT), both $O\left(N_B \log N_b\right)$, or
  - a **direct summation on the GPU** of $O\left(N_b^2\right)$ but with very small prefactor!

Block-Diagonal Preconditioner

- We have had great success with the indefinite **block-diagonal preconditioner** [3]

$$\mathcal{P} = \begin{bmatrix} \widetilde{\mathcal{M}} & -\mathcal{K} \\ -\mathcal{K}^T & \mathbf{0} \end{bmatrix} \quad (18)$$

where we neglect all hydrodynamic interactions between blobs on distinct bodies in the preconditioner,

$$\widetilde{\mathcal{M}}^{(pq)} = \delta_{pq} \mathcal{M}^{(pp)}. \quad (19)$$

- Note that the complete hydrodynamic interactions are taken into account by the Krylov iterative solver.
- For the **mobility problem**, we find a **small constant number of GMRES iterations** independent of the number of particles (rigid multiblobs), growing only weakly with density.
- But the **resistance problem is harder** (but fortunately less important to us!), we get $O\left(N_b^{4/3}\right)$ in 3D.

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, Lancsoz 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 $\breve{\mathbf{u}}$,

$$\begin{bmatrix} \mathcal{M} & -\mathcal{K} \\ -\mathcal{K}^T & \mathbf{0} \end{bmatrix} \begin{bmatrix} \lambda \\ \mathbf{U} \end{bmatrix} = - \begin{bmatrix} \breve{\mathbf{u}} = (2k_B T)^{1/2} \mathcal{M}^{\frac{1}{2}}\mathbf{W} \\ \mathbf{F} \end{bmatrix}, \tag{20}$$

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

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 [1], such as the following **Euler scheme**:

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

$$\begin{bmatrix} \mathcal{M} & -\mathcal{K} \\ -\mathcal{K}^T & \mathbf{0} \end{bmatrix}^n \begin{bmatrix} \lambda^{RFD} \\ \mathbf{U}^{RFD} \end{bmatrix} = \begin{bmatrix} \mathbf{0} \\ -\widetilde{\mathbf{W}} \end{bmatrix}. \tag{21}$$

2. Compute **random finite differences**:

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

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

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

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:

$$\begin{bmatrix} \mathcal{M} & -\mathcal{K} \\ -\mathcal{K}^T & \mathbf{0} \end{bmatrix}^n \begin{bmatrix} \boldsymbol{\lambda}^n \\ \mathbf{U}^n \end{bmatrix} = - \begin{bmatrix} \breve{\mathbf{u}}^n + \breve{\mathbf{u}}^{RFD} \\ \mathbf{F}^n - \mathbf{F}^{RFD} \end{bmatrix}. \qquad (22)$$

3. Move the particles (rotate for orientation)

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

.

Random Slip Trapezoidal Scheme

- One can make more efficient temporal integrators (work by Brennan Sprinkle and Florencio Balboa) that are more accurate and require less GMRES solves per time step, for example, the following **trapezoidal scheme**:

1. Solve a mobility problem with an **uncorrelated random slip**:

$$\begin{bmatrix} \mathcal{M} & -\mathcal{K} \\ -\mathcal{K}^T & \mathbf{0} \end{bmatrix}^n \begin{bmatrix} \lambda^{RFD} \\ \mathbf{U}^{RFD} \end{bmatrix} = \begin{bmatrix} -\widetilde{\mathbf{W}} \in \text{Range}\left(\mathcal{M}^n\right) \\ 0 \end{bmatrix}. \tag{23}$$

2. Compute **random finite differences**:

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

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

Random Slip Trapezoidal Scheme 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. Take a **predictor FBEM** step:

$$\left[\begin{array}{cc} \mathcal{M} & -\mathcal{K} \\ -\mathcal{K}^T & \mathbf{0} \end{array}\right]^n \left[\begin{array}{c} \lambda^p \\ \mathbf{U}^p \end{array}\right] = -\left[\begin{array}{c} \breve{\mathbf{u}}^n \\ \mathbf{F}^n \end{array}\right]. \quad (24)$$

3. Compute predicted $\mathbf{Q}^p = \mathbf{Q}^n + \Delta t\, \mathbf{U}^n$.
4. Take a **trapezoidal corrector FBEM** step:

$$\left[\begin{array}{cc} \mathcal{M} & -\mathcal{K} \\ -\mathcal{K}^T & \mathbf{0} \end{array}\right]^p \left[\begin{array}{c} \lambda^c \\ \mathbf{U}^c \end{array}\right] = -\left[\begin{array}{c} \breve{\mathbf{u}}^n + 2\breve{\mathbf{u}}^{RFD} \\ \mathbf{F}^p - 2\mathbf{F}^{RFD} \end{array}\right]. \quad (25)$$

5. Complete the update, $\mathbf{Q}^{n+1} = \mathbf{Q}^n + \frac{\Delta t}{2}\left(\mathbf{U}^p + \mathbf{U}^c\right)$.

Conclusions

- We have constructed **linear-scaling** algorithms for Brownian dynamics of **nonspherical colloids in the presence of boundaries**.
- Key to generating **Brownian increments** efficiently in any **finite domain** is to use **fluctuating hydrodynamics** (Stokes solver using FFTs or multigrid) to handle the far-field hydrodynamic interactions.
- **Can a similar idea be used with grid-free fast multipole methods in unbounded domains?**
- Specialized temporal integrators employing **random finite differences** are required to obtain the correct stochastic drift terms.
- Higher accuracy can be reached by using our recently-developed **fluctuating boundary integral method (FBIM)** [6], which uses the same ideas I described here for rigid multiblobs but replaces the RPY tensor with a **high-order singular quadrature**.
- FBIM is so far only developed in two dimensions as a proof-of-concept: we need **singular quadratures in 3D that are SPD**.

References

B. Sprinkle, F. Balboa Usabiaga, N. A. Patankar, and A. Donev.
Large Scale Brownian Dynamics of Confined Suspensions of Rigid Particles.
Submitted to J. Chem. Phys., ArXiv:1709.02410, 2017.

Florencio Balboa Usabiaga, Blaise Delmotte, and Aleksandar Donev.
Brownian dynamics of confined suspensions of active microrollers.
J. Chem. Phys., 146(13):134104, 2017.
Software available at https://github.com/stochasticHydroTools/RigidMultiblobsWall.

F. Balboa Usabiaga, B. Kallemov, B. Delmotte, A. P. S. Bhalla, B. E. Griffith, and A. Donev.
Hydrodynamics of suspensions of passive and active rigid particles: a rigid multiblob approach.
Communications in Applied Mathematics and Computational Science, 11(2):217–296, 2016.
Software available at https://github.com/stochasticHydroTools/RigidMultiblobsWall.

A. M. Fiore, F. Balboa Usabiaga, A. Donev, and J. W. Swan.
Rapid sampling of stochastic displacements in brownian dynamics simulations.
J. Chem. Phys., 146(12):124116, 2017.
Software available at https://github.com/stochasticHydroTools/PSE.

S. Delong, F. Balboa Usabiaga, R. Delgado-Buscalioni, B. E. Griffith, and A. Donev.
Brownian Dynamics without Green's Functions.
J. Chem. Phys., 140(13):134110, 2014.
Software available at https://github.com/stochasticHydroTools/FIB.

Y. Bao, M. Rachh, E. Keaveny, L. Greengard, and A. Donev.
A fluctuating boundary integral method for Brownian suspensions.
Submitted to J. Comp. Phys., preprint ArXiv:1709.01480, 2017.