Hydrodynamics of Suspensions of Passive and Active Rigid Particles

Aleksandar Donev, CIMS
Collaborators: **Blaise Delmotte** and **Florencio Balboa**, CIMS
Contributors: Steven Delong, Michelle Driscoll, Paul Chaikin

Courant Institute, *New York University*

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Non-Spherical Colloids near Boundaries

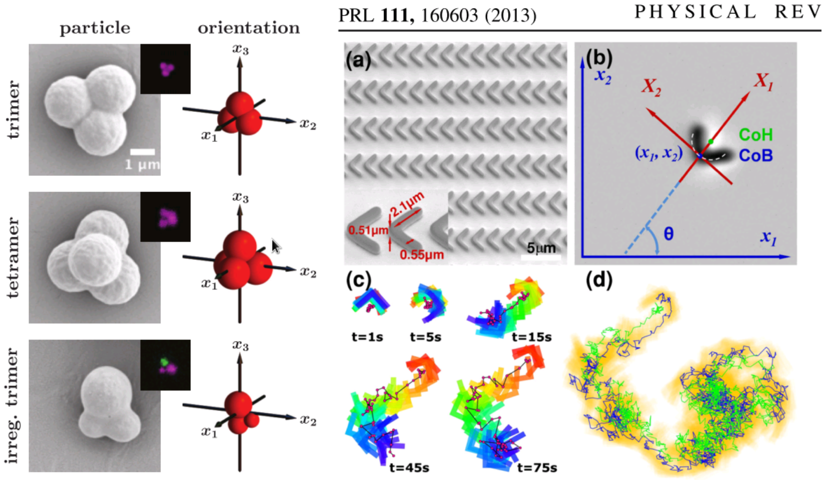


Figure: (Left) Cross-linked spheres from Kraft et al. (Right) Lithographed boomerangs in a microchannel from Chakrabarty et al.

Active Nanorod Clusters

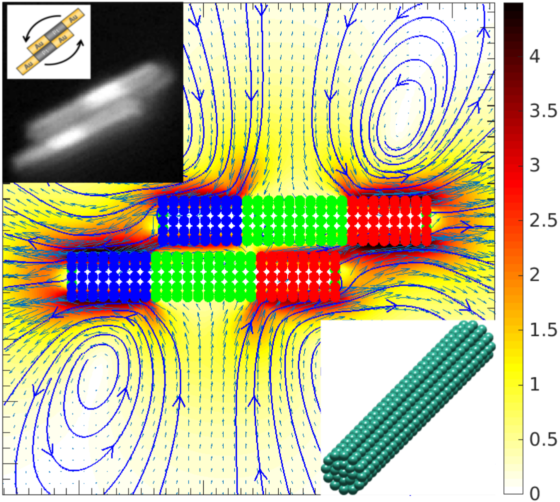
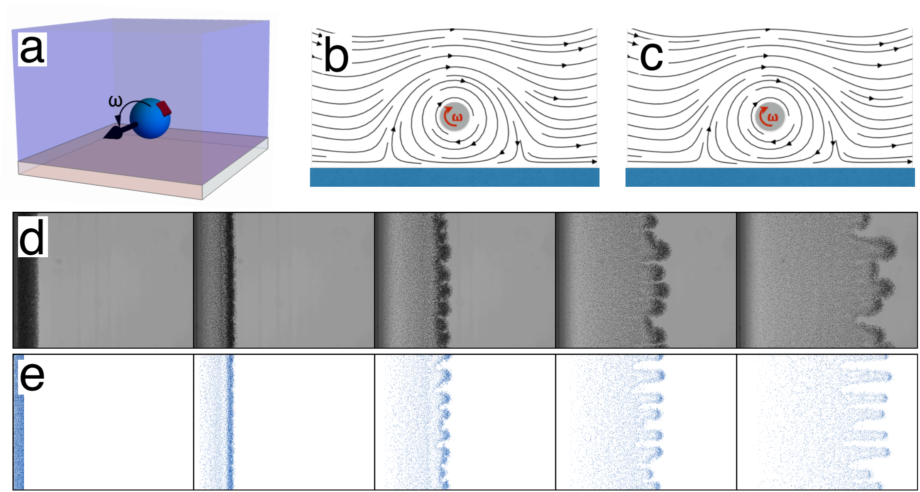


Figure: From Megan Davies Wykes [1] in the Courant Applied Math Lab.

Magnetic Spherical Rollers



Collaboration of Michelle Driscoll (lab of Paul Chaikin, NYU Physics) and Blaise Delmotte (Courant, Donev group)

RigidMultiBlob Models

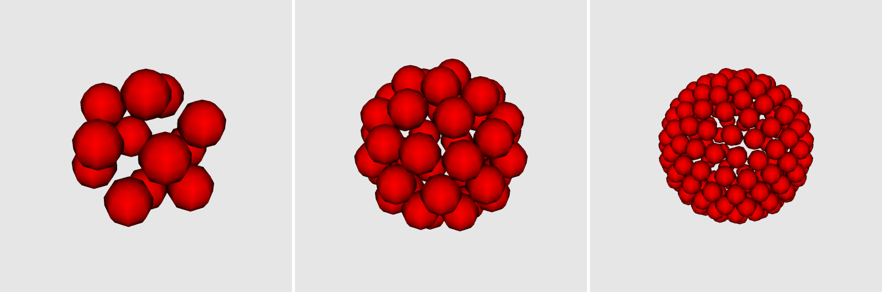


Figure: Blob or "raspberry" models of a spherical colloid.

- 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** [2].
- Can we do this efficiently for $10^4 - 10^5$ particles?
  Yes, if we use iterative linear solvers!

Fluctuating Hydrodynamics

We consider a rigid body Ω immersed in an unbounded fluctuating fluid. In the fluid domain

$$-\boldsymbol{\nabla}\cdot\boldsymbol{\sigma}=\boldsymbol{\nabla}\pi-\eta\boldsymbol{\nabla}^{2}\mathbf{v}-\left(2k_{B}T\eta\right)^{\frac{1}{2}}\boldsymbol{\nabla}\cdot\boldsymbol{\mathcal{Z}}=0$$
$$\boldsymbol{\nabla}\cdot\mathbf{v}=0,$$

where the fluid stress tensor

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

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

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

Fluid-Body Coupling

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

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

with the **force and torque balance**

$$\int_{\partial\Omega} \boldsymbol{\lambda}(\mathbf{q})\, d\mathbf{q} = \mathbf{F} \quad \text{and} \quad \int_{\partial\Omega} [\mathbf{q} \times \boldsymbol{\lambda}(\mathbf{q})]\, d\mathbf{q} = \boldsymbol{\tau}, \tag{3}$$

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 add **active slip $\breve{\mathbf{u}}$** due to active boundary layers.

Steady Stokes Flow ($\mathrm{Re} \to 0$, $\mathrm{Sc} \to \infty$)

- Consider a suspension of N_b rigid bodies with **configuration** $\mathbf{Q} = \{\mathbf{q}, \theta\}$ consisting of **positions and orientations** (described using **quaternions** [3]).
- For viscous-dominated flows we can assume **steady Stokes flow** and define the **body mobility matrix** $\mathcal{N}(\mathbf{Q})$,

$$\frac{d\mathbf{Q}(t)}{dt} = \mathbf{U} = \mathcal{N}\mathbf{F} - \breve{\mathcal{M}}\breve{\mathbf{u}} + (2k_B T \mathcal{N})^{\frac{1}{2}} \diamond \mathcal{W}(t),$$

  where $\mathbf{U} = \{\mathbf{u}, \omega\}$ collects the **linear and angular velocities** $\mathbf{F}(\mathbf{Q}) = \{\mathbf{f}, \tau\}$ collects the **applied forces and torques**
- **How to compute (the action of) $\mathcal{N}$ and $\mathcal{N}^{\frac{1}{2}}$ and simulate the Brownian motion of the bodies?**

Difficulties/Goals

Stochastic drift It is crucial to handle stochastic calculus issues carefully for **overdamped Langevin** dynamics. Since diffusion is slow we also want to be able to take **large time step sizes**.

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. It is preferred to use **no Green's functions** but rather work in complex geometry.

Gravity Observe that in all of the examples above there is gravity and the particles sediment toward the bottom wall, often **very close to the wall** ($\sim$ 100nm). This is a general feature of all active suspensions but this is almost always neglected in theoretical models.

Many-body Want to be able to scale the algorithms to suspensions of **many particles**–nontrivial **numerical linear algebra**.

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:

$$\mathbf{v}(\mathbf{r}) \approx \mathbf{w} = \mathcal{M}\boldsymbol{\lambda}. \tag{4}$$

- 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**

$$\mathcal{M}_{ij} \approx \eta^{-1} \left(\mathbf{I} + \frac{a^2}{6}\boldsymbol{\nabla}^2_{\mathbf{r}'}\right) \left(\mathbf{I} + \frac{a^2}{6}\boldsymbol{\nabla}^2_{\mathbf{r}''}\right) \mathbb{G}(\mathbf{r}', \mathbf{r}'')\Big|^{\mathbf{r}'=\mathbf{r}_j}_{\mathbf{r}''=\mathbf{r}_i} \tag{5}$$

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!),

$$\boldsymbol{\mathcal{M}}_{ij} = f\left(r_{ij}\right)\boldsymbol{\mathcal{I}} + g\left(r_{ij}\right)\hat{\mathbf{r}}_{ij} \otimes \hat{\mathbf{r}}_{ij}, \tag{6}$$

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

$$\mathbb{G}(\mathbf{r}, \mathbf{r}') \equiv \mathbb{O}(\mathbf{r} - \mathbf{r}') = \frac{1}{8\pi r}\left(\mathbf{I} + \frac{\mathbf{r} \otimes \mathbf{r}}{r^2}\right). \tag{7}$$

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

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

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.
- A generic procedure for how to **generalize RPY** has been proposed, but to my knowledge there is no simple analytical formula even for a single wall.
- For non-overlapping blobs next to a wall the **Rotne-Prager-Blake** tensor has been computed [4] and we will use it here.
- 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 $\mathcal{M}$.
- It all depends on **boundary conditions**:
  - In unbounded domains we can just use the **RPY tensor** (always SPD!).
  - For single wall we can use the **Rotne-Prager-Blake** tensor [4].
  - For periodic domains we can use the **spectral Ewald method** [5].
  - In more general cases we can use a **FD/FE/FV fluid Stokes solver** [2]
    To compute the (action of the) **Green's functions on the fly** [6]
    In the grid-based approach adding thermal fluctuations (Brownian motion) can be done using **fluctuating hydrodynamics** (not discussed here).

Nonspherical Rigid Multiblobs

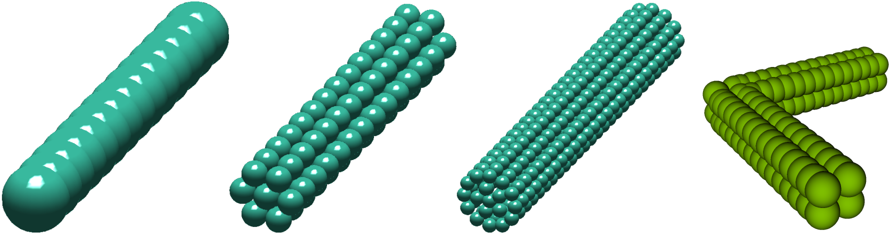


Figure: Rigid multiblob models of colloidal particles manufactured in recent experimental work.

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,

$$\begin{aligned} \sum_j \mathcal{M}_{ij} \boldsymbol{\lambda}_j =& \mathbf{u}_p + \boldsymbol{\omega}_p \times \left(\mathbf{r}_i - \mathbf{q}_p \right) + \breve{\mathbf{u}}_i \\ \sum_{i \in \mathcal{B}_p} \boldsymbol{\lambda}_i =& \mathbf{f}_p \\ \sum_{i \in \mathcal{B}_p} (\mathbf{r}_i - \mathbf{q}_p) \times \boldsymbol{\lambda}_i =& \boldsymbol{\tau}_p. \end{aligned} \quad (8)$$

  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 $\mathbf{U}$,

$$\begin{bmatrix} \mathcal{M} & -\mathcal{K} \\ -\mathcal{K}^T & \mathbf{0} \end{bmatrix} \begin{bmatrix} \boldsymbol{\lambda} \\ \mathbf{U} \end{bmatrix} = \begin{bmatrix} \breve{\mathbf{u}} \\ -\mathbf{F} \end{bmatrix}. \tag{9}$$

- 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**,

$$\mathcal{N} = \left(\mathcal{K}^T\mathcal{M}^{-1}\mathcal{K}\right)^{-1} \tag{10}$$

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 $\mathcal{M}\boldsymbol{\lambda}$.
  2. A suitable **preconditioner**, which is an approximate solver for (9), to bound the number of GMRES iterations.
- How to do the fast $\mathcal{M}\boldsymbol{\lambda}$ 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** [2]

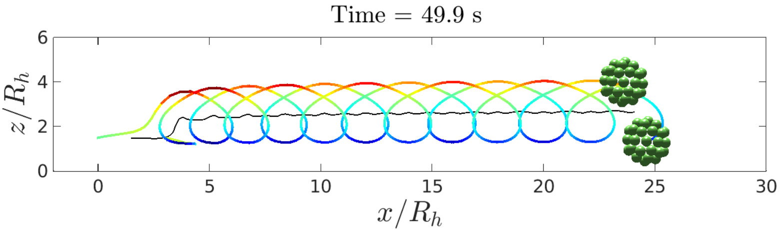
$$\mathcal{P} = \begin{bmatrix} \widetilde{\mathcal{M}} & -\mathcal{K} \\ -\mathcal{K}^T & \mathbf{0} \end{bmatrix} \tag{11}$$

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

$$\widetilde{\mathcal{M}}^{(pq)} = \delta_{pq} \mathcal{M}^{(pp)}. \tag{12}$$

- Note that the complete hydrodynamic interactions are taken into account by the Krylov iterative solver.
- For the **mobility problem**, we find a **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.

Example: Dimer of sedimented rollers



Regularized On-the-Fly Green's Function

- For **fully confined suspensions**, compute the **Green's function on the fly** using a **discrete Stokes solver**:

$$\boldsymbol{\mathcal{M}}_{ij}(\mathbf{r}_i, \mathbf{r}_j) = \eta^{-1} \int \delta_a(\mathbf{r}_i - \mathbf{r}')\mathbb{G}(\mathbf{r}', \mathbf{r}'')\delta_a(\mathbf{r}_j - \mathbf{r}'') \, d\mathbf{r}' d\mathbf{r}'' \qquad (13)$$

  which is a **generalized RPY tensor** that with suitable modifications of δ_a next to a boundary has all of the desired properties I wrote earlier!

- This is consistent with the Faxen formula for far-away blobs,

$$\int \delta_a(\mathbf{r}_i - \mathbf{r})\mathbf{v}(\mathbf{r})d\mathbf{r} \approx \left(\mathbf{I} + \frac{a_F^2}{6}\boldsymbol{\nabla}^2\right)\mathbf{v}(\mathbf{r})\,\Big|_{\mathbf{r}=\mathbf{r}_i},$$

  with a **Faxen blob radius** $a_F \equiv \left(3 \int x^2 \delta_a(x)\, dx\right)^{1/2}$.

- The effective **hydrodynamic blob radius** $a \approx a_F$ is

$$\boldsymbol{\mathcal{M}}_{ii} = \frac{1}{6\pi\eta a}\mathbf{I} = \eta^{-1} \int \delta_a(\mathbf{r}')\mathbb{O}(\mathbf{r}' - \mathbf{r}'')\delta_a(\mathbf{r}'') \, d\mathbf{r}' d\mathbf{r}''$$

Suspension of rods (cylinders) next to wall

ϕ_a	Resolution	Wall-corrected	Unbounded
0.01	21	12	17
	98	16	28
0.1	21	19	23
	98	22	32
0.2	21	20	25
	98	23	34
0.4	21	25	29
	98	27	33
0.6	21	30	33
	98	31	43

Table: Suspension of cylinders sedimented against a no-slip boundary. Number of GMRES iterations required to reduce the residual by a factor of 10^8 for several surface packing fractions and two different resolutions (number of blobs per rod), for $H/D = 0.75$ and $N_r = 1000$ rods.

Suspension of rods (cylinders) next to wall

N_r	Resolution	$H/D = 0.75$	$H/D = 2$
10	21	7	7
	98	8	9
100	21	14	13
	98	19	18
1000	21	19	16
	98	22	20
5000	21	18	16
	98	23	22
10000	21	20	17
	98	23	21

Table: Suspension of cylinders sedimented against a no-slip boundary. (Right) Number of GMRES iterations required to reduce the residual by a factor of 10^8 for $\phi_a = 0.1$ and different number of rods.

Active dimer of extensors

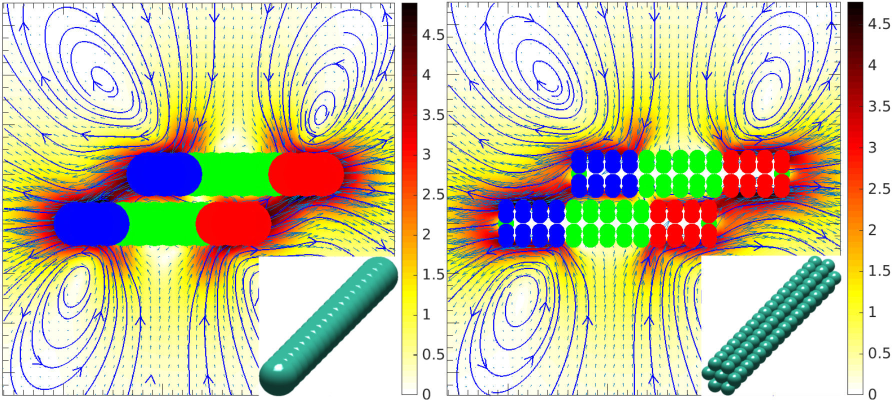


Figure: Active flow around a pair of extensile three-segment nanorods (Au-Pt-Au) sedimented on top of a no-slip boundary (the plane of the image) and viewed from above. The dimers are rotating together at ≈ 0.7Hz in the counter-clockwise direction, consistent with recent experimental observations.

Bodies with rotation

- We can extend our work to simulate bodies with **rotational DOFs** by formulating the appropriate Langevin equation and using a RFD approach to for temporal integration.
- For simplicity, first we consider a single body with only rotational degrees of freedom.
- Orientation is an element of SO(3) so we need to parameterize it: we use **normalized quaternion** (point on the unit 4-sphere)

$$\boldsymbol{\theta} \in \mathbb{R}^4, \quad \|\boldsymbol{\theta}\|_2 = \boldsymbol{\theta} \cdot \boldsymbol{\theta} = 1.$$

- This offers several advantages over several other common approaches, such as rotation angles, rotation matrices, and Euler angles.

Quaternions

- Successive rotations can be accumulated by **quaternion multiplication.**
- In three dimensions, there exists a 4×3 matrix $\boldsymbol{\Psi}(\boldsymbol{\theta})$ such that, given a conservative potential $U(\boldsymbol{\theta})$,

$$\dot{\boldsymbol{\theta}} = \boldsymbol{\Psi}\boldsymbol{\omega}, \quad \boldsymbol{\tau} = \boldsymbol{\Psi}^T \partial_{\boldsymbol{\theta}} U(\boldsymbol{\theta}).$$

  Here $\boldsymbol{\tau}$ is the torque applied to the body, and $\boldsymbol{\omega}$ is the angular velocity.
- One can also rotate a body by an oriented angle $\boldsymbol{\phi}$, denoted as

$$\boldsymbol{\theta}^{n+1} = \text{Rotate}\left(\boldsymbol{\theta}^n, \, \boldsymbol{\phi}\right).$$

Rotational Langevin Equation

- We assume now that we know the mobility tensor $\mathbf{M}_{\omega\tau}$,

$$\boldsymbol{\omega} = \mathbf{M}_{\omega\tau}\boldsymbol{\tau}.$$

- Given $\mathbf{M}_{\omega\tau}$ and a potential $U(\boldsymbol{\theta})$, the **Overdamped Langevin Equation** for orientation is

$$\partial_t\boldsymbol{\theta} = -\left(\boldsymbol{\Psi}\mathbf{M}_{\omega\tau}\boldsymbol{\Psi}^T\right)\partial_{\boldsymbol{\theta}}U + \sqrt{2k_BT}\boldsymbol{\Psi}\mathbf{M}^{\frac{1}{2}}_{\omega\tau}\boldsymbol{\mathcal{W}} + k_BT\partial_{\boldsymbol{\theta}}\cdot\left(\boldsymbol{\Psi}\mathbf{M}_{\omega\tau}\boldsymbol{\Psi}^T\right).$$

- This equation preserves the unit norm constraint and is time reversible w.r.t. the **Gibbs-Boltzmann distribution**

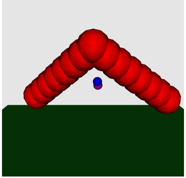
$$P_{\text{eq}}(\boldsymbol{\theta}) = Z^{-1}\exp\left(-U(\boldsymbol{\theta})/k_BT\right)\delta\left(\boldsymbol{\theta}^T\boldsymbol{\theta}-1\right).$$

Random Finite Difference

- To take a time step in a **Brownian Dynamics** algorithm with rotational diffusion we do:

$$
\begin{aligned}
\tilde{\mathbf{v}} &= \widetilde{\mathbf{W}} \\
\tilde{\mathbf{q}} &= \mathbf{q}^n + \delta \tilde{\mathbf{u}} \\
\tilde{\boldsymbol{\theta}} &= \text{Rotate}\left(\boldsymbol{\theta}^n, \delta \tilde{\boldsymbol{\omega}}\right) \\
\mathbf{v}^n &= -\left(\mathbf{N}\boldsymbol{\Xi}^T \partial_{\mathbf{x}} U\right)^n + \sqrt{\frac{2k_B T}{\Delta t}} \left(\mathbf{N}^{\frac{1}{2}}\right)^n \mathbf{W}^n + \frac{k_B T}{\delta} \left(\widetilde{\mathbf{N}} - \mathbf{N}^n\right) \widetilde{\mathbf{W}} \\
\mathbf{q}^{n+1} &= \mathbf{q}^n + \Delta t \mathbf{u}^n \\
\boldsymbol{\theta}^{n+1} &= \text{Rotate}\left(\boldsymbol{\theta}^n, \Delta t \boldsymbol{\omega}^n\right).
\end{aligned}
$$

Diffusion of a Confined Boomerang



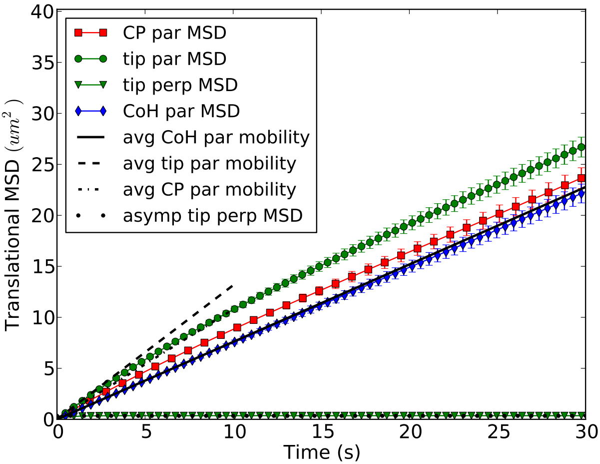
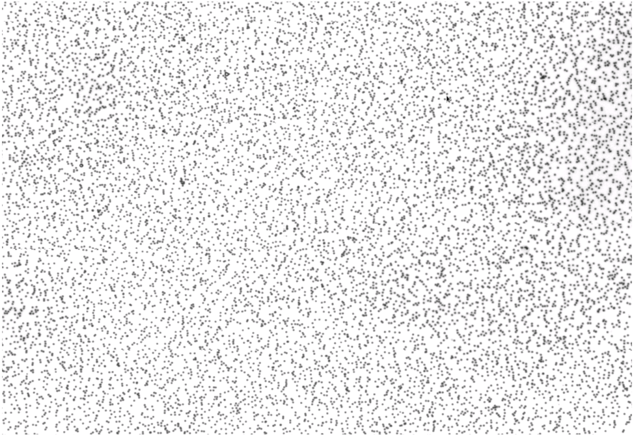


Figure: Translational MSD for a boomerang

Active Micro-rollers

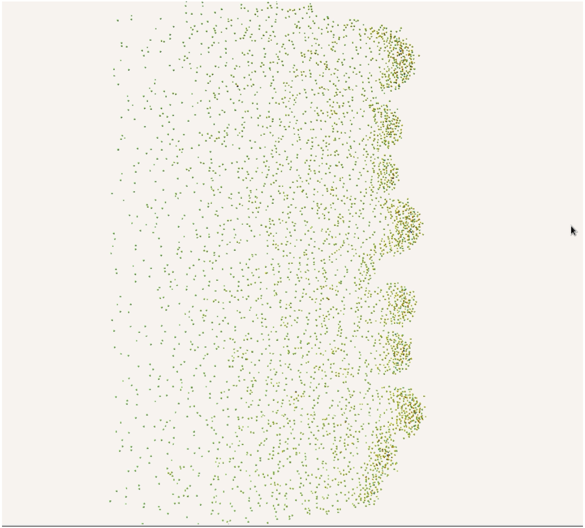


Experiments performed by **Michelle Driscoll** in lab of **Paul Chaikin**, NYU CSMR Physics

Active Roller Interfacial Instability

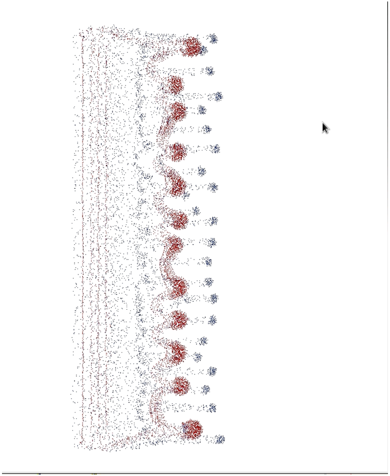
Larger Density

Active Roller Simulations



Simulations performed by **Blaise Delmotte**, Courant

Advantages of Simulation: 2D motion

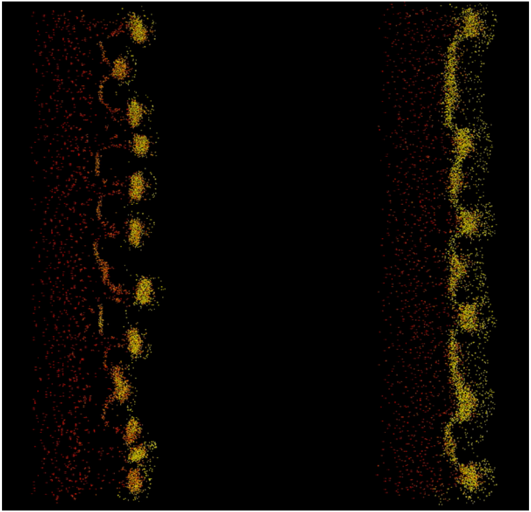


Simulations confirm **instability is purely hydrodynamic** and develops similarly even in a suspension of singular rotlets with no steric interactions.

3D Stable Critters: Wall Repulsion

3D side view

Brownian Diffusion



Left: Without + Right: With **Brownian motion**

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