Brownian Suspensions of Rigid Particles

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Non-Spherical Colloids near Boundaries

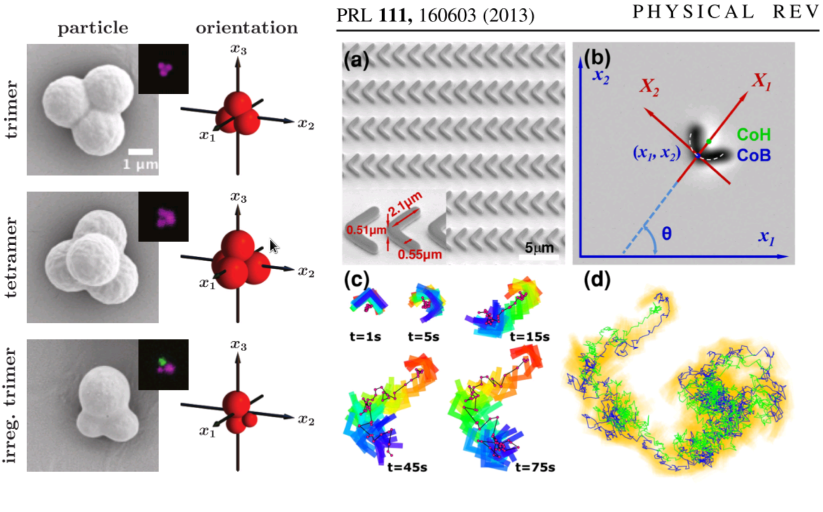


Figure: (Left) Cross-linked spheres; Kraft et al. [1]. (Right) Lithographed boomerangs; Chakrabarty et al. [2].

Bent Active Nanorods

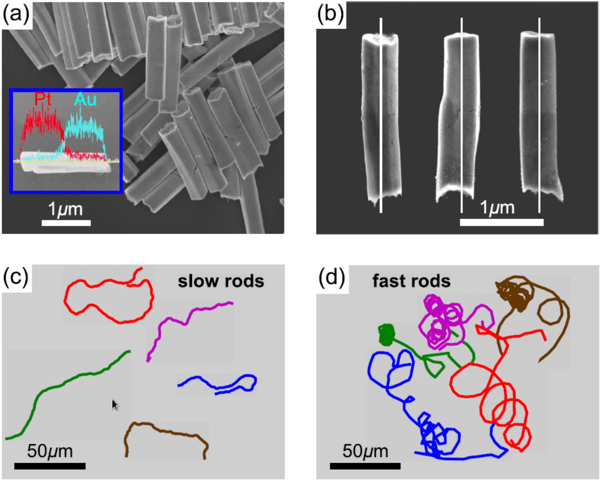
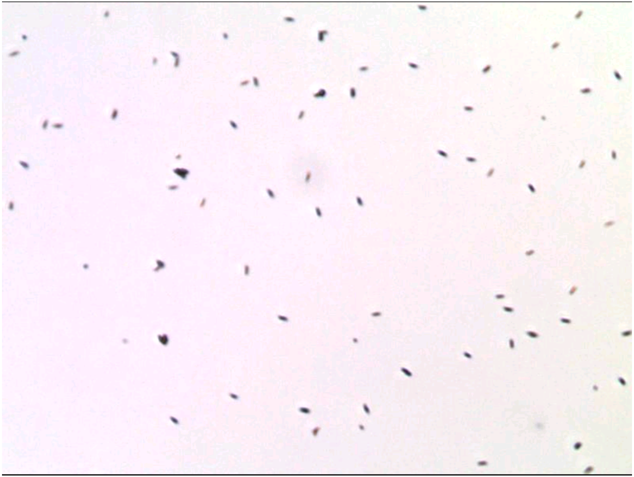


Figure: From the Courant Applied Math Lab of Zhang and Shelley [3]

Thermal Fluctuation Flips



QuickTime

Steady Stokes Flow (Re $\rightarrow$ 0)

- Consider a **suspension of** N_b **rigid bodies** with positions $\mathcal{Q} = \left\{\boldsymbol{\varrho}_1, \ldots, \boldsymbol{\varrho}_{N_b}\right\}$ and orientations $\boldsymbol{\Theta} = \{\boldsymbol{\theta}_1, \ldots, \boldsymbol{\theta}_{N_b}\}$.
  We describe orientations using **quaternions**.
- For viscous-dominated flows we can assume **steady Stokes flow** and define the **body mobility matrix** $\mathcal{N}(\mathcal{Q}, \boldsymbol{\Theta})$,

$$[\mathcal{U}, \, \boldsymbol{\Omega}]^T = \mathcal{N} \, [\mathcal{F}, \, \mathcal{T}]^T \, ,$$

  where the left-hand side collects the **linear** $\mathcal{U} = \{\boldsymbol{\upsilon}_1, \ldots, \boldsymbol{\upsilon}_{N_b}\}$ and **angular** $\boldsymbol{\Omega} = \{\boldsymbol{\omega}_1, \ldots, \boldsymbol{\omega}_{N_b}\}$ **velocities**,
  and the right hand side collects the **applied forces** $\mathcal{F}(\mathcal{Q}, \boldsymbol{\Theta}) = \{\mathbf{F}_1, \ldots, \mathbf{F}_{N_b}\}$ and **torques** $\mathcal{T}(\mathcal{Q}, \boldsymbol{\Theta}) = \{\boldsymbol{\tau}_1, \ldots, \boldsymbol{\tau}_{N_b}\}$.

Brownian Motion

- The Brownian motion of the rigid bodies is described by the **overdamped Langevin equation**, symbolically:

$$\begin{bmatrix} d\mathcal{Q}/dt \\ d\boldsymbol{\Theta}/dt \end{bmatrix} = \begin{bmatrix} \mathcal{U} \\ \boldsymbol{\Omega} \end{bmatrix} = \mathcal{N} \begin{bmatrix} \mathcal{F} \\ \mathcal{T} \end{bmatrix} + \left(2k_B T \mathcal{N}\right)^{\frac{1}{2}} \diamond \mathcal{W}(t) \, .$$

- How to represent orientations using normalized quaternions and handle the constraint $\|\boldsymbol{\Theta}_k\| = 1$?
- What is the correct thermal drift (i.e., what does $\diamond$ mean)?
- **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**.

Blob/Bead Models

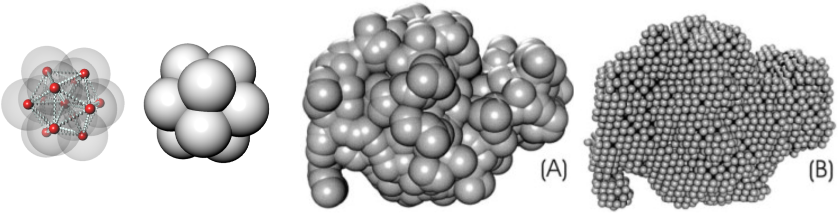


Figure: Blob or "raspberry" models of: a spherical colloid, and a lysozyme [4].

- The rigid body is discretized through a number of "**beads**" or "**blobs**" with positions $\mathbf{Q} = \{\mathbf{q}_1, \ldots, \mathbf{q}_N\}$.
- Describe the fluid-blob interaction using a localized smooth **kernel** $\delta_a(r)$ with compact support of size a giving the effective hydrodynamic radius of the blob (diffuse sphere).
- Standard in fluctuating/stochastic immersed boundary methods but with **stiff springs** instead of **truly rigid agglomerates**.

Rigidly-Constrained Blobs

$$\begin{aligned}
\boldsymbol{\nabla}\pi-\eta\boldsymbol{\nabla}^{2}\mathbf{v} &= \sum_{i=1}^{N}\boldsymbol{\lambda}_{i}\delta_{a}\left(\mathbf{q}_{i}-\mathbf{r}\right)+\sqrt{2\eta k_{B}T}\;\boldsymbol{\nabla}\cdot\boldsymbol{\mathcal{W}} \\
\boldsymbol{\nabla}\cdot\mathbf{v} &= 0\;(\text{Lagrange multiplier is }\pi) \\
\sum_{i=1}^{N}\boldsymbol{\lambda}_{i} &= \mathbf{F}\;(\text{Lagrange multiplier is }\boldsymbol{\upsilon}) \\
\sum_{i=1}^{N}\left(\mathbf{q}_{i}-\boldsymbol{\varrho}^{0}\right)\times\boldsymbol{\lambda}_{i} &= \boldsymbol{\tau}\;(\text{Lagrange multiplier is }\boldsymbol{\omega}), \\
\forall i:\quad\int\delta_{a}\left(\mathbf{q}_{i}-\mathbf{r}\right)\mathbf{v}\left(\mathbf{r},t\right)\,d\mathbf{r} &= \boldsymbol{\upsilon}+\boldsymbol{\omega}\times\left(\mathbf{q}_{i}-\boldsymbol{\varrho}^{0}\right)+\textbf{slip}\;(\text{Multiplier is }\boldsymbol{\lambda}_{i})
\end{aligned} \tag{1}$$

Notation

- Composite velocity $\mathbf{U} = \{\mathbf{u}_1, \ldots, \mathbf{u}_N\}$ and rigidity forces $\boldsymbol{\Lambda} = \{\boldsymbol{\lambda}_1, \ldots, \boldsymbol{\lambda}_N\}$.
- Define the composite local **averaging** linear operator $\mathcal{J}(\mathbf{Q})$ operator, and the composite **spreading** linear operator, $\mathcal{S}(\mathbf{Q}) = \mathcal{J}^{\star}(\mathbf{Q})$,

$$\mathbf{u}_i = (\mathcal{J}\mathbf{v})_i = \int \delta_a(\mathbf{q}_i - \mathbf{r})\,\mathbf{v}(\mathbf{r}, t)\; d\mathbf{r}$$

$$\boldsymbol{\lambda}(\mathbf{r}) = (\mathcal{S}\boldsymbol{\Lambda})(\mathbf{r}) = \sum_{i=1}^{N} \boldsymbol{\lambda}_i \delta_a(\mathbf{q}_i - \mathbf{r}).$$

- Denote the (potentially discrete) operators scalar gradient $\mathbf{G} \equiv \boldsymbol{\nabla}$, vector divergence $\mathbf{D} = -\mathbf{G}^{\star} \equiv \boldsymbol{\nabla}\cdot$, tensor divergence $\mathbf{D}_{\mathbf{v}}$, and vector Laplacian $\mathbf{L} = -\mathbf{D}_{\mathbf{v}}\mathbf{D}_{\mathbf{v}}^{\star} \equiv \boldsymbol{\nabla}^2$.

Saddle-Point Problem

- Define the geometric matrix $\mathcal{K}$ that converts body kinematics to blob kinematics,

$$\mathbf{U} = \mathcal{K}\mathcal{Y} = \mathcal{K}\left[\mathcal{U}, \boldsymbol{\Omega}\right]^T = \mathcal{U} + \boldsymbol{\Omega} \times \left(\mathbf{Q} - \mathcal{Q}^0\right).$$

- We get the symmetric **constrained Stokes saddle-point problem**,

$$\begin{bmatrix} -\eta\mathbf{L} & \mathbf{G} & -\mathcal{S} & \mathbf{0} \\ -\mathbf{D} & \mathbf{0} & \mathbf{0} & \mathbf{0} \\ -\mathcal{J} & \mathbf{0} & \mathbf{0} & \mathcal{K} \\ \mathbf{0} & \mathbf{0} & \mathcal{K}^\star & \mathbf{0} \end{bmatrix} \begin{bmatrix} \mathbf{v} \\ \pi \\ \boldsymbol{\Lambda} \\ \mathcal{Y} \end{bmatrix} = \begin{bmatrix} \boldsymbol{\nabla} \cdot \left(\sqrt{2\eta k_B T}\,\mathcal{W}\right) \\ 0 \\ 0 \\ \mathcal{R} \end{bmatrix},$$

where $\mathcal{Y} = [\mathcal{U},\, \boldsymbol{\Omega}]^T$ and $\mathcal{R} = [\mathcal{F},\, \mathcal{T}]^T$, and recall that $\mathcal{S} = \mathcal{J}^\star$.

Mobility Matrix

- Eliminate velocity and pressure using the Schur complement

$$\begin{bmatrix} \mathcal{M} & -\mathcal{K} \\ -\mathcal{K}^{\star} & \mathbf{0} \end{bmatrix} \begin{bmatrix} \boldsymbol{\Lambda} \\ \mathcal{Y} \end{bmatrix} = \begin{bmatrix} \text{slip} \\ -\left(\mathcal{R} + \widetilde{\mathcal{R}}\right) \end{bmatrix},$$

where $\widetilde{\mathcal{R}} = \sqrt{2\eta k_B T}\, \mathcal{K}^{\star} \mathcal{M}^{-1} \mathcal{J} \mathcal{L}^{-1} \mathbf{D}_v \mathcal{W}$ are the random (stochastic) forces and torques.

- Here the all-important $3N \times 3N$ blob **mobility matrix** $\mathcal{M}$ is

$$\mathcal{M} = \mathcal{J} \mathcal{L}^{-1} \mathcal{S},$$

where $\mathcal{L}^{-1} = -\mathbf{L}^{-1} + \mathbf{L}^{-1}\mathbf{G}\left(\mathbf{D}\mathbf{L}^{-1}\mathbf{G}\right)^{-1}\mathbf{D}\mathbf{L}^{-1}$ denotes the Stokes solution operator.

Rigidly-Constrained Blobs

- The physical interpretation is simple:

$$\boldsymbol{\mathcal{M}}\boldsymbol{\Lambda} = \boldsymbol{\mathcal{K}}\boldsymbol{\mathcal{Y}} + \text{slip}$$
$$\boldsymbol{\mathcal{K}}^{\star}\boldsymbol{\Lambda} = \boldsymbol{\mathcal{R}} + \widetilde{\boldsymbol{\mathcal{R}}},$$

  where the unknown $\boldsymbol{\mathcal{Y}} = [\boldsymbol{\mathcal{U}}, \boldsymbol{\Omega}]^T$ are the body kinematics, $\boldsymbol{\mathcal{R}} = [\boldsymbol{\mathcal{F}}, \boldsymbol{\mathcal{T}}]^T$ are the applied forces and torques and $\widetilde{\boldsymbol{\mathcal{R}}}$ are the random (stochastic) forces and torques.
- Here $\boldsymbol{\Lambda}$ are the *unknown* rigidity forces (Lagrange multipliers) acting on the blobs that needs to be solved for.
- The $3N \times 3N$ block **mobility matrix** $\boldsymbol{\mathcal{M}}$ has a **simple pairwise physical interpretation**:
  The 3×3 block $\mathbf{M}_{ij}$ maps a force on blob j to a velocity of blob i,

$$\mathbf{M}_{ij} \approx \eta^{-1} \int \delta_a(\mathbf{q}_i - \mathbf{r}) \mathbf{G}(\mathbf{r}, \mathbf{r}') \delta_a(\mathbf{q}_j - \mathbf{r}') \, d\mathbf{r} d\mathbf{r}' \qquad (2)$$

  where $\mathbf{G}$ is the Green's function (**Oseen tensor** for unbounded).

Suspensions of Rigid Bodies

- Taking yet one more Schur complement we get

$$\left[\begin{array}{c}\mathcal{U}\\ \boldsymbol{\Omega}\end{array}\right]=\mathcal{N}\left[\begin{array}{c}\mathcal{F}\\ \mathcal{T}\end{array}\right]+\left(2k_{B}T\mathcal{N}\right)^{\frac{1}{2}}\mathcal{W}.$$

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

$$\mathcal{N}=\left(\mathcal{K}^{\star}\mathcal{M}^{-1}\mathcal{K}\right)^{-1}.$$

- If a fluctuating fluid solver is used it gives an **explicit square root** of

$$\mathcal{N}^{\frac{1}{2}}=\sqrt{2k_{B}T}\,\mathcal{N}\mathcal{K}^{\star}\mathcal{M}^{-1}\mathcal{J}\mathcal{L}^{-1}\mathbf{D}_{v}\mathcal{W}.$$

  Observe that **discrete fluctuation-dissipation balance** is guaranteed,

$$\mathcal{N}^{\frac{1}{2}}\left(\mathcal{N}^{\frac{1}{2}}\right)^{\star}=\mathcal{N}\mathcal{K}^{\star}\mathcal{M}^{-1}\left(\mathcal{J}\mathcal{L}^{-1}\mathbf{L}\mathcal{L}^{-1}\mathcal{S}\right)\mathcal{M}^{-1}\mathcal{K}\mathcal{N}=$$

$$\mathcal{N}\mathcal{K}^{\star}\mathcal{M}^{-1}\mathcal{M}\mathcal{M}^{-1}\mathcal{K}\mathcal{N}=\mathcal{N}\left(\mathcal{K}^{\star}\mathcal{M}^{-1}\mathcal{K}\right)\mathcal{N}=\mathcal{N}\mathcal{N}^{-1}\mathcal{N}=\mathcal{N}.$$

How to Approximate the Mobility

- If we have a way to approximate the (action of) the mobility matrix $\mathcal{M}$ we can also do this **without invoking a fluid solver**.
- We need to be able to solve

$$\mathcal{N}^{-1}\mathcal{Y} = \left(\mathcal{K}^{\star}\mathcal{M}^{-1}\mathcal{K}\right)\mathcal{Y} = \mathcal{R} + \widetilde{\mathcal{R}},$$

  which we can either do using direct or iterative solvers.
- There are different ways to obtain $\mathcal{M}$:
  - In unbounded domains we can just use the **Rotne-Prager-Yamakawa tensor** (RPY) (always SPD!).
  - In simple geometries such as a single wall we can use a **generalization of RPY** [5].
  - For periodic domains we can use Ewald-type summations or **non-uniform FFTs** with a fluctuating **spectral fluid solver**.
  - In more general cases we can use a fluctuating **FEM/FVM fluid Stokes solver** [6, 7].

Brownian motion under gravity

- We consider the Brownian motion of a single rigid body near a no-slip boundary.
- Temporal integration of the overdamped equations is done using a **random finite different (RFD)** approach as described by Steven Delong.
- Number of blobs is small and we have a simple geometry so we use approximate **Blake-Rotne-Prager tensor** (Brady & Swan [8])
- For this test we use **direct linear algebra** to compute $\mathcal{N}$ and Cholesky factorization to compute $\mathcal{N}^{\frac{1}{2}}$.
- We add gravity which makes the equilibrium **Gibbs-Boltzmann distribution** be

$$P_{GB}(\mathcal{Q}, \boldsymbol{\Theta}) \sim \exp\left[-\frac{mgh + U_{\text{steric}}}{k_B T}\right],$$

where h is the center-of-mass height and U_{steric} is a Yukawa-type repulsion with the wall.

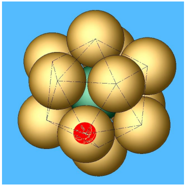
Quasi-2D Diffusion

- Brownian motion is confined near the bottom wall so it **quasi-two dimensional**.
- Without external forcing the Brownian motion along the wall should be isotropic diffusive at long time scales.
- A naive guess for the **effective 2D diffusion coefficient** would be the Gibbs-Boltzmann average of the parallel translational mobility:

$$D_{\parallel} = k_B T \left\langle \mu_{\parallel} \right\rangle_{\text{GB}} .$$

- This is in fact a theorem for a sphere because rotational Brownian motion does not change the mobility.
  Is it true for non-spherical particles?

MSD for a sphere



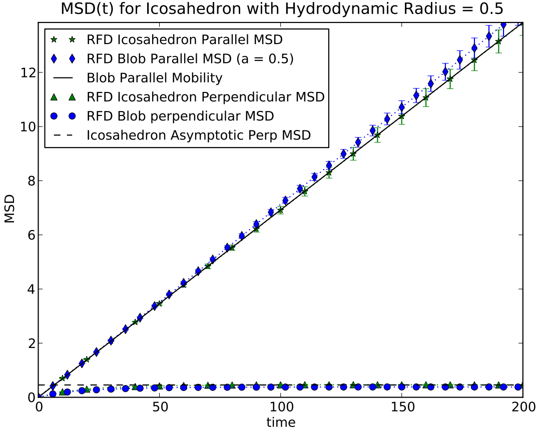
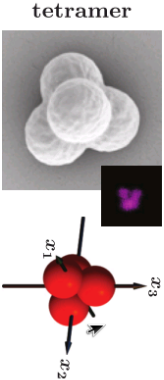


Figure: Mean square displacement (MSD) for a non-uniform **spherical particle** of unit diameter discretized as an icosahedron of 12 blobs or just a single blob.

MSD for a tetrahedron



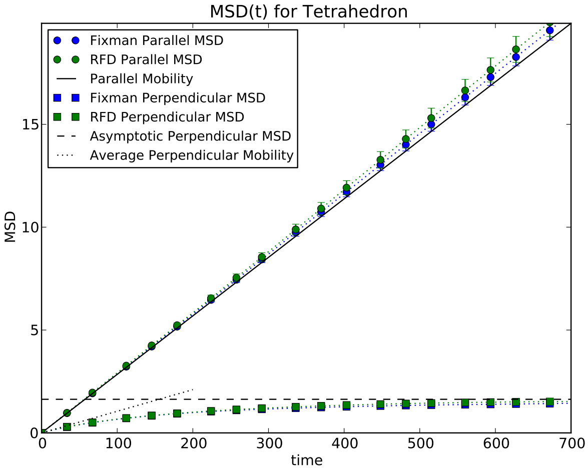
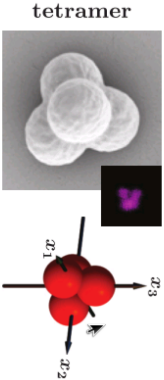
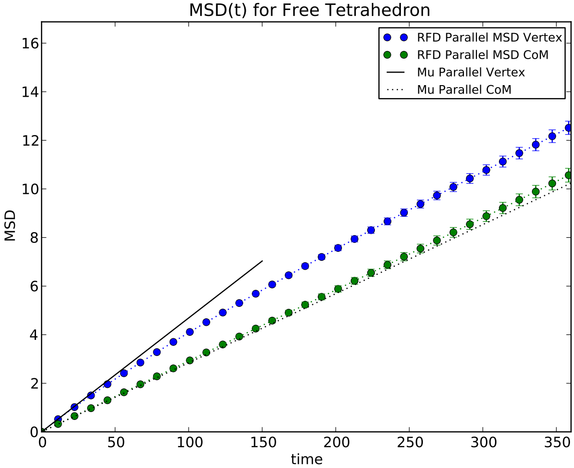


Figure: MSD for a **non-spherical particle** (tetrahedron/tetramer).

The choice of tracking point matters





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Resolving lubrication forces

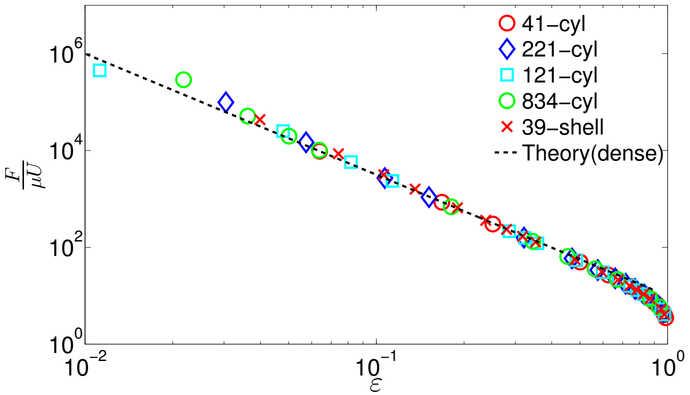

Figure: The drag coefficient for a periodic array of cylinders in steady Stokes flow for close-packed arrays with inter-particle gap ε, showing the correct asymptotic $\varepsilon^{-\frac{5}{2}}$ lubrication force divergence.

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