Brownian Dynamics without Green's Functions

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~~Particle~~ Methods for Micro- and Nano-flows
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Outline

Levels of Coarse-Graining

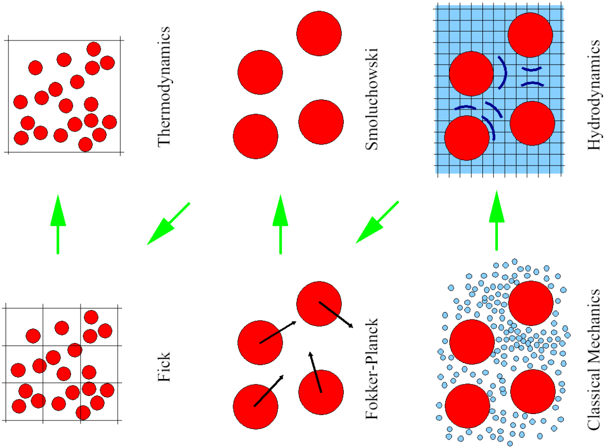

Figure: From Pep Español, "Statistical Mechanics of Coarse-Graining"

Incompressible Fluctuating Hydrodynamics

- The colloidal are immersed in an incompressible fluid that we assume can be described by the time-dependent **fluctuating incompressible Stokes** equations,

$$\begin{aligned} \rho\partial_t \mathbf{v} + \boldsymbol{\nabla}\pi &= \eta\boldsymbol{\nabla}^2\mathbf{v} + \mathbf{f} + \sqrt{2\eta k_B T}\ \boldsymbol{\nabla}\cdot\boldsymbol{\mathcal{Z}} \\ \boldsymbol{\nabla}\cdot\mathbf{v} &= 0, \end{aligned} \tag{1}$$

  along with appropriate boundary conditions.

- Here the **stochastic momentum flux** is modeled via a random Gaussian tensor field $\boldsymbol{\mathcal{Z}}(\mathbf{r},t)$ whose components are white in space and time with mean zero and covariance

$$\langle \mathcal{Z}_{ij}(\mathbf{r},t)\mathcal{Z}_{kl}(\mathbf{r}',t')\rangle = (\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk})\,\delta(t-t')\delta(\mathbf{r}-\mathbf{r}'). \tag{2}$$

Brownian Particle Model

- Consider a **Brownian "particle"** of size a with position $\mathbf{q}(t)$ and velocity $\mathbf{u} = \dot{\mathbf{q}}$, and the velocity field for the fluid is $\mathbf{v}(\mathbf{r}, t)$.
- We do not care about the fine details of the flow around a particle, which is nothing like a hard sphere with stick boundaries in reality anyway.
- Take an **Immersed Boundary Method** (IBM) approach and describe the fluid-blob interaction using a localized smooth **kernel** $\delta_a(\Delta\mathbf{r})$ with compact support of size a (integrates to unity).
- Often presented as an interpolation function for point Lagrangian particles but here a is a **physical size** of the particle (as in the **Force Coupling Method** (FCM) of Maxey *et al*).
- We will call our particles "**blobs**" since they are not really point particles.

Local Averaging and Spreading Operators

- Postulate a **no-slip condition** between the particle and local fluid velocities,

$$\dot{\mathbf{q}} = \mathbf{u} = [\mathbf{J}(\mathbf{q})]\,\mathbf{v} = \int \delta_a(\mathbf{q} - \mathbf{r})\,\mathbf{v}(\mathbf{r}, t)\,d\mathbf{r},$$

  where the *local averaging* linear operator $\mathbf{J}(\mathbf{q})$ averages the fluid velocity inside the particle to estimate a local fluid velocity.
- The **induced force density** in the fluid because of the force $\mathbf{F}$ applied on particle is:

$$\mathbf{f} = -\mathbf{F}\delta_a(\mathbf{q} - \mathbf{r}) = -[\mathbf{S}(\mathbf{q})]\,\mathbf{F},$$

  where the *local spreading* linear operator $\mathbf{S}(\mathbf{q})$ is the reverse (adjoint) of $\mathbf{J}(\mathbf{q})$.
- The physical **volume** of the particle ΔV is related to the shape and width of the kernel function via

$$\Delta V = (\mathbf{JS})^{-1} = \left[\int \delta_a^2(\mathbf{r})\,d\mathbf{r}\right]^{-1}. \tag{3}$$

Fluctuation-Dissipation Balance

- One must ensure **fluctuation-dissipation balance** in the coupled fluid-particle system.
- The **stationary** (equilibrium) distribution must be the **Gibbs distribution**

$$P_{eq}(\mathbf{q}) = Z^{-1} \exp\left(-U(\mathbf{q})/k_B T\right), \tag{4}$$

where $\mathbf{F}(\mathbf{q}) = -\partial U(\mathbf{q})/\partial \mathbf{q}$ with $U(\mathbf{q})$ a conservative potential.

- No entropic contribution to the coarse-grained free energy because our formulation is isothermal and the particles do not have internal structure.
- In order to ensure that the dynamics is time reversible with respect to an appropriate Gibbs-Boltzmann distribution), the thermal or **stochastic drift** forcing

$$\mathbf{f}_{\text{th}} = (k_B T)\, \partial_{\mathbf{q}} \cdot \mathbf{S}\,(\mathbf{q}) \tag{5}$$

needs to be added to the fluid equation [1, 2, 3].

Viscous-Dominated Flows

- We consider n spherical neutrally-buoyant particles of radius a in d dimensions, having spatial positions $\mathbf{q} = \{\mathbf{q}_1, \dots, \mathbf{q}_N\}$ with $\mathbf{q}_i = (q_i^{(1)}, \dots, q_n^{(d)})$.
  Let script $\mathcal{J}$ and $\mathcal{S}$ denote composite fluid-particles interaction operators.
- Let us assume that the Schmidt number is very large,

$$\mathrm{Sc} = \eta / (\rho\chi) \gg 1,$$

  where $\chi \approx k_B T / (6\pi\eta a)$ is a typical value of the diffusion coefficient of the particles [4].
- To obtain the asymptotic dynamics in the limit $\mathrm{Sc} \to \infty$ heuristically, we delete the inertial term $\rho\partial_t \mathbf{v}$ in (1), $\boldsymbol{\nabla} \cdot \mathbf{v} = 0$ and

$$\begin{aligned} \boldsymbol{\nabla}\pi &= \eta\boldsymbol{\nabla}^2\mathbf{v} + \boldsymbol{\mathcal{S}}\mathbf{F} + \sqrt{2\eta k_B T}\;\boldsymbol{\nabla}\cdot\boldsymbol{\mathcal{Z}} \quad \Rightarrow \\ \mathbf{v} &= \eta^{-1}\boldsymbol{\mathcal{L}}^{-1}\left(\boldsymbol{\mathcal{S}}\mathbf{F} + \sqrt{2\eta k_B T}\;\boldsymbol{\nabla}\cdot\boldsymbol{\mathcal{Z}}\right), \end{aligned} \tag{6}$$

  where $\boldsymbol{\mathcal{L}}^{-1} \succeq \mathbf{0}$ is the Stokes solution operator.

Overdamped Limit

- A rigorous adiabatic mode elimination procedure informs us that the correct interpretation of the noise term in this equation is the **kinetic stochastic integral**,

$$\frac{d\mathbf{q}(t)}{dt} = \mathcal{J}(\mathbf{q})\mathcal{L}^{-1}\left[\frac{1}{\eta}\mathcal{S}(\mathbf{q})\mathbf{F}(\mathbf{q}) + \sqrt{\frac{2k_B T}{\eta}}\,\boldsymbol{\nabla}\diamond\boldsymbol{\mathcal{Z}}(\mathbf{r},t)\right]. \tag{7}$$

- This is equivalent to the standard equations of **Brownian Dynamics** (BD),

$$\frac{d\mathbf{q}}{dt} = \boldsymbol{\mathcal{M}}\mathbf{F} + (2k_B T\boldsymbol{\mathcal{M}})^{\frac{1}{2}}\,\boldsymbol{\mathcal{W}}(t) + k_B T\,(\partial_{\mathbf{q}}\cdot\boldsymbol{\mathcal{M}}), \tag{8}$$

where $\boldsymbol{\mathcal{M}}(\mathbf{q}) \succeq \mathbf{0}$ is the symmetric positive semidefinite (SPD) mobility matrix

$$\boldsymbol{\mathcal{M}} = \eta^{-1}\mathcal{J}\mathcal{L}^{-1}\mathcal{S}.$$

Brownian Dynamics via Fluctuating Hydrodynamics

- It is not hard to show that $\mathcal{M}$ is very similar to the **Rotne-Prager mobility** used in BD, for particles i and j,

$$\mathcal{M}_{ij} = \eta^{-1} \int \delta_a(\mathbf{q}_i - \mathbf{r})\mathbf{K}(\mathbf{r}, \mathbf{r}')\delta_a(\mathbf{q}_j - \mathbf{r}') \; d\mathbf{r}d\mathbf{r}' \tag{9}$$

  where $\mathbf{K}$ is the Green's function for the Stokes problem (**Oseen tensor** for infinite domain).

- The self-mobility defines a consistent **hydrodynamic radius** of a blob,

$$\mathcal{M}_{ii} = \mathcal{M}_{\text{self}} = \frac{1}{6\pi\eta a}\mathbf{I}.$$

- For well-separated particles we get the correct Faxen expression,

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

- At smaller distances the mobility is regularized in a natural way and positive-semidefiniteness ensured automatically.

Numerical Methods

- Both **compressible and incompressible, inertial and overdamped**, numerical methods have been implemented by Florencio Balboa (UAM) on GPUs for periodic BCs (public-domain!), and in the parallel **IBAMR code** of Boyce Griffith by Steven Delong for general boundary conditions (to be made public-domain next fall!).
- Spatial discretization is based on previously-developed **staggered schemes** for fluctuating hydro [5] and the **immersed-boundary method kernel functions** of Charles Peskin.
- Temporal discretization follows a second-order **splitting algorithm** (move particle + update momenta), and is limited in **stability** only by **advective CFL**.
- We have constructed specialized temporal integrators that ensure **discrete fluctuation-dissipation balance**, including for the overdamped case.

(Simple) Midpoint Scheme

Fluctuating Immersed Boundary Method (FIBM) method:

- Solve a **steady-state Stokes problem** (here $\delta \ll 1$)

$$\begin{aligned} \boldsymbol{\nabla}\boldsymbol{\pi}^{n} &= \eta\boldsymbol{\nabla}^{2}\mathbf{v}^{n}+\sqrt{2\eta k_{B}T}\,\boldsymbol{\nabla}\cdot\boldsymbol{\mathcal{Z}}^{n}+\mathbf{S}^{n}\mathbf{F}\left(\mathbf{q}^{n}\right) \\ &+ \frac{k_{B}T}{\delta}\left[\mathbf{S}\left(\mathbf{q}^{n}+\frac{\delta}{2}\widetilde{\mathbf{W}}^{n}\right)-\mathbf{S}\left(\mathbf{q}^{n}-\frac{\delta}{2}\widetilde{\mathbf{W}}^{n}\right)\right]\widetilde{\mathbf{W}}^{n} \\ \boldsymbol{\nabla}\cdot\mathbf{v}^{n} &= 0. \end{aligned}$$

- **Predict** particle position:

$$\mathbf{q}^{n+\frac{1}{2}}=\mathbf{q}^{n}+\frac{\Delta t}{2}\mathcal{J}^{n}\mathbf{v}$$

- **Correct** particle position,

$$\mathbf{q}^{n+1}.=\mathbf{q}^{n}+\Delta t\mathcal{J}^{n+\frac{1}{2}}\mathbf{v}.$$

Slit Channel

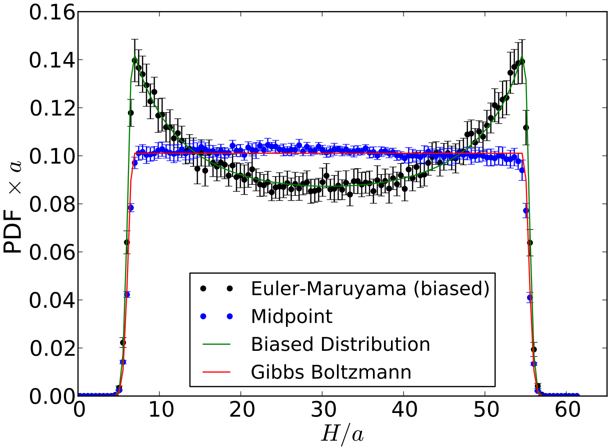

Figure: Probability distribution of the distance H to one of the walls for a freely-diffusing blob in a two dimensional slit channel.

Equilibrium Radial Correlation Function

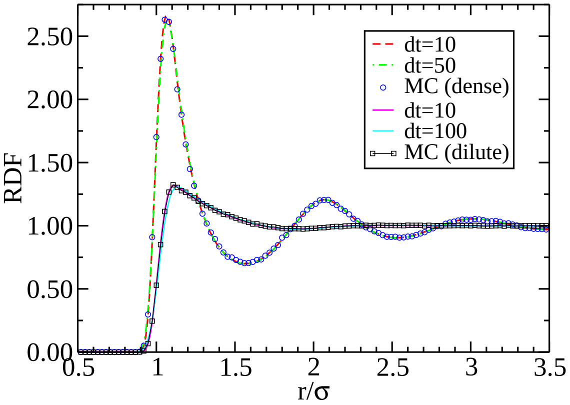

Figure: Equilibrium radial distribution function $g_2(\mathbf{r})$ for a suspension of blobs interacting with a repulsive LJ (WCA) potential.

Hydrodynamic Interactions

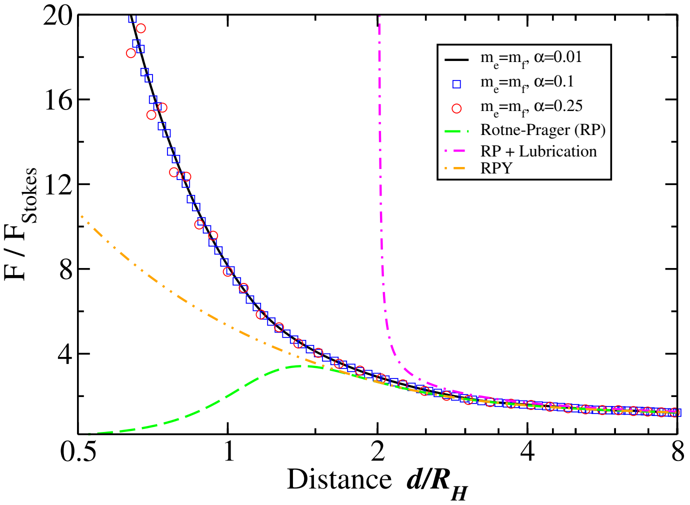

Figure: Effective hydrodynamic force between two approaching blobs at small Reynolds numbers, $\frac{F}{F_{\text{St}}} = -\frac{2F_0}{6\pi\eta R_H v_r}$.

Diffusive Dynamics

- At long times, the motion of the particle is diffusive with a diffusion coefficient $\chi = \lim_{t\to\infty} \chi(t) = \int_{t=0}^{\infty} C(t)dt$, where

$$\chi(t) = \frac{\Delta q^2(t)}{2t} = \frac{1}{2dt} \left\langle \left[\mathbf{q}(t) - \mathbf{q}(0)\right]^2 \right\rangle .$$

- The Stokes-Einstein relation predicts

$$\chi = \frac{k_B T}{\mu} \text{ (Einstein) and } \chi_{SE} = \frac{k_B T}{6\pi\eta a} \text{ (Stokes)}, \quad (10)$$

where for our blob a is on the order of the fluid solver grid spacing.

- The dimensionless Schmidt number $S_c = \nu/\chi_{SE}$ controls the separation of time scales.
- Self-consistent theory predicts a correction to Stokes-Einstein's relation for small S_c,

$$\chi \left(\nu + \frac{\chi}{2}\right) = \frac{k_B T}{6\pi\rho a}.$$

Stokes-Einstein Corrections

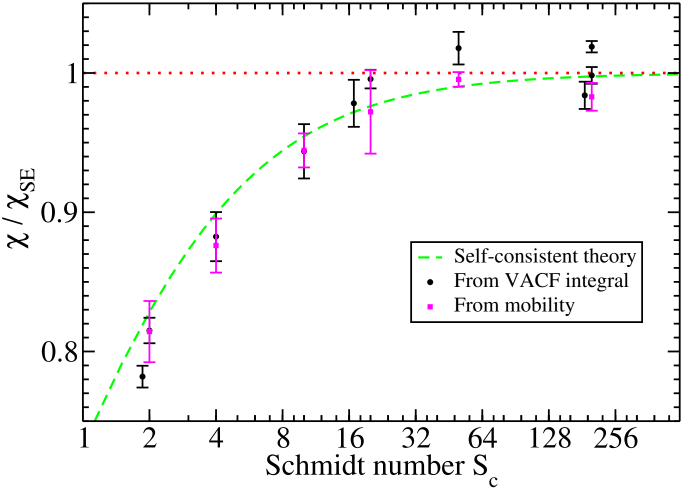

Figure: Corrections to Stokes-Einstein with changing viscosity $\nu = \eta/\rho$, $m_e = m_f = \rho \Delta V$.

Colloidal Gellation: Cluster collapse

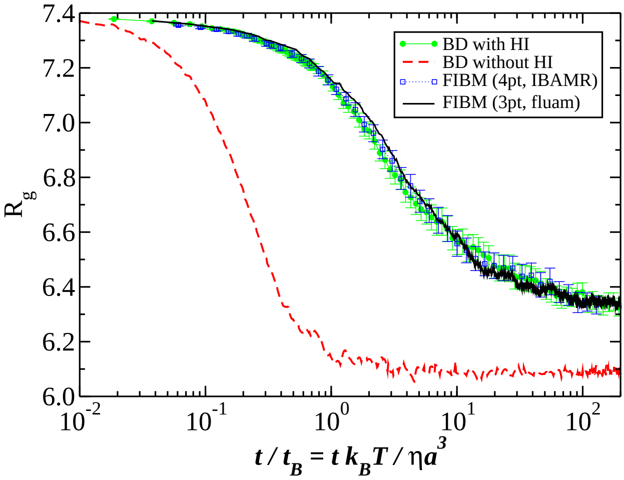

Figure: Relaxation of the radius of gyration of a colloidal cluster of 13 spheres toward equilibrium, taken from Furukawa+Tanaka.

Immersed Rigid Blobs

- Unlike a **rigid sphere**, a blob particle would not perturb a pure shear flow.
- In the far field our blob particle looks like a force monopole (**stokeset**), and does not exert a force dipole (**stresslet**) on the fluid.
- Similarly, since here we do not include **angular velocity** degrees of freedom, our blob particle does not exert a **torque** on the fluid (rotlet).
- It is possible to include rotlet and stresslet terms, as done in the fluctuating force coupling method [6] and **Stokesian Dynamics** in the deterministic setting.
- Maintaining **fluctuation-dissipation balance** more challenging.

Conclusions

- **Fluctuating hydrodynamics** seems to be a very good coarse-grained model for fluids, and coupled to immersed particles to model Brownian suspensions.
- The **minimally-resolved blob approach** provides a low-cost but reasonably-accurate representation of rigid particles in flow.
- We have recently successfully extended the blob approach to **reaction-diffusion problems** (with Amneet Bhalla and Neelesh Patankar).
- Particle and fluid **inertia** can be included in the description, or, an **overdamped limit** can be taken if $S_c \gg 1$.
- More **complex particle shapes** can be built out of a collection of blobs to form a **rigid body**.

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