Brownian Dynamics without Green’s Functions

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Outline

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Levels of Coarse-Graining

Figure: From Pep Español, “Statistical Mechanics of Coarse-Graining”
The colloidal are immersed in an incompressible fluid that we assume can be described by the time-dependent fluctuating incompressible Stokes equations,

\[
\rho \partial_t v + \nabla \pi = \eta \nabla^2 v + f + \sqrt{2\eta k_B T} \nabla \cdot \mathcal{Z} \tag{1}
\]
\[
\nabla \cdot v = 0,
\]

along with appropriate boundary conditions.

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

\[
\langle \mathcal{Z}_{ij}(r, t) \mathcal{Z}_{kl}(r', t') \rangle = (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk}) \delta(t - t') \delta(r - r'). \tag{2}
\]
Consider a Brownian “particle” of size $a$ with position $q(t)$ and velocity $u = \dot{q}$, and the velocity field for the fluid is $v(r, t)$.

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 $\delta_a(\Delta r)$ 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.
Postulate a no-slip condition between the particle and local fluid velocities,

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

where the local averaging linear operator \( J(q) \) averages the fluid velocity inside the particle to estimate a local fluid velocity.

The induced force density in the fluid because of the force \( F \) applied on particle is:

\[ f = -F\delta_a(q - r) = -[S(q)]F, \]

where the local spreading linear operator \( S(q) \) is the reverse (adjoint) of \( J(q) \).

The physical volume of the particle \( \Delta V \) is related to the shape and width of the kernel function via

\[ \Delta V = (JS)^{-1} = \left[ \int \delta_a^2(r) \, dr \right]^{-1}. \]
Fluid-Particle Coupling

**Fluctuation-Dissipation Balance**

- One must ensure **fluctuation-dissipation balance** in the coupled fluid-particle system.
- The **stationary** (equilibrium) distribution must be the **Gibbs distribution**
  \[ P_{eq}(q) = Z^{-1} \exp \left( -\frac{U(q)}{k_B T} \right), \]
  \[ (4) \]
  where \( F(q) = -\frac{\partial U(q)}{\partial q} \) with \( U(q) \) 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
  \[ f_{th} = (k_B T) \partial_q \cdot S(q) \]
  \[ (5) \]
  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 $\mathbf{q} = \{\mathbf{q}_1, \ldots, \mathbf{q}_N\}$ with $\mathbf{q}_i = (q_i^{(1)}, \ldots, q_i^{(d)})$.

Let script $\mathcal{J}$ and $\mathcal{S}$ denote composite fluid-particles interaction operators.

- Let us assume that the Schmidt number is very large,
  \[ \text{Sc} = \frac{\eta}{(\rho \chi)} \gg 1, \]
  where $\chi \approx \frac{k_B T}{6\pi\eta a}$ is a typical value of the diffusion coefficient of the particles [4].

- To obtain the asymptotic dynamics in the limit $\text{Sc} \to \infty$ heuristically, we delete the inertial term $\rho \partial_t \mathbf{v}$ in (1), $\nabla \cdot \mathbf{v} = 0$ and
  \[ \nabla \pi = \eta \nabla^2 \mathbf{v} + \mathcal{S} \mathbf{F} + \sqrt{2\eta k_B T} \nabla \cdot \mathbf{Z} \Rightarrow \]
  \[ \mathbf{v} = \eta^{-1} \mathcal{L}^{-1} \left( \mathcal{S} \mathbf{F} + \sqrt{2\eta k_B T} \nabla \cdot \mathbf{Z} \right), \]
  where $\mathcal{L}^{-1} \succeq 0$ is the Stokes solution operator.
A rigorous adiabatic mode elimination procedure informs us that the correct interpretation of the noise term in this equation is the kinetic stochastic integral,

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

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

$$\frac{dq}{dt} = \mathcal{M}F + (2k_B T \mathcal{M})^{\frac{1}{2}} \mathcal{W}(t) + k_B T (\partial_q \cdot \mathcal{M}), \quad (8)$$

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

$$\mathcal{M} = \eta^{-1} \mathcal{J} \mathcal{L}^{-1} S.$$
It is not hard to show that $\mathcal{M}$ is very similar to the Rotne-Prager mobility used in BD, for particles $i$ and $j$,

$$
\mathcal{M}_{ij} = \eta^{-1} \int \delta_a(q_i - r) K(r, r') \delta_a(q_j - r') \, dr \, dr'
$$

where $K$ 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,

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

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

$$
\mathcal{M}_{ij} \approx \eta^{-1} \left( I + \frac{a^2}{6} \nabla_r^2 \right) \left( I + \frac{a^2}{6} \nabla_{r'}^2 \right) K(r - r')|_{r'=q_i}.
$$

At smaller distances the mobility is regularized in a natural way and positive-semidefiniteness ensured automatically.
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.
Fluctuating Immersed Boundary Method (FIBM) method:

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

\[
\nabla \pi^n = \eta \nabla^2 v^n + \sqrt{2\eta k_B T} \nabla \cdot \mathcal{Z}^n + S^n F(q^n) \\
+ \frac{k_B T}{\delta} \left[ S \left( q^n + \frac{\delta}{2} \tilde{W}^n \right) - S \left( q^n - \frac{\delta}{2} \tilde{W}^n \right) \right] \tilde{W}^n
\]

\[\nabla \cdot v^n = 0.\]

- **Predict** particle position:

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

- **Correct** particle position,

\[q^{n+1} = q^n + \Delta t \mathcal{J}^{n+\frac{1}{2}} v.\]
**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 $g_2(r)$ for a suspension of blobs interacting with a repulsive LJ (WCA) potential.
Figure: Effective hydrodynamic force between two approaching blobs at small Reynolds numbers, \( \frac{F}{F_{St}} = -\frac{2F_0}{6\pi \eta R_H v_r} \).
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
  \[ \chi(t) = \frac{\Delta q^2(t)}{2t} = \frac{1}{2t} \langle [q(t) - q(0)]^2 \rangle. \]

- The Stokes-Einstein relation predicts
  \[ \chi = \frac{k_B T}{\mu} \text{ (Einstein)} \quad \text{and} \quad \chi_{SE} = \frac{k_B T}{6\pi \eta a} \text{ (Stokes)}, \]
  \[ \text{ (10)} \]
  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 \( S_c \),
  \[ \chi \left( \nu + \frac{\chi}{2} \right) = \frac{k_B T}{6\pi \rho a}. \]
**Results**

**Stokes-Einstein Corrections**

![Graph showing corrections to Stokes-Einstein with changing viscosity.](image)

**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.
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**.
P. J. Atzberger.

Inertial Coupling Method for particles in an incompressible fluctuating fluid.

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F. Balboa Usabiaga, X. Xie, R. Delgado-Buscalioni, and A. Donev.
The Stokes-Einstein Relation at Moderate Schmidt Number.

Staggered Schemes for Fluctuating Hydrodynamics.

Eric E. Keaveny.
Fluctuating force-coupling method for simulations of colloidal suspensions.