Fast Brownian Dynamics for Colloidal Suspensions

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Modeling Complex Fluids and Gels for Biological Applications
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Colloidal Gelation

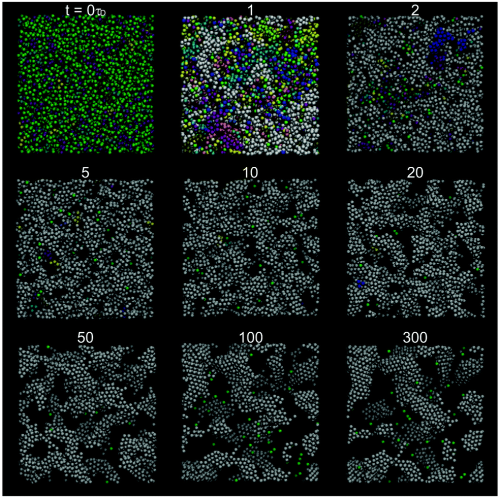

Figure : Colloidal gelation simulated using Brownian Dynamics with HIs (from work of James Swan, MIT Chemical Engineering).

Diffusion in Crowded Environments

- Diffusion of proteins in the cell cytoplasm is strongly dominated by **hydrodynamics and crowding** (Skolnick) [1].
- Modeling this requires **Brownian Dynamics with Hydrodynamic Interactions** that can handle hundreds of thousands of particles of **different sizes and shapes**, some rigid, some perhaps flexible.
- Thus, we have to **give up on high accuracy** methods such as boundary integral formulations, but we cannot throw the baby out with the bathwater and completely neglect hydrodynamics.
- Paper most closely associated to this talk [2]:
  "**Rapid Sampling of Stochastic Displacements in Brownian Dynamics Simulations**"
  A. M. Fiore, F. Balboa Usabiaga, A. Donev and J. W. Swan
  J. Chem. Phys., 146, 124116, 2017 [**Arxiv:1611.09322**].
  Codes at `https://github.com/stochasticHydroTools/PSE`

Goal of this talk

- **Brownian Dynamics with Hydrodynamic Interactions**: **How to efficiently capture the effect of long-ranged hydrodynamic correlations (interactions) in the Brownian motion of 10^6 spherical colloids?**
- Because we want to simulate huge numbers of particles we have to sacrifice accuracy and use a very low-resolution (far-field) approximation for the hydrodynamics: "**long-ranged hydrodynamic interactions** are sufficient for establishing the gel boundary, structure and coarsening kinetics observed in experiments..." (Varga, Wang, Swan, 2015)
- Note: The problem of generating Gaussian variates with a covariance specified by a long-ranged kernel has many **other applications** as well, e.g., in data science, not discussed here.

Brownian Dynamics with Hydrodynamic Interactions (BD-HI)

- The Ito equations of **Brownian Dynamics** (BD) for the (correlated) positions of the N spherical particles $\mathbf{Q}(t) = \{\mathbf{q}_1(t), \dots, \mathbf{q}_N(t)\}$ are

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

  where $\mathcal{B}(t)$ is a vector of Brownian motions, and $\mathbf{F}(\mathbf{Q})$ are forces.
- Here $\mathbf{M}(\mathbf{Q}) \succeq \mathbf{0}$ is a symmetric positive semidefinite (SPD) **mobility matrix**, assumed to have a far-field **pairwise approximation**

$$\mathbf{M}_{ij}(\mathbf{Q}) \equiv \mathbf{M}_{ij}(\mathbf{q}_i, \mathbf{q}_j) = \mathcal{R}(\mathbf{q}_i - \mathbf{q}_j).$$

- Here we use the **Rotne-Prager-Yamakawa (RPY) kernel**:

$$\mathcal{R}(\mathbf{r}) = \frac{k_B T}{6\pi\eta a} \begin{cases} \left(\frac{3a}{4r} + \frac{a^3}{2r^3}\right)\mathbf{I} + \left(\frac{3a}{4r} - \frac{3a^3}{2r^3}\right)\frac{\mathbf{r}\otimes\mathbf{r}}{r^2}, & r > 2a \\ \left(1 - \frac{9r}{32a}\right)\mathbf{I} + \left(\frac{3r}{32a}\right)\frac{\mathbf{r}\otimes\mathbf{r}}{r^2}, & r \le 2a \end{cases}$$

  where a is the radius of the colloidal particles.

Hydrodynamic Correlations

- Observe that in the far-field, $r \gg a$, the RPY tensor becomes the **long-ranged** Oseen tensor

$$\mathcal{R}\left(r \gg a\right) \to \frac{1}{8\pi r}\left(\mathbf{I} + \frac{\mathbf{r} \otimes \mathbf{r}}{r^2}\right). \qquad (2)$$

- To solve the equations of BD numerically (*not* the subject of this talk), one needs two fast routines:
  - A fast matrix-vector product to compute **MF**.
    This can be done using **Fast Multipole Methods (FMM)** (Greengard) in an unbounded domain or using the **Spectral Ewald (SE) Method** [3] (Tornberg) for periodic domains.
  - A fast method to compute $\mathbf{M}^{\frac{1}{2}}\mathbf{W}$, where $\mathbf{W}$ is a vector of Gaussian random variables. More precisely, we want to sample Gaussian random variables with mean zero and covariance $\mathbf{M}$.
    First part of this talk: **How to compute $\mathbf{M}^{\frac{1}{2}}\mathbf{W}$ using a fast method.**

Existing Approaches

- The product $\mathbf{M}^{\frac{1}{2}}\mathbf{W}$ is usually computed iteratively by **repeated multiplication** of a vector by $\mathbf{M}$.
- Traditionally chemical engineers have used an approach by **Fixman** based on a Chebyshev polynomial approximation to the square root.
- Recently, Chow and Saad have developed Krylov subspace **Lanczos methods** [4] for multiplying a vector with the principal square root of $\mathbf{M} = \mathbf{U}\mathbf{\Lambda}\mathbf{U}^T$,

$$\mathbf{M}^{\frac{1}{2}}\mathbf{W} \equiv \mathbf{U}\mathbf{\Lambda}^{\frac{1}{2}}\mathbf{U}^T\mathbf{W} \approx \|\mathbf{W}\|_2 \, \mathbf{V}_m \mathbf{H}_m^{1/2}\mathbf{e}_1,$$

  where $\mathbf{V}_m$ is an orthonormal basis for the Krylov subspace of order m, and $\mathbf{H}_m = \mathbf{V}_m^T\mathbf{M}\mathbf{V}_m$ is a tridiagonal matrix, both computed in the course of a Lanczos iteration through m matrix-vector multiplies.
- The Krylov method is vastly superior, but, because of the long-ranged nature of the Oseen kernel the number of iterations is found to grow with the number of particles, leading to an overall complexity of at least $O\left(N^{4/3}\right)$.

Near-Far field decomposition

- Work done by **Andrew Fiore and James Swan** (MIT Chemical Engineering), with help from **Florencio Balboa** (Courant).
- We don't really need to multiply any particular matrix "square root" by $\mathbf{W}$, rather, we want to generate a Gaussian random vector $\delta\mathbf{U}$ with specified covariance, $\langle(\delta\mathbf{U})(\delta\mathbf{U})^T\rangle = \mathbf{M}$.
- *First key idea*: Use (Spectral) Ewald approach to decompose $\mathbf{M} = \mathbf{M}^{(w)} + \mathbf{M}^{(r)}$ into a **far-field wave-space part** $\mathbf{M}^{(w)}$ and a **near-field real space part** $\mathbf{M}^{(r)}$, then in law,

$$\mathbf{M}^{\frac{1}{2}}\mathbf{W} \overset{\mathrm{d}}{=} \left(\mathbf{M}^{(w)}\right)^{\frac{1}{2}}\mathbf{W}^{(w)} + \left(\mathbf{M}^{(r)}\right)^{\frac{1}{2}}\mathbf{W}^{(r)},$$

  if **both** $\mathbf{M}^{(w)}$ **and** $\mathbf{M}^{(r)}$ **are SPD** and $\langle\mathbf{W}^{(w)}\mathbf{W}^{(r)}\rangle = \mathbf{0}$.
- For the real-space part, use the Krylov Lanczos method to compute $\left(\mathbf{M}^{(r)}\right)^{\frac{1}{2}}\mathbf{W}^{(r)}$ since $\mathbf{M}^{(r)}$ is **sparse and well-conditioned**.
- *Second key idea*: Compute $\mathbf{M}^{(w)}\mathbf{F}$ and $\left(\mathbf{M}^{(w)}\right)^{\frac{1}{2}}\mathbf{W}^{(w)}$ in **Fourier space (using FFTs)** as in fluctuating hydrodynamics.

Spectral RPY

- We need to find an Ewald-like decomposition where both the real space and wave space kernels decay exponentially and are SPD.
- The most physically-relevant and simplest definition of RPY is the integral representation:

$$\mathcal{R}\left(\mathbf{r}_1, \mathbf{r}_2\right) = \mathcal{R}\left(\mathbf{r}_1 - \mathbf{r}_2\right) = \int \delta_a\left(\mathbf{r}_1 - \mathbf{r}'\right) \mathbb{G}\left(\mathbf{r}', \mathbf{r}''\right) \delta_a\left(\mathbf{r}_2 - \mathbf{r}''\right) d\mathbf{r}' d\mathbf{r}'',$$

  where δ_a denotes a surface delta function on a sphere of radius a.
- In other $O(N)$ methods for BD other regularized delta functions have been used (Peskin's in fluctuating immersed boundary methods and Gaussians in the fluctuating force coupling method).
- Here the Green's function for periodic Stokes flow is given by

$$\mathbb{G}\left(\mathbf{x}, \mathbf{y}\right) = \frac{1}{\mu V} \sum_{\mathbf{k} \neq 0} e^{i\mathbf{k}\cdot(\mathbf{x}-\mathbf{y})} \frac{1}{k^2} \left(\mathbf{I} - \hat{\mathbf{k}}\hat{\mathbf{k}}\right).$$

- The surface delta functions in Fourier space give us a sinc factor.

Positively Split Ewald RPY

- This gives us a previously-unappreciated simple spectral representation of the periodic RPY tensor:

$$\mathcal{R}(\mathbf{r}) = \frac{1}{\mu V} \sum_{\mathbf{k} \neq 0} e^{i \mathbf{k} \cdot \mathbf{r}} \frac{1}{k^2} \text{sinc}^2(ka) \left(\mathbf{I} - \hat{\mathbf{k}}\hat{\mathbf{k}} \right). \tag{3}$$

- We can now directly apply Hasimoto's Ewald-like decomposition [3] to RPY to get the desired **Positively Split Ewald (PSE) RPY tensor**, $\mathcal{R} = \mathcal{R}_\xi^{(w)} + \mathcal{R}_\xi^{(r)}$,

$$\mathcal{R}_\xi^{(w)}(\mathbf{r}) = \frac{1}{\mu V} \sum_{\mathbf{k} \neq 0} e^{i \mathbf{k} \cdot \mathbf{r}} \frac{\text{sinc}^2(ka)}{k^2} H(k, \xi) \left(\mathbf{I} - \hat{\mathbf{k}}\hat{\mathbf{k}} \right), \tag{4}$$

where the Hasimoto splitting function is determined by the **splitting parameter** ξ,

$$H(k, \xi) = \left(1 + \frac{k^2}{4\xi^2} \right) e^{-k^2/4\xi^2}. \tag{5}$$

Real-space part

- Converting back to real space we get

$$\mathcal{R}_{\xi}^{(r)}(\mathbf{r}) = F(r,\xi)\,(\mathbf{I} - \hat{\mathbf{r}}\hat{\mathbf{r}}) + G(r,\xi)\,\hat{\mathbf{r}}\hat{\mathbf{r}}, \tag{6}$$

  where and $F(r,\xi)$ and $G(r,\xi)$ are scalar functions that **both decay exponentially** in $r^2\xi^2$.
  Analytical formulas are complicated but these can easily be **tabulated** for fast evaluation.

- Diagonal part is well-defined,

$$\mathbf{M}_{ii}^{(r)} = \mathcal{R}^{(r)}(\mathbf{0}) = \frac{1}{24\pi^{3/2}\mu\xi a^2}\left(1 - e^{-4a^2\xi^2} + 4\pi^{1/2}a\xi\,\mathrm{erfc}\,(2a\xi)\right)\mathbf{I}.$$

- If we choose $0 \leq H(k,\xi) \leq 1$ (satisfied by Hasimoto but not Beenakker) we obtain SPD real and wave space parts.

Conditioning

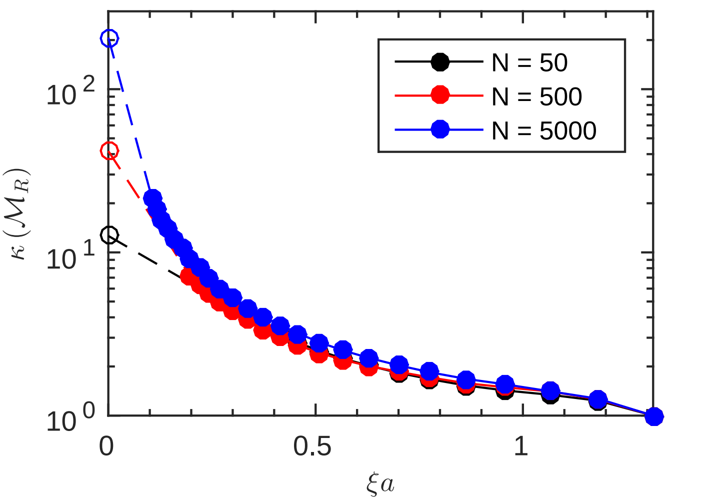

Figure : Condition number of $\mathbf{M}^{(r)}$ for varying number of particles N [2].

Fourier-space part

- The wave space component of the mobility can be applied efficiently using FFTs as

$$\mathbf{M}^{(w)} = \mathbf{D}^{-1}\mathbf{B}\mathbf{D} = \left(\mathbf{D}^{\dagger}\mathbf{B}^{1/2}\right)\left(\mathbf{D}^{\dagger}\mathbf{B}^{1/2}\right)^{\dagger}, \tag{7}$$

where $\mathbf{D}$ is the non-uniform FFT (NUFFT) of Greengard/Lee [3] and

$$\mathbf{B}^{1/2} = \mathrm{Diag}\left(\frac{1}{\mu V}\frac{\mathrm{sinc}^2\left(ka\right)}{k^2}H(k,\xi)\right)^{1/2}.$$

- This shows that the wave space Brownian displacement can be calculated with a single call to the NUFFT,

$$\left(\mathbf{M}^{(w)}\right)^{\frac{1}{2}}\mathbf{W}^{(w)} \equiv \mathbf{D}^{\dagger}\mathbf{B}^{1/2}\mathbf{W}^{(w)}. \tag{8}$$

- This is basically **equivalent to fluctuating hydrodynamics** (putting stochastic forcing on fluid rather than on particles) as in existing methods, but now corrected in the near field.

Efficiency

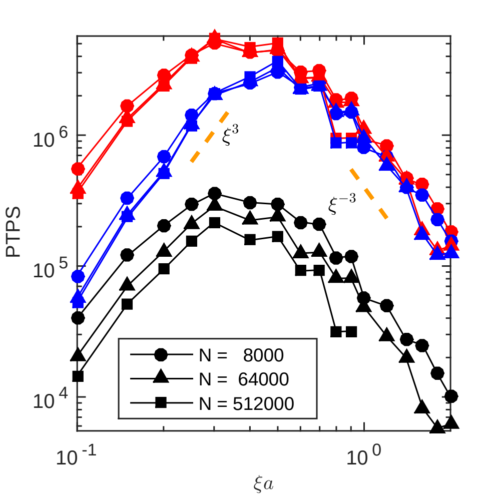

Figure : Particle timesteps per second (PTPS) for a random suspension of hard spheres ($\phi = 0.1$) implemented as a plugin to the **HOOMD GPU framework**. Red=**MF**, blue=$\mathbf{M}^{\frac{1}{2}}\mathbf{W}$ using PSE, black=$\mathbf{M}^{\frac{1}{2}}\mathbf{W}$ without PSE.

Rigid Multiblob Models

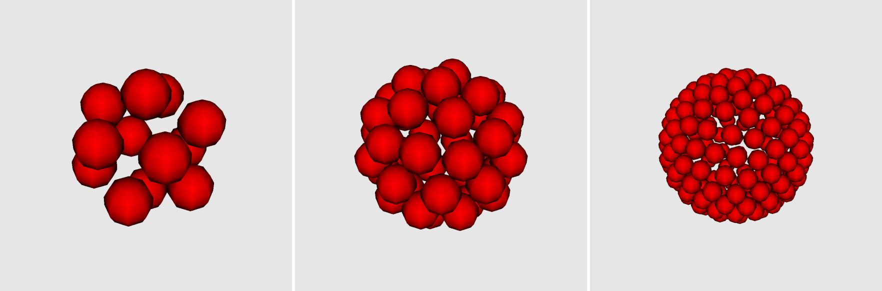

Figure : Blob or "raspberry" models of a spherical colloid.

- The rigid body is discretized through a number of spherical "**beads**" or "**blobs**" which interact via the **Rotne-Prager-Yamakawa** tensor.
- Standard is **stiff springs** but we want **rigid multiblobs** [5].
- We do this efficiently for $10^3 - 10^4$ particles (10^5 blobs) using **iterative solvers** and specially-designed **temporal integrators**.

Nonspherical Rigid Multiblobs

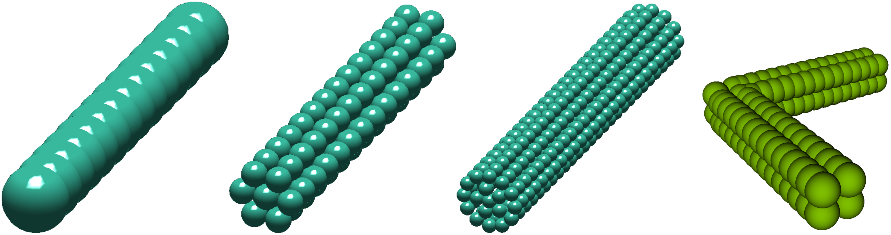

Figure : Rigid multiblob models of colloidal particles manufactured in recent experimental work.

Example: Confined Boomerang Suspension

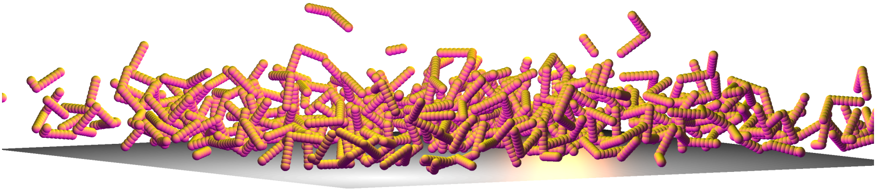

Figure : Quasi-periodic suspension of sedimented colloidal boomerangs.

Conclusions

- **Ewald (Hasimoto) splitting** can be used to accelerate both deterministic and stochastic colloidal simulations in periodic domains.
- Key is to ensure that **both the near-field and far-field are (essentially) SPD** so one piece of the noise is generated using FFTs and the other using an iterative method.
- Using these principles we have constructed a **linear-scaling Brownian dynamics** method (10^6 particles on a GPU), and a linear-scaling fluctuating boundary element method.
- The far-field can be done in **non-periodic but finite domains** using a discrete Stokes solver and fluctuating hydrodynamics. **How about unbounded or doubly-periodic suspensions, e.g., diffusion on interfaces (e.g., lipid bi-layers)?**
- **Can a similar idea be used with grid-free fast multipole methods?**

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