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Fluctuating force-coupling method for interacting colloids

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Abstract Brownian motion plays an important role in the dynamics of colloidal suspensions. It affects rheological properties, influences the self-assembly of structures, and regulates particle transport. While including Brownian motion in simulations is necessary to reproduce and study these effects, it is computationally intensive due to the configuration dependent statistics of the particles' random motion. We will present recent work that speeds up this calculation for the force-coupling method (FCM), a regularized multipole approach to simulating suspensions at large-scale. We show that by forcing the surrounding fluid with a configuration-independent, white-noise stress, fluctuating FCM yields the correct particle random motion, even when higher-order terms, such as the stresslets, are included in the multipole expansion. We present results from several simulations demonstrating the effectiveness of this approach for modern problems in colloidal science and discuss open questions such as the extension of fluctuating FCM to dense suspensions.

Keywords: Brownian motion, colloid simulations, suspensions, Stokes flow

1. Introduction

Brownian motion is the random movement of particles suspended in liquid [33] resulting from the many collisions between the particles and the molecules that make up the surrounding fluid. In addition to altering the motion of individual particles, Brownian motion can also affect the mechanical and rheological properties of suspensions themselves [5, 15, 7]. Brownian motion also plays a strong role in microparticle self-assembly and aggregation-based fabrication techniques [40, 17], as well as in the dynamics and pattern formation of active and field-responsive colloids [31, 36, 30, 39].

Characterising and quantifying the role of Brownian motion in these contexts where interparticle forces, hydrodynamic interactions, and geometric constraints play a strong role presents a current computational challenge. Traditional techniques such as Brownian dynamics [12] and Stokesian dynamics [6], rely on introducing random particle velocities at each time step. These random particle velocities, however, must have correlations proportional to the hydrodynamic mobility matrix [11, 4] and generating such random velocities requires computing the square root of the mobility matrix, an $\mathscr{O}(N^3)$ computation, at every time step.

While several approaches [14, 8] have been devised to overcome these speed up this calculation, we will instead utilise random flow fields generated by a fluctuating stress [27, 16] to determine random particle motion with the correct statistics. The advantage of this approach is that the fluctuating stress is spatial white noise and the matrix square root computation is not required. As a result, the usage of fluctuating stresses to resolve Brownian motion has been demonstrated in a variety of simulation techniques such as the Lattice-Boltzmann method [22, 24, 25, 23], distributed Lagrange multiplier (DLM) method [35], and implementations of the immersed-boundary method [32, 2, 1, 37, 10]. The fluctuating stress approach can also be used to resolve the fluctuations of flexible structures, even in cases where inertial effects are present and lead to power-law tails in the time-correlations of the particle velocities [19, 33].

We recently developed the fluctuating force-coupling method (FCM) [20] which combines the deterministic FCM [29, 28, 9, 41] with the random fluid flows produced by a white-noise fluctuating stress. This paper presents an overview of this new method and summarises recent results obtained by using it. FCM itself is based on representing particles using regularised multipole expansions in the Stokes equations and recovering their motion through volume averaging of the resulting fluid motion. It has been used in a variety of scenarios, including microfluidic contexts, and can be enhanced to include finite Reynolds number effects and near-field lubrication hydrodynamics between particles. Here, we show that when coupled with the random fluid flows, FCM can also be extended to Brownian simulations.

2. The fluctuating force-coupling method

In this work, we will be considering a suspension of N rigid spherical particles, each having radius a. Each particle n, $(n = 1,\ldots,N)$, is centred at $\mathbf{Y}_n$ and can be subject to external forces $\mathbf{F}_n$, and external torques $\boldsymbol{\tau}_n$. In the over damped, or Brownian dynamics [12] limit, the equations of motion of these particles is given by

$$\frac{d\mathscr{Y}}{dt} = \mathscr{V} + \tilde{\mathscr{V}} + k_B T \nabla_{\mathscr{Y}} \cdot \mathscr{M}^{\mathscr{V}\mathscr{F}} \qquad (1)$$

where $\mathscr{Y}$ is the $3N \times 1$ vector containing the components of $\mathbf{Y}_n$ for all of the particles, $\mathscr{V}$ holds the components of the deterministic particle velocities, and $\tilde{\mathscr{V}}$ gives the random velocities of the particles due to Brownian motion. The Brownian drift term is given by $k_B T \nabla_{\mathscr{Y}} \cdot \mathscr{M}^{\mathscr{V}\mathscr{F}}$ where k_B is Boltzmann's constant, T is the temperature of the system, and $\mathscr{M}^{\mathscr{V}\mathscr{F}}$ is the translational mobility matrix [21] that relates particle velocities and forces. Brownian drift represents the mean particle velocities established during the inertial relaxation time not resolved in the over damped limit.

With FCM, the deterministic particle velocities are found by first solving the inhomogenous Stokes equations

$$\begin{aligned} -\boldsymbol{\nabla} p + \eta \nabla^2 \mathbf{u} + \mathbf{f}_{FCM} &= \mathbf{0} \\ \boldsymbol{\nabla} \cdot \mathbf{u} &= 0. \end{aligned} \qquad (2)$$

where $\mathbf{f}_{FCM} = \sum_n \mathbf{F}_n \Delta_n(\mathbf{x}) + (1/2)\boldsymbol{\tau}_n \times \boldsymbol{\nabla}\Theta_n(\mathbf{x}) + \mathbf{S}_n \cdot \boldsymbol{\nabla}\Theta_n(\mathbf{x})$ with $\mathbf{F}_n$, $\boldsymbol{\tau}_n$, and $\mathbf{S}_n$ being, respectively, the force, torque, and stresslet associated with particle n and

$$\begin{aligned} \Delta_n(\mathbf{x}) &= (2\pi\sigma_\Delta^2)^{-3/2} \mathrm{e}^{-|\mathbf{x}-\mathbf{Y}_n|^2/2\sigma_\Delta^2} \\ \Theta_n(\mathbf{x}) &= (2\pi\sigma_\Theta^2)^{-3/2} \mathrm{e}^{-|\mathbf{x}-\mathbf{Y}_n|^2/2\sigma_\Theta^2}. \end{aligned} \qquad (3)$$

The length scales σ_Δ and σ_Θ are related to the radius of the particles through $\sigma_\Delta = a/\sqrt{\pi}$ and $\sigma_\Theta = a/\left(6\sqrt{\pi}\right)^{1/3}$. After solving Eq. (2), the velocity, $\mathbf{V}_n$, angular velocity, $\boldsymbol{\Omega}_n$, and local rate-of-strain, $\mathbf{E}_n$, of each particle n are determined from

$$\mathbf{V}_n = \int \mathbf{u}\Delta_n(\mathbf{x}) d^3\mathbf{x} \qquad (4)$$

$$\boldsymbol{\Omega}_n = \frac{1}{2}\int \left[\boldsymbol{\nabla} \times \mathbf{u}\right] \Theta_n(\mathbf{x}) d^3\mathbf{x}. \qquad (5)$$

$$\mathbf{E}_n = \frac{1}{2}\int \left[\boldsymbol{\nabla}\mathbf{u} + (\boldsymbol{\nabla}\mathbf{u})^T\right] \Theta_n(\mathbf{x}) d^3\mathbf{x}. \qquad (6)$$

where the integration is performed over the volume occupied by the fluid. For rigid particles, the stresslets are found by enforcing the constraint $\mathbf{E}_n = \mathbf{0}$ for each n. This is equivalent to stating that the local rates-of-strain can do no work on the fluid [28].

To include the random particle velocities, we consider the flow generated by a white-noise, fluctuating stress, $\mathbf{P}$, in the Stokes equations,

$$\begin{aligned} -\boldsymbol{\nabla} p + \eta \nabla^2 \tilde{\mathbf{u}} &= -\boldsymbol{\nabla} \cdot \mathbf{P} \\ \boldsymbol{\nabla} \cdot \tilde{\mathbf{u}} &= 0. \end{aligned} \qquad (7)$$

As introduced in [27, 16], the statistics for the fluctuating stress, in index notation, are given by $\langle P_{jl} \rangle = 0$ and $\langle P_{jl}(\mathbf{x},t) P_{pq}(\mathbf{x}',t') \rangle = 2k_B T \eta \left(\delta_{jp}\delta_{lq} + \delta_{jq}\delta_{lp}\right) \delta(\mathbf{x}-\mathbf{x}')\delta(t-t')$. We use the brackets $\langle \cdot \rangle$ to denote the ensemble average of a quantity. These statistics for the fluctuating stress yields a velocity field [20] with $\langle \tilde{\mathbf{u}}(\mathbf{x},t) \rangle = \mathbf{0}$ and

$$\left\langle \tilde{\mathbf{u}}(\mathbf{x},t)\tilde{\mathbf{u}}^T(\mathbf{x}',t') \right\rangle = 2k_B T \mathbf{G}(\mathbf{x}-\mathbf{x}')\delta(t-t'). \qquad (8)$$

where $\mathbf{G}(\mathbf{x}-\mathbf{x}')$ is the Stokeslet, the Green's function for the Stokes equations.

In fluctuating FCM, we combine Eqs. (2) and (7) and determine the deterministic and random particle velocities simultaneously by solving

$$\begin{aligned} -\nabla p + \eta \nabla^2 \mathbf{u} &= -\nabla \cdot \mathbf{P} - \mathbf{f}_{FCM} \\ \nabla \cdot \mathbf{u} &= 0. \end{aligned} \quad (9)$$

As with deterministic simulations, the particle velocities, angular velocities, and local rates-of-strain are determined from Eqs. (4), (5), and (6) and, again, the stresslets are determined by enforcing the usual constraint, $\mathbf{E}_n = \mathbf{0}$. Using the statistics of the random fluid flow, Eq. (8), and the expressions for the particle velocities, Eq. (4), one can show analytically [20] that fluctuating FCM yields random particle velocities with the correct statistical properties.

3. Numerical simulations

To demonstrate that fluctuating FCM yields the correct random particle motion, we solve Eq. (9) in a triply periodic domain using a Fourier spectral method to obtain the flow field. Each side of the domain has length $L = 2\pi$ and we use M grid points in each direction, giving a total number of $N_g = M^3$ points. This sets the grid spacing to be $h = 2\pi/M$ and the grid points as $x_\alpha = \alpha h$ for $\alpha = 0, \ldots, M-1$. The statistics for the fluctuating stress need to be modified for the discretised system. As done in other methods that employ fluctuating stresses, see for example [35, 2], at each grid point, the fluctuating stress is an independent Gaussian random variable with $\langle P_{ij}(x_\alpha, x_\beta, x_\gamma) \rangle = 0$ and $\langle P_{ij}(x_\alpha, x_\beta, x_\gamma) P_{pq}(x_\alpha, x_\beta, x_\gamma) \rangle = (2k_B T \eta/(h^3 \Delta t)) (\delta_{ip}\delta_{jq} + \delta_{iq}\delta_{jp})$ where Δt is the timestep. After generating the random stress field and computing the grid values of the regularised multipole expansion, we compute the discrete Fourier transform (DFT) of the total force distribution and determine the fluid velocity field in Fourier space. After taking the inverse DFT, we determine the particle velocities by integrating numerically Eqs. (4 – 6) using the spectrally accurate trapezoidal rule.

We advance the particle positions using Fixman's midpoint scheme [13, 18] as it automatically accounts for the Brownian drift term. With

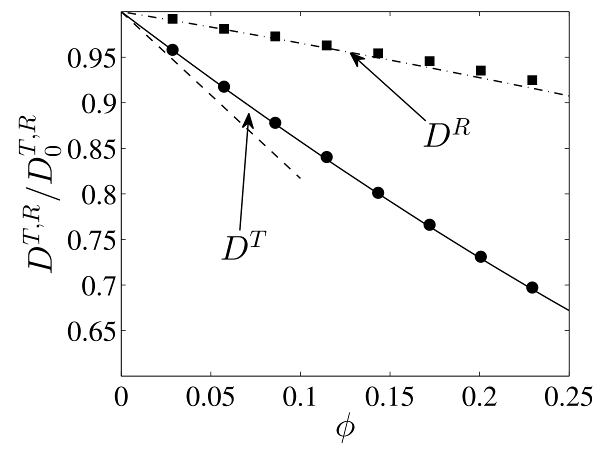


Figure 1: Short-time diffusion coefficient, D. The circular markers indicate the fluctuating FCM results, the dashed line corresponds with $D^T/D_0^T = 1 - 1.83\phi$ from [4] and the solid line show $D^T/D_0^T = 1 - 1.5\phi + 0.75\phi^2$ provided by [3]. The dash-dotted line shows $D^R/D_0^R = 1 - 0.33\phi - 0.16\phi^2$.

this scheme, however, one must utilise the random forces and torques that correspond to the random velocities and angular velocities that fluctuating FCM computes. The random forces and torques can be computed iteratively using a conjugate gradient scheme as described in [20].

3.1 Short-time Diffusion of a Random Suspension

Using fluctuating FCM, we compute the short-time diffusion coefficient

$$D^T = \frac{\Delta t}{6N} \sum_{n=1}^{N} \langle \tilde{\mathbf{V}}_n \cdot \tilde{\mathbf{V}}_n \rangle . \quad (10)$$

for a random suspension of $N = 50 - 400$ particles, corresponding to volume fractions $\phi = 0.0285 - 0.23$. The short-time diffusion coefficient was calculated for low volume fractions by Batchelor [4] who found $D^T/D_0 = 1 - 1.83\phi + O(\phi^2)$ with $D_0 = k_B T/(6\pi a \eta)$. This result has been subsequently confirmed experimentally and theoretically and a summary of these results is presented by Ladd [26]. For each volume fraction, we perform the ensemble average over 10^4 realisations of the fluctuating stress field. For these simulations, there are no forces or torques on the particles, though there are particle stresslets that are induced by the fluctuating stress field.

Fig. 2 shows the results from these simulations using a correction for the periodicity of the domain [26, 3]

$$D^T = D^T_{PER} + \frac{k_B T}{6\pi a \bar{\eta}}(1.7601(\phi/N)^{1/3} - \phi/N) \tag{11}$$

where D^T_{PER} is the short-time self-diffusion coefficient for the periodic domain and $\bar{\eta}$ is the bulk suspension viscosity given by FCM. Fig. 2 also shows results from far-field Stokesian Dynamics calculations [3] and Batchelor's asymptotic result. We see that the values given by fluctuating FCM coincide with those given by far-field Stokesian Dynamics as both provide the same approximation to the mobility matrix. There is a discrepancy, however, with Batchelor's result which includes the near-field lubrication hydrodynamics neglected in both fluctuating FCM and far-field Stokesian Dynamics. In Fig. 2, we also show the short-time rotation self-diffusion coefficient

$$D^R = \frac{\Delta t}{6N}\sum_{n=1}^{N}\left\langle \tilde{\Omega}_n \cdot \tilde{\Omega}_n \right\rangle \tag{12}$$

determined by fluctuating FCM and far-field Stokesian dynamics. As with D^T, we again see correspondence between these two methods with the slight discrepancy attributed to the periodicity of the domain.

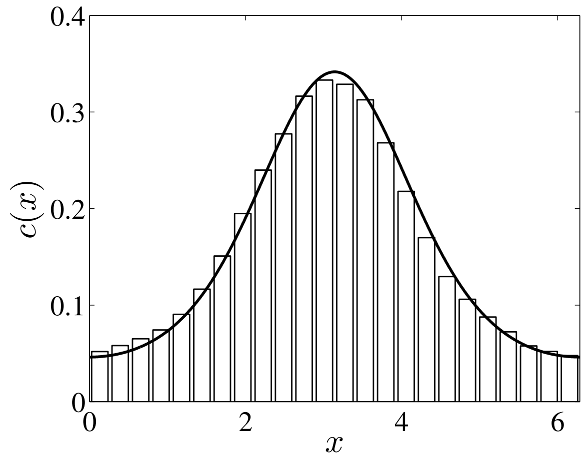
3.2 Concentration profiles in an external potential

In this set of simulations, we consider a suspension of particles subject to the periodic external potential

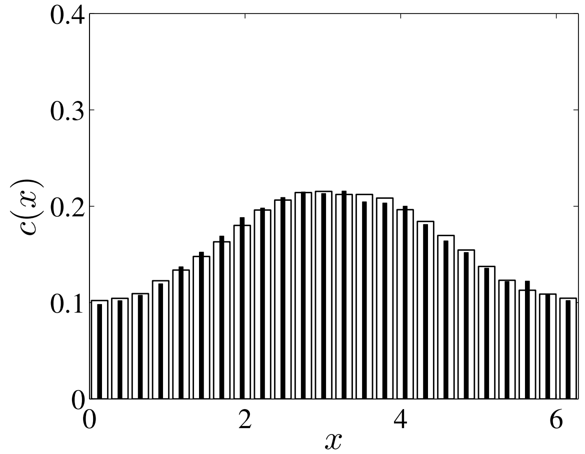
$$\Phi(x) = \Phi_0 \cos x. \tag{13}$$

For non-interacting particles, the equilibrium concentration profile will be given by the Boltzmann distribution

$$c(x) = \frac{1}{Z}\exp\left(-\Phi_0 \cos x / k_B T\right) \tag{14}$$



(a)



(b)

Figure 2: Concentration profiles for a suspension subject to the external potential $\Phi(x) = \Phi_0 \cos x$ (a) The solid line shows the Boltzmann distribution, see Eq. (14), while the bars show the concentration given by stresslet-free fluctuating FCM with no particle interactions. (b) Concentration profiles given by fluctuating FCM simulations with Yukawa interactions. The open bars correspond to stresslet-free simulations, while the closed bars show results from simulations where the particle stresslets are included.

where $Z = \int_0^{2\pi} \exp(-\Phi_0 \cos x / k_B T)\, dx$. We performed simulations with $N = 183$ particles in the external potential. Fig. 2(a) shows the time-averaged concentration from a fluctuating FCM simulation without the stresslets. Here, we recover the Boltzmann concentration profile, Eq. (14) for non-interacting particles. The concentration profile changes when we also include repulsive interactions between the particles. We do this here using the Yukawa potential

$$V(r) = \frac{U_0 \sigma_Y}{r} \exp(-\lambda(r - \sigma_Y)/\sigma_Y) \qquad (15)$$

from DLVO theory [38, 34]. In the simulations, we set $U_0 = k_B T$, $\sigma_Y = 2a$, and $\lambda = 8$. Fig. 2(b) shows the resulting equilibrium concentration when the Yukawa interactions are included. Our simulations with and without the particle stresslets coincide which indicates that the time-integration scheme is recovering the Brownian drift term.

4. Conclusions

We presented a summary of fluctuating FCM, a new approach for the efficient computation of Brownian motion in suspensions of hydrodynamically interacting particles. This technique relies on randomly forcing the fluid, rather than the particles, and using the framework of FCM to recover the motion of the particles. It can be shown that this yields random particle velocities with the correct statistics and consequently, a simulation with the correct diffusivity. To demonstrate the effectiveness of fluctuating FCM, we have presented a numerical implementation of the scheme and compared results with those from previous numerical and analytical studies.

While fluctuating stress based methods such as fluctuating FCM represent a significant advancement in the inclusion of Brownian effects in continuum level simulations, there are still several outstanding challenges in the field. The successful inclusion of near contact hydrodynamics and their effect on the random velocity statistics have yet to be incorporated into these methods. In addition, time-integration schemes that incorporate Brownian drift, but avoid the computation of random forces and torques would aid in increasing the speed of these simulations. Recently, there has been some successful work in this direction [10]. We aim to pursue both of these challenges in the context of the fluctuating FCM, moving to devise a simulation technique appropriate for large scale simulations of dense Brownian suspensions.

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