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Fluctuating force-coupling method for simulations of colloidal suspensions

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ABSTRACT

The resolution of Brownian motion in simulations of micro-particle suspensions can be crucial to reproducing the correct dynamics of individual particles, as well as providing an accurate characterisation of suspension properties. Including these effects in simulations, however, can be computationally intensive due to the configuration dependent random displacements that would need to be determined at every time step. In this paper, we introduce the fluctuating force-coupling method (FCM) to overcome this difficulty, providing a fast approach to simulate colloidal suspensions at large-scale. We show explicitly that by forcing the surrounding fluid with a fluctuating stress and employing the FCM framework to obtain the motion of the particles, one obtains the random particle velocities and angular velocities that satisfy the fluctuation–dissipation theorem. This result holds even when higher-order multipoles, such as stresslets, are included in the FCM approximation. Through several numerical experiments, we confirm our analytical results and demonstrate the effectiveness of fluctuating FCM, showing also how Brownian drift can be resolved by employing the appropriate time integration scheme and conjugate gradient method.

1. Introduction

Brownian motion, or the random movement of particles suspended in liquid [1], results from the many collisions between the particles and the molecules that make up the surrounding fluid. While this is inherently linked to the discrete, molecular nature of the fluid, the effects of Brownian motion extend upwards to longer, continuum length scales, affecting not only the dynamics of individual particles, but also the properties of suspensions themselves. For example, Brownian motion is known to affect the rheological properties of particulate suspensions, changing their linear response to applied stresses, as well as contributing to their non-Newtonian behaviour [2–4]. In biological systems, the diffusion of Brownian particles is a fundamental mechanism of transport, regulating rates of many life processes, especially those in crowded intracellular environments [5]. Characterising and quantifying the role of Brownian motion in these contexts where inter-particle forces, hydrodynamic interactions, and geometric constraints play a strong role presents a current computational challenge. Moreover, with the development of particle self-assembly and aggregation-based fabrication techniques [6,7], as well as the increasing number of highly engineered active, flow-generating and field-responsive micro-particles [8–10], accurately characterising the effects of Brownian motion on suspension dynamics and structure is of fundamental technological importance.

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In simulation techniques such as Brownian dynamics [11] and Stokesian dynamics [12], Brownian motion is incorporated by introducing random particle velocities at each time step. However, in order for either of these methods to yield the correct particle diffusion, the random particle velocities must follow precise statistics, where their correlations are proportional to the hydrodynamic mobility matrix [13,14]. This requires one to compute the square root of the mobility matrix, an $\mathcal{O}(N^3)$ calculation, at every time step. Thus, including the effects of Brownian motion adds significant computational overhead to both of these methods, and as a result, has limited such simulations to two extreme cases – small particle numbers with the hydrodynamic interactions adequately resolved, or larger-scale simulations in which the hydrodynamic interactions are ignored completely. Further, the multiplicative noise, or noise whose amplitude depends on the particle positions, introduced by the hydrodynamic interactions yields also a Brownian drift term [11] that is proportional to the divergence of the mobility matrix. This term also needs to be computed in order to produce the correct particle dynamics.

One approach to overcoming these limitations is to utilise a polynomial expansion of the matrix square root [15]. This method has been used successfully in conjunction with both Brownian and Stokesian dynamics [16,17], allowing for simulations with significantly more particles than would otherwise be possible. Another approach to increase the speed of Brownian simulations, and the one that we will pursue in this work, is to introduce a white-noise, fluctuating stress [18] to drive the surrounding fluid and require that the resulting velocity field satisfy the no-slip condition, or some approximation to it, on the particle surfaces. Indeed, Fox and Uhlenbeck [19] showed for rigid particles that this approach does yield the correct particle velocity correlations and, consequently, the correct diffusion matrix for the suspension. Since the fluctuating stress itself is *independent* of the particle configuration, the $\mathcal{O}(N^3)$ matrix square root computation is not required. While this approach does require one to solve for the random fluid flow, such fluid flow computations are typically already performed to find the deterministic motion of the particles. The effectiveness of fluctuating stresses in resolving Brownian motion has been demonstrated in a variety of simulation techniques. They have been successfully employed in large-scale Lattice-Boltzmann simulations of particulate suspensions [20–23], as well as more traditional, continuum mechanics based simulations of Brownian particles and structures. Fluctuating stresses have been used with the distributed Lagrange multiplier (DLM) method [24] where the induced fluctuating flow is constrained at the grid points within the solid particle. Recently, they have been successfully employed with immersed-boundary [25–27] and "blob" methods [28], resolving 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 [29,1].

Based on the success of these approaches, we utilise fluctuating stresses, the fluid flows they produce, and the simulation technique known as the force-coupling method (FCM) to develop a fast method for large-scale simulations of suspensions of interacting particles. FCM [30–33] employs regularised multipole expansions of the force distributions the particles exerts on the surrounding fluid and spatial averaging of the resulting flow to obtain the particle motion. It has been shown to be very effective for large-scale simulations of suspensions and particle-laden flows [34–37] over a wide range of volume fractions. Here, we show analytically that when the surrounding fluid is also forced by a fluctuating stress, FCM yields random particle velocity and angular velocity correlations consistent with the fluctuation–dissipation theorem [1]. A main result of this work is that fluctuating FCM gives the proper correlations even when higher-order multipoles, such as the rotlet and stresslet, are included in the multipole expansion. We provide numerical examples confirming these results. In addition, for dynamic fluctuating FCM simulations, we show how to recover Brownian drift using Fixman's midpoint time integration scheme [38,39] and the conjugate gradient method. We employ this scheme to examine long-time diffusion of interacting particles and suspension dynamics in cellular flow fields.

2. Particle motion

In this study, 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 $\boldsymbol{\tau}_n$. We will be considering the motion of these particles in the over-damped, or Brownian dynamics [11], limit where the Reynolds number [40,41] is low, and fluid and particle inertia are neglected. While working in this limit does not resolve the power-law decay of the velocity autocorrelation function, it provides an accurate description of diffusive motion for times $t \gg \rho a^2/\eta$ (ρ is the density of the fluid and η the shear viscosity) [1], making it appropriate for describing the dynamics of suspensions of micron-scale, colloidal particles. In this limit, the equations of motion can be written as

$$\frac{d\mathcal{Y}}{dt} = \mathcal{V} + \tilde{\mathcal{V}} + k_B T \boldsymbol{\nabla}_{\mathcal{Y}} \cdot \mathcal{M}^{\mathcal{VF}} \tag{1}$$

where $\mathcal{Y}$ is the $3N \times 1$ vector containing the components of $\mathbf{Y}_n$ for all of the particles, $\mathcal{V}$ holds the components of the deterministic particle velocities, and $\tilde{\mathcal{V}}$ gives the random velocities of the particles due to Brownian motion. The Brownian drift term is given by $k_B T \boldsymbol{\nabla}_{\mathcal{Y}} \cdot \mathcal{M}^{\mathcal{VF}}$ where k_B is Boltzmann's constant, T is the temperature of the system, and $\mathcal{M}^{\mathcal{VF}}$ is the translational mobility matrix as described below.

The deterministic velocities, $\mathcal{V}$, as well as the particle angular velocities, $\mathcal{W}$, are given by

$$\begin{bmatrix} \mathcal{V} \\ \mathcal{W} \end{bmatrix} = \begin{bmatrix} \mathcal{M}^{\mathcal{VF}} & \mathcal{M}^{\mathcal{VT}} \\ \mathcal{M}^{\mathcal{WF}} & \mathcal{M}^{\mathcal{WT}} \end{bmatrix} = \mathcal{M} \begin{bmatrix} \mathcal{F} \\ \mathcal{T} \end{bmatrix} \tag{2}$$