# Direct Numerical Simulation of Moving Charged, Flexible Bodies with Thermal Fluctuations

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## ABSTRACT

The interaction of sub-micron scale objects with fluids is an important problem encountered in miniaturized systems. Various physical phenomena should be modeled simultaneously for a fundamental investigation of these systems. Our objective is to develop a direct numerical simulation tool to better understand the motion of sub-micron objects in complex geometries.

We report: (a) A novel methodology to simulate the Brownian motion of objects in fluids, (b) A technique to simulate the motion of electrically charged bodies with thin Debye layers & (c) An innovative approach to simulate the motion of flexible bodies. The goal is to develop numerical techniques for each of these problems such that they can be unified or interfaced to allow simultaneous description of complex physical phenomena. Preliminary work has been accomplished.

*Keywords*: Numerical simulation, Brownian motion, hydrodynamic fluctuations, electrophoresis, flexible macromolecules.

## 1 INTRODUCTION

Some of the key issues, commonly encountered in sub-micron/nanoscale devices, that require further investigation are considered here. A brief introduction to the various methods we are pursuing will be given in this paper.

We consider numerical schemes within the realm of the continuum hypothesis. The key issues that require further investigation, and considered here, are the modeling of thermal fluctuations, the motion of flexible bodies and the motion of electrically charged bodies. Numerical techniques for each of these problems are to be developed such that they can be unified or interfaced to allow simultaneous description of complex physical phenomena.

In section 2 we will discuss the simulation method for Brownian motion. Electrophoresis will be considered in section 3 and the motion of flexible bodies in section 4.

## 2 BROWNIAN MOTION

The interaction of sub-micron/nanoscale objects (such as macromolecules or small particles or small devices) with fluids is an important problem in small scale devices. A better understanding of fluid dynamics is critical in e.g. bio-molecular transport, manipulating & controlling chemical & biological processes using small particles etc. These objects could be moving in an environment with varying temperatures and fluid properties (e.g. viscosity, density). Thermal fluctuations can influence the motion of such objects.

Direct Numerical Simulations (DNS) of particle motion in fluids is a tool that has been developed over the past ten years (see [1] & [7] and references therein). In this approach the fluid equations are solved coupled with the equations of motion of the particles. DNS allows investigation of a wide variety of problems including particles in Newtonian or viscoelastic fluids with constant or varying properties. DNS can be an excellent tool to investigate the motion of sub-micron particles in varying fluid environments. Currently, there is no convenient way to incorporate the effect of thermal fluctuations, important to simulate the motion of sub-micron particles or objects, in the governing equations. Here, we propose a new approach to overcome this shortcoming.

The conventional approach to perform Brownian dynamic (BD) simulations is based on the algorithm by Ermak and McCammon [2]. It is an algorithm for simulating the Brownian motion of $N$ particles with the inclusion of hydrodynamic interactions. For timescales larger than the momentum relaxation time of the particles or for inertia-less particles the algorithm updates the particle positions according to

$$x_i^{n+1} = x_i^n + \Delta t \sum_j \frac{\partial R_{ij}}{\partial x_j}\bigg|_{x^n}$$
$$+ \left(\frac{\Delta t}{k_B T}\right)\sum_j R_{ij}\left(x^n\right)F_j^n + X_i^n\left(\Delta t\right) \qquad (1)$$

where $x^n$ is the collective position vector of all the $N$ particles at the $n^{th}$ timestep, $\Delta t$ is the timestep, $k_B$ is the Boltzmann constant, $T$ is the temperature and $F$ is the collective vector of non-hydrodynamic forces on the particles. $\mathbf{R}(x^n)$ is the configuration dependent diffusion tensor where $R_{ij}$ is its component. $X_i$ is the random displacement vector that mimics thermal interactions with the suspending fluid. It is a Gaussian white noise process related to $\mathbf{R}$ by

$$\langle \mathbf{X}^n \rangle = \mathbf{0}, \langle \mathbf{X}^n (\mathbf{X}^m)^T \rangle = 2\Delta t \delta_{mn} \mathbf{R}(\mathbf{x}^n), \qquad (2)$$

where $\delta_{mn}$ is the Kronecker delta and angle brackets represent an average. The diffusion tensor $\mathbf{R}$ defines the hydrodynamic interactions.

Obtaining the random displacement turns out to expensive. Using the conventional Brownian dynamic simulation methodology to objects with irregular shapes and to cases where the fluid exhibits varying properties and non-linearities is not straightforward. This is mainly because of the presence of the diffusion tensor in the expression for the random term. For the same reason, it is difficult to use this approach to include thermal motion in DNS schemes. A different approach is preferable and is discussed below.

A particle suspended in a fluid experiences a hydrodynamic force due to the average motion of the fluid around it. The average motion of the fluid is represented by the usual continuum equations such as the Navier-Stokes equations. Small particles in fluids, in addition to the average force, experience a random force due to the thermal fluctuations in the fluid. In BD simulations the principle is to model this thermal force from the fluid in terms of a random term in the particle equation.

A different approach is to model the thermal fluctuations in the fluid itself via random stress and heat flux terms in its governing equations. A general theory of fluctuations in fluid dynamics is given in by Landau & Lifshitz [8]. The general formulas of fluctuation theory are used to obtain equations of motion for the fluctuating fluid. Given below is the momentum equation for a fluid with hydrodynamic fluctuations (valid for non-quantized fluctuations i.e. $\hbar\omega << k_B T$, where $\omega$ are the frequencies in the fluctuations) at equilibrium, in the creeping flow limit.

$$-\nabla p + \nabla \cdot \boldsymbol{\sigma} = \mathbf{0},$$
$$\boldsymbol{\sigma} = \eta(\nabla \mathbf{u} + \nabla \mathbf{u}^T) + \mathbf{s},$$
$$\langle \mathbf{s} \rangle = \mathbf{0},$$
$$\langle s_{ik}(\mathbf{x}_1, t_1) s_{lm}(\mathbf{x}_2, t_2) \rangle =$$
$$2k_B T(\eta[\delta_{il}\delta_{km} + \delta_{im}\delta_{kl}])\delta(\mathbf{x}_1 - \mathbf{x}_2)\delta(t_1 - t_2)$$
$$(3)$$

where $p$ is the pressure, $\boldsymbol{\sigma}$ is the total stress at a point in the fluid, $\mathbf{s}$ is the stress due to fluctuations, $\mathbf{x}_1$ & $\mathbf{x}_2$ are the position vectors of points in the fluid domain, $t_1$ & $t_2$ are two time instants and $\delta(t_1-t_2)$ is the Dirac delta function. An incompressible fluid is considered. More general governing equations for non-equilibrium, non-linear cases are available in literature (see [3] and references therein). It can be shown that the fluctuating hydrodynamic equations give rise to a fluctuating force on a particle [4]. The solution of

fluid equations coupled with the equations of motion of the particles leads to the Brownian motion of the particles.

We have used a moving mesh, Galerkin finite-element, technique to solve the coupled fluid-particle equations of motion [1]. This technique uses an unstructured body-fitted mesh and a mesh update scheme to handle the time-dependent fluid domain.

We added the fluctuating stress term in the fluid equations. This poses no fundamental difficulty especially because the covariance structure of the fluctuation stress is locally defined. The random stress can then be obtained from uncorrelated random numbers chosen from a Gaussian distribution (note that this simplicity does not exist in Equation 2 above).
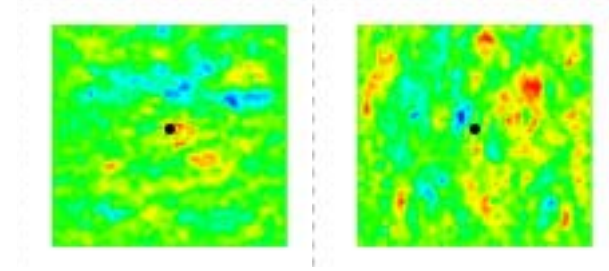


Figure 1: Contour plots of the horizontal and vertical velocity fluctuations in the fluid. The dark dot at the center is the particle. In this case we have a time-step of 100 ps for a particle in water at $20^o$ C.

A two-dimensional code for the direct simulation of the motion of particles in fluctuating fluids was developed. Thermal motion of a particle in a stationary fluid was considered (Figure 1). We assumed Stokes flow and the particle inertia was neglected. The particle translational and angular momentum equations, respectively, are

$$\oint \boldsymbol{\sigma} \cdot \mathbf{n} d\Gamma = \mathbf{0}, \quad \oint \mathbf{r} \times (\boldsymbol{\sigma} \cdot \mathbf{n}) d\Gamma = \mathbf{0}, \qquad (4)$$

where the integrals are over the particle surface, $\mathbf{n}$ is the unit outward normal and $\mathbf{r}$ is the radius vector at a point on the particle surface. The solution of Equation 4 coupled with Equation 3 and the incompressibility constraint, along with the no-slip boundary condition on the particle surface, gives a random velocity of the particle. These are quasi-steady simulations since the inertia terms are absent. The random velocity thus obtained is a measure of the random particle displacement, which is calculated by multiplying this velocity by the time-step. The particle positions are then explicitly updated. This approach is direct and can be applied to arbitrarily shaped devices or objects without any basic difficulty.

Rigorous testing of this approach is under way where we are comparing the results with known analytical values. Our subsequent steps will involve a more detailed investigation of appropriate boundary conditions for fluctuating hydrodynamics and the extension of this

approach to cases of particles in non-stationary fluids and in fluids with varying properties and constitutive forms.

# 3   ELECTROPHORESIS

In this section we motivate and briefly describe the simulation of the motion charged particles, with thin Debye layers, under the action of an external electric field.

An insulating charged particle, of any shape, freely suspended in an infinite viscous strong electrolyte of constant dielectric permittivity $\varepsilon$ and viscosity $\eta$, moves under the action of an external electric field. More importantly, the electric field and the velocity field are similar and differ only in magnitude [5]. The assumptions of a thin Debye layer and constant material properties are critical for this similarity because that leads to the following boundary condition at each point on the particle surface

$$u_{slip,t} = - \frac{\varepsilon \zeta E_t}{\eta} , \qquad (5)$$

where $u_{slip,t}$ is the tangential component of the slip between the fluid and the particle velocities at a point on the particle surface, $E_t$ is the tangential component of the electric field at that point and $\zeta$ is the 'zeta potential' on the particle surface. The zeta potential arises due to the charge density on the particle surface. The slip in the normal direction is zero and so is the normal component of the electric field on the surface of an insulating particle. If the zeta potential is constant then the velocity and electric fields differ only by a factor $\varepsilon \zeta / \eta$. Consequently, we obtain a potential flow even in the Stokes limit. This is due to the fact that the potential flow satisfies the Stokes equations with appropriate slip boundary conditions.

An immediate implication is that the disturbance due to a sphere of radius $a$ decays as $(a/r)^3$, where $r$ is the radial distance from the sphere, instead of the usual $a/r$ for the Stokes flow. The similarity may be extended to large numbers of particles at high concentrations if the material properties are constant and if all the particles have the same constant zeta potential.

The above similitude breaks down if the material properties are not constant or if the zeta potentials on all the surfaces are not same or constant. Non-constant zeta potentials are common in many applications in biotechnology and even in chemical engineering (colloidal science). The breakdown of similarity implies that there will be long range viscous interactions which can lead to important deviations from the similitude-based solutions. Better understanding of this phenomenon is critical for 'microfluidic devices' where the thin Debye layer assumption is often reasonable. The current understanding is largely restricted to single particle studies. A systematic investigation that also accounts for multiparticle interactions is yet to be accomplished.

We intend to study this problem using direct numerical simulation techniques for solid-liquid flows. We have developed a two-dimensional scheme for few particles in the Stokes limit (details of this method will appear elsewhere [6]). These are quasi-steady simulations where the fluid-particle motion is coupled to the applied electric field through the slip boundary condition on the particle surface. For a given particle configuration we first solve the Laplace equation for the electric potential with an insulating boundary condition on the particle surfaces. Then we solve the coupled fluid-particle equations of motion [1]. Instead of the usual no-slip boundary condition on the particle surface we use the slip boundary condition given in Equation 5 to model the effect of particle charge. The material properties and zeta potential need not be constant. The slip surface around each particle encloses a neutral body. Hence there is no net force or torque in the Stokes limit. The particle equations are therefore given by Equation 4. The fluid and particle velocities are obtained from the coupled solution procedure.
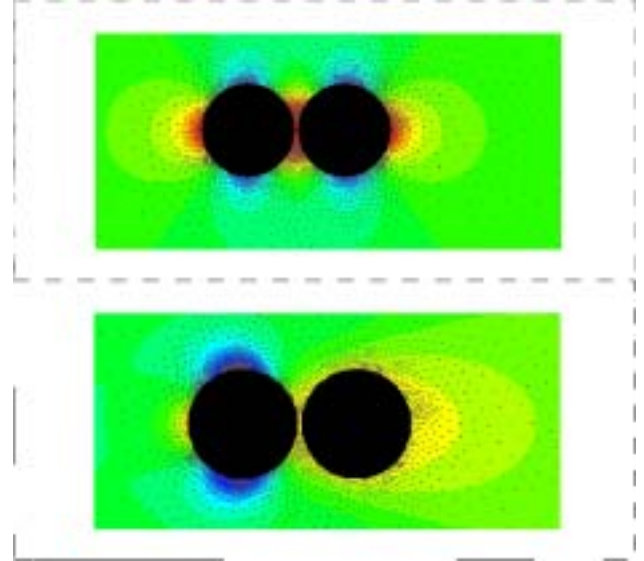


Figure 2: Numerical simulation of electrophoresis of two particles. Color contour plot of the horizontal velocity is shown for two cases: (a) Top figure: The two particles have the same zeta-potential. (b) Bottom figure: The two particles have different zeta potentials.

Figure 2 shows color contour plots of horizontal velocity in the fluid for the electrophoresis of two particles [6]. The external electric field is in the horizontal direction. In the top figure, the two particles have the same zeta potential. As expected the resultant velocity field of the fluid is similar to the electric field. The velocity disturbance decays fast and is short range. In the bottom figure we have two particles with different zeta potentials. The similitude solution is no longer valid. The Stokes flow interaction between the two particles lead to long range velocity disturbances in the fluid as evidenced by the velocity contour plot.

## 4 MOTION OF FLEXIBLE BODIES

Long macromolecules are flexible and may be modeled as continuum elastic rods. Coupled simulation of the motion of an elastic body in a fluid is not straightforward. Here, we discuss a formulation that could lead to the development robust schemes for such simulations.

Consider a linear elastic particle freely suspended in a fluid. For simplicity we assume that the inertia effects are negligible. The equation for the displacement field is given by

$$\nabla \cdot (2\sigma \mathbf{D}[\mathbf{d}] + \kappa(\nabla \cdot \mathbf{d})\mathbf{I}) = \mathbf{0} \quad \text{in P,}$$
$$(2\sigma \mathbf{D}[\mathbf{d}] + \kappa(\nabla \cdot \mathbf{d})\mathbf{I}) \cdot \mathbf{n} = \mathbf{t} \text{ on } \partial \text{P,}$$
$$(6)$$

where $\sigma$ and $\kappa$ are material parameters of the elastic particle, $\mathbf{d}$ is the displacement field, $\mathbf{D}[\mathbf{d}]$ denotes the symmetric part of the gradient of $\mathbf{d}$, $\mathbf{t}$ is the traction vector on the particle surface $\partial \text{P}$ and P is the particle domain. The net force and torque on the body is zero in the Stokes limit. Solution of Equation 6 yields the displacement field of the body.

N. Patankar *et al.* [7] recently proposed a formulation for the DNS of rigid particles that is relevant to the above problem. Their idea is to consider the rigid particle as a fluid and then to constrain this fluid to move rigidly by setting the deformation-rate tensor, $\mathbf{D}[\mathbf{u}]$, equal to zero. This constraint can be given as

$$\nabla \cdot (2\sigma \mathbf{D}[\mathbf{u}] + \kappa(\nabla \cdot \mathbf{u})\mathbf{I}) = \mathbf{0} \quad \text{in P,}$$
$$(2\sigma \mathbf{D}[\mathbf{u}] + \kappa(\nabla \cdot \mathbf{u})\mathbf{I}) \cdot \mathbf{n} = \mathbf{0} \quad \text{on } \partial \text{P,}$$
$$(7)$$

where $\mathbf{u}$ is the velocity field in the particle domain. Equation 7 is the equation of constraint that sets $\mathbf{D}[\mathbf{u}] = \mathbf{0}$ in the particle domain. Note that we have chosen a rigidity constraint using the material parameters for the elastic body in Equation 6. It can be shown that the rigidity constraint leads to a Lagrange multiplier $\boldsymbol{\lambda}$, which is a vector field [7]. Consequently, the momentum equation of the particle modeled as a fluid and the boundary condition on its surface become [7]

$$\nabla \cdot (2\sigma \mathbf{D}[\ddot{\mathbf{e}}] + \kappa(\nabla \cdot \ddot{\mathbf{e}})\mathbf{I}) = \mathbf{0} \quad \text{in P,}$$
$$(2\sigma \mathbf{D}[\ddot{\mathbf{e}}] + \kappa(\nabla \cdot \ddot{\mathbf{e}})\mathbf{I}) \cdot \mathbf{n} = \mathbf{t} \text{ on } \partial \text{P,}$$
$$(8)$$

where we have neglected inertia. The physical significance of $\boldsymbol{\lambda}$ is evident from the difference between Equations 6 and 8. It is clear from the comparison that $\boldsymbol{\lambda}$ is the displacement field of the elastic body whose parameters were used in the rigidity constraint (Equation 7).

The key conclusion of the above analysis is that the rigidity constraint can be used to obtain the displacement field of an elastic body. This result can be used to perform 'quasi-rigid' simulations of elastic bodies. In such an approach we would solve the fluid-particle equations by assuming the elastic particles to be rigid. The coupled solution would not only give the particle velocity (which represents its rigid motion) but also the displacement field of the elastic body [7]. The position and the shape of the particle would then be updated using this solution. This approach is applicable when the elastic deformation is small compared to its rigid motion (this requirement is mainly related to the no-slip boundary condition on the fluid-particle interface). We are investigating how these ideas could be extended to simulate the motion of objects with large deformations and to non-linear elastic bodies.

## 5 CONCLUSION(6)

In this paper we have briefly discussed the numerical techniques we are developing for the Brownian motion of bodies in fluids, electrophoresis and the motion of flexible bodies in fluids. Although each of these techniques is being independently explored they can ultimately be combined or interfaced, without any fundamental difficulty, to simulate complex multi-physics problems.

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