Numerical Simulation of Brownian Motion of Multiple Particles Via a Fluctuating Lattice Boltzmann Model

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Abstract. In this work the fluctuating lattice Boltzmann model was adopted to simulate the Brownian motion of multiple particles in two dimensions. The thermal equilibrium between the particles and the fluctuating fluid was evaluated. More importantly, both the translational and rotational velocity correlation functions (VCFs) were studied for different number of particles. It has been shown that the translational and rotational VCFs decay at $t^{-1}$ and $t^{-2}$ at long times, respectively, which the long time tails of Brownian particles are moving randomly in a fluid.

Introduction

Particles suspended in fluids experience a random force due to the thermal fluctuations in the fluid around them in addition to the average hydrodynamic force. Brownian motion may take place for those sub-micron/nanoscale particles. For many applications in microsystems, the ability to control and measure temperature inside microfluidic devices is critical since temperature often affects biological or chemical processes. Recent developments \cite{1, 2} demonstrate that the well-defined temperature dependence of the Brownian motion of nanoparticles could be used to present a temperature measurement technique which offers several benefits over existing methodologies. Brownian particle can be adopted to measure the local viscoelastic response of soft materials \cite{3} or the topography of a surrounding polymer network \cite{4}. The motion of a Brownian probe can also be used to characterize mechanical properties of molecular motors by analyzing the particle's trajectory \cite{5}. Moreover, the biased Brownian motions or rectified Brownian motions, induced by an energy source \cite{6} or by broken spatial reflection symmetry \cite{7}, provide a very effective technique for particle separation. Furthermore, it has been demonstrated \cite{8, 9} that nanoparticles in a conventional base fluid, known as nanofluids, tremendously enhance the heat transfer characteristics of the original fluid. At the same time, study \cite{10} has declared that Brownian motion is a key mechanism governing the thermal behavior of nanofluids. Due to its importance in engineering applications, there has always been a great deal of interest in developing algorithms that can provide a better understanding of particle's Brownian motion.

The most important and accurate approach to simulate particle Brownian motion may be the fluctuating hydrodynamics, which was proposed by Landau and Lifshitz \cite{11}. In this approach, the thermal fluctuations in the fluid, which result in the Brownian motion of particles, are modeled by adding a random stress tensor to Navier – Stokes equations. Solving the fluctuating hydrodynamic equations coupled with the particle equations of motion result in the Brownian motion of particles. As a direct numerical simulation
scheme, there is no need to add a random force term in the particles’ equations since random fluctuations are applied directly to the particles.

The effect of hydrodynamic interactions among particles is very important to the Brownian motion of multiple particles. As shown by Ladd [12], the diffusion coefficient of a sphere undergoing Brownian motion decreases as the volume fraction increases, which is suggesting that the motion of one particle may be suppressed by the particles around it. However, the research work on this subject is very limited. It is still unclear how the hydrodynamic interactions affect the Brownian motion when many particles are randomly moving in the fluid, because little attention has been paid in the past. Therefore, in this work the fluctuating dynamics was adopted to simulate the motion of multiple Brownian particles in two dimensions. This work aims to study the characteristics of Brownian motion of multiple particles and to present a better understanding of how the hydrodynamic interactions affect the Brownian motion.

**Numerical Model**

The fluid flow is solved by the LB method. The discrete LB equations of a single-relaxation-time model are described as [13],

\[ f_i(x, t + \Delta t) = f_i(x, t) - \frac{\tau}{2} \left[ f_i^{(0)}(x, t) - f_i(x, t) \right] + f_i'(x, t) \]  

(1)

where \( f_i(x, t) \) is the distribution function on the \( i \)-direction microscopic velocity \( e_i \), \( f_i^{(0)}(x, t) \) is the equilibrium distribution function, \( \Delta t \) is the time step of the simulation, \( f_i'(x, t) \) is a stochastic term representing the thermal fluctuations, which is related to the fluctuating stress in the Navier-Stokes equation [12]. The fluid density \( \rho \) and velocity \( u \) are determined by the distribution function,

\[ \rho = \sum_i f_i, \quad \rho \cdot u = \sum_i f_i e_i \]  

(2)

For the two-dimensional D2Q9 lattice model used here, the discrete velocity vectors are,

\[ e_i = \begin{cases} (0,0), & \text{for } i = 0, \\ (\pm 1,0)c, & \text{for } i = 1 \text{ to } 4, \\ (\pm 1,\pm 1)c, & \text{for } i = 5 \text{ to } 8, \end{cases} \]  

(3)

where \( c = \Delta x / \Delta t, \Delta x \) is the lattice spacing. The equilibrium distribution function is chosen as,

\[ f_i^{(0)}(x, t) = \omega_i \rho \left[ 1 + \frac{3e_i \cdot u}{c^2} + \frac{9(e_i \cdot u)^2}{2c^4} - \frac{3u^2}{2c^2} \right] \]  

(4)

As shown by Nie and Lin [13], the stochastic term is related to the fluctuating stress,

\[ \sigma_{\alpha\beta}' = -\tau \sum_i f_i' e_{\alpha\beta} \]  

(5)

According to the fluctuation-dissipation theorem, \( \sigma_{\alpha\beta}' \) has the following property [11],

\[ \left\langle \sigma_{\alpha\beta}' \right\rangle = 0 \]

\[ \left\langle \sigma_{\alpha\beta}'(x_1, t_1) \sigma_{\gamma\delta}''(x_2, t_2) \right\rangle = 2k_B T \mu \left( \delta_{\alpha\gamma} \delta_{\beta\delta} + \delta_{\alpha\delta} \delta_{\beta\gamma} - \frac{2}{3} \delta_{\alpha\beta} \delta_{\gamma\delta} \right) \delta(x_1 - x_2) \delta(t_1 - t_2) \]  

(6)
where <> denotes averaging over an ensemble, $k_B$ is the Boltzmann constant, $T$ is temperature of the fluid, $\mu$ is the dynamic viscosity of the fluid. The fluctuating stress is sampled from a Gaussian distribution with zero mean and a given variance of $2k_BT\mu$.

In this work we assume the stochastic term $f'_i(x, t)$ to be the following form to make sure of the conservation of mass and momentum [13],

\[
\begin{align*}
    f'_0 &= 0 \\
    f'_i &= f'_j = \frac{1}{2\tau}\sigma'_{yy} \\
    f'_i &= f'_j = \frac{1}{2\tau}\sigma'_{xx} \\
    f'_i &= f'_j = -\frac{1}{4\tau}(\sigma'_{xx} + \sigma'_{yy} + \sigma'_{ij}) \\
    f'_i &= f'_j = -\frac{1}{4\tau}(\sigma'_{xx} + \sigma'_{yy} - \sigma'_{ij})
\end{align*}
\]

**Numerical Results**

In this work, the Brownian motion of multiple particles in a periodic domain was numerically investigated. In the simulations only the hydrodynamic force was considered. The periodic domain is set to be $256 \times 256$. The density of the fluid is fixed at $\rho = 1$ and the non-dimensional relaxation time $\tau = 0.8$, which leads to the viscosity of the fluid $\nu = (2\tau - 1)/6 = 0.1$. The radius of particle is $a = 3.5$. The solid/fluid density ratio is fixed at $\rho_s/\rho = 11$. In order to determine the magnitude of the fluid fluctuation, the variance of fluctuating stress is chosen as $2k_BT\mu = 10^{-4}$. It should be stated that all the above-mentioned parameters are in lattice unit.

For $N = 16$, the instantaneous velocity field of fluid at different times is shown in Fig. 1. The Brownian particles are also shown in the figure. As one can see, the velocity vectors are disorderly and disorganized, which represents the random molecular motion of the fluid. This is the origin of the Brownian motion of particles in a fluid, resulting from the essence of the present fluctuating lattice Boltzmann method. As one can see in Fig. 1, the particles move randomly due to the fluid molecular collision. This suggests that it is impossible to predict the motion of particle, such as the velocity or the trajectory of particle. In addition, the rotation of particle is also attained in the simulation, which is different from the Langevin dynamics.

![Figure 1. Instantaneous flow of Brownian motion for $N=16$ at different times: (a) $t=1000$, (b) $t=20000$, (c) $t=50000$ and (d) $t=100000$. The white arrow on each particle is used to visually track its rotation with horizontal orientation at the beginning (the same as below).](image)

The instantaneous flow of Brownian motion at different times for $N = 100$ is shown in Fig. 2, which is the largest number of particles in this work. As shown in the figure, the particles are moving randomly as soon as they are released. Due to the
hydrodynamic interactions among particles, the motion of each particle not only depends on the fluid, but also the particles around it, because it can “feel” the other particles in the fluid. All the particles tend to spread out as they undergo Brownian motion due to the fluctuating fluid. Furthermore, because of the symmetry of flow conditions, the particles spread out homogeneously, displaying the classical motion of Brownian diffusion.

Figure 2. Instantaneous flow of Brownian motion for $N=100$ at different times: (a) $t=1000$, (b) $t=50000$, (c) $t=100000$ and (d) $t=200000$.

Figure 3. Translational/rotational velocity correlation functions for Brownian motion: (a) $N=16$, (b) $N=100$.

Fig. 3 shows the translational and rotational velocity correlation function (VCFs) for $N = 16$ and $N = 100$, respectively. All the results are normalized by their initial values, i.e. the values at $t = 0$. As demonstrated by Hauge and Martin-Löf [14], the translational and rotational VCFs of a sphere undergoing Brownian motion have power-law decays over long times that are $t^{-3/2}$ and $t^{-5/2}$, respectively. The long time tails are fundamental to help understanding the physics of Brownian motion. As shown in Fig. 3, the similar long time tails are observed for a circular particle, irrespective of the number of particles. The translation VCF decays at $t^{-1}$ at long times, while the rotational VCF decays at $t^{-2}$ at long times, which is different from the exponential decay predicted by Langevin dynamics.

It has been shown that thermal equilibrium between the Brownian particle and the surrounding fluid molecular will reach and that an equi-partition of energy for each degree of freedom will be observed, which can be described as,

$$\langle U^2 \rangle = \langle V^2 \rangle = k_B T / M \; ; \; \langle \Omega^2 \rangle = k_B T / J$$

(8)

where $U$ and $V$ refer to the translational velocity of $x$ and $y$ coordinate, respectively, and $\Omega$ refers to the rotational velocity of particle. $M$ and $J$ are the mass and inertial of the particle, respectively. An equi-partition of energy for $x$ and $y$ translational motion can
be observed, as well as the rotational motion, as displayed in Fig. 4, which shows the thermal equilibrium of \( N=16 \) and \( N=100 \), respectively. The temperatures characterizing translational motion of \( x \) and \( y \) coordinate, and the rotational motion agree with each other after a certain time, but all about 5% less than the effective temperature of the fluid, as one can see in Fig. 4. The similar results were also obtained by Ladd [13], which are however 15% less than the effective temperature of the fluid.

![Figure 4](image1.png)

Figure 4. Time history of translational/rotational velocity for Brownian motion: (a) \( N=16 \), (b) \( N=100 \).

Diffusion coefficient, also called diffusivity, is an important parameter indicative of the diffusion mobility, which is generally prescribed for a given pair of species. The higher the diffusivity (of one substance with respect to another), the faster they diffuse into each other. Fig. 5 shows the normalized translational and rotational diffusion coefficient depending on the volume fraction. As one can see, the diffusion coefficients are decreasing as volume fraction is increasing. The reason is that the hydrodynamic interaction among particles suppresses the diffusion of Brownian particle. The higher the volume fraction is, the more significantly the Brownian diffusion is suppressed. In addition, the translational diffusion coefficient is larger than that of rotational one, as shown in Fig. 5.

![Figure 5](image2.png)

Figure 5. Translational/rotational diffusion coefficient for different volume fraction.

**Conclusion**

In this work the previously developed fluctuating lattice Boltzmann method was adopted to simulate the Brownian motion of multiple circular particles in a fluid. It has been shown that the translational velocity correlation function has a long time tail, which decays at \( t^{-1} \) at long times. Similarly, the rotational velocity correction function decays at \( t^{-2} \) at long times. Furthermore, the thermal equilibrium between the Brownian particles and the fluid is also demonstrated in the present study. Results show that the difference between the particle temperature and the fluid temperature is within 5% irrespective of the number of particles.
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References