Mapping of dissipative particle dynamics in fluctuating hydrodynamics simulations

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Dissipative particle dynamics (DPD) is a method developed primarily for the simulation of complex fluids at mesoscales.[1,2] While it is often thought that DPD beads represent packets of atoms collectively moving, the statistical mechanical foundation of such a view remains obscure.[3] Associated with the ambiguity of the exact nature of DPD beads is the nonuniqueness of mapping DPD models to physical systems. In particular, the mapping of time is nonunique and is problem dependent, e.g., mapping of DPD time to physical time has been based on bead diffusion rate, bead thermal velocity, or externally imposed time scales,[4–6] to name just a few.

Our interest in DPD originates from the need to study transport phenomena in particulate suspension where thermal fluctuations may play a critical role.[7,8] Such transport can be described by the fluctuating hydrodynamics theories.[9] However, solving the hydrodynamics equations in particulate suspension that rigorously satisfy the fluctuation-dissipation theorem (FDT) is challenging.[10] Particle methods such as the stochastic rotation dynamics method have been used for such simulations.[11] DPD is also well suited for studying such phenomena as momentum/energy conservation, and FDT are guaranteed by the way DPD models are designed and colloidal particles can be modeled easily.[1,2,12] Two outstanding issues in applying DPD to fluctuating hydrodynamics simulations are as follows: (1) to devise a scheme to map between DPD models to physical systems in such simulations and (2) to establish that DPD model can model fluctuating hydrodynamics with good accuracy. An explicit demonstration of (2) is desirable because while such capability is *ideally* expected, it may not be achievable, *in practice*, due to various artifacts arises from the coarse graining.

To address the first issue, we propose to abandon the idea that DPD fluid beads are "clumps" of real fluid atoms but view them *together* as a "media" that provides an arena for the transport of momentum, energy, and particulates that satisfies the fluctuating hydrodynamics laws. This idea is inspired by a recent paper on coarse graining in colloidal suspensions.[3] To bridge the DPD and physical systems, we require that

(1) the time scale of diffusional transport of momentum (and energy if in nonisothermal simulations) inside the DPD fluids should match that of the real fluids; and

(2) DPD fluids should provide objects immersed in them the proper random "kicks" such that these objects exhibit correct fluctuation behaviors at the macroscopic scale.

We now consider the application of the above requirements in the simulation of colloidal particles (diameter, $\bar{d}$; density: $\bar{\rho}$) dispersed in fluids (density, $\bar{\rho}$; kinematic viscosity, $\bar{\nu}$; temperature, $\bar{T}$). Properties of corresponding DPD fluids are denoted by the same symbol as in real fluids but without the bar, e.g., density of DPD fluids is denoted as ρ. We will limit our discussion to isothermal simulations, and the DPD model reads[2]

$$d\boldsymbol{r}_i = \boldsymbol{v}_i dt, \quad m d\boldsymbol{v}_i = \boldsymbol{F}_i^C dt + \boldsymbol{F}_i^D dt + \boldsymbol{F}_i^R \sqrt{dt}, \tag{1}$$

where m, $\boldsymbol{r}_i$, and $\boldsymbol{v}_i$ are the mass, position, and velocity of bead i, respectively. T is the system temperature. $\boldsymbol{F}_i^C$, $\boldsymbol{F}_i^D$ and $\boldsymbol{F}_i^R$ are the conservative, dissipative, and random forces acting on bead i, respectively. These forces are given by[2,13]

$$\boldsymbol{F}_i^C = \sum_{j \neq i} a_{ij} w(r_{ij}/r_c) \boldsymbol{e}_{ij}, \tag{2}$$

$$\boldsymbol{F}_i^D = \sum_{j \neq i} -\gamma_{ij} w_d^2(r_{ij}/r_c)(\boldsymbol{e}_{ij} \cdot \boldsymbol{v}_{ij}) \boldsymbol{e}_{ij}, \tag{3}$$

$$\boldsymbol{F}_i^R = \sum_{j \neq i} \sigma_{ij} w_d(r_{ij}/r_c) \theta_{ij} \boldsymbol{e}_{ij}, \tag{4}$$

where a_{ij} is the conservative force coefficient and $r_{ij} = |\mathbf{r}_{ij}| = |\mathbf{r}_i - \mathbf{r}_j|$. w and w_d are the weighing functions with a cutoff distance of r_c. $\boldsymbol{e}_{ij} = \mathbf{r}_{ij}/r_{ij}$ and $\boldsymbol{v}_{ij} = \mathbf{v}_i - \mathbf{v}_j$. θ_{ij} is a symmetric random variable with zero mean and unit variance. γ_{ij} and σ_{ij} are related by the FDT as $\gamma_{ij} = \sigma_{ij}^2/2k_BT$, where k_B is the Boltzmann constant. Mass, length, and time in the above model are measured by m, r_c, and $t_c = r_c/\sqrt{k_BT/m}$, respectively. In this paper, all DPD quantities are rendered dimensionless by m, r_c, t_c, and their combinations. We assume that the unit length and time in DPD map to physical length $[L]$ and time $[t]$, respectively. To satisfy the proposed requirements, we enforce

$$d^2/\nu[t] = \bar{d}^2/\bar{\nu}, \tag{5}$$

$$\sqrt{k_BT/\rho d^3}[L]/[t] = \sqrt{\bar{k}_B\bar{T}/\bar{\rho}\bar{d}^3}. \tag{6}$$

Equation (6) uses the fact that the mass of the nanoparticle $\bar{m}=\pi/6\bar{\rho}\bar{d}^3$ is mapped to $m=\pi/6\rho d^3$ in the DPD model. Combining Eqs. (5) and (6),

$$[t]=[L]^2\nu/\bar{\nu}, \tag{7}$$

$$k_BT=\frac{\rho\nu^2}{\bar{\rho}\bar{\nu}^2}\frac{\overline{k_BT}}{[L]}. \tag{8}$$

Equations (7) and (8) can be used to setup and to analyze DPD simulations of fluctuating hydrodynamics. Equation (7) provides the mapping of time scale in DPD model. Equation (8) indicates that T, $[L]$, ρ, and transport properties of DPD fluids (represented by ν) are not independent but are subject to an additional constraint in fluctuating hydrodynamics simulations. Since $[L]$ and ρ are determined by the desired level of coarse graining, one can tune T and ν to satisfy Eqs. (7) and (8). There are many ways to achieve this, e.g., one can choose T first and then tune DPD parameters, such as a_{ij} and σ_{ij}, to obtain the necessary ν. However, given that a_{ij} only weakly affects the viscosity and choice of σ_{ij} is constrained by numerical stability,[2] we propose to set both a_{ij} and σ_{ij} first and then choose the appropriate T to satisfy Eqs. (7) and (8). In practice, since ν of DPD fluids depends on their temperature, the temperature can only be determined after such dependence is known. From Eq. (8), we observe that the temperature of DPD system generally decreases as the level of coarse graining increases (i.e., as $[L]$ increases). Since DPD fluids can become a weak solid at very low temperature, such a mapping scheme may be limited by such artifacts. However, for mesosystems with characteristic dimension smaller than 1 μm, we did not find such limitation to be important. Should such limitations become important, one may choose to tune the fluids viscosity by using different thermostatting techniques.[14]

To demonstrate the above mapping scheme and DPD's ability in modeling fluctuating hydrodynamics, we study the diffusion of a neutrally buoyant nanoparticle immersed in water ($\bar{\rho}=10^3$ kg/m^3, $\bar{\nu}=0.89\times10^{-6}$ m^2/s) at 300 K. We set $[L]=10$ nm and $\rho=6.0$. The nanoparticle is built by bonding 117 DPD beads together and is modeled as a rigid body. We choose $w(r_{ij}/r_c)=1-r_{ij}/r_c$ and $w_d(r_{ij}/r_c)=\sqrt[4]{1-r_{ij}/r_c}$ with $r_c=1.0$[13]. $a_{\mathrm{ff}}=10.0$ and $a_{\mathrm{fp}}=17.0$, where ff and fp denote fluid-fluid and fluid-nanoparticle interactions, respectively. $\sigma_{ij}=5.0$ for all bead pairs. We measured the viscosity of DPD fluid as a function of temperature, and the temperature in DPD model of the nanoparticle-fluid system is then determined via Eq. (8) to be 0.3875. Next, we computed the diffusion coefficient D_p of the particle, which is a macroscopic manifest of the thermal fluctuations. We run five simulations with different initial configurations each for 10^6 steps (simulating box size, $18\times18\times18$; time step, 0.001). From the mean square displacement, shown in Fig. 1(a), a D_p of $(1.59\pm0.12)\times10^{-4}$ was obtained. We also computed D_p by using the Einstein–Stokes law $D_p=k_BT/6\pi R\rho\nu$. From the particle-fluid pair correlation function, shown in Fig. 1(b), the particle radius R was determined to be 1.65, with which a D_p of 1.55×10^{-4} was then computed. Given the ambiguity in the definition of particle diameter and the statistical uncertainty of DPD results, the agreement between DPD and Einstein–Stokes prediction is good. This verifies the ability of DPD in capturing the macroscopic effects of thermal fluctuations in particulate suspensions and in modeling fluctuating hydrodynamics.

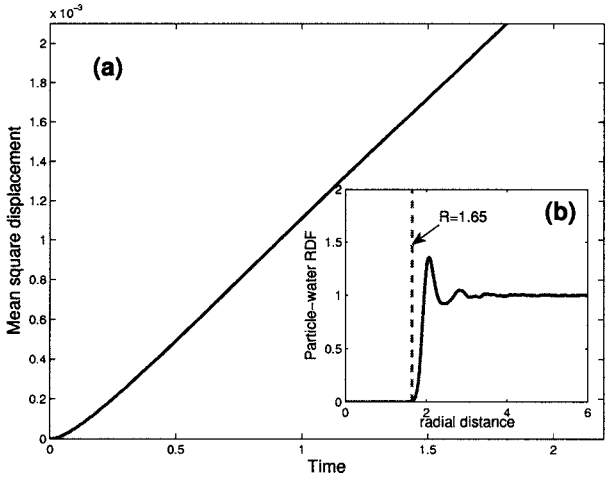

FIG. 1. (a) Mean square displacement of the nanoparticle immersed in fluids. (b) Particle-fluid pair correlation obtained from DPD simulation.

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