From Molecular Dynamics to Dissipative Particle Dynamics

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A procedure is introduced for deriving a coarse-grained dissipative particle dynamics from molecular dynamics. The rules of the dissipative particle dynamics are derived from the underlying molecular interactions, and a Langevin equation is obtained that describes the forces experienced by the dissipative particles and specifies the associated canonical Gibbs distribution for the system.

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Hydrodynamic simulations of complex fluids remain a major challenge in most cases of interest. Such fluids include particulate and colloidal suspensions, polymeric liquids, emulsions and other self-assembling amphiphilic fluids, and fluids where Brownian motion is important. For such fluids it is often necessary to base the modeling on a microscopic picture of the system, thus working from the bottom upwards. Over the last decade several such "bottom up" strategies have been introduced. Hydrodynamic lattice gases [1], which model the fluid as a discrete set of particles, represent a computationally efficient discretization of the more conventional molecular dynamics (MD) [2].

A recent contribution to the family of bottom-up approaches is the dissipative particle dynamics (DPD) method introduced by Koelman and Hoogerbrugge in 1992 [3]. Applications of the technique include colloidal suspensions [4], polymer solutions [5], and binary immiscible fluids [6]. For specific applications where comparison is possible, this model is orders of magnitude faster than MD [7].

The basic components of DPD are particles that are thought to represent mesoscopic elements of the underlying molecular fluid. These dissipative particles then evolve just as MD particles but with different interparticle forces: Since the DPD particles are pictured as having internal degrees of freedom, the forces between them have both a fluctuating and a dissipative component in addition to the conservative forces that are present already at the MD level. Nevertheless, momentum conservation along with mass conservation produce hydrodynamic behavior at the macroscopic level.

Dissipative particle dynamics has been demonstrated to connect correctly to the macroscopic continuum theory; that is, for a one-component DPD fluid, it is possible to derive the Navier-Stokes equations and to compute the viscosity in the large scale limit [8,9]. However, thus far no attempt has been made to link DPD to the underlying microscopic dynamics. This is the purpose of the present Letter. We define the dissipative particles (DP) by appropriate weight functions that sample a portion of the underlying conservative MD particles, and we derive the forces between the DP's from the hydrodynamic description of the MD system.

The present development has two main virtues, one fundamental and one practical. From a fundamental point of view our work gives a microscopic foundation to DPD and thus provides a quantitative meaning to the term "mesoscopic." On the practical side this foundation may be used to deal with physical systems where the modeling is challenged by the simultaneous presence of several different length scales. While conventional DP's are spheres of fixed size and mass, the current DP's are defined as cells on a Voronoi lattice with variable sizes. This provides us with the freedom to define particle sizes according to the local resolution requirements—a particle analog to adaptive meshes in finite—element simulations [10]. The concept is illustrated by the simulation of a

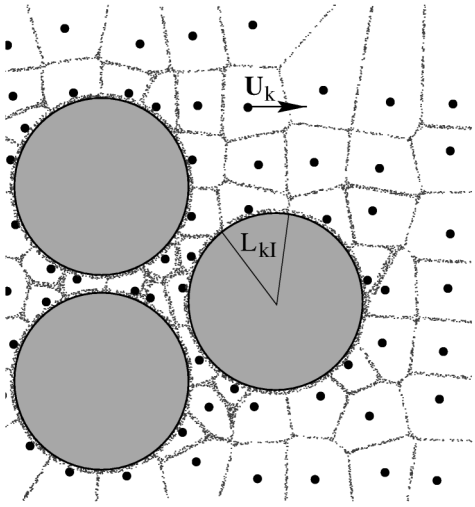

FIG. 1. Multiscale modeling: The dissipative particles are defined as cells in the Voronoi lattice, moving with velocity $\mathbf{U}_k$. There are four relevant length scales: The scale of the large, gray colloid particles, the two scales of the dissipative particles in between and away from the colloids, and finally the scale of the MD particles, which are shown as the little dots that form the boundaries between the DP's.

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colloidal suspension, which is shown in Fig. 1. Here the computational effort is adapted to meet the local need for detail of description: it is larger in narrow regions between the particles than in the bulk. Previous DPD simulations have had difficulty with dense colloidal suspensions precisely because the technique is unable to handle multiple length scale phenomena [4]. Other complex systems where modeling and simulation frequently involve several simultaneous length scales include polymeric and amphiphilic fluids, particularly in porous media and restricted geometries [11].

The basic ingredient in our derivation of DPD is an appropriate coarse-graining scheme. The dissipative particles are defined as clusters of MD particles in such a way that the MD particles are all represented by the dissipative particles. A general way to achieve this is via the sampling function $f_k(\mathbf{x}) = s(\mathbf{x} - \mathbf{r}_k)/\sum_l s(\mathbf{x} - \mathbf{r}_l)$. Here the positions $\mathbf{r}_k$ and $\mathbf{r}_l$ define the DP centers, which initially may be distributed arbitrarily in space, $\mathbf{x}$ is an arbitrary position and $s(\mathbf{x})$ is some localized function, which we choose as a Gaussian $s(\mathbf{x}) = \exp(-x^2/a^2)$; the distance a sets the scale of the sampling function. The mass M_k, momentum $\mathbf{P}_k$, and internal energy E_k of the kth DP are then defined as

$$
\begin{aligned}
M_k &= \sum_i f_k(\mathbf{x}_i) m \,, \\
\mathbf{P}_k &= M_k \mathbf{U}_k = \sum_i f_k(\mathbf{x}_i) m \mathbf{v}_i \,, \\
\frac{1}{2} M_k U_k^2 + E_k &= \sum_i f_k(\mathbf{x}_i) \left(\frac{1}{2} m v_i^2 + \frac{1}{2} \sum_{j \neq i} V(r_{ij}) \right),
\end{aligned}
\tag{1}
$$

where $\mathbf{x}_i$ and $\mathbf{v}_i$ are the position and velocity of the ith MD particle, all assumed to have identical masses m, $V(r_{ij})$ is the MD interparticle potential, and $\mathbf{U}_k$ is the velocity of the kth DP. The kinematic condition $\dot{\mathbf{r}}_k = \mathbf{U}_k$ completes the definition of the DPD. The normalization property $\sum_k f_k(\mathbf{x}) = 1$ implies directly that $\sum_k M_k = \sum_i m$ and $\sum_k M_k \mathbf{U}_k = \sum_i m \mathbf{v}_i$, so that if mass, momentum, and energy are conserved at the MD level, they are also conserved at the DP level.

In order to obtain the equations of motion for the DPD we now take the time derivatives of Eqs. (1). The Gaussian form of s makes it possible to write the time-derivative $\dot{f}_k(\mathbf{x}_i) = f_{kl}(\mathbf{x}_i)(\mathbf{v}_i' \cdot \mathbf{r}_{kl} + \mathbf{x}_i' \cdot \mathbf{U}_{kl})$ where the function f_{kl} is defined as $f_{kl}(\mathbf{x}) \equiv (2/a^2) f_k(\mathbf{x}) f_l(\mathbf{x})$ and $\mathbf{v}_i' = \mathbf{v}_i - (\mathbf{U}_k + \mathbf{U}_l)/2$, $\mathbf{x}_i' = \mathbf{x}_i - (\mathbf{r}_k + \mathbf{r}_l)/2$, $\mathbf{U}_{kl} = \mathbf{U}_k - \mathbf{U}_l$, $\mathbf{r}_{kl} = \mathbf{r}_k - \mathbf{r}_l$. After some algebra [12] the microscopic equations of motion then take the form

$$
\begin{aligned}
\frac{dM_k}{dt} &= \sum_l \dot{M}_{kl} \equiv \sum_{li} f_{kl}(\mathbf{x}_i) m (\mathbf{v}_i' \cdot \mathbf{r}_{kl} + \mathbf{x}_i' \cdot \mathbf{U}_{kl}) \,, \\
\frac{d\mathbf{P}_k}{dt} &= M_k \mathbf{g} + \sum_l \dot{M}_{kl} \frac{\mathbf{U}_k + \mathbf{U}_l}{2} + \sum_{li} f_{kl}(\mathbf{x}_i) \mathbf{\Pi}_i' \cdot \mathbf{r}_{kl} \,, \\
\frac{dE_k}{dt} &= \sum_l \frac{\dot{M}_{kl}}{2} \left(\frac{\mathbf{U}_{kl}}{2} \right)^2 + \sum_{li} f_{kl}(\mathbf{x}_i) \left(\mathbf{J}_i' - \mathbf{\Pi}_i' \cdot \frac{\mathbf{U}_{kl}}{2} \right) \cdot \mathbf{r}_{kl} \,,
\end{aligned}
\tag{2}
$$

where we have defined the general momentum-flux tensor $\mathbf{\Pi}_i' = m \mathbf{v}_i' \mathbf{v}_i' + (1/2) \sum_j \mathbf{F}_{ij} \Delta \mathbf{x}_{ij}$, where $\mathbf{F}_{ij}$ is the force between MD particles i and j, the microscopic energy flux vector $\mathbf{J}_i' = \epsilon_i \mathbf{v}_i' + (1/4) \sum_{i \neq j} \mathbf{F}_{ij} \cdot (\mathbf{v}_i' + \mathbf{v}_j') \Delta \mathbf{x}_{ij}$, and $m\mathbf{g}$ is the external force on an MD particle. In the mass equation above the $\mathbf{x}_i' \cdot \mathbf{U}_{kl}$ term may be shown to be negligible upon averaging, as it samples the difference in mass density rather than the average of this quantity across the region where $f_{kl} \neq 0$ [12]. For that reason it will be omitted, and we have already omitted the corresponding terms in the momentum and energy equations.

All the interaction terms in the above transport equations are weighted by the overlap function $f_{kl}(\mathbf{x})$. If only two DP's, k and l say, are present it may be shown that $f_{kl}(\mathbf{x}) = [1/(2a^2)] \cosh^{-2}\{[\mathbf{x} - (\mathbf{r}_k + \mathbf{r}_l)/2] \cdot (\mathbf{r}_k - \mathbf{r}_l)/a^2\}$. This function becomes exponentially small away from the dividing line that is equally far from $\mathbf{r}_k$ and $\mathbf{r}_l$, as is illustrated in Fig. 2. The set of all such dividing lines defines a Voronoi lattice. In Fig. 1 fictitious MD particles are plotted where $f_{kl}(\mathbf{x}) > 0.2a^2$. This happens in the neighborhood of the dividing lines. When additional DP's are present their contribution to the $\cosh^{-2}$ result may be shown to be negligible in the

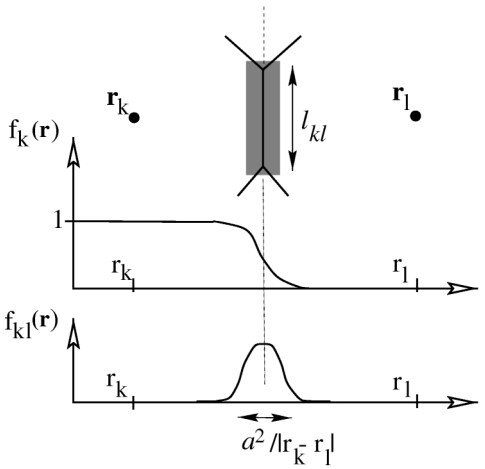

FIG. 2. The overlap region between two Voronoi cells is shown in grey. The sampling function $f_k(\mathbf{r})$ is shown in the upper graph and the overlap function in the lower graph. The width of the overlap region is $a^2/|\mathbf{r}_k - \mathbf{r}_l|$, and its length is denoted by l_{kl}.

vicinity of the dividing line, except at the corners, where dividing lines meet. In the end the DPD equations of motion will turn out to be independent of a, and only the length l_{kl} shown in Fig. 2 will remain. At this point it suffices to construct the Voronoi lattice itself, and there is no need to evaluate the overlap functions. Standardized algorithms and software for the construction of Voronoi lattices exist [13].

Splitting Eqs. (2) into fluctuating and average parts gives

$$\begin{aligned}
\frac{dM_k}{dt} &= \sum_{li} f_{kl}(\mathbf{x}_i) m \langle \mathbf{v}_i' \rangle \cdot \mathbf{r}_{kl} + \sum_l \dot{\tilde{M}}_{kl} \\
\frac{d\mathbf{P}_k}{dt} &= M_k \mathbf{g} + \sum_l \left[\sum_i f_{kl}(\mathbf{x}_i) \langle \mathbf{\Pi}^i \rangle \cdot \mathbf{r}_{kl} \right] + \tilde{\mathbf{F}}_{kl} \\
\frac{dE_k}{dt} &= \sum_{li} f_{kl}(\mathbf{x}_i) \left[\langle \mathbf{J}_i' \rangle - \langle \Pi_i' \rangle \cdot \frac{\mathbf{U}_{kl}}{2} \right] \cdot \mathbf{r}_{kl} + \sum_l \tilde{\mathbf{F}}_{kl} \cdot \frac{\mathbf{U}_{kl}}{2} + \tilde{q}_{kl} ,
\end{aligned} \tag{3}$$

where $\dot{\tilde{M}}_{kl}$ is the fluctuating part of the mass flux, $\tilde{F}_{kl}$ is the fluctuating part of the momentum flux $\sum_i f_{kl}(\mathbf{x}_i) \mathbf{\Pi}^i \cdot \mathbf{r}_{kl} + \dot{M}_{kl}(\mathbf{U}_k + \mathbf{U}_l)/2$, and $\tilde{q}_{kl}$ is the fluctuating part of the energy flux $\sum_i f_{kl}(\mathbf{x}_i) \mathbf{J}_i' \cdot \mathbf{r}_{kl} + (1/2)\dot{M}_{kl}(\mathbf{U}_{kl}/2)^2$. Note that we have absorbed the contributions from the mass variations in $\tilde{\mathbf{F}}_{kl}$ and $\tilde{q}_{kl}$. The thermal averages, $\langle ... \rangle$, are computed by means of an ensemble of systems with common *instantaneous* values of the mesoscopic variables $\{\mathbf{r}_k, M_k, \mathbf{U}_k, E_k\}$. This means that only the time derivatives of this set have a fluctuating part.

It is necessary to introduce *some* average description of the MD system. For this purpose we assume the scale separation $a \ll |\mathbf{r}_k - \mathbf{r}_l|$, for all k and l, and that the width of the overlap region a^2/r_{kl} is larger than the mean free path of the MD particles. For simplicity, we choose the momentum flux tensor of a simple Newtonian fluid which has the form $\langle \mathbf{\Pi}_{kl}^i \rangle = \mathbf{I}P - \eta[\nabla \mathbf{v} + (\nabla \mathbf{v})^T]$, where η is the dynamic viscosity and P the pressure of the MD fluid, ^{T} denotes the transpose, and $\mathbf{I}$ is the identity tensor [14]. We shall make the approximation that the average molecular particle velocity $\langle \mathbf{v} \rangle$ interpolates linearly between the DP's.

It follows that the average mass current $\langle \mathbf{v}' \rangle = \mathbf{0}$ and that the velocity gradients in the momentum-flux tensor take the form $\nabla \mathbf{v} + (\nabla \mathbf{v})^T = 1/r_{kl}(\mathbf{e}_{kl}\mathbf{U}_{kl} + \mathbf{U}_{kl}\mathbf{e}_{kl})$, where $\mathbf{e}_{kl} = (\mathbf{r}_k - \mathbf{r}_l)/|\mathbf{r}_k - \mathbf{r}_l|$. Since $\langle \mathbf{v}' \rangle \approx \mathbf{0}$ we may choose the heat flux according to Fouriers law $\langle \mathbf{J}' \rangle = \lambda \nabla T$, where T is the temperature and λ is the thermal conductivity. In other words, in the frame of reference of the overlap region the energy flux is simply the heat flux since work terms proportional to the velocity vanish [14]. With this input we get $\dot{M}_k = \sum_l \dot{\tilde{M}}_{kl}$ and

$$\begin{aligned}
\frac{d\mathbf{P}_k}{dt} &= M_k \mathbf{g} - \sum_l l_{kl} \left\{ \frac{p_{kl}}{2} \mathbf{e}_{kl} + \frac{\eta}{r_{kl}} [\mathbf{U}_{kl} + (\mathbf{U}_{kl} \cdot \mathbf{e}_{kl}) \mathbf{e}_{kl}] \right\} + \sum_l \tilde{\mathbf{F}}_{kl} \\
\frac{dE_k}{dt} &= \sum_l l_{lk} \left\{ \frac{p_k + p_l}{2} \mathbf{e}_{kl} + \frac{\eta}{r_{kl}} [\mathbf{U}_{kl} + (\mathbf{U}_{kl} \cdot \mathbf{e}_{kl}) \mathbf{e}_{kl}] \right\} \cdot \frac{\mathbf{U}_{kl}}{2} + \sum_l l_{lk} \lambda \frac{T_{kl}}{r_{kl}} + \tilde{\mathbf{F}}_{kl} \cdot \frac{\mathbf{U}_{kl}}{2} + \tilde{q}_{kl} ,
\end{aligned} \tag{4}$$

where we have assumed that the pressure p and temperature T, as well as the average velocity, interpolates linearly between DP centers, $p_{kl} = p_k - p_l$ and $T_{kl} = T_k - T_l$. The pressure will eventually follow from an equation of state of the form $p_k = p(E_k, V_k)$ where V_k is the volume of DP k. The pressure and temperature must be obtained via a thermodynamic description, i.e., an equation of state that relates pressure and temperature to energy E_k and volume V_k. In the special case of an ideal gas (see Ref. [12] for a more general treatment), these relations simplify to $dp_k = p_k(dE_k/E_k - dV_k/V_k)$ and $dT_k = dE_k/(Nk_B)$.

The average rate of change of M_k vanishes. This allows us to neglect mass variations altogether in the DPD equations, since the effect of mass fluctuations may then be absorbed in $\tilde{\mathbf{F}}_{kl}$ and $\tilde{q}_{kl}$. Had there been a coupling, say, between the *averaged* momentum and mass values the mass would have had to be updated as well. With nothing but fluctuations in M_k, the only change introduced by the $M_k = \text{const}$ approximation is the loss of the drift in the M_k's around their constant average, caused by the fluctuations. Nothing is neglected in the instantaneous changes of momentum and energy.

In general, force fluctuations will cause mass fluctuations, which in turn will couple back to cause momentum fluctuations. The time scale over which this will happen is $t_\eta = r_{kl}^2/\eta$, where η is the dynamic viscosity of the MD system. This is the time it takes for a velocity perturbation to decay over a distance r_{kl}. We shall need to make the assumption that the fluctuating forces are Markovian, and it is clear that this assumption may be valid only on time scales larger than t_η. Since the time scale of a hydrodynamic perturbation of size l, say, is also given as l^2/η this restriction implies the scale separation requirement $r_{kl}^2 \ll l^2$, consistent with the scale r_{kl} being mesoscopic.

In "conventional" DPD [3,9,15] the forces are pairwise and act parallel to $\mathbf{e}_{kl}$. They have a conservative part that

depends only on r_{kl} and a dissipative part proportional to $(\mathbf{U}_{kl} \cdot \mathbf{e}_{kl})\mathbf{e}_{kl}$. Here the same terms are present. The conservative force is seen to arise from the pressure and the dissipative part from dissipation in the underlying fluid with the MD viscosity taking the place of a postulated friction coefficient. In addition, there is a dissipative term parallel to $\mathbf{U}_{kl}$. The energy part of Eq. (4) is identical in form to the energy equation postulated by Avalos and Mackie [16] and similar to the equation studied by Español [17], save for the fact that here the work done by the conservative force $(p_k + p_l)\mathbf{e}_{kl} \cdot \mathbf{U}_{kl}/4$ is present. The principal difference is associated with the Voronoi lattice: Our DP's fill space and change their shape, a key feature that enables this new DPD to treat a multitude of length scales within a single simulation.

In order to obtain $\tilde{\mathbf{F}}$ and $\tilde{q}$ we invoke the Markovian approximation to write $\tilde{\mathbf{F}} = \boldsymbol{\omega}_{kl\parallel} W_{kl\parallel} + \boldsymbol{\omega}_{kl\perp} W_{kl\perp}$, where $\tilde{\mathbf{F}}_{kl}$ is decomposed into components parallel and perpendicular to $\mathbf{e}_{kl}$, and the W's are defined as Gaussian random variables with the correlation function $\langle W_{kl\alpha}(t) W_{nm\beta}(t')\rangle = \delta_{\alpha\beta}\delta(t - t')(\delta_{kn}\delta_{lm} + \delta_{km}\delta_{ln})$ where α and β denote either $\perp$ or $\parallel$. Newton's third law guarantees that $\boldsymbol{\omega}_{kl} = -\boldsymbol{\omega}_{lk}$. We have the similar expression $\tilde{q}_{kl} = \Lambda_{kl} W_{kl}$ where $\Lambda_{kl} = -\Lambda_{lk}$ and W_{kl} satisfies the above equation for the W's without the $\delta_{\alpha\beta}$-factor.

In Refs. [8,12,16] the magnitudes of $\tilde{\mathbf{F}}_{kl}$ and $\tilde{q}_{kl}$ are obtained on the basis of the Fokker-Planck equation which derives from equations like Eqs. (4). The isothermal results, adapted to the present case, are the fluctuation dissipation relations $\omega^2_{kl\parallel} = 2\omega^2_{kl\perp} = 4\eta k_B T(l_{kl}/r_{kl})$ for the force $\tilde{\mathbf{F}}_{kl}$ and for the heat fluctuations $\Lambda^2_{kl} = 2k_B T\lambda(l_{kl}/r_{kl})$ [16]. It is also possible to show that detailed balance [18] holds, and that the DP's obey the Gibbs distribution [12] $\rho^{\mathrm{eq}} = Z^{-1}(T, V)\exp\{-\beta\sum_k [P_k^2/2M_k + V(\mathbf{r}_k)]\}$, where the potential $V(\mathbf{r}_k)$ is responsible for the pressure force in Eq. (4), and $T = 1/\beta k_B$ is the temperature characterizing the MD system [12].

Considering now the application illustrated in Fig. 1 we need to define DP-colloid forces. Taking the hydrodynamic momentum flux tensor and Eq. (4) as a starting point, we observe that the DP-colloid interaction may be obtained in the same form as the DP-DP interaction of Eq. (4) with the replacement of $l_{kl} \rightarrow L_{kI}$, where L_{kI} is the length (area in 3D) of the arc segment where the DP meets the colloid (see Fig. 1). The velocity gradient is that between the DP and the colloid surface. The latter may be computed by linear interpolation using $\mathbf{U}_k$ and the velocity of the colloid surface together with a no-slip boundary condition on this surface. In the momentum fluctuation-dissipation relation too the replacement $l_{kl} \rightarrow L_{kI}$ must be made. In order to increase the spatial resolution where colloidal particles are close, it is necessary to introduce a higher DP density there; this ensures that fluid lubrication effects are maintained. After these particles have moved it may be necessary to retile the DP system, as is done in finite element calculations. This is simply achieved by distributing the mass, momentum, and energy of the old DP's on the new ones according to their area (or volume in 3D).

The DPD which we have derived in the present work is similar to conventional DPD. But the forces conventionally used to define DPD have now been given a microscopic basis. More important, however, is the fact that our analysis permits the introduction of specific physical interactions at the mesoscopic level, together with a well-defined meaning for this mesoscale. Finally, we note the similarity of the present particulate description, which is based on a bottom-up approach, to existing continuum approaches, which start out from a macroscopic description. Such top-down approaches include in particular smoothed particle hydrodynamics [19] and finite-element simulations. In these descriptions too the computational method is based on tracing the motions of elements of the fluid on the basis of the forces acting between them [20]. We stress, however, that while such top-down computational strategies require as initial input a macroscopic phenomenological description, the present approach relies on a microscopic representation from the outside.

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