Coarse-Graining and Scaling in Dissipative Particle Dynamics

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Dissipative particle dynamics (DPD) is now a well-established method for simulating soft matter systems. However, its applicability was recently questioned because some investigations showed an upper coarse-graining limit that would prevent the applicability of the method to the whole mesoscopic range. This article aims to reestablish DPD as a truly scale-free method by analyzing the problems reported by other authors and by presenting a scaling scheme that allows one to gauge a DPD-simulation to any desired length scale.

Keywords: mesoscopic simulation, dissipative particle dynamics, scaling, renormalization, calibration

I. INTRODUCTION

Dissipative particle dynamics (DPD) was introduced in 1992 by Hoogerbrugge and Koelma as a novel method for performing mesoscopic simulations of complex fluids. Since then, the method has gained significant theoretical support and refinement, and has been applied to fluid dynamics in numerous research areas such as rheology, material sciences, and molecular biology, where membranes, vesicles, and micellar systems have been modeled.

Initially, DPD was understood to be a truly mesoscopic method able to bridge the whole gap between the underlying atomistic scale (in the range of nanometers and nanoseconds) that is accessible by molecular dynamics (MD) simulations and the macroscopic scale (in the range of micrometers and milliseconds) considered by continuum descriptions. To fulfill this promise, it is crucial that the method is scalable, meaning that its coarse-graining level can be adjusted without introducing serious artifacts that would render the method worthless.

Coarse-graining in DPD translates into having a number of physical molecules represented by a single DPD particle. By $N$, we denote the total number of DPD particles in a simulation and it holds $\nu N = N_{\text{phys}}$, with $N_{\text{phys}}$ the number of physical molecules the simulation represents. The main objective of this article is the comparison of DPD simulations with different coarse-graining levels $\nu$ and $\nu'$. This motivates the introduction of the scaling ratio $\phi = N/N' = \nu'/\nu$. In the following, functions of $\phi$ will be used to describe the scaling of various quantities at different coarse-graining levels. (One may set $\nu = 1$ so that scaling ratio and coarse-graining level coincide and thereby simplify the notation and providing a direct link to physical constants such as the Boltzmann constant; this is at the price of using DPD concepts at a length scale on which their interpretation becomes difficult.)

It was originally stated that the method is scale-free, meaning that the parameters used in the simulation do not depend on the level of coarse-graining. In a later publication, this earlier finding was declared erroneous, and it was proposed that interaction parameters determining the conservative forces between DPD particles scale linearly with $\phi$ when one goes from one coarse-graining level $\nu$ to another $\nu'$.

Based on this scaling relation, the performance of DPD was analyzed for various coarse-graining levels. It was found that there exists an upper coarse-graining level above which the simulated fluid freezes. Trovimof reported that this coarse-graining limit is disappointingly low, and only allows up to about 10 water molecules to be grouped together into one DPD particle. This limit would prevent DPD from covering the whole mesoscopic range and confines its applicability essentially to the order of magnitude of MD simulations. Dzwinel and Yuen even concluded that the DPD method would be best suited for the simulation of vapors and gases (where the freezing artifact would happen only for much higher coarsening levels).

In this article, we argue that the original statement that DPD is scale free is in fact correct, and that the later refinement declaring that the DPD interaction parameter scales linearly with the scaling ratio $\phi$ is based on an inappropriate scaling assumption. More specifically, we show that if a scaling method that preserves physical quantities is used and the DPD calculations are performed in reduced units (which is standard practice) then the results are independent of the level of coarse-graining. Expressed in more technical terms, if a physically consistent scaling of all simulation parameters is maintained, the velocity increments calculated from integrating the equations of motion of the DPD particles (c.f. Eqns. (1) below) do not depend on the coarse-graining value used for the DPD particles. This is in contrast to earlier publications.

In this paper, the physical molecules will refer to bulk fluid particles (such as water), which is in accordance with the cited literature. Systems including surfaces, such as binary fluids, will be commented on in the discussion.
The article is structured as follows: in Sec. II A we discuss the results of Groot and Rabone and specify where we deviate from their analysis. We show that their approach of decreasing the number of DPD particles (namely the particle density) while keeping relevant properties, in particular their radius of interaction, constant, is not appropriate. The alternative scaling process we employ is schematically shown in Fig. 1A and 1B. When we change the level of coarse-graining for the DPD-particles, we accordingly scale their number and adjusting their size (radius of interaction). Fig. 1C depicts the main result to be shown in this article, namely that by employing the correct scaling relations and unit reduction one has a complete equivalence of a simulation performed at the scale of Fig. 1A with the zoomed version in Fig. 1C. In other words, we demonstrate that one single DPD-simulation represents the dynamics of a whole family of physical systems at different levels of coarse-graining.

This demonstration is split into two parts. In Sec. II B and II C we change the interaction parameters according to the necessities of the scaling procedure we adopt. The interaction parameters have to be changed such that a system with many DPD-particles is mapped onto one with fewer, but larger and heavier particles. This is done without changing the physical properties of the complete system, namely the compressibility and thermal energy. Sec. II D presents simulation results that corroborate the correctness of the derived scaling relations.

In Sec. III we analyze the behavior of the DPD algorithm when the rescaled system is expressed in its own set of reduced units. The change of units affects the time scale and we will show that as a consequence, the numerical values that appear in the reduced unit system (Fig. II C) are identical to those of the original one and, as a consequence, their dynamics are equivalent.

In order to present our argument, we need to define the nomenclature used and introduce some notation. By “coarse-graining” we understand the operation of coalescing \( \nu \) physical particles into one DPD particle. By “scaling” we refer to the functional relation between the respective parameters of two systems with different coarse-graining resolutions.

Conventionally, DPD operates in reduced units, such that energy is measured in units of \( kT \) (\( T \) being the temperature and \( k \) a scaled version of the Boltzmann constant to be introduced later), length in units of \( r_c \), and mass in units of \( m \), the mass of a single particle; in these units, length, mass, time and energy are dimensionless. We will use two different sets of reduced units, one for the coarse-graining resolution \( \nu \), and another one for \( \nu' \). We denote by \( X \) a quantity expressed in reduced units with respect to coarse-graining level \( \nu \). With \( X' \) we denote the corresponding value of the same quantity calculated in the units of \( X \) but with a different level of coarse-graining. In other words: moving from \( X \) to \( X' \) denotes the scaling operation. Finally, by \( \bar{X} \) we refer to the quantity \( X' \) in its own set of reduced units given by the coarse-graining resolution \( \nu' \). That is, going from \( X' \) to \( \bar{X} \) denotes the reduction of units. One may summarize the combined coarse-graining and change of units with the following diagram:

![Diagram](image)

Excellent descriptions of the DPD-method are given in various articles; we will not recapitulate the method itself, but instead refer to Groot and Warren. Here, we give only the definitions of the conservative, dissipative, and random forces in order to define the notation of the parameters:

\[
\begin{align*}
F_{ij}^C &= a_{ij} \chi_{ij} (1 - \frac{r_{ij}}{r_c}) \hat{r}_{ij}, \\
F_{ij}^D &= -\gamma \omega^D (r_{ij}) [(v_i - v_j) \cdot \hat{r}_{ij}] \hat{r}_{ij}, \\
F_{ij}^R &= \sigma \omega^R (r_{ij}) \zeta_{ij} \hat{r}_{ij},
\end{align*}
\]

where \( r_{ij} \) is the Euclidean distance between particles \( i \) and \( j \), \( \hat{r}_{ij} \) is the unit vector pointing from particle \( j \) to particle \( i \), and \( \chi_{ij} \) equals one for pairs of particles separated by distances less than the force cut-off radius \( r_c \) and equals zero otherwise. The parameter \( a_{ij} \) determines the magnitude of the conservative interaction and will be regarded in this work as being the same for all pairs of particles: \( a_{ij} = a \). \( \zeta_{ij} \) is a random variable with Gaussian statistics, a vanishing mean and a
variance of $1/\Delta t$ for the numerical time step $\Delta t$ (see e.g. Gardiner[21]). As a consequence, the unit of $\zeta_{ij}$ is time$^{-1/2}$. Furthermore, the values of $\zeta_{ij}$ in two different time intervals are uncorrelated. In general, if $(i,j)$ and $(k,l)$ are different pairs of particles, $\zeta_{ij}$ and $\zeta_{kl}$ are independent; however, in order to guarantee the centrality of all forces, one must require that $\zeta_{ij} = \zeta_{ji}$.

The following dissipation-fluctuation relation leads to a thermal equilibrium at a given temperature $T$:

$$2k_BT\gamma\omega^D(r) = \sigma^2[\omega^R(r)]^2.$$  

(2)

Without loss of generality, we may take $\omega^D(r) = [\omega^R(r)]^2$ for the dimensionless weighing functions $\omega^D(r)$ and $\omega^R(r)$. The dissipation-fluctuation relation (2) then reduces to

$$\sigma^2 = 2k_BT\gamma.$$  

(3)

One is free to choose either $\omega^D(r)$ or $\omega^R(r)$ without changing the thermodynamic equilibrium, but it is customary in the literature to take $\omega^R(r) = 1 - r/r_c$ mimicking the conservative force $F^C$. One also notes that through Eqns. (2) and (3), the relations between the parameters therein are dependent on the temperature.

II. SCALING DPD

A. Compressibility and Equation of State

Following Groot and Warren[23] we analyze the scaling behavior of the conservative interaction parameter $a$ by relating the thermodynamic definition of the isothermal compressibility $\kappa_T$ to the equation of state (involving $a$) of a system of DPD-particles. The isothermal compressibility is defined as the fractional change in the volume $V$ that results from a change in the pressure $P$ of the system, in a process where the temperature $T$ is constant:

$$\kappa_T = -\left.\frac{1}{V}\frac{\partial V}{\partial P}\right|_T = \frac{1}{\rho}\left.\frac{\partial P}{\partial \rho}\right|_T.$$  

(4)

Here $\rho$ denotes the number density of particles, i.e. $N/V$. It is convenient to define the dimensionless parameter $\kappa^{-1}$ by

$$\kappa^{-1} = \frac{1}{k_BT\rho\kappa_T} = \frac{1}{k_BT} \left.\frac{\partial P}{\partial \rho}\right|_T,$$  

(5)

which we require to be invariant under scaling.

$$\left.\frac{1}{k_BT\rho\kappa_T}\right|_{\text{sim}} = \frac{1}{k_BT} \left.\frac{\partial P}{\partial \rho}\right|_{\text{sim}} = \frac{1}{k_BT} \left.\frac{\partial P}{\partial \rho}\right|_{\text{exp}} = \text{const.}$$  

(6)

Here, $n = \nu\rho$ denotes the molecular number density of the physical system and the subscript “exp” relates to the experimental value, while “sim” refers to the simulation value.

The equation of state relates the pressure with the particle number density $\rho$. From simulation results, Groot and Warren[23] evaluated the virial expression

$$P = \rho k_BT + \frac{1}{3V} \left\langle \sum_{j>i} r_{ij} F^C_{ij} \right\rangle,$$

$$= \rho k_BT + \frac{2\pi}{3} r_c^2 \int_0^{r_c} r a \left( 1 - \frac{r}{r_c} \right) g(r) r^2 dr.$$  

(7)

Here, $g(r)$ denotes the radial distribution function. For densities $\rho > 2$ in reduced units (i.e. more than 2 particles in a cubic box with linear dimensions of $r_c$), the following equation of state is a good approximation to the numerical simulations:

$$P = \rho k_BT + a\alpha \rho^2,$$  

$$\alpha = 0.101 \pm 0.001.$$  

(8)

From this, one concludes that the part of the pressure caused by the conservative interaction scales linearly in $a$. From Eqn. (4), we obtain that at constant temperature

$$\frac{1}{k_BT} \left.\frac{\partial}{\partial \rho} (k_BT a\alpha \rho^2)\right|_\gamma = \text{const.}$$  

(9)

Using $\rho = n/\nu$ where $n$ is the molecular number density and $\nu$ the chosen coarse-graining parameter, it follows that

$$1 + 2aan = \text{const.}$$  

(10)

Since $n$ and $T$ are constant, Groot and Rabone[23] concluded that $a$ must scale linearly with $\phi = \nu'/\nu$ in order to maintain isothermal compressibility under a change of the coarse-graining level.

Several authors regard this scaling as an inherent drawback of the DPD-method, since on the micrometer scale the method would appear to be effectively thwarted. We claim that this scaling – and hence its implications – is wrong. The problem is rooted in the construction of the equation of state (Eqn. 8). In their computer simulations, Groot and Rabone[23] decreased the density of DPD particles while keeping the interaction cutoff radius $r_c$ constant. This approach allows one to keep the system behavior invariant by scaling the interaction parameters while changing the DPD particle density, without altering their properties. However, proper scaling means lowering the number of employed DPD particles while simultaneously enlarging their interaction radius. The difference is illustrated in Fig. 2. Frame 2A is taken to be a system with fine coarse-graining. Frame 2B represents a scaled system with a lower DPD particle density but unchanged particle diameters. The result is that the mutual overlap of the soft particles is smaller (as seen in Frame 2B). Hence it is intuitively clear that the interaction parameter has to be increased in order to keep the system properties constant; formally, this argument is reflected in Eqn. (10). In contrast, Frame 2C shows the system with the same
scaling ratio as for Frame 2B, but with the relative overlap of the interacting particles kept constant, which is accomplished by scaling $r_c$. A closer examination of Frame 2C shows that it is part of a magnified version of Frame 2A, namely a system where all the lengths associated with a single DPD particle have been uniformly scaled by a factor $\phi$ while keeping the system size constant ($L' = L$). This results in the following scaling relations for the coarse-graining level, number, mass, and size of DPD particles:

\[
\nu' = \phi \nu \\
N' = \phi^{-1} N \\
m' = \phi m \\
r_c' = \phi^{1/d} r_c,
\]

where $d$ is the number of dimensions of the system (see Fig. 2C).

![Fig. 2: Schematic of the scaling process: Frame A shows a DPD simulation with a cutoff radius of $r_c$. Frame B depicts the coarse-graining procedure performed in Groot and Rabone. With changing particle density, the particle diameter is kept constant while the interparticle force is increased to maintain the system pressure. However, in order to properly conserve systemic parameters like compressibility, both the interaction parameter and the interaction cut-off radius need to be increased as measured in physical units. Frame C depicts the proper scaling in coarse-graining. Along with a decrease of the particle density (in physical units), the interaction range is increased. In this case, the interaction parameter $a$ scales differently than in Frame B in order to preserve systemic properties.](image)

### B. Scaling of the Potential Energy

We start by calculating the change of potential energy $U$ of a system of DPD particles enclosed in a box that undergoes compression. This change is related to the compressibility of the system and is required to be invariant under scaling. In practice, we require the dependence of $a$ on $\phi$, such that the chosen coarse-graining level does not affect the compressibility. For the un compressed system, we have

\[
U_0 = \sum_{i>j} \frac{\chi_{ij}a}{2r_c} (r_{ij} - r_c)^2.
\]

This equation holds for soft core repulsions which are used throughout the literature. In general, the potential may be viewed as a harmonic approximation of any potential close to an energy minimum. For an isotropically compressed system with box length $(1 - \epsilon)L$, where $\epsilon \ll 1$ is the relative compression parameter, the change in the interparticle distance $\Delta r_{ij}(\epsilon)$ is not assumed to be the same for all pairs of particles. However, we require that

\[
\Delta r_{ij}(\epsilon) = \epsilon r_{ij} + O(\epsilon^2),
\]

which means that we rule out (first order) phase transitions under compression. The total potential energy of the compressed system is then given by

\[
U_c = \sum_{i>j} \frac{\chi_{ij}a}{2r_c} (r_{ij} - \Delta r_{ij}(\epsilon) - r_c)^2.
\]

To first order in $\epsilon$, we obtain for the change of internal energy

\[
\Delta U = U_c - U_0 = \sum_{i>j} \chi_{ij}a \left(1 - \frac{r_{ij}}{r_c}\right) \epsilon r_{ij}.
\]

Because the change in internal energy of the system as a whole has to be invariant under scaling, we have

\[
\sum_{i>j} \chi_{ij}a \left(1 - \frac{r_{ij}}{r_c}\right) \epsilon r_{ij} = \sum_{i>j} N' \chi_{ij} a' \left(1 - \frac{r_{ij}}{r'_c}\right) \epsilon r_{ij}'.
\]

Due to the scaling of $N$, the number of terms in the sum of the left hand side of Eqn. (16) is proportional to $\phi^{-1}$. Since we require $\Delta U$ to be invariant under scaling, the force constant $a$ has to scale as

\[
a' = \phi^{1-1/d} a,
\]

the $\phi^1$ coming from the change in the number of terms in the sum and the $\phi^{-1/d}$ from the change in length scale. We note that this scaling differs from that found by Groot and Rabone where $a$ scaled linearly with $\phi$ (see Eqn. 6). This scaling is the result of our requirement of maintaining the fractional particle overlap during the
change of the coarse-graining level. Everything else being equal, this scaling would seemingly still imply an upper coarse-graining limit, although not as severe as the initial result of Groot and Rabone. However, scaling affects not only length scales and the interaction parameter, but also the energy and time scales implicit in the simulation.

C. Scaling of the Kinetic Energy

Since the unprimed and the primed systems should be physically equivalent, we further require that \( T' = T \) and \( Q' = Q \), where \( Q \) is the thermal energy, i.e. heat content, of the whole system; with these requirements, physical properties become independent of the coarse-graining, and the coarse-graining level is solely a simulation parameter. In order to be consistent with statistical mechanics, a proper consideration of the reduction of the number of degrees of freedom resulting from coalescing several physical particles into a DPD particle is required. The heat content in a 3-D system consisting of particles with no internal structure is given by

\[
Q_{\text{phys}} = \frac{f_{\text{phys}}}{2} k_B T = \frac{N_{\text{phys}}}{2} \langle m_{\text{phys}} v_{\text{phys}}^2 \rangle, \tag{18}
\]

where \( f_{\text{phys}} = dN_{\text{phys}} \) is the number of degrees of freedom. Forming \( N \) DPD-particles, i.e. coherently moving groups of \( \nu \) physical particles, changes the relation between kinetic energy, temperature and number of particles. It holds

\[
Q = \frac{f}{2} kT = \frac{N}{2} \langle mv^2 \rangle \tag{19}
\]

with \( f = 3N, k = \nu k_B, N = N_{\text{phys}}/\nu, m = \nu m_{\text{phys}} \). Consequently, from the required invariance \( Q' = Q = Q_{\text{phys}} \) and \( T' = T \) for two simulations, we get a scaling relation

\[
k' = \phi k \tag{20}
\]

We introduce the parameter \( k \) that has the role of a Boltzmann constant for systems with reduced number of degrees of freedom. (This scaling affects the dissipation-fluctuation relation Eqn. 8 which now reads \( \sigma^2 = 2kT\gamma \)) That Eqn. 12 and hence \( Q = Q' \) is consistent with the DPD method is discussed and explicitly corroborated in Sec. III.

The behavior of \( \gamma \) and \( \sigma \) under scaling remains to be determined. Examining Eqn. 7, we note that the pressure is independent of \( \gamma \) and \( \sigma \). This means that, with respect to static compressibility, we have significant freedom in the choice of the scaling function \( \Gamma \):

\[
\gamma' = \Gamma(\phi) \gamma
\]

\[
\sigma' = (\Gamma(\phi) \phi)^{1/2} \sigma \tag{21}
\]

the latter equation is a consequence of \( \sigma' = \sqrt{2\gamma'/kT'} \). Dimensional analysis motivates the choice

\[
\Gamma(\phi) = \phi^{1-1/d}, \tag{22}
\]

which in turn implies that

\[
\gamma' = \phi^{1-1/d} \gamma
\]

\[
\sigma' = \phi^{1-2/d} \sigma. \tag{23}
\]

This specific choice will later be shown to be crucial for establishing the scalability of the method, but investigations with other goals (such that deciding on a coarse-graining level \( \nu \) with least artefacts) may require alternative gauges.

To summarize what has been established throughout the last two sections, changing the level of coarse-graining in DPD requires the following scaling relations:

\[
N' = \phi^{-1} N \quad a' = \phi^{1-1/d} a \quad m' = \phi m \quad \gamma' = \phi^{1-1/d} \gamma \quad r'_c = \phi^{1/4} r_c \quad \sigma' = \phi^{1-2/d} \sigma \tag{24}
\]

D. Experimental validation

In order to illustrate the correctness of the above scaling arguments, we have measured the pressure in simulations as a function of \( a \) for different coarse-graining levels and temperatures. In these simulations, the DPD particles are confined to a box with hard walls. When a particle collides with a wall, it is reflected elastically and the instantaneous impulse normal to the wall is measured. The pressure is measured as the time-average of the normal forces on the walls divided by the surface area of the cube: \( P = \langle m \Delta v_{\perp} / (A \Delta t) \rangle \) where \( A \) is the area of the box and \( \Delta v_{\perp} \) is the component of the particle velocity orthogonal to the wall.

In Fig. 3 we show a comparison between \( r_c = 1 \) (white squares), corresponding to normal DPD, and a rescaled system with \( r'_c = 2r_c \), corresponding to \( \phi = 8 \) diamonds with black dots). Particle density and box length are set to \( 3r_c^{-3}, 10r_c, \) and \( 3r'_c^{-3}, 10r'_c, \) respectively. Different temperatures \( T_1, T_2, \) and \( T_3 \) are considered, such that \( kT_1 = 1, kT_2 = 2, kT_3 = 3, \) and \( kT'_1 = 1, kT'_2 = 2, kT'_3 = 3 \), respectively (bottom to top in Fig. 3). This is a slight deviation from the conventional usage of reduced units. For the rescaled simulation, all parameters (\( N, m, r_c, a, k, \gamma, \) and \( \sigma \)) have been scaled according to Eqn. 24. We want to show that the pressure of this system is invariant under scaling, for all values of \( a \) and \( T \), if we follow the described scaling relations.

In Fig. 3 the pressure is plotted as a function of \( a \phi^{1-1/d} \), so that we should obtain the same curve independent of the coarse-graining level. On the one hand, for \( a = 0 \), the case of an ideal gas, the pressure should be given by

\[
P|_{a=0} = \phi^{-1} N \phi kT/V = \rho kT, \tag{25}
\]

which it indeed is seen to be. On the other hand, kinetic gas theory establishes

\[
P = \rho \langle m v^2 \rangle / 3. \tag{26}
\]
This constitutes a relationship between the thermostat and the conservative mechanical interactions with the walls, which is non-trivial for our simulation. The way we measure the pressure gives a direct relation to \( \langle \mathbf{v} \mathbf{v} \rangle \). The pressure calculated from kinetic quantities (Eqn. (25)) coincides with the value required from the thermodynamic relation Eqn. (19). This shows that Eqn. (19) holds for our simulation and thereby confirms the scaling relation for \( k \) given in Eqn. (20).

Eqn. (8) estimates a linear relation between \( a \) and \( P \). This relation is shown as solid lines, whereby the Boltzmann factor in Eqn. (8) has been replaced by its scaled version \( k \). For each line, \( P|_{a=0} \) is the theoretical value. The average obtained from each data set \( P(a) - P(0) \) / \( a \). Unless \( a \) is too large, the pressure increases linearly with \( a \). For high values of \( a \), one finds that Eqn. (8) is not a good approximation to the measured pressure, anymore.

The plot also shows that the scaling of \( a \) is correct as the pressure values of the original and the rescaled systems fall right on top of each other, regardless of the value of \( a \). The conclusion from these comparisons is that the pressure is indeed invariant when we use the scaling relations given in Eqn. (24).

![Graph showing the calculated pressure as a function of the interaction strength \( a \).](image)

**FIG. 3:** The calculated pressure as a function of the interaction strength \( a \). We show the pressure for the basic DPD model, corresponding to \( r_c = 1 \) (white squares), and a rescaled simulation of the same 3D system with \( \phi = 8 \) corresponding to \( r'_c = 2r_c \) (diamonds with black dots). Particle density and box length are set to \( 3r_c^{-3}, 10r_\varnothing, \) and \( 3r_c^{-3}, 10r'_\varnothing, \) respectively. Different temperatures \( T_1, T_2, \) and \( T_3 \) are considered, such that \( kT_1 = 1, kT_2 = 2, kT_3 = 3, \) and \( kT'_1 = 1, kT'_2 = 2, kT'_3 = 3, \) respectively (bottom to top). Solid lines show the prediction of the equation of state (Eqn. (8)) with \( \alpha \) estimated from the simulation results (for the case of \( r_c = 1 \)). The pressure increases approximately linearly for \( a \) not too large. Furthermore, the values for the original and the rescaled simulations coincide.

**III. REDUCTION OF UNITS**

Our goal is now to show that the velocity increments \( \Delta \mathbf{v} \) obtained from integrating the forces are unchanged when the scaling is combined with the according reduction of units: \( \Delta \mathbf{v}' = \Delta \mathbf{v}' \), which implies that the relative particle motions are unaffected by scaling in the reduced unit systems.

In the DPD literature, length, mass, and energy are considered primary units, leading to a derived unit of time \( \tau \) given by

\[
\tau = r_c \sqrt{\frac{m}{kT}},
\]

From the above arguments we obtain the time-scale in the rescaled system:

\[
\tau' = r'_c \sqrt{\frac{m'}{k'T'}} = \phi^{1/4} r_c \sqrt{\frac{\phi m}{\phi kT}} = \phi^{1/4} \tau.
\]

Thus, length and time scale in the same way. This also implies that the velocities are invariant as they are given by \( \Delta \mathbf{r}/\Delta t \).

The random variable \( \zeta_{ij} \) has the unit \( \tau^{-1/2} \), as noted in the discussion following Eqn. (1). Given the scaling of \( \tau \), it follows that

\[
\zeta'_{ij} = \phi^{-1/(2d)} \zeta_{ij},
\]

and therefore

\[
\sigma'_{ij} = \phi^{-1/d} \sigma_{ij}.
\]

Since the other terms in Eqn. (1) are all scale-free, the three force components of DPD all scale by a factor \( \phi^{1/4} \).

When velocity increments are calculated during one time step, one finds that the force scaling is canceled by the scaling of mass and time. This is shown below for the conservative force:

\[
[\Delta \mathbf{v}_i^c]' = \sum_{j \neq i} \frac{\Delta t' \mathbf{F}_{ij}^c}' a' m' \frac{\Delta t'}{m} \sum_{j \neq i} \chi_{ij} \left(1 - \frac{r_{ij}}{r_c'}\right) \hat{r}_{ij} = \frac{\phi^{1/4} \phi_k}{\phi m} \frac{\Delta t}{\alpha} \sum_{j \neq i} \chi_{ij} \left(1 - \frac{\phi^{1/4} d r_{ij}^c}{\phi k T} \right) \hat{r}_{ij} = \Delta \mathbf{v}_i^c.
\]

The calculations for \( [\Delta \mathbf{v}_i^d]' \) and \( [\Delta \mathbf{v}_i^R]' \) give similar re-
sults:

\[ \Delta v_i = \sum_{j \neq i} \gamma_{ij} \rho \omega D (r_{ij}) [(v_i - v_j) \cdot \hat{r}_{ij}] \Delta t \]

\[ = \sum_{j \neq i} \gamma_{ij} \rho \omega \cdot \frac{\phi^{\frac{1}{2}}}{\phi} \cdot \frac{\gamma}{m} \omega D (r_{ij}) [(v_i - v_j) \cdot \hat{r}_{ij}] \Delta t \]

\[ = \Delta v_i^D \]

\[ \Delta v_i^R = \sum_{j \neq i} \sigma_{ij} \rho \omega R (r_{ij}) \Delta t \]

\[ = \sum_{j \neq i} \sigma_{ij} \rho \omega \cdot \frac{\phi^{\frac{1}{2}}}{\phi} \cdot \frac{\gamma}{m} \omega R (r_{ij}) \Delta t \]

\[ = \Delta v_i^R \]

Since \( \Delta \vec{r} = \Delta r'/r_c \) and \( \Delta \vec{t} = \Delta t'/\tau' \), we get for the velocity increment by considering

\[ \Delta \vec{v}_i = \Delta \vec{r}_i / \Delta \vec{t}_i = \Delta v_i' / \tau' / r_c. \]

Because time and length scale in the same way we get \( \tau'/\tau' = \tau/r_c \). Combining this with \( \Delta \vec{v} = \Delta v' \) (Eqns. (31) to (33)) one finally obtains

\[ \Delta \vec{n}_i = \Delta v_i, \]

which implies

\[ \vec{r}(\ell) = \vec{r}(t). \]

What remains to be shown is the scaling of the reduced parameters \( a, \gamma, \) and \( \sigma \). Since \( a \) scales like energy over length, when going to the reduced units of the primed system, we have

\[ a = a' \cdot \frac{r_c}{k'T} = \frac{\phi^{\frac{1}{2}}}{\phi} \cdot \frac{a}{k'T} = a, \]

and similarly, since \( \gamma \) scales like energy over length and velocity, from \( \gamma = \gamma' \cdot \frac{r_c^2}{k'T'\tau'} \) we get

\[ \gamma = \gamma' \cdot \frac{r_c^2}{k'T'\tau'} = \frac{\phi^{\frac{1}{2}}}{\phi} \cdot \frac{\gamma}{k'T\tau} = \gamma. \]

From the fluctuation-dissipation relation it follows again that

\[ \sigma = \sigma. \]

Hence, scaling and unit reduction precisely cancel each other. As a result, the DPD formalism is scale-free. This means that the calculation with one and same set of parameter values represents systems at arbitrary lengths scales.

Note that in order to understand the according physical time scales we have to comment on transport properties of the method and the scaling of fluctuations. We base our argument on diffusion, but could equally well consider viscosity since the two are related by the Schmidt-number, which is dimensionless. The numerical equivalence of the measured diffusion constants \( D = D \) (being a consequence of the equality of the measured displacement) and the fact that diffusion scales like length squared over time causes an apparent problem: it seemingly implies that relative fluctuations \( D/L \) stay constant instead of vanishing. This problem disappears when one calibrates the simulation to an actual physical system. Assume that the cutoff radius is related to a physical length by \( r_c = l[cm] \). We then have

\[ D \tau^2 / \tau = D_{phys} \cdot cm^2 sec, \]

with \( D_{phys} = \sqrt{<\Delta r(t)^2>} \) referring to the diffusion constant in physical units. We get

\[ \tau = \frac{D}{D_{phys}} l^2 [sec]. \]

Consequently, expressed in physical units, it holds for the fluctuations

\[ \lim_{l \to \infty} \frac{D_{phys}}{L_{phys}} \cdot \sqrt{t} = 0. \]

IV. SUMMARY AND DISCUSSION

We have shown that each individual term of a reduced unit DPD calculation is scale-free, and hence so is DPD as a whole. We emphasize that this result was only achieved by a combined effect of scaling, the reduction of units, and finally by requiring a specific but obvious scaling of \( \gamma \) and \( \sigma \).

The scaling of energy per particle (or equivalently the number of degrees of freedom in the system) is shown to require increasing the value of \( k \) by the same factor of \( \phi \) that the number of particles is reduced by. This scaling of \( k \) is required to keep the total system energy constant. The increase in \( k \) compensates for the freezing effect seen by other authors who failed to take this into account and who also did not uniformly scale all length parameters.

In this article, we have shown that the DPD-method is scale-free for the simulation of bulk fluids. This is not necessarily the case e.g. for binary mixtures of liquids \( A \) and \( B \) where several conservative interaction parameters occur, e.g. \( a_{AA}, a_{AB}, a_{BB} \). Whereas bulk interactions given by \( a_{AA}, a_{BB} \) scale as discussed in this article, \( a_{AB} \) is a surface term that determines interfacial energy and therefore scales differently. This is, however, beyond the scope of this article.

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