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On the connection between dissipative particle dynamics and the Itô-Stratonovich dilemma

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Dissipative Particle Dynamics (DPD) is a popular simulation model for investigating hydrodynamic behavior of systems with non-negligible equilibrium thermal fluctuations. DPD employs soft core repulsive interactions between the system particles, thus allowing them to overlap. This supposedly permits relatively large integration time steps, which is an important feature for simulations on large temporal scales. In practice, however, an increase in the integration time step leads to increasingly larger systematic errors in the sampling statistics. Here, we demonstrate that the prime origin of these systematic errors is the multiplicative nature of the thermal noise term in Langevin's equation, i.e., the fact that it depends on the instantaneous coordinates of the particles. This lead to an ambiguity in the interpretation of the stochastic differential Langevin equation, known as the Itô-Stratonovich dilemma. Based on insights from previous studies of the dilemma, we propose a novel algorithm for DPD simulations exhibiting almost an order of magnitude improvement in accuracy, and nearly twice the efficiency of commonly used DPD Langevin thermostats. © *2016 AIP Publishing LLC.* [http://dx.doi.org/10.1063/1.4942114]

Dissipative Particle Dynamics (DPD)[1] is a popular method employed for simulations of diverse molecular systems including colloidal suspensions, liquid crystals, polymers, and bilayer membranes.[2–5] It is particularly appealing for investigations of multiscale phenomena since, typically, the DPD particles do not represent individual atoms, but rather coarse-grained (CG) collections of molecules. DPD was originally introduced by Hoogerbrugge and Koelman[6] as a method for simulating hydrodynamic phenomena in complex fluids. Specifically, the method targets *fluctuating hydrodynamics*, i.e., hydrodynamics at the mesoscopic scales where thermal fluctuations are important.[7,8] This is achieved in DPD simulations by considering Langevin Dynamics (LD), where each particle experiences a conservative force and, additionally, friction and random thermal forces that satisfy the fluctuation-dissipation theorem.[9] However, in *conventional* LD,[10] the dissipative force acting on the particle is given by $\vec{f}^D = -\gamma\vec{v}$, where $\vec{v}$ is the velocity of the particle and $\gamma > 0$ is a constant friction coefficient. The random force is given by $\vec{f}^R = \sqrt{2k_BT\gamma}\vec{R}(t)$, where k_B is Boltzmann's constant, T is the temperature, and $\vec{R}(t)$ is a Gaussian-distributed white noise with vanishing mean $\langle R(t)_\alpha\rangle = 0$ and memory-less auto-correlation $\langle R(t)_\alpha R(t')_\beta\rangle = \delta(t-t')\delta_{\alpha\beta}$ (α and β denote Cartesian coordinates). In contrast, in DPD, the friction and random forces act in a pairwise fashion and are directed along the line connecting the centers of the particles. This ensures that the total momentum of the system is conserved, which is essential in order to capture the correct hydrodynamic behavior of fluids at large scales.[11] Explicitly, the friction force associated with each pair of particles $i \neq j$ is given by

$$\vec{f}_{ij}^D = -\gamma\omega\left(r_{ij}\right)\left(\hat{r}_{ij}\cdot\vec{v}_{ij}\right)\hat{r}_{ij}, \tag{1}$$

where $r_{ij} = |\vec{r}_i - \vec{r}_j|$ is the pair-distance, $\hat{r}_{ij} = \left(\vec{r}_i - \vec{r}_j\right)/r_{ij}$, and $\vec{v}_{ij} = \vec{v}_i - \vec{v}_j$ is the relative velocity of the particles. The random force is given by

$$\vec{f}_{ij}^R = \sqrt{2k_BT\gamma\omega\left(r_{ij}\right)}\theta_{ij}\hat{r}_{ij}, \tag{2}$$

where θ_{ij} is Gaussian white noise satisfying $\langle\theta_{ij}(t)\rangle = 0$ and $\langle\theta_{ij}(t)\theta_{kl}(t')\rangle = \left(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}\right)\delta(t-t')$. The friction and stochastic forces are modulated with a weight function, $\omega\left(r_{ij}\right)$, that determines their range, r_c. The most commonly used weight function is[12]

$$\omega_{ij} = \begin{cases} \left(1 - \dfrac{r_{ij}}{r_c}\right)^2 & r_{ij} < r_c \\ 0 & r_{ij} \geq r_c \end{cases}. \tag{3}$$

The total friction and stochastic forces exerted on the ith particle are given by $\vec{f}_i^D = \sum_{j\neq i}\vec{f}_{ij}^D$ and $\vec{f}_i^R = \sum_{j\neq i}\vec{f}_{ij}^R$, respectively.

As noted above, the particles in DPD simulations often represent CG groups of atoms and molecules. Coarse-graining is believed to lead to effective soft repulsive pair potentials; but, obviously, one has to keep in mind that due to the softening of the effective interaction potential, some features of the simulated system may change.[13] The conservative force used in DPD simulations is usually given by[12]

$$\vec{f}_{ij}^{C} = \begin{cases} a_{ij}\left(1 - \dfrac{r_{ij}}{r_c}\right)\hat{r}_{ij} & r_{ij} < r_c \\ 0 & r_{ij} \geq r_c \end{cases}, \tag{4}$$

where $a_{ij} = a_{ji}$ are parameters determining the strength of the repulsion, and the range r_c is the same as in Eq. (3). The total conservative force acting on the ith particle is $\vec{f}_i^{C} = \sum_{j\neq i} \vec{f}_{ij}^{C}$. One of the frequently proclaimed advantages of DPD simulations is that the soft-core pair potential allows the particles to overlap and, therefore, permits relatively large integration time steps dt, which speeds up the simulations. In practice, however, it is known that all integration methods for DPD exhibit increasing artificial changes in the sampling statistics as the discretization time step is enlarged.[14] Thus, despite the enhanced numerical stability limit obtained by softening the potentials, this feature imposes severe restrictions on the size of the allowed time steps and, moreover, requires one to validate results in order to assess the statistical errors.

In this paper, we present a novel DPD integrator that, in comparison to other DPD integrators, shows considerably smaller errors in the computed averages of configurational thermodynamic quantities. The new integration method is based on an integrator recently presented by the authors (G-JF Integrator) that exhibits minimal systematic errors in the sampling statistics of conventional LD simulations,[15,16] and on insights gained by implementing the integrator to study LD in systems with spatially varying friction coefficients.[17,18] In the latter case, an ambiguity, known in the literature as the *Itô-Stratonovich dilemma*, arises about the integration of the stochastic noise term in Langevin's equation.[10,19] DPD belongs to the same class of problems of stochastic dynamics with multiplicative (state-dependent) noise, and it poses several unique complications that we address in what follows.

In order to understand the origin of the problems in numerical integration of DPD, we start by writing Langevin's equation of motion

$$m_i d\vec{v}_i = \left(\vec{f}_i^{C} + \vec{f}_i^{D} + \vec{f}_i^{R}\right) dt, \tag{5}$$

where m_i is the mass of the ith particle. The G-JF integrator preserves the fluctuation-dissipation theorem in discrete time by using the *exact* relationships,

$$\int_{t_n}^{t_{n+1}} \vec{f}_i^{D} dt = -\int_{t_n}^{t_{n+1}} \gamma \vec{v}_i dt = -\gamma\left(\vec{r}_i^{n+1} - \vec{r}_i^{n}\right), \tag{6}$$

$$\int_{t_n}^{t_{n+1}} \vec{f}_i^{R} dt = \sqrt{2k_B T\gamma}\int_{t_n}^{t_{n+1}} \vec{R}_i(t) dt = \sqrt{2k_B T\gamma dt}\,\vec{R}_i^{n+1}, \tag{7}$$

where $t^{n+1} = t^n + dt$ denotes discrete time, $\vec{r}_i^{n} = \vec{r}_i(t_n)$, and $\vec{R}_i^{n+1}$ is a vector whose coordinates are Gaussian random numbers of zero mean and unity variance. Combining Eqs. (6) and (7) with the (second order in dt) approximations used in the derivation of the Verlet algorithm[20] for molecular dynamics simulations in microcanonical ensembles, one arrives at the G-JF algorithm for constant-temperature LD simulations, which (after some rearrangement) reads

$$\vec{r}_i^{n+1} = \vec{r}_i^{n} + b\left(v_i^n dt + \frac{dt^2}{2m_i}\vec{f}_i^{n} + \frac{dt}{2m_i}\vec{R}_i^{n+1}\right), \tag{8}$$

$$\vec{v}_i^{n+1} = a\vec{v}_i^{n} + \frac{dt}{2m_i}\left(a\vec{f}_i^{n} + \vec{f}_i^{n+1}\right) + \frac{b}{m_i}\vec{R}_i^{n+1}, \tag{9}$$

where $\vec{v}_i^{n} = \vec{v}_i(t_n)$, $\vec{f}_i^{n} = \vec{f}_i^{C}(t_n)$ is the *conservative* force, and the constants

$$b = \left(1 + \frac{\gamma dt}{2m}\right)^{-1}, \; a = \left(1 - \frac{\gamma dt}{2m}\right) b. \tag{10}$$

It has been demonstrated that, unlike other integrators, the G-JF algorithm exhibits minimal changes in the configurational sampling statistics as dt is varied, up to the stability limit of the integrator.[15,16,21]

When $\gamma = \gamma(\vec{r}_i)$ depends on the coordinate of the particle, one needs to specify where along the path from $\vec{r}_i^{n}$ to $\vec{r}_i^{n+1}$, the friction coefficient used in Eqs. (7) and (10) is evaluated. This ambiguity leads to the problem known as the Itô-Stratonovich dilemma, after the conventions $\gamma_{i(\mathrm{I})}^{n+1} = \gamma\left(\vec{r}_i^{n}\right)$ of Itô,[22] and $\gamma_{i(\mathrm{S})}^{n+1} = \left[\gamma\left(\vec{r}_i^{n}\right) + \gamma\left(\vec{r}_i^{n+1}\right)\right]/2$ of Stratonovich.[23] The vast majority of the literature on this topic focuses on the overdamped (strictly non-inertial) limit of Langevin's equation [$m_i \equiv 0$ in Eq. (5)], where different conventions lead to different statistical ensembles even for infinitesimally small integration steps, $dt \to 0$. In the case of full (inertial) dynamics (i.e., when the l.h.s. of Langevin's equation does *not* vanish completely), the different conventions lead to the same statistical sampling when $dt \to 0$. However, in numerical simulations with non-vanishing time steps, the error caused by employing different conventions varies considerably from one choice to another, and this error adds to the general error caused by the integrator itself. In a previous study,[17,18] we used the accurate G-JF integrator to study LD of a single particle in a medium with space-dependent friction coefficient. We demonstrated that both Itô and Stratonovich interpretations lead to noticeable deviations that scale linearly with dt from the equilibrium Boltzmann distribution. We proposed a new interpretation that produces markedly smaller discrepancies between the computed and the correct distributions and which, moreover, shows very little sensitivity to dt (and, thus, enables larger integration time steps). The newly proposed convention for choosing the value of γ_i^{n+1} is based on the recognition that the random collision forces between the Brownian particle and the molecules of the heat bath (which are not accounted for explicitly at molecular resolution) are decomposed in Langevin's equation into two contributions. The friction term represents the mean change in the momentum of the particle due to the collisions, while the noise accounts for the Gaussian statistical fluctuations around the mean value.[24] We, therefore, consider the *deterministic* part of Langevin's equation without the random component and define $\vec{s}_i^{n+1} = \vec{r}_i^{n} + \vec{v}_i^{n} dt + \left[\vec{f}_i^{n} - \gamma\left(\vec{r}_i^{n}\right)\vec{v}_i^{n}\right]\left(dt^2/2m_i\right)$ that satisfies $\langle \vec{r}_i^{n+1} \rangle \simeq \vec{s}_i^{n+1} + O\left(dt^3\right)$. Our new convention for γ_i^{n+1} reads $\gamma_i^{n+1} = \left[\gamma\left(\vec{r}_i^{n}\right) + \gamma\left(\vec{s}_i^{n+1}\right)\right]/2$. This definition resembles the Stratonovich interpretation for γ_i^{n+1}; yet, it does not create *spurious drift*. For a detailed discussion on the spurious drift problem, we refer to Refs. 17, 18, 25, and 26 (and references therein). In short, the fact that the noise term in Eq. (5) generates the distribution of momentum changes around the mean value, implies that the r.h.s. of Eq. (7)

must satisfy

$$\langle\sqrt{2k_BT\gamma_i^{n+1}dt}\,\vec{R}_i^{n+1}\rangle = 0. \tag{11}$$

However, because $\vec{R}_i^{n+1}$ is a Gaussian random number with zero mean, condition (11) can only be fulfilled if γ_i^{n+1} and $\vec{R}_i^{n+1}$ are independent of each other, which is *not* the case with the seemingly physical Stratonovich interpretation, where γ_i^{n+1} depends on $\vec{r}_i^{n+1}$, which itself depends on $\vec{R}_i^{n+1}$. Itô's interpretation satisfies Eq. (11); however, it uses a poor estimation for γ_i^{n+1} (the initial value—completely ignoring the path of the particle) and, therefore, also fails to produce accurate statistical sampling for large dt. The newly proposed interpretation satisfies condition (11) (like Itô), but with a value of γ_i^{n+1} representing a spatial average over the ensemble of trajectories of the particle during the time step (like Stratonovich).

DPD simulations present an even more challenging task of handling multiplicative (state-dependent) noise. The complexity is mainly linked to the fact that friction and noise forces act in a pairwise fashion and that they depend on both the relative coordinates and velocities of the particles. Nevertheless, a considerably improved DPD integrator can be devised, based on insights gained from our previous investigations of the Itô-Stratonovich dilemma. In order for the fluctuation-dissipation theorem to be implemented appropriately in discrete time, it is necessary to ensure that the friction and noise forces associated with each pair act along the same direction, and they must be weighted in a manner that on one hand represents an average over the time step (*a-la* Stratonivich convention), but on the other hand independent of the random noise (in order to avoid spurious drift, *a-la* Itô). We, therefore, start by advancing the system *without random forces*, which gives the deterministic estimations for the new coordinates, $\vec{s}_i^{n+1}$, and then compute the averages

$$\begin{aligned}\vec{s}_i^{n+1/2} &\equiv \left(\vec{r}_i^n + \vec{s}_i^{n+1}\right)/2 \\ &= \vec{r}_i^n + \vec{v}_i^n\frac{dt}{2} + \left[\vec{f}_i^C\left(\vec{r}^n\right) + \vec{f}_i^D\left(\vec{r}^n,\vec{v}^n\right)\right]\frac{dt^2}{4m_i},\end{aligned} \tag{12}$$

where $\vec{r}$ and $\vec{v}$ (to be distinguished from $\vec{r}_i$ and $\vec{v}_i$) denote dependence on coordinates and velocities of all the particles. The coordinates $\vec{s}^{n+1/2}$ define the directions of the friction and noise forces within the time step, as well as the values of the friction coefficients. We, thus, continue with calculating the random forces acting on the particles

$$\vec{f}_i^{R^{n+1/2}}dt = \sum_{i\neq j}\sqrt{2k_BT\gamma\omega\left(s_{ij}^{n+1/2}\right)dt}\;\theta_{ij}^{n+1}\hat{s}_{ij}^{n+1/2}, \tag{13}$$

where $s_{ij} = |\vec{s}_i - \vec{s}_j|$ and $\hat{s}_{ij} = \left(\vec{s}_i - \vec{s}_j\right)/s_{ij}$. We note the following important technical point: In order to avoid the necessity of recalculating the list of interacting particles associated with the coordinates $\vec{s}_i^{n+1/2}$, we perform the summation in Eq. (13) [as well as in Eqs. (15) and (17) below] over the list of interacting pairs corresponding to $\vec{r}_i^n$. This excludes from the summation the pairs with $s_{ij}^{n+1/2} < r_c$, for which $r_{ij}^n \geq r_c$. The fraction of such pairs diminishes with dt and their contribution to the friction and noise forces is, anyhow, small. We have tested and verified that including them in the sum has, indeed, almost no effect on the computational results.

The calculation of the friction force poses a problem unique to DPD simulations. In conventional LD, the impulse of the friction force on each particle can be related to the displacement of the same particle [see Eq. (6)], while in DPD the displacements of all the particles are coupled. This precludes us from following the route leading to Eqs. (8) and (9) and enforces the approximation of defining the velocity

$$\vec{u}_i^{n+1/2} \equiv \vec{v}_i^n + \left[\vec{f}_i^C\left(\vec{r}^n\right) + \vec{f}_i^D\left(\vec{r}^n,\vec{v}^n\right) + \vec{f}_i^{R^{n+1/2}}\right]\frac{dt}{2m_i}, \tag{14}$$

and the associated friction forces

$$\vec{f}_i^{D^{n+1/2}} = -\sum_{i\neq j}\gamma\omega\left(s_{ij}^{n+1/2}\right)\left(\hat{s}_{ij}^{n+1/2}\cdot\vec{u}_{ij}^{n+1/2}\right)\hat{s}_{ij}^{n+1/2}, \tag{15}$$

where $\vec{u}_{ij} = \vec{u}_i - \vec{u}_j$. The new coordinates of the particles can now be computed using

$$\vec{r}_i^{n+1} = \vec{r}_i^n + \vec{v}_i^n dt + \left[\vec{f}_i^C\left(\vec{r}^n\right) + \vec{f}_i^{D^{n+1/2}} + \vec{f}_i^{R^{n+1/2}}\right]\frac{dt^2}{2m_i}. \tag{16}$$

Once the new coordinates are determined, we can calculate the change in the relative coordinates $\vec{\delta}_{ij}^{n+1/2} = \left(\vec{r}_i^{n+1} - \vec{r}_j^{n+1}\right) - \left(\vec{r}_i^n - \vec{r}_j^n\right)$, and the associated velocities $\vec{w}_{ij}^{n+1/2} \equiv \vec{\delta}_{ij}^{n+1/2}/dt$, and replace approximation (15) with

$$\vec{f}_i^{D^{n+1/2}} = -\sum_{i\neq j}\gamma\omega\left(s_{ij}^{n+1/2}\right)\left(\hat{s}_{ij}^{n+1/2}\cdot\vec{w}_{ij}^{n+1/2}\right)\hat{s}_{ij}^{n+1/2}. \tag{17}$$

We also compute the new deterministic forces, $\vec{f}_i^C\left(\vec{r}^{n+1}\right)$, and then evaluate the new velocities via

$$\vec{v}_i^{n+1} = \vec{v}_i^n + \left[\frac{\vec{f}_i^C\left(\vec{r}^n\right) + \vec{f}_i^C\left(\vec{r}^{n+1}\right)}{2} + \vec{f}_i^{D^{n+1/2}} + \vec{f}_i^{R^{n+1/2}}\right]dt. \tag{18}$$

We "close the loop" by calculating the friction forces $\vec{f}_i^D\left(\vec{r}^{n+1},\vec{v}^{n+1}\right)$ to be used at the next application of Eq. (12).

The sequence of Eqs. (12)-(18) constitutes our proposed new DPD integrator, which we term DPD-DE after the "deterministic estimation" of $\vec{s}_i^{n+1}$ in Eq. (12). To test the algorithm, we simulate a system of $N = 500$ identical particles in a cubic box of length $L = 5$ with the parameter set $r_c = 1$, $k_BT = 1$, $m_i = 1$, $a_{ij} = 25$, and $\gamma = 4.5$. This system, with the same set of parameters, has recently been used in Ref. 27 for comparison between several DPD integrators. As a benchmark, we use the DPD Velocity-Verlet (DPD-VV) method of Besold *et al.*,[28] which is implemented in several popular simulation packages. We note that in the simulations of the very same system in Ref. 27, the accuracy and efficiency of the DPD-VV algorithm were found to be almost identical to other commonly used DPD Langevin integrators such as Shardlow's splitting method.[29,30] Therefore, the DPD-VV results also allow comparison with other integration schemes for constant-temperature DPD. We also note that methods for DPD simulations with energy conservation exist (e.g., Refs. 31 and 32), but the discussion of constant-energy DPD is beyond the scope of this paper. The performance of the integrator is evaluated by measuring the mean and standard deviation of the potential energy of the system

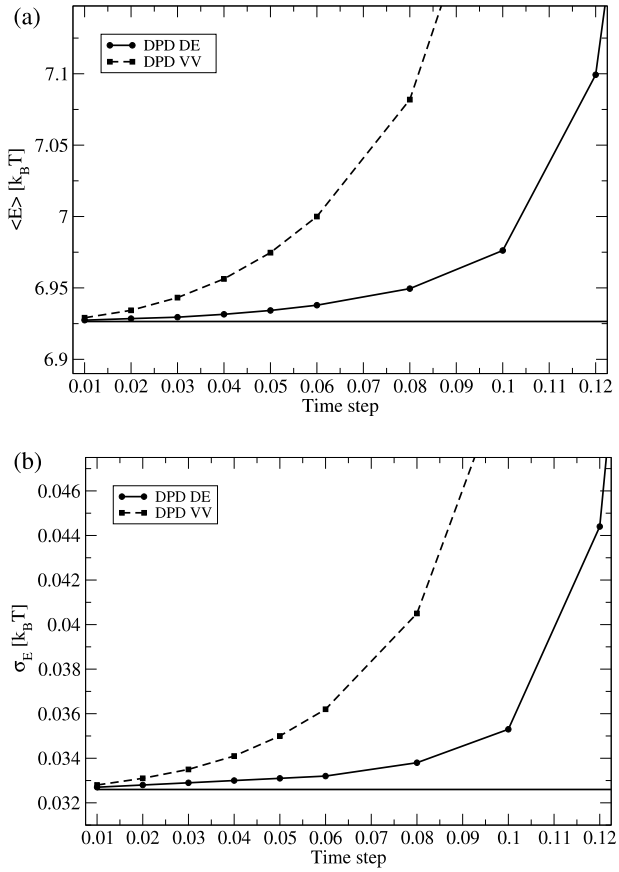

FIG. 1. Mean (a) and standard deviation (b) of the potential energy per particle, computed with different integration time steps. Results for the DPD-DE and DPD-VV methods are plotted in circles (with solid line) and squares (with dashed line), respectively. The lines serve as guides to the eye.

in simulations with increasing time steps. These quantities characterize the quality of configurational sampling. For each time step, ranging from $dt = 0.01$ and up to a time step showing significant deviations from the asymptotic $dt \to 0$ limit, we simulated the system for 1.44×10^6 time units and sampled the energy at intervals of 1.2 time units. Our results for the mean ($\langle E \rangle$) and standard deviation (σ_E) of the potential energy (normalized per particle) are plotted, respectively, in Fig. 1. We observe that both methods exhibit an increase in the measured $\langle E \rangle$ and σ_E with dt indicating unwanted changes in the sampling statistics. However, per dt, the results of the DPD-DE integrator of this work appear to be about 6-7 times more accurate (i.e., exhibiting smaller relative errors) than the results of the DPD-VV method. A similar degree of improvement in accuracy has been found in simulations of both denser and more dilute systems, and for different values of the parameter a_{ij} representing stronger/weaker repulsion between the particles.

Fig. 2 depicts the results for the simulated kinetic temperature, $T_k = (2/3)\langle K \rangle/(N-1)$ (where K is the kinetic energy), as a function of dt. One may erroneously conclude from the results for T_k that DPD-VV performs better than DPD-DE. This impression, however, is incorrect. It has been now well established (see numerous discussions on this point in, e.g., Refs. 15, 16, 21, 27, and 33) that in contrast to the potential energy, the simulated kinetic energy is not very

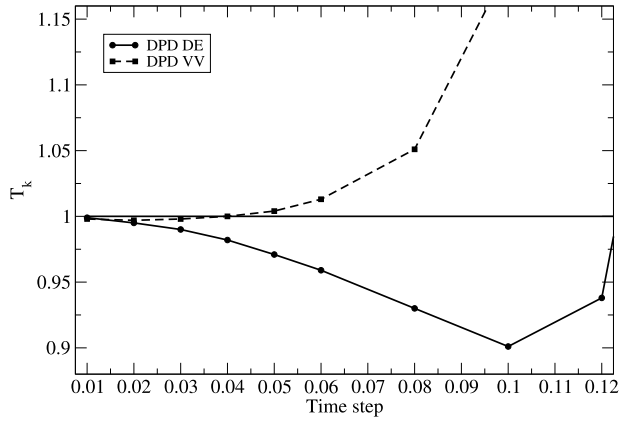

FIG. 2. The computed kinetic temperature as a function of the integration time steps. Results for the DPD-DE and DPD-VV methods are plotted in circles (with solid line) and squares (with dashed line), respectively. The lines serve as guides to the eye.

important and cannot be taken as a reliable measure for the accuracy of a simulation method. This feature of numerical integrators does not originate from the discretization of the friction and noise forces. This is an inherent property of the classic Verlet algorithm where the discrete-time momentum $\vec{v}_i^n$ is not exactly conjugated to the coordinate $\vec{r}_i^n$. For this reason, one should not attempt to use (with any integrator) quantities, such as momentum autocorrelations, for precise measures, unless very small integration time steps are applied.

To ensure that the new integrator is useful for DPD simulations, it is necessary to also demonstrate that it produces the correct dynamics, at least as accurately as other algorithms. As a measure for the dynamical evolution of the system, we consider the diffusion coefficient[14]

$$D = \lim_{t\to\infty} \frac{1}{6Nt} \left\langle \sum_{i=1}^{N} [\vec{r}_i(t) - \vec{r}_i(0)]^2 \right\rangle, \tag{19}$$

whose value depends only on the discrete-time coordinates $\{\vec{r}_i^n\}$, but not on the discrete-time momenta $\{\vec{v}_i^n\}$. Results for D as a function of dt are shown in Fig. 3. As expected, the results

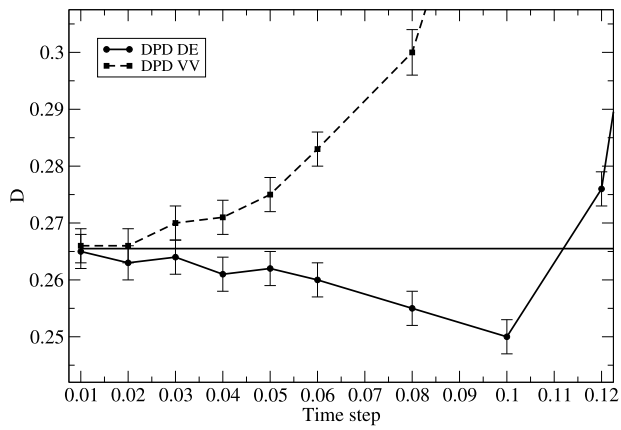

FIG. 3. The computed diffusion coefficient as a function of the integration time steps. Results for the DPD-DE and DPD-VV methods are plotted in circles (with solid line) and squares (with dashed line), respectively. The lines serve as guides to the eye.

of both integrators converge to the same limit when $dt \to 0$, indicating convergence to the correct dynamical behavior. Interestingly, the trends in the variations of D resemble the trends in T_k (Fig. 2). Also noticeable, the discretization time errors of the DPD-DE algorithm are always smaller the errors of the DPD-VV algorithm. The last observation suggests that the new DPD-DE method improves not only configurational sampling but also provide a better dynamical description of DPD systems.

The improvement by a factor of 6-7 in configurational sampling accuracy is outstanding considering that, per dt, all currently available Langevin thermostats for DPD simulations exhibit relative errors essentially identical to the one of the DPD-VV method.[27] This property suggests that the main source of numerical error in Langevin DPD thermostats is the application of Itô's interpretation to the friction coefficients, which is the common feature of all of these methods. The DPD-DE integrator of this work uses a different convention, which is based on spatial averaging of the friction along the trajectory that the particle would follow had the random noise force been turned off.[34] This new convention differs from the *seemingly more physical* Stratonovich convention that is based on the actual trajectory of the particle, and which also takes into account the influence of the random force along the trajectory. The Stratonovich interpretation represents an incorrect reading of Langevin's differential equation. In Langevin's equation, the friction force represents the mean change in the momentum of a particle, while the noise term accounts for the statistical distribution *around* the mean value. The Stratonivich interpretation "mixes" the two terms and, therefore, it leads to spurious drift.[17,18]

We close by noting that in order to assess the computational efficiency of integrators, one also needs to take into account the CPU time required to perform a single time step. For that purpose, we adopt the criterion suggested in Ref. 27, which defines the numerical efficiency as the step size giving the same relative accuracy as the DPD-VV method with step size $dt = 0.05$, divided by the CPU time. From Fig. 1 we read that the DPD-DE method with $dt = 0.1$ has the same accuracy as the DPD-VV method with $dt = 0.05$. Simulations on several different machines also reveal that the run time of DPD-DE is about 1.15-1.3 larger than that of DPD-VV.[35] Thus, the scaled efficiency of DPD-DE is about 155%–175%, placing it second in the list of integrators examined in Ref. 27 in terms of computational efficiency, just an inch behind the method that came first with scaled efficiency of 187%. However, the latter method, as well as *all* other integration methods ranked at the top places of the list, is based on a Nosé-Hoover thermostat. Such methods are more complicated for implementation, and their optimization requires fine-tuning of additional friction parameters. In contrast, DPD-DE is a *pure* Langevin thermostat having only a single tunable friction parameter γ [see Eqs. (1) and (2)]. It, thus, offers both ease of implementation and benefit of accuracy.

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