A stochastic Trotter integration scheme for dissipative particle dynamics

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Abstract

In this article we show in detail the derivation of an integration scheme for the dissipative particle dynamic model (DPD) using the stochastic Trotter formula [1]. We explain some subtleties due to the stochastic character of the equations and exploit analyticity in some interesting parts of the dynamics. The DPD-Trotter integrator demonstrates the inexistence of spurious spatial correlations in the radial distribution function for an ideal gas equation of state. We also compare our numerical integrator to other available DPD integration schemes.

Key words: Trotter stochastic formula, dissipative particle dynamics
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1 Introduction

Mesoscopic models require the use of stochastic differential equations (SDEs) to include the effects of thermal fluctuations so the selection of an efficient stochastic integration scheme is crucial to simulate correctly these systems. In this article we focus on dissipative particle dynamics (DPD) [2,3] as one of the most simple and widely used model. Although nowadays the conventional DPD model has a good theoretical basis, in the past few years there has been quite a controversy reported in literature about practical aspects of the simulations: the appearance of spurious effects related to time discretization, in concrete, the unphysical systematic drift of the temperature from the value predicted by the fluctuation-dissipation theorem and uncontrolled spatial correlations

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among particles. This is the reason of the increasing interest in developing good integrator methods for the DPD model. Several authors [4–6] have considered improvements to the basic stochastic Euler scheme through the use of solvers that have been successfully employed for *deterministic* dynamical systems in molecular dynamics (MD) simulations [7] such as the velocity Verlet algorithm (DPD-VV [4]).

Recently in [1] we investigated the applicability of the Trotter formula (widely used in molecular simulations) to a general SDEs and discuss the optimum way to split the dissipative-stochastic generators. It resulted that the Trotter formula cannot be applied without considering the special stochastic character of the equations. In general, different variables depending on the same noise should not be split. In the DPD equations this does not happen which allowed us to write a integration scheme. In [1] we concluded that, considering the accuracy of the equilibrium temperature and the computational cost, DPD-Trotter is among the best integrators for the DPD equations.

In this article we explain in detail how to apply the stochastic Trotter formula to the particular case of DPD. The aim is to furnish a non-trivial example to be used as a reference when one wishes to derive new integration schemes based on the stochastic Trotter formula for a general set of SDEs. We also test the behavior of the radial distribution function in the DPD model. For an ideal gas equation of state we find the DPD-Trotter scheme presents no spatial correlations at any scale.

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The DPD model consists of a set of N particles moving in continuous space. Each particle k is defined by its position $\mathbf{r}_k$ and its momentum $\mathbf{p}_k$ and mass m. The dynamics is specified by a set of Langevin equations very similar to the molecular dynamics equations, but where in addition to the conservative forces there are dissipative and fluctuating forces as well

$$d\mathbf{r}_k = \frac{\mathbf{p}_k}{m}dt,$$
$$d\mathbf{p}_k = \sum_{l \neq k}^{N} \mathbf{e}_{kl} \left[\left(a_{kl} F_c(r_{kl}) - \frac{\gamma}{m}\omega^2(r_{kl})(\mathbf{e}_{kl} \cdot \mathbf{p}_{kl}) \right) dt + \sqrt{2\gamma k_B T_0}\omega(r_{kl})dW_{kl}^t \right],$$

$$(1)$$

where $F_c(r)$ is the conservative pair interaction force weighted by positive and symmetric parameter a_{kl}, $\mathbf{r}_{kl} = \mathbf{r}_k - \mathbf{r}_l$ is the distance between the particle k and particle l, r_{kl} its length and $\mathbf{e}_{kl} = \mathbf{r}_{kl}/r_{kl}$. The weight function ω usually has a finite range r_c. A typical selection is $\omega(r) = 1 - r/r_c$ for $r < r_c$ and $\omega(r) = 0$ for $r \geq r_c$. The conservative force is usually chosen to be of the form $F_c(r_{kl}) = w(r_{kl})$. This system has a well defined Gibbsian equilibrium

state at a temperature T_0. Because the stochastic term in conventional DPD does not depend on the momenta, note the Itô or Stratonovich interpretation are exactly equivalent (additive noise) and we can apply the standard rules of ordinary calculus formally treating dW as $''dt''$.

The global state is $\mathbf{x} = (\mathbf{r}_1, ..., \mathbf{r}_N, \mathbf{p}_1, ..., \mathbf{p}_N)$ and the SDEs (1) can be expressed as $d\mathbf{x} = \mathcal{L}[\mathbf{x}]dt$ with formal solution $\mathbf{x}(t) = \mathcal{T}e^{\mathcal{L}t}[\mathbf{x}](\mathbf{x}_0)$ where $\mathcal{T}$ is the time-ordered operator (see [8]). The global time generator is divided in two operators generating "orthogonal dynamics" $\mathcal{L} = \mathcal{L}_r + \mathcal{L}_p$, where $\mathcal{L}_r = \sum_k \mathcal{L}_r^k$ and $\mathcal{L}_p = \sum_{k,l>k} \mathcal{L}_p^{kl}$. Because in the DPD model the forces between interacting particles k and l satisfy action-reaction (Newton's third law), the momentum is locally (and totally) conserved. For each partial dynamics, the generator can be subdivided in components $\mathcal{L}_r^k = \sum_\mu \mathcal{L}_{\mathbf{r}_k^\mu}$ and $\mathcal{L}_p^{kl} = \sum_\mu \mathcal{L}_{\mathbf{p}_{kl}^\mu}$ with

$$\mathcal{L}_{\mathbf{r}_k^\mu} = \frac{\mathbf{p}_k^\mu}{m}\partial_{\mathbf{r}_k^\mu}, \qquad \mathcal{L}_{\mathbf{p}_{kl}^\mu} = \mathcal{D}_{\mathbf{p}_{kl}^\mu} + \mathcal{S}_{\mathbf{p}_{kl}^\mu}(t),$$

$$\mathcal{D}_{\mathbf{p}_{kl}^\mu} = \left[a_{kl}F_c(r_{kl}) - \frac{\gamma}{m}\omega_D(r_{kl})(\mathbf{e}_{kl}\cdot\mathbf{p}_{kl})\right]\mathbf{e}_{kl}^\mu(\partial_{\mathbf{p}_k^\mu} - \partial_{\mathbf{p}_l^\mu}),$$

$$\mathcal{S}_{\mathbf{p}_{kl}^\mu}(t) = f(t)\sqrt{2\gamma k_B T_0}\omega(r_{kl})\mathbf{e}_{kl}^\mu(\partial_{\mathbf{p}_k^\mu} - \partial_{\mathbf{p}_l^\mu}). \tag{2}$$

Note that the momentum operator has two contributions, the deterministic and the stochastic which is an explicit function of time with $f(t) = dW_{kl}^t/dt$.

The formal solution of our system $\mathbf{x}(t) = \mathcal{T}e^{\mathcal{L}t}[\mathbf{x}](\mathbf{x}_0)$ corresponds to a continuous time evolution. In order to devise any integrator scheme we must discretize the continuous time in finite steps. The continuum time propagator can be approximated by discrete time steps of size $\Delta t = t/P$, recursively applying P times the exponential operator $e^{\mathcal{L}t} \equiv \left(e^{\mathcal{L}t/P}\right)^P \approx e^{\mathcal{L}\Delta t}\cdots e^{\mathcal{L}\Delta t}$. Note that when the generator depends explicitly on time $\mathcal{L}(t) \equiv \mathcal{L}^t$, the time-ordered exponential is relevant and the recursively nested exponentials become $\mathcal{T}e^{\mathcal{L}^t t} \approx e^{\mathcal{L}^{t+P\Delta t}\Delta t}\cdots e^{\mathcal{L}^{t+2\Delta t}\Delta t}e^{\mathcal{L}^{t+\Delta t}\Delta t}$ ([8]). At this point we must provide some approximation of the discrete time propagator of a "generic" global dynamics $e^{\mathcal{L}\Delta t}$. As we have mentioned in the previous paragraph, in general the generator $\mathcal{L}$ is formed by many generators $\sum \mathcal{L}_i$ each of them corresponding to a particular dynamics i. The generalized Trotter formula (Strang [9]) generates a straightforward approximation to the time propagator exact up to second order in time

$$e^{\sum_{i=1}^M \mathcal{A}_i t} = \left(\prod_{i=M}^1 e^{\mathcal{A}_i \frac{\Delta t}{2}} \prod_{j=1}^M e^{\mathcal{A}_j \frac{\Delta t}{2}}\right)^P + O(\Delta t^3). \tag{3}$$

Because it can be performed in many possible ways, the most important practical issue to apply formula (3) is the selection of a particular splitting of the global dynamics. One reasonable criteria is to keep the minimum number of

generators and exploit analyticity for each of them whenever possible. For the stochastic equations of DPD, the splitting we propose consist in $1 + \frac{N(N-1)}{2}$ operators: the global $\mathcal{L}_r$ and a $\mathcal{L}_p^{kl}$ for each pair.

The Baker-Campbell-Haussdorff (BCH) formula reads

$$e^{\mathcal{A}} e^{\mathcal{B}} = e^{\mathcal{A}+\mathcal{B}+\frac{1}{2}[\mathcal{A},\mathcal{B}]+\frac{1}{12}[\mathcal{A},[\mathcal{A},\mathcal{B}]]+\frac{1}{12}[\mathcal{B},[\mathcal{B},\mathcal{A}]]+\cdots} \tag{4}$$

so for $[\mathcal{A},\mathcal{B}] = 0$ we have the exact formula $e^{\mathcal{A}+\mathcal{B}} = e^{\mathcal{A}} e^{\mathcal{B}} = e^{\mathcal{B}} e^{\mathcal{A}}$. The DPD position generator $\mathcal{L}_r = \sum_{k,\mu} \mathcal{L}_{r_k^\mu}$ is composed by many simple individual generators per particle and components that satisfy $[\mathcal{L}_{\mathbf{r}_k^\mu}, \mathcal{L}_{\mathbf{r}_l^\nu}] = 0$ for all particles k, l $(k \neq l)$ and components μ, ν except $\mu \neq \nu$. Therefore we can use the exact formula

$$e^{\mathcal{L}_r \Delta t} = e^{\sum_k \mathcal{L}_r^k \Delta t} \equiv \prod_{k=1}^{N} \left(\prod_{\mu=1}^{d} e^{\mathcal{L}_{\mathbf{r}_k^\mu} \Delta t} \right) \tag{5}$$

with d the dimensionality. In MD (DPD without dissipative and random forces) the momentum generator $\mathcal{L}_p = \sum_{k,\mu} \mathcal{L}_{p_{kl}^\mu}$ also satisfies $[\mathcal{L}_{\mathbf{p}_{kp}^\mu}, \mathcal{L}_{\mathbf{p}_{lq}^\nu}] = 0$ for all pairs of particles kp, lq, with $k \neq l$, $p \neq q$, $p \neq q$ and components $\mu, \nu, \mu \neq \nu$, such that the ordering of the individual-component momentum generators is absolutely irrelevant. On the contrary in DPD, the forces depend on the other components of the velocity of the particle and also on other particles velocities and the operator $e^{\mathcal{L}_p \Delta t} = e^{\sum_{k,l>k} \mathcal{L}_p^{kl} \Delta t}$ cannot be globally integrated and has to be approximated in some way. This is the reason for the splitting it in $\frac{N(N-1)}{2}$ momentum operators. Due to this splitting and formula (3), the DPD scheme is finally given by the following Trotter integrator

$$\mathbf{x}(t + \Delta t) = \left(\prod_{q=1,r>1}^{N} e^{\mathcal{L}_p^{qr} \frac{\Delta t}{2}} \right) \left(\prod_{i=1}^{N} e^{\mathcal{L}_r^i \Delta t} \right) \left(\prod_{k=N,l<N}^{1} e^{\mathcal{L}_p^{kl} \frac{\Delta t}{2}} \right) \mathbf{x}(t). \tag{6}$$

The propagator that corresponds to the generator $\mathcal{L}_{\mathbf{r}_k}^\mu$ produces the position update which is analytically given by

$$e^{\mathcal{L}_{r_k^\mu}^k \Delta t}[\mathbf{x}]: \quad r_k^\mu(t + \Delta t) = r_k^\mu(t) + \frac{p_k^\mu(t)}{m} \Delta t \tag{7}$$

because the momentum is a constant in this step of the scheme. The next step is to solve the propagator of the momenta of the interaction pair k, l (corresponding to the generator $\mathcal{L}_p^{kl}$) independently of the positions. We have mentioned before that DPD forces satisfy action-reaction, so for a particular interacting pair k, l we propose to make a change of variables from $\mathbf{p}_k, \mathbf{p}_l$ to $\mathbf{p}_k + \mathbf{p}_l$, $\mathbf{p}_{kl} = \mathbf{p}_k - \mathbf{p}_l$. The new system to solve is $d(\mathbf{p}_k + \mathbf{p}_l) = 0$ and $d\mathbf{p}_{kl} = 2d\mathbf{p}_k$. Because the positions of the particles are "frozen" at this step of the Trotter scheme, the equation for $d\mathbf{p}_{kl}$ can be solved more easily for the

projection on the radial direction $p_{kl}^e = \mathbf{p}_{kl} \cdot \mathbf{e}_{kl}$

$$dp_{kl}^e = A dt - B p_{kl}^e dt + C dW_{kl}^t, \qquad (8)$$

where $A = 2a_{kl}F_c(r_{kl})$, $B = 2\gamma/m\omega^2$ and $C = 2\sqrt{2\gamma k_B T_0}\omega$. This equation is an Ornstein-Uhlenbeck process with analytical solution [10]

$$p_{kl}^e(t) = e^{-B\Delta t} p_{kl}^e(t_0) + A \int_{t_0}^t e^{B(s-t)} ds + C \int_{t_0}^t e^{B(s-t)} dW_s, \qquad (9)$$

where $\Delta t = t - t_0$, t_0 being the initial time. The solution of (9) requires the generation of colored noise based on a numerical scheme itself. A version of the method to generate coloured noise [1,11] adapted to Eq.(9) results

$$\Delta p_{kl}^e = \left(\mathbf{p}_{kl} \cdot \mathbf{e}_{kl} - \frac{a_{kl}F_c}{\tau} \right) \left(e^{-\frac{2\Delta t}{\tau}} - 1 \right) + \sqrt{2k_B T_0 m \left(1 - e^{-\frac{4\Delta t}{\tau}} \right)} \xi^{kl}, \qquad (10)$$

where $\tau = \gamma/m\omega^2$, $\xi^{kl} = \xi^{lk}$ are normal distributed with zero mean and variance one $(N(0,1))$ and $\Delta p_{kl}^e = p_{kl}^e(t) - p_{kl}^e(t_0)$. The propagator $e^{\mathcal{L}_p^{kl}\Delta t}$ for $\mathbf{p}_k$ and $\mathbf{p}_l$ gives

$$e^{\mathcal{L}_p^{kl}\Delta t}[\mathbf{x}] : (\mathbf{p}_k^{t+\Delta t}, \mathbf{p}_l^{t+\Delta t}) = \left(\mathbf{p}_k(t) + \frac{\Delta p_{kl}^e}{2} \mathbf{e}_{kl}(t), \quad \mathbf{p}_l(t) - \frac{\Delta p_{kl}^e}{2} \mathbf{e}_{kl}(t) \right). \qquad (11)$$

So in DPD we can solve the dynamics corresponding to the generator $\mathcal{L}_p^{kl}$ (globally for all components at the same time) without the need to go to the scalar operator $\mathcal{L}_{\mathbf{p}_{kl}^\mu}$ corresponding to the coordinate μ. In practice the Trotter integration algorithm (6) consists of the following steps: for the interaction pairs k, l update the momentum half timestep according to the propagator (11) with a noise ξ^{kl}; iterate over particles k updating the position according to (7); finally, update pairs k, l in reverse order again using the propagator (11) but with new noises ξ'_{kl}. This algorithm requires the calculation of the pair-list only once per iteration and has the same complexity as a simple DPD velocity-Verlet scheme (DPD-VV [4]).

We tested in [1] this integration scheme using the open-source code *mydpd* [12] for the equilibrium temperature with $N = 4000$ particles, $\gamma = 4.5$, $k_B T_0 = 1$, $m = 1$, $r_c = 1$ in a three dimensional periodic box (L, L, L) with $L = 10$ with periodic boundary conditions. These settings give a particle density $\rho = 4$. Here, we show in left Fig.1 the radial distribution function for $a_{kl} = 0$ (corresponding to an ideal gas equation of state) and a time step $\Delta t = 0.05$. We compare the results for three methods: the velocity Verlet (DPD-VV) [4], the Shardlow scheme [13] and DPD-Trotter [1]. We find good agreement with the theoretical value 1 for Shardlow and DPD-Trotter integrators but DPD-VV is notably wrong displaying spurious spatial correlations at distances less than the finite range r_c. In right Fig.1 we show the radial distribution function for a simulation with $a_{kl} = 25$ and a time step $\Delta t = 0.01$. As we see the three methods perform very similarly.
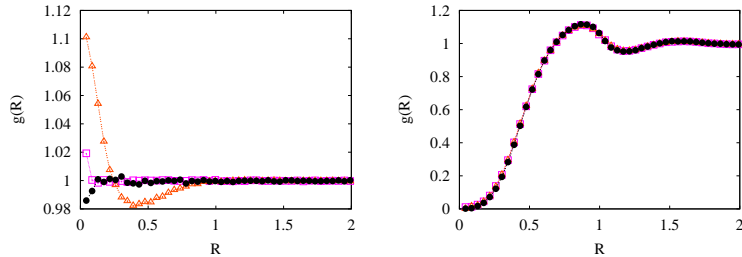
Fig. 1. Radial distribution function for three integrator methods. Velocity Verlet with ($\triangle$) symbols, Shadlow scheme with ($\square$) symbols and Trotter DPD with ($\bullet$) symbols. Left figure corresponds to an ideal gas simulation $a_{kl} = 0$. Right figure corresponds to simulations including conservative forces with $a_{kl} = 25$.

3 Conclusions

The stochastic Trotter formula can be successfully applied to the DPD model and the procedure to tailor the integrator scheme has been explained in detail. In the scheme we have also exploited the exact integration of important parts of the dynamics like the conservation of total momentum of an interacting pair of particles. The DPD-Trotter integrator displays correctly the radial distribution functions for an ideal gas (no conservative forces among particles) and also for a non ideal gas. Following this important example and [1] it should be strathforward to apply the stochastic Trotter formula to new mesoscopic models and more general SDEs.

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