SPLITTING FOR DISSIPATIVE PARTICLE DYNAMICS[*]

TONY SHARDLOW[†]

Abstract. We study numerical methods for dissipative particle dynamics, a system of stochastic differential equations for simulating particles interacting pairwise according to a soft potential at constant temperature where the total momentum is conserved. We introduce splitting methods and examine the behavior of these methods experimentally. The performance of the methods, particularly temperature control, is compared to the modified velocity Verlet method used in many previous papers.

Key words. stochastic differential equations, numerical methods, molecular dynamics

AMS subject classifications. 60H10, 82C80

PII. S1064827501392879

1. Introduction. Dissipative particle dynamics (DPD) was first introduced by Hoogerbrugge and Koelman in [9] to simulate complex hydrodynamic behavior. The technique is based on simulation of particles that evolve according to a system of SDEs (see (1) below). DPD particles should not be thought of as individual molecules; the particles represent mesoscopic groups of fluid molecules that interact at short range and according to a soft potential. This coarse graining provides a model, thought to be realistic in some regimes, that is convenient computationally, allowing for large time steps in simulations and for the exploration of long time scales. Recent applications of DPD include polymer simulation, spinodal decomposition, and suspension rheology [21, 12, 7, 4, 1]. Theoretical investigations of DPD have focused on understanding the physics (see [5, 24, 8, 18]). The issue we tackle is the numerical simulation of a DPD fluid.

DPD is described by the following system of SDEs. Consider N particles with positions $\mathbf{q}_i$ and momenta $\mathbf{p}_i$ for $i = 1, \ldots, N$ evolving in dimension d:

(1)
$$d\mathbf{q}_i = \mathbf{p}_i \, dt,$$
$$d\mathbf{p}_i = -\sum_{j \neq i} a_{ij} V'(q_{ij}) \hat{\mathbf{q}}_{ij} \, dt - \gamma \sum_{j \neq i} w^D(q_{ij})(\hat{\mathbf{q}}_{ij} \cdot \mathbf{p}_{ij}) \hat{\mathbf{q}}_{ij} \, dt + \sigma \sum_{j \neq i} w^R(q_{ij}) \hat{\mathbf{q}}_{ij} \, d\beta_{ij}(t).$$

We take the simplest case and prescribe periodic boundary conditions on the positions $\mathbf{q}_i$ in the domain $[0, L]^d$ (see [18] for a discussion of other boundary conditions). The relative positions and momenta are denoted by $\mathbf{q}_{ij}$ and $\mathbf{p}_{ij}$ and the unit direction from $\mathbf{q}_j$ to $\mathbf{q}_i$ by $\hat{\mathbf{q}}_{ij}$ and the length of $\mathbf{q}_{ij}$ by q_{ij}. We define $\hat{\mathbf{q}}_{ij} = 0$ for $q_{ij} = 0$. The pair potential

$$V(r) = \begin{cases} \frac{1}{2}(1 - r/r_c)^2, & r < r_c; \\ 0 & \text{otherwise}, \end{cases}$$

where r_c is the radius of interaction. The parameters a_{ij} are positive and symmetric ($a_{ij} = a_{ji} \geq 0$) and describe the strength of repulsion between particles i and j. For

$i < j$, β_{ij} are IID Brownian motions and for $i > j$, $\beta_{ij} = -\beta_{ji}$. We will use $\mathbf{E}$ to denote averages with respect to realizations of the Brownian motion.

The functions w^D and w^R describe the dissipative and random forces,

$$w^D(r) = w^R(r)^2 = \begin{cases} (1 - r/r_c)^2, & r < r_c; \\ 0 & \text{otherwise.} \end{cases}$$

The strength of the dissipation is parameterized by γ and of the noise by σ.

The mathematical theory of solutions of SDEs is discussed in [10], for example. A rigorous study of existence, uniqueness, and regularity is beyond the scope of the present paper. The difficulty in (1) is that neither drift nor diffusion coefficients are continuous at $q_{ij} = 0$, and this could lead to nonuniqueness of solutions (even of weak solutions) for initial data with $\mathbf{q}_{ij} = \mathbf{p}_{ij} = 0$, some $i \neq j$. This can be understood by considering

$$d\mathbf{q} = \mathbf{p}\, dt, \quad d\mathbf{p} = \hat{\mathbf{q}}\, d\beta(t), \qquad \mathbf{q}(0) = \mathbf{Q}, \quad \mathbf{p}(0) = \mathbf{P},$$

for a standard Brownian motion $\beta(t)$ and $\hat{\mathbf{q}} := \mathbf{q}/\|\mathbf{q}\|$ (where $\|\cdot\|$ denotes the standard Euclidean norm). For initial data $\mathbf{Q} = \mathbf{P} = 0$, no matter how $\hat{\mathbf{q}}$ is defined at $\mathbf{q} = 0$, the following

$$\mathbf{q}(t) = \hat{\mathbf{e}} \int_0^t \beta(s)\, ds, \qquad \mathbf{p}(t) = \hat{\mathbf{e}}\, \beta(t)$$

is a solution on a random time interval $[0, \tau]$, where $\tau > 0$ and $\hat{\mathbf{e}} \in \mathbf{R}^d$ is a unit vector. For dimension $d > 1$, this gives rise to multiple weak solutions. Even if we assume the initial data is nonzero, there is a positive probability of reaching the zero state. An extra condition is needed at $\mathbf{q} = \mathbf{p} = 0$ to specify the solution uniquely. To the author's knowledge, a rigorous analysis of this issue for DPD has not been undertaken. We assume the existence of a Markov process $(\{\mathbf{q}_i\}, \{\mathbf{p}_i\})$ on the space of configurations, which converges to the Gibbs canonical distribution (defined below) and whose generator A satisfies

$$
\begin{aligned}
A = &\sum_i \mathbf{p}_i \cdot \nabla_{\mathbf{q}_i} - \sum_{i \neq j} a_{ij} V'(q_{ij}) \hat{\mathbf{q}}_{ij} \cdot \nabla_{\mathbf{p}_i} - \gamma w^D(q_{ij})(\hat{\mathbf{q}}_{ij} \cdot \mathbf{p}_{ij}) \hat{\mathbf{q}}_{ij} \cdot \nabla_{\mathbf{p}_i} \\
&+ \tfrac{1}{2}\sigma^2 \sum_{i < j} w^R(q_{ij})^2 \hat{\mathbf{q}}_{ij}^T (\nabla_{\mathbf{p}_i} - \nabla_{\mathbf{p}_j})^2 \hat{\mathbf{q}}_{ij}
\end{aligned}
\tag{2}
$$

and has domain $\mathcal{D}(A)$, a subset of the space of continuous functions on $\mathbf{R}^{dN} \times \mathbf{R}^{dN}$ that have period L in the spatial components.

The fluctuation dissipation theorem applies to (1): Suppose that $\sigma^2 = 2\gamma k_B T$, where k_B is Boltzmann's constant and T is the equilibrium temperature. If

$$H(\{\mathbf{q}_i\}, \{\mathbf{p}_i\}) := \tfrac{1}{2} \sum_i \|\mathbf{p}_i\|^2 + \tfrac{1}{2} \sum_{i \neq j} a_{ij} V(q_{ij}),$$

then the Gibbs canonical distribution

$$\mu(\{\mathbf{q}_i\}, \{\mathbf{p}_i\}) := \frac{1}{Z} \exp\left[\frac{-H(\{\mathbf{q}_i\}, \{\mathbf{p}_i - \mathbf{p}\})}{k_B T} \right], \tag{3}$$

where Z is chosen so that μ is the density of a probability measure and $\mathbf{p}$ is the average momentum (over the N particles). As shown in [5], μ is an invariant measure of the SDEs (1) and

$$(4) \qquad \frac{1}{d}\frac{1}{N}\mathbf{E}^\mu \sum_{i=1}^N \|\mathbf{p}_i\|^2 - \frac{\|\mathbf{p}\|^2}{d} = k_B T.$$

($\mathbf{E}^\mu$ denotes expectation with respect to μ.)

The main emphasis of this paper is numerical methods for solving (1). Many papers have used a modified version of the velocity Verlet method (defined in section 2.1) to do this [8, 5, 6, 24]. The velocity Verlet scheme itself is a method for solving the Hamiltonian equations for $H(q,p) = \frac{1}{2}p^2 + V(q)$ and consists of iterating the following for a time step Δt:

$$p_{n+\frac{1}{2}} = p_n - \tfrac{1}{2}\Delta t\, V'(q_n), \quad q_{n+1} = q_n + \Delta t\, p_{n+\frac{1}{2}}, \quad p_{n+1} = p_{n+\frac{1}{2}} - \tfrac{1}{2}\Delta t\, V'(q_{n+1}).$$

This scheme is second order, symplectic, and will conserve linear and angular momentum if this holds for the Hamiltonian system. The force function, which is the most expensive part of the iteration, is computed only once per time step. For further discussion, see, for example, [19]. The modified Verlet method accounts for the noise and dissipation terms in (1). In DPD linear momentum is conserved; angular momentum is conserved when $\mathbf{q}_i$ evolve on the spatial domain $\mathbf{R}^d$ rather than the periodic domain $[0,L]^d$. The modified Verlet scheme inherits these properties. The method gives satisfactory results in many applications, but the time step must be small for the method to be stable. This becomes a more severe problem when the dissipation parameter γ or the density of particles is large. Marsh and Yeomans [13] discuss this issue in some detail and derive critical temperatures and densities for the method to be stable.

A large density or dissipation in (1) gives rise to stiffness, which is usually dealt with by applying an implicit numerical method. This approach has been tried by Pagonabarraga, Hagen, and Frenkel [15], who investigate an implicit method but find it expensive to run. Another implicit method was attempted during the writing of the present paper, based on replacing the relative momenta $\mathbf{p}_{ij}^n$ at time step n with a semi-implicit momenta $\mathbf{p}_i^n - \mathbf{p}_j^{n+1}$. This leads to an implicit numerical method where only block diagonal matrices need be inverted. This method is cheap to run but destroys the underlying conservation of linear and angular momentum, and poor behavior is observed [22].

Splitting is a widely used and important technique for solving differential equations; one important application to the Langevin equations is described in [20]. The main contribution of this paper is an implicit method based on splitting the vector field into a sum of conservative terms and pairwise fluctuation-dissipation terms. We solve the conservative system defined by the Hamilitonian H (and the boundary conditions) using the velocity Verlet method. Following this, we solve an SDE for the fluctuation and dissipation between particles i and j. We choose an implicit method that conserves the invariance of the momenta and that may be solved efficiently. This is possible because of the simple structure of the pair equations. Looping over all possible pairs, we can find an approximation for the equation as a whole. There are a number of ways of combining the solvers for the split systems.

To introduce the splitting method, we restrict attention to weak convergence. That is, for a functional ϕ (such as temperature) on the set of configurations $\{\mathbf{q}_i\}, \{\mathbf{p}_i\}$,

we want to generate approximations $\{\mathbf{q}_i^n\}$, $\{\mathbf{p}_i^n\}$ such that the averages (over the set of Brownian paths) of $\phi(\{\mathbf{q}_i^n\}, \{\mathbf{p}_i^n\})$ converge to the average of $\phi(\{\mathbf{q}_i(n\Delta t)\}, \{\mathbf{p}_i(n\Delta t)\})$. Weak convergence is of most interest in molecular dynamics and gives convergence of approximations to time correlations and, under ergodic assumptions, to averages with respect to the canonical distribution.

For weak convergence, we approximate the action of the semigroup, e^{At}, with generator A on the functional ϕ. This allows us to introduce splitting methods by writing the generator $A = A_1 + A_2$. We will consider first order splitting,

$$e^{An\Delta t} = (e^{A_1\Delta t} e^{A_2\Delta t})^n + \mathcal{O}(n\Delta t^2),$$

and the following second order splitting,

$$e^{An\Delta t} = (e^{A_1\Delta t/2} e^{A_2\Delta t} e^{A_1\Delta t/2})^n + \mathcal{O}(n\Delta t^3).$$

The first order splitting was introduced by Trotter; the second order splitting was introduced by Strang [23] for numerical approximation. Numerical analysts exploit these relations by approximating $e^{A_1\Delta t}$, $e^{A_2\Delta t}$ separately and using the above relations to generate a consistent approximation to $e^{A\Delta t}$. The splitting formula may be applied recursively to deal with a generator A split into multiple components. A rigorous statement of the convergence properties depends on the regularity of the test function ϕ and the generators $A_{1,2}$ and is not described herewith. We do define split methods for (1) in section 2.2 and demonstrate that the methods are valuable for DPD. It is not trivial to establish regularity of the generator in (1), because the diffusion is not uniformly elliptic nor are the coefficients smooth. Further discussion of splitting methods for SDEs includes [17, 3, 14].

DPD is used to simulate fluids in thermal equilibrium, where the solution of (1) is evolving in the canonical distribution (3). The temperature with respect to the canonical distribution is known and this provides a convenient way to evaluate the numerical methods. Assuming ergodicity of DPD, we expect time averages of the temperature to converge to the average given in (4). To evaluate the methods, we compute the time average

$$(5) \qquad \frac{1}{d} \frac{1}{N-1} \frac{1}{T_1 - T_0} \sum_{n=\lfloor T_0/\Delta t \rfloor}^{\lfloor T_1/\Delta t \rfloor} \sum_{i=1}^{N} \|\mathbf{p}_i^n\|^2 \Delta t,$$

where T_0 is chosen to allow the system to reach equilibrium and T_1 is chosen large to reduce the variance in the result. Notice that the average is computed by dividing by $N-1$ (the number of degrees of freedom) rather than N (the number of particles). We will take initial conditions with zero total linear momentum, in which case the computed temperature (5) will be compared to $k_B T$.

After defining the methods in section 2, results are presented in section 3 for parameter values that test the modified Verlet scheme with large density and dissipation. The results indicate the advantage of using the first order splitting method: 1% temperature control is achieved with $\Delta t = 0.04$ in section 3.1 for the first order splitting scheme whilst the modified Verlet method needs $\Delta t = 0.02$. Though the modified Verlet method is less computationally expensive per time step, it is faster to use the splitting technique to get 1% temperature control (the first order splitting method with $\Delta t = 0.04$ is 40% faster than the modified Verlet method with $\Delta t = 0.02$). The improvements are more dramatic when the dissipation (see section 3.2) or density is increased (see section 3.3).

Temperature control is a convenient way to evaluate the methods, because we know the correct value when in equilibrium, but it does not describe the physical system completely. The velocity autocorrelation function

$$(6) \qquad \frac{1}{d(N-1)} \mathbf{E} \sum_i (\mathbf{p}_i(t) - \mathbf{p}) \cdot (\mathbf{p}_i(0) - \mathbf{p})$$

and the radial distribution function (the density of the average number of particles distance r from a fixed particle normalized by the density under the uniform distribution with the same density of particles) are two important averages that should be reproduced correctly by the numerical methods. In section 3, we describe computations of velocity autocorrelation and radial distribution functions. The computations indicate agreement between the methods, with much poorer behavior for the modified Verlet method as the dissipation is increased.

The computations for the second order splitting show good behavior, but there is no evidence that the results are substantially better than the first order splitting method. This may indicate that the regularity of (1) is insufficient to gain second order convergence.

In section 4 we go some way to explain the stability properties of the splitting method of section 2.2 by generalizing an argument used by [16, 2] for the approximation to the Langevin equations

$$dq = p \, dt,$$
$$dp = -\gamma p \, dt + \sigma \, d\beta(t)$$

given by the Bruenger, Brooks, Karplus (BBK) method

$$p^{n+1/2} = p^n - \tfrac{1}{2}\gamma p^n \Delta t + \tfrac{1}{2}\sigma\beta^n(\Delta t),$$
$$q^{n+1} = q^n + p^{n+1/2}\Delta t,$$
$$p^{n+1} = p^{n+1/2} - \tfrac{1}{2}\gamma p^{n+1}\Delta t + \tfrac{1}{2}\sigma\beta^n(\Delta t),$$

where $\beta^n(\Delta t) := \beta((n+1)\Delta t) - \beta(n\Delta t)$. It is not hard to verify in this case that $\mathbf{E}p_n^2$ converges to $\sigma^2/2\gamma$, the correct equilibrium temperature, as $n \to \infty$. It would be inefficient to apply the BBK method to the full DPD system because of the structure of the linear equations. We choose a generalization of this approximation to solve the linear SDE for (i,j) fluctuation-dissipation equations. The behavior of the temperature in the numerical solution is discussed in section 4. We describe a sense in which the average temperature of the pair of particles i,j will approach that given by (4) with $\mathbf{p} = (\mathbf{p}_i + \mathbf{p}_j)/2$.

2. The numerical methods. Let Δt denote the time step and define

$$f_i^D(\{\mathbf{q}_i\}, \{\mathbf{p}_i\}) := -\gamma \sum_{j \neq i} w^D(q_{ij})(\hat{\mathbf{q}}_{ij} \cdot \mathbf{p}_{ij})\hat{\mathbf{q}}_{ij}\Delta t,$$
$$f_i^C(\{\mathbf{q}_i\}) := -\sum_{j \neq i} a_{ij}V'(q_{ij})\hat{\mathbf{q}}_{ij}\Delta t,$$
$$f_i^{R,n}(\{\mathbf{q}_i\}) := \sigma \sum_{j \neq i} w^R(q_{ij})\hat{\mathbf{q}}_{ij}W_{ij}^n(\Delta t).$$

$W_{ij}^n(\Delta t)$ for $n = 0, 1, 2, \ldots$ are normally distributed random variables with mean zero and variance Δt, with $W_{ij}^n(\Delta t)$ independent for $i < j$ and $n = 0, 1, \ldots$ and with

$W_{ij}^n(\Delta t) = -W_{ji}^n(\Delta t)$. We again use $\mathbf{E}$ to denote averages with respect to realizations of the $W_{ij}^n(\Delta t)$.

In practise, for those only interested in average properties, it is sufficient to generate $W_{ij}^n(\Delta t)$ that satisfy certain moment conditions; see [11]. For instance, for second order numerical methods, it is convenient to generate $W_{ij}^n(\Delta t)$ such that for $i < j$, $W_{ij}^n(\Delta t)$ are IID with

$$\mathbf{P}(W_{ij}^n(\Delta t) = \pm\sqrt{3\Delta t}) = \frac{1}{6}, \qquad \mathbf{P}(W_{ij}^n(\Delta t) = 0) = \frac{2}{3},$$

and $W_{ij}^n(\Delta t) = -W_{ji}^n(\Delta t)$ for $i > j$. The computations in section 3 use this rule to generate $W_{ij}^n(\Delta t)$.

2.1. Modified Verlet method. Consider initial conditions $\mathbf{q}_i^0 = \mathbf{Q}_i$ and $\mathbf{p}_i^0 = \mathbf{p}_i^{-1/2} = \mathbf{P}_i$. The modified Verlet method is the following iteration:

$$\mathbf{p}_i^{n+1/2} = \mathbf{p}_i^n + \tfrac{1}{2}\Big[f_i^D(\{\mathbf{q}_i^n\}, \{\mathbf{p}_i^{n-1/2}\}) + f_i^C(\{\mathbf{q}_i^n\}) + f_i^{R,n}(\{\mathbf{q}_i^n\})\Big],$$

$$\mathbf{q}_i^{n+1} = \mathbf{q}_i^n + \Delta t\mathbf{p}_i^{n+1/2},$$

$$\mathbf{p}_i^{n+1} = \mathbf{p}_i^{n+1/2} + \tfrac{1}{2}\Big[f_i^D(\{\mathbf{q}_i^{n+1}\}, \{\mathbf{p}_i^{n+1/2}\}) + f_i^C(\{\mathbf{q}_i^{n+1}\}) + f_i^{R,n}(\{\mathbf{q}_i^{n+1}\})\Big].$$

The algorithm is described in detail in Groot and Warren [8]. The method conserves linear and angular momentum. Without the random or dissipative forces, the algorithm is second order, but the dissipative term in the above algorithm introduces errors at order Δt^2.

By defining $\hat{\mathbf{q}} = 0$ at $\mathbf{q} = 0$, we see that $\mathbf{q}_{ij}^n = \mathbf{p}_{ij}^n = 0$ if and only if $\mathbf{q}_{ij}^{n+1} = \mathbf{p}_{ij}^{n+1} = 0$, for some $i \neq j$. This property guarantees that particles do not coalesce and the system cannot reduce to a system where $\mathbf{q}_i = \mathbf{q}, \mathbf{p}_i = \mathbf{p}$ for all i. To ensure the particles have distinct dynamics for all time, we require that the initial data obey $(\mathbf{Q}_{ij}, \mathbf{P}_{ij}) \neq 0$ for all $i \neq j$.

2.2. Splitting. We describe the splitting method for (1).

Consider the conservative terms

$$(7) \qquad \frac{d\mathbf{q}_i}{dt} = \mathbf{p}_i, \qquad \frac{d\mathbf{p}_i}{dt} = -\sum_{j \neq i} a_{ij} V'(q_{ij})\hat{\mathbf{q}}_{ij}$$

and select a numerical method for this system. Denote by $S^{C,\Delta t}(\{\mathbf{q}_i^n\}, \{\mathbf{p}_i^n\})$ the result of applying the method with step Δt to initial data $(\{\mathbf{q}_i^n\}, \{\mathbf{p}_i^n\})$.

The (i, j) fluctuation-dissipation terms are

$$(8) \qquad \begin{aligned} d\mathbf{q}_k &= 0\, dt \qquad \text{for all } k = 1, \ldots, N, \\ d\mathbf{p}_k &= 0\, dt \qquad \text{for all } k \neq i, j, \\ d\mathbf{p}_i &= -\gamma w^D(q_{ij})(\hat{\mathbf{q}}_{ij} \cdot \mathbf{p}_{ij})\hat{\mathbf{q}}_{ij}\, dt + \sigma w^R(q_{ij})\hat{\mathbf{q}}_{ij}\, d\beta_{ij}(t), \\ d\mathbf{p}_j &= -\gamma w^D(q_{ij})(\hat{\mathbf{q}}_{ij} \cdot \mathbf{p}_{ji})\hat{\mathbf{q}}_{ij}\, dt - \sigma w^R(q_{ij})\hat{\mathbf{q}}_{ij}\, d\beta_{ij}(t). \end{aligned}$$

Choose a one step method for this problem and denote the solution operator by $S^{i,j,\Delta t,n}$; that is, $S^{i,j,\Delta t,n}(\{\mathbf{q}_i^n\}, \{\mathbf{p}_i^n\})$ is the result of applying the method at time level n to initial data $(\{\mathbf{q}_i^n\}, \{\mathbf{p}_i^n\})$.

Notice that the generator of (8) is

$$\tfrac{1}{2}\sigma^2 w^R(q_{ij})^2 \hat{\mathbf{q}}_{ij}^T (\nabla_{\mathbf{p}_i} - \nabla_{\mathbf{p}_j})^2 \hat{\mathbf{q}}_{ij} - \gamma w^D(q_{ij})(\hat{\mathbf{q}}_{ij} \cdot \mathbf{p}_{ij})\hat{\mathbf{q}}_{ij} \cdot (\nabla_{\mathbf{p}_i} - \nabla_{\mathbf{p}_j})$$

and of (7) is

$$\sum_i \mathbf{p}_i \nabla_{\mathbf{q}_i} - \sum_{i \neq j} a_{ij} V'(q_{ij}) \hat{\mathbf{q}}_{ij} \cdot \nabla_{\mathbf{p}_i}.$$

The sum of the generators of (7) and of (8) for $i < j$ equals the generator of the DPD equations (2). If $S^{C,\Delta t}$ and $S^{ij,\Delta t}$ weakly converge with rate Δt, the following

(9) $$S^{C,\Delta t} \circ S^{1,1,\Delta t,n} \circ S^{1,2,\Delta t,n} \circ \cdots \circ S^{N-1,N,\Delta t,n}$$

will generate an order Δt weak approximation. (Regularity of the densities would need to be established before this could be made rigorous.) Moreover, if the methods converge weakly with rate Δt^2, we expect the following

(10)
$$S^{N-1,N,\frac{\Delta t}{2},n} \circ \cdots \circ S^{1,2,\frac{\Delta t}{2},n} \circ S^{1,1,\frac{\Delta t}{2},n} \circ S^{C,\Delta t} \circ S^{1,1,\frac{\Delta t}{2},n} \circ S^{1,2,\frac{\Delta t}{2},n} \circ \cdots \circ S^{N-1,N,\frac{\Delta t}{2},n}$$

to weakly converge with rate Δt^2 (again depending on regularity of the underlying process).

Choose $S^{C,\Delta t}$ to be the Verlet method and $S^{ij,\Delta t,n}$ to be the BBK method. That is,

(i) let $S^{C,\Delta t}(\{\mathbf{q}_i^n\}, \{\mathbf{p}_i^n\}) = (\{\mathbf{q}_i^{n+1}\}, \{\mathbf{p}_i^{n+1}\})$, where

$$\mathbf{p}_i^{n+1/2} = \mathbf{p}_i^n + \tfrac{1}{2} f_i^C(\{\mathbf{q}_i^n\}), \quad \mathbf{q}_i^{n+1} = \mathbf{q}_i^n + \Delta t \, \mathbf{p}_i^{n+1/2},$$

$$\mathbf{p}_i^{n+1} = \mathbf{p}_i^{n+1/2} + \tfrac{1}{2} f_i^C(\{\mathbf{q}_i^{n+1}\});$$

(ii) let $S^{i,j,\Delta t,n}(\{\mathbf{q}_i^n\}, \{\mathbf{p}_i^n\}) = (\{\mathbf{q}_i^{n+1}\}, \{\mathbf{p}_i^{n+1}\})$, where

$$\mathbf{q}_k^{n+1} = \mathbf{q}_k^n \qquad \text{for all } k,$$

$$\mathbf{p}_k^{n+1} = \mathbf{p}_k^n \qquad \text{for all } k \neq i,j,$$

$$\mathbf{p}_i^{n+1/2} = \mathbf{p}_i^n - \tfrac{1}{2}\gamma w^D(q_{ij}^n)(\hat{\mathbf{q}}_{ij}^n \cdot \mathbf{p}_{ij}^n)\hat{\mathbf{q}}_{ij}^n \Delta t + \tfrac{1}{2}\sigma w^R(q_{ij}^n)\hat{\mathbf{q}}_{ij}^n W_{ij}^n(\Delta t),$$

$$\mathbf{p}_j^{n+1/2} = \mathbf{p}_j^n + \tfrac{1}{2}\gamma w^D(q_{ij}^n)(\hat{\mathbf{q}}_{ij}^n \cdot \mathbf{p}_{ij}^n)\hat{\mathbf{q}}_{ij}^n \Delta t - \tfrac{1}{2}\sigma w^R(q_{ij}^n)\hat{\mathbf{q}}_{ij}^n W_{ij}^n(\Delta t),$$

$$\mathbf{p}_i^{n+1} = \mathbf{p}_i^{n+1/2} - \tfrac{1}{2}\gamma w^D(q_{ij}^n)(\hat{\mathbf{q}}_{ij}^n \cdot \mathbf{p}_{ij}^{n+1})\hat{\mathbf{q}}_{ij}^n \Delta t + \tfrac{1}{2}\sigma w^R(q_{ij}^n)\hat{\mathbf{q}}_{ij}^n W_{ij}^n(\Delta t),$$

$$\mathbf{p}_j^{n+1} = \mathbf{p}_j^{n+1/2} + \tfrac{1}{2}\gamma w^D(q_{ij}^n)(\hat{\mathbf{q}}_{ij}^n \cdot \mathbf{p}_{ij}^{n+1})\hat{\mathbf{q}}_{ij}^n \Delta t - \tfrac{1}{2}\sigma w^R(q_{ij}^n)\hat{\mathbf{q}}_{ij}^n W_{ij}^n(\Delta t).$$

Notice that the equations for $\mathbf{p}_i^{n+1}$ and $\mathbf{p}_j^{n+1}$ are implicit, but can be simplified considerably by using

$$\begin{pmatrix} I + \alpha \mathbf{q}\mathbf{q}^T & -\alpha \mathbf{q}\mathbf{q}^T \\ -\alpha \mathbf{q}\mathbf{q}^T & I + \alpha \mathbf{q}\mathbf{q}^T \end{pmatrix}^{-1} = \begin{pmatrix} I + \beta \mathbf{q}\mathbf{q}^T & -\beta \mathbf{q}\mathbf{q}^T \\ -\beta \mathbf{q}\mathbf{q}^T & I + \beta \mathbf{q}\mathbf{q}^T \end{pmatrix},$$

where $\beta = -\alpha/(1 + 2\alpha)$. Apply this equation with $\alpha = \tfrac{1}{2}\gamma w^D(q_{ij}^n)\Delta t$ and $\mathbf{q} = \hat{\mathbf{q}}_{ij}^n$ to get

$$\mathbf{p}_i^{n+1} = \mathbf{p}_i^{n+1/2} + \tfrac{1}{2}\sigma w^R(q_{ij}^n)\hat{\mathbf{q}}_{ij}^n W_{ij}^n(\Delta t)$$
$$- \tfrac{1}{2}\frac{\gamma w^D(q_{ij}^n)\Delta t}{1 + \gamma w^D(q_{ij}^n)\Delta t}\Big((\hat{\mathbf{q}}_{ij}^n \cdot \mathbf{p}_{ij}^{n+1/2})\hat{\mathbf{q}}_{ij}^n + \sigma w^R(q_{ij}^n)\hat{\mathbf{q}}_{ij}^n W_{ij}^n(\Delta t)\Big),$$

$$\mathbf{p}_j^{n+1} = \mathbf{p}_j^{n+1/2} - \tfrac{1}{2}\sigma w^R(q_{ij}^n)\hat{\mathbf{q}}_{ij}^n W_{ij}^n(\Delta t)$$
$$+ \tfrac{1}{2}\frac{\gamma w^D(q_{ij}^n)\Delta t}{1 + \gamma w^D(q_{ij}^n)\Delta t}\Big((\hat{\mathbf{q}}_{ij}^n \cdot \mathbf{p}_{ij}^{n+1/2})\hat{\mathbf{q}}_{ij}^n + \sigma w^R(q_{ij}^n)\hat{\mathbf{q}}_{ij}^n W_{ij}^n(\Delta t)\Big).$$
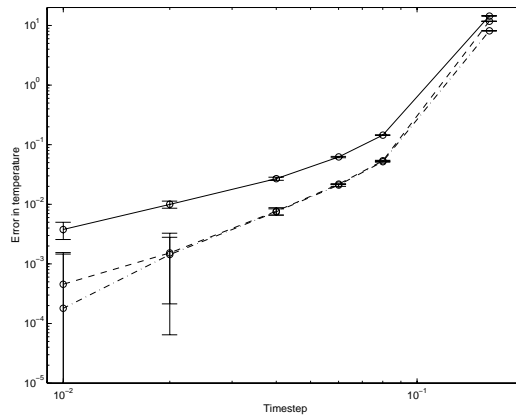
FIG. 1. *Error in temperature for $\gamma = 4.5$, $\sigma = 3$ with $L = 10$ for methods V (solid line), S1 (dashed), and S2 (dash dot).*

These equations are shown to give a weak second order approximation in the appendix (subject to regularity of the underlying SDE). It is easily checked that both methods preserve linear and angular momentum. Again, $(\mathbf{q}_{ij}^n, \mathbf{p}_{ij}^n) = 0$ if and only if $(\mathbf{q}_{ij}^{n+1}, \mathbf{p}_{ij}^{n+1})$, so that each particle has a different trajectory if the initial data satisfy $(\mathbf{Q}_{ij}, \mathbf{P}_{ij}) \neq 0$ for all $i \neq j$.

3. Computations. We evaluate three numerical methods:

V—the Verlet method of section 2.1,
S1—the first order splitting method (9) described in section 2.2,
S2—the second order splitting method (10) described in section 2.2.

For the following experiments, we take $N = 4000$ identical particles on a domain $[0, L]^3$ with the repulsion parameter $a_{ij} = 25$. The critical radius $r_c = 1$. The initial distribution $\mathbf{Q}_i$ of positions is IID uniformly distributed over $[0, L]^3$. The initial momenta $\mathbf{P}_i = \tilde{\mathbf{P}}_i - \frac{1}{N} \sum \tilde{\mathbf{P}}_i$, where $\tilde{\mathbf{P}}_i$ are IID normally distributed with mean zero and variance $k_B T$. We describe three cases with $k_B T = 1$: $(\gamma, \sigma) = (4.5, 3)$ for $L = 10$ (this case is used as a test in [8]) and $(\gamma, \sigma) = (40.5, 9)$ for $L = 10$ (increasing the dissipation) and $(\gamma, \sigma) = (4.5, 3)$ for $L = 7$ (increasing the density).

The time averaged temperature given in (5) is compared with the average $k_B T = 1$ under the canonical distribution (3). The time averaged temperature was computed for ten different realizations of the Brownian motions. The tables in sections 3.1–3.3 give the mean (column 1) and variance (column 2) of the ten computed average temperatures for the three sets of parameter values. Figures 1, 2, and 4 plot the log of the error against the log of the time steps Δt together with 90 percent confidence intervals. Figure 3 indicates the error in using the methods for a given CPU time. The benefit in using the splitting methods, in terms of the amount of temperature control available for a given CPU time, is clearly described by Figure 3. As can be seen in Figure 2, the computed error in temperature for $\Delta t = 0.02$ and 0.01 for methods S1 and S2 is dominated by the sampling error; i.e., the error in temperature is the same magnitude as the confidence interval. This explains why the lower plot in Figure 3 is flat for very small errors.

Two radial distribution functions are plotted in Figure 5, which indicates significant errors in the approximation by method V at $\Delta t = 0.04$. The resulting error
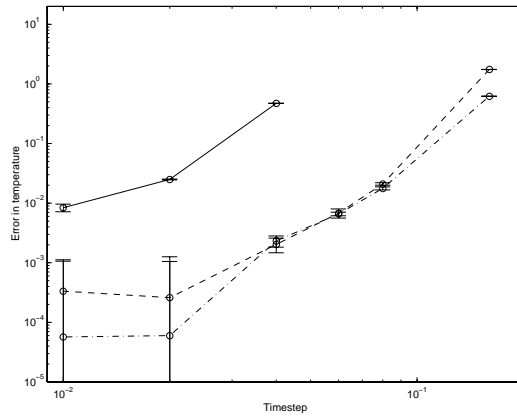
FIG. 2. *Error in temperature for $\gamma = 40.5$, $\sigma = 9$ with $L = 10$ for methods V (solid), S1 (dashed), and S2 (dash dot).*
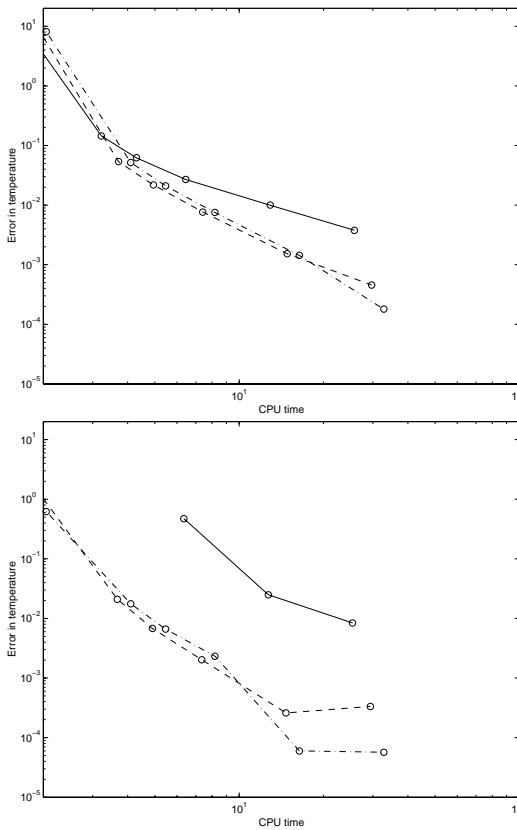

FIG. 3. *Plots of CPU time against error for the methods V (solid), S1 (dashed), and S2 (dash dot). Upper plot is $(\gamma, \sigma) = (4.5, 3)$ with $L = 10$ and lower plot is $(\gamma, \sigma) = (40.5, 9)$ with $L = 10$.*
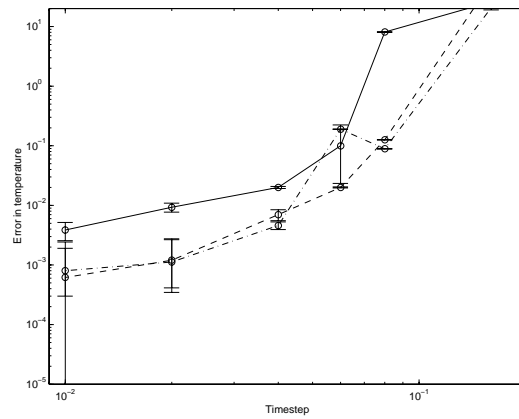
FIG. 4. *Error in temperature for $\gamma = 4.5$, $\sigma = 3$ with $L = 7$ for methods V (solid), S1 (dashed), and S2 (dash dot).*
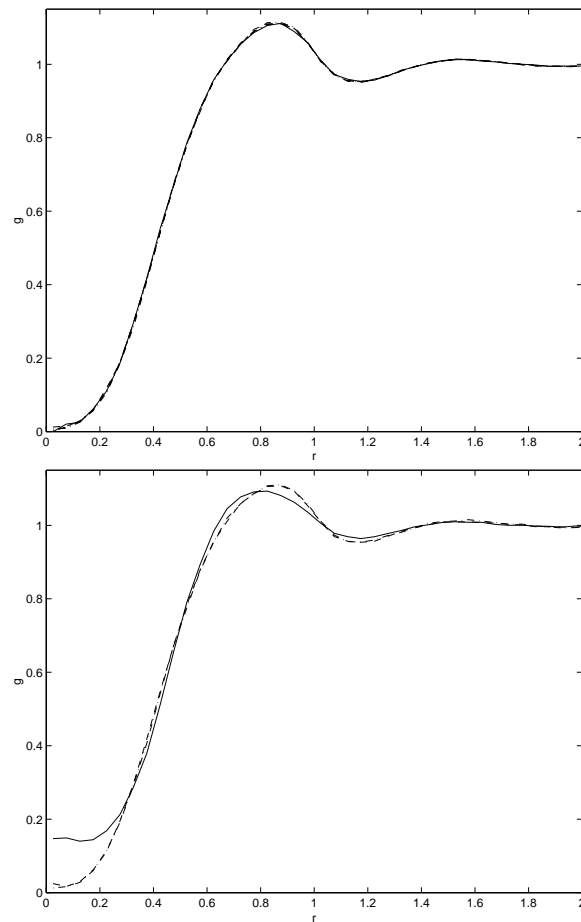

FIG. 5. *The computed radial distribution function $g(r)$ for $(\gamma, \sigma) = (4.5, 3)$ with $L = 10$ (top) and $(\gamma, \sigma) = (40.5, 9)$ with $L = 10$ (bottom) for $\Delta t = 0.04$ and methods V (solid), S1 (dashed), and S2 (dash dot).*
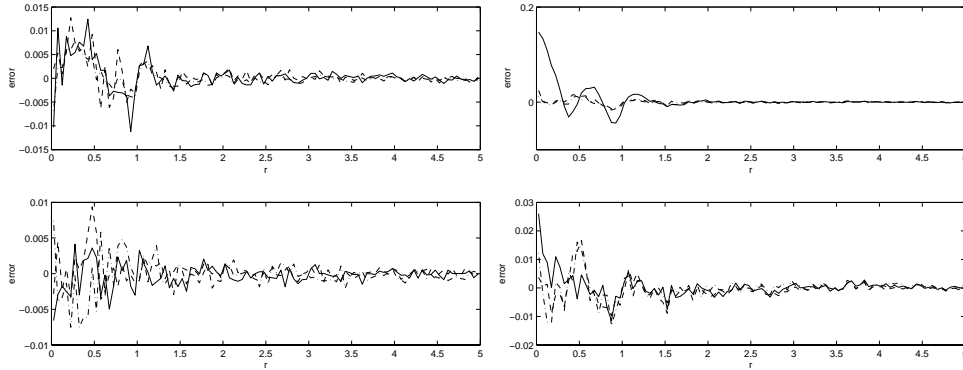
FIG. 6. *Errors in computing $g(r)$ comparing $\Delta t = 0.04$ (top) and $\Delta t = 0.02$ (bottom) with that computed with $\Delta t = 0.01$ by method $S2$. Parameter values $(\gamma, \sigma) = (4.5, 3)$ with $L = 10$ (left) and $(\gamma, \sigma) = (40.5, 9)$ with $L = 10$ (right), for methods V (solid), $S1$ (dashed), and $S2$ (dash dot).*

depends strongly on the parameter values chosen, and it is not until the dissipation is increased that the error for method V becomes significantly larger than that given by the splitting methods. The error can be seen in greater detail in Figure 6, where the computations for $\Delta t = 0.04, 0.02$ are compared to a well-resolved computation. The largest error in the computation of $g(r)$ occurs when r is small. A velocity autocorrelation function (see (6)) is plotted in Figure 7, together with comparisons of computations for $\Delta t = 0.02, 0.04$ with a well-resolved computation. Method V gives poor results for computations of the velocity autocorrelation function. Even when the equilibrium temperature has less than 2.5 percent error, the error in velocity autocorrelation may become out of control for small time.

3.1. Parameter values $(\gamma, \sigma) = (4.5, 3)$ and $L = 10$. We compute the average temperature (5) with $T_0 = 4$ and $T_1 = 200$. See Figure 1.

Δt	V		S2		S1	
0.16	15.481	1.7e-2	9.13177	8.7e-3	12.7872	1.9e-2
0.08	1.1448	8.04e-4	1.05193	6.2e-4	1.0536	7.0e-4
0.06	1.06211	9.6e-4	1.02122	6.14e-4	1.0217	1.2e-3
0.04	1.02687	9.6e-4	1.00748	5.5e-4	1.00768	6.6e-4
0.02	1.00996	8.2e-4	1.00143	8.3e-4	1.00153	1.06e-3
0.01	1.00378	7.4e-4	1.00018	7.8 e-4	0.999544	6.65e-4

The following table gives CPU times (in seconds) per unit time.

Δt	V	S2	S1
0.04	6.445	8.197	7.42

3.2. Parameter values $(\gamma, \sigma) = (40.5, 9)$ and $L = 10$. We compute the average (5) with $T_0 = 1$ and $T_1 = 100$. See also Figure 2.

Δt	V		S2		S1	
0.16	unstable		1.6229	1e-3	2.748	4e-3
0.08	unstable		1.01769	6.2e-4	1.0208	6e-4
0.06	unstable		1.00659	2.83e-4	1.0068	7.17e-4
0.04	1.47304	1.5e-3	1.00232	3.02e-4	1.00203	3.4e-4
0.02	1.02499	2.4e-4	1.00006	7.3e-4	0.99974	4.88e-4
0.01	1.0084	7.5e-4	0.999943	6.1e-4	0.99967	4.88e-4

The following table gives CPU times (in seconds) per unit time.

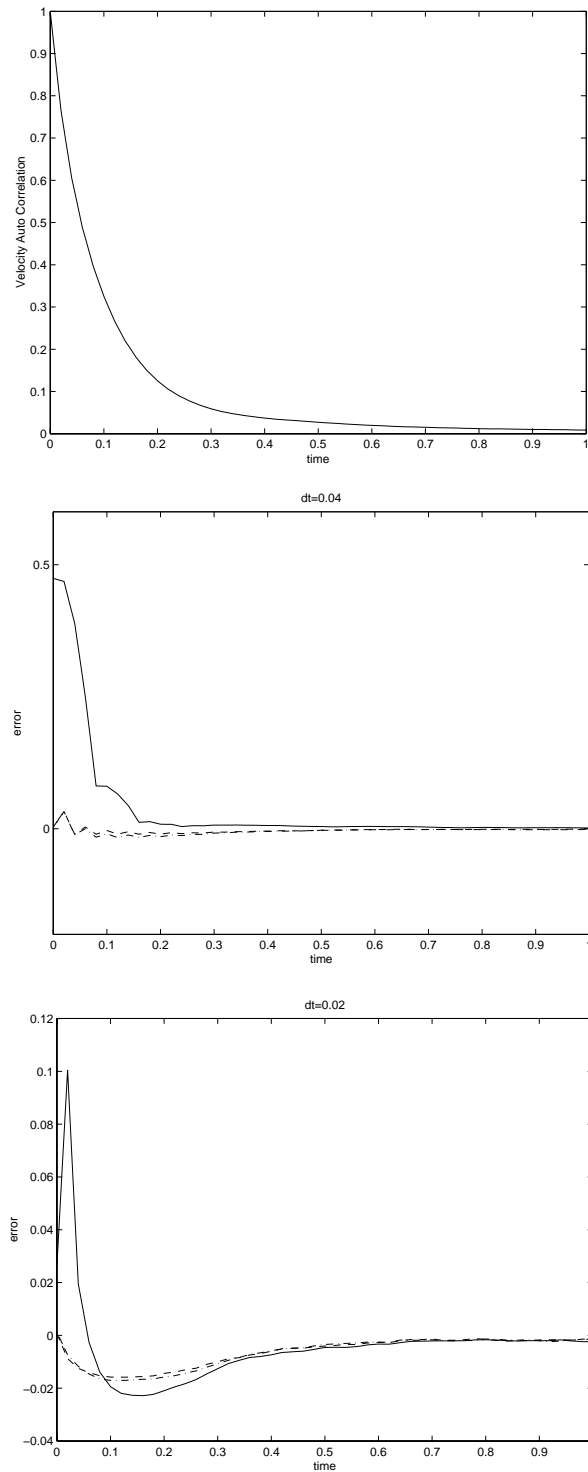| Δt	V	S2	S1
0.04	6.35	8.2	7.35

Fig. 7. *Velocity autocorrelation function for* $(\gamma, \sigma) = (40.5, 9)$ *with* $L = 10$ *computed by* $S2$ *with* $\Delta t = 0.02$ *(top). The differences in computations of the velocity autocorrelations are plotted for* $\Delta t = 0.04$ *(middle) and* $\Delta t = 0.02$ *(bottom) compared to the result with* $S2$ *with* $\Delta t = 0.01$ *for methods* V *(solid),* $S1$ *(dashed), and* $S2$ *(dash dot).*

3.3. Parameter values $(\gamma, \sigma) = (4.5, 3)$ and $L = 7$. We compute the average (5) with $T_0 = 1$ and $T_1 = 150$. See also Figure 4.

Δt	V		S2		S1	
0.16	27.1299	4.8e-2	21.809	1.1	56.06	1.01937
0.08	9.08654	8.01e-2	1.0888	3.7e-4	1.126	6.7e-4
0.06	1.10077	7.5e-2	1.01895	4.64e-4	1.02163	2.58e-4
0.04	1.0243	4.8e-4	1.0046	4.08e-4	1.00713	8.84e-4
0.02	1.00929	9.74e-4	1.00112	9.43e-4	1.00127	9.4e-4
0.01	1.00386	7.94e-4	0.9992	6.69e-4	0.999383	1.15e-3

The following table gives CPU times (in seconds) per unit time.

Δt	V	S2	S1
0.04	5.679	7.94	7.16

4. Temperature control in the splitting method. Consider the method described in section 2.2 for solving the (i, j) fluctuation-dissipation equations. We explain how the temperature of particles i and j evolves according to this method.

LEMMA 4.1. *Let* $(\{\mathbf{q}_i^{n+1}\}, \{\mathbf{p}_i^{n+1}\}) := S^{i,j,\Delta t,n}(\{\mathbf{q}_i^n\}, \{\mathbf{p}_i^n\})$; *then*

$$\mathbf{E}(\mathbf{p}_i^{n+1} \cdot \hat{\mathbf{q}}_{ij}^n)^2 + \mathbf{E}(\mathbf{p}_j^{n+1} \cdot \hat{\mathbf{q}}_{ij}^n)^2 - k_B T - \mathbf{E}(\mathbf{p}^n \cdot \hat{\mathbf{q}}_{ij}^n)^2$$
$$= \lambda\Big(\mathbf{E}(\mathbf{p}_i^n \cdot \hat{\mathbf{q}}_{ij}^n)^2 + \mathbf{E}(\mathbf{p}_j^n \cdot \hat{\mathbf{q}}_{ij}^n)^2 - k_B T - \mathbf{E}(\mathbf{p}^n \cdot \hat{\mathbf{q}}_{ij}^n)^2\Big),$$

where $\lambda := \left(\frac{1 - \gamma w^D(q_{ij})\Delta t}{1 + \gamma w^D(q_{ij})\Delta t}\right)^2$ *and* $\mathbf{p}^n := \frac{1}{2}(\mathbf{p}_i^n + \mathbf{p}_j^n)$.

This equation describes precisely how the numerical method for the (i, j) fluctuation-dissipation equations effects the temperature of the two particles. If the particles interact ($q_{ij} < r_c$), then $0 \le \lambda < 1$. Therefore, when $\mathbf{p}_i^n + \mathbf{p}_j^n = 0$ and the particles interact, the average temperature in the interparticle direction of the two particles i and j approaches $k_B T$. If $\mathbf{p}_i^n + \mathbf{p}_j^n \ne 0$ and the particles interact, the pair temperature in the interparticle direction approaches the correct equilibrium temperature given by (3) for the pair rather than for the whole ensemble of particles. The pair system cannot see the total momentum for the whole system and so will not get the equilibrium temperature correct.

Proof. First recall that if for scalars a, b and for mean zero, variance Δt IID random variables $W^n(\Delta t)$,

$$X_{n+1} = aX_n + bW^n(\Delta t),$$

then

$$X_n = a^n X_0 + b \sum_{i=0}^{n-1} a^{n-i-1} W^i(\Delta t).$$

Thus,

$$\mathbf{E}X_n^2 = a^{2n} X_0^2 + b^2 \sum_{i=0}^{n-1} a^{2(n-i-1)} \Delta t = a^{2n} X_0^2 + b^2 \frac{1 - a^{2n}}{1 - a^2} \Delta t,$$

where $\mathbf{E}$ denotes average over the realizations of $W^i(\Delta t)$. Assume $a^2 < 1$. Let $X_\infty^2 = b^2 \Delta t / (1 - a^2)$. Then

$$\mathbf{E}X_{n+1}^2 - X_\infty^2 \le \Big[\mathbf{E}X_n^2 - X_\infty^2\Big]a^2.$$

We apply this to the equation for $p_{ij}^n := \mathbf{p}_{ij}^n \cdot \hat{\mathbf{q}}_{ij}^n$ given by

$$(11) \quad p_{ij}^{n+1} = \frac{1 - \gamma w^D(q_{ij}^n)\Delta t}{1 + \gamma w^D(q_{ij}^n)\Delta t} p_{ij}^n + \sigma(w^R(q_{ij}^n) + w^R(q_{ij}^n)) \frac{1}{1 + \gamma w^D(q_{ij}^n)\Delta t} W_{ij}^n(\Delta t).$$

Now,

$$\Delta t \frac{\sigma^2(w^R(q_{ij}^n) + w^R(q_{ij}^n))^2}{(1 + \gamma w^D(q_{ij}^n))^2} \left(1 - \left(\frac{1 - \gamma w^D(q_{ij}^n)\Delta t}{1 + \gamma w^D(q_{ij}^n)\Delta t}\right)^2\right)^{-1}$$

$$= \frac{\Delta t \sigma^2(w^R(q_{ij}^n) + w^R(q_{ij}^n))^2}{2\gamma(w^D(q_{ij}^n) + w^D(q_{ij}^n))\Delta t + \gamma^2 \Delta t^2(w^D(q_{ij}^n)^2 - w^D(q_{ij}^n)^2)}$$

$$= 2k_B T.$$

Hence,

$$(12) \qquad \mathbf{E}(p_{ij}^{n+1})^2 - 2k_B T = \lambda(\mathbf{E}(p_{ij}^n)^2 - 2k_B T), \qquad \lambda := \left(\frac{1 - \gamma w^D(q_{ij})\Delta t}{1 + \gamma w^D(q_{ij})\Delta t}\right)^2.$$

Suppose that the total linear momentum of the two particles is zero; then $\mathbf{p}_i = -\mathbf{p}_j$ and we have

$$\mathbf{E}(\mathbf{p}_i^{n+1} \cdot \hat{\mathbf{q}}_{ij}^n)^2 + \mathbf{E}(\mathbf{p}_j^{n+1} \cdot \hat{\mathbf{q}}_{ij}^n)^2 - k_B T = \lambda\left(\mathbf{E}(\mathbf{p}_i^n \cdot \hat{\mathbf{q}}_{ij}^n)^2 + \mathbf{E}(\mathbf{p}_j^n \cdot \hat{\mathbf{q}}_{ij}^n)^2 - k_B T\right).$$

For general initial momentum, the above argument can be repeated with $\mathbf{p}_i - \mathbf{p}^n$ instead of $\mathbf{p}_i$. $\square$

5. Appendix. Consider the following SDE:

$$dq = p\, dt,$$
$$dp = f(q,p)dt + g(q)\Theta\, dW(t),$$

where $q, p \in \mathbf{R}^d$ and $f \in C^2(\mathbf{R}^d, \mathbf{R}^d; \mathbf{R}^d)$ and $g \in C^2(\mathbf{R}^d; \mathbf{R}^{d \times d})$ and Θ is a $d \times m$ matrix. $W(t)$ is a standard Brownian motion in $\mathbf{R}^m$. Note that these assumptions do not apply to the DPD equations. We show that the methods used for the (i, j) fluctuation-dissipation equations in section 2.2 are weakly second order consistent [11]. That is, we show the Itô–Taylor expansions agree to terms with coefficients 1, Δt, Δt^2, and $W(\Delta t)$ and $\Delta t W(\Delta t)$. The method can be written more simply for the above SDE as

$$q^{n+1} = q^n + p^n \Delta t + \tfrac{1}{2} f(q^n, p^n)\Delta t^2 + \tfrac{1}{2}\Delta t\, g(q^n)\Theta W^n(\Delta t),$$
$$p^{n+1} = p^n + \tfrac{1}{2}\Delta t\left[f(q^n, p^n) + f(q^{n+1}, p^{n+1})\right] + \tfrac{1}{2}\left(g(q^n) + g(q^{n+1})\right)\Theta W^n(\Delta t).$$

Expand out the terms around (q^n, p^n) and denote higher order terms by hot. First

$$g(q^{n+1}) = g(q^n) + g_q(q^n)p^n \Delta t + hot.$$

Expand $f(q^{n+1}, p^{n+1})$ around (q^n, p^n),

$$f(q^{n+1}, p^{n+1}) = f(q^n, p^n) + f_q(q^n, p^n)p^n \Delta t$$
$$+ f_p(q^n, p^n)\left[\tfrac{1}{2}\Delta t(f(q^n, p^n) + f(q^{n+1}, p^{n+1})) + \tfrac{1}{2}(g(q^n) + g(q^{n+1}))W^n(\Delta t)\right]$$
$$+ \tfrac{1}{2}(g(q^n)\Theta\Theta^T g(q^n)^T)_{ij} f_{p_i p_j}(q^n, p^n)\Delta t + hot,$$

where the sum over i, j is implied in the end term. Eliminating $f(q^{n+1}, p^{n+1})$ terms on the right-hand side,

$$
\begin{aligned}
f(q^{n+1}, p^{n+1}) = {} & f(q^n, p^n) + f_q(q^n, p^n)p^n \Delta t \\
& + f_p(q^n, p^n)\Big[f(q^n, p^n)\Delta t + (g(q^n) + g(q^{n+1}))\Theta W^n(\Delta t) \Big] \\
& + \tfrac{1}{2}(g(q^n)\Theta\Theta^T g(q^n)^T)_{ij} f_{p_i p_j}(q^n, p^n)\Delta t + hot.
\end{aligned}
$$

Consequently,

$$
\begin{aligned}
p^{n+1} = {} & p^n + \frac{\Delta t}{2}\Big[2f(q^n, p^n) + f_q(q^n, p^n)p^n \Delta t \\
& + f_p(q^n, p^n)\Big(f(q^n, p^n)\Delta t + (g(q^n) + g(q^{n+1}))\Theta W^n(\Delta t) \Big) \Big] \\
& + \tfrac{1}{2}\Delta t \tfrac{1}{2}(g(q^n)\Theta\Theta^T g(q^n)^T)_{ij} f_{p_i p_j}(q^n, p^n)\Delta t \\
& + \tfrac{1}{2}(g(q^n) + g(q^{n+1}))W^n(\Delta t) + hot \\
= {} & p^n + f(q_n, p_n)\Delta t + f_p(q^n, p^n)\, g(q^n)\Theta W_n(\Delta t) \\
& + f_p(q^n, p^n)g(q^n)\,\Theta\, W^n(\Delta t)\Delta t \\
& + \frac{\Delta t^2}{2}\Big[f_q(q^n, p^n)p^n + f_p(q^n, p^n)f(q^n, p^n) + \tfrac{1}{2}(g(q^n)\Theta\Theta^T g(q^n)^T)_{ij} f_{p_i p_j} \Big] \\
& + hot.
\end{aligned}
$$

We now compute the Itô–Taylor expansion for this problem: let $t_n = n\Delta t$, then

$$
p(t_{n+1}) = p(t_n) + \int_{t_n}^{t_{n+1}} f(q(s), p(s))\, ds + \int_{t_n}^{t_{n+1}} g(q(s))\Theta\, dW(s).
$$

Now, for $t_n \le s \le t_{n+1}$,

$$
\begin{aligned}
f(q(s), p(s)) = {} & f(q(t_n), p(t_n)) + \int_{t_n}^{s} f_q(q(s'), p(s'))p(s')\, ds' \\
& + \int_{t_n}^{s} f_p(q(s'), p(s'))\Big(f(q(s'), p(s'))\, ds' + g(q(s'))\Theta\, dW(s') \Big) \\
& + \tfrac{1}{2}\int_{t_n}^{s} [g(q(s'))\Theta\Theta^T g(q(s'))]_{ij} f_{p_i p_j}(q^n, p^n)\, ds'.
\end{aligned}
$$

Thus,

$$
\begin{aligned}
p(t_{n+1}) = {} & p(t_n) + \int_{t_n}^{t_{n+1}} f(q(t_n), p(t_n))\, ds + \int_{t_n}^{t_{n+1}}\int_{t_n}^{s} f_q(q(s'), p(s'))p(s')\, ds'\, ds \\
& + \int_{t_n}^{t_{n+1}}\int_{t_n}^{s} f_p(q(s'), p(s'))\Big(f(q(s'), p(s'))\, ds' + g(q(s'))\Theta\, dW(s') \Big)\, ds \\
& + \tfrac{1}{2}\int_{t_n}^{t_{n+1}}\int_{t_n}^{s} [g(q(s'))\Theta\Theta^T g(s'))^T]_{ij} f_{p_i p_j}(q(s'), p(s'))\, ds'\, ds \\
= {} & p(t_n) + \Delta t f(q(t_n), p(t_n)) + \tfrac{1}{2}\Delta t^2 f_q(q(t_n), p(t_n))p(t_n) \\
& + \tfrac{1}{2}\Delta t^2 f_p(q(t_n), p(t_n))f(q(t_n), p(t_n)) \\
& + \int_{t_n}^{t_{n+1}}\int_{t_n}^{s} f_p(q(t_n), p(t_n))g(q(t_n))\Theta\, dW(s')\, ds \\
& + \tfrac{1}{4}\Delta t^2 [g(q)\Theta\Theta^T g(q)^T]_{ij} f_{p_i p_j}(q(t_n), p(t_n)) + hot.
\end{aligned}
$$

In this case, we see the two expansions agree and we conclude that the method is weakly second order.

REFERENCES

[1] E. S. BOEK, P. V. COVENEY, H. N. W. LEKKERKERKER, AND P. VAN DER SCHOOT, *Simulating the rheology of dense colloidal suspensions using dissipative particle dynamics*, Phys. Rev. E (3), 55 (1997), pp. 3124–3133.

[2] A. BRUENGER, C. L. BROOKS III, AND M. KARPLUS, *Stochastic boundary conditions for molecular dynamics simulations of ST2 water*, Chem. Phys. Lett., 105 (1984), pp. 495–500.

[3] K. BURRAGE AND P. M. BURRAGE, *High strong order methods for non-commutative SDE systems and the Magnus formula*, Phys. D, 133 (1999), pp. 34–48.

[4] P. COVENEY AND K. E. NOVICK, *Computer simulations of domain growth and phase separation in two dimensional binary immiscible fluids using dissipative particle dynamics*, Phys. Rev. E (3), (1996), pp. 5134–5141.

[5] P. ESPANOL AND P. WARREN, *Statistical mechanics of dissipative particle dynamics*, Europhys. Lett., 30 (1995), pp. 191–196.

[6] J. B. GIBSON, K. CHEN, AND S. CHYNOWETH, *The equilibrium of a velocity-Verlet type algorithm for DPD with finite time steps*, Internat. J. Modern Phys. C, 10 (1999), pp. 241–261.

[7] R. D. GROOT AND T. MADDEN, *Dynamic simulation of diblock copolymer microphase separation*, J. Chem. Phys., 108 (1998), pp. 8713–8724.

[8] R. D. GROOT AND P. B. WARREN, *Dissipative particle dynamics: Bridging the gap between atomistic and mesoscopic simulation*, J. Chem. Phys., 107 (1997), pp. 4423–4435.

[9] P. J. HOOGERBRUGGE AND J. M. V. A. KOELMAN, *Simulating microscopic hydrodynamic phenomena with dissipative particle dynamics*, Europhys. Lett., 19 (1992), pp. 155–160.

[10] I. KARATZAS AND S. E. SHREVE, *Brownian Motion and Stochastic Calculus*, Springer–Verlag, New York, 1991.

[11] P. E. KLOEDEN AND E. PLATEN, *Numerical Solution of Stochastic Differential Equations*, Appl. Math. 23, Springer–Verlag, New York, 1992.

[12] Y. KONG, C. W. MANKE, W. G. MADDEN, AND A. G. SCHLIJPER, *Effect of solvent quality on the conformation and relaxation of polymers via the dissipative particle dynamics*, J. Chem. Phys., 107 (1997), pp. 592–602.

[13] C. A. MARSH AND J. M. YEOMANS, *Dissipative particle dynamics: The equilibrium for finite time steps*, Europhys. Lett., 37 (1997), pp. 511–516.

[14] T. MISAWA, *A Lie algebraic approach to numerical integration of stochastic differential equations*, SIAM J. Sci. Comput., 23 (2001), pp. 866–890.

[15] I. PAGONABARRAGA, M. H. J. HAGEN, AND D. FRENKEL, *Self-consistent dissipative particle dynamics algorithm*, Europhys. Lett., 42 (1998), pp. 377–382.

[16] R. W. PASTOR, B. R. BROOKS, AND A. SZABO, *An analysis of the accuracy of Langevin and molecular dynamics algorithms*, Molecular Phys., 65 (1988), pp. 1409–1419.

[17] W. P. PETERSEN, *A general implicit splitting for stabilizing numerical simulations of Itô stochastic differential equations*, SIAM J. Numer. Anal., 35 (1998), pp. 1439–1451.

[18] M. REVENGA, I. ZUNIGA, AND P. ESPANOL, *Boundary model in DPD*, Int. J. Mod. Phys., 9 (1998), pp. 1319–1328.

[19] J. M. SANZ-SERNA AND M. P. CALVO, *Numerical Hamiltonian Problems*, Chapman and Hall, London, 1994.

[20] T. SCHLICK, E. BARTH, AND M. MANDZIUK, *Biomolecular dynamics at long timesteps*, Annu. Rev. Biophys. Biomol. Struct., 26 (1997), pp. 181–222.

[21] A. G. SCHLIJPER, P. J. HOOGERBRUGGE, AND C. W. MANKE, *Computer simulation of dilute polymer solutions with the dissipative particle dynamics method*, J. Rheol., 39 (1995), pp. 567–579.

[22] T. SHARDLOW, *Numerical Methods for DPD*, Tech. Rep. NA-01/06, University of Durham, Durham, UK, 2001.

[23] G. STRANG, *On the construction and comparison of difference schemes*, SIAM J. Numer. Anal., 5 (1968), pp. 506–517.

[24] P. B. WARREN, *Dissipative particle dynamics*, Current Opinion in Colloid & Interface Science, 3 (1998), pp. 620–624.