An extended dissipative particle dynamics model

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Abstract. – The method of dissipative particle dynamics (DPD) was introduced by HOOGERBRUGGE & KOELMAN [8] to study meso-scale material processes. The theoretical investigation of the DPD method was initiated by ESPANOL [4] who used a Fokker-Planck formulation of the DPD method and applied the Mori-Zwanzig projection operator calculus to obtain the equations of hydrodynamics for DPD. A current limitation of DPD is that it requires a clear separation of scales between the resolved and unresolved processes. In this note, we suggest a simple extension of DPD that allows for inclusion of unresolved processes with exponentially decaying variance for any value of the decay rate. The main point of the extension is that it is as easy to implement as DPD in a numerical algorithm.

Dissipative Particle Dynamics. – Following the notation of ESPANOL [4], the standard DPD method can be formulated as a stochastic differential equation (SDE):

$$d\mathbf{r}_i = \frac{\mathbf{p}_i}{m_i}dt, \tag{1}$$

$$d\mathbf{p}_i = \left[\mathbf{F}_i - \gamma \sum_{j\neq i} \omega(r_{ij})(\mathbf{e}_{ij}\cdot\mathbf{v}_{ij})\mathbf{e}_{ij}\right]dt + \sigma\sum_{j\neq i}\omega^{1/2}(r_{ij})\mathbf{e}_{ij}dW_{ij}, \tag{2}$$

where m_i is the mass of particle i with position vector $\mathbf{r}_i = (x_i, y_i, z_i)^T$, $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$, $r_{ij} = |\mathbf{r}_i - \mathbf{r}_j|$, $\mathbf{e}_{ij} = \mathbf{r}_{ij}/r_{ij}$, $\mathbf{v}_{ij} = \mathbf{v}_i - \mathbf{v}_j$, $\mathbf{v}_i = \mathbf{p}_i/m_i$, and $\mathbf{F}_i$ is the conservative force acting on particle i. The dimensionless weight function $\omega(r)$ can be chosen in a rather arbitrary manner. However, to reproduce the constant temperature macro-canonical ensemble, the friction coefficient γ and the noise amplitude σ have to satisfy the fluctuation dissipation relation

$$\sigma = \sqrt{2k_BT\gamma},$$

where T is the temperature of the equilibrium state and k_B is Boltzmann's constant. Finally, $W_{ij}(t) = W_{ji}(t)$ are independent Wiener processes.

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Let us write the equations (1)-(2) in a more compact and general manner:

$$\begin{aligned} d\mathbf{r} &= \mathbf{M}^{-1}\mathbf{p}\,dt, && (3)\\ d\mathbf{p} &= -\nabla_{\mathbf{r}}V(\mathbf{r})\,dt - \sum_{k=1}^{K}\nabla_{\mathbf{r}}h_k(\mathbf{r})\left[\gamma\dot{h}_k(\mathbf{r})\,dt + \sigma dW_k\right], && (4)\end{aligned}$$

where $\mathbf{r}$ is the collection of the N particle position vectors $\mathbf{r}_i$, $\mathbf{p}$ is the associated momentum vector, $\mathbf{M}$ is the diagonal mass matrix, $\mathbf{v} = \mathbf{M}^{-1}\mathbf{p}$, $V(\mathbf{r})$ is the potential energy,

$$\dot{h}_k(\mathbf{r}) = \nabla_{\mathbf{r}}h_k(\mathbf{r})\cdot\mathbf{v},$$

and the functions $h_k(\mathbf{r})$ can again be chosen quite arbitrarily. It is essential, however, that the matrix $\boldsymbol{D}(\boldsymbol{r})$ is positive definite, where

$$\mathbf{D}(\mathbf{r}) = \sum_k \nabla_{\mathbf{r}}h_k(\mathbf{r})\nabla_{\mathbf{r}}h_k(\mathbf{r})^T. \qquad (5)$$

The choice

$$h_k(\mathbf{r}) = \phi(r_{ij}), \qquad \phi'(r) = \omega^{1/2}(r),$$

$k = 1,\ldots,(N-1)N/2$, in (4) leads back to the standard DPD model. However, one can also set $K = 3N$ and

$$h_i(\mathbf{r}) = x_i, \qquad h_{i+N}(\mathbf{r}) = y_i, \qquad h_{i+2N}(\mathbf{r}) = z_i,$$

$i = 1,\ldots,N$, in (4), which leads to the standard Langevin model

$$\begin{aligned} d\mathbf{r} &= \mathbf{M}^{-1}\mathbf{p}\,dt, && (6)\\ d\mathbf{p} &= -\left[\nabla_{\mathbf{r}}V(\mathbf{r}) + \gamma\mathbf{v}\right]dt + \sigma d\mathbf{W}, && (7)\end{aligned}$$

where $\mathbf{W}(t)$ is now a N-vector of independent random Wiener processes. Yet another variant of (4) is obtained in the context of the Hamiltonian particle mesh (HPM) method [5,6], which itself is an application of the classical particle-in-cell (PIC) or particle-mesh methods [1,7] to geophysical fluid dynamics (GFD) [17]. Here the functions $h_k(\mathbf{r})$ would refer to some cell averaged quantity and k would be its cell index. For example, the fluid density at a grid point $\mathbf{x}_k$ can be approximated by

$$h_k(\mathbf{r}) = \sum_{i=1}^{N} m_i\psi(|\mathbf{x}_k - \mathbf{r}_i|),$$

with $\psi(r)$ some proper shape function such as a tensor product cubic B-spline. We will come back to this application in §4.

We finally mention an application to molecular dynamics (MD) suggested by Ma & Izaguirre [11]. Here the stochastic part of the dynamics is used to stabilize long-time step methods and the functions h_k correspond to entries in the MD potential energy function. For example, if one would like to stabilize a bond stretching mode between atoms i and j, then

$$h_k(\mathbf{r}) = |\mathbf{r}_i - \mathbf{r}_j|.$$

Generalized Langevin Dynamics. – ESPANOL [4] gave a justification of the DPD equations (1)-(2) using the Mori-Zwanzig projection operator calculus [13,19] under the assumption of a time-scale separation between the memory kernel and the dynamic variables of interest. This assumption allows one to replace the memory kernel by a Dirac δ-function and to obtain a Markovian SDE model. Let us step back for a moment and consider the equations of motion for an arbitrary memory kernel, *i.e.* the generalized (non-Markovian) Langevin equations:

$$\dot{\mathbf{r}} = \mathbf{M}^{-1}\mathbf{p}, \tag{8}$$

$$\dot{\mathbf{p}} = -\nabla_{\mathbf{r}} V(\mathbf{r}) - \sum_{k=1}^{K} \nabla_{\mathbf{r}} h_k(\mathbf{r}) \left[\mathcal{K}_k(t-s)\dot{h}_k(\mathbf{r}(s))ds + U_k(t) \right], \tag{9}$$

where $\mathcal{K}_k(\tau)$ is the memory kernel and $U_k(t)$ is a stationary zero-mean Gaussian process. Its auto-covariance function satisfies the fluctuation dissipation relation

$$\mathbb{E}[U_k(t)U_k(s)] = k_B T \mathcal{K}_k(t-s).$$

It is often reasonable to assume that the unresolved process exhibits an exponentially decaying covariance in which case the associated memory kernel can be approximated by

$$\mathcal{K}_k(\tau) = \lambda_k e^{-\alpha_k|\tau|},$$

with α_k the decay rate and λ_k a scaling (coupling) parameter. If this is the case, then $U_k(t)$ is, in fact, an Ornstein-Uhlenbeck (OU) process and the integro-differential equation (8)-(9) can be reformulated as an extended Markovian SDE [10,16]. These two references also provide a derivation of such SDEs from large Hamiltonian systems subject to random initial data.

Let us assume, for simplicity, that all unresolved processes have the same parameter, *i.e.* $\alpha = \alpha_i$ and $\lambda = \lambda_i$. Then the following equivalent SDE can be derived:

$$d\mathbf{r} = \mathbf{M}^{-1}\mathbf{p}\, dt, \tag{10}$$

$$d\mathbf{p} = -\left[\nabla_{\mathbf{r}} V(\mathbf{r}) + \sum_{k=1}^{K} \nabla_{\mathbf{r}} h_k(\mathbf{r}) s_k \right] dt, \tag{11}$$

$$ds_k = -\left[\alpha s_k - \lambda \dot{h}_k(\mathbf{r}) \right] dt + \sqrt{2k_B T\lambda\alpha}\, dW_k, \qquad k = 1, \ldots, K, \tag{12}$$

where $W_k(t)$ is a Wiener process for each k. To see how this relates to the equations (8-9), consider the Ito formula (see page 44 of [14]) for $d\left(e^{\alpha t}s_k\right)$:

$$d\left(e^{\alpha t}s_k\right) = \alpha e^{\alpha t}s_k\, dt + e^{\alpha t}\, ds_k$$

which means that equation (12) may be written (after multiplying through by the integrating factor $e^{\alpha t}$) as

$$d\left(e^{\alpha t}s_k\right) = -e^{\alpha t}\lambda\dot{h}(\boldsymbol{r})dt + \sqrt{2k_B T\lambda\alpha}\, dW_k.$$

We may now integrate immediately to give

$$s_k = U_k - \lambda\int_0^t e^{\alpha(t'-t)}\dot{h}(\boldsymbol{r}(t'))\, dt', \qquad U_k = e^{-\alpha t}s_k(0) + \sqrt{2k_B T\lambda\alpha}\int_0^t e^{\alpha(t'-t)}\, dW_k(t')$$

where $s_k(0)$ is a constant randomly sampled from the $\mathcal{N}(0, \lambda k_B T)$ distribution, independently of dW_k. This is to ensure that the fluctuation dissipation relation is satisfied:

$$
\begin{aligned}
\mathbb{E}[U_k(t)U_k(u)] &= \mathbb{E}\Big[e^{-2\alpha(t+u)} s_k^2(0) + s_k(0)e^{-\alpha t}\int_0^u \sqrt{2k_BT\lambda\alpha}e^{\alpha(t'-u)}\,dW_k(t') \\
&\quad + s_k(0)e^{-\alpha u}\int_0^t \sqrt{2k_BT\lambda\alpha}e^{\alpha(t'-t)}\,dW_k(t') \\
&\quad + \int_0^t \sqrt{2k_BT\lambda\alpha}e^{\alpha(t'-t)}\,dW_k(t')\int_0^u \sqrt{2k_BT\lambda\alpha}e^{\alpha(t'-u)}\,dW_k(t')\Big] \\
&= e^{-2\alpha(t+u)}\lambda k_BT + \int_0^{\min(u,t)} 2k_BT\lambda\alpha e^{\alpha(2t'-t-u)}\,dt \qquad (13)\\
&= \lambda k_B T e^{\alpha|t-u|}
\end{aligned}
$$

where equation (13) is obtained after combining the Ito isometry with the facts that

1. $s_k(0)$ is independent of the Wiener process dW_k.
2. dW_k has independent increments.

See [14] for more details. Generalized DPD models for other Markovian memory kernels can be found following [3, 9, 12, 16].

We call the equations (10)-(12) the extended dissipative particle dynamics (EDPD) model. The standard DPD model (3)-(4) can be recovered in the limit of $\alpha \gg 1$ subject to $\lambda/\alpha = const.$, in which case equation (12) reduces to

$$s_k dt = \frac{\lambda}{\alpha}\dot{h}_k(\mathbf{r})dt + \sqrt{\frac{2k_BT\lambda}{\alpha}}dW_k$$

and we identify $\gamma = \lambda/\alpha$ and $\sigma = \sqrt{2k_BT\lambda/\alpha}$. A rigorous proof of this statement could be performed along the lines of [15].

A Time Discretization for the EDPD Model. – The following modification of the popular Störmer-Verlet method [18] suggests itself:

$$\mathbf{p}^{n+1/2} = \mathbf{p}^n - \frac{\Delta t}{2}\left[\nabla_{\mathbf{r}}V(\mathbf{r}^n) + \sum_k \nabla_{\mathbf{r}}h_k(\mathbf{r}^n)s_k^n\right], \qquad (14)$$

$$\mathbf{r}^{n+1} = \mathbf{r}^n + \Delta t\mathbf{M}^{-1}\mathbf{p}^{n+1/2}, \qquad (15)$$

$$\left(1 + \frac{\alpha_k\Delta t}{2}\right)s_k^{n+1} = \left(1 - \frac{\alpha_k\Delta t}{2}\right)s_k^n + \lambda_k\left[h_k(\mathbf{r}^{n+1}) - h_k(\mathbf{r}^n)\right] + \sqrt{2\Delta t k_BT\lambda_k\alpha_k}\,w_k^{n+1/2} \qquad (16)$$

$$\mathbf{p}^{n+1} = \mathbf{p}^{n+1/2} - \frac{\Delta t}{2}\left[\nabla_{\mathbf{r}}V(\mathbf{r}^{n+1}) + \sum_k \nabla_{\mathbf{r}}h_k(\mathbf{r}^{n+1})s_k^{n+1}\right] \qquad (17)$$

where $w_k^{n+1/2} \sim \mathcal{N}(0,1)$ are independent random variables.

For large values of α_k, it might be preferable to replace (17) by the implicit Euler approximation

$$(1 + \alpha_k\Delta t)\,s_k^{n+1} = s_k^n + \lambda_k\left[h_k(\mathbf{r}^{n+1}) - h_k(\mathbf{r}^n)\right] + \sqrt{2k_BT\lambda_k\alpha_k}\,w_k^{n+1}.$$

Note that (14)-(17) requires only one force field evaluation and one evaluation of $h_k(\mathbf{r})$ per time-step.

A Numerical Experiment. – In [5], FRANK, GOTTWALD & REICH suggested a particle-mesh method, called the Hamiltonian particle-mesh (HPM) method, for the solution of the two-dimensional rotating shallow-water equations (SWEs). The Lagrangian formulation of the SWEs is

$$\ddot{\mathbf{X}} = -f_0 \mathbf{X}^\perp - c_0^2 \nabla_{\mathbf{X}} h(\mathbf{X}),$$

where $\mathbf{X} = (X,Y)^T$ are the particle positions, $\mathbf{X}^\perp = (Y,-X)^T$. The layer-depth h is given by the convolution

$$h(\mathbf{x},t) = \int h_0(\mathbf{a})\, \delta(\mathbf{x} - \mathbf{X}(\mathbf{a},t))\, d\mathbf{a},$$

where $\mathbf{a} = \mathbf{X}(\mathbf{a},0)$ are the initial particle positions and $h_0(\mathbf{a})$ is the initial layer-depth at $\mathbf{a}$. See [5,6] for more details and [17] for the significance of the shallow-water model to GFD.

One introduces a regular grid $\mathbf{x}_{kl} = (k\,\Delta x, l\,\Delta y)^T$, particles $\mathbf{X}_i = (X_i, Y_i)^T$, grid-centred basis functions $\psi_{kl}(\mathbf{X})$, and the layer-depth approximation

$$h_{kl}(t) = h(\mathbf{x}_{kl}, t) = \sum_{i=1}^{N} m_i \psi_{kl}(\mathbf{X}_i).$$

The basis functions form a partition of unity, *i.e.* $\sum_{k,l} \psi_{kl}(\mathbf{x}) = 1$.

The finite-dimensional Hamiltonian equations of motion are now given by

$$\ddot{\mathbf{X}}_i = -f_0 \mathbf{X}_i^\perp - c_0^2 \sum_{k,l} \nabla_{\mathbf{X}_i} \psi_{kl}(\mathbf{X}_i) h_{kl}.$$

It is essential for the method to work that the basis functions $\psi_{kl}(\mathbf{x})$ provide enough smoothing. In the HPM method this is achieved by applying an inverse modified Helmholtz operator with smoothing length $\Lambda = 4\Delta x$ over the grid $\mathbf{x}_{kl}$. For further implementation details see again [5,6].

A EDPD extension of the HPM equations is straightforward:

$$\begin{aligned} d\mathbf{X}_i &= \mathbf{V}_i\, dt, && (18)\\ d\mathbf{V}_i &= -f_0 \mathbf{V}_i^\perp - c_0^2 \sum_{k,l} [h_{kl} + s_{kl}]\, \nabla_{\mathbf{X}_i} \psi_{kl}(\mathbf{X}_i)\, dt, && (19)\\ ds_{kl} &= [-\alpha s_{kl} + \lambda p_{kl}]\, dt + c_0^{-1} (2 k_B T \alpha \lambda)^{1/2} dW_{kl}, && (20) \end{aligned}$$

where p_{kl} is defined by

$$p_{kl} = \sum_i m_i \nabla_{\mathbf{X}_i} \psi_{kl}(\mathbf{X}_i) \cdot \mathbf{V}_i.$$

The only necessary modifications are (i) a scaling of the term multiplying dW_{kl} by c_0^{-1} and (ii) a sign change in (20) since the associated matrix (5) is negative semi-definite. The scaling is necessary because c_0^2 multiplies the force term in (19).

The parameter values $f_0 = 2\pi$, $c_0 = 2\pi$, a mesh-size $\Delta x = \Delta y = 2\pi/64$, and a total of $N = 65536$ particles are used in the numerical experiments. The initial conditions for $\mathbf{X}_i$ and $\mathbf{V}_i$ correspond to an unstable shear flow and are chosen as in [5]. We set $k_B T = 4 \times 10^{-4}$, $\lambda = \epsilon\, \Lambda^2$ and $\alpha = \epsilon\, \Lambda / c_0$ with ϵ as a free parameter and $\Lambda = 4\Delta x$. This choice corresponds to $\gamma = \Lambda^3/c_0$ in the standard DPD method. We also have $r_{kl}(0) \sim (k_B T \lambda)^{1/2}/c_0 \mathcal{N}(0,1)$. Experiments are conducted for values of ϵ varying between $\epsilon = 0.1, \ldots, 10$. The value $\epsilon = 1$ is the most natural choice [2]. Varying ϵ allows us the demonstrate the importance of the

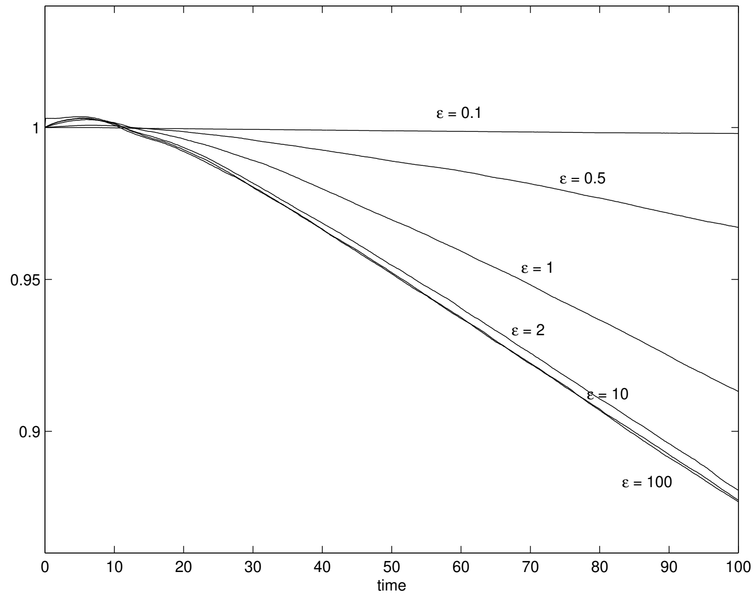

Fig. 1 – Decay of scaled energy $E(t)/E(0)$ for various values of the parameter ϵ. The value $\epsilon = 1$ is the most realistic one in the context of the shallow-water equations. The limit $\epsilon \gg 1$ corresponds to the standard DPD model.

finite decay of correlation in the EDPD model with the standard DPD model corresponding to $\epsilon \gg 1$.

One should keep in mind that the initial data for the macroscopic variables are far from thermal equilibrium. In fact, for the given initial conditions and parameters, the particles lose energy to the heat bath. This can be seen from the decay of the total particle energy

$$E = \frac{1}{2}\sum_i m_i \|\dot{\mathbf{X}}\|^2 + \frac{c_0^2}{2}\sum_{k,l} h_{kl}^2,$$

which is a conserved quantity for the HPM method. In Fig. 1, we plot the decay of the scaled energy $E(t)/E(0)$ for various values of ϵ. We note that the decay rate changes dramatically for $\epsilon \in [0.1, 2]$ while for $\epsilon \geq 10$ the energy decay rate becomes independent of ϵ. This is the regime where the standard DPD method gives essentially identical results.

Conclusion. – The proposed EDPD method allows one to take finite temporal decay of correlation into account. We have demonstrated the effect of such a modification for the two dimensional rotating shallow-water equations. The limit $\alpha \to \infty$ subject to constant $\gamma = \lambda/\alpha$ leads back to the standard DPD model.

There remains, however, the issue of possible spatial correlations in the unresolved degrees of freedom. In that respect the choice of the weight function $\omega(r)$ in (1)-(2) implies a certain spatial correlation. For example, the HPM method of §4 uses the Green's function of a modified Helmholtz operator with smoothing length Λ and the basis functions $\psi_{kl}(\mathbf{x})$ correspond in this case to a nearly exponential decay of spatial correlation [2]. Another possibility to account for spatial correlations is to modify equation (11) to

$$d\mathbf{p} = -\left[\nabla_{\mathbf{r}} V(\mathbf{r}) + \sum_{k,l=1}^{K} \nabla_{\mathbf{r}} h_k(\mathbf{r}) a_{kl} s_l\right] dt,$$

and equation (12) to

$$ds_k = -\alpha s_k\, t + \lambda \sum_{l=1}^{K} a_{kl} \dot{h}_l(\mathbf{r})\, dt + \sqrt{2k_B T \lambda\alpha}\, dB_k,$$

where the weights a_{kl} model the desired spatial correlation kernel. This approach seems particularly suited for particle-mesh methods such as the HPM method described in §4.

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