MOLECULAR PHYSICS, 2002, VOL. 100, NO. 24, 3885–3891

An impulse integrator for Langevin dynamics

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(*Received 2 January 2002; revised version accepted 10 July 2002*)

The best simple method for Newtonian molecular dynamics is indisputably the leapfrog Störmer–Verlet method. The appropriate generalization to simple Langevin dynamics is unclear. An analysis is presented comparing an 'impulse method' (kick; fluctuate; kick), the 1982 method of van Gunsteren and Berendsen, and the Brünger–Brooks–Karplus (BBK) method. It is shown how the impulse method and the van Gunsteren–Berendsen methods can be implemented as efficiently as the BBK method. Other considerations suggest that the impulse method is the best basic method for simple Langevin dynamics, with the van Gunsteren–Berendsen method a close contender.

1. Introduction

This paper considers simple numerical integrators for Langevin equations of the form

$$\mathrm{d}x = v\,\mathrm{d}t, \qquad M\,\mathrm{d}v = F(x)\,\mathrm{d}t - k_{\mathrm{B}}T D^{-1} v\,\mathrm{d}t + 2^{1/2} k_{\mathrm{B}}T D^{-1/2}\,\mathrm{d}W(t), \tag{1}$$

where x and v are collections of position and velocity coordinates to be determined as functions of time t, M is a diagonal matrix of masses, $F(x)$ is the collective force vector, k_{B} is Boltzmann's constant, T is the temperature, D is a constant diagonal diffusion tensor, and $W(t)$ is a collection of independent standard Wiener processes. One application of this type of Langevin equation is the modelling of implicit solvent with diffusion coefficients $D_{ii} = (k_{\mathrm{B}}T)/(6\pi\eta a_i)$ where η is the solvent viscosity and the a_i are particle radii. Another application is NVT sampling (and the equilibration phase of an NVE simulation), in which $D_{ii} = (k_{\mathrm{B}}T)/(\gamma m_i)$ where γ is the collision parameter or damping constant. For Newtonian molecular dynamics (MD), where $\gamma = 0$, the leapfrog Störmer–Verlet scheme is the basic method from which most advanced methods are constructed. Considered here is the question of how to generalize the leapfrog method to Langevin dynamics. There are a number of generalizations. Two popular ones are the scheme of Brünger, Brooks and Karplus [1] (BBK) and the 1982 scheme of van Gunsteren and Berendsen [2] (vGB82). A third scheme considered here is a 'Langevin impulse' (LI) method, briefly outlined in [3] and tested in [4]. Presented in this article is analytical evidence favouring the LI and vGB82 schemes over the BBK scheme for problems given by equation (1). Additional numerical evidence is needed before drawing practical conclusions.

The more general Langevin equation, derived in [5], has a dense array $D(x)$ of 3×3 tensors instead of D. Possible diffusion tensors are given in [6]. See [7] for appropriate numerical methods.

Section 2 presents basic information needed to derive Langevin integrators. Section 3 derives a simple one-parameter family of numerical integrators that are exact for constant force. These methods seem to require a pair of Gaussian random variables per timestep as well as exponentials of $-\gamma_i \Delta$ where $\gamma_i = (k_{\mathrm{B}}T)/(D_{ii}m_i)$, the m_i are masses, and Δ is the length of a timestep. The vGB82 scheme is such a method, and it is used, for example, in [8]. Another such method, simpler than the vGB82 scheme, is the LI scheme, which is based on the splitting 'kick; fluctuate; kick' (as opposed to the 'kick; drift; kick' of leapfrog). Section 4 discusses the BBK and other schemes, that are not exact for constant force. The BBK scheme is, for example, implemented in the parallel molecular dynamics program NAMD [9].

An earlier analytical comparison of simple Langevin integrators appears in [10]. The study was limited to schemes that require only one independent Gaussian random variable per timestep per velocity component, and among such schemes the BBK scheme is shown to be the best. Section 5 of this article shows that yet other schemes, notably the vGB82 and LI schemes, also may be implemented to attain the same economy in random numbers.

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Molecular Physics ISSN 0026–8976 print/ISSN 1362–3028 online © 2002 Taylor & Francis Ltd
http://www.tandf.co.uk/journals
DOI: 10.1080/0026897021000018321

Section 6 examines the limiting case where the inertia term in equation (1) is negligible compared with the friction term. Both the vGB82 and LI schemes give correct behaviour in the Brownian dynamics limit with the vGB82 scheme giving higher accuracy. The BBK scheme does not behave correctly.

Section 7 analyses stability for the BBK and LI schemes for the harmonic oscillator and concludes that the LI scheme is stable for longer timesteps Δ. (Here stability means that the energy is bounded.) Theoretically, the advantage is only slight for realistic values of γ; however, significant differences are observed in the numerical tests in [4] in the context of multiple timestepping.

Section 8 shows that the generalization of the LI scheme to multiple timesteps is particularly simple.

An analysis of the accuracy of these methods is given elsewhere [11, see also 12]. It is shown that the methods that are exact for constant force are second-order accurate, but that the BBK method is only first-order accurate. Also the argument in [10] favouring the BBK method is reexamined, and it is shown by construction that the BBK method is not unique in its ability to satisfy exactly the virial relation for a harmonic oscillator but that this is possible for other methods including the LI method.

2. Langevin integrators

The analysis in this article is given for the special case of the Langevin equation (1) where the friction tensor $k_{\mathrm{B}}T D^{-1} = \gamma M$ for some scalar $\gamma \geqslant 0$:

$$\mathrm{d}x = v\,\mathrm{d}t, \qquad \mathrm{d}v = M^{-1}F(x)\,\mathrm{d}t - \gamma v\,\mathrm{d}t + (2\gamma k_{\mathrm{B}}T)^{1/2}M^{-1/2}\,\mathrm{d}W(t). \tag{2}$$

The results generalize immediately to γ being a diagonal matrix.

A Wiener process $W(t)$, $t \geqslant 0$, is a one-parameter family of Gaussian random variables with expectations zero and covariances $E(W(s)W(t)) = \min\{s, t\}$. Because the $W(t)$ are all Gaussian, this information suffices to determine joint probabilities. Alternatively, $W(t)$ may be viewed as a 'random' continuous function with $W(0) = 0$. For the Langevin equation a Riemann–Stieltjes integral is adequate (and an Ito or Stratonovich interpretation is unnecessary). A Wiener process may be generated at consecutive grid points t^n by

$$W(0) = 0, \qquad W(t^n) = W(t^{n-1}) + (t^n - t^{n-1})^{1/2}Z^n, \tag{3}$$

where Z^n denotes a sequence of independent standard Gaussian random variables (with mean 0 and variance 1).

Simple numerical Langevin integrators use integrals $\int_a^b f(t)\,\mathrm{d}W(t)$ for deterministic functions $f \in \mathrm{C}^1[a, b]$. These integrals are Gaussian random variables with expectations zero and covariances given by

$$E\left(\int_a^b f_1(t)\,\mathrm{d}W(t)\int_a^b f_2(t)\,\mathrm{d}W(t)\right) = \int_a^b f_1(t)f_2(t)\,\mathrm{d}t. \tag{4}$$

Thus the computation reduces to generating Gaussians of given covariances. A good reference on numerical methods for stochastic differential equations is [13], and other references are [14, 15].

3. Derivation of integrators exact for constant force

We derive the methods in a form that uses positions only. This form is suitable as a canonical form because it is expressed in terms of values x at which the force $F(x)$ is evaluated, which leaves little room for reinterpretation [16]. Also, those same x values are good candidates for assessing the accuracy of a numerical integrator. Integrating equation (2) by parts gives

$$x(t) = x(t^n) + \frac{1 - \mathrm{e}^{-\gamma(t-t^n)}}{\gamma}v(t^n) + \int_{t^n}^{t}\frac{1 - \mathrm{e}^{-\gamma(t-s)}}{\gamma}M^{-1}F(x(s))\,\mathrm{d}s + (2\gamma k_{\mathrm{B}}T)^{1/2}M^{-1/2}\int_{t^n}^{t}\frac{1 - \mathrm{e}^{-\gamma(t-s)}}{\gamma}\,\mathrm{d}W(s). \tag{5}$$

By setting $n = 0$ and $t = t^1$ in equation (5), we may obtain an equation for $x(t^1)$ in terms of $x(t^0)$ and $v(t^0)$. By setting $t = t^{n-1}$ and then $t = t^{n+1}$ in equation (5), we obtain two equations from which we may eliminate $v(t^n)$ to yield an equation for $x(t^{n+1})$ in terms of $x(t^n)$ and $x(t^{n-1})$. More specifically,

$$x(t^1) = x(t^0) + \frac{1 - \mathrm{e}^{-\gamma\Delta}}{\gamma} \times \left(v(t_0) + \int_{t^0}^{t^1}\psi(t - t^0)M^{-1}F(x(t))\,\mathrm{d}t + R_+^0\right) \tag{6}$$

and

$$x(t^{n+1}) = (1 + \mathrm{e}^{-\gamma\Delta})x(t^n) - \mathrm{e}^{-\gamma\Delta}x(t^{n-1}) + \frac{1 - \mathrm{e}^{-\gamma\Delta}}{\gamma} \times \left(\int_{t^{n-1}}^{t^{n+1}}\psi(t - t^n)M^{-1}F(x(t))\,\mathrm{d}t + R_-^n + R_+^n\right), \tag{7}$$

$n = 1, 2, \ldots, N - 1$, where

$$\psi(t) = \begin{cases} \dfrac{\mathrm{e}^{\gamma t} - \mathrm{e}^{-\gamma\Delta}}{1 - \mathrm{e}^{-\gamma\Delta}}, & -\Delta \leqslant t \leqslant 0, \\ \dfrac{1 - \mathrm{e}^{\gamma(t-\Delta)}}{1 - \mathrm{e}^{-\gamma\Delta}}, & 0 \leqslant t \leqslant \Delta, \end{cases} \tag{8}$$

and

$$R_-^n = (2\gamma k_B T)^{1/2} M^{-1/2} \int_{t^{n-1}}^{t^n} \psi(t - t^n)\, dW(t),$$

$$R_+^n = (2\gamma k_B T)^{1/2} M^{-1/2} \int_{t^n}^{t^{n+1}} \psi(t - t^n)\, dW(t).$$

The forces $F(x(t))$ in the integrands of equations (6) and (7) are to be approximated using values of $F(x(t))$ at positions $x^n \approx x(t^n)$ that have already been calculated. For a method based on positions, three formulas may be needed.

3.1. *Starting formula*. There is little choice but to use $F(x(t)) \approx F(x^0)$:

$$x^1 = x^0 + \frac{1 - e^{-\gamma\Delta}}{\gamma}(v^0 + \Delta w_+ M^{-1} F(x^0) + R_+^0), \quad (10)$$

where $x^0 = x(t^0)$, $v^0 = v(t^0)$, and

$$w_+ = w_+(\gamma\Delta) = \frac{e^{-\gamma\Delta} - 1 + \gamma\Delta}{\gamma\Delta(1 - e^{-\gamma\Delta})}. \quad (11)$$

3.2. *Continuation formula*. Here we interpolate $F(x^{n-1})$ and $F(x^n)$, which we express as $F(x(t)) \approx F(x^n) + \chi(t - t^n)(F(x^n) - F(x^{n-1}))$ for some $\chi(t)$ satisfying $\chi(-\Delta) = -1$ and $\chi(0) = 0$. (The choice of $\chi(t)$ is discussed below.) The formula is

$$x^{n+1} = (1 + e^{-\gamma\Delta})x^n - e^{-\gamma\Delta} x^{n-1} + \frac{1 - e^{-\gamma\Delta}}{\gamma}(\Delta M^{-1} F(x^n) + \Delta S M^{-1}(F(x^n) - F(x^{n-1})) + R^n), \quad (12)$$

$n = 1, 2, \ldots, N - 1$, where

$$R^n = R_-^n + R_+^n \quad \text{and} \quad S = \frac{1}{\Delta}\int_{-\Delta}^{\Delta} \psi(t)\chi(t)\, dt. \quad (13)$$

3.3. *Finishing formula*. A finishing formula to get $v^N \approx v(t^N)$ can be chosen so that if it is followed by a restart it is equivalent to one use of the continuation formula

$$v^N = \frac{\gamma e^{-\gamma\Delta}}{1 - e^{-\gamma\Delta}}(x^N - x^{N-1}) + \Delta w_- M^{-1} F(x^N) + \Delta S M^{-1}(F(x^N) - F(x^{N-1})) + R_-^N, \quad (14)$$

where

$$w_- = w_-(\gamma\Delta) = 1 - w_+(\gamma\Delta). \quad (15)$$

A different criterion, and hence a different formula, could be used to define velocities.

A method as defined above is exact for constant force.

The Langevin impulse method is defined by replacing $F(x)$ by $\Delta(w_+\delta(t - t_+^{n-1}) + w_-\delta(t - t_-^n)) \times F(x)$ for $t^{n-1} \leqslant t \leqslant t^n$ in equation (2), where the weights $0 < w_- \leqslant w_+ < 1$, defined by equations (11) and (15), are such that the method is exact for constant force. (As originally proposed [3], $w_+ = w_- = \frac{1}{2}$. The choice of weights does not affect the continuation formula, but the choice given here makes the starting formula exact for constant force.) This gives a formula that is identical to equation (10) and equation (12) for $S = 0$. Note the time symmetry of the impulse method: if we substitute $-\Delta$ for Δ and interchange $n + 1$ and $n - 1$, equation (12) remains the same.

The derivation of the vGB82 scheme and an alternative derivation of the LI scheme proceeds from two choices of basis functions for interpolation in the key approximation (12). To assess how $F(x(t))$ behaves, we should first look at the behaviour of $x(t)$. Neglecting noise, we have from equation (5) that, for constant force,

$$x(t) \approx x(t^n) + \frac{1 - e^{-\gamma(t-t^n)}}{\gamma} v(t^n) + \frac{e^{-\gamma(t-t^n)} - 1 + \gamma(t - t^n)}{\gamma^2} M^{-1} F, \quad (16)$$

so that $x(t)$, $t^{n-1} \leqslant t \leqslant t^{n+1}$, is approximately a linear combination of 1, $e^{-\gamma(t-t^n)}$, and $t - t^n$. The last function is least important because it is not present if $F = 0$. If the force $F(x)$ is linear, then $F(x(t))$ is likewise a linear combination of these functions, approximately. The LI method chooses 1, $e^{-\gamma(t-t^n)}$ as basis functions for interpolation of F^n, F^{n-1}, so that $\chi(t) = (1 - e^{-\gamma(t-t^n)})/(e^{\gamma\Delta} - 1)$ and

$$S = 0. \quad (17)$$

The vGB82 method chooses 1, $t - t^n$ as basis functions for interpolation of F^n, F^{n-1}, so $\chi(t) = (t - t^n)/\Delta$ and

$$S = \tfrac{1}{2}(w_+ - w_-). \quad (18)$$

The weights $w_\pm = \frac{1}{2} \pm \frac{1}{12}\gamma\Delta + O((\gamma\Delta)^3)$.

4. Other simple Langevin integrators

Discussed here are integrators that are not exact for constant force.

The BBK integrator [1] is motivated by the desire to use just one independent random variable per step per velocity component. One integration of equation (2) yields

$$\dot{x}(t^{n+1/2}) = \dot{x}(t^{n-1/2}) + \int_{t^{n-1/2}}^{t^{n+1/2}} (M^{-1}F(x(t) - \gamma\dot{x}(t))\,\mathrm{d}t + (2\gamma k_{\mathrm{B}}T)^{1/2}M^{-1/2}(W(t^{n+1/2}) - W(t^{n-1/2})), \tag{19}$$

and centred difference approximations give the continuation formula

$$x^{n+1} = 2x^n - x^{n-1} + \Delta^2 M^{-1}F(x^n) - \frac{\Delta\gamma}{2}(x^{n+1} - x^{n-1}) + \Delta^{3/2}(2\gamma k_{\mathrm{B}}T)^{1/2}M^{-1/2}Z^n, \tag{20}$$

$n = 1, 2, \ldots, N-1$. A similar argument gives a starting formula for obtaining x^1 in terms of x^0 and v^0:

$$x^1 = x^0 + \Delta v^0 + \tfrac{1}{2}\Delta^2 M^{-1}F(x^0) - \tfrac{1}{2}\Delta^2\gamma v^0 + \tfrac{1}{2}\Delta^{3/2}(2\gamma k_{\mathrm{B}}T)^{1/2}M^{-1/2}Z^0. \tag{21}$$

Another discretization that generalizes the leapfrog method is proposed in [17]. It is implemented in the Brownian dynamics program UHBD [18]. Implicit discretization schemes have also been proposed for Langevin dynamics [19].

5. Efficient implementation

The values R_+^0, R^1, R^2, ..., R^{N-1}, R_-^N are joint Gaussian random variables of zero mean and known covariance matrix C, which has the form

$$C = \begin{bmatrix} a & b & & & \\ b & c+a & b & & \\ & b & \ddots & \ddots & \\ & & \ddots & c+a & b \\ & & & b & c \end{bmatrix}, \tag{22}$$

where $a = E(R_+^n R_+^n)$, $b = E(R_+^n R_-^{n+1})$ and $c = E(R_-^{n+1} R_-^{n+1})$. Using equation (4), it can be shown that

$$a = k_{\mathrm{B}}T\,M^{-1}(2w_+^2\gamma\Delta + w_+ - w_-), \tag{23}$$

$$b = k_{\mathrm{B}}T\,M^{-1}(2w_+w_-\gamma\Delta + w_- - w_+), \tag{24}$$

$$c = k_{\mathrm{B}}T\,M^{-1}(2w_-^2\gamma\Delta + w_+ - w_-). \tag{25}$$

To calculate the random numbers, we may use $[R_+^0, R^1, R^2, \ldots, R^{N-1}, R_-^N]^{\mathrm{T}} = C_{1/2}[Z^0, Z^1, Z^2, \ldots, Z^{N-1}, Z^N]^{\mathrm{T}}$ where the Z^n are independent Gaussian random numbers of mean 0 and variance 1 and where $C_{1/2}$ is a square matrix satisfying $C_{1/2}C_{1/2}^{\mathrm{T}} = C$. For a Cholesky decomposition, $C_{1/2}$ is a bidiagonal matrix with main diagonal elements, denoted here by α^0, α^1, α^2, ..., α^N, and first subdiagonal elements, denoted here by β^0, β^1, ..., β^{N-1}. A simple recurrence for these elements is given in the algorithm that follows.

Note the efficient use of random numbers. The number of random variables needed is just one (set) per step if we forgo the calculation of accurate velocities at each step. If we want to apply the finishing formula at every step, then the continuation formula is not needed and we have to generate two rather than one random number per step.

The position-only form (12) suffers from excessive roundoff error in floating-point arithmetic [20]. This may be avoided through use of the summed form ([20], p. 353), in which the older value x^{n-1} is represented in terms of x^n and an increment, which we take to be $v^{n-1/2} = \gamma\mathrm{e}^{-\gamma\Delta/2}(1 - \mathrm{e}^{-\gamma\Delta})^{-1}(x^n - x^{n-1})$. Suppose that velocity output is desired after $N \geqslant 1$ steps of length Δ and that we are given the initial collective position vector x^0 and velocity vector v^0. We start with

$$F^0 = F(x^0), \tag{26}$$

$$\alpha^0 = a^{1/2}, \tag{27}$$

$$R_+^0 = \alpha^0 Z^0, \tag{28}$$

$$v^{1/2} = \mathrm{e}^{-\gamma\Delta/2}(v^0 + \Delta w_+ M^{-1}F^0 + R_+^0), \tag{29}$$

$$x^1 = x^0 + \frac{1 - \mathrm{e}^{-\gamma\Delta}}{\gamma\mathrm{e}^{-\gamma\Delta/2}}v^{1/2}. \tag{30}$$

We continue for $n = 1, 2, \ldots, N-1$ with

$$F^n = F(x^n), \tag{31}$$

$$\beta^{n-1} = b/\alpha^{n-1}, \qquad \alpha^n = [a + c - (\beta^{n-1})^2]^{1/2}, \tag{32}$$

$$R^n = \beta^{n-1}Z^{n-1} + \alpha^n Z^n, \tag{33}$$

$$v^{n+1/2} = \mathrm{e}^{-\gamma\Delta/2}(\mathrm{e}^{-\gamma\Delta/2}v^{n-1/2} + \Delta M^{-1}F^n + \Delta SM^{-1}(F^n - F^{n-1}) + R^n), \tag{34}$$

$$x^{n+1} = x^n + \frac{1 - \mathrm{e}^{-\gamma\Delta}}{\gamma\mathrm{e}^{-\gamma\Delta/2}}v^{n+1/2}. \tag{35}$$

We finish with

$$F^N = F(x^N), \tag{36}$$

$$\beta^{N-1} = b/\alpha^{N-1}, \qquad \alpha^N = [c - (\beta^{N-1})^2]^{1/2}, \tag{37}$$

$$R_-^N = \beta^{N-1}Z^{N-1} + \alpha^N Z^N, \tag{38}$$

$$v^N = \mathrm{e}^{-\gamma\Delta/2}v^{N-1/2} + \Delta w_- M^{-1}F^N + \Delta SM^{-1}(F^N - F^{N-1}) + R_-^N. \tag{39}$$

The required constants are defined by equations (11), (15), (23)–(25), and (17), (18).

If the velocity v^N is not needed, the arithmetic of a Cholesky decomposition may be avoided by choosing $C_{1/2}$ to be an $N \times N+1$ matrix with main diagonal elements α_0, α, α, ..., α, and with first superdiagonal elements β, β, ..., β where

$$\alpha^2 = \tfrac{1}{2}(c + a + [(c+a)^2 - 4b^2]^{1/2}), \tag{40}$$

$$\beta^2 = \tfrac{1}{2}(c + a - [(c+a)^2 - 4b^2]^{1/2}), \tag{41}$$

$$\alpha_0^2 = \alpha^2 - c. \tag{42}$$

After an initialization

$$F^0 = F(x^0), \tag{43}$$

$$R^0 = \alpha^0 Z^0 + \beta Z^1, \tag{44}$$

$$\bar{v}^0 = v^0, \tag{45}$$

each step is the same:

$$v^{n+1/2} = \mathrm{e}^{-\gamma\Delta/2}(\bar{v}^n + w_+(\Delta M^{-1}F^n + R^n)), \tag{46}$$

$$x^{n+1} = x^n + \frac{1 - \mathrm{e}^{-\gamma\Delta}}{\gamma \mathrm{e}^{-\gamma\Delta/2}} v^{n+1/2}, \tag{47}$$

$$F^{n+1} = F(x^{n+1}), \tag{48}$$

$$R^{n+1} = \alpha Z^{n+1} + \beta Z^{n+2}, \tag{49}$$

$$\bar{v}^{n+1} = \mathrm{e}^{-\gamma\Delta/2} v^{n+1/2} + \Delta S M^{-1}(F^{N+1} - F^N) + w_-(\Delta M^{-1}F^{n+1} + R^{n+1}). \tag{50}$$

6. Robustness: the limiting case of Newtonian and Brownian dynamics

In the limit of Newtonian dynamics, as $\gamma \to 0$, all three integrators become the leapfrog Störmer–Verlet method. More specifically, the starting formulas equations (10) and (21) become

$$x^1 = x^0 + \Delta v^0 + \tfrac{1}{2}\Delta^2 M^{-1} F(x^0), \tag{51}$$

and the continuation formulas equations (12) and (20) become

$$x^{n+1} = 2x^n - x^{n-1} + \Delta^2 M^{-1} F(x^n), \tag{52}$$

$n = 1, 2, \ldots, N-1$.

A high friction limit approximation is appropriate if the friction effect $k_\mathrm{B}T D^{-1}$ is much greater than the inertial effect M/t. This limit is attained by setting the masses M to zero in equation (1) and the resulting equations are those of Brownian dynamics:

$$\mathrm{d}x = \frac{1}{k_\mathrm{B}T} DF(x)\,\mathrm{d}t + 2^{1/2} D^{1/2}\,\mathrm{d}W(t). \tag{53}$$

This approximation is derived in [6], section III.

In the limit of Brownian dynamics, as $M \to 0$ with $\gamma = k_\mathrm{B}T D^{-1} M^{-1}$, the starting formula, equation (10), becomes

$$x^1 = x^0 + \frac{\Delta}{k_\mathrm{B}T} DF(x^0) + (2D)^{1/2} \int_{t^0}^{t^1} \mathrm{d}W(t), \tag{54}$$

and the continuation formula, equation (12), becomes

$$x^{n+1} = x^n + \frac{\Delta}{k_\mathrm{B}T} D(F(x^n) + S(F(x^n) - F(x^{n-1}))) + (2D)^{1/2} \int_{t^n}^{t^{n+1}} \mathrm{d}W(t), \tag{55}$$

$n = 1, 2, \ldots, N-1$, where $S = 0$ for the LI scheme and $S = \frac{1}{2}$ for the vGB82 scheme. The value $S = 0$ corresponds to the Euler–Maruyama method ([13], p. 305), of weak order 1, which is a popular choice for Brownian dynamics, introduced in this context by Ermak and McCammon [6]. The value $S = \frac{1}{2}$ corresponds to the second-order explicit Adams formula.

In the Brownian limit, the BBK scheme becomes the explicit midpoint formula, which is unconditionally unstable ([20], p. 373]).

7. Stability

We compare the stability of the impulse integrator to that of the BBK scheme. For the analysis we consider the harmonic oscillator, for which $M^{-1}F(x) = -\omega^2 x$, and omit the inhomogeneous terms in the numerical scheme, since when studying perturbations they cancel out. The outcome of such an analysis is a condition involving the timestep Δ and problem parameters γ and ω that guarantees that the homogeneous difference equation has only decaying solutions. For this purpose we need conditions for the solutions of a general quadratic equation

$$\lambda^2 + 2c_1\lambda + c_0 = 0 \tag{56}$$

both to be less than one in modulus. A detailed case analysis (involving a plot of c_0 versus c_1) shows this to be the case if and only if

$$|c_0| < 1 \quad \text{and} \quad |c_1| < \tfrac{1}{2}(1 + c_0). \tag{57}$$

For the harmonic oscillator with the inhomogeneous term omitted, the impulse integrator, given by equation by equation (12), simplifies to

$$x^{n+1} = (1 + \mathrm{e}^{-\gamma\Delta})x^n - \mathrm{e}^{-\gamma\Delta} x^{n-1} - \frac{1 - \mathrm{e}^{-\gamma\Delta}}{\gamma} \Delta\omega^2 x^n. \tag{58}$$

The characteristic equation that determines the general solution (obtained by trying $x^n = (\lambda)^n$) is given by

$$\lambda^2 - \left((1 + \mathrm{e}^{-\gamma\Delta}) - \frac{1 - \mathrm{e}^{-\gamma\Delta}}{\gamma} \Delta\omega^2 \right) \lambda + \mathrm{e}^{-\gamma\Delta} = 0, \quad (59)$$

which we express as

$$\lambda^2 - 2B\mathrm{e}^{-g/2}\lambda + \mathrm{e}^{-g} = 0, \quad (60)$$

where $B = \cosh \frac{1}{2}g - w^2 g^{-1} \sinh \frac{1}{2}g$, with $g = \gamma\Delta$, and $w = \omega\Delta$.

Applying the stability conditions given by equation (57) to characteristic equation (60) for the impulse method gives

$$|B| < \cosh \frac{g}{2}, \quad (61)$$

which simplifies to

$$w < \left(2g \coth \frac{g}{2} \right)^{1/2}, \quad (62)$$

which is shown in figure 1 as the region under the solid curve.

It is interesting to find the condition for the critical damping. This is done by setting the discriminant in equation (60) to zero. We find the two solutions

$$w^2 = g \tanh \frac{g}{4}, \quad (63)$$

$$w^2 = g \coth \frac{g}{4}, \quad (64)$$

both shown in figure 1 as dotted curves. For comparison against the expression $\gamma = 2\omega$ for the critical damping of the analytical damped oscillator, equation (63) may be expressed as

$$\gamma = 2\omega \left(\frac{4}{\gamma\Delta} \tanh \frac{\gamma\Delta}{4} \right)^{-1/2}. \quad (65)$$

Now we consider the stability analysis of the BBK scheme. With the same simplifications given previously, equation (20) becomes

$$x^{n+1} = 2x^n - x^{n-1} - \Delta^2\omega^2 x^n - \frac{\Delta\gamma}{2}(x^{n+1} - x^{n-1}). \quad (66)$$

The characteristic equation that determines the general solution is given by

$$\left(1 + \tfrac{1}{2}\gamma\Delta\right)\lambda^2 - (2 - \omega^2\Delta^2)\lambda + \left(1 - \tfrac{1}{2}\gamma\Delta\right) = 0. \quad (67)$$

Normalizing equation (67) and applying the stability conditions of equation (57) gives

$$\omega\Delta < 2. \quad (68)$$

The region for critical damping corresponds to a curve

$$\left(\frac{\gamma}{\omega}\right)^2 + (\omega\Delta)^2 = 4. \quad (69)$$

Figure 2 shows a plot of the stability boundary for the impulse integrator and the BBK scheme. The x axis has been transformed from $\gamma\Delta$ to γ/ω to illustrate how much damping is needed for stability as Δ increases, with a fixed ω.

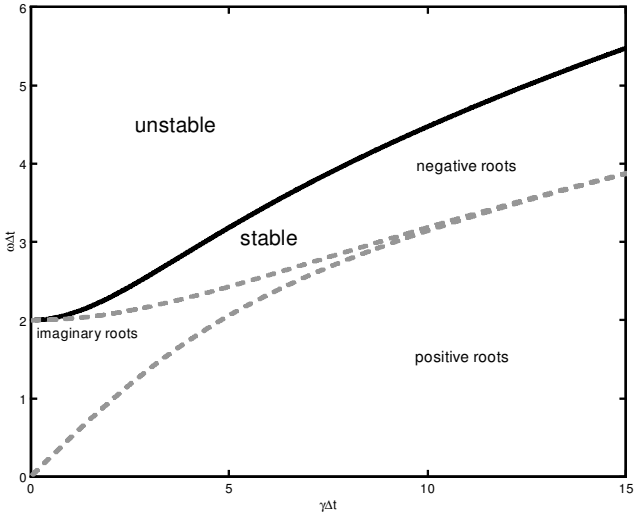

Figure 1. Stability region for the impulse integrator (LI). The boundaries of critical damping, along with regions of imaginary, positive and negative roots are illustrated. Note that the method is unstable only for $\omega\Delta > (2\gamma\Delta \coth(\gamma\Delta))^{1/2}$, where the roots of the characteristic polynomial are negative.

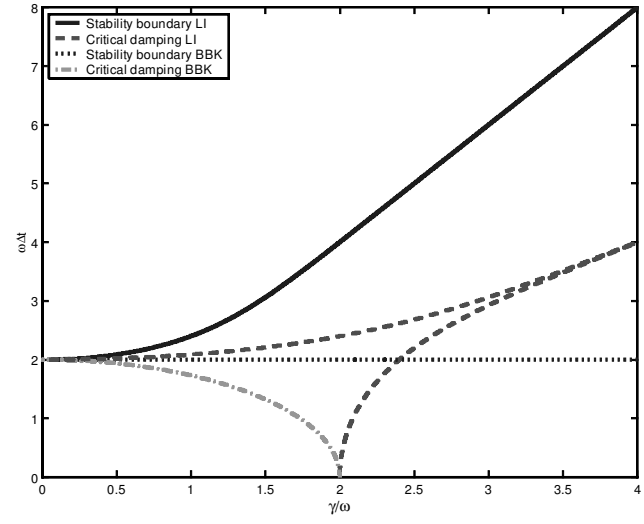

Figure 2. Comparison of the stability boundary and the critical damping boundary for the BBK scheme and the im-pulse integrator (LI) applied to the scalar problem $\ddot{x} + \gamma\dot{x} + \omega^2 x = 0$.

8. Multiple timestepping

Because of damping, Langevin dynamics are not nearly as prone to instability as Newtonian MD, so multiple timestepping is especially advantageous, as illustrated by the integration method LN [21].

Generalization of methods based on splitting, such as the LI method, to multiple timesteps is particularly simple. (Other methods may be generalized using the 'equivalence' approach of [22], although this does not lead to a unique formula.) Suppose $F = F_{\text{fast}} + F_{\text{slow}}$ where F_{fast} is to be evaluated every step and F_{slow} every m steps. The MTS impulse method is equivalent to replacing $F(x)$ by

$$\sum_{n=pm-m+1}^{pm} \Delta(w_+(\gamma\Delta)\delta(t-t_+^{n-1}) + w_-(\gamma\Delta)\delta(t-t_-^{n}))F_{\text{fast}}(x)$$

$$+ m\Delta(w_+(m\gamma\Delta)\delta(t-t_+^{pm-m}) + w_-(m\gamma\Delta)\delta(t-t_-^{pm}))F_{\text{slow}}(x) \tag{72}$$

for $t^{n-1} \leqslant t \leqslant t^n$. It follows from superposition that the MTS impulse method is exact if both F_{slow} and F_{fast} are constant. Let w be one of w_-, 1, or w_+. The method is implemented by replacing $\Delta\, w(\gamma\Delta)F^n$ by $\Delta\, w(\gamma\Delta)F^n_{\text{fast}} + m\Delta\, w(m\gamma\Delta)F^n_{\text{slow}}$ if n is a multiple of m and by $\Delta\, w(\gamma\Delta)F^n_{\text{fast}}$ otherwise.

R.D.S. was supported in part by NSF Grants DMS-9971830 and DBI-9974555 and NIH Grant P41RR05969. J.A.I. was supported in part by NSF Grant BIOCOMPLEXITY-PHY-0083653 and a grant from the University of Notre Dame.

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