

# An impulse integrator for Langevin dynamics

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## Abstract

The best simple method for Newtonian molecular dynamics is indisputably the leapfrog/Störmer/Verlet method. The appropriate generalization to simple Langevin dynamics is unclear. Presented is an analysis 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 method 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 article considers simple numerical integrators for Langevin equations of the form

$$dx = vdt, \quad Mdv = F(x)dt - k_B T D^{-1} vdt + \sqrt{2k_B T D^{-1/2}} dW(t) \quad (1)$$

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 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 modeling of implicit solvent with diffusion coefficients  $D_{ii} = (k_B T)/(6\pi\eta a_i)$  where  $\eta$  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  $D_{ii} = (k_B T)/(\gamma m_i)$  where  $\gamma$  is the collision parameter or damping constant. For Newtonian molecular dynamics (MD), where  $D = 0$ , 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 LI and vGB82 over BBK for problems given by equation (1).

The more general Langevin equation, derived in [5], has a dense array  $D(x)$  of 3 by 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 time step as well as exponentials of  $-\gamma_i \Delta$  where  $\gamma_i = (k_B T)/(D_{ii} m_i)$  and the  $m_i$  are masses. The vGB82 scheme is such a method, and it is used, for example, in [8]. Another such method, simpler than vGB82, 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 BBK and other schemes, which are not exact for constant force. BBK 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 time step per velocity component, and among such schemes BBK is shown to be the best. Section 5 of this article shows that yet other schemes notably vGB82 and LI can also be implemented to attain the same economy in random numbers.

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

Section 7 analyzes stability for BBK and LI for the harmonic oscillator and concludes that LI is more stable. This conclusion is supported by the numerical tests in [4].

Section 8 shows that the generalization of LI to multiple time steps is particularly simple.

An analysis of the accuracy of these methods is given in a companion paper [11, 12]. There 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 BBK is reexamined, and it is shown by construction that the BBK method is not unique in its ability to exactly satisfy the virial relation for a harmonic oscillator but that this is possible for other methods including LI.

## 2 Langevin integrators

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

$$dx = v dt, \quad dv = M^{-1} F(x) dt - \gamma v dt + \sqrt{2\gamma k_B T} M^{-1/2} dW(t). \quad (2)$$

The results generalize immediately to  $\gamma$  being a diagonal matrix.

A Wiener process  $W(t)$ ,  $t \geq 0$ , 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,  $W(t)$  can 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 can be generated at consecutive grid points  $t^n$  by

$$W(0) = 0, \quad W(t^n) = W(t^{n-1}) + \sqrt{t^n - t^{n-1}} Z^n \quad (3)$$

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

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

given by

$$E \left( \int_a^b f_1(t) dW(t) \int_a^b f_2(t) dW(t) \right) = \int_a^b f_1(t) f_2(t) dt. \quad (4)$$

Thus the computation reduces to generating Gaussians of given covariances. A good reference on numerical methods for stochastic differential equations is the book [13]. 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 re-interpretation [16]. Also, those same  $x$  values are good candidates for assessing accuracy of a numerical integrator. Integrating equation (2) by parts gives

$$\begin{aligned} x(t) = & x(t^n) + \frac{1 - e^{-\gamma(t-t^n)}}{\gamma} v(t^n) + \int_{t^n}^t \frac{1 - e^{-\gamma(t-s)}}{\gamma} M^{-1} F(x(s)) ds \\ & + \sqrt{2\gamma k_B T} M^{-1/2} \int_{t^n}^t \frac{1 - e^{-\gamma(t-s)}}{\gamma} dW(s). \end{aligned} \quad (5)$$

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

$$x(t^1) = x(t^0) + \frac{1 - e^{-\gamma\Delta}}{\gamma} \left( v(t^0) + \int_{t^0}^{t^1} \psi(t - t^0) M^{-1} F(x(t)) dt + R_+^0 \right) \quad (6)$$

and

$$\begin{aligned} x(t^{n+1}) = & (1 + e^{-\gamma\Delta}) x(t^n) - e^{-\gamma\Delta} x(t^{n-1}) \\ & + \frac{1 - e^{-\gamma\Delta}}{\gamma} \left( \int_{t^{n-1}}^{t^{n+1}} \psi(t - t^n) M^{-1} F(x(t)) dt + R_-^n + R_+^n \right), \end{aligned} \quad (7)$$

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

$$\psi(t) = \begin{cases} \frac{e^{\gamma t} - e^{-\gamma\Delta}}{1 - e^{-\gamma\Delta}}, & -\Delta \leq t \leq 0, \\ \frac{1 - e^{\gamma(t-\Delta)}}{1 - e^{-\gamma\Delta}}, & 0 \leq t \leq \Delta, \end{cases} \quad (8)$$

and

$$R_-^n = \sqrt{2\gamma k_B T} M^{-1/2} \int_{t^{n-1}}^{t^n} \psi(t - t^n) dW(t), \quad R_+^n = \sqrt{2\gamma k_B T} M^{-1/2} \int_{t^n}^{t^{n+1}} \psi(t - t^n) dW(t). \quad (9)$$

The forces  $F(x(t))$  in the integrands of Eqs. (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:

I. *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

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

II. *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

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

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

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

III. *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  $t^{n-1} \leq t \leq t^n$  in the given equation (2) where the weights  $0 < w_- \leq w_+ < 1$ , defined by Eqs. (11) and (15), are such that the method is exact for constant force.<sup>1</sup> 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  $\Delta$  and interchange  $n + 1$  and  $n - 1$ , equation (12) remains the same.

The derivation of vGB82 and an alternative derivation of LI 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  $x(t)$ ,  $t^{n-1} \leq t \leq t^{n+1}$ , 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

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

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<sup>1</sup>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.

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

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

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

$$\dot{x}(t^{n+1/2}) = \dot{x}(t^{n-1/2}) + \int_{t^{n-1/2}}^{t^{n+1/2}} (M^{-1}F(x(t)) - \gamma\dot{x}(t)) dt + \sqrt{2\gamma k_B T} M^{-1/2} (W(t^{n+1/2}) - W(t^{n-1/2})), \quad (19)$$

and centered difference approximations give the continuation formula

$$x^{n+1} = 2x^n - x^{n-1} + \Delta^2 M^{-1} F(x^n) - \frac{\Delta\gamma}{2}(x^{n+1} - x^{n-1}) + \Delta^{3/2} \sqrt{2\gamma k_B T} M^{-1/2} Z^n, \quad (20)$$

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

$$x^1 = x^0 + \Delta v^0 + \frac{1}{2}\Delta^2 M^{-1} F(x^0) - \frac{1}{2}\Delta^2 \gamma v^0 + \frac{1}{2}\Delta^{3/2} \sqrt{2\gamma k_B T} M^{-1/2} Z^0. \quad (21)$$

Another discretization which 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  $R_+^0, R^1, R^2, \dots, R^{N-1}, R_-^N$  are joint Gaussian random variables of zero mean and known covariance matrix  $C$ , which has the form

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

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

$$a = k_B T M^{-1} (2w_+^2 \gamma \Delta + w_+ - w_-), \quad (23)$$

$$b = k_B T M^{-1} (2w_+ w_- \gamma \Delta + w_- - w_+), \quad (24)$$

$$c = k_B T M^{-1} (2w_-^2 \gamma \Delta + w_+ - w_-). \quad (25)$$

To calculate the random numbers, we can use  $[R_+^0, R^1, R^2, \dots, R^{N-1}, R_-^N]^T = C_{1/2} [Z^0, Z^1, Z^2, \dots, Z^{N-1}, Z^N]^T$  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  $C_{1/2} C_{1/2}^T = C$ . For a Cholesky decomposition,  $C_{1/2}$  is a bidiagonal matrix with main diagonal elements, denoted here by  $\alpha^0, \alpha^1, \alpha^2, \dots, \alpha^N$ , and first subdiagonal elements, denoted here by  $\beta^0, \beta^1, \dots, \beta^{N-1}$ . 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 can 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  $v^{n-1/2} = \gamma e^{-\gamma\Delta/2}(1 - e^{-\gamma\Delta})^{-1}(x^n - x^{n-1})$ . Suppose that velocity output is desired after  $N \geq 1$  steps of length  $\Delta$  and that we are given initial collective position vector  $x^0$  and velocity vector  $v^0$ . We start with

$$F^0 = F(x^0), \quad (26)$$

$$\alpha^0 = \sqrt{a}, \quad (27)$$

$$R_+^0 = \alpha^0 Z^0, \quad (28)$$

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

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

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

$$F^n = F(x^n), \quad (31)$$

$$\beta^{n-1} = b/\alpha^{n-1}, \quad \alpha^n = \sqrt{a + c - (\beta^{n-1})^2}, \quad (32)$$

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

$$v^{n+1/2} = e^{-\gamma\Delta/2} \left( e^{-\gamma\Delta/2} v^{n-1/2} + \Delta M^{-1} F^n + \Delta S M^{-1} (F^n - F^{n-1}) + R^n \right), \quad (34)$$

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

We finish with

$$F^N = F(x^N), \quad (36)$$

$$\beta^{N-1} = b/\alpha^{N-1}, \quad \alpha^N = \sqrt{c - (\beta^{N-1})^2}, \quad (37)$$

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

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

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

If the velocity  $v^N$  is not needed, the arithmetic of a Cholesky decomposition can be avoided by choosing  $C_{1/2}$  to be an  $N$  by  $N + 1$  matrix with main diagonal elements  $\alpha_0, \alpha, \alpha, \dots, \alpha$ , and with first superdiagonal elements  $\beta, \beta, \dots, \beta$  where

$$\alpha^2 = \frac{1}{2}(c + a + ((c + a)^2 - 4b^2)^{1/2}), \quad (40)$$

$$\beta^2 = \frac{1}{2}(c + a - ((c + a)^2 - 4b^2)^{1/2}), \quad (41)$$

$$\alpha_0^2 = \alpha^2 - c. \quad (42)$$

After an initialization

$$F^0 = F(x^0), \quad (43)$$

$$R^0 = \alpha^0 Z^0 + \beta Z^1, \quad (44)$$

$$\bar{v}^0 = v^0, \quad (45)$$

each step is the same:

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

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

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

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

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

## 6 Robustness—the limiting case of Newtonian and Brownian dynamics

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

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

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

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

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

A high friction limit approximation is appropriate if the friction effect  $k_B T D^{-1}$  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:

$$dx = \frac{1}{k_B T} DF(x)dt + \sqrt{2D}^{1/2} dW(t). \quad (53)$$

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

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

$$x^1 = x^0 + \frac{\Delta}{k_B T} DF(x^0) + (2D)^{1/2} \int_{t^0}^{t^1} dW(t) \quad (54)$$

and the continuation formula equation (12) becomes

$$x^{n+1} = x^n + \frac{\Delta}{k_B T} D (F(x^n) + S(F(x^n) - F(x^{n-1}))) + (2D)^{1/2} \int_{t^n}^{t^{n+1}} dW(t), \quad (55)$$

$n = 1, 2, \dots, N - 1$ , where  $S = 0$  for LI and  $S = \frac{1}{2}$  for vGB82. 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, BBK 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 BBK. For the analysis we consider the harmonic oscillator, for which  $M^{-1}F(x) = -\omega^2x$ , 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 time step  $\Delta$  and problem parameters  $\gamma$  and  $\omega$  which guarantees that the homogeneous difference equation has only decaying solutions. For this purpose we need conditions for the solutions of a general quadratic equation

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

both to be less than one in modulus. A detailed case analysis shows this to be the case if and only if

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

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

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

The characteristic equation that determines the general solution is given by

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

which we express as

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

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

Applying stability conditions 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) can 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

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

The characteristic equation that determines the general solution is given by

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

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 BBK. The x axis has been transformed from  $\gamma\Delta$  to  $\gamma/\omega$  to illustrate how much damping is needed for stability as  $\Delta$  increases, with a fixed  $\omega$ .

## 8 Multiple time stepping

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

Generalization of methods based on splitting, such as LI, to multiple time steps is particularly simple. (Other methods can 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  $F_{\text{fast}}$  is to be evaluated every step and  $F_{\text{slow}}$  every  $m$  steps. The MTS impulse method is equivalent to replacing  $F(x)$  by

$$\begin{aligned} & \sum_{n=pm-m+1}^{pm} \Delta \left( w_+(\gamma\Delta)\delta(t - t_+^{n-1}) + w_-(\gamma\Delta)\delta(t - t_-^n) \right) F_{\text{fast}}(x) \\ & + m\Delta \left( w_+(m\gamma\Delta)\delta(t - t_+^{pm-m}) + w_-(m\gamma\Delta)\delta(t - t_-^{pm}) \right) F_{\text{slow}}(x) \end{aligned} \quad (70)$$

for  $t^{n-1} \leq t \leq t^n$ . It follows from superposition that the MTS impulse method is exact if both  $F_{\text{slow}}$  and  $F_{\text{fast}}$  are constant. Let  $w$  be one of  $w_-$ , 1, or  $w_+$ . The method is implemented by replacing

$\Delta w(\gamma\Delta)F^n$  by  $\Delta w(\gamma\Delta)F_{\text{fast}}^n + m\Delta w(m\gamma\Delta)F_{\text{slow}}^n$  if  $n$  is a multiple of  $m$  and by  $\Delta w(\gamma\Delta)F_{\text{fast}}^n$  otherwise.

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## Figure Captions

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. Notice 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 BBK and the impulse integrator (LI) applied to the scalar problem  $\ddot{x} + \gamma\dot{x} + \omega^2x = 0$ .



