# FLUCTUATION-DISSIPATION THEOREM CONSISTENT APPROXIMATION OF THE LANGEVIN DYNAMICS MODEL*

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**Abstract.** We present a numerical method for solving the Langevin dynamics model. Rather than the trajectory-wise accuracy, we emphasize on the consistency to the equilibrium statistics at the discrete level. A discrete fluctuation-dissipation theorem is imposed to ensure that the statistical properties are preserved.

**Key words.** Molecular Dynamics; Langevin Dynamics; Fluctuation-dissipation theorem.

**AMS subject classifications.** 65C30, 60G10.

**1. Introduction** This paper is concerned with the numerical methods for solving the Langevin dynamics model. Such models arise when a molecular system is embedded in a medium, e.g., solvent, or when a reversible dynamics is coarse-grained to fewer degrees of freedom. They play a crucial role in the modeling of bio-molecular systems. We write the Langevin dynamics in the following first-order form.

$$\begin{cases} \dot{x}_i = v_i, \\ m_i \dot{v}_i = -\nabla_{x_i} V(x) - \gamma m_i v_i + w_i(t), \quad 1 \le i \le N. \end{cases} \tag{1.1}$$

The right hand side consists of a conservative force, friction and a random noise respectively. In general, the friction coefficient $\gamma$ is a matrix. The random force is a white Gaussian noise. A key property of the Langevin dynamics is that the covariance of the noise has to be consistent with the friction coefficients in the form of the fluctuation-dissipation theorem (FDT) [**?**],

$$\langle w_i(t) w_i(t')^T \rangle = 2k_B T \gamma \delta(t-t'). \tag{1.2}$$

This property ensures that the correct equilibrium distribution will be reached [**?**]. For this reason, the Langevin dynamics can be used as a 'thermostat' to equilibrate a system to a desired temperature, and then sample relevant physical quantities [**?**, **?**].

In practice, the equation (**??**) has to be solved numerically, and with shorthand notations $y = (x, v)$ and $y(n\Delta t) \approx y_n$, most existing methods can be recast into the form,

$$y_{n+1} = \Phi(y_n) + \xi_n, \tag{1.3}$$

which consists of a deterministic part that resembles a numerical method for the ODE, along with a numerical noise term $\xi_n$.

As a stochastic differential equation, there are many methods available in the literature, especially the Runge-Kutta type of methods [**?**]. Often favored by practitioners are methods that reduce to symplectic methods for solving the Hamiltonian systems, i.e., when the damping coefficient is zero. Such methods date back as early as to the work of van Gunsteren and Berendsen in [**?**], in which a third order method has been derived by first solving the second equation assuming that the force is constant or linear in time, and then update the position component accordingly. When the system

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needs to be constrained, the SHAKE algorithm [**?**] can also be incorporated. Many other integrators have also been developed. For example, the boundary method [**?**] by Brooks-Brünger and Karplus, the Langevin impulse integrator using a partition of the force into fast and slow part so that they are evaluated on different scales to minimize the force calculation effort [**?**], and the approach by Ricci and Ciccotti [**?**] using Trotter formula to split time ordered exponentials. Many of these methods have been discussed in the book [**?**]. For the random term, methods have also been proposed to construct a random walk based on the probability density associated with the Fokker-Planck equation [**?**]. The integration scheme adopted in this paper is a stochastic velocity Verlet method [**?**], which can be viewed as a second order truncation of the method from van Gunsteren and Berendsen [**?**]. This method has been selected here due to the fact that it has been widely implemented in computer packages.

Analysis of various Langevin integrators can be found in [**?**, **?**, **?**]. The focus of the work of Burrage et al [**?**] and Leimkuhler et al [**?**] is probably the closest to the present paper. In [**?**] they studied several numerical schemes, obtained the covariance of the solution, and examined the consistency with the corresponding property of the exact solution. In particular, they emphasized the influence of the damping coefficient. In [**?**], the analysis was aimed at the error estimate in terms of the equilibrium probability density, especially for several operator-splitting methods.

From a practical viewpoint, one would like to take a stepsize $\Delta t$ as large as possible to reach a longer time scale. This is owing to the multiscale nature of most of the molecular systems. For the deterministic part, the maximum stepsize, $\Delta t_{\max}$, as determined from a linear stability analysis, is typically inversely proportional to the maximum frequency (the square root of the maximum eigenvalue of the Hessian) of the molecular system, is typically on the scale of femto-seconds ($10^{-15}s$), while the motions of biomolecules range from nano-seconds $10^{-9}s$ to seconds [**?**]. When the friction coefficient is large, this threshold might be increased. Within such linear analysis, the stochastic dynamics will remain stable under such condition, since the stochastic forcing only acts as a non-homogeneous term, and it will not alter the stability property. However, for nonlinear problems, the stability issue becomes quite subtle, e.g., see the mathematical analysis in [**?**]. Nevertheless, our experiences with several existing methods are that the covariance of the displacement tends to be over-estimated, especially when the system is projected to low-frequency modes. Further, when such methods are applied to a bio-molecular model with complex force field, e.g., CHARMM [**?**], the over-estimated variance would destroy the rigid molecular structure and the simulations will have to be terminated. Therefore, the stepsize is usually far below $\Delta t_{\max}$.

Motivated by these observations, we propose to study a discrete analog of the FDT so that statistical properties are correctly predicted even when $\Delta t$ is relatively large. We notice that at the continuous level, the FDT can be established by requiring that the resulting solution follows the correct statistics. At the discrete level, it is difficult to derive a parallel result that is as explicit as the continuous FDT. One exception, nevertheless, is linear problems where the interactions are harmonic. In this case, the solution can be found by using a discrete analogue of the variation of constant formula. In particular, the equilibrium properties (covariance) and dynamic properties (time-correlation functions) can be expressed explicitly in terms of the propagation matrix.

With these results at hand, we derive the FDT *at the discrete level*. The main result is a formula for the covariance matrix associated with the discrete noise $\xi_n$. Thanks to the consistency with the discrete FDT, the computational cost is greatly reduced without compromising the statistical accuracy. Such discrete FDT is expressed as a

discrete Lyapunov equation, which also arises in linear control theory [**?**]. Therefore, the first point raised by this paper is that any one-step ODE method can be turned into a method for the Langevin dynamics by introducing the numerical additive noise according to the discrete FDT. Alternatively, one can modify a Langevin solver by retaining the deterministic part, and define the numerical noise using the discrete FDT. We will refer to this approach as a FDT-consistent method.

We will demonstrate that the widely used Euler-Maruyama method can never be FDT-consistent, regardless of the size of the time steps, implying that no stationary process can be generated from this numerical method. To further investigate the method, we consider a popular Langevin solver in molecular modeling – the stochastic velocity Verlet's method [**?**], and test our approach via a one-dimensional Lennard-Jones system. Although the analysis was for linear stochastic systems, the numerical tests will show that the FDT-consistent method can be applied to nonlinear systems as well.

Meanwhile, we have found that for sufficiently small step size, the matrix obtained from the Lyapunov equation is always positive-definite, and it can be used as the covariance of the added noise. On the other hand, for larger step sizes, we observe that depending on the numerical method, this matrix may or may not be positive-definite. Therefore, the second point raised by this paper is that in addition to the stability threshold $\Delta t_{\max}$, there is another critical threshold for the step size, under which a covariance matrix can be defined for the numerical additive noise to satisfy the discrete FDT. To elaborate on the second point, we consider several one-step methods, and compute the thresholds, with the hope to understand this new property of the solvers.

## 2. The fluctuation-dissipation theorem for general linear models

We start with a linear model and briefly discuss the general theory regarding the FDT. Consider a linear stochastic dynamics with variable $y \in \mathbf{R}^n$,

$$\dot{y} = Ay + \xi, \tag{2.1}$$

where $A$ is a matrix and $\xi(t)$ is a white noise. For stability considerations, we make the standard assumption that $A$ has eigenvalues with non-positive real parts, and those eigenvalues with zero real parts must be simple. This means that the deterministic part of the dynamics is stable [**?**]. The Gaussian noise $\xi(t)$ has mean zero and covariance $S$,

$$\left\langle \xi(t)\xi(t')^T \right\rangle = S\delta(t-t'). \tag{2.2}$$

It seems that the coefficient matrix $A$ and the covariance of the random force are independent; however, they have to satisfy certain conditions in order for the system to admit a solution as a stationary process. This is stated in the following theorem, which can be found in [**?**]. Here a stationary process $y(t)$ is defined as one with a constant mean $\mu$ and its covariance $\langle (y(t_1)-\mu)(y(t_2)^T-\mu^T)\rangle$ only depends on $t_1-t_2$ [**?**].

THEOREM 2.1. *Suppose that the initial condition $y(0)$ is Gaussian with zero mean and covariance $Q$. Then the process $y(t)$ is stationary with correlation function given by,*

$$\left\langle y(t)y(t')^T \right\rangle = e^{(t-t')A}Q, \tag{2.3}$$

*for any $t' \le t$, provided that,*

$$AQ + QA^T = -S. \tag{2.4}$$

Equation (**??**) is known as the Lyapunov equation, which explicitly expressed the relation between the covariance of the solution $y$ and the random noise $\xi$.

Now we turn to numerical methods for the stochastic model. In particular, we consider a discretization of the SDE in the following general form,

$$y_{n+1} = Gy_n + \xi_n, \quad n \geq 0. \tag{2.5}$$

In accordance with the assumption on the matrix $A$, we assume that the matrix $G$ has eigenvalues with magnitude less or equal to 1, and those eigenvalues with modulus one must be simple. This implies that the stability of the ODE $\dot{y} = Ay$ is inherited by the numerical method $y_{n+1} = Gy_n$. Interested readers are referred to [**?**] for details.

The correlation of the discrete noise is denoted by $\Sigma$. Namely, $\langle \xi_n \xi_m^T \rangle = \Sigma \delta_{n-m}$. This parameter will be determined based on an analogous result, stated as follows.

THEOREM 2.2. *Suppose that $y_0$ is Gaussian with zero mean and covariance $Q$. Then the process $\{y_n\}_{n\geq 0}$ is stationary with correlation function given by,*

$$\langle y_n \, y_m^T \rangle = G^{n-m} Q, \tag{2.6}$$

*for any $m \leq n$, provided that*

$$\Sigma = Q - GQG^T. \tag{2.7}$$

The last equation is known as the *discrete* Lyapunov equation [**?**]. Again, it dictates a relation between the covariance of the numerical solution and the random noise. This equation will be referred to as the *discrete FDT*. It provides an important guidance for choosing the additive noise in the numerical method.

To get (**??**), we start with the discrete solution $y_n$, which can be represented by

$$y_n = G^n y_0 + \sum_{\ell=0}^{n-1} G^{n-\ell-1} \xi_\ell, \tag{2.8}$$

given $y_0$ and $\xi_\ell$ as independent variables. Thus, the correlation of $y$ is given by

$$\langle y_n \, y_m^T \rangle = G^n Q (G^T)^m + \sum_{l=0}^{n-1} \sum_{\ell'=0}^{m-1} G^{n-l-1} \langle \xi_l \xi_{\ell'}^T \rangle (G^T)^{m-\ell'-1}.$$

So for $m \leq n$, we have

$$\langle y_n \, y_m^T \rangle = G^n Q (G^T)^m + \sum_{l'=0}^{m-1} G^{n-l'-1} \Sigma (G^T)^{m-l'-1}.$$

Now with (**??**), the equation (**??**) immediately follows.

The same identity has also been shown in [**?**] (equation (2.4)), but only considering the covariance matrix from numerical scheme (8). Here we also showed the correlation of the solution at different times. The deterministic term $Gy_n$, from the numerical method (**??**), can be viewed as an approximation of the SDE without the noise, i.e., the ODE. In particular, for sufficient accuracy, we must have [**?**], $G = e^{\Delta t A} + \mathcal{O}(\Delta t^{k+1})$, where $k$ is the order of the accuracy.

This provides a useful view: when $G$ is an accurate approximation of the ODE, *and* the discrete FDT holds, the covariance of the numerical solution will be exactly correct, and the time correlation is also correct within order $\Delta t^k$ over a finite period of time.

Furthermore, when the numerical method $y_{n+1}=Gy_n$ for the ODE $\dot{y}=Ay$ is stable, the corresponding numerical method for the SDE is also stable in the sense that the correlation function is bounded. Therefore, this analysis suggests that when only the equilibrium properties are of interest, one can turn any one-step ODE integrator into a stochastic one by introducing the numerical noise based on the discrete FDT (**??**).

**3. Application to Langevin dynamics models.**

The Langevin dynamics in a mass-scaled, vector form can be written as follows,

$$\begin{aligned}\dot{x}&=v,\\ \dot{v}&=-f(x)-\gamma v+w,\end{aligned} \tag{3.1}$$

with the following mass scaling introduced to (**??**): $x_i\to m_i^{\frac{1}{2}}x_i$, $v_i\to m_i^{\frac{1}{2}}v_i$, $w_i\to m^{\frac{1}{2}}w_i$. Clearly, the FDT still holds after this scaling, $\langle w_i(t)w_i(t')^T\rangle=2k_BT\gamma\delta(t-t')$. To obtain a linearized model, we simply change $f(x)$ to $-Kx$, where the matrix $K$ is the hessian matrix of the potential energy around some reference state.

For this Langevin dynamics model, there are many existing numerical methods, with different integrators and different ways to treat the additive noise. We will consider a popular method, the stochastic velocity-Verlet method (SVV), which has been discussed in several places [**?**, **?**, **?**].

**3.1. The stochastic velocity-Verlet method.** We first consider the SVV method, which has been widely implemented in software packages. We start by defining

$$c_0=e^{-\gamma\Delta t},c_1=(1-c_0)/(\gamma\Delta t),c_2=(1-c_1)/(\gamma\Delta t). \tag{3.2}$$

For the simplicity of the discussion, we have assumed the damping coefficient to be scalar constant. However, the extension to matrix-valued $\gamma$ is straightforward: We can simply use matrix exponentials to represent these parameters.

The original SVV method updates the solutions as follows,

$$x_{n+1}=x_n+c_1\Delta tv_n+c_2\Delta t^2f(x_n)+\delta x_n, \tag{3.3}$$

$$v_{n+1}=c_0v_n+(c_1-c_2)\Delta tf(x_n)+c_2\Delta tf(x_{n+1})+\delta v_n. \tag{3.4}$$

Here $\xi_n=(\delta x_n,\delta v_n)^T$ is the discrete noise. The covariance is given by,

$$\langle\delta x_n,\delta x_n\rangle=\frac{k_BT\Delta t}{\gamma}\big(2-\frac{3-4c_0+c_0^2}{\gamma\Delta t}\big), \tag{3.5}$$

$$\langle\delta v_n,\delta v_n\rangle=k_BT(1-c_0^2),\quad \langle\delta x_n,\delta x_v\rangle=k_BT\gamma\Delta t^2c_1^2. \tag{3.6}$$

For the linear model, the above formulation is equivalent to

$$\begin{pmatrix}x_{n+1}\\ v_{n+1}\end{pmatrix}=G\begin{pmatrix}x^n\\ v_n\end{pmatrix}+\xi_n. \tag{3.7}$$

The propagation matrix for the deterministic part, denoted by $G$, is given by,

$$G=\begin{bmatrix}1-c_2K\Delta t^2 & c_1\Delta t\\ c_2^2K^2\Delta t^3-c_1K\Delta t & -c_1c_2K\Delta t^2+c_0\end{bmatrix}. \tag{3.8}$$

**3.2. FDT-consistent methods.** We continue from previous section, and consider the stochastic algorithm (**??**) According to Theorem 2.2, we need to choose $\xi_n$ such that (**??**) is satisfied.

We first discuss the initial covariance $Q$. Recall the linearized Langevin equation,

$$\begin{aligned} \dot{u} &= v, \\ \dot{v} &= -Ku - \gamma v + w. \end{aligned} \tag{3.9}$$

Here we introduce the variable $u$ to represent the displacement. The total energy is written as, $H = \frac{1}{2}v^T v + \frac{1}{2}u^T K u$. For the potential energy, the matrix may have a null space, corresponding to the conservation of momentum. Let $\Phi$ be the matrix with columns that span the null space. Then the probability density is defined as follows,

$$\rho = \frac{1}{Z} e^{-\beta H} \delta(\Phi^T u)\delta(\Phi^T v) du dv, \quad \beta = \frac{1}{k_B T}. \tag{3.10}$$

We have the following theorem for the correlation of the displacement:

THEOREM 3.1. *The correlation of the displacement is given by,*

$$\langle uu^T \rangle = k_B T K^-, \tag{3.11}$$

*where $K^-$ is the generalized inverse, defined to satisfy the property that $KK^-K = K$.* This can be proved by a change of variables using the eigenvectors of the symmetric matrix $K$. Since $\Phi$ already contains some of the eigenvectors, the delta functions can be integrated out. The remaining part becomes a Gaussian integral and the covariance can be easily calculated.

Now with the initial covariance $Q$, the FDT-consistent scheme becomes straight forward to implement. In practice, $Q$ may be obtained from a normal mode analysis.

**3.3. The positive-definiteness of the covariance matrix** In implementing the FDT-consistent method, the covariance matrix $\Sigma$ needs to be at least semi-positive definite, so that a singular value decomposition (SVD) type of algorithm can be used to sample $\xi_n$. We first show that this is the case for sufficiently small step size.

THEOREM 3.2. *Assume that the numerical method $y_{n+1} = Gy_n$ is a consistent approximation of the ODE: $\dot{y} = Ay$, and the noise in the stochastic model (**??**) is non-degenerate. Then $\Sigma$ is semi positive-definite for sufficiently small $\Delta t$.*

The consistency of the approximation $y_{n+1} = Gy_n$ implies that [**?**],

$$G = I + \Delta t A + \mathcal{O}(\Delta t^2).$$

A direct substitution into the discrete FDT (**??**) yeilds

$$\Sigma = Q - GQG^T = -\Delta t(AQ + QA^T) - \mathcal{O}(\Delta t^2).$$

Let us revisit the SVV method and demonstrate how the covariance matrix $\Sigma$ can be obtained. To begin, we define,

$$L = \begin{bmatrix} I & 0 \\ c_2 K \Delta t & I \end{bmatrix}, \quad R = \begin{bmatrix} I - c_2 K \Delta t^2 & c_1 \Delta t I \\ (c_2 - c_1) K \Delta t & c_0 I \end{bmatrix}. \tag{3.12}$$

We may write the SVV scheme in the following compact form,

$$Ly_{n+1} = Ry_n + \xi. \tag{3.13}$$

Therefore, the discrete FDT reads,

$$LQL^T - RQR^T = \Sigma. \tag{3.14}$$

With direct calculations, we find that,

$$\Sigma = k_B T \begin{bmatrix} (2c_2 - c_1^2)\Delta t^2 I - c_2^2 K \Delta t^4 & c_1(1-c_0)\Delta t I + (c_2^2 - c_1 c_2) K \Delta t^3 \\ c_1(1-c_0)\Delta t I + (c_2^2 - c_1 c_2) K \Delta t^3 & (1-c_0^2) I + (2c_1 c_2 - c1^2) K \Delta t^2 \end{bmatrix}. \tag{3.15}$$

As $\Delta t \to 0$, we have,

$$(2c_2 - c_1^2)\Delta t^2 = \frac{2}{3}\gamma \Delta t^3 + \mathcal{O}(\Delta t^4), c_1(1-c_0)\Delta t = \gamma \Delta t^2 + \mathcal{O}(\Delta t^3), 1 - c_0^2 = 2\gamma \Delta t + \mathcal{O}(\Delta t^2).$$

Therefore, the leading term in $\Sigma$ is given by,

$$\Sigma \approx k_B T \begin{bmatrix} \frac{2}{3}\gamma \Delta t^3 & \gamma \Delta t^2 \\ \gamma \Delta t^2 & 2\gamma \Delta t \end{bmatrix}, \tag{3.16}$$

which is clearly positive-definite when the step size is sufficiently small. Meanwhile, for the popular Euler-Maruyama method, a direct calculation yields the covariance matrix,

$$\Sigma \approx k_B T \begin{bmatrix} -\Delta t^2 & \gamma \Delta t^2 \\ \gamma \Delta t^2 & 2\gamma \Delta t - (\gamma^2 + K)\Delta t^2 \end{bmatrix}, \tag{3.17}$$

which clearly is indefinite. Therefore, no FDT-consistent method can be built upon it.

At this point, we can invoke the FDT in the continuous case (**??**). Since the noise is non-degenerate, the covariance is positive-definite, so we obtain, $\Sigma = \Delta t S + \mathcal{O}(\Delta t^2)$, which shows the stated result. However, for a large step size, $S$ might be indefinite. Then some modification needs to be introduced. For example, a natural approximation is given by,

$$\Sigma \approx \Sigma_1 \overset{\text{def}}{=} \Delta t S \tag{3.18}$$

If the numerical method $y_{n+1} = G y_n$ is a higher-order approximation of $y' = Ay$, i.e.,

$$G = I + \Delta t A + \frac{1}{2}\Delta t^2 A^2 + \mathcal{O}(\Delta t^3),$$

a similar substitution leads to,

$$\Sigma = Q - GQG^T = -\Delta t(AQ + QA^T) - \frac{1}{2}\Delta t^2 (A^2 Q + 2AQA^T + Q(A^T)^2) + \mathcal{O}(\Delta t^3).$$

Together with the FDT (**??**), one can find another approximation,

$$\Sigma \approx \Sigma_2 \overset{\text{def}}{=} \Delta t S - \frac{1}{2}\Delta t^2 (AS + SA^T). \tag{3.19}$$

**4. Extension to nonlinear problems.** Here we briefly discuss the extension to nonlinear problems. We again consider the SVV's method as a concrete example. We will treat the nonlinear force as a separate variable, and define,

$$\widehat{L} = \begin{bmatrix} I & 0 & 0 \\ 0 & I & (1 - c_2 \Delta t) I \end{bmatrix}, \quad \widehat{R} = \begin{bmatrix} I & c_1 I \Delta t & c_2 \Delta t^2 I \\ 0 & c_0 I & (c_2 - c_1) \Delta t I \end{bmatrix}, \quad \widehat{y} = (x, v, f(x))^T. \tag{4.1}$$

Using previous notations, we can write the SVV method as: $\widehat{L}\widehat{y}_{n+1} = \widehat{R}\widehat{y}_n + \xi_n$. Further, one can show that,

$$\widehat{Q} \overset{\text{def}}{=} \langle \widehat{y}\widehat{y}^T \rangle = k_B T \begin{bmatrix} M & 0 & -I \\ 0 & I & 0 \\ -I & 0 & K \end{bmatrix}, \quad \text{with} \quad M = \beta \langle x x^T \rangle, \quad K = \beta \langle f f^T \rangle. \tag{4.2}$$

With direct calculations, one can show that the variance $\Sigma$ is exactly the same as the one in the linear case (**??**), so long as $K$ is defined according to (**??**) as the force-force correlation. For some stochastic models, e.g., the discrete Burgers equations, the equilibrium density is Gaussian and the force terms are quadratic, and the explicit expression of the matrix $K$ is available. In more general cases, the matrix $K$ would need to be pre-computed, e.g., by Monte Carlo methods.

Finally, we summarize the steps needed to obtain and implement an FDT-consistent method: (1) Starting with an ODE solver, identify the matrices in (**??**); (2) Compute the force correlation in (**??**); (3) Compute the covariance matrix $\Sigma$ from (**??**); (4) Sample $\xi$ and apply the formula (**??**) to get solutions at the next time step.

**5. Numerical tests.** In out tests, we consider a chain of 128 atoms, interacting via the Lennard-Jones potential, with spacing $2^{\frac{1}{6}}$ nearest neighbor interactions. $k_B T = 10^{-4}$. By choosing the energy and length unit to be 1, we found the maximum frequency to be around 8. For the ODE, the stability threshold for a Verlet-type of method is about 1/4. For comparison, we first generate a solution using $\Delta t = 0.008$. We will use this result as the "exact" solution. The duration is set to $T = 80000$ to ensures that we have enough data for the analysis. We perform simulations using stochastic velocity-Verlet (SVV) method, and the FDT-consistent method with the same deterministic part.

**5.1. Comparison with the stochastic Velocity-Verlet** We first show the results from an experiment with step size $\Delta t = 0.16$, which is close to the threshold. The results are shown in Figure **??**. While the time correlation of the velocity has been well predicted by both the SVV and the FDT-consistent method, for the displacement, the FDT-consistent method is superior to the original SVV method.

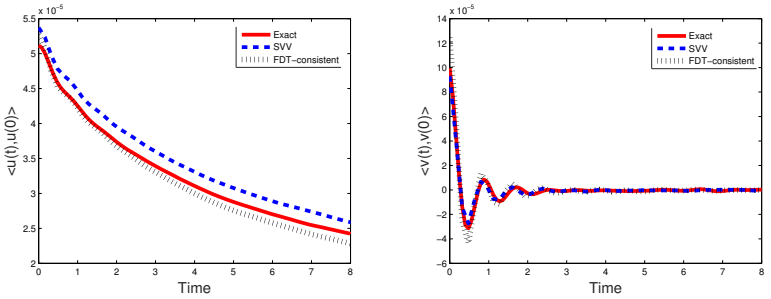


Fig. 5.1: The auto-correlations ($\Delta t = 0.16$). Left: Displacement; Right: Velocity.

Next we show simulation results obtained at much higher temperature $k_B T = 10^{-2}$ and step size $\Delta t = 0.2$, which is even closer to the threshold. In this case, the linear approximation is too crude since the system is no longer confined to the bottom of the energy well. Therefore, we use the nonlinear version, in which the matrix $K$ is

precomputed. More specifically, we consider the atom chain with nearest neighbor interaction under periodic boundary conditions (PBC). In this case, the many particle probability density can be converted into a one-dimensional one [**?**]. In particular, the matrix $K$ is simply a tri-diagonal Toeplitz matrix, and the elements can be computed using a straightforward quadrature approximation. We show the resulting statistics in Figure **??** for the velocity of the 10th atom and the relative displacement $u_{10}-u_9$, which can be interpreted as the strain. Again, compared to the original SVV method, the FDT-consistent scheme produces more accurate statistics.

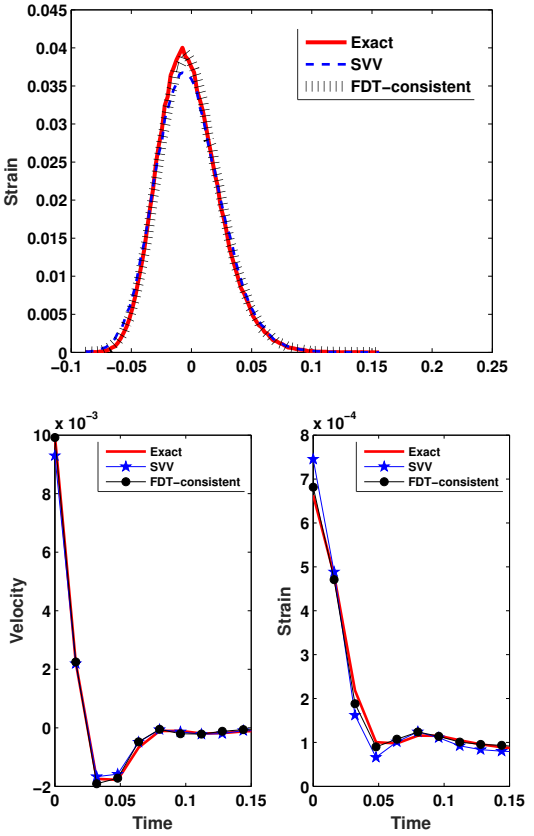


Fig. 5.2: Comparison of the statistics among the exact solution, the SVV method, and the modification using a **nonlinear** FDT-consistent approach with $\Delta t = 0.2$. Top: histogram of the strain; Bottom: auto-correlation functions.

**5.2. The positive-definiteness of $S$ for several integrators.** When the step size is increased to $\Delta t = 0.2$, the matrix $\Sigma$ obtained from the SVV method becomes indefinite. This property is associated with the particular method. Therefore, we picked several other numerical methods, including the SVV method, the operator-splitting method (OS) [**?**], the second and third order Runge-Kutta methods and the one-stage implicit Runge-Kutta method derived from the mid-point rule [**?**], and examined the corresponding matrix $G$ and $S$. In particular, we computed $\lambda_{\max}(G)$, which is pertinent to the numerical stability, and $\lambda_{\min}(\Sigma)$, which indicates the positive-definiteness. The

| Methods | $\Delta t = 0.08$ | | $\Delta t = 0.2$ | |
|---|---|---|---|---|
| | $\lambda_{\min}(\Sigma)$ | $\lambda_{\max}(G)$ | $\lambda_{\min}(\Sigma)$ | $\lambda_{\max}(G)$ |
| SVV | 0.0000 | 0.9963 | -0.0119 | 0.9991 |
| OS | 0.0001 | 0.9963 | -0.0117 | 0.9991 |
| RK2 | -0.0001 | 0.9963 | -0.81971 | 1.2840 |
| RK3 | 0.0002 | 0.9963 | 0.0019 | 0.9991 |
| Im RK | 0.0000 | 0.9963 | 0.0000 | 0.9991 |

Table 5.1: The comparison of several ODE methods.

results are shown in the table **??**. We observe the first three methods yield a matrix $S$ that is not positive-definite. The 3rd order Runge-Kutta method and the implicit Runge-Kutta method are more robust from this perspective, in that they give a positive-definite matrix $S$ even when the step size is large. However, compared to the SVV method, their algorithms involve more force evaluations within one step or solutions of nonlinear equations, and therefore are more expensive to implement.

**6. Conclusion** We proposed a FDT-consistent method to deal with the additive noise in Langevin equations with the goal to guarantee that the result has sufficient statistical accuracy. A discrete FDT is derived based on the general linear Langevin dynamics model. We made the observation that our FDT-consistent method is straightforward to construct, and works well in terms of predicting the time correlation functions. It would be of practical interest to apply such methods to bio-molecular systems to assess the computational speedup. This work is underway.

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