

EXACT AND NON-STIFF SAMPLING OF HIGHLY OSCILLATORY SYSTEMS: AN IMPLICIT MASS-MATRIX PENALIZATION APPROACH

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Abstract. We propose and analyze an implicit mass-matrix penalization (IMMP) technique which enables efficient and exact sampling of the (Boltzmann/Gibbs) canonical distribution associated with highly oscillatory systems. The penalization is based on an extended Hamiltonian with artificial constraints associated with each expected fast degree of freedom (fDOF). The penalty parameters enable arbitrary tuning of the timescale for the selected fDOFs. Associated (stochastic) numerical methods are shown to be dynamically consistent when the penalty vanishes with the time-step, and always statistically exact with respect to canonical distributions for any chosen penalty. Moreover, the IMMP method is shown to be asymptotically stable in the infinite stiffness limit, converging towards standard effective dynamics on the slow manifold. It can be easily implemented from standard geometric integrators with algebraic constraints given by the slow manifold, and has no additional complexity in terms of enforcing the constraint and force evaluations. For high dimensional systems, the IMMP method enables a tunable slowdown of high frequencies thereby relaxing time-step stability restrictions, while, at the same time, conserving macroscopic features of the system's dynamics. This property is proved rigorously for a linear harmonic atomic chain, and numerical evidence is given in the case of non-linear interactions.

Key words. stochastic differential equations, Monte Carlo methods, Langevin dynamics, equilibrium measure, NVT ensemble, constrained Hamiltonian dynamics.

AMS subject classifications. 65C05, 65C20, 82B20, 82B80, 82-08

Introduction. This paper deals with stochastic numerical integration and sampling of thermostatted Hamiltonian systems with multiple timescales. The main motivation is to develop numerical methods which sample *exactly* canonical distributions, while integrating the fastest degrees of freedom only statistically. Furthermore, one also seeks good approximation of dynamical behavior at large temporal scales.

Hamiltonian systems with multiple timescales typically appear in molecular dynamics (MD) simulations, which have become, with the aid of increasing computational power, a standard tool in many fields of physics, chemistry and biology. However, extending the simulations to physically relevant time-scales remains a major challenge for various large molecular systems. Due to the complexity of implicit methods the time scales reachable by standard numerical methods are usually limited by the rapid oscillations of some particular degrees of freedom. Yet the physical necessity of resolving the fast degrees of freedom in simulations is often ambiguous. Thus efficient treatment of the fast time scales has motivated new interest in developing numerical schemes for the integration of such stiff systems. Broadly speaking, one may start by recognizing two approaches to the numerical treatment of such stiff oscillatory systems:

- (i) Semi-implicit, multi-step integrators and their variants (e.g., the textbooks [21, 13], or the review paper [8] and references therein), which attempt to resolve microscopic highly oscillatory dynamical behavior.
- (ii) Methods with direct constraints, where the highly oscillatory degrees of freedom are constrained to their equilibrium value (e.g., [21, 16, 27] and references therein).

In spite of their differences, the common key feature of all these methods is to balance, at some point, a trade-off between stability restrictions and implicit time-stepping form. Equivalently, this can be summarized as the trade-off between the computational effort associated with small time steps, and the computational cost of solving implicit equations implied by the stiffness.

The method developed in this paper belongs to the second family of these two approaches. Although constrained dynamics removes, in principle, the stiffness of the associated numerical scheme, it introduces new difficulties and numerical problems. As an approximation to the original dynamics, it modifies important features of the system; for example, the associated statistical distribution. In this paper we treat

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the case of canonical distributions only. The principal goal of the proposed method is to replace direct constraints by implicit mass-matrix penalization (IMMP), Section 2, which integrates fDOFs, but with a tunable mass penalty. This approach guarantees that the canonical distribution is computed *exactly*, and arguably, some “macroscopic dynamics” are partially preserved. The idea of adjusting mass tensors goes back to [1], however, to our knowledge, such strategies has not been explicitly developed in practical MD simulations. We present numerical analysis of the method and simulations that demonstrate properties of the proposed algorithms.

The “fast” degrees of freedom (denoted by ξ) are chosen arbitrarily, and are penalized with an additional mass-matrix $\nu^2 M_z$ orthogonal to the expected slow manifold (the parameter ν denotes the penalty intensity). If a stiffness parameter ϵ is introduced, the penalty intensity ν can be tuned in order to obtain asymptotically stable dynamics in the limit $\epsilon \rightarrow 0$. More precisely, we obtain the following desirable properties:

1. The canonical equilibrium distribution in position is independent of the penalty ν .
2. The limit of vanishing penalization ($\nu = 0$) is the original full dynamics, thus enabling the construction of consistent numerical schemes.
3. The limit of the infinite stiffness ($\epsilon \rightarrow 0$ with $\epsilon\nu \rightarrow \bar{\nu}$) is the effective constrained dynamics on the slow manifold.
4. The computational complexity of numerical integrators is equivalent to the complexity of standard integrators for effective dynamics on the slow manifold.

From the practical point of view, the key point is to use an implicit representation of the mass penalty with the aid of an extended Hamiltonian with artificial constraints. The “stiff” part of the potential is applied to auxiliary degrees of freedom endowed with a “virtual” mass-matrix $\nu^2 M_z$. The extended Hamiltonian obtained in this way is then constrained by identifying the auxiliary variables and the fDOFs ξ . The typical time scale of the fDOFs is thus tuned by the mass penalty ν , while keeping the canonical distribution on positions invariant.

When working with the constrained dynamics it is important to take into account the geometric bias introduced by non-linear constraints. This bias is corrected by introducing an effective potential, the so-called Fixman corrector, which can be computed explicitly using the co-area formula. The Fixman corrector of the IMMP methods is a ν -perturbation of the Fixman corrector associated with constraints on the slow manifold. Note, that by using a Metropolis acceptance/rejection rule, which is reminiscent of Hybrid Monte-Carlo (HMC) methods, the forces associated with the Fixman corrector need not be computed. We introduce highly oscillatory systems and their statistical properties in Section 1.

The numerical method is presented as an exact discretization of Langevin processes with constraints, using splitting between the Hamiltonian dynamics and the fluctuation/dissipation perturbation. A Metropolis correction (HMC) is then added to obtain a Markov chain which samples exactly, under the usual ergodicity assumptions, the Boltzmann canonical distribution in positions of the original highly oscillatory system, Section 3. This choice is only a presentation matter, and for practical purposes one can refer to ones’ favorite numerical integrator for Hamiltonian systems with or without fluctuation/dissipation perturbations. We analyze the infinite stability limit and stability properties of the proposed scheme in Section 4.

High-dimensional systems usually contain a large variety of timescales, and are therefore challenging test cases. For a given re-scaled mass-penalty $\bar{\nu} = \frac{\nu}{N}$, it is shown that the IMMP induces a high frequency filter with tunable cut-off (slowdown), which relaxes time-step stability restrictions typically by a factor N . Then numerical evidence are given of the continuous dependence of the macroscopic dynamics of a tagged particle of an anharmonic chain with respect to the scaled penalty $\bar{\nu}$. In particular, the convergence rate towards equilibrium of the position of the tagged particle is affected by a small penalty $\bar{\nu}$ in a continuous manner. A rigorous analysis for the linear case (i.e., harmonic potentials) is provided in Section 5, and the method is numerically tested for a non-linear chain of oscillators in Section 6. The numerical study demonstrates the ability of the IMMP method to be statistically exact and to approximate macroscopic dynamical properties such as equilibrium convergence, while enabling arbitrary large relaxation of time-step

restriction.

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1. Highly oscillatory systems with a thermostat. We consider an Hamiltonian system in the phase-space $\mathbb{R}^d \times \mathbb{R}^d$ with the Hamiltonian H in the following form

$$H(p, q) = \frac{1}{2}p^T M^{-1}p + V(q, \xi(q)). \quad (1.1)$$

We use the notation in which the Euclidean scalar product of two vectors $p_1, p_2 \in \mathbb{R}^d$ is $p_1^T p_2$. The mapping $\xi : \mathbb{R}^d \rightarrow \mathbb{R}^n$, defines $n \leq d$ degrees of freedom, given by smooth functions $q = (q_1, \dots, q_d) \mapsto \xi_1(q), \dots, \xi_n(q)$ taking values in a neighborhood of the origin. We assume that the mapping ξ is regular (i.e., with non-degenerate Jacobian) in an open δ -neighborhood $\mathcal{O}_\delta = \{q \mid \|\xi(q)\| < \delta\}$ of $\xi^{-1}(0)$, hence defining a smooth sub-manifold of \mathbb{R}^d denoted $\mathcal{M}_0 = \xi^{-1}(0)$.

The dependence of the potential $z \mapsto V(q, z)$ is expected to be “stiff” in the second variable, and the functions ξ will thus be called “fast” degrees of freedom (fDOFs). Throughout the paper, we often assume that a stiffness parameter ϵ can be explicitly identified, and that the potential energy V can be written in the following form

$$V(q, \xi(q)) = V^\epsilon(q, \xi(q)) = U(q, \frac{\xi(q)}{\epsilon}),$$

with a confining assumption

$$\inf_{q \in \mathbb{R}^d} U(q, z) \geq K(z),$$

where $\lim_{z \rightarrow \infty} K(z) = +\infty$ strictly faster than at any logarithmic rate. The fast degrees of freedom ξ of states at a given energy remain in a closed neighborhood of the origin as the stiffness parameter vanishes ($\epsilon \rightarrow 0$). In this limit the system is confined to the sub-manifold \mathcal{M}_0 , which is usually called the “slow manifold”. For the sake of simplicity, the symmetric positive definite mass-matrix M of the system is taken independent of positions, and typically it is diagonal.

When the system is thermostatted, i.e., kept at the constant temperature, the long time distribution of the system in the phase-space is given by the canonical equilibrium measure at the inverse temperature β (also called NVT distribution) given by

$$\mu(dp dq) = \frac{1}{Z} e^{-\beta H(p, q)} dp dq, \quad Z = \int_{\mathbb{R}^d \times \mathbb{R}^d} e^{-\beta H(p, q)} dp dq, \quad (1.2)$$

with the normalization constant assumed to be finite. The standard dynamics used to model thermostatted systems are given by Langevin processes.

DEFINITION 1.1 (Langevin process). *A Langevin process at the inverse temperature β with the Hamiltonian $H(q, p)$, $(p, q) \in \mathbb{R}^d \times \mathbb{R}^d$, the $d \times d$ dissipation matrix γ , and the fluctuation matrix σ is given by the stochastic differential equations*

$$\begin{cases} \dot{q} = \nabla_p H \\ \dot{p} = -\nabla_q H - \gamma \dot{q} + \sigma \dot{W}, \end{cases} \quad (1.3)$$

where \dot{W} is the standard multi-dimensional white noise, and the fluctuation matrix $\sigma \in \mathbb{R}^d \times \mathbb{R}^d$ satisfies the fluctuation-dissipation identity

$$\sigma\sigma^T = \frac{2}{\beta}\gamma.$$

For any γ , the process is reversible with respect to the stationary canonical distribution (1.2). Furthermore, if γ is strictly positive, the process is ergodic (see Appendix B). Throughout this paper, the usual global Lipschitz conditions (see [17]) on H and ξ are assumed, thus ensuring well-posedness of the considered stochastic differential equations.

REMARK 1.2. The analysis does not exclude a more general case when the dissipation matrix γ can be a position dependent tensor $\gamma(q)$.

When a stiffness parameter is introduced, the canonical distribution becomes

$$\mu_\epsilon(dp dq) = \frac{1}{Z_\epsilon} e^{-\beta(\frac{1}{2}p^T M^{-1}p + U(q, \frac{\xi(q)}{\epsilon}))} dp dq. \quad (1.4)$$

In the infinite stiffness limit ($\epsilon \rightarrow 0$), the measure concentrates on the slow manifold \mathcal{M}_0 . The limit is computed using the co-area formula (see Appendix A for relevant definitions of surface measures). In order to characterize the limiting measure we introduce the effective potential

$$V_{\text{eff}}(q) = -\frac{1}{\beta} \ln \int e^{-\beta V(q,z)} dz. \quad (1.5)$$

LEMMA 1.3. In the infinite stiffness limit ($\epsilon \rightarrow 0$), the highly oscillatory canonical distribution (1.4) converges $\mu_\epsilon \rightarrow \mu_0$ (in distribution) towards $\mu_0(dp dq)$, which is supported on \mathcal{M}_0 , and defined as

$$\mu_0(dp dq) = \frac{1}{Z_0} e^{-\beta(\frac{1}{2}p^T M^{-1}p + V_{\text{eff}}(q))} dp \delta_{\xi(q)=0}(dq). \quad (1.6)$$

Its marginal distribution in position is given, up to the normalization, by

$$e^{-\beta V_{\text{eff}}(q)} \delta_{\xi(q)=0}(dq). \quad (1.7)$$

Proof. It is sufficient to consider distributions in the position variable q only. Let \mathcal{U}^δ be a δ -neighborhood of \mathcal{M}_0 where

$$dq = \epsilon^n \delta_{\xi(q)=\epsilon z}(dq) dz.$$

We construct a decomposition $\varphi = \varphi_1 + \varphi_2$ of continuous bounded observables such that $\text{supp } \varphi_1 \subset \mathcal{U}^\delta$ and $\text{supp } \varphi_2 \cap \mathcal{U}^\delta = \emptyset$. Using the confining property of $U(q, \cdot)$ we obtain

$$\int \varphi(q) e^{-\beta U(q, \frac{\xi(q)}{\epsilon})} dq = \epsilon^n \int \varphi_1(q) e^{-\beta U(q,z)} \delta_{\xi(q)=\epsilon z}(dq) dz + \mathcal{O}(e^{-\beta K(\delta/\epsilon)}).$$

By continuity of $\epsilon \mapsto \int \varphi_1(q) e^{-\beta U(q,z)} \delta_{\xi(q)=\epsilon z}(dq)$ and by the dominated convergence theorem

$$\int \varphi_1(q) e^{-\beta U(q,z)} \delta_{\xi(q)=\epsilon z}(dq) dz \rightarrow \int \varphi_1(q) e^{-\beta V_{\text{eff}}(q)} \delta_{\xi(q)=0}(dq) = \int \varphi(q) e^{-\beta V_{\text{eff}}(q)} \delta_{\xi(q)=0}(dq),$$

and the result follows after normalization. \square

The infinite stiffness limit ($\epsilon \rightarrow 0$) of highly oscillatory dynamics has been studied in a series of papers [22, 25, 15, 3, 19, 20]. The limiting dynamics can be fully characterized in special cases. For

example, when the highly oscillatory potential is linear and non-resonant (at least almost everywhere on the trajectory, see [25]), it can be described through adiabatic effective potentials. However, when the system is thermostatted, one can postulate an “ad hoc” effective dynamics ([20]) exhibiting the appropriate limiting canonical distribution given by (1.7). Such dynamics can be obtained by constraining the system to the slow manifold \mathcal{M}_0 , and adding a correcting entropic potential (sometimes called “Fixman” corrector from [12]), which is due to the geometry of \mathcal{M}_0 . This correcting potential is given by

$$V_{\text{fix}}(q) = \frac{1}{2\beta} \ln (\det G(q)) , \quad (1.8)$$

where $G(q)$ is the $n \times n$ Gram matrix associated with the fast degrees of freedom

$$G(q) = \nabla_q^T \xi M^{-1} \nabla_q \xi ,$$

where we use the notation

$$(\nabla_q \xi)_{ij} = (\nabla_q^T \xi)_{ji} = \frac{\partial \xi_i}{\partial q_j} , \quad i = 1, \dots, n , \quad j = 1, \dots, d .$$

In general, since the effective potential (1.5) is not explicit, one may need to couple the system with virtual fast degrees of freedom to enforce the appropriate effective dynamics associated with (1.5). The resulting extended Hamiltonian is then defined on the state space $T^*(\mathcal{M}_0 \times \mathbb{R}^n)$ (the cotangent bundle) and is given by

$$\begin{cases} H_{\text{eff}}(p, p_z, q, z) = \frac{1}{2} p^T M^{-1} p + \frac{1}{2} p_z^T M_z^{-1} p_z + U(q, z) + V_{\text{fix}}(q) \\ \xi(q) = 0 . \end{cases} \quad (C) \quad (1.9)$$

DEFINITION 1.4 (Effective Langevin process with constraints). *The constrained Langevin process associated with Hamiltonian (1.9) is defined by the following stochastic differential equations*

$$\begin{cases} \dot{q} = M^{-1} p \\ \dot{z} = M_z^{-1} p_z \\ \dot{p} = -\nabla_q U(q, z) - \nabla_q V_{\text{fix}}(q) - \gamma \dot{q} + \sigma \dot{W} - \nabla_q \xi \dot{\lambda} \\ \dot{p}_z = -\nabla_z U(q, z) - \gamma_z \dot{z} + \sigma_z \dot{W}_z \\ \xi(q) = 0 , \end{cases} \quad (C) \quad (1.10)$$

where \dot{W} (resp. \dot{W}_z) is the standard multi-dimensional white noise, γ (resp. γ_z) a $d \times d$ (resp. $n \times n$) non-negative symmetric dissipation matrix, σ (resp. σ_z) is the fluctuation matrix satisfying $\sigma \sigma^T = \frac{2}{\beta} \gamma$ (resp. $\sigma_z \sigma_z^T = \frac{2}{\beta} \gamma_z$). The processes $\lambda \in \mathbb{R}^n$ are Lagrange multipliers associated with the constraints (C) and adapted with respect to the white noise.

We formulate reversibility of this process as a separate lemma.

LEMMA 1.5. *The process defined in (1.10) is reversible with respect to the stationary canonical distribution given by*

$$e^{-\beta H_{\text{eff}}(p, p_z, q, z)} dp_z dz \sigma_{T^* \mathcal{M}_0}(dp dq)$$

with the q -marginal

$$e^{-\beta V_{\text{eff}}(q)} \delta_{\xi(q)=0}(dq) .$$

When γ and γ_z are strictly positive, the process is ergodic.

Proof. The process (1.10) is a Langevin process with mechanical constraints, exhibiting reversibility properties with respect to the associated Boltzmann canonical measure (see the summary in Appendix B). Then the q -marginal is obtained by remarking that the integration of any function of $\frac{1}{2}p^T M^{-1}p + \frac{1}{2}p_z^T M_z^{-1}p_z$ with respect to $dp_z \sigma_{T_q^* \mathcal{M}_0}(dp)$ results in a constant independent of q . \square

The properties of thermostatted highly oscillatory systems are summarized in Table 1.1.

	Finite stiffness $\epsilon > 0$	Infinite stiffness limit $\epsilon \rightarrow 0$	Infinite stiffness $\epsilon = 0$
Dynamics	Highly oscillatory + fluct./diss.	Adiabatic (if non-resonant) + non-Markov fluct./diss.	Effective with constraints + fluct./diss.
Statistics	Canonical	Positions on \mathcal{M}_0 , free velocities.	Canonical on $T^* \mathcal{M}_0$, geometric corrector.
Numerics	Leapfrog/Verlet + fluct./diss.	Time-step restrictions ($\delta t = o(\epsilon)$)	Leapfrog/Verlet with constraints + fluct./diss.

TABLE 1.1

Stiff Hamiltonian systems and associated commonly used numerical methods. Two different schemes are required for the stiff system and its effective Markovian approximation. (\mathcal{M}_0 denotes the slow manifold).

2. Implicit mass-matrix penalization. We now turn to the definition of the implicit mass-matrix penalization (IMMP) strategy. Implicitness refers to the fact that the penalization is carried out using an extension of the state space balanced by some additional constraints. In this section and the following sections, which are devoted to the definition of the modified dynamics and its numerical integration, the stiffness parameter ϵ does not play any role.

A virtual $n \times n$ mass-matrix denoted M_z is associated with the n selected degrees of freedom ξ . M_z can be seen as a pre-conditioning matrix penalizing the dynamics of the selected degrees of freedom. The intensity of the penalty is given by the parameter ν , which enables the arbitrary tuning of the frequencies for the selected degrees of freedom. The new, penalized mass-matrix of the system is a position dependent tensor

$$M_\nu(q) = M + \nu^2 \nabla_q \xi M_z \nabla_q^T \xi, \quad (2.1)$$

with the associated modified impulses

$$p_\nu = M_\nu(q) M^{-1} p. \quad (2.2)$$

When ν becomes large, the velocities are constrained to remain tangent to the slow manifold \mathcal{M}_0 , and orthogonal motions are arbitrarily slowed down. Conversely when $\nu = 0$, one recovers the original highly oscillatory system. Since the modification in M_ν depends on the position q , new geometry is introduced and an additional correction in the potential energy is required in order to preserve original statistics in the position variable. The correction is derived from the usual Fixman corrector (1.8) as a smooth perturbation of order ν^{-2}

$$V_{\text{fix},\nu}(q) = \frac{1}{2\beta} \ln(\det(M_\nu(q))). \quad (2.3)$$

PROPOSITION 2.1. *Up to an additive constant, we have*

$$V_{\text{fix},\nu}(q) = \frac{1}{2\beta} \ln \det \left(G(q) + \frac{1}{\nu^2} M_z^{-1} \right), \quad (2.4)$$

and thus (up to additive constants)

$$\lim_{\nu \rightarrow +\infty} V_{\text{fix},\nu} = V_{\text{fix}}, \text{ and } \lim_{\nu \rightarrow 0} V_{\text{fix},\nu} = 0.$$

Proof. Using the identity for a non-diagonal matrix J of dimension $n_1 \times n_2$:

$$\det(\text{Id}_{n_1} + JJ^T) = \det(\text{Id}_{n_2} + J^T J),$$

one observes

$$\det(M_\nu) = \det(M) \det(\nu^2 M_z) \det\left(G + \frac{1}{\nu^2} M_z^{-1}\right)$$

from which the expression for the corrected Fixman potential follows. \square

The associated modified Hamiltonian is then given by

$$H_\nu(p_\nu, q) = \frac{1}{2} p_\nu^T M_\nu^{-1} p_\nu + V(q, \xi(q)) + V_{\text{fix},\nu}(q), \quad (2.5)$$

and $H_0 = H$ is simply the original Hamiltonian (1.1).

By construction, statistics of positions q of the mass penalized Hamiltonian are independent of the penalization, leading to *exact canonical statistics* in position variables.

PROPOSITION 2.2. *The canonical distribution associated with the mass-penalized Hamiltonian (2.5) is defined by*

$$\mu_\nu(dp_\nu dq) = \frac{1}{Z_\nu} e^{-\beta H_\nu(p_\nu, q)} dp_\nu dq. \quad (2.6)$$

It has the marginal probability distribution in q , up to the normalization, $e^{-\beta V(q, \xi(q))} dq$, which agrees with the original canonical distribution (1.2) in the position variables, and is independent of the mass penalization parameter ν .

Proof. The normalization of Gaussian integrals in the p_ν variables yields

$$\int e^{-\beta \frac{1}{2} p_\nu^T M_\nu^{-1} p_\nu} dp_\nu = \left(\frac{2\pi}{\beta}\right)^{d/2} \sqrt{\det(M_\nu)},$$

which is cancelled out by the Fixman corrector $V_{\text{fix},\nu}$ and the result follows. \square

The sampling of such a system can be done using the standard Langevin stochastic perturbation as detailed in Definition 1.1. However, the direct discretization of the equation of motion given by H_ν (e.g., by an explicit scheme) is bound to be unstable (from non-linear instabilities) when the fast degrees of freedom are not affine functions of the system. To construct stable schemes, one may rather use an implicit formulation of the Hamiltonian (2.5), in conjunction with a solver which enforces the constraints. To obtain such a formulation, we extend the state space with n new variables (z_1, \dots, z_n) , and associated moments $(p_{z_1}, \dots, p_{z_n})$. The auxiliary mass-matrix for the new degrees of freedom is then given by M_z . The new extended Hamiltonian of the system is now given in $\mathbb{R}^{d+n} \times \mathbb{R}^{d+n}$ with n additional constraints denoted by (C_ν)

$$\begin{cases} H_{\text{IMMP}}(p, p_z, q, z) = \frac{1}{2} p^T M^{-1} p + \frac{1}{2} p_z^T M_z^{-1} p_z + V\left(q, \frac{z}{\nu}\right) + V_{\text{fix},\nu}(q), \\ \xi(q) = \frac{z}{\nu}. \end{cases} \quad (C_\nu) \quad (2.7)$$

The equivalence of the two formulations is stated as a simple separate lemma.

LEMMA 2.3. *The equations of motion associated with the penalized mass-matrix Hamiltonian (2.5) or the extended Hamiltonian with constraints (2.7) are identical.*

Proof. The Lagrangian associated with H_{IMMP} is given by

$$L_{\text{IMMP}}(\dot{q}, \dot{z}, q, z) = \frac{1}{2} \dot{q}^T M \dot{q} + \frac{1}{2} \dot{z}^T M_z \dot{z} - V\left(q, \frac{z}{\nu}\right) - V_{\text{fix}, \nu}(q),$$

and includes hidden constraints on velocities implied by the constraints (C_ν) on positions variables

$$\dot{z} = \nu \nabla_q^T \xi \dot{q}.$$

Replacing \dot{z} and z in L_{IMMP} by their expressions as functions of \dot{q} and q , one gets the Lagrangian associated with H_ν . \square

The stochastically perturbed equations of motion of Langevin type associated with (2.7) are given by:

DEFINITION 2.4 (IMMP). *The implicit Langevin process associated with Hamiltonian H_{IMMP} and constraints (C_ν) is defined by the following equations of motion*

$$\begin{cases} \dot{q} = M^{-1} p \\ \dot{z} = M_z^{-1} p_z \\ \dot{p} = -\nabla_1 V(q, \xi(q)) - \nabla_q V_{\text{fix}, \nu}(q) - \gamma \dot{q} + \sigma \dot{W} - \nabla_q \xi \dot{\lambda} \\ \dot{p}_z = -\frac{1}{\nu} \nabla_2 V(q, \xi(q)) - \gamma_z \dot{z} + \sigma_z \dot{W}_z + \frac{1}{\nu} \dot{\lambda} \\ \xi(q) = \frac{z}{\nu}, \end{cases} \quad (2.8) \quad (C_\nu)$$

where ∇_1 and ∇_2 are respectively derivatives with respect to the first and second variable of the function $V(q, z)$. The process \dot{W} (resp. \dot{W}_z) is the standard multi-dimensional white noise, γ (resp. γ_z) a $d \times d$ (resp. $n \times n$) non-negative symmetric dissipation matrix, σ (resp. σ_z) is the fluctuation matrix satisfying $\sigma \sigma^T = \frac{2}{\beta} \gamma$ (resp. $\sigma_z \sigma_z^T = \frac{2}{\beta} \gamma_z$). The processes $\lambda \in \mathbb{R}^n$ are Lagrange multipliers associated with the constraints (C_ν) and adapted with the white noise.

This process is naturally equivalent to the explicit mass-penalized Langevin process in $\mathbb{R}^d \times \mathbb{R}^d$ associated with M_ν . Moreover, when the penalization vanishes ($\nu \rightarrow 0$), the evolution law of the process $\{p_t, q_t\}_{t \geq 0}$ or $\{(p_\nu)_t, q_t\}_{t \geq 0}$ converges towards the original dynamics.

PROPOSITION 2.5. *The stochastic process with constraints (2.8) is well-posed and equivalent to the Langevin diffusion in $\mathbb{R}^d \times \mathbb{R}^d$ (see Definition 1.1), with the mass-penalized Hamiltonian H_ν (2.5), and the dissipation matrix given by*

$$\gamma_\nu(q) = \gamma + \nu^2 \nabla_q \xi \gamma_z \nabla_q^T \xi.$$

Furthermore, the process is reversible with respect to $Z_\nu^{-1} e^{-\beta H_\nu(p_\nu, q)} dp_\nu dq$ (the stationary canonical distribution associated with H_ν). Its marginal in position variables is given, up to the normalization, by the original potential, i.e., $e^{-\beta V(q, \xi(q))} dq$.

Proof. Imposing the constraints implies $\nabla_q^T \xi M^{-1} p = \frac{1}{\nu} M_z^{-1} p_z$. Thus by the definition of p_ν we have

$$p_\nu = p + \nu \nabla_q \xi p_z.$$

Since the position process $\{q_t\}_{t \geq 0}$ is of finite variation, a short computation shows that for each coordinate $i = 1, \dots, d$

$$\dot{p}_\nu^i = \dot{p}^i + \nu \partial_{q_i} \xi \dot{p}_z + \nu^2 \dot{q}^T \nabla_q (\partial_{q_i} \xi) p_z. \quad (2.9)$$

Furthermore,

$$-\partial_{q_i} \left(\frac{1}{2} p_\nu^T M_\nu^{-1} p_\nu \right) = \partial_{q_i} \left(\frac{1}{2} \dot{q}^T M_\nu \dot{q} \right) = \nu^2 \dot{q}^T \nabla_q (\partial_{q_i} \xi) M_z \nabla_q^T \xi \dot{q},$$

and thus

$$\dot{p}_\nu = \dot{p} + \nu \nabla_q \xi \dot{p}_z - \nabla_q \left(\frac{1}{2} p_\nu^T M_\nu^{-1} p_\nu \right).$$

On the other hand

$$\nabla_q V(q, \xi(q)) = \nabla_1 V(q, \frac{z}{\nu}) + \nabla_q \xi \nabla_2 V(q, \frac{z}{\nu}).$$

Using the expressions for \dot{p} and \dot{p}_z from (2.8) in (2.9) we obtain

$$\dot{p}_\nu = -\frac{1}{2} p_\nu^T \nabla_q M_\nu^{-1} p_\nu - \nabla_q V(q, \xi(q)) - \nabla_q V_{\text{fix}, \nu}(q) - \gamma \dot{q} - \nu \nabla_q \xi \gamma_z \dot{z} + \sigma \dot{W} + \nu \nabla_q \xi \sigma_z \dot{W}_z, \quad (2.10)$$

which gives the result. \square

PROPOSITION 2.6 (Consistency). *When the penalization $\nu \rightarrow 0$, the evolution law of the processes $\{p_t, q_t\}_{t \geq 0}$ or $\{(p_\nu)_t, q_t\}_{t \geq 0}$ defined by the implicit equations (2.8) converges (in the sense of probability distributions on continuous paths endowed with the uniform convergence) towards the process solving the original Langevin dynamics (1.3).*

Proof. The stochastic differential equation defined by $\dot{q} = M_\nu^{-1} p_\nu$ and (2.10) has smooth coefficients which depend on ν in a continuous fashion ($\nu \mapsto M_\nu$ and $\nu \mapsto V_{\text{fix}, \nu}$ are continuous). Standard results on weak convergence ([9]) of stochastic processes imply the result as stated. \square

We conclude this section by discussing some consequences for numerical computations.

First, proposition 2.6 implies that numerical schemes consistent with the penalized dynamics (2.8) used with a penalization which vanishes with the time-step (at a given order) $\nu = \mathcal{O}(\delta t^{k_1})$ gives numerical schemes consistent with the original Langevin dynamics (1.3). Moreover, since the perturbation M_ν is of order ν^2 , the standard Gronwall argument shows that the time-step order of the scheme is given by $\min(2k_1, k_2)$ where k_2 is the order of the scheme with respect to the penalized dynamics (2.8).

On the other hand, for ν large enough, the mass penalization in the implicit equation of motion (2.8) can remove possibly stiff behavior of the dependence $z \mapsto V(q, z)$.

Finally, in the infinite stiffness limit (see Section 4), one has to consider large mass-penalization parameter, which results in the IMMP dynamics (2.8) convergence to standard effective constrained dynamics similar to (1.10).

3. Numerical integration. The key ingredient for achieving efficient numerical simulation is to use an integrator that enforces the constraints associated with the implicit formulation of the mass penalized dynamics (2.8). The implicit structure of (2.8) leads to numerical schemes that are potentially asymptotically stable in stiff cases, converging to usual schemes for effective dynamics on the slow manifold \mathcal{M}_0 , such as (1.10).

On the other hand, when the penalization ν vanishes with the time-step, the scheme becomes consistent with respect to the original dynamics (1.3). One may then consider the mass-penalization introduced here as a special method of pre-conditioning for a stiff ODE system with an “implicit” (in the time evolution sense) structure. Here, the “implicit” structure amounts to solving the imposed constraints $\xi(q) = \frac{z}{\nu}$ in (2.8).

It lies outside the scope of this paper to make a review of standard numerical methods for constrained mechanical systems, we refer to [13] as a classical textbook, and to the series ([27, 23, 28, 5, 6]) as a sample of works on practical developments of numerical methods. In the appendix we describe in

a greater detail the classical leapfrog/Verlet scheme that enforces constraints, which is usually called RATTLE. The scheme is second order, reversible and symplectic. As an option, one can also add a Metropolis acceptance/rejection rule at each time step. If the underlying integrator is reversible and preserves the phase space measure, this leads to a Hybrid Monte Carlo scheme which *exactly* preserves canonical distributions (see [7, 14, 24] for more details on HMC methods). The numerical discretization of the Langevin process with constraints (2.8) is then obtained for instance by splitting the Hybrid Monte Carlo part and the Gaussian fluctuation/dissipation perturbation. A review of standard numerical schemes for Langevin dynamics is found, for example, in [4].

We recall that we consider the IMMP dynamics (2.8), which consists of a Langevin process with constraints defined by the following elements:

(i) the Hamiltonian (2.7)

$$\begin{cases} H_{\text{IMMP}}(p, p_z, q, z) = \frac{1}{2}p^T M^{-1}p + \frac{1}{2}p_z^T M_z^{-1}p_z + V(q, \frac{z}{\nu}) + V_{\text{fix},\nu}(q), \\ \xi(q) = \frac{z}{\nu}, \end{cases} \quad (C_\nu)$$

(ii) the dissipation matrix $\begin{pmatrix} \gamma & 0 \\ 0 & \gamma_z \end{pmatrix}$, and (iii) the inverse temperature β . A key point in Monte Carlo methods is that the Hamiltonian of the integrator can be modified as follows

$$\begin{cases} \tilde{H}_{\text{IMMP}}(p, p_z, q, z) = \frac{1}{2}p^T M^{-1}p + \frac{1}{2}p_z^T M_z^{-1}p_z + \tilde{V}_\nu(q) \\ \xi(q) = \frac{z}{\nu} \end{cases} \quad (C_\nu) \quad (3.1)$$

where \tilde{V}_ν is a potential that can be chosen arbitrarily. Indeed, only the underlying phase space structure is necessary, and (3.1) has the same hidden constraints on impulses and thus the same phase space structure as (2.7). In our context, it can be beneficial to use the simplified potential

$$\tilde{H}_{\text{IMMP}}(p, p_z, q, z) = \frac{1}{2}p^T M^{-1}p + \frac{1}{2}p_z^T M_z^{-1}p_z + V(q, \frac{z}{\nu}),$$

which avoids potentially costly evaluations of the gradient of the Fixman corrector $V_{\text{fix},\nu}$.

SCHEME 3.1 (Numerical discretization of IMMP (2.8)). *For a given numerical flow $\Phi_{\delta t}^\nu$ of a reversible, phase-space measure preserving integrator associated with (3.1), construct a Markov chain (of a Hybrid Monte Carlo type) $\{p_n, p_n^z, q_n, z_n\}_{n \geq 1}$*

Step 1: Compute $\Phi_{\delta t}^\nu(p_n, p_n^z, q_n, z_n)$ and $H_{n+1} = H_{\text{IMMP}}(\Phi_{\delta t}^\nu(p_n, p_n^z, q_n, z_n))$.

Step 2: Accept the step

$$(q_{n+1/2}, z_{n+1/2}, p_{n+1/2}, p_{n+1/2}^z) = \Phi_{\delta t}^\nu(p_n, p_n^z, q_n, z_n)$$

with the probability $\min(1, e^{-\beta(H_{n+1} - H_n)})$, otherwise reverse impulses

$$(q_{n+1/2}, z_{n+1/2}, p_{n+1/2}, p_{n+1/2}^z) = (q_n, z_n, -p_n, -p_n^z).$$

Step 3: Integrate the Gaussian fluctuation/dissipation part (see Appendix C.3).

For example, one can chose as $\Phi_{\delta t}^\nu$ the leapfrog/Verlet scheme

$$\left\{ \begin{array}{l} p_{n+1/2} = p_n - \frac{\delta t}{2} \nabla_1 V(q_n, \xi(q_n)) - \nabla_q \xi(q_n) \lambda_{n+1/2} \\ p_{n+1/2}^z = p_n^z - \frac{\delta t}{2\nu} \nabla_2 V(q_n, \xi(q_n)) + \frac{1}{\nu} \lambda_{n+1/2} \\ q_{n+1} = q_n + \delta t M^{-1} p_{n+1/2} \\ z_{n+1} = z_n + \delta t M_z^{-1} p_{n+1/2}^z \\ \xi(q_{n+1}) = \frac{z}{\nu} \\ p_{n+1} = p_{n+1/2} - \frac{\delta t}{2} \nabla_1 V(q_{n+1}, \xi(q_{n+1})) - \nabla_q \xi(q_{n+1}) \lambda_{n+1} \\ p_{n+1}^z = p_{n+1/2}^z - \frac{\delta t}{2\nu} \nabla_2 V(q_{n+1}, \xi(q_{n+1})) + \frac{1}{\nu} \lambda_{n+1} \\ \nabla_q^T \xi(q_{n+1}) M^{-1} p_{n+1} = \frac{1}{\nu} M_z^{-1} p_{n+1}^z \end{array} \right. \quad (C_{1/2}) \quad (3.2)$$

$$(C_1).$$

This numerically constructed Markov chain preserves the canonical distribution.

PROPOSITION 3.1 (Exact sampling). *Suppose the numerical flow $\Phi_{\delta t}^\nu$ is globally well-defined. Then the numerical discretization of (2.8) described in Scheme 3.1 generates a well-defined Markov chain that leaves the canonical distribution (2.6) invariant. The marginal distribution in position variables is the original distribution $e^{-\beta V(q, \xi(q))} dq$, which is independent of the mass-penalization ν .*

Proof. The statement follows from properties of Hybrid Monte Carlo Markov kernels (see Definition C.2), and from the construction of the mass-penalized Hamiltonian (Proposition 2.5). \square

One can then construct consistent schemes by letting the penalty $\nu \sim \delta t^{k_1}$ go to zero with the time-step, for some $k_1 > 0$. Indeed, Proposition 2.6 shows that the mass-penalized dynamics (2.8) is smooth with respect to ν^2 at $\nu = 0$. Consequently most of the usual numerical schemes will be consistent at a computable order bounded above by k_1^2 . In the context of this work, one could be interested in weak convergence of numerical Markov chains towards limiting diffusion processes. However, the splitting between the Hamiltonian part and the fluctuation/dissipation part (see Step 3 of Scheme 3.1) is bound to restrict the order of convergence to order 1. Arguably, only the independent order of convergence of each part of the splitting is relevant, and in the following analysis, the fluctuation/dissipation step is omitted ($\gamma = 0$). Thus in a deterministic setting the order of convergence refers to the maximal integer k such that the convergence of trajectories with respect to the uniform norm occurs at the rate of order $\mathcal{O}(\delta t^k)$.

PROPOSITION 3.2 (Time-step consistence). *Let the numerical flow $\Phi_{\delta t}^\nu$ be defined globally and suppose the dissipation vanishes ($\gamma = 0$). Assume that $\Phi_{\delta t}^\nu$ is of the order k_2 uniformly in ν . Then the numerical flow $\Phi_{\delta t}^{\delta t^{k_1}}$ is convergent of the order $\min(2k_1, k_2)$ with respect to the original limiting process (1.3).*

Proof. The mass-penalized dynamics (2.8) is a differential equation (deterministic here) whose coefficients differ from the original process by a smooth perturbation of order ν^2 . The result thus follows from applying a simple Gronwall argument. \square

A more precise statement can be given for the case of the RATTLE integrator, see Appendix C.1.

LEMMA 3.3. *The RATTLE numerical flow defined by (3.2) is of order 2 convergent with respect to time-step, uniformly in ν .*

Proof. [Sketch of the proof] The idea is to consider the scheme (3.2) on the variables $(p, q, \nu p^z, \nu q^z)$. Then the force field only depends on q and the global mass-matrix is smooth with respect to ν^2 . The sub-manifold defining the constraints ($\nu^2 \xi(q) = \nu z$) is also smooth with respect to ν^2 . By the implicit function theorem we have that locally the RATTLE scheme is the standard leapfrog scheme (see [13, 21]), and the local mapping depends smoothly on ν^2 . Therefore the result follows from the standard calculation of the order of the leapfrog scheme, [13]. \square

4. The infinite stiffness limit: stability properties. In this section, the stiffness parameter ϵ is re-introduced, by assuming that the potential V can be written in the form

$$V(q, z) = V^\epsilon(q, z) = U\left(q, \frac{z}{\epsilon}\right).$$

We shall then assume that the mass-matrix penalty parameter $\nu = \nu_\epsilon$ grows to infinity in such a way that $\lim_{\epsilon \rightarrow 0} \epsilon \nu_\epsilon = \bar{\nu}$. It will be proven that the IMMP numerical scheme (Scheme (3.1)) is then *asymptotically stable* in the infinite stiffness regime ($\epsilon \rightarrow 0$).

The original Hamiltonian with the stiffness parameter is expressed explicitly as

$$H_\epsilon(p, q) = \frac{1}{2} p^T M^{-1} p + U\left(q, \frac{\xi(q)}{\epsilon}\right), \quad (4.1)$$

and including the mass-matrix penalization one gets

$$H_{\nu_\epsilon}(p_{\nu_\epsilon}, q) = \frac{1}{2} p_{\nu_\epsilon}^T M_{\nu_\epsilon}^{-1} p_{\nu_\epsilon} + U\left(q, \frac{\xi(q)}{\epsilon}\right) + V_{\text{fix}, \nu_\epsilon}(q), \quad (4.2)$$

or in its implicit formulation

$$\begin{cases} H_{\text{IMMP}}(q, z, p, p_z) = \frac{1}{2} p^T M^{-1} p + \frac{1}{2} p_z^T M_z^{-1} p_z + U\left(q, \frac{z}{\nu_\epsilon \epsilon}\right) + V_{\text{fix}, \nu_\epsilon}(q), \\ \xi(q) = \frac{1}{\nu_\epsilon} z. \end{cases} \quad (C_{\nu_\epsilon}) \quad (4.3)$$

One immediately sees that H_{IMMP} is non-singular when $\epsilon \rightarrow 0$ and converges to the effective Hamiltonian, similar to H_{eff} , (1.9), on the slow manifold,

$$\begin{cases} H_{\text{eff}, \bar{\nu}}(q, z, p, p_z) = \frac{1}{2} p^T M^{-1} p + \frac{1}{2} p_z^T M_z^{-1} p_z + U\left(q, \frac{z}{\bar{\nu}}\right) + V_{\text{fix}}(q) \\ \xi(q) = 0. \end{cases} \quad (C_{\nu_\epsilon})$$

The continuity in ϵ of H_{IMMP} will imply stability of the associated dynamics and their numerical integrators.

4.1. Statistical and dynamical limits. One first derives the limits of the original and penalized canonical distribution. Recall the definition of the effective potential from (1.5) ($U_{\text{eff}} = V_{\text{eff}}$ up to an additive constant)

$$U_{\text{eff}}(q) = -\frac{1}{\beta} \ln \int e^{-\beta U(q, z)} dz.$$

PROPOSITION 4.1 (Limits of canonical distributions). *Consider the canonical distributions μ_ϵ and μ_{ν_ϵ} (see (1.4) and (2.6)) respectively associated with the original stiff Hamiltonian (4.1) and the mass penalized one (4.2), and both considered with respect to the $(p = M M_{\nu_\epsilon}^{-1} p_{\nu_\epsilon}, q)$ variables. In the sense of weak convergence of measures we have $\mu_\epsilon \rightarrow \mu_0$ and $\mu_{\nu_\epsilon} \rightarrow \mu_{\text{eff}}$ as $\epsilon \rightarrow 0$; where μ_0 is defined by (1.6), and μ_{eff} is defined as follows*

$$\mu_{\text{eff}}(dp dq) = \frac{1}{Z_{\text{eff}}} e^{-\beta(\frac{1}{2} p^T M^{-1} p + U_{\text{eff}}(q) + V_{\text{fix}}(q))} \sigma_{T^* \mathcal{M}_0}(dp dq). \quad (4.4)$$

In both cases, positions q lie almost surely in the slow manifold \mathcal{M}_0 , and the marginal in position variables is given up to the normalization by

$$e^{-\beta V_{\text{eff}}(q)} \delta_{\xi(q)=0}(dq). \quad (4.5)$$

Proof. The first convergence is proved in Lemma 1.3. For the second one, the following notation will be used

$$\begin{cases} \delta_{q,\epsilon z}(dq) = \delta_{\xi(q)=\epsilon z}(dq) \\ \delta_{p,\epsilon p_z}(dp) = \delta_{p^T M^{-1} \nabla \xi(q) = \epsilon M_z^{-1} p_z}(dp). \end{cases}$$

To prove the convergence of μ_{eff} we consider a δ -neighborhood \mathcal{U}^δ of \mathcal{M}_0 where

$$dp dq = \frac{\epsilon^{2n}}{\det M_z} \delta_{q,\epsilon z}(dq) dz \delta_{p,\epsilon p_z}(dp) dp_z$$

and a decomposition of the bounded observable (in (p, q) variables) $\varphi = \varphi_1 + \varphi_2$ such that $\text{supp } \varphi_1 \subset \mathcal{U}^\delta$ and $\text{supp } \varphi_2 \cap \mathcal{U}^\delta = \emptyset$. Thus, keeping in mind that $p_{\nu_\epsilon} = M_{\nu_\epsilon} M^{-1} p$, and using the confining property of the potential $U(q, \cdot)$ we obtain

$$\int \varphi(p, q) e^{-\beta H_{\nu_\epsilon}} dp_{\nu_\epsilon} dq = \int \varphi_1(p, q) e^{-\beta H_{\nu_\epsilon}} dp_{\nu_\epsilon} dq + \mathcal{O}(e^{-\beta K(\delta/\epsilon)}) \equiv I_\epsilon + \mathcal{O}(e^{-\beta K(\delta/\epsilon)}). \quad (4.6)$$

Applying the change of variables $p_{\nu_\epsilon} = M_{\nu_\epsilon} M^{-1} p$ gives

$$dp_{\nu_\epsilon} = \det(M_{\nu_\epsilon} M^{-1}) dp = \nu_\epsilon^{2n} \det M_z \det(G + \frac{1}{\nu_\epsilon^2} M_z^{-1}) dp,$$

and setting $\epsilon M_z^{-1} p_z \equiv \nabla_q \xi M^{-1} p$ and $\epsilon z \equiv \xi(q)$ we get

$$H_{\nu_\epsilon}(p_{\nu_\epsilon}, q) = \frac{1}{2} p^T M^{-1} p + \nu_\epsilon^2 \epsilon^2 p_z^T M_z^{-1} p_z + U(q, \frac{z}{\nu_\epsilon \epsilon}) + V_{\text{fix}, \nu_\epsilon}(q) = H_{\text{IMMP}}(q, z, p, p_z).$$

Thus substituting back to (4.6) we obtain

$$I_\epsilon = (\nu_\epsilon \epsilon)^{2n} \int \varphi_1 e^{-\beta H_{\text{IMMP}}(q, z, p, p_z)} \det(G + \frac{1}{\nu_\epsilon^2} M_z^{-1}) \delta_{p,\epsilon p_z}(dp) dp_z \delta_{q,\epsilon z}(dq) dz,$$

and thus

$$I_\epsilon \xrightarrow{\epsilon \rightarrow 0} \bar{\nu}^{2n} \int \varphi_1 e^{-\beta H_{\text{eff}, \bar{\nu}}(q, z, p, p_z)} \det(G) \delta_{p,\epsilon p_z}(dp) dp_z \delta_{\xi(q)=0}(dq) dz.$$

Using the co-area formula yields

$$\det(G) \delta_{\nabla \xi(q) M^{-1} p = 0}(dp) \delta_{\xi(q)=0}(dq) = \sigma_{T^* \mathcal{M}_0}(dp dq),$$

which leads to the final result after integration of the (p_z, z) variables and normalization. \square

REMARK 4.2. Due to the fast oscillations, the distribution of impulses in the limiting distribution μ_0 in (1.6) is free, whereas after the mass-matrix penalization, the limiting distribution (2.6) has almost surely co-tangent impulses (i.e., satisfying the constraints $\nabla_q \xi M^{-1} p = 0$). This explains the role of the corrected potential energy V_{fix} taking into account the curvature of \mathcal{M}_0 .

In the next step we inspect the infinite stiffness asymptotic of the penalized dynamics.

PROPOSITION 4.3 (Infinite stiffness limit). *When $\epsilon \rightarrow 0$ with $\nu = \nu_\epsilon \sim \frac{\bar{\nu}}{\epsilon}$ and $V(q, \xi(q)) = U(q, \frac{\xi(q)}{\epsilon})$, the IMMP Langevin stochastic process (2.8) converges weakly towards the following coupled limiting processes with constraints*

$$\begin{cases} \dot{q} = M^{-1} p, \\ \dot{p} = -\nabla_1 U(q, \frac{z}{\bar{\nu}}) - \nabla_q V_{\text{fix}}(q) - \gamma \dot{q} + \sigma \dot{W} - \nabla_1 \xi \dot{\lambda}, \\ \xi(q) = 0, \\ \dot{z} = M_z^{-1} p_z, \\ \dot{p}_z = -\frac{1}{\bar{\nu}} \nabla_2 U(q, \frac{z}{\bar{\nu}}) - \gamma_z \dot{z} + \sigma_z \dot{W}_z. \end{cases} \quad (C) \quad (4.7)$$

where ∇_1 and ∇_2 are respectively derivatives with respect to the first and second variable of the function $U(q, z)$, and $\{\lambda_t\}_{t \geq 0}$ are adapted stochastic processes defining the Lagrange multipliers associated with the constraints (C).

The process $\{q_t, p_t\}_{t \geq 0}$ defines an effective dynamics with constraints (Definition 1.4) for thermostatted highly oscillatory systems. It is reversible with respect to its stationary canonical distribution given by μ_{eff} (4.4), and is ergodic when (γ, γ_z) is strictly positive.

Proof. Satisfying the constraint (C_{ν_ϵ}) in (2.8) implies a hidden constraint in the momentum space, $\nabla_q \xi M^{-1} p = \frac{1}{\nu_\epsilon} M_z^{-1} p_z$. Differentiating this expression with respect to time and replacing the result in (2.8) yields an explicit formula for the Lagrange multipliers

$$\dot{\lambda} = (G + \frac{1}{\nu_\epsilon^2} M_z^{-1}) (\text{Hess}(\xi)(M^{-1} p, M^{-1} p) + \nabla_q \xi M^{-1} f_q - \frac{1}{\nu_\epsilon} M_z^{-1} f_z), \quad (4.8)$$

with forces (f_q, f_z)

$$\begin{aligned} f_q &= -\nabla_1 U(q, \frac{z}{\epsilon \nu_\epsilon}) - \nabla_q V_{\text{fix}, \nu_\epsilon}(q) - \gamma M^{-1} p + \sigma \dot{W}, \\ f_z &= -\frac{1}{\epsilon \nu_\epsilon} \nabla_2 U(q, \frac{z}{\epsilon \nu_\epsilon}) - \gamma_z M_z^{-1} p_z + \sigma_z \dot{W}_z, \end{aligned}$$

and the Hessian $\text{Hess}(\xi)$ of the mapping $\xi(q)$ acting on the velocities $M^{-1} p$. This calculation shows that (2.8) is in fact a standard stochastic differential equation with smooth coefficients, and thus has a unique strong solution. Since by assumption $\epsilon \nu_\epsilon \rightarrow \bar{\nu}$, the coefficients of this stochastic differential equations are continuous with respect to $\epsilon \rightarrow 0$ in a δ -neighborhood of \mathcal{M}_0 . The formally computed limiting process is given by (4.7) with the Lagrange multipliers solving

$$\dot{\lambda} = G^{-1} (\text{Hess}(\xi)(M^{-1} p, M^{-1} p) + \nabla_q^T \xi M^{-1} f_q).$$

By construction, this limiting process satisfies the constraint $\xi(q) = 0$, and therefore it belongs to the slow manifold \mathcal{M}_0 . Its coefficients are Lipschitz and the process is well-posed. As a result of those properties, the rigorous proof of weak convergence follows classical arguments, see [9], that are split into three steps

- (i) First, we truncate the process (2.8) to a compact neighborhood of \mathcal{M}_0 .
- (ii) The continuity of the Markov generator with respect to ϵ implies tightness for the associated ϵ -sequence of truncated processes as well as weak convergence.
- (iii) Well-posedness of the limiting process implies weak convergence for the ϵ -sequence of processes without truncation.

□

REMARK 4.4. When $\bar{\nu} \rightarrow +\infty$, by a classical averaging argument (see, e.g., [15]), one can check that the limiting dynamics are the effective dynamics pointed out in [20]

$$\begin{cases} \dot{q} = M^{-1} p \\ \dot{p} = -\nabla_q U_{\text{eff}}(q) - \nabla_q V_{\text{fix}} - \gamma \dot{q} + \sigma \dot{W} - \nabla_q \xi \dot{\lambda} \\ \xi(q) = 0. \end{cases} \quad (C) \quad (4.9)$$

with the stationary canonical distribution (4.4).

4.2. Asymptotic stability of the numerical integrator. The numerical scheme (Scheme 3.1) proposed for the IMMP method (2.8) is also stable in the limit of infinite stiffness $\epsilon \rightarrow 0$. Recall that we assume the scaling $\epsilon \nu_\epsilon \rightarrow \bar{\nu}$, and a reversible, measure preserving numerical flow $\Phi_{\delta t}^{\nu_\epsilon}(p, p_z, q, z)$ associated with any modified Hamiltonian with constraints (see (3.1))

$$\begin{cases} \tilde{H}_{\text{IMMP}}(p, p_z, q, z) = \frac{1}{2} p^T M^{-1} p + \frac{1}{2} p_z^T M_z^{-1} p_z + \tilde{V}_{\nu_\epsilon}(q) \\ \xi(q) = \frac{z}{\nu_\epsilon}, \end{cases} \quad (C) \quad (4.10)$$

which approximates H_{IMMP} , (4.3). We assume that there is a smooth potential $\tilde{V}_{\bar{\nu}}$ such that $\lim_{\nu_\epsilon \rightarrow +\infty} \tilde{V}_{\nu_\epsilon} = \tilde{V}_{\bar{\nu}}$.

PROPOSITION 4.5 (Asymptotic stability). *In the limit $\epsilon\nu_\epsilon \rightarrow \bar{\nu}$, the numerical flow $\Phi_{\delta t}^{\nu_\epsilon}$ associated with the leapfrog/Verlet integrator with constraints for the IMMP Hamiltonian (4.3) converges towards the numerical flow $\Phi_{\delta t}^{\bar{\nu}}$, which is the leapfrog/Verlet integrator with geometric constraints associated with*

$$\begin{cases} \tilde{H}_{\text{eff},\bar{\nu}}(p, p_z, q, z) = \frac{1}{2}p^T M^{-1}p + \frac{1}{2}p_z^T M_z^{-1}p_z + \tilde{V}_{\bar{\nu}}(q) \\ \xi(q) = 0. \end{cases} \quad (\text{C}) \quad (4.11)$$

Proof. The statement is a direct consequence of the implicit function theorem and the continuity of the leapfrog integrator with constraints (3.2) with respect to the parameter $\bar{\nu} = \epsilon\nu_\epsilon$. Indeed, we have $\frac{1}{\nu}\nabla_2 V = \frac{1}{\epsilon\nu_\epsilon}\nabla_2 U$, and in the considered limit we obtain the following leapfrog scheme

$$\begin{cases} p_{n+1/2} = p_n - \frac{\delta t}{2}\nabla_1 U(q_n, \frac{z_n}{\bar{\nu}}) - \nabla_q \xi(q_n)\lambda_{n+1/2} \\ p_{n+1/2}^z = p_n^z - \frac{\delta t}{2\bar{\nu}}\nabla_2 U(q_n, \frac{z_n}{\bar{\nu}}) \\ q_{n+1} = q_n + \delta t M^{-1}p_{n+1/2} \\ z_{n+1} = z_n + \delta t M_z^{-1}p_{n+1/2}^z \\ \xi(q_{n+1}) = 0 \end{cases} \quad (\text{C}_{1/2})$$

$$\begin{cases} p_{n+1} = p_{n+1/2} - \frac{\delta t}{2}\nabla_1 U(q_{n+1}, \frac{z_{n+1}}{\bar{\nu}}) - \nabla_q \xi(q_{n+1})\lambda_{n+1} \\ p_{n+1}^z = p_{n+1/2}^z - \frac{\delta t}{2\bar{\nu}}\nabla_2 U(q_{n+1}, \frac{z_{n+1}}{\bar{\nu}}) \\ \nabla_q \xi(q_{n+1})M^{-1}p_{n+1} = 0. \end{cases} \quad (\text{C}_1)$$

□

Similar properties hold for the case when a Metropolis step is introduced.

PROPOSITION 4.6. *Consider the limit of infinite stiffness $\epsilon \rightarrow 0$. Suppose the reversible, measure preserving numerical flow $\Phi_{\delta t}^{\nu_\epsilon}$ converges pointwise to the (reversible measure preserving) $\Phi_{\delta t}^{\bar{\nu}}$ associated with (4.11). Furthermore, suppose the initial state of the system is convergent. Then the numerical Markov chain (Scheme 3.1) associated with the IMMP Hamiltonian (4.3) converges almost surely to the scheme*

Step 1 Compute $\Phi_{\delta t}^{\bar{\nu}}(p_n, p_n^z, q_n, z_n)$ and $H_{n+1} = H_{\text{IMMP}}(\Phi_{\delta t}^{\bar{\nu}}(p_n, p_n^z, q_n, z_n))$.

Step 2 Accept the step $(q_{n+1/2}, z_{n+1/2}, p_{n+1/2}, p_{n+1/2}^z) = \Phi_{\delta t}^{\bar{\nu}}(p_n, p_n^z, q_n, z_n)$ with probability $\min(1, e^{-\beta(H_{n+1} - H_n)})$, otherwise reject and reverse impulses

$$(q_{n+1/2}, z_{n+1/2}, p_{n+1/2}, p_{n+1/2}^z) = (q_n, z_n, -p_n, -p_n^z).$$

Step 3 Integrate the fluctuation/dissipation as described, for example, in Appendix C.3.

Proof. The statement follows from the continuity of the Hamiltonian H_{IMMP} (4.3) with respect to the parameter $\bar{\nu} = \epsilon\nu_\epsilon$. □

The results and properties discussed in this section are summarized in Table 4.1.

	Null mass penalization $\nu = 0$	Positive mass-penalization $\epsilon, \nu > 0$	Infinite stiffness limit $\epsilon = 0, \frac{\nu}{\epsilon} \rightarrow \bar{\nu}$
Dynamics	Highly oscillatory + fluct./diss.	IMMP + fluct./diss.	Effective with constraints + fluct./diss.
Statistics	Canonical	Canonical with correlated velocities	Canonical on $T^*\mathcal{M}_0$
Numerics	Leapfrog/Verlet with IMMP + fluct./diss.		

TABLE 4.1

The IMMP dynamics and the Verlet numerical integration are both asymptotically stable in the infinite stiffness regime if $\frac{\nu}{\epsilon} \rightarrow \bar{\nu} < +\infty$. If the mass-penalization vanishes ($\nu = 0$) one recovers the original physical stiff system. The canonical distribution is always exact in the position variable. Notice that due to the penalized mass-matrix ($\nu > 0$) the statistics have correlated velocities.

5. A high dimensional test case. The behavior of numerical statistical methods for Hamiltonian systems is of particular interest in simulating high-dimensional systems. Such systems in general exhibit many different time-scales and one seeks to go beyond stability constraints implied by the fastest microscopic degrees of freedom. As mentioned in the introduction, globally implicit methods are usually too costly, and splitting methods using minimal implicitness represent an active research area. On the other hand, direct constraints on microscopic fast degrees of freedom are still used in practice and might introduce a strong bias in the macroscopic behavior of the whole system.

We show in this section that IMMP strategies can dramatically slow down the fastest timescales while at the same time preserving the macroscopic statistical distribution as well as some features of macroscopic dynamics. More precisely, it will be argued, using a specific example, that macroscopic dynamics relying on low frequencies are correctly approximated by the IMMP dynamics.

The model we consider consists of a chain of oscillators fixed at one end. The particles in the chain interact through a pairwise potential (possibly non-linear) and each particle is also individually submitted to a macroscopic exterior potential. It is assumed that after converting to the non-dimensional form and an appropriate space-time rescaling at the mass-transport level, the chain is considered in the unit interval $[0, 1]$ and its dynamics is described by the Hamiltonian

$$H_N(q, p) = \frac{1}{2}p^T p + \sum_{i=1}^N v_{\text{int}}(\nabla_i^d q) + \sum_{i=1}^N v_{\text{ext}}\left(\frac{i}{N}, q_i\right), \quad (5.1)$$

and a coupling with an exterior thermal bath at the inverse temperature $\beta_N = \beta N^{-1}$. In (5.1) the particles are represented by the deviations $q = (q_1, \dots, q_N)$ from their equilibrium position. The function $y \in \mathbb{R} \mapsto v_{\text{int}}(y) \in \mathbb{R}$ (resp. $(x, d) \in \mathbb{R} \times \mathbb{R} \mapsto v_{\text{ext}}(x, d) \in \mathbb{R}$) is a non-dimensional smooth interaction potential (resp. an exterior potential). The linear operator $\nabla^d : \mathbb{R}^N \rightarrow \mathbb{R}^N$ having the components

$$\nabla_i^d q = \frac{q_i - q_{i-1}}{1/N}, \quad i = 1, \dots, N$$

is the discrete gradient associated to the chain with the Dirichlet condition $q_0 = 0$ at the left end $x = 0$. In the same way,

$$(\nabla_i^d)^T q = \frac{q_i - q_{i+1}}{1/N}, \quad i = 1, \dots, N,$$

with $q_{N+1} = 0$. Differentiation with respect to $d \in \mathbb{R}$ is denoted $v'_{\text{int}}(y)$ and $v'_{\text{ext}}(x, y)$; and $v'_{\text{int}}(\nabla^d q) = (v'_{\text{int}}(\nabla_1^d q), \dots, v'_{\text{int}}(\nabla_N^d q))$ as well as $v'_{\text{ext}}(q) = (v'_{\text{ext}}(1/N, \nabla_1^d q), \dots, v'_{\text{ext}}(1, \nabla_N^d q))$. Using the matrix notation,

the stochastically perturbed equations of motion are

$$\begin{aligned}\dot{q} &= p \\ \dot{p} &= -(\nabla^d)^T v'_{\text{int}}(\nabla^d q) - v'_{\text{ext}}(q) - \gamma p + \sqrt{N} \sigma \dot{W}\end{aligned}$$

with the fluctuation/dissipation identity $\sigma \sigma^T = 2\beta^{-1}\gamma$.

Considering the chain in $[0, 1]$, the degrees of freedom q_i represent the deviation from the equilibrium positions at $x = \frac{i}{N}$. In the thermodynamic limit $N \rightarrow \infty$ the Hamiltonian (5.1) formally converges to

$$\lim_{N \rightarrow \infty} \frac{1}{N} H_N(q, p) = \int_0^1 \frac{1}{2} p(x)^2 dx + \int_0^1 v_{\text{int}} \left(\frac{dq}{dx} \right) dx + \int_0^1 v_{\text{ext}}(x, q(x)) dx$$

assuming that $x \mapsto q(x)$ is a smooth function on $[0, 1]$ with mixed boundary conditions $q(0) = q'(1) = 0$. When v_{int} is a bounded perturbation of an harmonic potential, the convergence towards a stochastic partial differential equation (SPDE) is a standard rigorous result ([18]). According to [26] such an atomic chain at a mass-transport scale is a particularly relevant benchmark for numerical methods in molecular dynamics simulations.

Next we turn to the mass-penalized Hamiltonian model for this linear chain of atoms. The number of independent fast degrees of freedom that will be penalized is equal to the size of the system N , and they are given by the inter-particle distances $\xi_i(q) = z_i = q_i - q_{i-1}$. The virtual mass penalty associated with fDOFs is still denoted M_z . The mass-penalization intensity scales as $\nu_N = \bar{\nu}N$, and we define the perturbed mass-matrix in \mathbb{R}^N

$$M_{\nu_N} = \text{Id} + \bar{\nu}^2 O,$$

with the symmetric positive operator $O = (\nabla^d)^T M_z \nabla^d$. When $M_z = \text{Id}$, the matrix $-O$ is the discrete Laplacian

$$O = (\nabla^d)^T \nabla^d = -\Delta_d.$$

Following our general construction we obtain the mass-penalized Hamiltonian

$$H_{\nu_N}(p_{\nu_N}, q) = \frac{1}{2} p_{\nu_N}^T M_{\nu_N}^{-1} p_{\nu_N} + \sum_{i=1}^N v_{\text{int}}(\nabla_i^d q) + \sum_{i=1}^N v_{\text{ext}}\left(\frac{i}{N}, q_i\right), \quad (5.2)$$

and the stochastically perturbed equations of motion are given by

$$\ddot{q} = (\text{Id} + \bar{\nu}^2 O)^{-1} \left(-(\nabla^d)^T v'_{\text{int}}(\nabla^d q) - v'_{\text{ext}}(q) - \gamma \dot{q} + \sigma \sqrt{N} \dot{W} \right). \quad (5.3)$$

5.1. Low-pass filtering and conservation of macroscopic dynamics. The form (5.3) of the equations of motion clearly demonstrates the regularizing effect of the mass-matrix penalization. Indeed, all the high frequencies generated by the microscopic forces $-(\nabla^d)^T v'_{\text{int}}(\nabla^d q)$ are *filtered* by the regularizing operator $\text{Id} + \bar{\nu}^2 O$. When the interaction potential is quadratic ($v_{\text{int}}(y) = y^2/2$), the operator O commutes with discrete Laplacian, and γ is proportional to the identity matrix. Hence we can consider the discrete Fourier spectral decomposition with the projection $PP^T = \text{Id}$

$$-\Delta_d = P^{-1} \text{diag}(\delta_1, \dots, \delta_N) P, \quad \text{and} \quad O = P^{-1} \text{diag}(o_1, \dots, o_N) P,$$

with the eigenvalues of the discrete Laplacian

$$\delta_k = 4N^2 \sin^2 \left(\frac{2k-1}{2N+1} \frac{\pi}{2} \right) \underset{N \rightarrow \infty}{\sim} \left(k + \frac{1}{2} \right)^2 \pi^2.$$

The system of equations (5.3) then becomes

$$\ddot{q} = (\text{Id} + \bar{\nu}^2 O)^{-1} \left(\Delta_d q - v'_{\text{ext}}(q) - \gamma \dot{q} + \sigma \sqrt{N} \dot{W} \right), \quad (5.4)$$

and applying the spectral projection with the Fourier image of the position variable $\hat{q} = N^{-1/2} P q$, we obtain (5.3) in the Fourier space for the spectral coefficients of order $k = 1, \dots, N$

$$\ddot{\hat{q}}^k = \frac{1}{1 + \bar{\nu}^2 o_k} \left(-\delta_k \hat{q}^k - (\widehat{v'_{\text{ext}}(q)})^k - \gamma \dot{\hat{q}}^k + \sigma \dot{W}^k \right), \quad (5.5)$$

where $\{W_t^k\}_{t \geq 0}$ are standard mutually independent Wiener processes. The Fourier representation shows that short wavelengths of the Fourier order k are slowed down by a factor $(1 + \bar{\nu}^2 o_k)^{-1}$, thus preventing instabilities.

In fact when $N \rightarrow +\infty$ and then $\bar{\nu} \rightarrow 0$, one gets the convergence of the macroscopic dynamics $t \mapsto q_t$ in the $L^2([0, 1])$ norm towards the original non-penalized dynamics. In order to treat the limit $N \rightarrow \infty$ we introduce the ℓ_2 -norm $\|q\|_0 = 1/N \sum_{i=1}^N |q_i|^2 \equiv 1/N q^T q$, $\|q\|_{\bar{\nu}} = \|(\text{Id} + \bar{\nu}^2 O)^{1/2} q\|_0$ and the norm $\|p\|_{-1} = \|\Delta_d^{-1/2} p\|_0$ defined for all, possibly infinite, sequences (q_1, \dots, q_N, \dots) and (p_1, \dots, p_N, \dots) . We write $X = (p, q)$ and $\|X\|_{\bar{\nu}} = \|q\|_{\bar{\nu}} + \|p\|_{-1}$. The convergence of the dynamics as $N \rightarrow \infty$ can be then studied in the space $\mathcal{X} = \{X = (p, q) \mid \|X\|_{\bar{\nu}} < \infty\}$. The analysis is carried out by straightforward application of semi-group properties of the linear equation (5.4). Thus we write the equation (5.4) in the operator form

$$\frac{dX_t}{dt} - A_{\bar{\nu}} X_t = F(X_t) + \Sigma \dot{W}_t, \quad (5.6)$$

where

$$A_{\bar{\nu}} X = \begin{pmatrix} 0 & \Delta_d \\ (\text{Id} + \bar{\nu}^2 O)^{-1} & 0 \end{pmatrix} \begin{pmatrix} p \\ q \end{pmatrix}, \quad \text{and} \quad F(X) = \begin{pmatrix} -v'_{\text{ext}}(q) - (\text{Id} + \gamma \bar{\nu}^2 O)^{-1} p \\ 0 \end{pmatrix}, \quad \Sigma = \begin{pmatrix} \sqrt{N} \sigma \\ 0 \end{pmatrix}.$$

PROPOSITION 5.1 (Macroscopic dynamical convergence). *Let the exterior force $v'_{\text{ext}}(x, \cdot)$ satisfies the Lipschitz condition*

$$\|v'_{\text{ext}}(q_2) - v'_{\text{ext}}(q_1)\|_{-1} \leq L_v \|q_2 - q_1\|_0$$

for almost all $x \in [0, 1]$ and uniformly in N . Given an initial condition $\|X_0^{\bar{\nu}}\|_{\bar{\nu}} < \infty$ such that $\lim_{\bar{\nu} \rightarrow 0} \|X_0^{\bar{\nu}} - \tilde{X}_0\|_0 = 0$ let $X_t^{\bar{\nu}}$ be the solution for $t \in [0, T]$ of (5.4), i.e.,

$$\frac{dX_t^{\bar{\nu}}}{dt} - A_{\bar{\nu}} X_t^{\bar{\nu}} = F(X_t^{\bar{\nu}}) + \Sigma \dot{W}_t. \quad (5.7)$$

Then as $\bar{\nu} \rightarrow 0$ the solution $\{X_t^{\bar{\nu}}\}_{t \geq 0}$ converges in the mean-square sense and uniformly in N to the solution of (5.7) with $\bar{\nu} = 0$ and the initial condition \tilde{X}_0 . More precisely,

$$\mathbb{E} (\|X_t^{\bar{\nu}} - X_t^0\|_0^2) \leq C_T \bar{\nu}^2,$$

where the constant C_T is independent of N but may depend on T .

Proof. The proof follows standard arguments and we outline it for the sake of completeness. We write an explicit solution formula for the equation (5.7)

$$X_t^{\bar{\nu}} = e^{A_{\bar{\nu}} t} X_0^{\bar{\nu}} + \int_0^t e^{A_{\bar{\nu}}(t-s)} (F(X_s^{\bar{\nu}}) ds + \Sigma dW_s). \quad (5.8)$$

In the spectral coordinates we have

$$(\widehat{e^{A_{\bar{\nu}}t}})^k = \begin{pmatrix} \cos(\omega_k t) & -\delta_k^{1/2}(1 + \bar{\nu}^2 o_k)^{1/2} \sin(\omega_k t) \\ \delta_k^{-1/2}(1 + \bar{\nu}^2 o_k)^{-1/2} \sin(\omega_k t) & \cos(\omega_k t) \end{pmatrix},$$

with $\omega_k = \delta_k^{1/2}(1 + \bar{\nu}^2 o_k)^{-1/2}$. Thus $e^{A_{\bar{\nu}}t}$ is in fact an isometry with respect to $\|\cdot\|_{\bar{\nu}}$ and $\|e^{A_{\bar{\nu}}t}\|_{\bar{\nu}} = 1$. Denoting $\langle \cdot, \cdot \rangle_{\bar{\nu}}$ the scalar product on \mathcal{X} we have

$$\mathbb{E} [\langle X, \Sigma W_t \rangle_{\bar{\nu}}^2] = \sigma^2 \left\| \begin{pmatrix} \Delta_d^{-1} & 0 \\ 0 & 0 \end{pmatrix} X \right\|_{\bar{\nu}}^2, \quad \text{with } \text{Tr} \begin{pmatrix} \Delta_d^{-1} & 0 \\ 0 & 0 \end{pmatrix} < \infty, \quad \text{as } N \rightarrow \infty.$$

Thus following [18], we have rigorous existence of the limiting equation (5.8) as $N \rightarrow \infty$. In particular, by a simple Gronwall argument, we check that $\mathbb{E}[\|X_t\|_{\bar{\nu}}^2]$ is bounded uniformly in N . We consider two solutions of (5.8) (for a given $\bar{\nu}$ and $\bar{\nu} = 0$) and the same noise. Subtracting the solutions we have

$$X_t^{\bar{\nu}} - X_t^0 = e^{A_{\bar{\nu}}t}(X_0^{\bar{\nu}} - \tilde{X}_0) + \int_0^t e^{A_{\bar{\nu}}(t-s)} (F(X_s^{\bar{\nu}}) - F(X_s^0) + (A_{\bar{\nu}} - A_0)X_s) ds.$$

Direct calculation shows

$$\|A_{\bar{\nu}} - A_0\|_0 = \left\| \begin{pmatrix} 0 & \\ (\text{Id} + \bar{\nu}^2 O)^{-1} - \text{Id} & 0 \end{pmatrix} \right\|_0 \leq \bar{\nu}^2,$$

leading to the upper bound (where L_F is the Lipschitz constant of the mapping $F(\cdot)$)

$$\|X_t - X_t^0\|_0 \leq t(\bar{\nu}^2 \|X_t\|_0 + L_F \|X_t - X_t^0\|_0) + \|X_0 - \tilde{X}_0\|_0.$$

The proof is concluded by applying the Gronwall inequality. \square

The continuity of macroscopic dynamics in the $L^2([0, 1])$ sense for the quasi-linear case (harmonic interactions) is a strong argument in favor of the following heuristic behavior: For high dimensional systems the IMMP method behaves as a low-pass filter. The high-frequency cut-off is tuned by the mass-matrix penalization parameter $\bar{\nu}$, and for vanishing $\bar{\nu}$, some macroscopic features of the dynamics can be recovered. It is shown in the next section that dramatic improvement of time-step stability can be obtained even for the vanishing re-scaled penalty $\bar{\nu}$.

5.2. Relaxation of time-step stability restriction. To demonstrate improved stability properties of time integration algorithms we consider the leapfrog/Verlet scheme associated with the mass-matrix penalized Hamiltonian (5.2). Note that when the constraints are linear, the leapfrog scheme (RATTLE) applied to an implicit Hamiltonian is identical to the usual leapfrog scheme for the explicit Hamiltonian (5.2). Again we restrict the rigorous analysis to the quadratic interaction potential ($v_{\text{int}}(y) = \frac{y^2}{2}$), and to the mass-matrix penalization operator O commuting with the discrete Laplacian. The leapfrog scheme is defined as

$$\begin{cases} p_{n+1/2} = p_n + \frac{\delta t}{2}(-\Delta_d)q_n \\ q_{n+1} = q_n + \delta t M_{\nu_N}^{-1} p_{n+1} \\ p_{n+1} = p_{n+1/2} + \frac{\delta t}{2}(-\Delta_d)q_{n+1}. \end{cases}$$

Denoting the spectral variables

$$\begin{cases} \hat{v} = \sqrt{N} P(-\Delta_d)^{1/2} M_{\nu_N}^{-1/2} p_{\nu_N} \\ \hat{x} = \sqrt{N} P(-\Delta_d)^{-1/2} M_{\nu_N}^{1/2} q, \end{cases} \quad (5.9)$$

we write the scheme in the form

$$\begin{pmatrix} \widehat{v}_{n+1}^k \\ \widehat{x}_{n+1}^k \end{pmatrix} = L_k \begin{pmatrix} \widehat{v}_n^k \\ \widehat{x}_n^k \end{pmatrix},$$

where we denote

$$L_k = \begin{pmatrix} 1 - \frac{h_k^2}{2} & -h_k + \frac{h_k^3}{4} \\ h_k & 1 - \frac{h_k^2}{2} \end{pmatrix}, \quad \text{and} \quad h_k = \delta t \frac{\delta_k^{1/2}}{(1 + \bar{\nu}^2 o_k)^{1/2}}.$$

Since $\det(L_k) = 1$, the standard CFL stability condition is equivalent to

$$|\text{Tr}(L_k)| \leq 2$$

which is fulfilled if and only if $h_k \leq 2$. Thus we arrive at a bound on the time step

$$\delta t \leq 2 \frac{(1 + \bar{\nu}^2 o_k)^{1/2}}{\delta_k^{1/2}}.$$

Summarizing the above calculations we have the following characterization of the stability properties.

LEMMA 5.2 (Stability). *Suppose $v_{\text{ext}} = 0$ and consider an harmonic interaction potential $v_{\text{int}}(y) = \frac{y^2}{2}$. Let the mass-matrix penalization $M_{\nu_N} = \text{Id} + \bar{\nu}^2 O$ commute with the discrete Laplacian Δ_d , and denote by $\{o_1, \dots, o_N\}$ the eigenvalues of O . The leapfrog/Verlet integration of the IMMP harmonic Hamiltonian (5.1) is stable in the spectral sense if and only if*

$$\delta t \leq \inf_{k=1, \dots, N} \frac{(1 + \bar{\nu}^2 o_k)^{1/2}}{N \sin\left(\frac{2k-1}{2N+1} \frac{\pi}{2}\right)}. \quad (5.10)$$

In particular, if the implicit mass-matrix $M_z = \text{Id}$ is chosen, the stability condition (5.10) is, for large N , equivalent to

$$\delta t \leq \left(\frac{1}{N^2} + 4\bar{\nu}^2 \right)^{1/2},$$

consequently any mass-penalization ($\bar{\nu} > 0$) induces a relative increase of the order N , compared to the original dynamics $\bar{\nu} = 0$, for the boundary of numerical stability.

Since we work with a Metropolis correction of the Hybrid Monte-Carlo type, we are also interested in the limiting behavior of the energy variation compared to the temperature, i.e.,

$$\beta_N (H(p_{n+1}, q_{n+1}) - H(p_n, q_n)),$$

when (p_n, q_n) is distributed according to the canonical distribution. This quantity gives the average acceptance rate of the Metropolis correction. This result is similar to [2], where the authors analyze infinite dimensional sampling with the standard Metropolis-Hastings Markov chains.

PROPOSITION 5.3. *Suppose $v_{\text{ext}} = 0$ and the interaction potential is harmonic ($v_{\text{int}}(y) = \frac{y^2}{2}$). Let the mass-matrix penalization $M_{\nu_N} = \text{Id} + \bar{\nu}^2 O$ commute with the discrete Laplacian Δ_d . We denote by $\{o_1, \dots, o_N\}$ the eigenvalues of O .*

Suppose the state variable $X = (p_{\nu_N}, q)$ is a random variable distributed according to the canonical distribution associated with mass-matrix penalized Hamiltonian (5.1). Then the energy variation $\beta_N \Delta H$ after one step of the leapfrog integration scheme converges in distribution (up to normalization and centering) to the Gaussian random variable

$$\frac{\beta_N \Delta H - m_N}{\sigma_N} \xrightarrow[N \rightarrow +\infty]{\text{Law}} \mathcal{N}(0, 1),$$

with the mean and variance

$$m_N = \sum_{k=1}^N \frac{h_k^6}{2^5}, \quad \text{and} \quad \sigma_N^2 = \sum_{k=1}^N \frac{h_k^6}{2^4} + \frac{h_k^{12}}{2^9}.$$

In the special case $M_z = \text{Id}$, one gets the infinite size asymptotics for $\bar{\nu} > 0$ and $\delta t \equiv \delta t_N = o(1)$:

$$m_N \underset{N \rightarrow +\infty}{\sim} \frac{N \delta t_N^6}{32 \bar{\nu}^6}, \quad \text{and} \quad \sigma_N^2 \underset{N \rightarrow +\infty}{\sim} \frac{N \delta t_N^6}{16 \bar{\nu}^6},$$

and for $\bar{\nu} = 0$ and $\delta t \equiv \delta t_N = o(1/N)$:

$$m_N \underset{N \rightarrow +\infty}{\sim} \frac{5}{8} N^7 \delta t_N^6, \quad \text{and} \quad \sigma_N^2 \underset{N \rightarrow +\infty}{\sim} \frac{5}{4} N^7 \delta t_N^6.$$

Thus in the above case, the time-step restriction imposed in the case with $\bar{\nu} = 0$ is relaxed by a factor of order N when $\bar{\nu} > 0$.

Proof. We start with a canonically distributed state $X = (p_{\nu_N}, q)$, which is, by assumption on the form of interaction potential, a Gaussian random vector. After changing to the spectral coordinates (5.9) we have the spectral representation of the Hamiltonian

$$\beta_N H = \beta \sum_{k=1}^N \frac{\delta_k^{1/2}}{2(1 + \bar{\nu}^2 o_k)^{1/2}} ((\hat{v}^k)^2 + (\hat{x}^k)^2),$$

and introducing Gaussian random vectors U and V with the identity co-variance matrix we can write

$$\hat{x}^k = \beta^{-1/2} \frac{(1 + \bar{\nu}^2 o_k)^{1/4}}{\delta_k^{1/4}} U_k, \quad \text{and} \quad \hat{v}^k = \beta^{-1/2} \frac{(1 + \bar{\nu}^2 o_k)^{1/4}}{\delta_k^{1/4}} V_k.$$

We then compute explicitly the change of the Hamiltonian after one step of the leapfrog integration

$$\beta_N \Delta H = \sum_{k=1}^N \frac{1}{2} \begin{pmatrix} U_k \\ V_k \end{pmatrix}^T (L_k^T L_k - \text{Id}) \begin{pmatrix} U_k \\ V_k \end{pmatrix}.$$

Since $\det(L_k^T L_k) = 1$, $L_k^T L_k - \text{Id}$ has two positive eigenvalues $(\lambda_k - 1, 1/\lambda_k - 1)$, which satisfy

$$\begin{aligned} \lambda_k + 1/\lambda_k - 2 &= \text{Tr}(L_k^T L_k - \text{Id}) = \frac{h_k^6}{16}, \\ (\lambda_k - 1)^2 + (1/\lambda_k - 1)^2 &= \text{Tr}(L_k^T L_k)^2 - 2\text{Tr}(L_k^T L_k) = \frac{h_k^{12}}{256} + \frac{h_k^6}{8}, \end{aligned}$$

and we find

$$m_N \equiv \mathbb{E}[\beta_N \Delta H] = \sum_{k=1}^N \frac{h_k^6}{2^5}, \quad \text{and} \quad \sigma_N^2 \equiv \text{Var}[\beta_N \Delta H] = \sum_{k=1}^N \frac{h_k^6}{2^4} + \frac{h_k^{12}}{2^9}.$$

Moreover, the Lindenberg or simply Lyapunov condition in the general central limit theorem (see [11]) is verified since we work with a sum of χ^2 random variables, thus concluding the first part of the proof.

When $M_z = \text{Id}$

$$h_k = \delta t \frac{\sin(\frac{2k-1}{2N+1} \frac{\pi}{2})}{(\frac{1}{4N^2} + \bar{\nu}^2 \sin^2(\frac{2k-1}{2N+1} \frac{\pi}{2}))^{1/2}},$$

and for $\bar{\nu} \neq 0$, and $p = 6$ or $p = 12$ the Riemann sums converge

$$\lim_{N \rightarrow \infty} \frac{1}{N} \sum_{k=1}^N h_k^p = \frac{1}{\bar{\nu}^p}.$$

For $\bar{\nu} = 0$ we have

$$\begin{aligned} \lim_{N \rightarrow \infty} \frac{1}{N^{p+1}} \sum_{k=1}^N h_k^p &= \lim_{N \rightarrow \infty} \frac{1}{N} \sum_{k=1}^N 2^p \sin^p \left(\frac{2k-1}{2N+1} \frac{\pi}{2} \right) \\ &= 2^p \int_0^1 \sin^{2p} \left(\frac{\pi}{2} x \right) dx \\ &= 20 \quad \text{for } p = 6. \end{aligned}$$

Then from the assumptions it follows $\delta t_N^{12} \ll \delta t_N^6$, and similarly in the case $\bar{\nu} = 0$ $\delta t_N^{12} N^{12} \ll \delta t_N^6 N^6$ which gives the result. \square

6. Numerical experiments. In this section, the behavior of the IMMP method ((5.2),(5.3)) is tested for a non-linear interaction potential, and different values of the mass-matrix penalization intensity $\bar{\nu}$, of the time step parameter δt , and of the system size N . The reader should bear in mind that the mass-matrix penalization intensity $\bar{\nu}$ can be chosen as a vanishing function of the time-step although one usually seeks the largest stable time-step in such multi-scale models. This point of view makes the IMMP method a *dynamically consistent scheme* for highly oscillatory problems. As already proven, it is also *statistically exact*.

The simulations have been carried out with no exterior potential $v_{\text{ext}} = 0$, with the quartic interaction potential $v_{\text{int}}(y) = \frac{y^4}{4}$, and a penalizing mass-matrix given by the identity matrix $M_z = \text{Id}$. The following parameters have been used: the inverse temperature $\beta = 10$, the dissipation parameter $\gamma = 1$, and the chain size $N = 500$ (if not stated otherwise).

Relaxation of time-step restrictions. We first study the dependence between the time-step δt , the mass penalty $\bar{\nu}$, and the system size N . In Figure 6.1(a), the average Metropolis acceptance rate is computed for different time-steps and different values of the mass-matrix penalization ($\bar{\nu} = \{0, .01, .03, .1, .32\}$). Two different points are observed in this figure:

- (i) The time-step is relaxed by a factor comparable to the chain size (~ 300 for $N = 500$).
- (ii) The time-step is relaxed by a factor broadly proportional to the mass-matrix penalization, in a good agreement with Proposition (5.3) for the linear case.

In Figure 6.1(b), the time-step relaxation is studied for different values of the chain size N . The typical time-step (giving a Metropolis average acceptance rate 0.80) for the mass-matrix penalization ($\bar{\nu} = 0.32$) is compared to the non-penalized case ($\bar{\nu} = 0$), and the relative improvement is given on the vertical axis (“healing order”). Observe that the time-step relaxation increases with the system size N (in a broadly linear fashion). This observation shows that for a large system ($N \rightarrow \infty$), it is possible to choose a vanishing re-scaled penalization ($\bar{\nu} = o_N(1)$) while still performing arbitrary large relaxation of time-step restriction.

Approximate macroscopic dynamics. We now analyze the dependence of the macroscopic dynamics of a tagged particle with respect to the mass-penalization $\bar{\nu}$. Figure 6.2 depicts the dynamical behavior (for short times) of the chain extremity (i.e., the distance between the fixed end of the chain and the last, tagged particle) for different values of the mass-matrix penalization intensity $\bar{\nu} = \{0, 0.07, 0.22\}$. The typical (macroscopic) timescale is continuously perturbed by the mass penalization. For instance, typical dynamical quantities as the average transition time (from “low” positions to “high” positions) of the tagged particle is computed in Table 6.1. The convergence to equilibrium of the tagged extremal particle is shown in Figure 6.3. The Cramér–Von Mises test is computed between the time empirical distribution of the

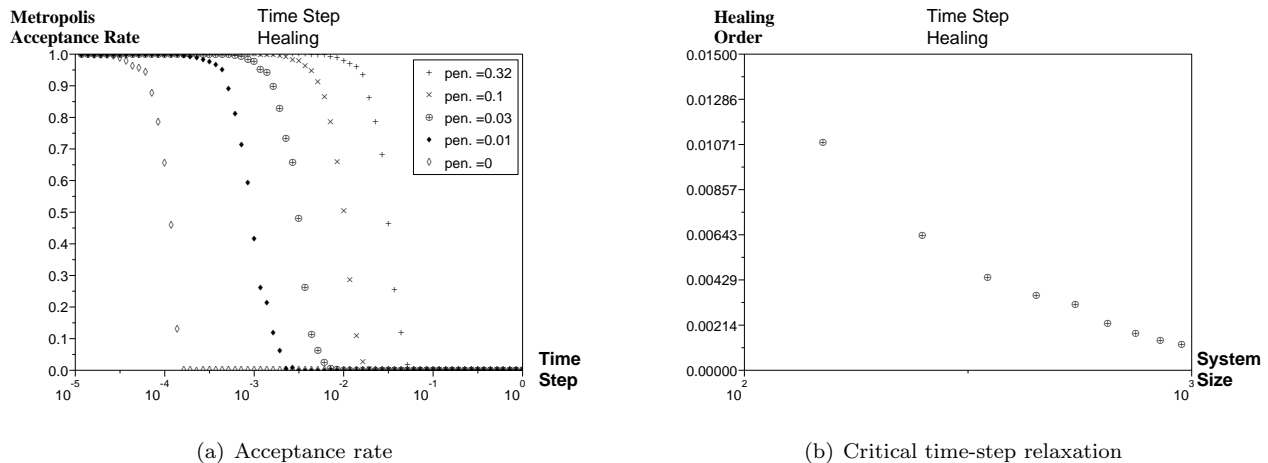


FIGURE 6.1. (a) Average acceptance rate of the Metropolis step for different value of the mass-matrix penalization $\text{pen.} = \bar{\nu} = \{0, 0.01, 0.03, 0.1, 0.32\}$, with $N = 500$. (b) Typical time-step comparison between the IMMP dynamics ($\bar{\nu} = .32$), and the free dynamics ($\bar{\nu} = 0$), for different values of the system size N . The vertical axis shows the relaxation (increasing coefficient or “healing order” between $\bar{\nu} = .32$ and $\bar{\nu} = 0$) of the typical time-step which gives a Metropolis acceptance rate of .8.

tagged particle for different mass penalizations, and the time empirical distribution of a reference run without mass-penalty. In the same plot, the average value (with the standard error bound) of a Cramér–Von Mises test for i.i.d. empirical distributions with the number of samples proportional to the final time T is displayed. The test shows that the decorrelation time of the tagged particle is of comparable order whatever the penalization $\bar{\nu}$.

Mass-Matrix Penalization	$\bar{\nu} = 0$	$\bar{\nu} = 0.07$	$\bar{\nu} = 0.22$
Transition Time	0.50	0.55	0.65

TABLE 6.1

Average transition time from the position $q_N = 0.9$ to the position $q_N = 1.1$ of the tagged particle of the chain. Estimated from simulations on a time interval with the final time $T > 1000$.

Conclusions. This numerical study shows that IMMP allows for arbitrary large relaxation of the time-step restriction which occurs even for a small re-scaled penalisation $\bar{\nu} = o_N(1)$. Furthermore, it demonstrates that some macroscopic quantities that depend on the dynamics can also be approximated.

Appendix A. Surface measures. Let \mathbb{R}^d be endowed with the scalar product given by the positive definite matrix M , and consider \mathcal{M}_z a family of sub-manifolds of co-dimension n implicitly defined by n independent functions $\mathcal{M}_z = \{x \in \mathcal{M} \mid \xi_1(q) = z_1, \dots, \xi_n(q) = z_n\}$ for z in a neighborhood of the origin. For each z in a neighborhood of the origin the *conditional measure* $\delta_{\xi(q)=z}(dq)$ is a measure on \mathcal{M}_z defined in such a way that it satisfies the chain rule for conditional expectations with respect to the Lebesgue measure dq , i.e.,

$$dq = \delta_{\xi(q)=z}(dq) dz. \quad (\text{A.1})$$

The *surface measure* $\sigma_{T_q^* \mathcal{M}_z}(dp)$ is the Hausdorff measure induced by the metric M^{-1} on the co-tangent space $T_q^* \mathcal{M}_z = \{p \mid \nabla_q^T \xi(q) M^{-1} p = 0\}$; and in the same way, $\sigma_{\mathcal{M}_z}(dq)$ is the Hausdorff measure induced by the metric M on the sub-manifold \mathcal{M}_z . It is important to note that, although this is not explicit in notation, σ is defined with respect to the mass-tensor M of the mechanical system. The Liouville measure

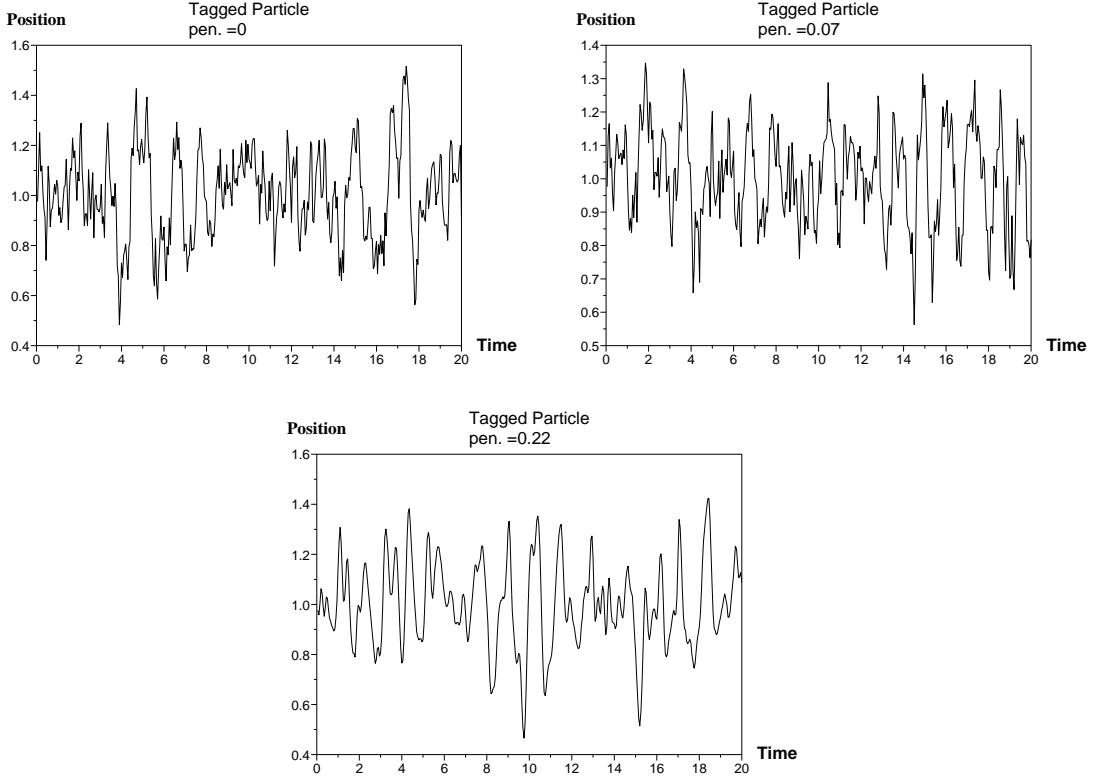


FIGURE 6.2. Evolution with respect of the extremity of the atomic chain, depicted for different values of the mass-penalization $\text{pen.} = \bar{\nu} = \{0, 0.07, 0.22\}$.

$\sigma_{T^*\mathcal{M}_z}(dp dq)$ on the co-tangent bundle $T^*\mathcal{M}_z$ is the volume form induced on

$$T^*\mathcal{M}_z = \{(p, q) \mid \nabla^T \xi(q) M^{-1} p = 0, \quad \xi(q) = z\}$$

by the usual symplectic form $dp \wedge dq$. It can be described in terms of surface measures as follows

$$\sigma_{T^*\mathcal{M}_z}(dp dq) = \sigma_{T_q^*\mathcal{M}_z}(dp) \sigma_{\mathcal{M}_z}(dq).$$

Finally, the co-area formula (see [10] for a general reference) gives out the relative probability density between $\delta_{\xi(q)=z}(dq)$ and $\sigma_{\mathcal{M}_z}(dq)$.

PROPOSITION A.1 (Co-area formula). *Given the invertible Gram matrix associated with the constraints $\xi(q) = z$ in a neighborhood of $\mathcal{M}_z = \{q \mid \xi(q) = z\}$*

$$G(q) = \nabla_q^T \xi M^{-1} \nabla_q \xi,$$

one has

$$\delta_{\xi(q)=z}(dq) = \frac{1}{\sqrt{\det G(q)}} \sigma_{\mathcal{M}_z}(dq).$$

Appendix B. Langevin processes. Defining the Poisson bracket

$$\{\varphi_1, \varphi_2\} = \nabla_p^T \varphi_1 \nabla_q \varphi_2 - \nabla_p^T \varphi_2 \nabla_q \varphi_1,$$

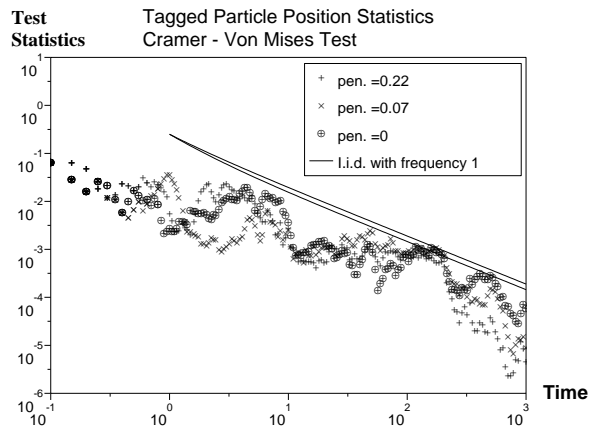


FIGURE 6.3. Convergence of the time empirical distribution of the tagged extremal particle for different mass penalizations, computed with a two-sample Cramér–Von Mises test. The solid lines depict the Cramér–Von Mises test (with error bounds) of a “benchmark” empirical distribution of i.i.d. samples with the frequency equal to 1.

and the dissipation tensor

$$d(q) = \sigma q,$$

where σ is the fluctuation matrix in Definition 1.1, the Markov generator of the Langevin process in Definition 1.1 is

$$\mathcal{L} = \{\cdot, H\} + \frac{1}{\beta} \{d, \{d^T, \cdot\} e^{-\beta H}\} e^{\beta H}.$$

We assume that the process is reversible and thus the generator \mathcal{L} satisfies

$$\int \varphi_1 \mathcal{L}(\varphi_2) e^{-\beta H} dp dq = \int \mathcal{L}^*(\varphi_1) \varphi_2 e^{-\beta H} dp dq,$$

where

$$\mathcal{L}^* = \{\cdot, -H\} + \frac{1}{\beta} \{d, \{d^T, \cdot\} e^{-\beta H}\} e^{\beta H}.$$

The generator \mathcal{L}^* defines the same Langevin process as \mathcal{L} but with the time-reversed Hamiltonian ($-H$). Reversibility of the process implies that the canonical measure is stationary. Furthermore, if the initial state of the system is a canonically distributed random variable, the probability distribution of a trajectory after the time-reversal is given by a Langevin process with the generator \mathcal{L}^* . When H has the form $H(p, q) = \frac{1}{2} p^T M^{-1} p + V_p(q)$, reversal of impulses ($p \rightarrow -p$) leads to time-reversed dynamics, and a process with generator \mathcal{L}^* can be constructed by the following simple steps:

1. Reverse momenta ($p \rightarrow -p$).
2. Draw a random path with generator \mathcal{L} .
3. Reverse again momenta ($p \rightarrow -p$).

When holonomic constraints, for example, of the form

$$\Xi(p, q) = \zeta \Leftrightarrow \begin{cases} p^T M^{-1} \nabla_q \xi = 0 \\ \xi(q) = z \end{cases}$$

are added, it is useful to introduce the Poisson bracket on the co-tangent bundle $T^*\mathcal{M}_z$

$$\{\varphi_1, \varphi_2\}_{\mathcal{M}_z} = \{\varphi_1, \varphi_2\} - \sum_{a,b} \{\varphi_1, \Xi^a\} \Gamma_{a,b}^{-1} \{\Xi^b, \varphi_2\},$$

where Γ is the symplectic Gram matrix of the full constraints

$$\Gamma^{a,b} = \{\Xi^a, \Xi^b\}.$$

As a basic result of symplectic geometry one recovers the divergence formula with respect to the bracket $\{\cdot, \cdot\}_{\mathcal{M}_z}$ and the Liouville measure $\sigma_{T^*\mathcal{M}_z}(dp dq)$

$$\int \{\cdot, \cdot\}_{\mathcal{M}_z} \sigma_{T^*\mathcal{M}_z}(dp dq) = 0.$$

Given a constrained Langevin process in a stochastic differential equation form

$$\begin{aligned} \dot{q} &= \nabla_p H, \\ \dot{p} &= -\nabla_q H - \gamma \dot{q} + \sigma \dot{W} - \nabla_q \xi \dot{\lambda}, \end{aligned}$$

where λ are Lagrange multipliers associated with the constraints $\xi(q) = 0$, adapted with respect to the noise \dot{W} , then the process $\{p_t, q_t\}_{t \geq 0}$ obeys hidden velocity constraints and is characterized by the SDE

$$\begin{aligned} \dot{q} &= \nabla_p H + \nabla_p \Xi \dot{\Lambda}, \\ \dot{p} &= -\nabla_q H - \gamma \dot{q} + \sigma \dot{W} - \nabla_q \Xi \dot{\Lambda}, \end{aligned}$$

where Λ are Lagrange multipliers associated with the full constraints $\Xi(p, q) = 0$. The Markov generator of this process can be written in the form

$$\mathcal{L}_{\mathcal{M}_z} = \{\cdot, H\}_{\mathcal{M}_z} + \frac{1}{\beta} \left\{ \text{d}, \left\{ \text{d}^T, \cdot \right\}_{\mathcal{M}_z} e^{-\beta H} \right\}_{\mathcal{M}_z} e^{\beta H},$$

demonstrating the reversibility with respect to the constrained canonical measure $e^{-\beta H} \sigma_{T^*\mathcal{M}_z}(dp dq)$.

Appendix C. Exact hybrid scheme for Langevin dynamics with constraints.

C.1. RATTLE integrator. The RATTLE algorithm is a leapfrog integrator with algebraic constraints. It is given by a "kick-move-kick" splitting with a procedure that enforces constraints at the velocity level. The flow defined by the integrator is symplectic, symmetric, and of the second order (see [13]). We denote the phase-space variables of the system at the discrete time n by $(p_n, q_n) \in T^*\mathbb{R}^d$ and its Hamiltonian

$$H(p, q) = \frac{1}{2} p^T M p + V(q),$$

imposing the holonomic constraints

$$\begin{cases} \xi(q_n) = z, \\ p_n^T M \nabla_q \xi(q_n) = 0. \end{cases}$$

SCHEME C.1 (RATTLE). Suppose that (p_n, q_n) satisfy the constraints, (p_{n+1}, q_{n+1}) is defined by the following set of equations

$$\begin{cases} p_{n+1/2} = p_n - \frac{\delta t}{2} \nabla_q V(q_n) - \nabla_q \xi(q_n) \lambda_{n+1/2} \\ q_{n+1} = q_n + \delta t M^{-1} p_{n+1/2} \\ \xi(q_{n+1}) = z \\ p_{n+1/2} = p_n - \frac{\delta t}{2} \nabla_q V(q_{n+1}) - \nabla_q \xi(q_n) \lambda_{n+1} \\ p_{n+1}^T M^{-1} \nabla_q \xi(q_{n+1}) = 0, \end{cases} \quad (C_{1/2}) \quad (C.1) \quad (C_1)$$

where the Lagrange multipliers $\lambda_{n+1/2}$, λ_{n+1} are associated with the constraints $(C_{1/2})$ and (C_1) , respectively. For a given initial condition (p_n, q_n) and sufficiently small time step δt , the solution of equations (C.1) exists and is unique.

We state the standard properties of the RATTLE flow as a separate lemma, for details see [13].

LEMMA C.1. The numerical flow $\Phi_{\delta t}$ defined by (C.1)

- (i) is symplectic as a mapping of $T^*\mathcal{M}_z$ and preserves the Liouville measure $\sigma_{T^*\mathcal{M}_z}(dp dq)$,
- (ii) is time-reversible under reversal of impulses,
- (iii) is of the second order, i.e., $\mathcal{O}(\delta t^2)$.

C.2. Metropolis rule. The time-reversibility and phase-space measure preserving property of RATTLE enables us to introduce a Metropolis acceptance/rejection rule (Hybrid Monte-Carlo). We obtain a Markov chain reversible with respect to the canonical distribution which is used for sampling the canonical distribution exactly.

SCHEME C.2 (Hybrid Monte-Carlo). Let $\Phi_{\delta t}$ be a deterministic numerical flow associated with the Hamiltonian

$$H(q, p) = \frac{1}{2} p^T M p + V(q),$$

on the phase-space $T^*\mathcal{M}_z$.

The Hybrid Monte-Carlo acceptance/rejection rule consists of building a Markov kernel $M(p_n, q_n, dp_{n+1} dq_{n+1})$, using the following steps: For $(p_n, q_n) \in T^*\mathcal{M}_z$

STEP 1: compute the energy variation

$$\Delta H = H \circ \Phi_{\delta t}(p_n, q_n) - H(p_n, q_n),$$

STEP 2: with probability $\min(1, e^{-\beta \Delta H})$ accept the step and set

$$(p_{n+1}, q_{n+1}) = \Phi_{\delta t}(p_n, q_n),$$

STEP 3: otherwise reject the step and reverse impulses

$$(p_{n+1}, q_{n+1}) = (-p_n, q_n).$$

The algorithm allows for sampling the exact canonical distribution, a property which we state as a separate proposition.

PROPOSITION C.2. Suppose the numerical flow $\Phi_{\delta t}$ associated with the Hamiltonian H is globally defined, time-reversible, and phase-space measure preserving. Then the Markov kernel $M(q_n, p_n, dp_{n+1} dq_{n+1})$ defined by the Metropolis rule of Scheme C.2 is reversible with respect to the canonical measure $\mu(dp dq) = Z^{-1} e^{-\beta H} \sigma(dp dq)$ on $T^*\mathcal{M}_z$, and

$$M(p_n, q_n, dp_{n+1} dq_{n+1}) \mu(dp_n dq_n) = M(-p_{n+1}, q_{n+1}, dp_n dq_n) \mu(dp_{n+1} dq_{n+1}). \quad (C.2)$$

Proof. For the sake of completeness, we give a proof of this proposition. We use the following abbreviated notation: $x_n \equiv (p_n, q_n)$, $h(\cdot) \equiv \min(1, \cdot)$, $\sigma(dx) \equiv \sigma(dp dq)$ is the phase-space measure, S is the reversal of impulses, and $\Delta H = H \circ \Phi_{\delta t} - H$ is the energy variation. We summarize the properties of the numerical flow

- (a) The numerical flow preserve the phase-space measure $\Phi_{\delta t}^{-1}(\sigma(dx)) = \sigma(dx)$.
- (b) The numerical flow is time-reversible $S \circ \Phi_{\delta t} \circ S = \Phi_{\delta t}^{-1}$.
- (c) Energy and phase-space measure conservation of the symmetry: $H \circ S = H$ and $S^{-1}(\sigma(dx)) = \sigma(dx)$.
- (d) Metropolis rate function h satisfies $\forall \theta \in \mathbb{R}, h(\theta) = \frac{h(1/\theta)}{\theta}$.

The symmetry property (c) leads to the relation

$$\Delta H \circ \Phi_{\delta t}^{-1} = H \circ S - H \circ S^{-1} \circ \Phi_{\delta t}^{-1} = -\Delta H \circ S^{-1},$$

which together with the property (d) proves the detailed balance condition

$$(h(e^{-\beta\Delta H})e^{-\beta H}) \circ \Phi_{\delta t}^{-1} = (h(e^{-\beta\Delta H})e^{-\beta H}) \circ S^{-1}. \quad (\text{C.3})$$

On the other hand, the property (a) gives

$$\delta_{\Phi_{\delta t}(x_n)}(dx_{n+1})\sigma(dx_n) = \delta_{\Phi_{\delta t}^{-1}(x_{n+1})}(dx_n)\sigma(dx_{n+1}),$$

and

$$\delta_{S(x_n)}(dx_{n+1})\sigma(dx_n) = \delta_{S^{-1}(x_{n+1})}(dx_n)\sigma(dx_{n+1}).$$

Recalling the form of the Metropolis kernel we have

$$M(x_n, dx_{n+1}) = h(e^{-\beta\Delta H(x_n)})\delta_{\Phi_{\delta t}(x_n)}(dx_{n+1}) + (1 - h(e^{-\beta\Delta H(x_n)}))\delta_{S(x_n)}(dx_{n+1}).$$

and composing with $e^{-\beta H(x_n)}\sigma(dx_n)$ we obtain

$$\begin{aligned} e^{-\beta H(x_n)}M(x_n, dx_{n+1})\sigma(dx_n) &= \\ &= (h(e^{-\beta\Delta H})e^{-\beta H})(\Phi_{\delta t}^{-1}(x_{n+1}))\delta_{\Phi_{\delta t}^{-1}(x_{n+1})}(dx_n)\sigma(dx_{n+1}) + \\ &+ (1 - h(e^{-\beta\Delta H}))e^{-\beta H}(S^{-1}(x_{n+1}))\delta_{S^{-1}(x_{n+1})}(dx_n)\sigma(dx_{n+1}), \end{aligned}$$

and substituting (C.3) in the first part of the right hand side yields (C.2). \square

C.3. Exact sampling of fluctuation/dissipation perturbations. In this section, we recall how to perform exact sampling of fluctuation/dissipation perturbations. Since we only work with impulses, we refer to the system by using the impulse variables p only. Note that throughout the paper, we also use extended variables (p, p_z) , however, the presentation that follows covers general cases. The kinetic energy of the system is $\frac{1}{2}p^T M p$. We impose constraints $p^T M^{-1} \nabla_q \xi = 0$ on impulses, thus $p \in T_q^* \mathcal{M}$ and hence the associated orthogonal projector on $T_q^* \mathcal{M}$ is

$$P = \text{Id} - \nabla_q \xi G^{-1} \nabla_q^T \xi M^{-1}.$$

The stochastic differential equations of motion on impulses that are integrated on a time-step interval are

$$\begin{cases} \dot{p} = -\gamma M^{-1} p + \sigma \dot{W} - \nabla_q \xi \dot{\lambda}, \\ p^T M^{-1} \nabla_q \xi = 0, \end{cases} \quad (\text{C.4})$$

with the usual fluctuation/dissipation relation $\sigma \sigma^T = 2\beta^{-1}\gamma$. The Gaussian distribution of impulses

$$\frac{1}{Z} e^{-\frac{\beta}{2} p^T M^{-1} p} \sigma_{T_q^* \mathcal{M}}(dp) \quad (\text{C.5})$$

is invariant under the dynamics (C.4).

PROPOSITION C.3 (Exact sampling of stochastic perturbation). *Given the mass matrix M , suppose either δt or γ are small enough so that the condition*

$$\frac{\delta t}{2} M^{-1} \leq \gamma \quad (\text{C.6})$$

holds in the sense of non-negative symmetric matrices. Let U be a centered and normalized Gaussian vector. Consider the mid-point Euler scheme with constraints

$$\begin{cases} p_{n+1} = p_n - \frac{\delta t}{2} \gamma M^{-1} (p_n + p_{n+1}) + \sqrt{\delta t} \sigma U - \nabla_q \xi \lambda_{n+1} \\ p_{n+1}^T M^{-1} \nabla_q \xi = 0, \end{cases} \quad (\text{C.7}) \quad (C_p)$$

where λ_{n+1} is the Lagrange multiplier associated with the constraint (C_p) . The Markov kernel defined by the transition $p_n \rightarrow p_{n+1}$ is reversible with respect to the Gaussian distribution (C.5).

Proof. After calculating the Lagrange multiplier, (C.7) can be written as

$$p_{n+1} = p_n - \frac{\delta t}{2} P \gamma P^T M^{-1} (p_n + p_{n+1}) + \sqrt{\delta t} P \sigma U.$$

Consider the new variable $\tilde{p} = \beta^{1/2} M^{-1/2} p$, and define the symmetric matrix

$$L \equiv \frac{\delta t}{2} M^{-1/2} P \gamma P^T M^{-1/2},$$

as well as K , such that $KK^T = L$. In terms of these new variables we obtain from (C.7)

$$\tilde{p}_{n+1} = (\text{Id} + L)^{-1} (\text{Id} - L) \tilde{p}_n + 2(\text{Id} + L)^{-1} K U. \quad (\text{C.8})$$

Moreover, the product measure $\sigma_{T_q^* \mathcal{M}_0}(dp_n) \sigma_{T_q^* \mathcal{M}_0}(dp_{n+1})$ is the measure induced on the linear subspace of constraints by the scalar product M^{-1} and the Lebesgue measure $dp_n dp_{n+1}$. Thus in the variables $(\tilde{p}_n, \tilde{p}_{n+1})$ this measure becomes, up to a constant, the measure induced by the usual Euclidean structure. As a consequence the log density of the random variable $(\tilde{p}_n, \tilde{p}_{n+1})$ defined by (C.8) with respect to this latter measure equates to

$$\begin{aligned} & -\frac{1}{2} |\tilde{p}_n|^2 - \frac{1}{8} (\tilde{p}_{n+1} - (\text{Id} + L)^{-1} (\text{Id} - L) \tilde{p}_n)^T L^{-1} (\text{Id} + L)^2 (\tilde{p}_{n+1} - (\text{Id} + L)^{-1} (\text{Id} - L) \tilde{p}_n) \\ & = -\frac{1}{8} \tilde{p}_{n+1}^T L^{-1} (\text{Id} + L)^2 \tilde{p}_{n+1} - \frac{1}{8} \tilde{p}_n^T L^{-1} (\text{Id} + L)^2 \tilde{p}_n, \end{aligned}$$

which is indeed symmetric between \tilde{p}_n and \tilde{p}_{n+1} . Hence we have shown the reversibility of the induced Markov kernel and consequently stationarity of the canonical Gaussian distribution.

□

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