Geometric Numerical Integration
5. Adaptive geometric integrators

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BCAM, May 20-24 2019

Universidad de Oviedo

Index course

1. Hamiltonian mechanics and numerical methods
2. Symplectic integration
3. Modified equations
4. Constrained mechanical systems
5. **Adaptive geometric integrators**

Molecular simulation

Molecular simulation

Motivation

- **Importance:** Information about the macroscopic behavior of molecules can be obtained from simulations of a system at an atomistic level.

- **Applications:** Drug design, computational biology, nanoscience, materials science...

- **Challenges:** Improve the accuracy and the sampling quality of simulations without increasing the computational cost.

- **Sampling techniques:** Molecular Dynamics **MD**,

  Monte Carlo **MC**, Hybrid Monte Carlo **HMC**.

Molecular simulation
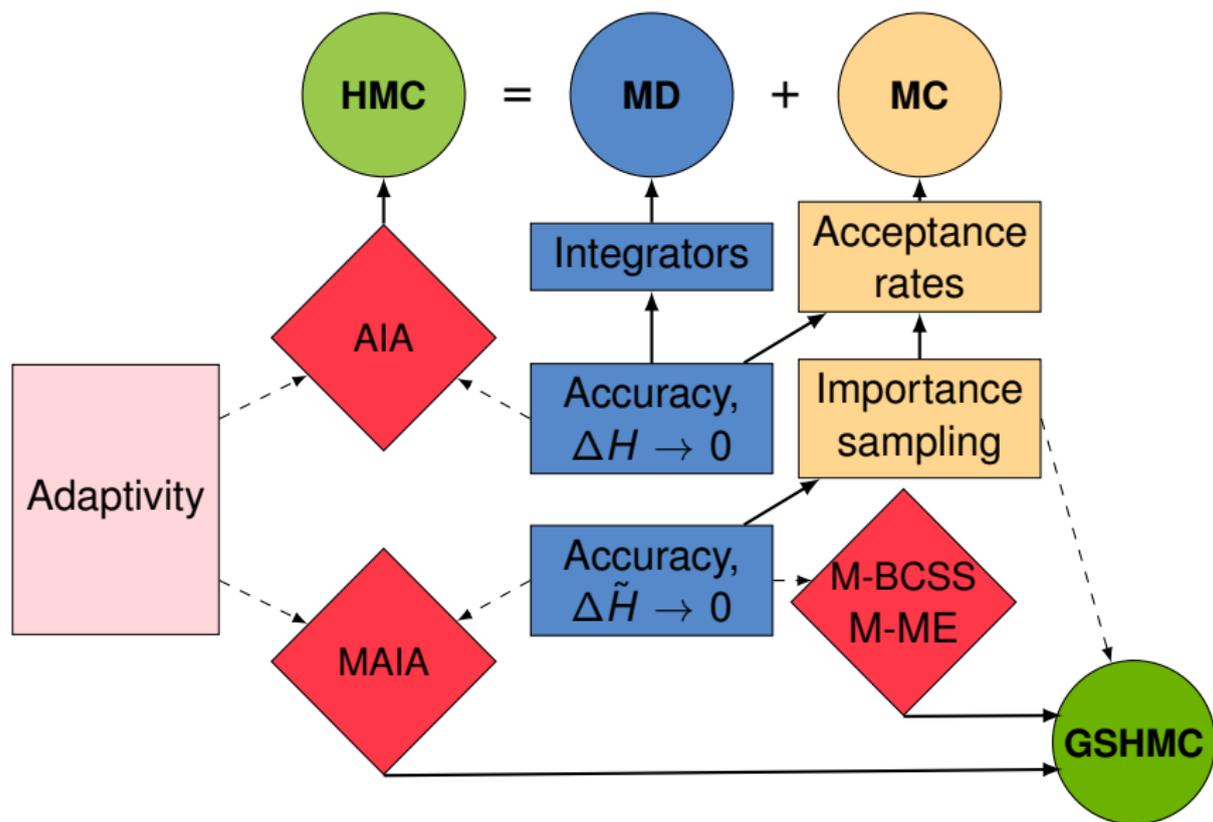
Monte Carlo

Monte Carlo

Metropolis algorithm is a Markov Chain Monte Carlo (MCMC) algorithm.

▶ It is associated with a computer called MANIAC built in Los Alamos under the direction of Nicolas Metropolis in 1952.

▶ The method was designed for simulation of a liquid in equilibrium with its gas phase and it was published in 1953.

▶ The idea of the method was to create a Markov chain (a random walk in thermodynamic phase space) of molecular configurations whose appearance is proportional to their statistical probability.

4

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▶ The idea of the method was to create a Markov chain (a random walk in thermodynamic phase space) of molecular configurations whose appearance is proportional to their statistical probability.

Generalized as the **Metropolis-Hastings** algorithm in 1970.

Monte Carlo

We are interested in **sampling** the positions $\mathbf{q} \in \mathbb{R}^d$ distributed according to the Boltzmann distribution:

$$\pi(\mathbf{q}) = \frac{1}{\mathcal{Z}} \exp\left(-\beta U(\mathbf{q})\right),$$

where $\mathcal{Z}$ is a normalizing constant, $U(\mathbf{q})$ the potential energy and β the inverse of the thermodynamic temperature $k_B T$.

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$$\pi(\mathbf{q}) \propto \exp\left(-\beta U(\mathbf{q})\right).$$

Monte Carlo

- ▶ Configurations are generated as a Markov chain defined by a conditional transition probability densities $\rho_T(\mathbf{q} \to \mathbf{q}')$.
- ▶ **Detailed balance** condition

$$\pi(\mathbf{q})\rho_T(\mathbf{q} \to \mathbf{q}') = \pi(\mathbf{q}')\rho_T(\mathbf{q}' \to \mathbf{q})$$

ensures that the Markov chain converges to the unique stationary probability distribution $\pi(\mathbf{q})$.

- ▶ One step $\mathbf{q} \to \mathbf{q}'$ is realized by proposing $\mathbf{q}'$ according to a proposal probability density $\rho_P(\mathbf{q} \to \mathbf{q}')$ and accepting it with the probability

$$P_A(\mathbf{q} \to \mathbf{q}') = \min \left\{ 1, \frac{\pi(\mathbf{q}')\rho_P(\mathbf{q}' \to \mathbf{q})}{\pi(\mathbf{q})\rho_P(\mathbf{q} \to \mathbf{q}')} \right\} = \min \left\{ 1, \exp\left(-\beta \Delta U\right) \right\}.$$

Monte Carlo

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- ▶ Thus,

$$\rho_T(\mathbf{q} \to \mathbf{q}') = \rho_P(\mathbf{q} \to \mathbf{q}')P_A(\mathbf{q} \to \mathbf{q}').$$

Monte Carlo

Drawbacks

- ► Only local moves are performed. It may lead to slow exploration of phase space.
- ► Highly correlated samples.
- ► No information on the dynamics.

Molecular dynamics

Molecular dynamics

- ▶ It estimates equilibrium and dynamic properties of complex systems by numerically solving equations of motion.

- ▶ Inspired by the success of the Monte Carlo simulations (Metropolis et al., 1953), Fermi, Pasta, Ulam and Tsingou first suggested MD in the mid-50s.

- ▶ In 1957, Alder and Wainwright used an IBM 704 computer to simulate elastic collisions between hard spheres.

- ▶ In 1964, Rahman published simulations of liquid argon that used a Lennard-Jones potential.

Molecular dynamics

Let a **Hamiltonian** be

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

with **p** the momenta and M a mass matrix. Then, the associated **equations of motion** are

$$\frac{\mathrm{d}}{\mathrm{d}t}\mathbf{q} = M^{-1}\mathbf{p}, \quad \frac{\mathrm{d}}{\mathrm{d}t} = \mathbf{p} - \nabla_{\mathbf{q}} U(\mathbf{q}).$$

Molecular dynamics

Knowing the **flow** $\varphi_t : \mathbb{R}^D \to \times \mathbb{R}^D$

$$(\mathbf{q}(t), \mathbf{p}(t)) = \varphi_t(\mathbf{q}(0), \mathbf{p}(0)),$$

the system is moved **deterministically** through phase space.

As we have seen, we simulate Hamiltonian dynamics **numerically**.

Molecular dynamics

Important features

- **Low integration error**.
- **Time-reversible**.
- **Symplectic**.
- **Computationally efficient**.
- **Stable**.

Molecular dynamics

Important features

- ► **Low integration error**.
- ► **Time-reversible**.
- ► **Symplectic**.
- ► **Computationally efficient**.
- ► **Stable**.

Velocity Verlet: a golden standard for Hamiltonian-dynamics-based simulation methods.

Molecular dynamics

Velocity Verlet

- ▶ Second order,
- ▶ time-reversible,
- ▶ symplectic,
- ▶ computationally efficient,
- ▶ **easy to implement**.

Molecular dynamics

Drawbacks

- ▶ Not free of discretization errors.
- ▶ Large moves are not allowed. The step size Δt has to be small.
- ▶ No rigorous temperature control.

Hybrid Monte Carlo
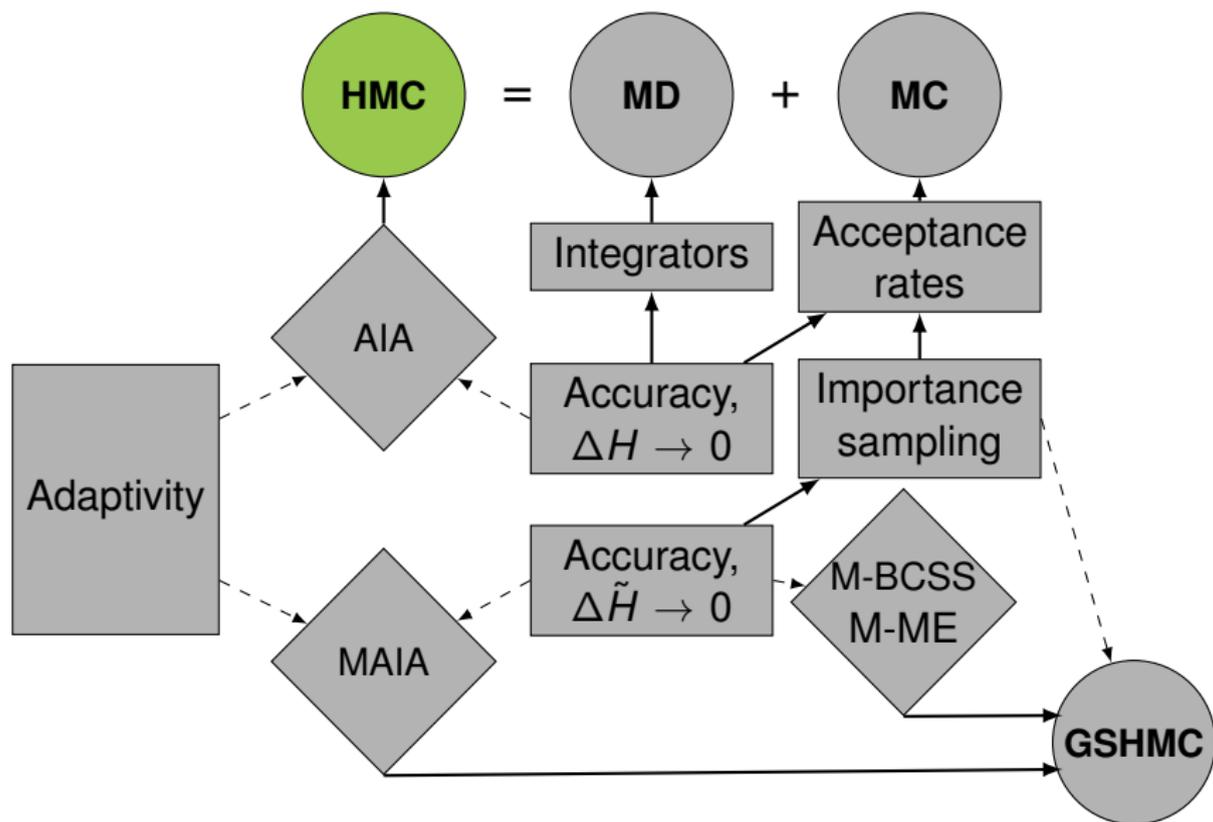
Hybrid Monte Carlo

Hybrid Monte Carlo

▶ HMC is a popular technique for sampling of high dimensional and complex systems proposed in Duane et al., "Hybrid Monte Carlo", 1987.

▶ Originally applied to lattice field theory simulations.

▶ Applications from molecular to statistical problems.

▶ Combines deterministic (Hamiltonian dynamics) with stochastic approaches (Metropolis Monte Carlo sampling).

Hybrid Monte Carlo

Objective

To sample positions $\mathbf{q}$ from the **target density**
$\pi(\mathbf{q}) \propto \exp(-\beta U(\mathbf{q}))$ **taking advantage of the main features of both molecular dynamics (MD) and Monte Carlo (MC)**.

Method

- ▶ Introduce the **auxiliary momenta $\mathbf{p} \in \mathbb{R}^d$**, independent of $\mathbf{q}$ and Maxwell-Boltzmann distributed.
- ▶ Define an **augmented target density** $\pi(\mathbf{q}, \mathbf{p}) \propto \exp(-\beta H(\mathbf{q}, \mathbf{p}))$, where $H(\mathbf{q}, \mathbf{p}) = K(\mathbf{p}) + U(\mathbf{q})$ is the Hamiltonian with $K(\mathbf{p})$ and $U(\mathbf{q})$ being the kinetic and potential energy, respectively.
- ▶ **Generate a proposal using Hamiltonian dynamics** and **accept/reject it with a Metropolis test**.
- ▶ Simulate a **Markov chain** $\{\mathbf{q}_n, \mathbf{p}_n\}_{n=1}^{N}$ with **invariant distribution** $\pi(\mathbf{q}, \mathbf{p})$ and **recover the target** $\pi(\mathbf{q})$ by marginalizing out $\mathbf{p}$.

Hybrid Monte Carlo
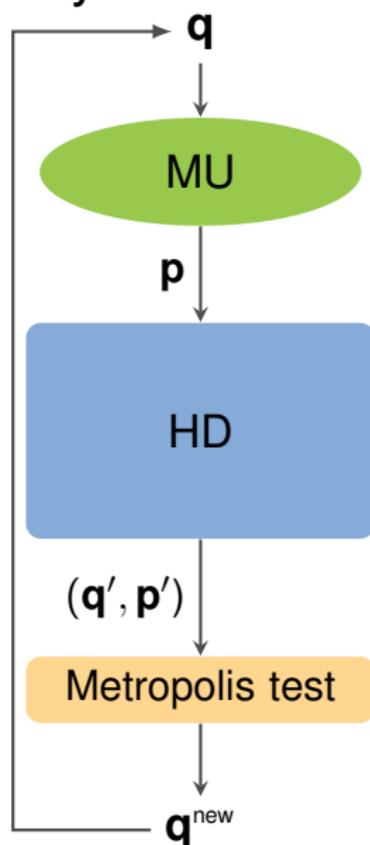
Momentum update

Draw momenta from Maxwell-Boltzmann distribution $\exp(-\beta\frac{1}{2}\mathbf{p}^T M^{-1}\mathbf{p})$ with M the mass matrix

Hamiltonian dynamics

L steps of integration of equations of motion with a symplectic numerical integrator $\Psi_{\Delta t,L}$ and step size Δt generating proposal

$$(\mathbf{q}', \mathbf{p}') = \Psi_{\Delta t,L}(\mathbf{q}, \mathbf{p}).$$

Metropolis test

$$\mathbf{q}^{\text{new}} = \left\{ \begin{array}{ll} \mathbf{q}' & \text{with probability } \alpha = \min\left\{1, \exp(-\beta\Delta H)\right\} \\ \mathbf{q} & \text{otherwise} \end{array} \right.$$

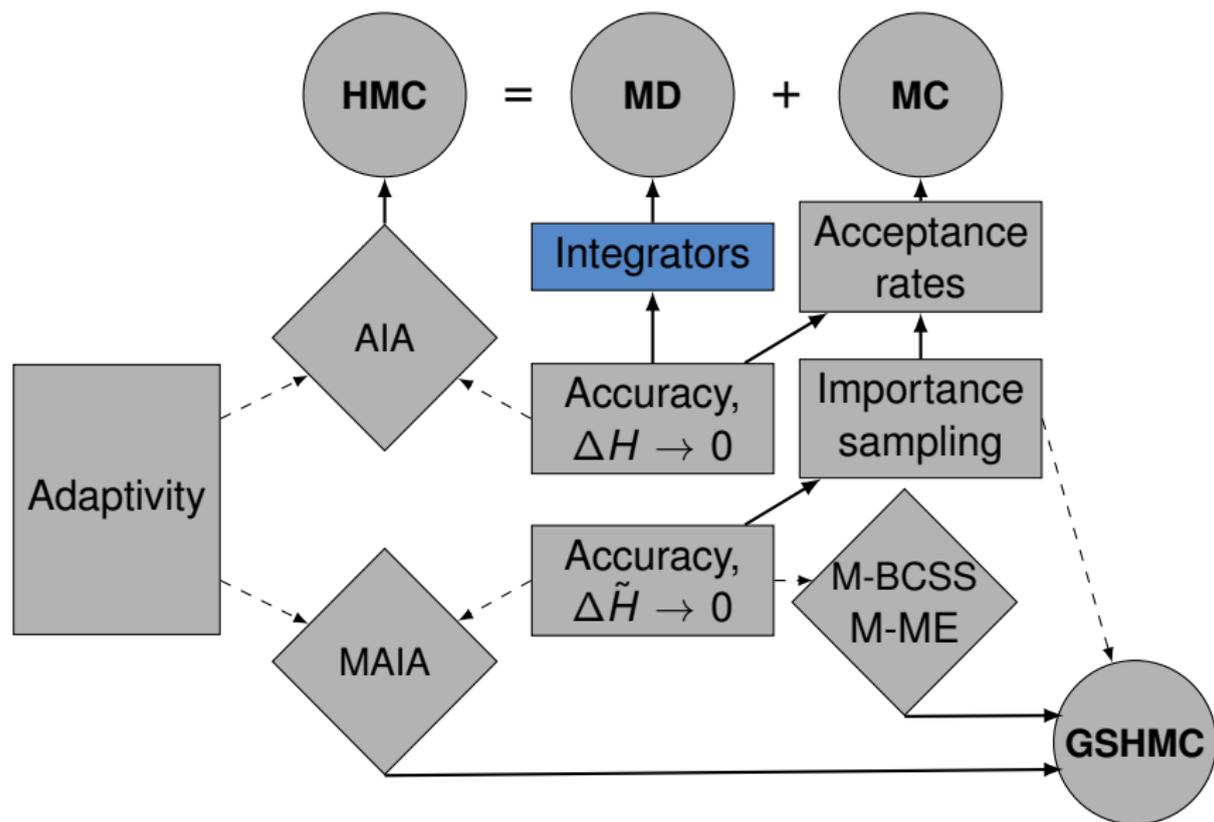$\Delta H = H(\mathbf{q}', \mathbf{p}') - H(\mathbf{q}, \mathbf{p}) \neq 0$ (due to numerical error)

19

Hybrid Monte Carlo

Drawbacks

► No information on the dynamics.
► Loses accuracy with increasing the dimension D or the step size Δt.

Integrators

Integrators

Integrators

Equations of motion

Consider a **separable Hamiltonian**

$$H(\mathbf{q}, \mathbf{p}) = \frac{1}{2}\mathbf{p}^T M^{-1}\mathbf{p} + U(\mathbf{q}) \equiv A(\mathbf{p}) + B(\mathbf{q}),$$

with associated **equations of motion**:

$$\frac{\mathrm{d}}{\mathrm{d}t}\mathbf{q} = M^{-1}\mathbf{p}, \quad \frac{\mathrm{d}}{\mathrm{d}t}\mathbf{p} = -\nabla U(\mathbf{q}).$$

Integrators

Solution flows

The equations of motion can be integrated in closed form and their **solution flows** at a time t are given by

$$(\mathbf{q}(t), \mathbf{p}(t)) = \varphi_t^{(A)}(\mathbf{q}(0), \mathbf{p}(0)), \quad \mathbf{q}(t) = \mathbf{q}(0) + t\, M^{-1} \mathbf{p}(0), \quad \mathbf{p}(t) = \mathbf{p}(0),$$

and

$$(\mathbf{q}(t), \mathbf{p}(t)) = \varphi_t^{(B)}(\mathbf{q}(0), \mathbf{p}(0)), \quad \mathbf{q}(t) = \mathbf{q}(0), \quad \mathbf{p}(t) = \mathbf{p}(0) - t\, \nabla U(\mathbf{q}(0)).$$

$\phi_t^{(X)}$ t-**flow** of $X = \{A, B\}$ are **symplectic** and **time-reversible**.

Integrators

We have seen that *Velocity Verlet* is a golden standard for Hamiltonian-dynamics-based simulation methods.
Our aim here is to outperform it using more sophisticated symplectic methods.

Integrators

One-parameter family of **two-stage schemes** are competitors to Verlet:

$$\psi_{\Delta t} = \varphi^{(B)}_{b\Delta t} \circ \varphi^{(A)}_{\Delta t/2} \circ \varphi^{(B)}_{(1-2b)\Delta t} \circ \varphi^{(A)}_{\Delta t/2} \circ \varphi^{(B)}_{b\Delta t},$$

with $b \in (0, 0.25]$.

- ► Higher accuracy (substeps $\Delta t/2$).
- ► Bigger step sizes (stability interval up to $(0, 4)$).
- ► Verlet is a special case with $b = 0.25$: $\psi^{\text{Verlet}}_{\Delta t} = \phi^B_{\Delta t/2} \circ \phi^A_{\Delta t} \circ \phi^B_{\Delta t/2}$. - velocity Verlet (VV).

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Remark

Two force evaluations per step. Fair comparison with Verlet:

$$\Delta t_{\text{Verlet}} = \Delta t_{\text{two-stage}}/2, \quad L_{\text{Verlet}} = 2L_{\text{two-stage}}.$$

Integrators

ME (McLachlan, "On the Numerical Integration of Ordinary Differential Equations by Symmetric Composition Methods", 1995)

- $b = 0.1932$, the parameter that minimizes the **energy error**.
- Outperforms velocity Verlet for **small step sizes**.
- Performance **degrades for large step sizes**.

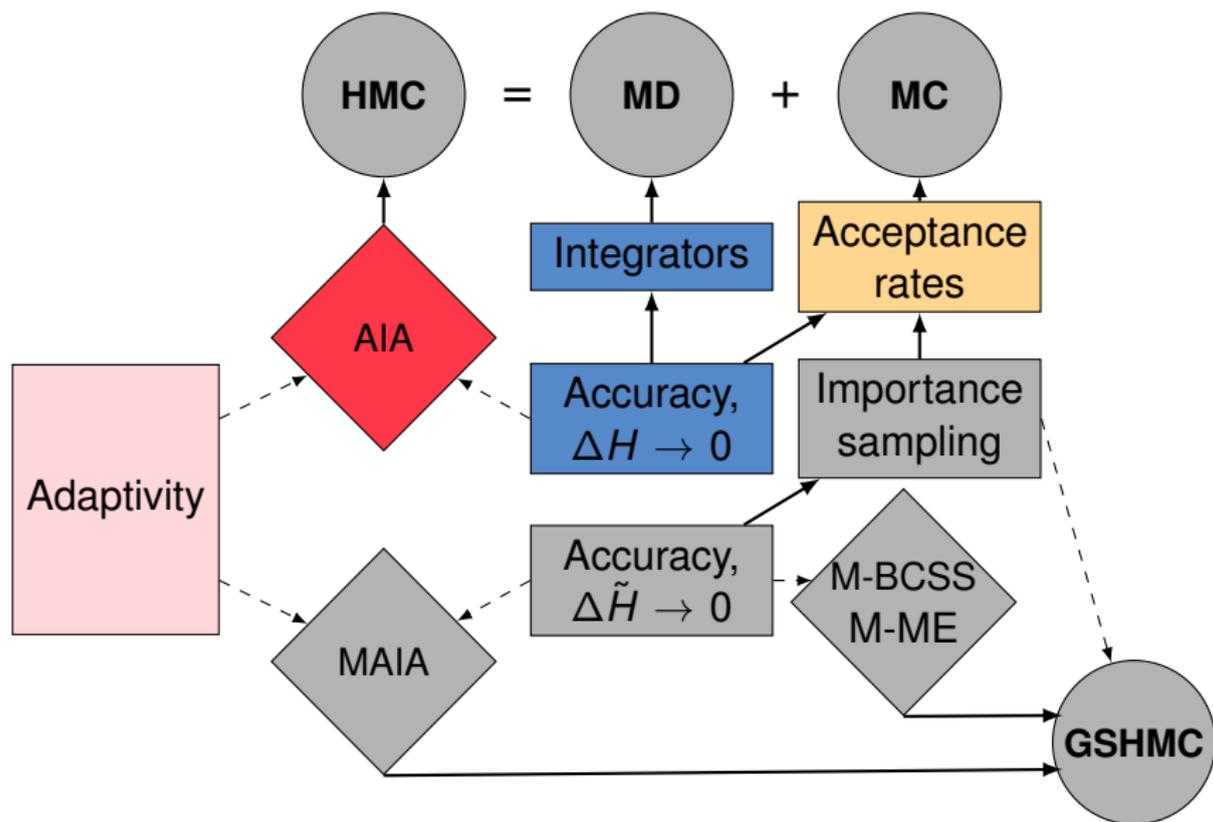**BCSS** (Blanes, Casas, and Sanz-Serna, "Numerical Integrators for the Hybrid Monte Carlo Method", 2014)

- $b = \arg\min\|\rho\|_{(2)} = \arg\min \max_{0<h<2} \rho(h, b) = 0.2113$, where $\rho(h, b)$ is an **upper bound for the expectation of the energy error** and h a **dimensionless step size**.
- Shows the best performance **around** $h = 2$.
- Performance may drop for **smaller or larger** h.

Integrators

What are the best values of b?

Adaptive Integration Approach

Adaptive Integration Approach

Adaptive Integration Approach

The **Adaptive Integration Approach (AIA)** method was presented in Fernández-Pendás, Akhmatskaya, and Sanz-Serna, "Adaptive multi-stage integrators for optimal energy conservation in molecular simulations", 2016.

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- ▶ **Objective**: To define a **system specific**, unique for each application, **two-stage integrator** which provides the best simulation **accuracy** and **performance**.

Adaptive Integration Approach

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- ▶ **Objective**: To define a **system specific**, unique for each application, **two-stage integrator** which provides the best simulation **accuracy** and **performance**.

- ▶ **Idea**: **Using the information about the system**, i.e., the highest oscillation frequency and the desired step size, find a parameter *b* for a two-stage integrator that **guarantees the best conservation of energy for harmonic forces**.

Adaptive Integration Approach

Then, we want to find b by minimizing $\max_{0<h<\tilde{h}} \rho(h, b)$, with $\tilde{h}(\Delta t, \omega)$.

$\rho(h, b)$ is an **upper bound for the expectation of the energy error** in the integration of the standard harmonic oscillator[1]:

$$\mathbb{E}(\Delta H) \leq \rho(h, b) = \frac{(B_h + C_h)^2}{2(1 - A_h^2)},$$

where A_h, B_h and C_h are functions of the integrator parameter b and the dimensionless step size h.

$$\rho(h, b) = \frac{h^4(2b^2(1/2 - b)h^2 + 4b^2 - 6b + 1)^2}{8(2 - bh^2)(2 - (1/2 - b)h^2)(1 - b(1/2 - b)h^2)}.$$

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[1] Blanes, Casas, and Sanz-Serna, "Numerical Integrators for the Hybrid Monte Carlo Method", 2014.

Adaptive Integration Approach

Algorithm

1. Given:
   - ▶ The integration step size Δt, in units of measure,
   - ▶ $\tilde{\omega}$, the fastest oscillatory frequency in the system.
2. Calculate the normalized step size

$$\tilde{h} := S\tilde{\omega}\Delta t,$$

   where $S = \sqrt{2}$.
3. Find $b^* = \arg\min\|\rho\|_{(\tilde{h})} = \arg\min \max_{0 < h < \tilde{h}} \rho(h, b)$.
4. The resulting adaptive integrator:

$$\psi_{\Delta t} = \varphi^B_{b^* \Delta t} \circ \varphi^A_{\Delta t/2} \circ \varphi^B_{(1-2b^*)\Delta t} \circ \varphi^A_{\Delta t/2} \circ \varphi^B_{b^* \Delta t}.$$

Adaptive Integration Approach

▶ **No computational overheads** in simulations.

▶ Extended to the use of **holonomic constraints**: $g(\mathbf{q}) = 0$.
  ▶ Presented the two-stage schemes as two concatenated velocity Verlet steps and combine with RATTLE.

Adaptive Integration Approach

Why $\sqrt{2}$?

We study in detail the behavior of the velocity Verlet scheme for the classic harmonic oscillator with potential energy $U(q) = (k/2)q^2$, where $k > 0$ is the force constant.
Thus, the forces are computed as $F(q) = -\omega^2 q$. The equations of motion are

$$\frac{\mathrm{d}}{\mathrm{d}t}q = \frac{p}{M}, \quad \frac{\mathrm{d}}{\mathrm{d}t}p = -kq.$$

The angular frequency is expressed in terms of the force constant as $\omega = \sqrt{k/M}$. We can assume for the sake of simplicity that the mass is trivial and then $p/M = v$ and $\omega = \sqrt{k}$.

Adaptive Integration Approach

A transformation matrix M is used to relate one phase point to the next[2]. Then, for a step size Δt,

$$\begin{bmatrix} \omega q(t + \Delta t) \\ v(t + \Delta t) \end{bmatrix} = M_{\Delta t} \begin{bmatrix} \omega q(t) \\ v(t) \end{bmatrix},$$

where

$$M_{\Delta t} = \begin{bmatrix} 1 - \frac{(\omega \Delta t)^2}{2} & \omega \Delta t \\ -\omega \Delta t + \frac{(\omega \Delta t)^3}{4} & 1 - \frac{(\omega \Delta t)^2}{2} \end{bmatrix}.$$

The numerical integrator is stable if the matrix $M_{\Delta t}$ is power bounded. This is satisfied if the eigenvalues of $M_{\Delta t}$ lie in the unit disc. The matrix S is power bounded if and only if

$$(\omega \Delta t)^2 < 4,$$

or, equivalently,

$$\Delta t < 2/\omega.$$

Adaptive Integration Approach

The restriction on the step size above is the **linear stability** condition for Verlet.

Under the linear stability assumption, the matrix $M_{\Delta t}$ has eigenvalues[3] $\exp(\pm i\theta)$, where

$$\theta = 2\arcsin(\omega\Delta t/2) \tag{1}$$

$$= \omega\Delta t + \frac{1}{24}(\omega\Delta t)^3 + \mathcal{O}(\omega\Delta t)^5. \tag{2}$$

Thus, **the angle θ depends on the step size and the frequency ω.**

---

[3] The angle is computed from the trace $\text{Tr}(M_{\Delta t}) = 2\cos\theta$. Thus, $\theta = \arccos\left(\text{Tr}(M_{\Delta t})/2\right) = \arccos\left(1 - (\omega\Delta t)^2/2\right) = 2\arcsin\left(\omega\Delta t/2\right)$.

Adaptive Integration Approach

The step size-dependent behavior of the transformation $M_{\Delta t}$ can be interpreted as follows. Equations (1) and (2) show that **the integrator uses θ as an approximation to the exact rotation** $\omega \Delta t$. *The smaller the step size, the closer the approximation is.*

Adaptive Integration Approach

The step size-dependent behavior of the transformation $M_{\Delta t}$ can be interpreted as follows. Equations (1) and (2) show that **the integrator uses θ as an approximation to the exact rotation $\omega \Delta t$.** *The smaller the step size, the closer the approximation is.*

Thus, one can define the **effective rotation θ_{eff}** as

$$\theta_{\text{eff}} = \omega_{\text{eff}} \, \Delta t.$$

For the Verlet method, the effective rotation is given by equation (1)

$$\theta_{\text{eff}}^{\text{Verlet}} = 2 \arcsin(\omega_{\text{eff}} \, \Delta t/2). \qquad (3)$$

Adaptive Integration Approach

For periodic motion with natural frequency ω, **nonphysical resonance (an artifact of the symplectic integrator) can occur when ω is related by relatively prime integers n and m to the forcing frequency ($2\pi/\Delta t$)** (cf. Arnold, *Mathematical methods of classical mechanics*, 1989):

$$\frac{n}{m}\omega = \frac{2\pi}{\Delta t}.$$

Here n is the **resonance order**.

Adaptive Integration Approach

Now, if we take the Verlet's step size-dependent frequency ω_{eff},

$$\frac{n}{m}\omega_{\text{eff}} = \frac{2\pi}{\Delta t}. \tag{4}$$

A **resonance of order** $n : m$ means that n phase space points are sampled in m revolutions:

$$n\,\theta_{\text{eff}} = n\,\Delta t\,\omega_{\text{eff}} = 2\pi m.$$

This special, finite-coverage of phase space can lead to incorrect, limited sampling of configuration space.

Adaptive Integration Approach

It has been shown[4] that equation (3) can be used to formulate **a condition for a resonant step size** for the harmonic oscillator system. That is, using

$$\omega_{\text{eff}}^{\text{Verlet}} = \frac{2 \sin^{-1}(\omega \, \Delta t/2)}{\Delta t},$$

with the resonance condition in (4), we have

$$\frac{\omega \, \Delta t}{2} = \sin\left(\frac{m\pi}{n}\right).$$

Equivalently,

$$\Delta t_{n:m}^{\text{Verlet}} = \frac{2}{\omega} \sin\left(\frac{m\pi}{n}\right).$$

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[4]Mandziuk and Schlick, "Resonance in the dynamics of chemical systems simulated by the implicit midpoint scheme", 1995.

41

Adaptive Integration Approach

To get the lowest-order resonances (which are the most severe) we take $m = 1$. *It is easy to see that for $n = 2$, we recover the linear stability condition*.

Since the limiting step sizes $\Delta t_{n:1}$ for resonance orders $n > 2$ are smaller than the linear stability limit $\Delta t_{2:1}$, **resonance limits the step size to values lower than classical stability**. Since the third-order resonance leads to instability and the fourth-order resonance often leads to instability in molecular simulation[5], in practice it is usually required that $\Delta t < \Delta t_{4:1}$.

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[5]It was predicted in Arnold, *Mathematical methods of classical mechanics*, 1989 that *instabilities are not observed for resonances of orders higher than four* and this assessment has been confirmed with experiments such as in Mandziuk and Schlick, "Resonance in the dynamics of chemical systems simulated by the implicit midpoint scheme", 1995.

Adaptive Integration Approach

This implies for Verlet a stricter restriction than (36)

$$\Delta t < \sqrt{2}/\omega, \tag{5}$$

which corresponds to the fourth-order resonance and is the **non linear stability** condition for Verlet. More resonance step size limits are summarized in the table below.

Order n	$\Delta t_{n:1}(\omega)$
2	$2/\omega$
3	$\sqrt{3}/\omega$
4	$\sqrt{2}/\omega$
5	$1.176/\omega$
6	$1/\omega$

Adaptive Integration Approach

▶ Testing system: **Spider toxin in membrane/water environment**[6]. Coarse grained **unconstrained** system with 7810 particles.
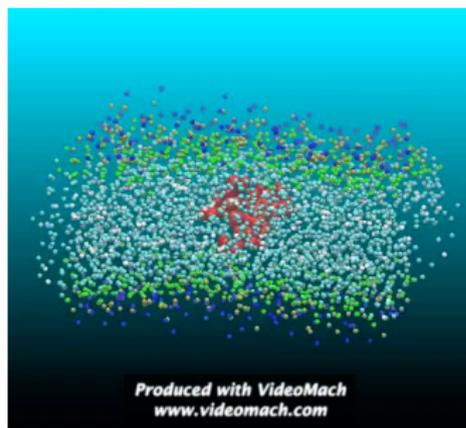
Toxin movement towards water/membrane interface.

All simulations correspond to 20 ns at 310 K.

[6]Wee et al., "Improved Sampling for Simulations of Interfacial Membrane Proteins: Application of Generalized Shadow Hybrid Monte Carlo to a Peptide Toxin/Bilayer System", 2008.

Adaptive Integration Approach

Dependence of b on Δt
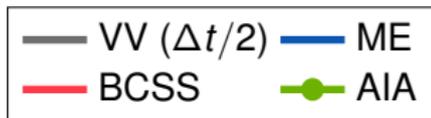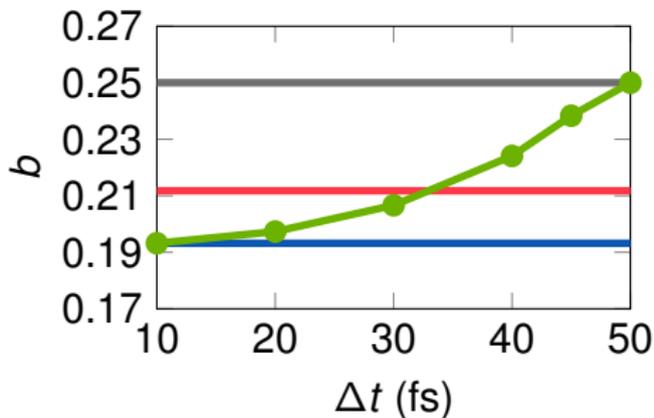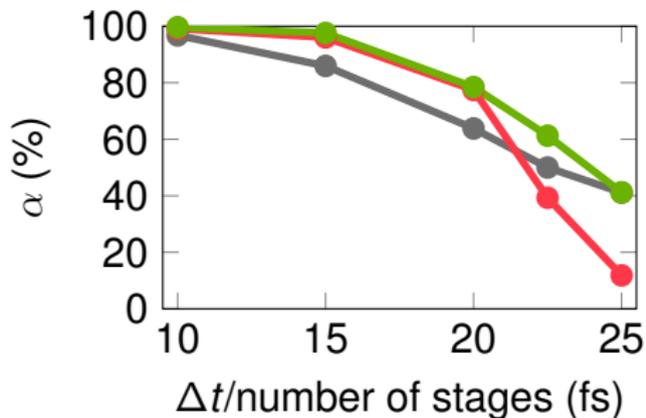
Acceptance rates

Adaptive Integration Approach

Dependence of b on Δt
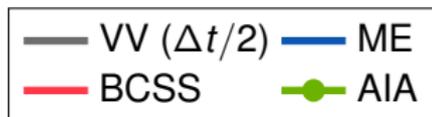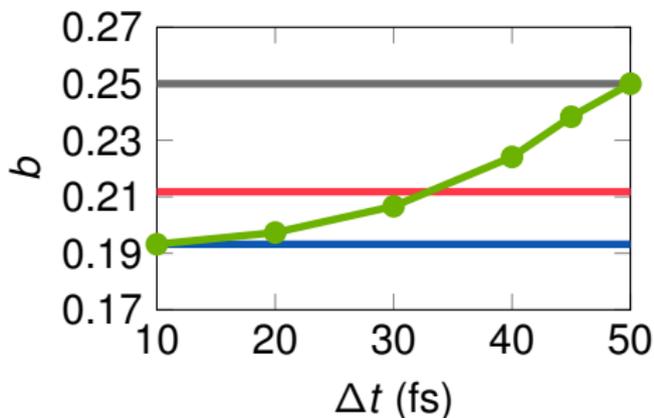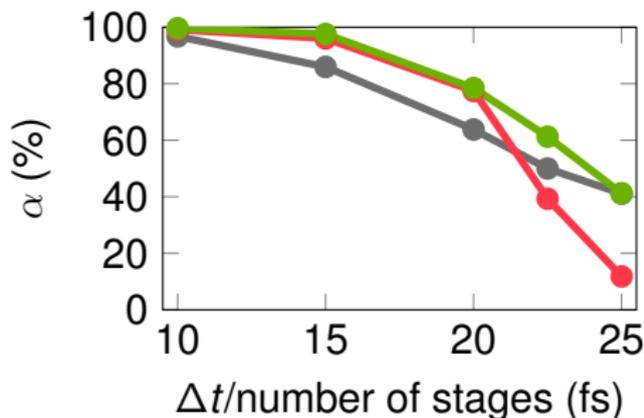
Acceptance rates

* The predicted by AIA values of b **confirm the known trends in behavior** of Verlet, ME and BCSS.
* AIA leads to the **best energy conservation** and thus to **the highest acceptance rates** for **all tested step sizes**.

Adaptive Integration Approach
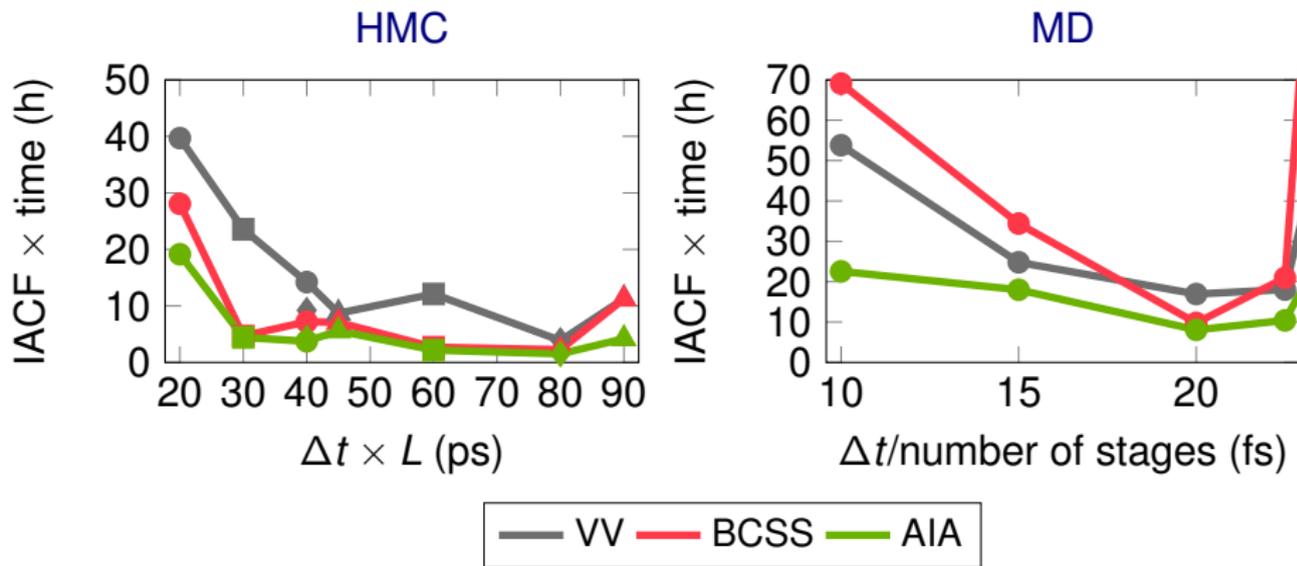
Time normalized Integrated Autocorrelation Functions (IACF)
Low IACF ⇒ efficient sampling
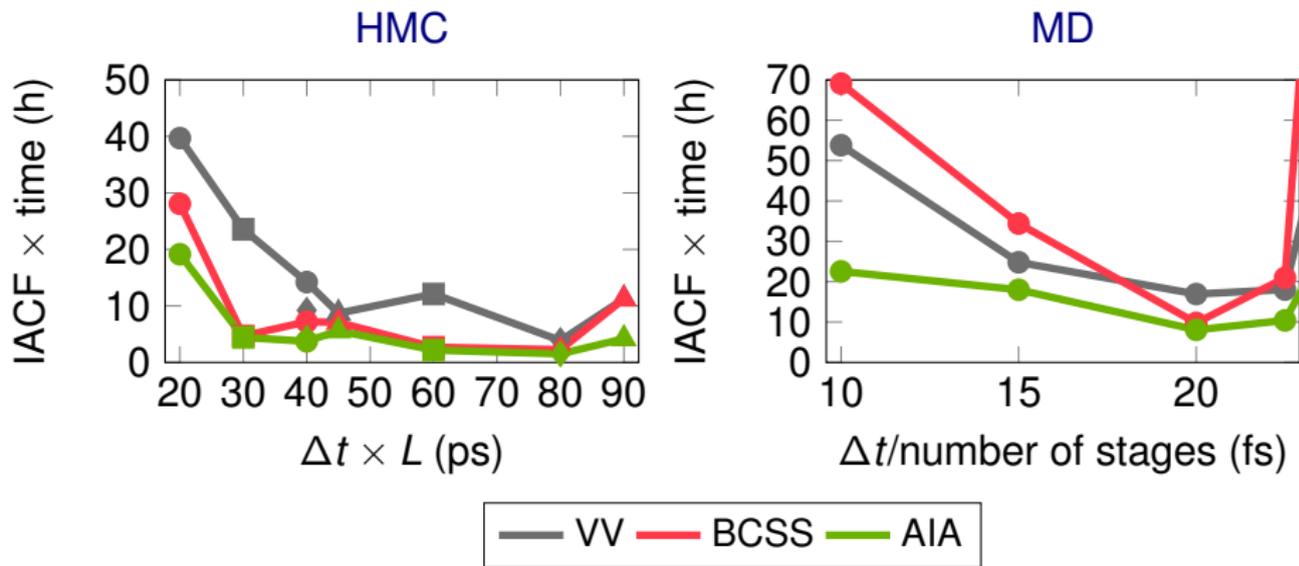
Adaptive Integration Approach

Time normalized Integrated Autocorrelation Functions (IACF)
Low IACF $\Rightarrow$ efficient sampling

* AIA provides the **lowest IACF** in both HMC and MD regardless a choice of step size Δt or trajectory length L. Up to 5 times better sampling.

46

Generalized Shadow Hybrid Monte Carlo

Generalized Shadow Hybrid Monte Carlo
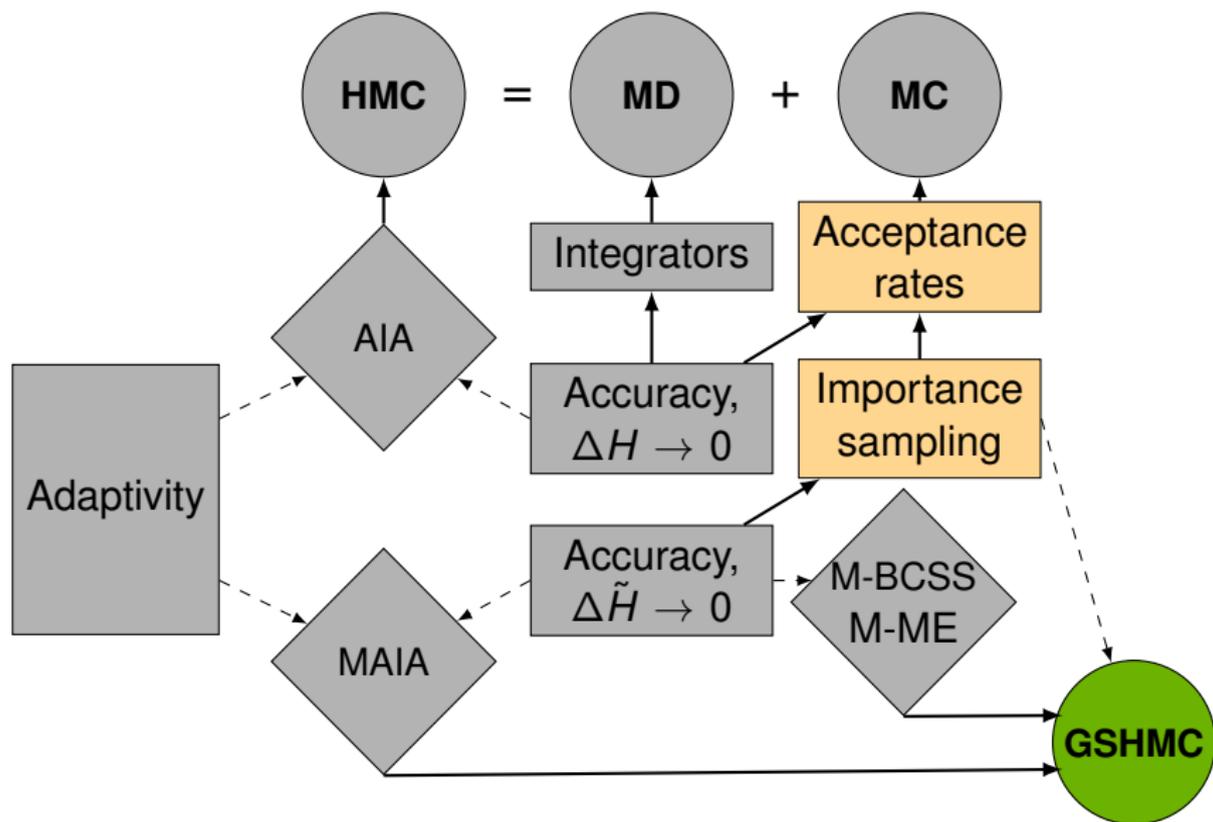
Generalized Shadow Hybrid Monte Carlo

Sampling efficiency of HMC can be enhanced through:

- **High acceptance rates**: Imply **less correlated sampling**.

- **Importance sampling**: A variance reduction method which **samples from an alternative distribution that overweights the important regions**.

---

[7]Akhmatskaya and Reich, "GSHMC: An efficient method for molecular simulation", 2008

Generalized Shadow Hybrid Monte Carlo

Sampling efficiency of HMC can be enhanced through:

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- **Importance sampling**: A variance reduction method which **samples from an alternative distribution that overweights the important regions**.

> Importance sampling HMC: GSHMC
>
> Generalized Shadow Hybrid Monte Carlo (GSHMC) method[7] improves sampling efficiency of HMC by sampling with respect to **modified Hamiltonians** and introducing a **partial momentum update**.

---

[7] Akhmatskaya and Reich, "GSHMC: An efficient method for molecular simulation", 2008

Generalized Shadow Hybrid Monte Carlo

▶ Sampling is performed with respect to a **modified canonical density**

$$\tilde{\pi} \propto \exp\left(-\beta \tilde{H}_{\Delta t}^{[m]}\right),$$

where

$$\tilde{H}_{\Delta t}^{[m]} = H + \Delta t^p H_{p+1} + \cdots + \Delta t^{m-1} H_m$$

is the *m*-th order modified or shadow Hamiltonian, an arbitrary accurate approximation of the Hamiltonian conserved by *p*-th order symplectic integrators to higher accuracy than true Hamiltonian: $\left\langle \Delta \tilde{H}_{\Delta t}^{[m]} \right\rangle = \mathcal{O}(D\Delta t^{2m})$ and $\langle \Delta H \rangle = \mathcal{O}(D\Delta t^{2p})$, $m > p$.

▶ Partial momentum update step combined with **modified Metropolis** test helps to retain dynamical information.

▶ **Reweighted averages** due to sampling in a modified ensemble:

$$\langle \Omega \rangle = \frac{\sum_{n=1}^{N} w_n \Omega_n}{\sum_{n=1}^{N} w_n}, \quad w_n = \exp\left(-\beta \left(H(\mathbf{q}_n, \mathbf{p}_n) - \tilde{H}_{\Delta t}^{[m]}(\mathbf{q}_n, \mathbf{p}_n)\right)\right),$$

with Ω_n values of an observable along a sequence of states $(\mathbf{q}_n, \mathbf{p}_n)$.

GSHMC
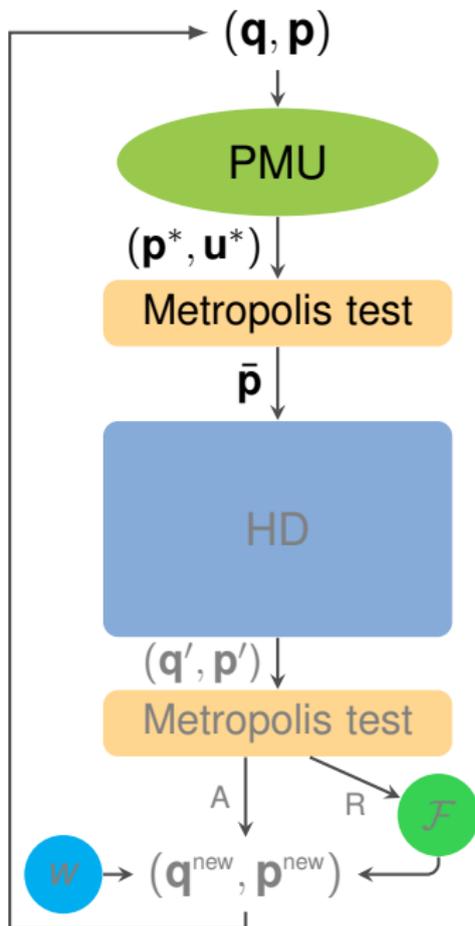
Partial Momentum Update

$$\mathbf{p}^* = \cos\varphi\, \mathbf{p} + \sin\varphi\, \mathbf{u}$$
$$\mathbf{u}^* = -\sin\varphi\, \mathbf{p} + \cos\varphi\, \mathbf{u},$$

where $\varphi \in (0, \pi/2]$.

Modified Metropolis test

$$\bar{\mathbf{p}} = \begin{cases} \mathbf{p}^* & \text{with prob. } \mathcal{P} \\ \mathbf{p} & \text{otherwise,} \end{cases}$$
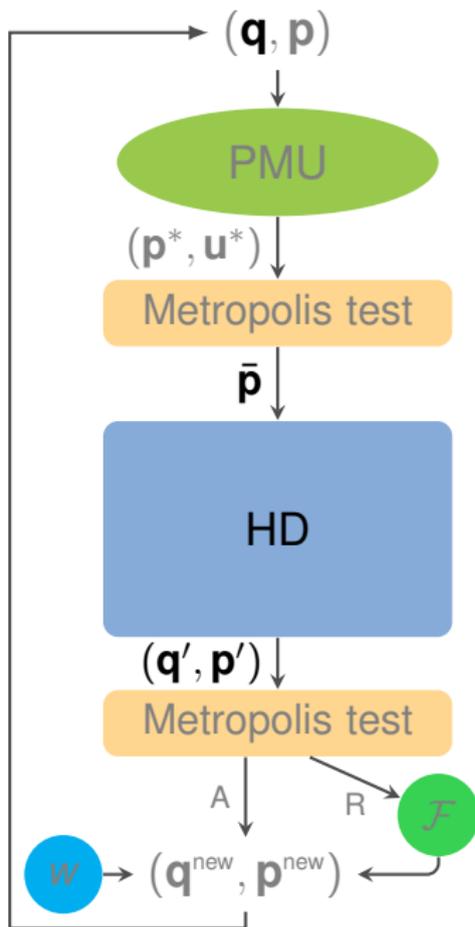
$$\mathcal{P} = \min\left\{ 1, \frac{\exp\left(-\beta\hat{H}(\mathbf{q}, \mathbf{p}^*, \mathbf{u}^*)\right)}{\exp\left(-\beta\hat{H}(\mathbf{q}, \mathbf{p}, \mathbf{u})\right)} \right\},$$

with the extended "Hamiltonian"

$$\hat{H}(\mathbf{q}, \mathbf{p}, \mathbf{u}) = \tilde{H}_{\Delta t}^{[m]}(\mathbf{q}, \mathbf{p}) + \frac{1}{2}\mathbf{u}^{\mathsf{T}} M^{-1}\mathbf{u}$$

defining the extended reference density
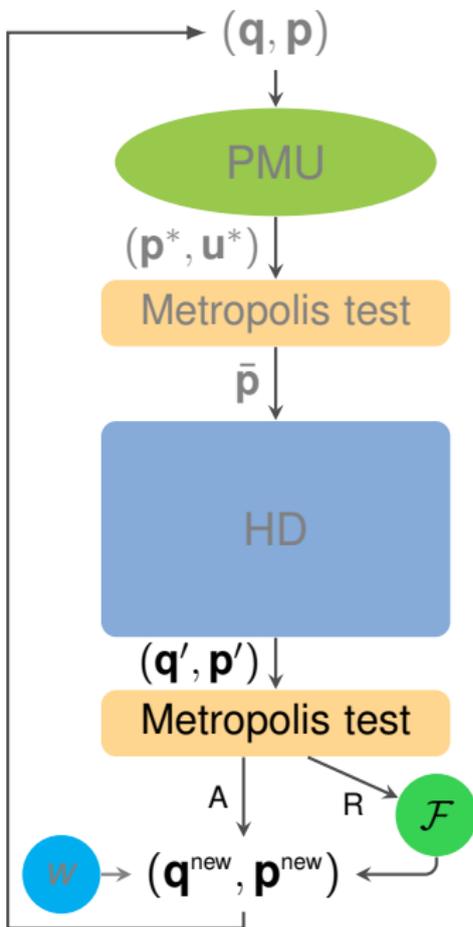$\hat{\pi}(\mathbf{q}, \mathbf{p}, \mathbf{u}) \propto \exp(-\beta\hat{H}(\mathbf{q}, \mathbf{p}, \mathbf{u})).$

51

Generalized Shadow Hybrid Monte Carlo

L steps of integration of equations of motion with a symplectic numerical integrator $\psi_{\Delta t, L}$ and step size Δt generating proposal

$$(\mathbf{q}', \mathbf{p}') = \psi_{\Delta t, L}(\mathbf{q}, \bar{\mathbf{p}}).$$
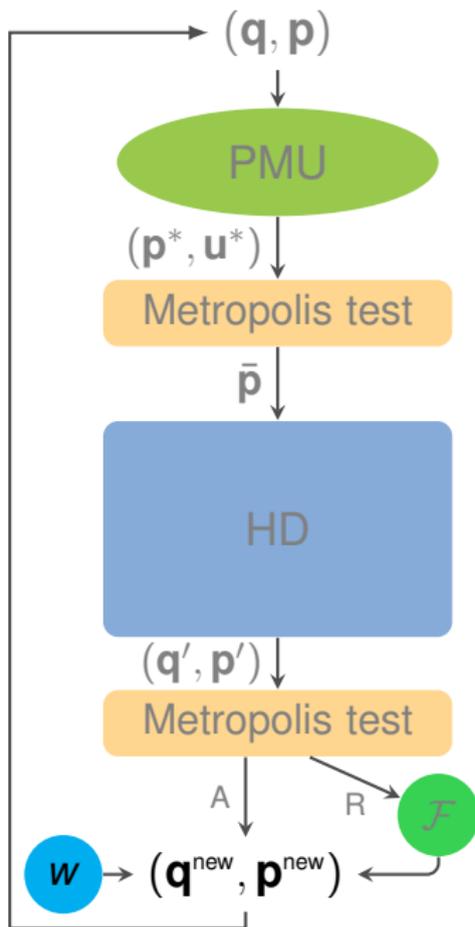
Generalized Shadow Hybrid Monte Carlo

$$(\mathbf{q}^{\text{new}}, \mathbf{p}^{\text{new}}) = \begin{cases} (\mathbf{q}', \mathbf{p}') & \text{with prob. } \alpha \\ \mathcal{F}(\mathbf{q}, \bar{\mathbf{p}}) & \text{otherwise,} \end{cases}$$

where $\mathcal{F}(\mathbf{q}, \mathbf{p}) = (\mathbf{q}, -\mathbf{p})$ and
$\alpha = \min\left\{1, \exp\left(-\beta\Delta\tilde{H}^{[m]}_{\Delta t}\right)\right\}.$

Generalized Shadow Hybrid Monte Carlo

$$w_n = \exp\left(-\beta\left(H(\mathbf{q}_n, \mathbf{p}_n) - \tilde{H}^{[m]}_{\Delta t}(\mathbf{q}_n, \mathbf{p}_n)\right)\right)$$
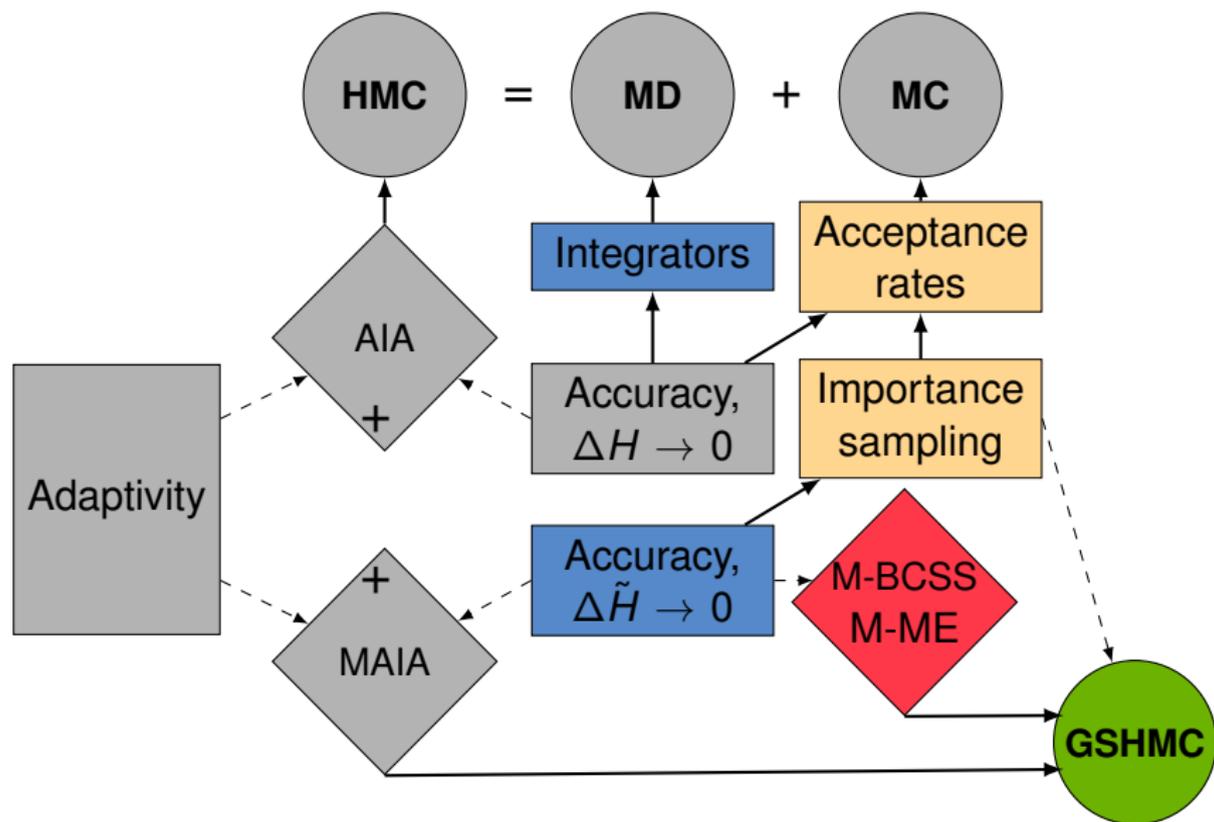stored for every $n = 1, \ldots, N$.

Generalized Shadow Hybrid Monte Carlo

Our objective: Increase the acceptance rates and investigate the potential of importance sampling.

Modified splitting schemes

Modified splitting schemes

Modified splitting schemes

> Modified multi-stage integrators
>
> ▶ The modified counterparts of BCSS and ME are designed by **minimizing (expected) error in the modified Hamiltonians**.
> ▶ Two- and three-stage versions are available.

One-parameter family of **three-stage schemes** are competitors to VV and two-stage integrators:

$$\psi_{\Delta t} = \phi^B_{b\Delta t} \circ \phi^A_{a\Delta t} \circ \phi^B_{(\frac{1}{2}-b)\Delta t} \circ \phi^A_{(1-2a)\Delta t} \circ \phi^B_{(\frac{1}{2}-b)\Delta t} \circ \phi^A_{a\Delta t} \circ \phi^B_{b\Delta t},$$

with $b \in (0, 0.16]$ and $a = (1 - 2b)/4(1 - 3b)$.

Modified splitting schemes

Modified Hamiltonians

4th order modified Hamiltonians for **two-stage splitting integrators** in terms of quantities available during a simulation[8]:

$$\tilde{H}_{\Delta t}^{[4]}(\mathbf{q}, \mathbf{p}) = H + \Delta t^2 \left(\lambda \mathbf{p}^T M^{-1} U_{\mathbf{qq}}(\mathbf{q}) M^{-1} \mathbf{p} + \mu U_{\mathbf{q}}(\mathbf{q})^T M^{-1} U_{\mathbf{q}}(\mathbf{q}) \right),$$

where $\lambda = \frac{6b-1}{24}$, $\mu = \frac{6b^2-6b+1}{12}$.

---

[8] Radivojević, "Enhancing Sampling in Computational Statistics Using Modified Hamiltonians", 2016.

Modified splitting schemes

Using harmonic oscillator analysis, upper bound for the **expectation w.r.t. the modified density of the modified energy error**[9]:

$$\mathbb{E}(\Delta \tilde{H}) \leq \tilde{\rho}(h, b) = \frac{(MB_h + C_h)^2}{2M(1 - A_h^2)},$$

where A_h, B_h and C_h are functions of the integrator parameter b and the dimensionless step size h, and $M = \frac{1 + 2h^2\mu}{1 + 2h^2\lambda}$.

---

[9] Radivojević, "Enhancing Sampling in Computational Statistics Using Modified Hamiltonians", 2016.

Modified splitting schemes

Given **modified Hamiltonians** and the expressions for upper bound $\tilde{\rho}$, the **counterparts of ME and BCSS** are derived specifically for **methods sampling with shadow Hamiltonians**, e.g. GSHMC.

It has been found in Radivojević et al., "Multi-stage splitting integrators for sampling with modified Hamiltonian Monte Carlo methods", 2018:

- ▶ Two-stage modified integrators: M-ME2 ($b = 0.230907$), M-BCSS2 ($b = 0.230907$).
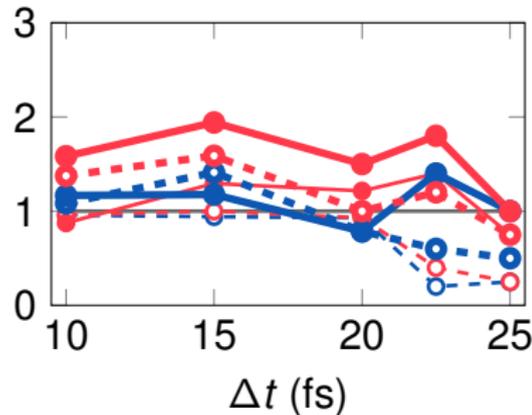- ▶ Three-stage modified integrators: M-ME3 ($b = 0.142757$), M-BCSS3 ($b = 0.144115$).
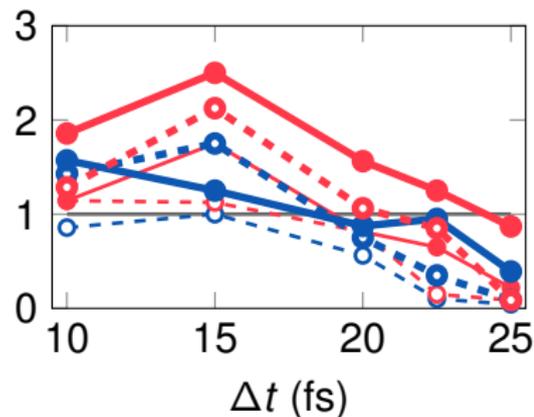
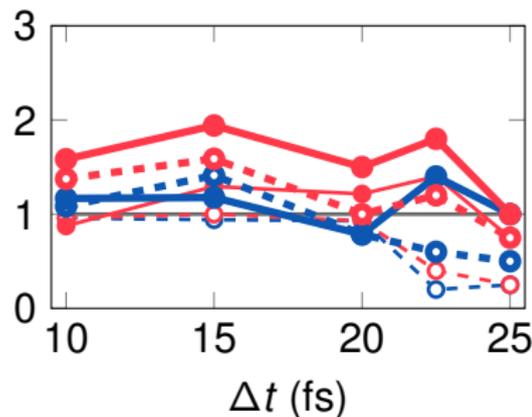# Modified splitting schemes

Effective Sample Size (ESS) normalized with respect to Verlet
Big ESS $\Rightarrow$ efficient sampling
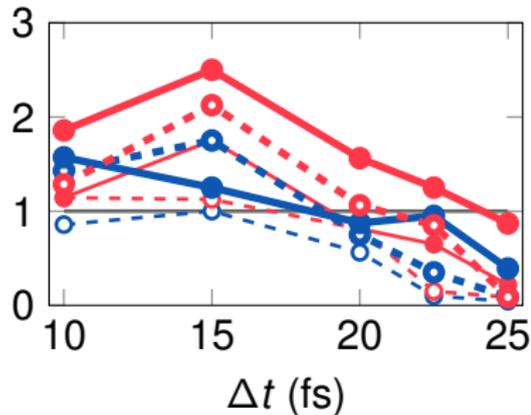
Modified splitting schemes
Effective Sample Size (ESS) normalized with respect to Verlet
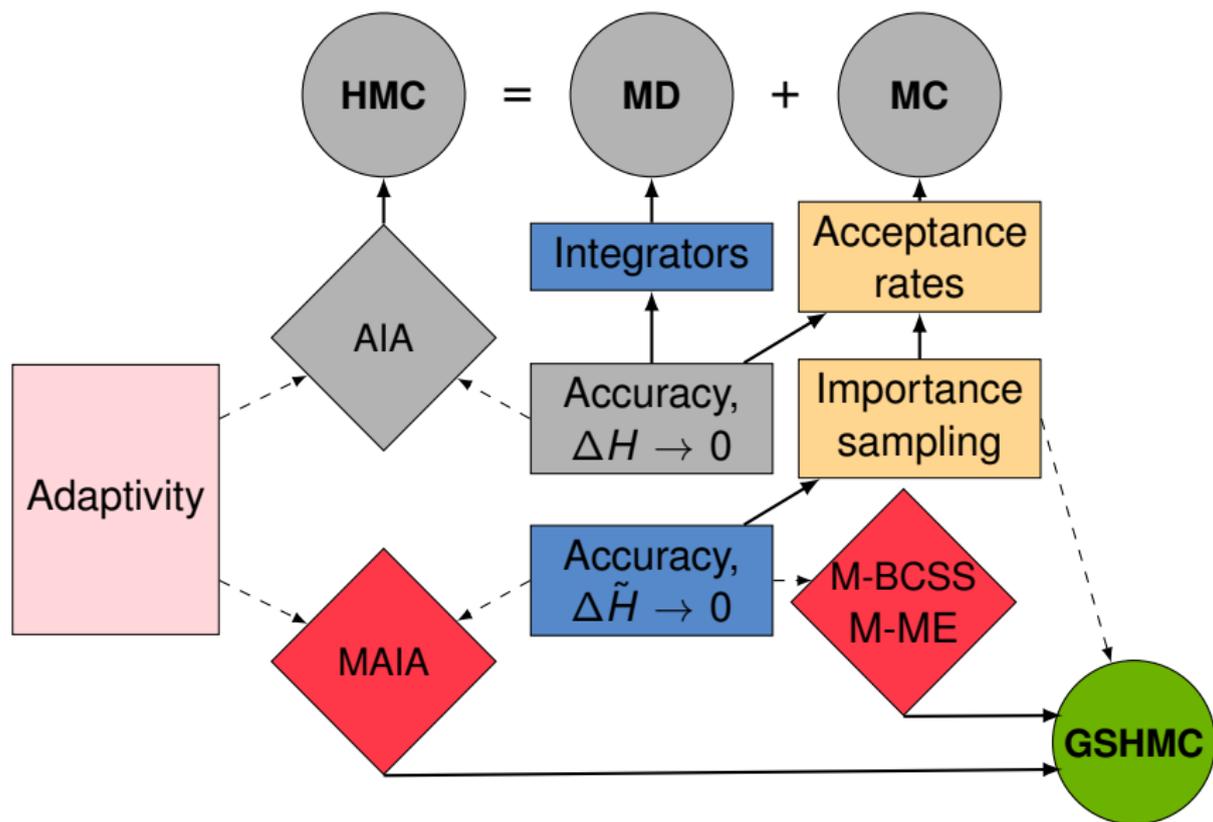### Big ESS ⇒ efficient sampling

* M-BCSS3 provided the **highest values of ESS** for all choices of step sizes but the last one. Up to 2.5 times better sampling.
* M-BCSS3 produced the **highest absolute ESS** at $\Delta t = 15$ fs: 53.

Modified splitting schemes

What are the best values of b?

Modified Adaptive Integration Approach

Modified Adaptive Integration Approach

Modified Adaptive Integration Approach

- ▶ **Objective**: To define a **system specific**, unique for each application, **two-stage integrator** which provides the best simulation **accuracy** and **performance** using **modified Hamiltonians**.

---

[10]Akhmatskaya et al., "Adaptive splitting integrators for enhancing sampling efficiency of modified Hamiltonian Monte Carlo methods in molecular simulation", 2017.

Modified Adaptive Integration Approach

- ▶ **Objective**: To define a **system specific**, unique for each application, **two-stage integrator** which provides the best simulation **accuracy** and **performance** using **modified Hamiltonians**.

- ▶ AIA for methods sampling with **modified Hamiltonians**.

- ▶ The same ideas as in AIA but the **expected error of modified energy** w.r.t. the **modified density** is minimized.

- ▶ The Modified Adaptive Integration Approach (MAIA) requires[10]:
    - ▶ **Modified Hamiltonians** for two-stage splitting integrators.
    - ▶ **Upper bound** for the expectation of modified energy error.

---

[10] Akhmatskaya et al., "Adaptive splitting integrators for enhancing sampling efficiency of modified Hamiltonian Monte Carlo methods in molecular simulation", 2017.

Modified Adaptive Integration Approach

The normalizationo factor. Non-linear stability condition[11]

$$h_{n:m} = \frac{2}{\tilde{\omega}} \sin\left(\frac{m\pi}{n}\right).$$

$m = 1$ gives the lowest-order resonances:

- ▶ If $n = 2$, $h < 2/\tilde{\omega}$, the linear stability condition of Verlet;
- ▶ if $n = 3$, $h < \sqrt{3}/\tilde{\omega}$;
- ▶ if $n = 4$, $h < \sqrt{2}/\tilde{\omega}$.

These factors were predicted by numerical experiments.
The smoother behaviour of shadow Hamiltonians in comparison with Hamiltonians can explain it.

---

[11]Schlick, *Molecular Modeling and Simulation: An Interdisciplinary Guide*, 2002.

Modified Adaptive Integration Approach

1. Given:
    - ▶ The integration step size Δt, in units of measure,
    - ▶ $\tilde{\omega}$, the fastest oscillatory frequency in the system.
2. Calculate the normalized step size

$$\tilde{h} := \tilde{S}\tilde{\omega}\Delta t,$$

where $\tilde{S} = \sqrt{3}$.
3. Find $b^* = \arg\min \|\tilde{\rho}\|_{(\tilde{h})} = \arg\min \max_{0 < h < \tilde{h}} \tilde{\rho}(h, b)$.
4. The resulting adaptive integrator:

$$\psi_{\Delta t} = \phi^B_{b^*\Delta t} \circ \phi^A_{\Delta t/2} \circ \phi^B_{(1-2b^*)\Delta t} \circ \phi^A_{\Delta t/2} \circ \phi^B_{b^*\Delta t}.$$

Extended Modified Adaptive Integration Approach

The performance of GSHMC depends on the **momentum update acceptance rate** α_p.

The Metropolis test uses the **extended "Hamiltonian" error** $\Delta\hat{H} = \hat{H}(\mathbf{q}, \mathbf{p}^*, \mathbf{u}^*) - \hat{H}(\mathbf{q}, \mathbf{p}, \mathbf{u})$, which **depends on the choice of an angle** φ.

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[12]Akhmatskaya et al., "Adaptive splitting integrators for enhancing sampling efficiency of modified Hamiltonian Monte Carlo methods in molecular simulation", 2017.

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- **Objective of Extended MAIA (e-MAIA)**[12]: To define a system specific, unique for each application, two-stage integrator which provides the best simulation accuracy and performance using modified Hamiltonians, and **adjusts the value of** φ **to achieve a desired acceptance rate for the momentum update step**.

---

[12] Akhmatskaya et al., "Adaptive splitting integrators for enhancing sampling efficiency of modified Hamiltonian Monte Carlo methods in molecular simulation", 2017.

Extended Modified Adaptive Integration Approach

Using harmonic oscillator analysis, the function which bounds the **expected extended Hamiltonian error** can be found as

$$\tau(\bar{h}, b^*, \varphi) = \frac{2\bar{h}^4 \lambda^{*2} \sin^2 \varphi}{1 + 2\bar{h}^2 \lambda^*},$$

where $\lambda^* = \frac{6b^* - 1}{24}$ and $\bar{h} = \bar{\omega} \Delta t$ with $\bar{\omega}$ the slowest oscillatory frequency in the system.

Extended Modified Adaptive Integration Approach

Algorithm

1. Given:
    - The integration step size Δt, in units of measure,
    - a target acceptance rate AR_p for the momentum update,
    - an initial value φ_0 of the angle φ,
    - $\bar{\omega}$ and $\tilde{\omega}$, the slowest and the fastest oscillatory frequencies in the system, respectively.

2. Find b^* as in MAIA.

3. Find the angle φ^* as

$$\varphi^* = \arg \min_{\varphi \in (0, \pi/2]} \left| -\frac{\log(AR_p)}{D} - \tau(\bar{h}, b^*, \varphi) \right|.$$
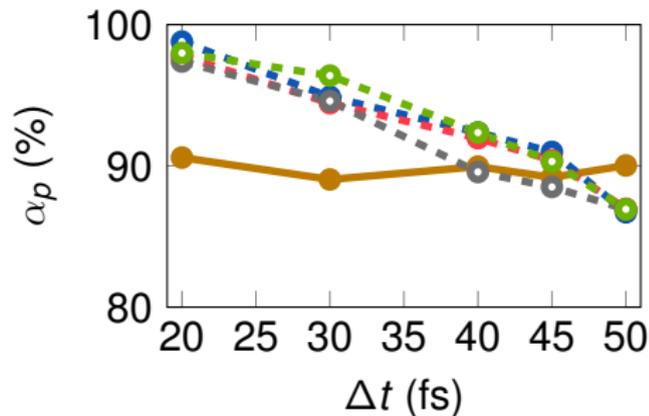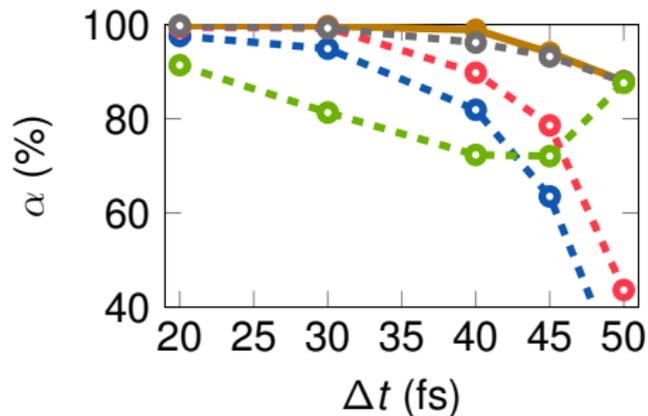
4. If $\varphi^* \leq \varphi_0$:
    - Decrease AR_p and go to step 3, or
    - define $\sigma(h, b, \varphi_0) := \rho(h, b) + \tau(h, b, \varphi_0)$ and choose b^{**} that minimizes $\max_{0 < h < \tilde{h}} \sigma(h, b, \varphi_0)$.

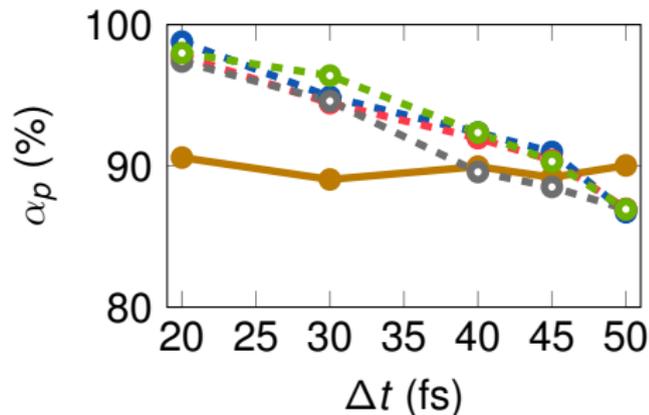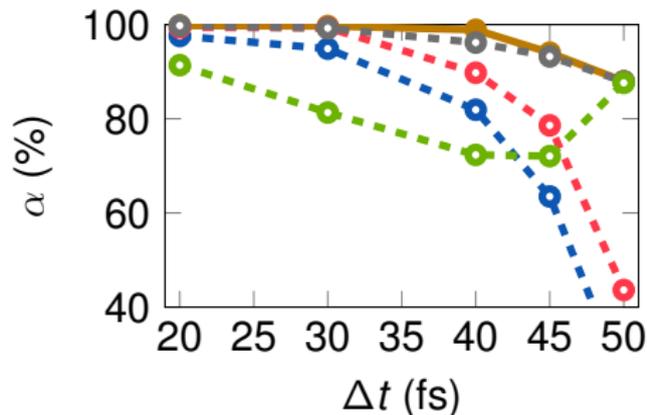# Extended Modified Adaptive Integration Approach

Position acceptance rates

Momenta acceptance rates

Extended Modified Adaptive Integration Approach

Position acceptance rates

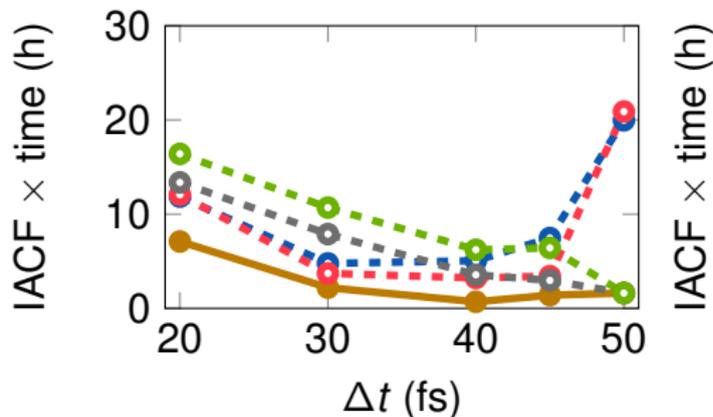Momenta acceptance rates

Legend: e-MAIA, M-ME, M-BCSS, VV, AIA

* e-MAIA provides the **best acceptance rates for positions** regardless a choice of the step size.
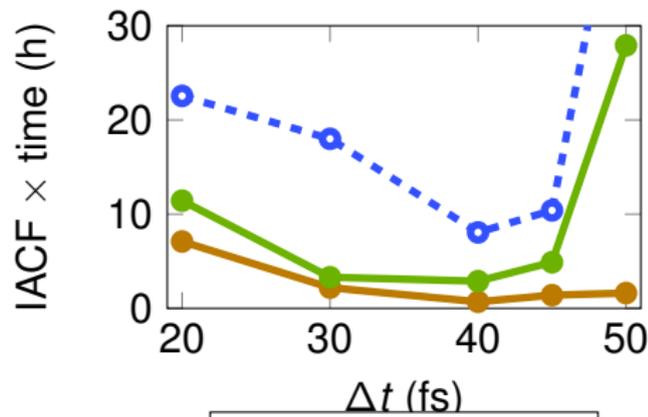* e-MAIA **reaches the target acceptance rate for momenta of 90 %** for each step size.

Extended Modified Adaptive Integration Approach

Time normalized Integrated Autocorrelation Functions (IACF)

Low IACF $\Rightarrow$ efficient sampling

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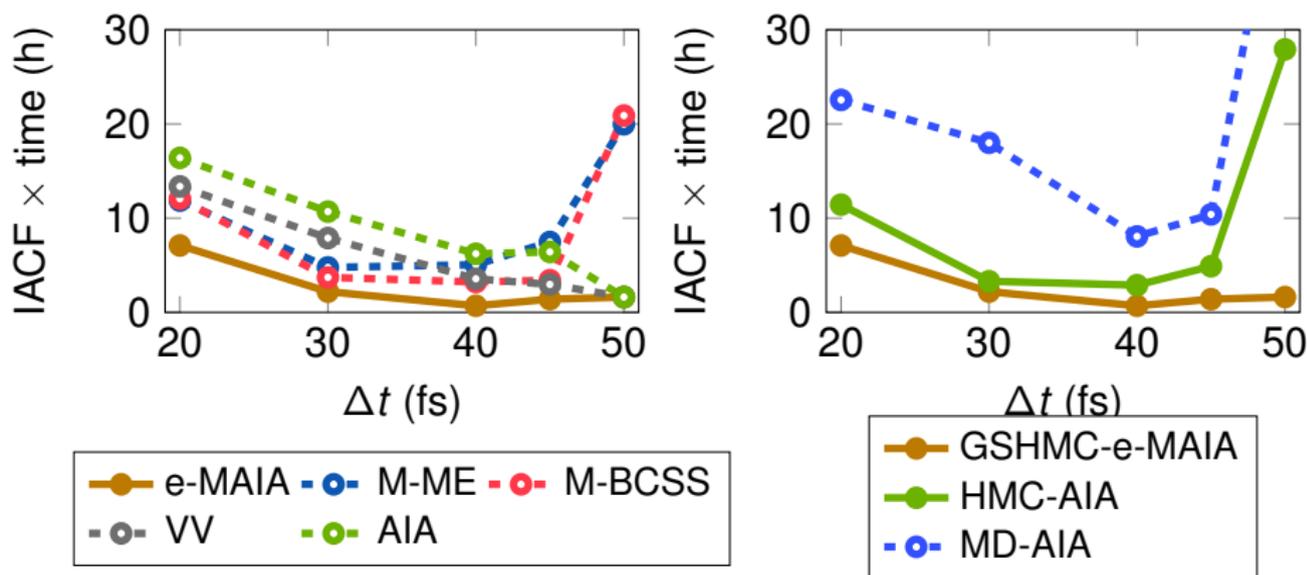
* e-MAIA provides the **lowest IACF** using GSHMC regardless a choice of step size Δt. Up to 3.5 times better sampling.
* e-MAIA with GSHMC provides the **lowest IACF** compared to HMC and MD using AIA. Up to 50 times better sampling.

49

Take home messages

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- **Constrained mechanical system**, while preserving the symplectic structure of Hamiltonian problems, allow us to model a broader range of realistic systems.
- **Multi-stage splitting integrators** and **adaptive** approaches allow us to outperform the standard method velocity Verlet.

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